# ADVANCED ANALYSIS OF SHORT-FIBER POLYMER COMPOSITE MATERIAL BEHAVIOR 

A Dissertation presented to the Faculty of the Graduate School University of Missouri - Columbia

In Partial Fulfillment of the Requirements for the Degree<br>Doctor of Philosophy

by<br>DAVID ABRAM JACK

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The undersigned, appointed by the Dean of the Graduate School, have examined the dissertation entitled

# ADVANCED ANALYSIS OF SHORT-FIBER POLYMER COMPOSITE MATERIAL BEHAVIOR 

Presented by David Abram Jack, a candidate for the degree of Doctor of Philosophy, and hereby certify that in their opinion it is worthy of acceptance.

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To my best friend and dear wife Trisha, who has patiently let me pursue my varied research interests.

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# ADVANCED ANALYSIS OF SHORT-FIBER POLYMER COMPOSITE MATERIAL BEHAVIOR 

David Abram Jack<br>Dr. Douglas E. Smith, Dissertation Supervisor<br>ABSTRACT

Short-fiber polymer composites experience widespread use in many industrial applications, where the orientation state of the short-fibers within the polymer matrix define the material properties of the composite structure. Due to the extensive use of these short fiber products, it is necessary to develop an accurate understanding of the fiber orientation kinematics and the resultant material characteristics of the processed part.

This dissertation presents techniques to accurately represent the orientation state of fibers during the part molding process, and from the orientation state within the processed part predict, statistically, the resulting elastic material characteristics. Higher-order representations of the fiber orientation distribution are presented through the sixth-order orientation tensor fitted closure, and results yield a material stiffness tensor with fewer planes of material symmetry than current fourth-order closures while retaining a more accurate representation of fiber orientation. Analytic expressions for material stiffness expectation and variance are developed and validated through the Monte-Carlo method, and provide a more thorough understanding into the statistical nature of the material stiffness tensor. This work concludes with the presentation of the directional diffusion model for fiber collisions, and results demonstrate a significant delay in fiber alignment beyond existing models while retaining an identical steady state orientation.

## CHAPTER 1

## INTRODUCTION

Short-fiber polymer composites experience widespread use in many industrial applications due in large part to their high strength to weight ratio. The orientation state of the short-fibers within the polymer matrix defines the material properties of the composite structure. Therefore a thorough understanding of fiber orientation during mold filling, and the associated material characteristics, is necessary for accurate product and process analysis. This dissertation seeks to present techniques for an accurate representation of the fiber orientation state during the part molding process, and from the orientation state within the processed part predict the resulting elastic material characteristics.

Perhaps the most important application for fiber orientation prediction is the mechanical property evaluation of the short fiber composite. Mechanical properties may be computed from the distribution function of fibers, ergo a more accurate representation of the distribution function is expected to yield more accurate mechanical properties. Current methods to predict material stiffness behavior from orientation tensors are based on the volume average of a transversely isotropic stiffness tensor. The sixth-order closures are not constrained by the transversely isotropic assumption, hence the formulation of material stiffness predictions warrant further investigation.

Fiber orientation closure methods and material stiffness prediction formulations are of little worth if the underlying model for fiber orientation flow kinematics is flawed. Current industrial models representing interacting fiber flow have been suggested to over predict the alignment of fibers during the transient solution, and for
simple flows the steady state solution is attained at a faster rate than experimental results indicate. Until recently, this characteristic has been acceptable for industrial applications due to significant uncertainty in the fabrication process. With increasing industrial demands it is necessary to develop methods to accurately model the entire transient solution of the fiber orientation probability distribution function.

The following dissertation investigates procedures to accurately predict the orientation state of fiber suspensions, with considerable effort devoted to material property predictions given an orientation state. The focus is on three interrelated topics to analyze short-fiber behavior in polymer products. The first presents accurate approximations of fiber distributions through the sixth-order closures. The second studies mechanical property analysis procedures through the employment of closure methods and mathematical expressions are developed for the statistical nature of short fiber distributions. The third presents an objective, phenomenological, directionally dependant diffusion model for fiber collisions shown to have several desirable characteristics to qualitatively represent fiber kinematics.

In Chapter 3, three sixth-order closures are presented along with an analysis of their effects to accurately represent the fiber orientation state. These closures allow fiber orientation representations unavailable through the fourth-order closures, and it is demonstrated that the sixth-order fitted invariant based closure is not constrained by the orthotropic assumption inherent to the fourth-order closures.

With the capability of sixth-order closures to represent material behavior without the orthotropic assumption, Chapter 4 investigates the effect of closures on material property predictions to determine the effectiveness in representing various orientation states. To develop material property prediction methods, this work introduces
the Laplace series of complex spherical harmonics to expand the fiber orientation probability distribution function assuming only that the distribution is symmetric about a single axis. This method makes no assumptions regarding the form of the orientation tensors beyond their inherent symmetry, thereby providing a means for computing material behavior from orientation tensors, and bypasses the issue of a closure selection. In Chapter 5, an analytic form is developed to evaluate the variance of the stiffness tensor's distribution, which requires orientation tensors through the eighth-order. Statistical results obtained from the method of Monte-Carlo to obtain the sample mean and variance of material stiffness tensor components are compared to computed results obtained from the analytically derived expectation and variance. Results are studied in depth for a simple analytic function and for industrially relevant center-gated disk flow obtained from numerical distribution function simulations to demonstrate the effectiveness of the proposed method.

Chapter 6 presents a directionally dependant diffusion model and results demonstrate the capability to adjust both the final orientation state and the transient solution through two scalable parameters. Shearing and elongational flows are investigated and illustrate the flexibility of the proposed model to diminish the orientation solution rate while retaining an accurate representation of the final orientation state.

The dissertation concludes with a discussion of the scientific advancements from the present work. Limitations of the fiber orientation models presented are discussed with suggestions for further enhancements of the current work. Particular emphasis is given to directions of future research endeavors and a listing is given of possible actions that may be undertaken to promote current scientific understanding.

## CHAPTER 2

## FIBER ORIENTATION ANALYSIS

Due to the extensive use of short-fiber polymer composites in industrial applications and the strong dependance of these products on their manufacturing process, a complete investigation into fiber orientation characteristics required. This chapter sets the framework to develop effective methods to analyze the flow kinematics and fiber orientation relationship. Development starts from the representation of a single fiber and transitions to the distribution of fibers necessary for effective industrial applications. Considerations such as bulk fluid deformation, fiber volume fraction, and fiber aspect ratio are incorporated to evaluate the motion of a single fiber. Current models to represent fiber interactions are introduced along with a discussion of known limitations. The fiber orientation tensor equations of motion allow rapid computations of industrial simulations for complex parts. Unfortunately, their use leads to the classical closure problem which has been extensively investigated in the literature. Several closures are introduced, along with the need for higher order closures. Since the material response of the final product is essential for a full analysis, current approaches for material property predictions from the orientation tensors are introduced. The discussion concludes with the introduction of the Laplace series of spherical harmonics which will prove essential in later chapters to allow development of a closed form solution for material property behavior from the fiber orientation distribution function.


Figure 2.1: Coordinate system defining $\boldsymbol{p}(\theta, \phi)$ along with the angles $\theta$ and $\phi$.

### 2.1 Fiber Orientation Representation

The orientation of a single rigid fiber within a polymer matrix can be described by the angle pair, $(\theta, \phi)$, or by the unit vector $\boldsymbol{p}(\theta, \phi)$ aligned along the axis of the fiber as shown in Figure 2.1 which are related through

$$
\boldsymbol{p}(\theta, \phi)=\left\{\begin{array}{c}
\sin \theta \cos \phi  \tag{2.1}\\
\sin \theta \sin \phi \\
\cos \theta
\end{array}\right\}
$$

Observe that a fiber along $\boldsymbol{p}(\theta, \phi)$ will be indiscernible from a fiber aligned along $-\boldsymbol{p}(\theta, \phi)$ since the two ends of the fiber are indistinguishable [1], therefore any description of the orientation must satisfy

$$
\begin{equation*}
(\theta, \phi) \rightarrow(\pi-\theta, \phi+\pi) \tag{2.2}
\end{equation*}
$$

Fibers are rarely, if ever, perfectly aligned along a single direction, and instead each discrete fiber will have an orientation that varies from nearby fibers. There are examples where discrete fibers have been modeled in simple flows for small numbers
of fibers [2], but for typical industrial use, modeling individual fibers is computationally impractical. Therefore, the discrete set of fibers is assumed to satisfy a given continuous probability distribution function $\psi(\theta, \phi)$ defined such that the probability of a fiber oriented between the angles $\theta_{i}$ and $\theta_{i}+d \theta$ and between $\phi_{i}$ and $\phi_{i}+d \phi$ is defined as [3]

$$
\begin{equation*}
P\left(\theta_{i} \leq \theta \leq \theta_{i}+d \theta, \phi_{i} \leq \phi \leq \phi_{i}+d \phi\right)=\psi(\theta, \phi) \sin \theta_{i} d \theta d \phi \tag{2.3}
\end{equation*}
$$

Observe that Equation (2.2) will cause the distribution to be an even function, i.e.

$$
\begin{equation*}
\psi(\theta, \phi)=\psi(\pi-\theta, \phi+\pi) \tag{2.4}
\end{equation*}
$$

Since every fiber is described by some angle pair $(\theta, \phi) \in \mathbb{S}^{2}$, the integral of the distribution function over the unit sphere $\mathbb{S}^{2}$ must equate to one

$$
\begin{equation*}
\oint_{\mathbb{S}^{2}} \psi(\theta, \phi) \mathrm{d} \mathbb{S}=\int_{0}^{2 \pi} \int_{0}^{\pi} \psi(\theta, \phi) \sin \theta \mathrm{d} \theta \mathrm{~d} \phi=1 \tag{2.5}
\end{equation*}
$$

which is often referred to as the normalization condition. The fiber distribution function $\psi(\theta, \phi)$ is a complete description if the orientation of a fiber is statistically uncorrelated with that of any of its neighbors, and is considered to be a continuous function that is assumed to vary smoothly with position [1]. The case of "orientational clustering" has been considered by Ranganathan and Advani [4] where the local clustering of the fibers causes a local inhomogeneity of the distribution function. This phenomena is relegated to complex flow fields, and is neglected here since all results will be for simple flow fields.

The fiber orientation probability distribution function presented in Equation (2.3) is defined on the surface of the unit sphere. Therefore an understanding of spherical coordinates and the derivatives of the spherical unit vectors is essential. The spherical
coordinate unit vectors $\boldsymbol{r}, \boldsymbol{\phi}$, and $\boldsymbol{\theta}$ are [5]

$$
\boldsymbol{r}=\left\{\begin{array}{c}
\sin \theta \cos \phi  \tag{2.6}\\
\sin \theta \sin \phi \\
\cos \theta
\end{array}\right\} \quad \boldsymbol{\phi}=\left\{\begin{array}{c}
-\sin \phi \\
\cos \phi \\
0
\end{array}\right\} \quad \boldsymbol{\theta}=\left\{\begin{array}{c}
\cos \phi \cos \theta \\
\sin \phi \cos \theta \\
-\sin \theta
\end{array}\right\}
$$

where the bold font on a lower case letter designates a vector belonging to $\mathbb{R}^{3}$. Observe, the spherical coordinate $\boldsymbol{r}$ is identical to the definition of the unit vector $\boldsymbol{p}$ aligned along the fiber axis given in Equation (2.1) and shown in Figure 2.1. Unlike Cartesian coordinates, derivatives of the coordinate unit vectors in spherical coordinates are not identically zero. The derivatives of the unit coordinate vectors are [5]

$$
\begin{array}{lll}
\frac{\partial \boldsymbol{r}}{\partial r}=\mathbf{0} & \frac{\partial \boldsymbol{r}}{\partial \phi}=\sin \theta \boldsymbol{\phi} & \frac{\partial \boldsymbol{r}}{\partial \theta}=\boldsymbol{\theta} \\
\frac{\partial \boldsymbol{\phi}}{\partial r}=\mathbf{0} & \frac{\partial \boldsymbol{\phi}}{\partial \phi}=-\cos \theta \boldsymbol{\theta}-\sin \theta \boldsymbol{r} & \frac{\partial \boldsymbol{\phi}}{\partial \theta}=\mathbf{0}  \tag{2.7}\\
\frac{\partial \boldsymbol{\theta}}{\partial r}=\mathbf{0} & \frac{\partial \boldsymbol{\theta}}{\partial \phi}=\cos \theta \boldsymbol{\phi} & \frac{\partial \boldsymbol{\theta}}{\partial \theta}=-\boldsymbol{r}
\end{array}
$$

Additionally, it will be useful to recognize that the Kronecker Delta $\delta_{i j}$ is written as

$$
\begin{equation*}
\delta_{i j}=r_{i} r_{j}+\theta_{i} \theta_{j}+\phi_{i} \phi_{j} \tag{2.8}
\end{equation*}
$$

where $r_{i}, \theta_{i}$ and $\phi_{i}$ are the $i^{\text {th }}$ components of the unit vectors $\boldsymbol{r}, \boldsymbol{\theta}$ and $\boldsymbol{\phi}$ given in Equation (2.6), respectively. The $i^{\text {th }}$ component of the gradient on the surface of the unit sphere where $r=1$ is

$$
\begin{equation*}
\nabla_{i}=\theta_{i} \frac{\partial}{\partial \theta}+\phi_{i} \frac{1}{\sin \theta} \frac{\partial}{\partial \phi} \tag{2.9}
\end{equation*}
$$

and the Laplacian in spherical coordinates on the surface of the unit sphere is simply

$$
\begin{equation*}
\nabla^{2}=\frac{1}{\sin ^{2} \theta} \frac{\partial^{2}}{\partial \phi^{2}}+\frac{\cos \theta}{\sin \theta} \frac{\partial}{\partial \theta}+\frac{\partial^{2}}{\partial \theta^{2}} \tag{2.10}
\end{equation*}
$$

The fiber orientation distribution function is a complete description of the fiber orientation state, but calculations with the distribution function are too cumbersome to apply to industrially relevant flows due to computational considerations. Additionally, $\psi(\theta, \phi)$ does not provide a convenient interpretation of physical behavior [1,6,7].

An alternative approach is to consider an average orientation property over a sufficiently large enough volume to contain many fibers, but small enough such that the statistics of the orientation are uniform throughout. This assumption is valid when the volume is small in regards to the dimensions of the part but large with respect to the fiber length [8]. The statistical behavior of the distribution function is captured in a compact form for rapid computations using the moments of the distribution, commonly referred to as the orientation tensors, defined as [6]

$$
\begin{array}{rlrl}
a_{i j} & =\oint_{\mathbb{S}^{2}} p_{i} p_{j} \psi(\theta, \phi) \mathrm{d} \mathbb{S} & a_{i j k l}=\oint_{\mathbb{S}^{2}} p_{i} p_{j} p_{k} p_{l} \psi(\theta, \phi) \mathrm{d} \mathbb{S} \\
a_{i j \ldots} & =\oint_{\mathbb{S}^{2}} p_{i} p_{j} \ldots \psi(\theta, \phi) \mathrm{d} \mathbb{S} \tag{2.11}
\end{array}
$$

Due to the symmetric nature of the fiber orientation probability distribution function in Equation (2.4), odd ordered orientation tensors integrate to zero and will not be discussed further. Through the application of Equations (2.1) and (2.11) orientation tensors can be shown to be completely symmetric

$$
\begin{gather*}
a_{i j}=a_{j i} \\
a_{i j k l}=a_{k l i j}=a_{j i k l}=a_{i l k j}=\cdots  \tag{2.12}\\
a_{i j k l m n}=a_{j i k l m n}=a_{k l i j m n}=a_{m n k l i j}=a_{i l k j m n}=a_{i n k l m j}=\cdots
\end{gather*}
$$

Note that $a_{i j k l}$ enjoys more symmetry than that of an anisotropic stiffness or compliance tensor (see e.g. [9, 10]). In addition, higher order orientation tensors completely describe lower order orientation tensors which can be shown using the normalization condition of the distribution function $\psi(\theta, \phi)$ from Equation (2.5) along
with Equation (2.1) and Equation (2.11)

$$
\begin{align*}
a_{i j \cdots \alpha \beta \gamma \gamma} & =\oint_{\mathbb{S}^{2}} p_{i} p_{j} \ldots p_{\alpha} p_{\beta} p_{\gamma} p_{\gamma} \psi(\theta, \phi) \mathrm{d} \mathbb{S} \\
& =\oint_{\mathbb{S}^{2}} p_{i} p_{j} \ldots p_{\alpha} p_{\beta}\left(p_{1} p_{1}+p_{2} p_{2}+p_{3} p_{3}\right) \psi(\theta, \phi) \mathrm{d} \mathbb{S} \\
& =\oint_{\mathbb{S}^{2}} p_{i} p_{j} \ldots p_{\alpha} p_{\beta}\left(\|\boldsymbol{p}\|^{2}\right) \psi(\theta, \phi) \mathrm{d} \mathbb{S} \\
& =\oint_{\mathbb{S}^{2}} p_{i} p_{j} \ldots p_{\alpha} p_{\beta}(1) \psi(\theta, \phi) \mathrm{d} \mathbb{S}=a_{i j \cdots \alpha \beta} \tag{2.13}
\end{align*}
$$

where here and throughout the remainder of the text repeated indices imply summation, i.e. $a_{i i}=\sum_{i=1}^{3} a_{i i}=a_{11}+a_{22}+a_{33}$. Due to the symmetry conditions of Equation (2.12), there are six independent components of $a_{i j}$ which is reduced to five components when accounting for the normalization condition of Equation (2.5). Similarly, the fourth-order orientation tensor $a_{i j k l}$ has 14 independent components, the sixth-order orientation tensor $a_{i j k l m n}$ has 27 independent components, the eighthorder orientation tensor $a_{i j k l m n o p}$ has 44 independent components, and the tenthorder orientation tensor $a_{i j k l m n o p q r}$ has 65 independent components when accounting for normalization and symmetry. Additionally, due to the normalization condition of Equation (2.13) it is trivially shown that lower ordered orientation tensors are completely contained within the higher ordered orientation tensors

$$
\begin{align*}
1 & =a_{i j}  \tag{2.14}\\
a_{i j} & =a_{i j p p}  \tag{2.15}\\
a_{i j k l} & =a_{i j k l q q} \tag{2.16}
\end{align*}
$$

Although the tensors may seem an unusual means to describe fiber orientation, they have a simple physical interpretation. For example, take an isotropic, i.e. 3-D random orientation, state with $\psi(\theta, \phi)=\frac{1}{4 \pi}$ where a sample set of fibers is shown in

Figure 2.2, then the second order orientation tensor is simply

$$
a_{i j}=\left[\begin{array}{ccc}
\frac{1}{3} & 0 & 0  \tag{2.17}\\
0 & \frac{1}{3} & 0 \\
0 & 0 & \frac{1}{3}
\end{array}\right]
$$

When all fibers lie along the $x_{1}$ axis such as the fiber set in Figure 2.3, the second-order orientation tensor is

$$
a_{i j}=\left[\begin{array}{lll}
1 & 0 & 0  \tag{2.18}\\
0 & 0 & 0 \\
0 & 0 & 0
\end{array}\right]
$$

It will be shown later that the orientation tensors are an incomplete representation of the fiber distribution function, but are very useful in practical industrial simulations of fiber orientation kinematics.

### 2.2 Fiber Orientation Kinematics

Most fiber orientation analysis are based on the Folgar and Tucker model [11] which incorporates fiber interaction effects through a phenomenological diffusion term. Their model adds to the Jeffery model [12] for fiber motion in a Newtonian solvent, and has enjoyed wide acceptance for simulations of the fiber orientation kinematics.

A comprehensive treatment of fiber suspension flow in complex geometries involves flow/orientation coupling. It is well understood that fibers orient in response to flow kinematics, while the suspension rheology is defined in part by the concentration and orientation of the suspended fibers. For example, Dinh and Armstrong [13] provide a rheological equation of state for semi-concentrated suspensions of stiff fibers in a Newtonian solvent. Flow simulations that incorporate flow/orientation coupling further emphasize the need for including these fully coupled evaluations to make accurate predictions in certain geometries. Lipscomb, et al., [14] showed that the size of the corner vortex in axisymmetric contraction changed significantly when fibers


Figure 2.2: Isotropic orientation state, $\psi(\theta, \phi)=\frac{1}{4 \pi}$.


Figure 2.3: Uniaxially aligned orientation state along $x_{1}$ axis.
were added, and VerWeyst and Tucker [15] exposed the influence of fiber concentration on flow near the gate of an injection molded part.

In the same simulations, however, VerWeyst and Tucker [15] demonstrated that the effects from incorporating the two-way flow/orientation coupling decays rapidly with increasing distance from the gate. This latter result supports the earlier conclusion by Tucker [16] that flow and fiber orientation evaluations may be decoupled for flows in narrow gaps where the lubrication approximation applies. Accordingly, throughout the present work the effect of fiber orientation on the flow kinematics will be neglected to retain the focus solely on fiber orientation analysis. It is left for future investigations to pursue the full coupling between the fiber orientation analysis and the flow kinematics.

### 2.2.1 Jeffery's Model

The dynamics of the motion of fibers in a flow field are given by Jeffery [12] where the motion of individual rigid ellipsoids in a Newtonian solvent is evaluated from first principles. The Jeffery model assumes the fluid velocity is a linear function of position in a neighborhood of the fiber and all inertia and body forces are negligible. Jeffery solved the particle motion by requiring the net forces and moments on the ellipsoid equal zero. Therefore the centroid of the particle moves with the bulk flow of the surrounding matrix. Jeffery's solution in vector notation and index notation are, respectively, written as

$$
\begin{align*}
\frac{D \boldsymbol{p}^{h}}{D t} & =\dot{\boldsymbol{p}}^{h}=-\frac{1}{2} \boldsymbol{\omega} \cdot \boldsymbol{p}+\frac{1}{2} \lambda(\dot{\gamma} \cdot \boldsymbol{p}-\dot{\boldsymbol{\gamma}}: \boldsymbol{p} \boldsymbol{p} \boldsymbol{p})  \tag{2.19}\\
\frac{D p_{i}^{h}}{D t} & =\dot{p}_{i}^{h}=-\frac{1}{2} \omega_{i j} p_{j}+\frac{1}{2} \lambda\left(\dot{\gamma}_{i j} p_{j}-\dot{\gamma}_{k l} p_{k} p_{l} p_{i}\right) \tag{2.20}
\end{align*}
$$

where $t$ is time, $\dot{\boldsymbol{p}}^{h}$ is defined as the time derivative of the hydraulic component of rotational motion of the ellipsoid, $\lambda$ depends on the fiber aspect ratio $a_{r}$ where $\lambda=\left(a_{r}^{2}-1\right) /\left(a_{r}^{2}+1\right)[6]$ (for an alternative formulation, see e.g. Harris and Pittman $[17,18])$. In Equations (2.19) and (2.20), $\dot{\gamma}_{i j}$ is the rate of deformation tensor and $\omega_{i j}$ is the vorticity tensor written, respectively, as

$$
\begin{align*}
\dot{\gamma}_{i j} & =\frac{\partial v_{j}}{\partial x_{i}}+\frac{\partial v_{i}}{\partial x_{j}}  \tag{2.21}\\
\omega_{i j} & =\frac{\partial v_{j}}{\partial x_{i}}-\frac{\partial v_{i}}{\partial x_{j}} \tag{2.22}
\end{align*}
$$

where $v_{i}$ is the $i^{\text {th }}$ component of the velocity of the fluid. Observe in Equations (2.19) and (2.20) the ordinary derivative $\frac{d}{d t}$ has been replaced with the material derivative $\frac{D}{D t}$ to recognize that the orientation may not be spatially uniform but may convect with the bulk motion of the fluid where $\frac{D}{D t}=\frac{\partial}{\partial t}+\mathbf{v} \cdot \nabla$.

It is interesting to note that Jeffery's equation predicts that an ellipsoid in simple shearing flow will experience periodic motion labeled the Jeffery orbits. The time average of the Jeffery orbits will describe the general tendency of alignment, but in practical applications the Jeffery model is insufficient. The Jeffery model predicts that in simple shear flow after a period $T$, every fiber will return to its original orientation state. While this has been shown to be the case for single fibers, Jeffery orbits are not seen when multiple interacting fibers exist. Instead, a steady state orientation for the bulk distribution is often obtained after a short period of time with no oscillations in the distribution [1, 11, 19-21].

### 2.2.2 Effects from Fiber Interactions

Jeffery's model for fiber suspension kinematics requires there exist no fiber interactions, and is thus valid in the dilute regime only [1,22]. This occurs when the inverse
of the square of the aspect ratio $a_{r}$ of the fiber, defined as $a_{r}=L / D$ where $L$ is the fiber length and $D$ the fiber diameter, is much greater than the volume fraction of fibers, i.e. $V_{f} \ll \frac{1}{a_{r}^{2}}$. Typical fiber aspect ratios range from 10 to 20 [23], therefore for a suspension with an aspect ratio of 10 to be dilute, the fiber volume fraction would have to be much less than $1 \%$. The semi-dilute regime is defined [1] such that $\frac{1}{a_{r}^{2}} \ll V_{f} \ll \frac{1}{a_{r}}$, therefore for an aspect ratio of 10 , the semi-dilute regime would encompass a volume fraction range of $1 \% \ll V_{f} \ll 10 \%$. The concentrated regime is defined as the region where the volume fraction is greater than the inverse of the aspect ratio, i.e. $V_{f}>\frac{1}{a_{r}}$. Most typical industrial applications are in the concentrated regime, and as such the Jeffery model of Equation (2.19) is clearly not sufficient for industrial use.

Modeling the interaction between two fibers in a fluid suspension is difficult, and is the subject of continuing work. It is assumed that fiber interactions are due to volume averaged effects and the model for fiber interactions is similar to the theory of rotary Brownian motion $[24,25]$ where each of the ellipsoids experience small forces as they collide within the suspension causing torques on each ellipsoid. Rotary Brownian motion has seen extensive use in the dynamics of polymeric liquids whereby interactions between individual polymer chains in a suspension are modeled as being caused by directionally dependant diffusion processes [25]. In traditional Brownian motion, if the particles are not aligned by some outside force, the orientations will eventually become random, whereas for short fiber composite flow fibers will not reorient unless subjected to a deformation of the surrounding fluid. Assuming rigid fibers of uniform density and aspect ratio, the model of Bird [25] for rotary diffusion is accepted whereby the flow of a fiber distribution is given by the equation of mass
continuity as [25]

$$
\begin{equation*}
\frac{D \psi}{D t}=-\nabla \cdot\left[\dot{\boldsymbol{p}}^{h} \psi(\boldsymbol{p})-\nabla\left(D_{r} \psi(\boldsymbol{p})\right)\right]=-\nabla \cdot\left[\dot{\boldsymbol{p}}^{h} \psi(\boldsymbol{p})\right]+\nabla^{2}\left[D_{r} \psi(\boldsymbol{p})\right] \tag{2.23}
\end{equation*}
$$

where $\nabla$ and $\nabla^{2}$ represent, respectively, the gradient operator and the Laplacian operator on the surface of the unit sphere, and $D_{r}$ is the rotary diffusivity. At the present, it is sufficient to assume that $D_{r}$ may be a function of $(\theta, \phi)$ and as such cannot be brought out of the Laplacian. When the assumptions from the Jeffery model are taken into consideration, i.e. no fiber interaction, the rotary diffusion expression $D_{r}$ is set to zero. The form of Equation (2.23) has been accepted for rigid polymer chains [25] and in the case of $D_{r}$ being independent of direction simplifies to existing theories for modeling the interaction of fibers. In later chapters, a model to incorporate a directional dependance into the rotary diffusivity will be introduced.

### 2.2.3 Current Models for Fiber Interactions

Kamel and Mutel [26] proposed a form for the rotary diffusivity $D_{r}$ that is a function of volume fraction and independent of shear rate for a sufficiently large volume fraction of fibers. Their $D_{r}$ is determined solely by the intensity of the interactions, and predicts a continually changing orientation independent of applied deformation on the surrounding fluid. This implies the fiber orientation tends toward an isotropic distribution for no applied deformation. Unfortunately, this result violates the physical system being modeled where fiber motion is only seen to occur during velocity changes of the surrounding fluid.

Current approaches to model fiber orientation while incorporating fiber interaction effects are based on the Folgar and Tucker model [11]. Their model introduces interaction between fibers through forces similar to Brownian motion where each of
the fibers experience small forces as they collide within the suspension causing induced torques upon each ellipsoid. Folgar and Tucker proposed a diffusivity function based on the rate of deformation tensor $\dot{\gamma}_{i j}$ and an empirically derived parameter $C_{I}$ termed the interaction coefficient as

$$
\begin{equation*}
D_{r}=C_{I}\|\dot{\gamma}\| \tag{2.24}
\end{equation*}
$$

where $C_{I}$ is assumed to be a function of volume fraction, and $\|\dot{\gamma}\|$ is the scalar magnitude of the rate-of-deformation tensor, $\|\dot{\gamma}\|=\sqrt{\frac{1}{2} \dot{\gamma}_{i j} \dot{\gamma}_{j i}}$. Their model neglects the directional dependance of the collisions notwithstanding the suggestion by Folgar and Tucker that "...while it is possible, and even likely, that these orientation changes have a directional bias, or are different in nearly random and nearly aligned suspensions, we have chosen to ignore these features." The Folgar and Tucker model yields exceptional results compared to previous theories, and has been considered the standard throughout both the industrial and academic communities. After much simplification, the time evolution for the fiber orientation distribution function of Equation (2.23) can be expressed with the Folgar and Tucker model as [23,27]

$$
\begin{align*}
\frac{D \psi}{D t}(\theta, \phi, t) & =C_{I} \dot{\gamma} \frac{\partial^{2} \psi}{\partial \theta^{2}}+\frac{C_{I} \dot{\gamma}}{\sin ^{2} \theta} \frac{\partial^{2} \psi}{\partial \phi^{2}}+\frac{\partial \psi}{\partial \theta}\left(C_{I} \dot{\gamma} \frac{\cos \theta}{\sin \theta}-\frac{\lambda-1}{2} \boldsymbol{\kappa}^{T}: \boldsymbol{p} \boldsymbol{\theta}-\frac{\lambda+1}{2} \boldsymbol{\kappa}: \boldsymbol{p} \boldsymbol{\theta}\right) \\
& +\frac{1}{\sin \theta} \frac{\partial \psi}{\partial \phi}\left(\frac{\lambda-1}{2} \boldsymbol{\kappa}^{T}: \boldsymbol{p} \boldsymbol{\phi}-\frac{\lambda+1}{2} \boldsymbol{\kappa}: \boldsymbol{p} \boldsymbol{\phi}\right)+\psi(3 \lambda \boldsymbol{\kappa}: \boldsymbol{p} \boldsymbol{p}) \tag{2.25}
\end{align*}
$$

Computer simulations that numerically evaluate $\psi(\theta, \phi, t)$ for simple flows may require several weeks to several months of computational resources to reach steady state [23] and as thus are impractical to be considered for industrial simulations.

Alternatively, the orientation tensor approach of Advani and Tucker [6] represents the distribution function of fibers in a concise form alleviating the overwhelming computational burden encountered when solving Equation (2.25) numerically. The Folgar
and Tucker model is considered to be the benchmark for fiber orientation analysis during processing and has found wide acceptance in the literature [4, 6, 15, 19, 27-39]. Unlike the Kamal and Mutal model, the degree of alignment at steady state for the Folgar and Tucker model is a function of the strain rate and as such $\psi(\theta, \phi)$ evolves due to deformations in the surrounding fluid. The empirically determined interaction coefficient $C_{I}$ ensures that the final orientation state is accurately captured. Yamane et al. [40] curve fit the fiber interaction coefficient based upon volume fraction over a given set of flow conditions, and similarly Bay [23] along with Tucker and Advani [1] developed a formulation for the fiber interaction coefficient based on a different set of experimental data. These two forms assume the rate of fiber interactions is solely dependant on the volume fraction of fibers and yet yield a range of interaction coefficients with several orders of magnitude difference over the same range of fiber volume fractions. Phan-Thien et al. [2] discuss the discrepancy, and suggest the variation may be due to neglecting the fiber aspect ratio in the calculations.

Although the Folgar and Tucker model is used extensively, recently the Folgar and Tucker model has been questioned in its accuracy to model the orientation of fibers during the transient solution $[2,21,40-45]$. With recent industrial demands for efficient and accurate production design and advances in repeatable production processes, it is necessary to ensure that accurate models exist to represent the orientation state.

Koch [46] presents a model for orientational diffusion resulting from hydrodynamic fiber-fiber interactions, but has seen little use in the literature. The model proposed by Koch is a function of the fourth- and sixth-order orientation tensor and has two scalable parameters that are fit to calculations of orientational diffusion in pure extensional flows. The Koch model is presented without any derivation, and
provides no experimental results to validate the model. Since the proposed model claims to only be valid for shearing type flows and low concentrations of fibers, it is not considered further in this work.

Phan-Thien et al. [2,42] assume the rotary diffusion of the fiber distribution is an anisotropic second-order tensor

$$
\begin{equation*}
\mathbf{D}_{r}=\mathbf{C}\|\dot{\gamma}\| \tag{2.26}
\end{equation*}
$$

where bold capital letters designate a tensor. Their form assumes the diffusion follows a "white noise" random force behavior and determine the six independent components of the symmetric tensor $\mathbf{C}$ from experimental sampling data of the steady state solution. They relate their results back to those of the Folgar and Tucker model by assuming the interaction coefficient is related to the trace of $\mathbf{C}$ as

$$
\begin{equation*}
C_{I}=\frac{1}{3} \operatorname{Tr} \mathbf{C} \tag{2.27}
\end{equation*}
$$

Fan et al. [21,41] use Equation (2.27) along with the steady state experimental results of Mondy et al. [47] for suspensions of fibers in Couette flow, neglecting the transient solution and demonstrate the validity of their method at steady state. Phan-Thien and coauthors [2] numerically solve the evolution equations by taking discrete fibers in a reference cell undergoing simple shear. The reference cell experiences periodic boundary conditions, i.e. a fiber leaving the reference cell from one side enters the opposite side with identical exit behavior. Phan-Thien et al. give all results in terms of the interaction coefficient and develop a relation between interaction coefficient and volume fraction similar to that of Bay [23]. Unfortunately, Phan-Thien et al. [2] never discuss the transient results and the effect their method has on the transient solution. They conclude that fiber interactions may not be best described by a simple diffusion
process, and clearly the Folgar and Tucker model must be further investigated and improved for accurate simulations of fiber orientation.

Tucker et al. [44] hypothesize that fibers experience a local strain lower than the average strain through the thickness of a part. This leads to requiring an understanding of the fluid flow regions between layers of fibers [19, 20, 23] where Tucker and co-workers hypothesize the strain is "absorbed". They introduce a strain reduction factor, $S R F$, to the equation of continuity from Equation (2.23) as

$$
\begin{equation*}
\frac{D \psi}{D t}=\frac{1}{S R F}\left(-\nabla \cdot\left(\dot{\boldsymbol{p}}^{h} \psi(\boldsymbol{p})-\nabla\left(D_{r} \psi(\boldsymbol{p})\right)\right)\right) \tag{2.28}
\end{equation*}
$$

Although results from the model by Tucker and co-workers appears to yield adequate results in short plaque flow, the details of their model are not currently available in the literature and as such results developed in the following chapters cannot be compared to their formulation.

### 2.2.4 Equation of Motion for $a_{i j}$

Advani and Tucker [6] extend the usefulness of the Folgar and Tucker model by introducing orientation tensors which represent the fiber orientation state using moments of the fiber orientation distribution function. Orientation tensors provide a computationally efficient means for evaluating the orientation state, even in complex flow fields, and have seen extensive use in simulations of many complex phenomena to model the orientation distribution function in a concise manner suitable for large scale computations (e.g. crystalline polymers [48, 49], short-fiber polymer composites [6], crack fabric tensors [50], and turbulence models [51]). A disadvantage in the application of orientation tensors arises during the solution whereby the next higherordered orientation tensor is required for a complete representation. Therefore, to
solve the orientation tensor evolution equation given by Advani and Tucker, a closure is required where higher-ordered orientation tensors are approximated as functions of lower-ordered orientation tensors.

The definition of orientation tensors from Equation (2.11) is applied to the equation of motion given in Equation (2.25) for the fiber orientation distribution function to obtain the equation of motion of the orientation tensors. Advani [3] assumed the Folgar and Tucker model of diffusion, and thus several key components necessary for a directionally dependant diffusion model were not included in the derivation. A full derivation of the equation of motion is undertaken in Jack [45]. The most common application of fiber orientation tensor evolution is that of the second-order which has seen wide use for industrial problems.

The evolution equations for $a_{i j}$ obtained by first considering the second-order tensor $\mathbf{B}(\boldsymbol{p})$ defined in component form as $B_{i j}=p_{i} p_{j}$ for $i, j \in\{1,2,3\}$ is related to the second-order orientation tensor through integration as $\oint_{\mathbb{S}^{2}} \psi(\theta, \phi) B_{i j} \mathrm{~d} \mathbb{S}=$ $\oint_{\mathbb{S}^{2}} \psi(\theta, \phi) p_{i} p_{j} \mathrm{dS}=a_{i j}$. To derive the equation of change associated with the tensor $\mathbf{B}(\boldsymbol{p})$, post-multiply the equation of continuity with rotary diffusion given in Equation (2.23), with the $i, j$ component of the test function $\mathbf{B}(\boldsymbol{p})$, where the integral over the unit sphere $\mathbb{S}^{2}$ is given as

$$
\begin{equation*}
\oint_{\mathbb{S}^{2}} \frac{\mathrm{D} \psi}{\mathrm{D} t} B_{i j} \mathrm{~d} \mathbb{S}=-\oint_{\mathbb{S}^{2}} \nabla \cdot\left(\dot{\boldsymbol{p}}^{h} \psi-\nabla\left(D_{r} \psi\right)\right) B_{i j} \mathrm{~d} \mathbb{S} \tag{2.29}
\end{equation*}
$$

where it is assumed that $B_{i j}, \psi$ and $D_{r}$ each depend on $\boldsymbol{p}$ and are as continuous as necessary. The left-hand side of the equation is the desired form for the equation of motion for the second-order orientation tensor $a_{i j}$ which is seen as

$$
\begin{equation*}
\oint_{\mathbb{S}^{2}} \frac{\mathrm{D} \psi}{\mathrm{D} t} B_{i j} \mathrm{~d} \mathbb{S}=\frac{\mathrm{D}}{\mathrm{D} t} \oint_{\mathbb{S}^{2}} \psi B_{i j} \mathrm{~d} \mathbb{S}=\frac{\mathrm{D}}{\mathrm{D} t} \oint_{\mathbb{S}^{2}} \psi p_{i} p_{j} \mathrm{~d} \mathbb{S}=\frac{D a_{i j}}{D t} \tag{2.30}
\end{equation*}
$$

where the material derivative $\frac{D}{D t}$ can brought outside the integration since integration over the unit sphere is independent of time and the velocity gradients within the material derivative are assumed constant over the surface of integration.

The chain rule may be used to expand the right hand side of Equation (2.29) while assuming that the rotary diffusion $D_{r}$ is first-order differentiable within and on the unit sphere. Recognizing both terms within Equation (2.29) are integrable, the equation of motion for the second-order orientation tensor becomes

$$
\begin{equation*}
\frac{D a_{i j}}{D t}=\oint_{\mathbb{S}^{2}} \dot{\boldsymbol{p}}^{h} \psi \cdot \nabla\left(p_{i} p_{j}\right) \mathrm{d} \mathbb{S}-\oint_{\mathbb{S}^{2}} \nabla\left(D_{r} \psi\right) \cdot \nabla\left(p_{i} p_{j}\right) \mathrm{d} \mathbb{S} \tag{2.31}
\end{equation*}
$$

where the first component on the right-hand side represents the rate of change of motion due to hydrodynamic forces (as given by Jeffery's equation), and the second component represents effects due to rotary diffusion. Jeffery's component of the second-order orientation tensor is rewritten using the unit vector derivatives in Equation (2.7) along with the chain rule to yield the well known expression for the hydrodynamic effects on $a_{i j}$ as

$$
\begin{equation*}
\oint_{\mathbb{S}^{2}} \dot{\boldsymbol{p}}^{h} \psi \cdot \nabla\left(p_{i} p_{j}\right) \mathrm{d} \mathbb{S}=-\frac{1}{2} \omega_{i k} a_{k j}+\frac{1}{2} a_{i k} \omega_{k j}+\frac{1}{2} \lambda\left(\dot{\gamma}_{i k} a_{k j}+a_{i k} \dot{\gamma}_{k j}-2 \dot{\gamma}_{k l} a_{i j k l}\right) \tag{2.32}
\end{equation*}
$$

where fiber collisions have no effect. Observe that the final term in Equation (2.32) contains the fourth-order orientation tensor $a_{i j k l}$, therein introducing the classical closure dilemma. To alleviate the need for $a_{i j k l}$ this higher order orientation tensor is typically approximated as a function of a lower ordered orientation tensor. This problem is not unique to polymer composite flow, but also appears in crystalline polymer flow [48, 49], crack fabric tensors [50], and turbulent flow [51].

The integration associated with the rotary diffusion on the right hand side of Equation (2.31) can be rewritten using the chain rule for gradients. After much
simplification (see e.g. [45]) along with the Laplacian in spherical coordinates from Equation (2.10) and the Kronecker Delta of Equation (2.8), the second term on the right hand side of Equation (2.31) simplifies to

$$
\begin{equation*}
-\oint_{\mathbb{S}^{2}} \nabla\left(D_{r} \psi\right) \cdot \nabla\left(B_{i j}\right) \mathrm{d} \mathbb{S}=\oint_{\mathbb{S}^{2}} D_{r} \psi\left(2 \delta_{i j}-6 p_{i} p_{j}\right) \mathrm{d} \mathbb{S} \tag{2.33}
\end{equation*}
$$

The equation of motion for the second-order orientation tensor is readily obtained by combining Equations (2.32) and (2.33) with Equation (2.31) to obtain

$$
\begin{align*}
\frac{D a_{i j}}{D t}= & -\frac{1}{2} \omega_{i k} a_{k j}+\frac{1}{2} a_{i k} \omega_{k j}+\frac{1}{2} \lambda\left(\dot{\gamma}_{i k} a_{k j}+a_{i k} \dot{\gamma}_{k j}-2 \dot{\gamma}_{k l} a_{i j k l}\right) \\
& +\oint_{\mathbb{S}^{2}} D_{r} \psi\left(2 \delta_{i j}-6 p_{i} p_{j}\right) \mathrm{d} \mathbb{S} \tag{2.34}
\end{align*}
$$

Notice the equation of motion for $a_{i j}$ is, at the least, a function of the fourthorder orientation tensor $a_{i j k l}$. When rotary diffusivity $D_{r}$ has no directional bias, the integration is easily written in terms of the orientation tensors, i.e. $\oint_{\mathbb{S}^{2}} D_{r} \psi\left(2 \delta_{i j}-6 p_{i} p_{j}\right) \mathrm{d} \mathbb{S}=D_{r}\left(2 \delta_{i j}-6 a_{i j}\right)$. For the development of the fitted closures, the rotary diffusivity is assumed to be of the form proposed by Folgar and Tucker [11] given by $D_{r}=C_{I}\|\dot{\gamma}\|$ where $C_{I}$ is the empirically determined interaction coefficient, and $\|\dot{\gamma}\|$ is the scalar magnitude of the strain rate tensor discussed in Equation (2.21). The resulting equation of motion of the second-order orientation tensor with the Folgar and Tucker model for rotary diffusion is written concisely as

$$
\begin{equation*}
\frac{D a_{i j}}{D t}=-\frac{1}{2} \omega_{i k} a_{k j}+\frac{1}{2} a_{i k} \omega_{k j}+\frac{1}{2} \lambda\left(\dot{\gamma}_{i k} a_{k j}+a_{i k} \dot{\gamma}_{k j}-2 \dot{\gamma}_{k l} a_{i j k l}\right)+C_{I}\|\dot{\gamma}\|\left(2 \delta_{i j}-6 a_{i j}\right) \tag{2.35}
\end{equation*}
$$

The time required to compute the evolution of the second-order orientation tensor is significantly less than the computational time required to evolve $\psi(\theta, \phi, t)$ with Equation (2.25) once an appropriate method is selected to approximate the fourthorder orientation tensor $a_{i j k l}$.

### 2.2.5 Equation of Motion for $a_{i j k l}$

A similar derivation as that outlined above for the equation of motion of the secondorder orientation tensor can be performed for the equation of motion for the fourthorder orientation tensor. Assuming the Folgar and Tucker model for diffusion, the equation of motion for the fourth-order orientation tensor is expressed as

$$
\begin{align*}
\frac{D a_{i j k l}}{D t}= & -\frac{1}{2}\left(\omega_{i m} a_{m j k l}-a_{i j k m} \omega_{m l}+\omega_{j m} a_{i k l m}-a_{i j l m} \omega_{m k}\right) \\
& +\frac{1}{2} \lambda\left(\dot{\gamma}_{i m} a_{j k l m}+\dot{\gamma}_{j m} a_{i k l m}+\dot{\gamma}_{k m} a_{i j l m}+\dot{\gamma}_{l m} a_{i j k m}-4 \dot{\gamma}_{m n} a_{i j k l m n}\right) \\
& +C_{I} \dot{\gamma}\left[-20 a_{i j k l}+2\left(a_{i j} \delta_{k l}+a_{i k} \delta_{j l}+a_{i l} \delta_{j k}+a_{j k} \delta_{i l}+a_{j l} \delta_{i k}+a_{k l} \delta_{i j}\right)\right] \tag{2.36}
\end{align*}
$$

Note that Equation (2.36) is a modification of that proposed by Advani and Tucker [6] as it includes the correction with the rate of deformation and vorticity tensors as proposed by Altan et al. [52].

### 2.2.6 Orientation Closure Overview

Unfortunately, the evolution equation of any even-ordered orientation tensor requires the next higher even-ordered orientation tensor, i.e. the solution of $a_{i j}$ requires $a_{i j k l}$ in Equation (2.35) and the solution of $a_{i j k l}$ from Equation (2.36) requires $a_{i j k l m n}$. To avoid solving higher order tensor evolution equations, closure approximations are introduced to represent a higher order tensor in terms of lower ordered tensors. A fourth-order closure may be expressed as

$$
\begin{equation*}
a_{i j k l} \approx F_{i j k l}\left(a_{m n}\right) \tag{2.37}
\end{equation*}
$$

where $F_{i j k l}$ is a function of the second-order tensor $a_{i j}$. A closure of the sixth-order orientation tensor may be written in a similar manner as

$$
\begin{equation*}
a_{i j k l m n} \approx G_{i j k l m n}\left(a_{o p q r}\right) \tag{2.38}
\end{equation*}
$$

where $G_{i j k l m n}$ represents a function of the fourth-order orientation tensor.
The fourth-order quadratic closure $\tilde{a}_{i j k l}$ is the simplest of the fourth-order closures and is exact for highly aligned distributions [48]

$$
\begin{equation*}
\tilde{a}_{i j k l}=a_{i j} a_{k l} \tag{2.39}
\end{equation*}
$$

Although the quadratic closure is frequently used in conjunction with Advani and Tucker's hybrid closure [6], the quadratic closure does not obey the symmetry requirements for a fourth-order orientation tensor in Equation (2.12), i.e. $\tilde{a}_{i j k l} \neq$ $\tilde{a}_{i l k j} \forall i, j, k, l$. As a result, the hybrid closure, which contains the quadratic closure, also does not obey the symmetries of Equation (2.12). Even with that being known, the hybrid closure is still regularly used in many process simulations of shortfibers $[36,37]$ perhaps due to the simplicity of its evaluation. Note that even though the hybrid closure is exact for perfectly aligned orientations of fibers and completely random orientations of fibers, it overpredicts the actual alignment of fibers for all other alignment states.

Cintra and Tucker [27] introduced the eigenvalue based fourth-order fitted closure which provides a significant accuracy increase over previous analytic closures. There exist several modifications to the Cintra and Tucker closure [33, 35, 49, 53], but the fundamental basis remains unchanged. The natural closure of Verleye and Dupret [54] and Dupret et al. [55] is formed from the general expression given by Lipscomb et al. [14] for a fully symmetric fourth-order orientation tensor obtained from the Cayley-Hamilton theorem in terms of a second-order orientation tensor through a fitting process. The invariant based closure of Chung and Kwon [56] modifies the natural closure of Dupret et al. to account for a range of fiber volume fractions while removing the singularity issues found in the natural closure. Petty et al. [31]
introduced the fully symmetric closure, a further enhancement to the natural closure, and has been demonstrated to yield realizable behavior in Couette flow. Schache [57] along with Smith et al. [58] developed the fourth-order neural-network based closure, and demonstrated its effective use in simple flows of short-fibers. Recently, Jack [59] and Jack and Smith [60] demonstrated, through the use of distribution reconstruction functions, that current closures of the fourth-order orientation tensor represent fiber orientation distribution functions nearly as accurately as does the true fourth-order reconstruction of the distribution function. In other words, further research on fourth-order closures is expected to yield only minor improvements in accuracy over current methods. To obtain a significant increase in accuracy it becomes necessary to investigate closures of higher-order orientation tensors.

The sixth-order closures of Doi [48], Advani and Tucker [6] and Altan et al. [61] have been shown to over predict the alignment state, and tend to diverge from the actual solution except in alignment states tending toward either a random or a perfectly aligned state (see e.g. Jack and Smith [60]). The sixth-order closure of Doi as used by Altan et al. [61] is simply written as

$$
\begin{equation*}
\tilde{a}_{i j k l m n} \simeq a_{i j k l} a_{m n p p} \tag{2.40}
\end{equation*}
$$

and is exact for perfectly aligned orientations, but in general flow conditions it overestimates the actual alignment of fibers (see e.g. [60]). The sixth-order linear closure $\hat{a}_{i j k l m n}$ from Advani and Tucker [6] given as

$$
\begin{align*}
\hat{a}_{i j k l m n}= & \frac{1}{693}\left[\delta_{i j} \delta_{k l} \delta_{m n}+\delta_{i k} \delta_{j l} \delta_{m n}+\cdots(15 \text { total terms })\right] \\
& -\frac{1}{99}\left[a_{i j} \delta_{k l} \delta_{m n}+\delta_{i j} a_{k l} \delta_{m n}+\cdots(45 \text { total terms })\right] \\
& +\frac{1}{11}\left[a_{i j k l} \delta_{m n}+a_{i j m n} \delta_{k l}+\cdots(15 \text { total terms })\right] \tag{2.41}
\end{align*}
$$

produces the exact solution for uniformly random fiber orientation states. The sixthorder hybrid closure $\bar{a}_{i j k l m n}$ given by Advani [3] and Advani and Tucker [6] is created by forming an approximation that is more accurate over the range of possible orientations. The sixth-order hybrid closure simply combines the sixth-order linear and quadratic closures through a scalar measure of orientation $f$ as

$$
\begin{equation*}
\bar{a}_{i j k l m n} \simeq(1-f) \hat{a}_{i j k l m n}+f \tilde{a}_{i j k l m n} \tag{2.42}
\end{equation*}
$$

where $f$ is zero for randomly orientated fibers and unity for perfectly aligned fibers.
The use of higher-order closures has been avoided, perhaps due in part to the conclusion by Cintra and Tucker [27] that fiber orientation predictions using sixthorder closures are less accurate than those based on current closures of the fourthorder orientation tensor, despite requiring more computational efforts. Alternatively, Altan et al. [52] state that lower- (e.g. fourth-) order approximations may result in "... errors for complex flow fields, where both shear and elongational velocity gradients exist in three unique planes. Therefore, higher-order approximations may be required for the accurate description of suspension mechanics." To date, the investigation into higher order tensors, i.e. closures of the sixth-order or higher, has yet to yield acceptable results for injection molding processes of concentrated suspensions of fibers $[6,52,61-63]$.

In the following chapter, several new fitted sixth-order closures will be introduced. The sixth-order fitted invariant based closure $\mathrm{INV}_{6}$ of Jack and Smith [38] will be demonstrated to more accurately predict the second-order orientation tensor than simulations that employ existing fourth-order and sixth-order closures. Additionally, it will be shown that the sixth-order $\mathrm{INV}_{6}$ closure more accurately represents the distribution function of fibers than any of the current closure methods.

### 2.3 Material Stiffness Predictions

Fiber orientation predictions from polymer flow simulations are often used to compute the material properties of the finished composite part. Material properties such as mechanical strength and stiffness are strongly dependant on the orientation of the fibers in a molded part $[3,10,23,29,36,37,52,55,64-69]$. In general, the tensile strength and modulus are higher where significant fiber alignment occurs [3,10,66,70]. Several theories have been developed to compute the mechanical properties of a short fiber composite once fiber orientation is known. Although there is some disagreement in the current theories, they all use a basic two step procedure. The first step estimates the properties of the unidirectional short-fiber reinforced composite [6, 10, 66, 71-74] , and the second step averages the properties according to the fiber orientation distribution function $[3,23,66,71,75-79]$. The second step is the focus of several of the following chapters, and will be discussed later in detail.

The computation of unidirectional material properties such as Young's Modulus, Poisson's Ratio and Shear Modulus begins with Hooke's Law [70]

$$
\begin{equation*}
\varepsilon_{i j}=S_{i j k l} \sigma_{k l} \quad \sigma_{i j}=C_{i j k l} \varepsilon_{i j} \tag{2.43}
\end{equation*}
$$

where $\varepsilon_{i j}$ represents the strain tensor, $\sigma_{k l}$ represents the stress tensor, $S_{i j k l}$ is the fourth-order compliance tensor, and $C_{i j k l}$ is the fourth-order stiffness tensor. Note that the stiffness tensor, $C_{i j k l}$ is the inverse of the compliance tensor, $S_{i j k l}$.

Tucker and Liang [80] review and discuss micromechanics models that are commonly used to calculate the elasticity tensor for unidirectionally aligned short-fiber composites. Their review includes dilute suspensions where elastic properties are based on Eshelby's equivalent inclusions [81], the Mori-Tanaka model [82], the HalpinTsai equations [70], the bound interpolation model of Lielens [83], and several others.

All of the models included in [80] are written in terms of the volume fraction of fibers $V_{f}$, Young's modulus and Poisson's ratio of the fiber, $E_{f}$ and $\nu_{f}$, respectively, Young's modulus and Poisson's ratio of the matrix, $E_{m}$ and $\nu_{m}$, respectively, and the fiber aspect ratio, $a_{r}$. Each of the various stiffness predictions investigated by Tucker and Liang yield five independent properties for perfectly aligned fibers along the $x_{1}$ axis. These properties are typically chosen to be $E_{1}, E_{2}, G_{12}, G_{23}$ and $\nu_{12}$ which are, respectively, Young's modulus in the $x_{1}$ direction, Young's modulus in the $x_{2}$ direction, shear modulus in the $x_{1}-x_{2}$ plane, shear modulus in the $x_{2}-x_{3}$ plane, and Poisson's ratio on the $x_{1}$ face in the $x_{2}$ direction. Tucker and Liang concluded that the Halpin-Tsai equations provide reasonable estimates for stiffness, with the best predictions coming from the Mori-Tanaka model and the bound interpolation model of Lielens. In this thesis, the Halpin-Tsai equations are employed to obtain the unidirectional material properties for simplicity in computations, but the reader is encouraged to consider either the Mori-Tanaka model or the Lielens method in actual part simulations. The Halpin-Tsai equations have seen extensive use in fiber-reinforced composites with isotropic fibers (see e.g. [6, 7, 66, 71]), and have been experimentally shown to be valid for volume fractions up to $70 \%$.

For a unidirectional fiber distribution aligned along the $x_{1}$ axis, the stiffness tensor $C_{i j k l}$ may be written as the $6 \times 6$ matrix $[\bar{C}]$ in contracted notation as a function of the five previously discussed material constants as $[9,10,84]$

$$
[C]=\left[\begin{array}{cccccc}
\frac{1}{E_{1}} & \frac{-\nu_{12}}{E_{1}} & \frac{-\nu_{12}}{E_{1}} & 0 & 0 & 0  \tag{2.44}\\
\frac{-\nu_{12}}{E_{1}} & \frac{1}{E_{2}} & \frac{-\nu_{23}}{E_{2}} & 0 & 0 & 0 \\
\frac{-\nu_{12}}{E_{1}} & \frac{-\nu_{23}}{E_{2}} & \frac{1}{E_{2}} & 0 & 0 & 0 \\
0 & 0 & 0 & \frac{1}{G_{23}} & 0 & 0 \\
0 & 0 & 0 & 0 & \frac{1}{G_{12}} & 0 \\
0 & 0 & 0 & 0 & 0 & \frac{1}{G_{12}}
\end{array}\right]^{-1}
$$

where $\nu_{23}$ is substituted in place of $\frac{E_{2}}{2 G_{23}}-1$ for simplicity. In Equation (2.44), and
throughout this paper, contracted notation is used to relate material property matrices (i.e., $[C]$ ) to material tensors (i.e., $C_{i j k l}$ ) in the usual manner. For example, contracted notation replaces indices of 11,22 , and 33 with 1,2 and 3 respectively, indices of 23 and 32 are replaced by 4 , indices of 13 and 31 are replaced by 5 , and indices of 12 and 21 are replaced by 6, i.e., $C_{2213}$ can be written as $C_{25}$ in contracted notation. The material stiffness tensor given in Equation (2.44) may be used to represent the corresponding stiffness tensor for the unidirectional composite with the material parameters, $E_{1}, E_{2}, E_{3}$, etc. of Equation (2.44) being determined from the Halpin-Tsai equations listed in Table 2.1, where the shear modulus $G$ and bulk modulus $K$ for the matrix (or fiber) are related to Young's Modulus and Poisson's Ratio of the matrix (or fiber) by $[9,10,84,85]$

$$
\begin{equation*}
G=\frac{E}{2(1+\nu)} \quad K=\frac{E}{3(1-2 \nu)} \tag{2.45}
\end{equation*}
$$

Wright [66] and Advani [3] provide expressions for computing unidirectional material properties from the orientation tensors of Equation (2.11). They consider a tensor property $\mathbf{T}(\boldsymbol{p})$ that is associated with the unidirectional microstructure aligned along the direction of $\boldsymbol{p}$. If $\mathbf{T}$ is assumed to be transversely isotropic with $\boldsymbol{p}$ as the axis of symmetry, then the orientation average of the tensor $\mathbf{T}$ is given as

$$
\begin{equation*}
\langle\mathbf{T}\rangle=\oint_{\mathbb{S}^{2}} \mathbf{T}(\boldsymbol{p}) \psi(\boldsymbol{p}) \mathrm{d} \mathbb{S} \tag{2.46}
\end{equation*}
$$

Both Wright [66] and Advani [3] state that a fourth-order property tensor that has the symmetries of a transversely isotropic tensor must take the following form

$$
\begin{align*}
T_{i j k l}(\boldsymbol{p}) & =B_{1}\left(p_{i} p_{j} p_{k} p_{l}\right)+B_{2}\left(p_{i} p_{j} \delta_{k l}+p_{k} p_{l} \delta_{i j}\right) \\
& +B_{3}\left(p_{i} p_{k} \delta_{j l}+p_{i} p_{l} \delta_{j k}+p_{j} p_{l} \delta_{i k}+p_{j} p_{k} \delta_{i l}\right)+B_{4}\left(\delta_{i j} \delta_{k l}\right)+B_{5}\left(\delta_{i k} \delta_{j l}+\delta_{i l} \delta_{j k}\right) \tag{2.47}
\end{align*}
$$

Table 2.1: Halpin-Tsai Equations for a unidirectional composite.

| Material Parameter | $\zeta$ | $\eta$ |
| :---: | :---: | :---: |
| $E_{1}=E_{m}\left(\frac{1+\zeta \eta V_{f}}{1-\eta V_{f}}\right)$ | $\zeta=2 a_{r}$ | $\eta=\frac{E_{f} / E_{m}-1}{E_{f} / E_{m}+\zeta}$ |
| $E_{2}=E_{m}\left(\frac{1+2 \eta V_{f}}{1-\eta V_{f}}\right)$ | $\eta=\frac{E_{f} / E_{m}-1}{E_{f} / E_{m}+2}$ |  |
| $E_{3}=E_{2}$ |  |  |
| $\nu_{12}=\nu_{f} V_{f}+\nu_{m}\left(1-V_{f}\right)$ |  |  |
| $G_{12}=G_{m}\left(\frac{1+\zeta \eta V_{f}}{1-\eta V_{f}}\right)$ | $\zeta=1+40 V_{f}^{10}$ | $\eta=\frac{G_{f} / G_{m}-1}{G_{f} / G_{m}+\zeta}$ |
| $G_{13}=G_{12}$ |  |  |
| $G_{23}=G_{m}\left(\frac{1+\zeta \eta V_{f}}{1-\eta V_{f}}\right)$ | $\zeta=\frac{K_{m} / G_{m}}{K_{m} / G_{m}+2}$ | $\eta=\frac{G_{f} / G_{m}-1}{G_{f} / G_{m}+\zeta}$ |
| $\nu_{23}=-1+\frac{E_{2}}{2 G_{23}}$ |  |  |

where the $B_{m}, m \in\{1,2, \ldots, 5\}$ are scalar constants estimated from independent components of the associated transversely isotropic elasticity tensor. From Equation (2.11) and Equation (2.47), fourth-order tensor properties may be obtained as [3, 66]

$$
\begin{align*}
\left\langle T_{i j k l}\right\rangle= & B_{1}\left(a_{i j k l}\right)+B_{2}\left(a_{i j} \delta_{k l}+a_{k l} \delta_{i j}\right)+B_{3}\left(a_{i k} \delta_{j l}+a_{i l} \delta_{j k}+a_{j l} \delta_{i k}+a_{j k} \delta_{i l}\right) \\
& +B_{4}\left(\delta_{i j} \delta_{k l}\right)+B_{5}\left(\delta_{i k} \delta_{j l}+\delta_{i l} \delta_{j k}\right) \tag{2.48}
\end{align*}
$$

Recall that the second-order orientation tensor may be written solely in terms of the fourth-order orientation tensor using Equation (2.15). Therefore, the orientation average of the fourth-order tensor is determined by the fourth-order orientation tensor $a_{i j k l}$ and the underlying unidirectional tensor $\delta_{i j}$. The relationship between the five coefficients $B_{i}$ and the five independent components of the stiffness tensor of Equation

$$
\left\{\begin{array}{c}
B_{1}  \tag{2.44}\\
B_{2} \\
B_{3} \\
B_{4} \\
B_{5}
\end{array}\right\}=\left\{\begin{array}{c}
C_{11}+C_{22}-2 C_{12}-4 C_{66} \\
C_{12}-C_{23} \\
C_{66}+\frac{1}{2}\left(C_{23}-C_{22}\right) \\
C_{23} \\
\frac{1}{2}\left(C_{22}-C_{23}\right)
\end{array}\right\}
$$

Currently in the literature, there does not exist a method for computing the material properties from the distribution function of fibers $\psi(\theta, \phi)$. Additionally, the calculations in Equations (2.48) provide the orientation average of a transversely isotropic stiffness tensor but do not address property variation. Recognizing the statistical nature of fiber orientations within the composite, commonly defined through the fiber orientation probability distribution function, an analytical function will be developed in later chapters to predict both the expectation and variance of the material stiffness tensor from the probability distribution function of fibers.

### 2.4 Spherical Harmonic Reconstruction

Reconstructions of the fiber orientation probability distribution function provide a quantitative means for assessing the effect of using closure approximations when representing $\psi(\theta, \phi)[6,60]$. Onat $[86,87]$ provided a reconstruction of the distribution function $\psi(\theta, \phi)$ as

$$
\begin{equation*}
\psi(\theta, \phi)=f_{0}(\theta, \phi) V_{0}+f_{i j}(\theta, \phi) V_{i j}+f_{i j k l}(\theta, \phi) V_{i j k l}+\cdots \tag{2.50}
\end{equation*}
$$

where $f_{0}(\theta, \phi), f_{i j}(\theta, \phi), f_{i j k l}(\theta, \phi), \cdots$ are referred to as the orthogonal Fourier basis functions and $V_{0}, V_{i j}, V_{i j k l}, \cdots$ are labeled the Fourier coefficients with $i, j, k, l, \ldots \in$ $\{1,2,3\}$. Onat's reconstruction has been used to describe fiber distributions under rigid body rotations of a material, but little information is given as to expansions beyond sixth-order. When Fourier basis reconstructions up to the sixth-order are
used, Equation (2.50) can be written exclusively in terms of the orientation tensors (see e.g. $[86,87]$ for the second- and fourth-order expansion, and [60] for the extension to the sixth-order). Unfortunately, expressions for expansions of eighth-order and higher were not included in Onat's original and subsequent works. To the best of our knowledge, these higher-order terms have yet to be defined and are instead described in terms of the conditions that the higher order expansions must satisfy.

To facilitate a derivation for the statistical behavior of the material stiffness tensor, it is necessary to develop a general expression for the tensors appearing in Equation (2.50). It has been shown by Gelfand [88] that any arbitrary real function defined on $\mathbb{S}^{2}$ expanded about a fixed point, may be resolved into irreducible representations forming a complete orthogonal system, which is often called a Laplace series or a generalized Fourier series. In general, the Laplace series states that any real function (such as the fiber orientation distribution function $\psi(\theta, \phi)$ ) where the square of the modulus is integrable over the surface of the sphere (i.e., $\oint_{\mathbb{S}^{2}}|\psi(\theta, \phi)|^{2} \mathrm{~d} \mathbb{S}$ exists and is finite), may be expanded as a series of complex spherical harmonics. The Laplace series for $\psi(\theta, \phi)$ is thus written as

$$
\begin{equation*}
\psi(\theta, \phi)=\sum_{l=0}^{\infty} \alpha_{l}(\theta, \phi) \tag{2.51}
\end{equation*}
$$

where each $\alpha_{l}(\theta, \phi)$ is within the invariant subspace for an irreducible representation [88], i.e., there is no subspace of $\mathbb{S}^{2}$ in which the representation $\alpha_{l}(\theta, \phi)$ remains invariant. Each of these invariant subspaces are formed by the given order spherical functions as

$$
\begin{equation*}
\alpha_{l}(\theta, \phi)=\sum_{m=-l}^{l} C_{l}^{m} Y_{l}^{m}(\theta, \phi) \tag{2.52}
\end{equation*}
$$

where the set $\left\{Y_{l}^{m}(\theta, \phi):|m| \in \mathbb{N}, l \in \mathbb{N},|m| \leq l\right\}$ form an orthogonal system on the unit sphere, $\mathbb{N}$ is the set of integer numbers $\mathbb{N}=\{1,2, \ldots, N\}$, and $Y_{l}^{m}(\theta, \phi)$ are the
$l^{\text {th }}$ order complex spherical harmonic functions given as

$$
\begin{equation*}
Y_{l}^{m}(\theta, \phi)=\frac{1}{\sqrt{4 \pi}} \mathrm{e}^{\mathrm{i} m \phi} P_{l}^{m}(\cos \theta) \tag{2.53}
\end{equation*}
$$

where i is the imaginary unit equal to the square root of -1 , and the functions $P_{l}^{m}(\cos \theta)$ are the associated Legendre polynomial solutions to the Legendre differential equation given as

$$
\begin{equation*}
P_{l}^{m}(\mu)=(-1)^{m} \sqrt{\frac{2 l+1}{2} \frac{(l-m)!}{(l+m)!}} \frac{1}{2^{l} \cdot l!}\left(1-\mu^{2}\right)^{m / 2} \frac{\mathrm{~d}^{m+l}\left(\mu^{2}-1\right)^{l}}{\mathrm{~d} \mu^{m+l}} \tag{2.54}
\end{equation*}
$$

It can be shown that the spherical harmonics satisfy the orthogonality condition [88],

$$
\begin{equation*}
\oint_{\mathbb{S}^{2}} Y_{l}^{m}(\theta, \phi) \bar{Y}_{l^{\prime}}^{m^{\prime}}(\theta, \phi) \mathrm{d} \mathbb{S}=\delta_{m m^{\prime}} \delta_{l l^{\prime}} \tag{2.55}
\end{equation*}
$$

for all $\left\{m, m^{\prime}, l, l^{\prime}:|m|,\left|m^{\prime}\right|, l, l^{\prime} \in \mathbb{N},|m| \leq l,\left|m^{\prime}\right| \leq l^{\prime}\right\}$ and $\delta_{m m^{\prime}}$ is the Kronecker delta such that $\delta_{m m^{\prime}}=0, \forall m \neq m^{\prime}$, otherwise $\delta_{m m^{\prime}}=1$. The process to determine the coefficients $C_{l}^{m}$ from Equation (2.52) is similar to that of determining the coefficients in a Fourier Series. Multiplying the function $g(\theta, \phi)$ with $\bar{Y}_{l^{\prime}}^{m^{\prime}}(\theta, \phi)$, the complex conjugate of $Y_{l^{\prime}}^{m^{\prime}}(\theta, \phi)$, and integrating over the unit sphere, the coefficient $C_{l}^{m}$ is determined with the orthogonality condition from Equation (2.55) as

$$
\begin{align*}
& \oint_{\mathbb{S}^{2}} \psi(\theta, \phi) \bar{Y}_{l^{\prime}}^{m^{\prime}}(\theta, \phi) \mathrm{d} \mathbb{S}=\oint_{\mathbb{S}^{2}}\left(\sum_{l=0}^{\infty} \sum_{m=-l}^{l} C_{l}^{m} Y_{l}^{m}(\theta, \phi)\right) \bar{Y}_{l^{\prime}}^{m^{\prime}}(\theta, \phi) \mathrm{d} \mathbb{S} \\
& =\sum_{l=0}^{\infty} \sum_{m=-l}^{l} C_{l}^{m} \oint_{\mathbb{S}^{2}} Y_{l}^{m}(\theta, \phi) \bar{Y}_{l^{\prime}}^{m^{\prime}}(\theta, \phi) \mathrm{d} \mathbb{S}=\sum_{l=0}^{\infty} \sum_{m=-l}^{l} C_{l}^{m} \delta_{m m^{\prime}} \delta_{l l^{\prime}}=C_{l}^{m} \tag{2.56}
\end{align*}
$$

The form of Equation (2.52) along with the integral definition of complex variables given in Equation (2.56) is somewhat cumbersome, and is simplified as

$$
\begin{equation*}
\alpha_{l}(\theta, \phi)=\sum_{m=0}^{l} \beta_{l}^{m}(\theta, \phi) \tag{2.57}
\end{equation*}
$$

where the function $\beta_{l}(\theta, \phi)$ is defined for $0 \leq m \leq l$ as

$$
\begin{equation*}
\beta_{l}^{m}(\theta, \phi) \equiv\left(1-\frac{1}{2} \delta_{m 0}\right)\left(C_{l}^{m} Y_{l}^{m}(\theta, \phi)+C_{l}^{-m} Y_{l}^{-m}(\theta, \phi)\right) \tag{2.58}
\end{equation*}
$$

such that when $m=0, \beta_{l}^{-0}(\theta, \phi)=\beta_{l}^{0}(\theta, \phi)$. Using the Euler identities, the functions $\beta_{l}^{m}(\theta, \phi)$ are simplified as

$$
\begin{align*}
\beta_{l}^{m}(\theta, \phi)=\left(1-\frac{1}{2} \delta_{m 0}\right) & \left(\frac{1}{\pi} P_{l}^{m}(\cos \theta) \cos (m \phi) \oint_{\mathbb{S}^{2}} \psi(\theta, \phi) P_{l}^{m}(\cos \theta) \cos (m \phi) d \mathbb{S}\right. \\
& \left.+\frac{1}{\pi} P_{l}^{m}(\cos \theta) \sin (m \phi) \oint_{\mathbb{S}^{2}} \psi(\theta, \phi) P_{l}^{m}(\cos \theta) \sin (m \phi) d \mathbb{S}\right) \tag{2.59}
\end{align*}
$$

It then follows for example that $\alpha_{0}(\theta, \phi)$ can be written succinctly as

$$
\begin{align*}
\alpha_{0}(\theta, \phi) & =\beta_{0}^{0}(\theta, \phi)=\left(1-\frac{1}{2} \delta_{00}\right) \frac{1}{\pi} P_{0}^{0}(\cos \theta) \oint_{\mathbb{S}^{2}} \psi(\theta, \phi) P_{0}^{0}(\cos \theta) \mathrm{d} \mathbb{S} \\
& =\frac{1}{2} \frac{1}{\pi} \frac{1}{\sqrt{2}} \oint_{\mathbb{S}^{2}} \psi(\theta, \phi) \frac{1}{\sqrt{2}} \mathrm{~d} \mathbb{S}=\frac{1}{4 \pi} \oint_{\mathbb{S}^{2}} \psi(\theta, \phi) \mathrm{d} \mathbb{S}=\frac{1}{4 \pi} \tag{2.60}
\end{align*}
$$

where it is recognized that the integral of the distribution function over all possible orientations is equal to one, i.e. $\oint_{\mathbb{S}^{2}} \psi(\theta, \phi) \mathrm{d} \mathbb{S}=1$ (see Equation (2.5)). Observe, the above expression is the same as the zeroth-order term from Onat's reconstruction $[86,87]$ where $V_{0} f_{0}(\theta, \phi)=\frac{1}{4 \pi}$.

The second-order spherical function $\alpha_{2}(\theta, \phi)$ is the summation of the three terms, $\alpha_{2}(\theta, \phi)=\beta_{0}^{2}(\theta, \phi)+\beta_{1}^{2}(\theta, \phi)+\beta_{2}^{2}(\theta, \phi)$ which can be derived using Equation (2.59), and after some simplification

$$
\begin{align*}
& \beta_{2}^{0}(\theta, \phi)=\frac{5}{32 \pi}(1+3 \cos (2 \theta))\left(3 a_{33}-1\right)  \tag{2.61}\\
& \beta_{2}^{1}(\theta, \phi)=\frac{15}{4 \pi} \cos \theta \sin \theta\left(a_{13} \cos \phi+a_{23} \sin \phi\right)  \tag{2.62}\\
& \beta_{2}^{2}(\theta, \phi)=\frac{15}{16 \pi} \sin ^{2} \theta\left(\left(a_{11}-a_{22}\right) \cos (2 \phi)+2 a_{12} \sin (2 \phi)\right) \tag{2.63}
\end{align*}
$$

where Equation (2.11) is used to replace $\oint_{\mathbb{S}^{2}} p_{i}(\theta, \phi) p_{j}(\theta, \phi) \psi(\theta, \phi) \mathrm{d} \mathbb{S}$ with $a_{i j}$, the second-order orientation tensor. Summing the terms from Equations (2.61)-(2.63)
provides the form for the spherical function $\alpha_{2}(\theta, \phi)$. A careful comparison shows that $\alpha_{2}(\theta, \phi)$ is identical to the second-order reconstruction function used by Onat for $V_{i j} f_{i j}(\theta, \phi)$ and may be written as

$$
\begin{equation*}
\alpha_{2}(\theta, \phi)=\frac{15}{8 \pi}\left(p_{i}(\theta, \phi) p_{j}(\theta, \phi)-\frac{1}{3} \delta_{i j}\right)\left(a_{i j}-\frac{1}{3} \delta_{i j}\right) \tag{2.64}
\end{equation*}
$$

Similarly, the fourth-order function $\alpha_{4}(\theta, \phi)$ can be constructed with Equation (2.59), and is identical to the fourth-order Onat reconstruction function, $V_{i j k l} f_{i j k l}(\theta, \phi)[86,87]$ with each function $\beta_{4}^{m}(\theta, \phi), m \in\{0,1,2,3,4\}$, given as

$$
\begin{align*}
\beta_{4}^{0}(\theta, \phi)= & \frac{9}{2048 \pi}(9+20 \cos (2 \theta)+35 \cos (4 \theta))\left(3-30 a_{33}+35 a_{3333}\right)  \tag{2.65}\\
\beta_{4}^{1}(\theta, \phi)= & \frac{45}{128 \pi}(9 \cos \theta+7 \cos (3 \theta)) \sin \theta\left(\cos \phi\left(7 a_{1333}-3 a_{13}\right)\right. \\
& \left.+\sin \phi\left(7 a_{2333}-3 a_{23}\right)\right)  \tag{2.66}\\
\beta_{4}^{2}(\theta, \phi)= & \frac{45}{128 \pi}(5+7 \cos (2 \theta)) \sin ^{2} \theta\left(\cos (2 \phi)\left(7\left(a_{1133}-a_{2233}\right)-\left(a_{11}-a_{22}\right)\right)\right. \\
& \left.+\sin (2 \phi)\left(14 a_{1233}-2 a_{12}\right)\right)  \tag{2.67}\\
\beta_{4}^{3}(\theta, \phi)= & \frac{315}{32 \pi} \cos \theta \sin ^{3} \theta\left(\cos (3 \phi)\left(4 a_{1113}-3 a_{13}+3 a_{1333}\right)\right. \\
& \left.+\sin (3 \phi)\left(3 a_{1123}-a_{2223}\right)\right)  \tag{2.68}\\
\beta_{4}^{4}(\theta, \phi)= & \frac{315}{256 \pi} \sin ^{4} \theta\left(\cos (4 \theta)\left(8 a_{1111}-8 a_{11}+8 a_{1133}+1-2 a_{33}+a_{3333}\right)\right. \\
& \left.+\sin (4 \phi)\left(8 a_{1112}-4 a_{12}+4 a_{1233}\right)\right) \tag{2.69}
\end{align*}
$$

Note that when $l$ is odd, the functions $\alpha_{l}(\theta, \phi)$ are zero since the fiber orientation distribution function is symmetric, $\psi(\boldsymbol{p})=\psi(-\boldsymbol{p})$. By recognizing that $f_{0}(\theta, \phi) V_{0}=\alpha_{0}(\theta, \phi), f_{i j}(\theta, \phi) V_{i j}=\alpha_{2}(\theta, \phi)$, and $f_{i j k l}(\theta, \phi) V_{i j k l}=\alpha_{4}(\theta, \phi)$, and setting $f_{i j \cdots 2 N}(\theta, \phi) V_{i j \cdots 2 N}=\alpha_{2 N}(\theta, \phi)$ for $\{N: N \in \mathbb{N}, N \geq 3\}$ the full reconstruction of Onat can be realized succinctly as that of Equation (2.51) or (2.57). Although the notation of the spherical harmonic expansion does not lend itself to the succinct
notation of Onat's expansion which allows easy rotations, it does provide a means for constructing an expansion up to a desired order. Additionally, it is essential in the derivation given in Chapter 5 to fully understand the construction for all terms in the expansion and not just the first few.

An $n^{\text {th }}$ order approximate reconstruction of the fiber orientation distribution can be given as

$$
\begin{equation*}
\hat{\psi}_{n}(\theta, \phi)=\sum_{l=0}^{n} \alpha_{l}(\theta, \phi) \tag{2.70}
\end{equation*}
$$

where $n \in\{0\} \cup \mathbb{N}$, and for each $\{l: l=2 m+1, m \in \mathbb{N}\}$ the function $\alpha_{l}(\theta, \phi)$ is zero. To asses the error between the distribution function $\psi(\theta, \phi)$ and the $n^{\text {th }}$ order reconstruction of the distribution function $\hat{\psi}_{n}$, the following error metric is employed (see e.g. [59,60] for a full discussion)

$$
\begin{equation*}
E R R_{n}=\sqrt{\oint_{\mathbb{S}^{2}}\left(\psi\left(\theta, \phi, t_{o}\right)-\hat{\psi}_{n}\left(\theta, \phi, t_{o}\right)\right)^{2} \mathrm{dS}} \tag{2.71}
\end{equation*}
$$

where the integration is performed over the unit sphere at the time $t_{o}$. Equation (2.71) may be used to form an $n^{\text {th }}$ order truncation limit from the exact $n^{\text {th }}$ order orientation tensors in Equation (2.11). To form the truncation limit for a particular flow field it is necessary to first numerically evaluate the exact distribution function $\psi(\theta, \phi, t)$. The orientation tensors are then computed from $\psi(\theta, \phi, t)$ with Equation (2.11) and the distribution function is reconstructed to the desired order with Equation (2.70). This reconstructed distribution function $\hat{\psi}_{n}(\theta, \phi, t)$ is then used with the exact distribution function $\psi(\theta, \phi, t)$ in Equation (2.71) to form $E R R_{n}$. When the exact orientation tensors $a_{i j}, a_{i j k l}, \ldots$ computed using Equations (2.25) and (2.11) are used in Equation (2.71) a truncation limit of the reconstruction is obtained. Alternatively, when the closures discussed above and in the following chapters are employed
to compute any of the orientation tensors that compose the Laplace series reconstruction in Equation (2.70), Equation (2.71) can be used to indicate the error introduced by the closure. Note that any $n^{\text {th }}$ order closure of an orientation tensor can only be as accurate in representing the distribution function as the exact $n^{\text {th }}$ order reconstruction. As discussed in Jack and Smith [60] existing fourth-order closures approach the fourth-order reconstruction limit, therefore any significant increase in the distribution function representation is expected to require a sixth- or higher-order representation.

## CHAPTER 3

## SIXTH-ORDER CLOSURE METHODS FOR MODELING SHORT-FIBER SUSPENSIONS

Orientation tensors experience widespread use in short-fiber reinforced injection molding simulations of industrial polymer composite products. As described in Chapter 2 , the evolution equation for each even-order orientation tensor is written in terms of the next higher even-order orientation tensor necessitating the use of a closure. It has been shown that current fourth-order closures approach the fourth-order truncation limit when representing the fiber orientation distribution function (see, e.g. Jack and Smith [60]), ergo an increase in accuracy necessitates the development of a robust sixth-order closure. The following chapter outlines three fitted sixth-order closures. The two preliminary sixth-order fitted closures presented in Jack and Smith [39], are expressed in terms of the second-order orientation tensor where it is assumed that the orthogonal planes of material symmetry of the sixth-order orientation tensor are defined by the principal directions of the second-order orientation tensor. The sixthorder tensor components are fit to either eigenvalues or invariants of the second-order orientation tensor over numerous flow conditions and interaction coefficients. The results from the two preliminary models illustrate the effect that sixth-order closures may have in surpassing accuracy limits over the existing fourth-order closures, but fail to provide the desired effects throughout the range of flows investigated. The final investigated closure is that of the sixth-order invariant based fitted closure $\mathrm{INV}_{6}$ appearing in Jack and Smith [38]. The $\mathrm{INV}_{6}$ is formed from a general expression for a fully symmetric sixth-order tensor written as a function of a fourth-order orientation tensor. The components of this sixth-order closure are fit to a linear polynomial
of the fourth-order orientation tensor invariants whose coefficients are computed by fitting the sixth-order components obtained from the closure to those computed from distribution function simulations obtained with the Folgar and Tucker model for fiber interaction [11] from a variety of flow fields and fiber interaction coefficients. The sixth-order $\mathrm{INV}_{6}$ closure is shown to more accurately predict the second-order orientation tensor than simulations that employ existing fourth-order and the preliminary sixth-order closures. Additionally, it is shown that the sixth-order $\mathrm{INV}_{6}$ closure more accurately represents the fiber orientation probability distribution function than any current closure method.

### 3.1 Sixth-Order Fitted Closures from $a_{i j}$

The first two sixth-order fitted closures considered here demonstrate accuracy improvements in the representation of the fiber orientation distribution function that may be attained through higher-order closures. These two new closures identified here as $\mathrm{EBF}_{6}$ and $\mathrm{IBF}_{6}$, appear in Jack and Smith [39]. The closures provide a means for computing $a_{i j k l m n}$ from $a_{i j}$, but have been shown to diverge into non-physical orientation states under some flow conditions.

### 3.1.1 Eigenvalue Based Sixth-order Fitted Closure

The eigenvalue-based sixth-order fitted closure $\left(\mathrm{EBF}_{6}\right)$ assumes (1) that the principal directions of the second-order orientation tensor correspond with the planes of orthogonal symmetry of the sixth-order orientation tensor, and (2) that each principal component of the sixth-order orientation tensor is a function of the independent principal components of the second-order orientation tensor and the principal components of the fourth-order orientation tensor.

The sixth-order orientation tensor has 729 components, 28 of which are independent due to the symmetries in Equation (2.12). If it is assumed that the principal frame of the second-order orientation tensor $a_{i j}$ forms the planes of orthogonal symmetry for both the fourth-order orientation tensor $a_{i j k l}$ and the sixth-order orientation tensor $a_{i j k l m n}$, then only ten nonzero components of $a_{i j k l m n}$ remain

| $a_{(111111)}$ | $a_{(111122)}$ | $a_{(111133)}$ | $a_{(112222)}$ | $a_{(112233)}$ |
| :--- | :--- | :--- | :--- | :--- |
| $a_{(113333)}$ | $a_{(222222)}$ | $a_{(222233)}$ | $a_{(223333)}$ | $a_{(333333)}$ |

where $(\cdots)$ indicates that components of $a_{i j k l m n}$ are given in the principal reference frame of $a_{i j}$. Using Equation (2.13) for the relation between $a_{i j}, a_{i j k l}$ and $a_{i j k l m n}$, the number of independent components in $a_{(i j k l m n)}$ reduces further from those listed in Equation (3.1) to just four (see e.g. [59]). The $\mathrm{EBF}_{6}$ closure is formed by arbitrarily selecting the four unknown independent components to be $a_{(111111)}, a_{(111122)}, a_{(222222)}$, and $a_{(333333)}$.

The second-order orientation tensor $a_{i j}$ has three eigenvalues, $a_{(1)}, a_{(2)}$, and $a_{(3)}$. From Equation (2.13) the trace of $a_{i j}$ is 1 , and selecting $a_{(1)} \geq a_{(2)} \geq a_{(3)}$ there remain only two independent principal values of the second-order tensor, $a_{(1)}$ and $a_{(2)}$. The four independent components from Equation (3.1) are found by fitting them to a second-order polynomial of the independent eigenvalues of the second-order orientation tensor

$$
\left[\begin{array}{c}
a_{(111111)}  \tag{3.2}\\
a_{(111122)} \\
a_{(222222)} \\
a_{(333333)}
\end{array}\right]=\left[\begin{array}{llllll}
\alpha_{11} & \alpha_{12} & \alpha_{13} & \alpha_{14} & \alpha_{15} & \alpha_{16} \\
\alpha_{21} & \alpha_{22} & \alpha_{23} & \alpha_{24} & \alpha_{25} & \alpha_{26} \\
\alpha_{31} & \alpha_{32} & \alpha_{33} & \alpha_{34} & \alpha_{35} & \alpha_{36} \\
\alpha_{41} & \alpha_{42} & \alpha_{43} & \alpha_{44} & \alpha_{45} & \alpha_{46}
\end{array}\right]\left\{\begin{array}{c}
1 \\
a_{(1)} \\
a_{(2)} \\
a_{(1)} a_{(2)} \\
a_{(1)}^{2} \\
a_{(2)}^{2}
\end{array}\right\}
$$

where $\alpha_{i j}$ are coefficients determined during the fitting procedure.

The $\mathrm{EBF}_{6}$ closure may be computed once $a_{i j}$ is known, where $a_{i j}$ is given in a reference frame not necessarily corresponding to the principal frame of $a_{i j}$. First, the eigenvalues $a_{(1)}$ and $a_{(2)}$ are computed and the rotation tensor is formed from the unit normalized eigenvectors of the second-order tensor (see e.g. Knowles [89]). Next, the principal components of the sixth-order orientation tensor $a_{(i j k l m n)}$ are computed from Equation (3.2). Finally, the sixth-order orientation tensor in the principal frame $a_{(i j k l m n)}$ is rotated into the reference frame yielding $a_{i j k l m n}$ for the given $a_{i j}$. Note that the rotation of a sixth-order orientation tensor is not trivial. A sixth-order closure formed in the principal frame will require the rotation of 729 tensor components (28 if tensor symmetries are accounted for) which is a significant computational cost.

### 3.1.2 Invariant Based Sixth-order Fitted Closure

To avoid the costly tensor rotations, an invariant-based sixth-order fitted closure may be formed in a similar manner to the fourth-order natural closure of Dupret et al. [55] and Chung and Kwon [56]. The sixth-order invariant based fitted closure $\left(\mathrm{IBF}_{6}\right)$ is formed by taking a general expression for a fully symmetric sixth-order tensor $a_{i j k l m n}$ in terms of the second-order tensor $a_{i j}$ and the unit tensor $\delta_{i j}$. The form of the invariant based sixth-order closure is defined by writing all possible combinations of a second-order tensor to define an approximation of $a_{i j k l m n}$ as

$$
\begin{align*}
a_{i j k l m n}^{\mathrm{IBF}} 6 & =\beta_{1} S\left(\delta_{i j} \delta_{k l} \delta_{m n}\right)+\beta_{2} S\left(\delta_{i j} \delta_{k l} a_{m n}\right)+\beta_{3} S\left(\delta_{i j} \delta_{k l} a_{m p} a_{p n}\right)+\beta_{4} S\left(\delta_{i j} a_{k l} a_{m n}\right) \\
& +\beta_{5} S\left(\delta_{i j} a_{k l} a_{m p} a_{p n}\right)+\beta_{6} S\left(\delta_{i j} a_{k p} a_{p l} a_{m q} a_{q n}\right)+\beta_{7} S\left(a_{i j} a_{k l} a_{m n}\right) \\
& +\beta_{8} S\left(a_{i j} a_{k l} a_{m p} a_{p n}\right)+\beta_{9} S\left(a_{i j} a_{k p} a_{p l} a_{m q} a_{q n}\right)+\beta_{10} S\left(a_{i p} a_{p j} a_{k q} a_{q l} a_{m r} a_{r n}\right) \tag{3.3}
\end{align*}
$$

where the expressions $\beta_{i}, i=1,2, \ldots 10$ are functions of the second-order orientation tensor, and the operator $S$ represents the symmetric part of its argument which can
be expressed for a general sixth-order tensor $B_{i j k l m n}$ as

$$
\begin{equation*}
S\left(B_{i j k l m n}\right)=\frac{1}{720}\left(B_{i j k l m n}+B_{k l i j m n}+B_{j i k l m n}+\cdots(720 \text { total terms })\right) \tag{3.4}
\end{equation*}
$$

Employing the relations between second-, fourth- and sixth-order orientation tensors from Equation (2.12), three of the ten $\beta_{i}$ in Equation (3.3) can be solved in terms of the remaining seven. In this work, $\beta_{1}, \beta_{2}$ and $\beta_{3}$ are selected as dependent to avoid singularity issues (see e.g. [30,54]) since arbitrarily selecting other combinations of three dependent $\beta_{i}$ terms may lead to singularities in the solution at some orientation states (i.e. perfect alignment or completely random in space). After some mathematical manipulation using the normalization and symmetry conditions of the second-order orientation tensor given in Equations (2.12) and (2.13), respectively, expressions for the dependant $\beta_{i}$ functions can be expressed in terms of second and third invariants of the second-order orientation tensor (see e.g. Boresi and Chong [90]), II and III, respectively, as

$$
\begin{align*}
\beta_{1}= & -\frac{2}{21}+\left(-\frac{4}{35} \mathrm{II}+\frac{3}{35}\right) \beta_{4}+\left(-\frac{8}{35} \mathrm{II}-\frac{2}{5} \mathrm{III}+\frac{3}{35}\right) \beta_{5} \\
& +\left(-\frac{12}{35} \mathrm{II}-\frac{12}{35} \mathrm{III}+\frac{8}{35} \mathrm{II}^{2}+\frac{3}{35}\right) \beta_{6}+\left(-\frac{8}{35} \mathrm{II}-\frac{8}{35} \mathrm{III}+\frac{2}{21}\right) \beta_{7} \\
+ & \left(-\frac{12}{35} \mathrm{II}-\frac{8}{35} \mathrm{III}+\frac{16}{105} \mathrm{II}^{2}+\frac{2}{21}\right) \beta_{8} \\
+ & \left(-\frac{16}{35} \mathrm{II}-\frac{4}{21} \mathrm{III}+\frac{16}{35} \mathrm{II}^{2}+\frac{8}{35} \mathrm{II} \mathrm{III}+\frac{2}{21}\right) \beta_{9} \\
+ & \left(-\frac{4}{7} \mathrm{II}-\frac{4}{35} \mathrm{III}+\frac{32}{35} \mathrm{II}^{2}+\frac{8}{35} \mathrm{II} \mathrm{III}-\frac{8}{35} \mathrm{III}^{2}-\frac{32}{105} \mathrm{II}^{3}+\frac{2}{21}\right) \beta_{10}  \tag{3.5}\\
\beta_{2} & =\frac{5}{7}-\frac{2}{7} \beta_{4}+\left(\frac{6}{7} \mathrm{II}-\frac{1}{7}\right) \beta_{5}+\left(\frac{4}{7} \mathrm{II}-\frac{4}{7} \mathrm{III}\right) \beta_{6}+\left(\frac{4}{7} \mathrm{II}-\frac{1}{7}\right) \beta_{7} \\
& +\left(\frac{16}{21} \mathrm{II}-\frac{4}{7} \mathrm{III}-\frac{2}{21}\right) \beta_{8}+\left(\frac{16}{21} \mathrm{II}-\frac{4}{7} \mathrm{III}-\frac{16}{21} \mathrm{II}^{2}-\frac{1}{21}\right) \beta_{9} \\
& +\left(\frac{4}{7} \mathrm{II}-\frac{4}{7} \mathrm{III}-\frac{8}{7} \mathrm{II}^{2}+\frac{8}{7} \mathrm{II} \mathrm{III}\right) \beta_{10} \tag{3.6}
\end{align*}
$$

$$
\begin{align*}
\beta_{3} & =-\frac{4}{7} \beta_{4}-\frac{5}{7} \beta_{5}+\left(\frac{8}{7} \mathrm{II}-\frac{6}{7}\right) \beta_{6}-\frac{4}{7} \beta_{7}+\left(\frac{4}{7} \mathrm{II}-\frac{13}{21}\right) \beta_{8}+\left(\frac{4}{3} \mathrm{II}-\frac{4}{7} \mathrm{III}-\frac{2}{3}\right) \beta_{9} \\
& +\left(\frac{16}{7} \mathrm{II}-\frac{8}{7} \mathrm{III}-\frac{8}{7} \mathrm{II}^{2}-\frac{5}{7}\right) \beta_{10} \tag{3.7}
\end{align*}
$$

Since a closed form solution to equate the second- and sixth-order orientation tensor exists only for perfectly aligned and random orientations, a fitting procedure is used to form the remaining functions $\beta_{i}$ for a wide range of fiber orientations. The $\mathrm{IBF}_{6}$ closure assumes that $\beta_{4}$ through $\beta_{10}$ are functions of the second- and third-invariant of the second-order orientation tensor, recognizing that the first-invariant is identically equal to 1 by the normalization property given in Equation (2.13). The 7 functions $\beta_{4}$ through $\beta_{10}$ are fit to a third order polynomial of the second- and third-invariant of the second-order orientation tensor as

$$
\begin{align*}
\beta_{i}= & \mathcal{B}_{i 1}+\mathcal{B}_{i 2} \mathrm{II}+\mathcal{B}_{i 3} \mathrm{III}+\mathcal{B}_{i 4} \mathrm{II}^{2}+\mathcal{B}_{i 5} \mathrm{II} \mathrm{III}+\mathcal{B}_{i 6} \mathrm{III}^{2}+\mathcal{B}_{i 7} \mathrm{II}^{3} \\
& +\mathcal{B}_{i 8} \mathrm{II}^{2} \mathrm{III}+\mathcal{B}_{i 9} \mathrm{II} \mathrm{III}^{2}+\mathcal{B}_{i 10} \mathrm{III}^{3} \\
i= & 4,5, \ldots, 10 \tag{3.8}
\end{align*}
$$

requiring the fitting of the $10 \times 7=70$ parameters $\mathcal{B}_{i j}$. This number of fitted parameters is not much different than that used in other fourth-order closures such as the orthotropic closure employed by VerWeyst [91] which contains 45 fitted parameters and the invariant based closure of Chung and Kwon [56] which requires the fitting of 63 parameters. A third-order polynomial of the invariants of the second-order orientation tensor is selected to diminish the number of fitted parameters while retaining an accurate representation of the sixth-order orientation tensor.


Figure 3.1: Eigenspace of possible orientations of the second-order orientation tensor.

### 3.1.3 Computed Results

As with previous fitted closures [27,33,56-58, 91], multiple flows are considered that produce second-order tensors that span the eigenspace as shown in Figure 3.1. The eigenvalues $a_{(i)}$ of the second-order orientation tensor are defined such that $a_{(1)} \geq$ $a_{(2)} \geq a_{(3)} \geq 0$. Then recognizing that $a_{(1)}+a_{(2)}+a_{(3)}=1$, there are only two independent eigenvalues of $a_{i j}$. Therefore all possible orientation states are contained within the shaded region of Figure 3.1. To encompass as much of the eigenspace of the second-order orientation tensor as possible, fourteen representative flows are selected. Five of the flows are similar to those presented by Cintra and Tucker [27], three of the flows are from Chung and Kwon [56], and the remaining six are variations of those from Cintra and Tucker and from Chung and Kwon, selected to fill the eigenspace of $a_{i j}$. The fourteen flows used here are:

1. Simple Shear, $C_{I}=10^{-3}, v_{1}=G x_{2}, v_{2}=v_{3}=0$
2. Biaxial Elongation, $C_{I}=10^{-3}, v_{1}=G x_{1}, v_{2}=G x_{2}, v_{3}=-2 G x_{3}$
3. Uniaxial Elongation, $C_{I}=10^{-3}, v_{1}=2 G x_{1}, v_{2}=-G x_{2}, v_{3}=-G x_{3}$
4. Shear Stretch A, $C_{I}=10^{-3}, v_{1}=-G x_{1}+10 G x_{2}, v_{2}=-G x_{2}, v_{3}=2 G x_{3}$
5. Shear Stretch B, $C_{I}=10^{-3}, v_{1}=-G x_{1}+G x_{2}, v_{2}=-G x_{2}, v_{3}=2 G x_{3}$
6. Simple Shear, $C_{I}=10^{-2}, v_{1}=G x_{2}, v_{2}=v_{3}=0$
7. Shear Stretch C, $C_{I}=10^{-2}, v_{1}=-G x_{1}+3.75 G x_{2}, v_{2}=-G x_{2}, v_{3}=2 G x_{3}$
8. Shear Stretch D, $C_{I}=10^{-2}, v_{1}=-G x_{1}+1.5 G x_{2}, v_{2}=-G x_{2}, v_{3}=2 G x_{3}$
9. Shear/Biaxial A, $C_{I}=10^{-2}$, $v_{1}=G x_{1}+2 G x_{3}, v_{2}=G x_{2}, v_{3}=-2 G x_{3}$
10. Shear/Biaxial B, $C_{I}=10^{-2}$, $v_{1}=G x_{1}+2.75 G x_{3}, v_{2}=G x_{2}, v_{3}=-2 G x_{3}$
11. Shear/Biaxial C, $C_{I}=10^{-2}, v_{1}=G x_{1}+1.25 G x_{3}, v_{2}=G x_{2}, v_{3}=-2 G x_{3}$
12. Shear/Planar A, $C_{I}=10^{-2}, v_{1}=-G x_{1}+10 G x_{3}, v_{2}=G x_{2}, v_{3}=0$
13. Shear/Planar B, $C_{I}=10^{-2}, v_{1}=-G x_{1}+G x_{3}, v_{2}=G x_{2}, v_{3}=0$
14. Shear/Uniaxial, $C_{I}=10^{-2}, v_{1}=2 G x_{1}+3 G x_{3}, v_{2}=-G x_{2}, v_{3}=-G x_{3}$
where $v_{i}$ are the flow velocity components in the $x_{i}$ direction and $G$ is a scaling parameter. Each fiber flow evolution starts from an isotropic orientation, often represented as $a_{(1)}=a_{(2)}=a_{(3)}=1 / 3$. The distribution function $\psi(\theta, \phi, t)$ is solved with Equation (2.25) through steady state using the finite difference technique of Bay [23] for each of the fourteen representative flows. The eigenspace with the flows of $C_{I}=10^{-2}$ are seen in Figure 3.2 and the eigenspace of the flows with $C_{I}=10^{-3}$ are shown in Figure 3.3. As demonstrated in the two figures, the fourteen representative flows encompass much of the eigenspace of the second-order orientation tensor.

For most fitted closures, the selection of the representative orientation state is taken at discrete time increments throughout the orientation evolution [27, 33, 56]. This tends to weigh the fitted solution more towards the steady state solution, while neglecting the transient orientations. To avoid favoring a particular alignment state, the eigenspace triangle in Figure 3.1 is divided into sub-regions, with one orientation


Figure 3.2: Representative flows with an interaction coefficient of $C_{I}=10^{-2}$ used in the fitting of the preliminary sixth-order closures.


Figure 3.3: Representative flows with an interaction coefficient of $C_{I}=10^{-3}$ used in the fitting of the preliminary sixth-order closures.


Figure 3.4: Eigenspace containing the 428 representative orientation states.
state selected within each of the smaller regions. This is similar to the method used in the eigenvalue based closure employed by VerWeyst [91] and the rational closure of Chaubal and Leal [49]. Various discretizations of the eigenspace triangle were considered where it was found that a minimum of 250 sub-regions were required to yield satisfactory results. The fitting procedure given in the following section employed 428 sub-regions that yield the orientation states shown in Figure 3.4.

The unknown coefficients $\alpha_{i j}$ of the eigenvalue based closure in Equation (3.2) and $\mathcal{B}_{i j}$ of the invariant based closure in Equation (3.8) are determined through a fitting procedure that minimizes the difference between the true sixth-order orientation tensor $a_{i j k l m n}$ computed from the actual distribution function $\psi(\theta, \phi, t)$, and the sixth-order orientation tensor evaluated with a given closure formula. Note that the
two closures are fit in slightly different manners. Determining a useful set of coefficients $\alpha_{i j}$ and $\mathcal{B}_{i j}$ is a nontrivial procedure since the optimization procedure suffers from local minima issues and is highly dependant on the selection of representative flow fields and the flow history.

The fourth-order orientation tensor components from each of the different orientation states are substituted into the $\mathrm{EBF}_{6}$ closure of Equation (3.2) and the $\mathrm{IBF}_{6}$ closure of Equation (3.3). For the $\mathrm{EBF}_{6}$ closure the fitted components of the sixthorder orientation tensor $a_{i j k l m n}^{\mathrm{EBF}_{6}}$ are compared with the sixth-order orientation tensor obtained from the actual distribution $a_{i j k l m n}$ using Equation (2.11) to minimize the cost function

$$
\begin{equation*}
\chi_{\mathrm{EBF}_{6}}^{2}=\sum_{N=1}^{n p t s} \sum_{i=1}^{3} \sum_{j=i}^{3} \sum_{k=j}^{3}\left(a_{(i i j j k k)}^{N}-a_{(i i j j k k))}^{N \mathrm{EBF}_{6}}\right)^{2} \tag{3.9}
\end{equation*}
$$

where npts represents the number of different orientation states over which the minimization is performed. Notice only the components of $a_{i j k l m n}$ that are assumed to be non-zero in the principal frame of $a_{i j}$ are used in the fitting for the $\mathrm{EBF}_{6}$ closure. For the $\mathrm{IBF}_{6}$ closure, the 28 independent components of the sixth-order orientation tensor $a_{i j k l m n}^{\mathrm{IBF}_{6}}$ are tabulated and compared with the sixth-order orientation tensor $a_{i j k l m n}$ obtained from the actual distribution function. Fitted components and the components of the actual sixth-order orientation tensor define the cost function $\chi_{\mathrm{IBF}_{6}}^{2}$ to be minimized as

$$
\begin{equation*}
\chi_{\mathrm{IBF}_{6}}^{2}=\sum_{N=1}^{n p t s} \sum_{i=1}^{3} \sum_{j=i}^{3} \sum_{k=j}^{3} \sum_{l=k}^{3} \sum_{m=l}^{3} \sum_{n=m}^{3}\left(a_{i j k l m n}^{N}-a_{i j k l m n}^{N \mathrm{IBF}_{6}}\right)^{2} \tag{3.10}
\end{equation*}
$$

Notice that the nonstandard summations in Equations (3.9) and (3.10) (i.e. $j=$ $i, \ldots, 3$ not $j=1, \ldots, 3)$ applies the summation over the 28 independent components of the sixth-order orientation tensor only. The minimizations of Equation (3.9) and

Table 3.1: Optimal Set of Coefficients for the $\mathrm{EBF}_{6}$ closure.

|  | $\alpha_{i 1}$ | $\alpha_{i 2}$ | $\alpha_{i 3}$ | $\alpha_{i 4}$ | $\alpha_{i 5}$ | $\alpha_{i 6}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| $i=1$ | 0.202748 | -0.255697 | -0.710519 | 0.723147 | 1.040096 | 0.608822 |
| $i=2$ | 0.039026 | -0.094871 | -0.082023 | 0.423829 | 0.062962 | -0.046934 |
| $i=3$ | 0.226678 | -0.592443 | -0.487907 | 0.939447 | 0.366080 | 1.191984 |
| $i=4$ | 1.041155 | -1.887184 | -2.028437 | 1.814210 | 0.847071 | 1.003185 |

Table 3.2: Optimal Set of Coefficients for the $\mathrm{IBF}_{6}$ closure.

|  | $\mathcal{B}_{4 j}$ | $\mathcal{B}_{5 j}$ | $\mathcal{B}_{6 j}$ | $\mathcal{B}_{7 j}$ | $\mathcal{B}_{8 j}$ | $\mathcal{B}_{9 j}$ | $\mathcal{B}_{10 j}$ |
| :--- | ---: | ---: | ---: | ---: | ---: | ---: | ---: |
| $j=1$ | 7.058430 | -13.455559 | 6.454477 | 6.327924 | -1.969256 | -8.883780 | 5.472371 |
| $j=2$ | -22.713077 | 30.725940 | -9.383351 | -6.965827 | -1.305849 | -2.275332 | 8.501474 |
| $j=3$ | -6.056070 | -1.578164 | -5.930348 | 1.697995 | 1.835046 | 2.275327 | 7.068016 |
| $j=4$ | 0.973595 | 15.735807 | -5.613790 | 3.535952 | 2.632386 | 1.356542 | 5.077645 |
| $j=5$ | 3.901555 | 2.836412 | 0.218869 | 3.341889 | 0.739824 | -1.204312 | -1.943925 |
| $j=6$ | 2.940500 | 2.119271 | 1.476051 | 3.474308 | 2.188826 | 1.381563 | 0.881585 |
| $j=7$ | 5.771155 | 5.469221 | -1.974298 | 2.035038 | -3.725924 | -11.211195 | -13.635644 |
| $j=8$ | 3.447005 | 2.442307 | 1.291376 | 3.316028 | 1.627336 | 0.518012 | -0.084504 |
| $j=9$ | 0.865905 | 0.634756 | 0.451750 | 0.976714 | 0.634758 | 0.404470 | 0.255297 |
| $j=10$ | 0.098945 | 0.072333 | 0.050396 | 0.108356 | 0.073258 | 0.046699 | 0.031920 |

Equation (3.10) were performed using the software VisualDoc 4.0 [92], with the 24 parameters from Equation (3.2) for the $\mathrm{EBF}_{6}$ closure and the 70 parameters from Equation (3.8) for the $\mathrm{IBF}_{6}$ closure. Optimal values of the fitted coefficients appear in Table 3.1 for the $\mathrm{EBF}_{6}$ closure and Table 3.2 for the $\mathrm{IBF}_{6}$.

To evaluate the computational effort associated with these new sixth-order closures, second-order tensors $a_{i j}$ from several flows are first computed from the distribution function evolution in Equation (2.25). Then fourth-order tensors are computed using the orthotropic closure employed by VerWeyst et. al [53]. Sixth-order orientation tensors are then computed from the same second-order tensor components using the $\mathrm{EBF}_{6}$ and the $\mathrm{IBF}_{6}$ closures described above. Due to computationally expensive sixth-order tensor rotations, the $\mathrm{EBF}_{6}$ requires about 4.5 times the computational effort of the VerWeyst closure. Conversely, the $\mathrm{IBF}_{6}$ has a $35 \%$ reduction in the
computational cost when compared to the VerWeyst closure. This decrease is due in large part to the costly tensor rotations required by the VerWeyst closure. In other simulations the time required to solve Equation (2.36) with the $\mathrm{IBF}_{6}$ was found to take about 2.9 times longer than the time required to solve Equation (2.35) using the orthotropic closure of VerWeyst, whereas the $\mathrm{EBF}_{6}$ closure required nearly 10 times the computational effort. The increased computational costs result from the calculation of the 14 independent $a_{i j k l}$ components in Equation (2.36), whereas the evolution of Equation (2.35) includes only 5 independent $a_{i j}$ components.

### 3.1.4 Investigation of Results

The accuracy of the sixth-order fitted closures presented above is assessed using the spherical harmonic reconstruction procedure of Equation (2.70) for several flows. All closure results are compared to the solutions from the evolution of the distribution function in Equation (2.25) (see e.g. Bay [23]), which is assumed here to be the true solution of the orientation distribution function. Results are obtained by computing the second-order orientation tensor $a_{i j}$ with Equation (2.11) from $\psi(\theta, \phi)$ evaluated with Equation (2.25). Then sixth-order tensor components $a_{i j k l m n}^{\mathrm{EBF}_{6}}$ and $a_{i j k l m n}^{\mathrm{IBF}_{6}}$ are computed from Equations (3.2) and (3.3) respectively. The approximate sixth-order orientation tensors are then used to reconstruct the distribution functions $\hat{\psi}_{6}^{\mathrm{EBF}_{6}}$ and $\hat{\psi}_{6}^{\mathrm{IBF}_{6}}$, and finally the error metrics $E R R_{6}^{\mathrm{EBF}_{6}}$ and $E R R_{6}^{\mathrm{IBF}_{6}}$ from Equation (2.71) are computed.

The average error $\overline{E R R}_{n}$ is defined over the transient solution $\psi(\theta, \phi, t)$ as

$$
\begin{equation*}
\overline{E R R}_{n}=\frac{\int \sqrt{\oint_{\mathbb{S}^{2}}\left(\psi(\theta, \phi, t)-\hat{\psi}_{n}(\theta, \phi, t)\right)^{2} \mathrm{~d}} d t}{\Delta t} \tag{3.11}
\end{equation*}
$$

where $\Delta t$ is the range of time considered, typically taken from the initial time to a
steady state orientation condition and $\hat{\psi}_{n}$ is the reconstructed distribution function from the orientation tensors as given in Equation (2.70). Truncation limits are selected to assess the accuracy of closures since this approach provides a meaningful comparison between closures of fourth- and sixth-order on the representation of the fiber orientation distribution function (see e.g. Jack [59] and Jack and Smith [60]).

### 3.1.4.1 Simple Shear Flow

The first example considers simple-shear flow (e.g. flow condition \#1 from above) with an interaction coefficient of $C_{I}=10^{-3}$ beginning from an initially random distribution of fibers. For reference, the second-order tensor components from the distribution function evolution are given in Figure 3.5. Notice that the fibers begin from a random distribution $\left(a_{11}=a_{22}=a_{33}=1 / 3\right)$, and obtain an orientation state where most of the fibers tend to align along the $x_{1}$ axis as seen by the large $a_{11}$ component. The errors in reconstruction $E R R_{2}, E R R_{4}$ and $E R R_{6}$ are formed from the actual second-, fourth- and sixth-order orientation tensors and appear in Figure 3.6. These represent truncation limits that occur when higher-order data is ignored [60]. Note also that $E R R_{4}$ and $E R R_{6}$, respectively, define accuracy limits when fourth- and sixth-order closures are employed. For example, an ideal sixth-order closure could at best approach the line $E R R_{6}$. From the distribution function evolution, the second-order orientation tensors are computed. Then the sixth-order orientation tensors $a_{i j k l m n}^{\mathrm{EBF}_{6}}$ and $a_{i j k l m n}^{\mathrm{IBF}_{6}}$, are formed from the $\mathrm{EBF}_{6}$ and $\mathrm{IBF}_{6}$ closures, respectively. Then the distribution function is reconstructed through Equation (2.70) and finally the errors of reconstruction $E R R_{6}^{\mathrm{EBF}_{6}}$ and $E R R_{6}^{\mathrm{IBF}_{6}}$ are formed from Equation (2.71). Notice that throughout the transient solution shown in Figure 3.6, the error in reconstruction from both the $\mathrm{EBF}_{6}$ and $\mathrm{IBF}_{6}$ closures approach the sixth-order truncation limit
$E R R_{6}$, with the $\mathrm{IBF}_{6}$ closure performing slightly better than the $E B F_{6}$ closure.

### 3.1.4.2 Shear Planar A Flow

The second example considers Shear/Planar A flow (e.g. flow condition \#12 from above) with $C_{I}=10^{-2}$ which superimposes shearing along the $x_{1}$-axis in the $x_{3}$ direction and planar elongation in the $x_{1}-x_{2}$ plane. The fiber alignment is initially random and exhibits a larger degree of fiber interaction than seen in the simple shear example. Second-order orientation tensor components are plotted in Figure 3.7. Notice for this example that alignment of the fibers begins in a manner similar to the example considered above for simple shear flow, with the fibers quickly becoming aligned along the $x_{1}$ axis, but as steady state approaches the fibers tend to orient in the $x_{1}-x_{2}$ plane. From the second-order orientation tensor, the approximate sixth-order tensors are computed with the $\mathrm{EBF}_{6}$ and $\mathrm{IBF}_{6}$ closures, and the error in reconstruction from the closures are given in Figure 3.8. For Shear/Planar A, the $\mathrm{IBF}_{6}$ closure behaves quite well in attaining the sixth-order truncation limit $E R R_{6}$. One the other hand, the $\mathrm{EBF}_{6}$ closure does not represent the distribution function as well as the $\mathrm{IBF}_{6}$ closure, but the error remains below the fourth-order truncation limit $E R R_{4}$.

### 3.1.4.3 Average Reconstruction Error for Investigated Flows

The error in reconstruction is computed for all 14 flows in Section 3.1.3 used in the fitting procedure described above. Instead of presenting transient plots for all the flows, the average of the reconstruction error $\overline{E R R}_{N}$, as given in Equation (3.11), is presented in Table 3.3 for the $\mathrm{EBF}_{6}$ closure and the $\mathrm{IBF}_{6}$ closure. Also given in Table 3.3 are the values $\overline{E R R}_{2}, \overline{E R R}_{4}$ and $\overline{E R R}_{6}$ computed from the orientation tensors obtained from the distribution function evolution. These three values represent the


Figure 3.5: Selected tensor components of $a_{i j}$ for Simple-Shear for $C_{I}=10^{-3}$.


Figure 3.6: Transient error for Simple-Shear for $C_{I}=10^{-3}$.


Figure 3.7: Selected tensor components of $a_{i j}$ for Shear/Stretch A for $C_{I}=10^{-2}$.


Figure 3.8: Transient error for Shear/Stretch A for $C_{I}=10^{-2}$.

Table 3.3: Comparison of $\overline{E R R}_{2}, \overline{E R R}_{4}$ and $\overline{E R R}_{6}$ to $\overline{E R R}_{6}^{\mathrm{EBF}_{6}}$ and $\overline{E R R}_{6}^{\mathrm{IBF}}{ }_{6}$ over the 14 flows used in the fitting analysis.

| Flow | $\overline{E R R}_{2}$ | $\overline{E R R}_{4}$ | $\overline{E R R}_{6}$ | $\overline{E R R}_{6}^{\mathrm{EBF}_{6}}$ | $\overline{E R R}_{6}^{\mathrm{IBF}_{6}}$ | $\% \overline{E R R}_{6}^{\mathrm{EBF}_{6}}$ | $\% \overline{E R R}_{6}^{\mathrm{IBF}_{6}}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| 1. | $1.43 \times 10^{0}$ | $1.15 \times 10^{0}$ | $8.80 \times 10^{-1}$ | $9.68 \times 10^{-1}$ | $8.81 \times 10^{-1}$ | 10.00 | 0.14 |
| 2. | $4.13 \times 10^{-1}$ | $3.53 \times 10^{-1}$ | $3.02 \times 10^{-1}$ | $3.23 \times 10^{-1}$ | $3.02 \times 10^{-1}$ | 6.97 | 0.03 |
| 3. | $1.51 \times 10^{1}$ | $1.46 \times 10^{1}$ | $1.40 \times 10^{1}$ | $1.41 \times 10^{1}$ | $1.40 \times 10^{1}$ | 0.82 | 0.01 |
| 4. | $3.07 \times 10^{0}$ | $2.72 \times 10^{0}$ | $2.35 \times 10^{0}$ | $2.42 \times 10^{0}$ | $2.35 \times 10^{0}$ | 3.08 | 0.20 |
| 5. | $1.65 \times 10^{1}$ | $1.61 \times 10^{1}$ | $1.54 \times 10^{1}$ | $1.56 \times 10^{-1}$ | $1.54 \times 10^{1}$ | 0.90 | 0.01 |
| 6. | $1.86 \times 10^{-1}$ | $8.92 \times 10^{-2}$ | $3.86 \times 10^{-2}$ | $4.72 \times 10^{-2}$ | $4.15 \times 10^{-2}$ | 22.08 | 7.51 |
| 7. | $9.97 \times 10^{-1}$ | $6.91 \times 10^{-1}$ | $4.30 \times 10^{-1}$ | $5.29 \times 10^{-1}$ | $4.46 \times 10^{-1}$ | 22.81 | 3.58 |
| 8. | $1.66 \times 10^{0}$ | $1.28 \times 10^{0}$ | $8.96 \times 10^{-1}$ | $9.02 \times 10^{-1}$ | $9.00 \times 10^{-1}$ | 0.70 | 0.49 |
| 9. | $2.08 \times 10^{-1}$ | $1.35 \times 10^{-1}$ | $8.07 \times 10^{-2}$ | $9.20 \times 10^{-2}$ | $8.10 \times 10^{-2}$ | 13.99 | 0.34 |
| 10. | $1.92 \times 10^{-1}$ | $1.20 \times 10^{-1}$ | $6.92 \times 10^{-2}$ | $8.09 \times 10^{-2}$ | $6.96 \times 10^{-2}$ | 16.79 | 0.57 |
| 11. | $2.21 \times 10^{-1}$ | $1.47 \times 10^{-1}$ | $9.04 \times 10^{-2}$ | $1.01 \times 10^{-1}$ | $9.06 \times 10^{-2}$ | 11.87 | 0.21 |
| 12. | $8.70 \times 10^{-2}$ | $4.05 \times 10^{-2}$ | $1.70 \times 10^{-2}$ | $2.97 \times 10^{-2}$ | $1.82 \times 10^{-2}$ | 74.62 | 7.02 |
| 13. | $1.19 \times 10^{0}$ | $8.66 \times 10^{0}$ | $5.67 \times 10^{-1}$ | $6.06 \times 10^{-1}$ | $5.76 \times 10^{-1}$ | 6.79 | 1.49 |
| 14. | $1.40 \times 10^{0}$ | $1.03 \times 10^{0}$ | $6.69 \times 10^{-1}$ | $6.98 \times 10^{-1}$ | $6.76 \times 10^{-1}$ | 4.43 | 1.04 |

limit of a second-, fourth- and sixth-order reconstruction. Notice the error in reconstruction $\overline{E R R}_{6}{ }^{\mathrm{EBF}}{ }_{6}$ from the $\mathrm{EBF}_{6}$ closure and $\overline{E R R}_{6}{ }^{\mathrm{IBF}}{ }_{6}$ from the $\mathrm{IBF}_{6}$ closure is always less than the fourth-order truncation limit $E R R_{4}$ for all flows investigated. In addition, the average error in reconstruction from the $\mathrm{IBF}_{6}$ closure is nearly the same as the average error in reconstruction $\overline{E R R}_{6}$ from the exact sixth-order orientation tensor. The percentage error in Table 3.3 is computed from

$$
\begin{equation*}
\% \overline{E R R}_{6}^{\text {closure }}=\frac{\overline{E R R}_{6}^{\text {closure }}-\overline{E R R}_{6}}{\overline{E R R}_{6}} \tag{3.12}
\end{equation*}
$$

where it is seen that the sixth-order closure $\mathrm{IBF}_{6}$ represents the sixth-order truncation limit quite well for nearly all flows investigated. For the worst case, the percentage error in reconstruction is less than $8 \%$.


Figure 3.9: Schematic for center-gated disk flow.

### 3.1.4.4 Center-Gated Disk

Since all of the results presented above involve flows employed in the fitting of the $\mathrm{EBF}_{6}$ and $\mathrm{IBF}_{6}$ closures, the performance of the closures is investigated in a more general flow condition that is not included in the fitting procedure. The nonhomogeneous flow field represented by a center gated disk, as illustrated in Figure 3.9, is often used for this purpose $[27,33,56]$, and provides an example for analyzing the usefulness of a closure. Unlike the homogeneous flow fields used in the fitting process, the velocity gradients of center-gated disk flow change with radial position and the height within the gap between the mold walls. For a Newtonian fluid, the velocity field for the center-gated disk is given as

$$
\begin{equation*}
v_{r}=\frac{3 Q}{8 \pi r b}\left(1-\frac{z^{2}}{b^{2}}\right) \quad v_{\theta}=v_{z}=0 \tag{3.13}
\end{equation*}
$$

where $r$ is the radial location, $z$ is the gap height between the mold walls (where $z=0$ defines the disk's midplane), $b$ is half the gap thickness, and $Q$ is the volumetric flow rate entering the gate. In a local Cartesian coordinate system where the local directions $\left(x_{1}, x_{2}, x_{3}\right)$ correspond to $(r, \theta, z)$, the velocity gradients are given as

$$
\frac{\partial v_{i}}{\partial x_{j}}=\frac{3 Q}{8 \pi r b}\left[\begin{array}{ccc}
-\frac{1}{r}\left(1-\frac{z^{2}}{b^{2}}\right) & 0 & \frac{2}{b} \frac{z}{b}  \tag{3.14}\\
0 & \frac{1}{r}\left(1-\frac{z^{2}}{b^{2}}\right) & 0 \\
0 & 0 & 0
\end{array}\right]
$$

Notice that for small radii, the flow is dominated by $\frac{\partial v_{1}}{\partial x_{1}}$ and $\frac{\partial v_{2}}{\partial x_{2}}$ providing significant out-of-plane stretching which causes fibers to orient normal to the plane of the flow. As $r$ increases, the flow becomes dominated by shearing from the $\frac{\partial v_{1}}{\partial x_{3}}$ component, causing the fibers to become oriented in the radial direction.

Two examples are presented for gap heights of $z / b=3 / 10$ and $z / b=7 / 10$ with $C_{I}=10^{-2}$. For $z / b=3 / 10$ the transition between out of plane stretching and shearing defines much of the flow history. The lengthy transition time can be easily seen in Figure 3.10 which plots the significant second-order orientation tensor components. For $z / b=7 / 10$, the transition period is relatively quick between the stretching and the shearing flow. The second-order orientation tensor components for $z / b=7 / 10$ are shown in Figure 3.11 where the shearing flow dominates much of the flow history. For the two examples, the error in reconstruction from the $\mathrm{EBF}_{6}$ closure and the $\mathrm{IBF}_{6}$ closure lies in strike contrast to each other. As shown in Figures 3.12 and 3.13 the $\mathrm{IBF}_{6}$ closure lies near the sixth-order truncation limit $E R R_{6}$ throughout the flow evolution for both flows, whereas the $\mathrm{EBF}_{6}$ closure behaves erratically during the entire transition region for $z / b=3 / 10$. The $\mathrm{EBF}_{6}$ closure has difficulty during the transition region for $z / b=7 / 10$ but behaves well during the shear portion of the flow.

The average error in reconstruction of Equation (3.12) is presented for gap heights of $z / b=1 / 10,{ }^{2} / 10, \ldots 9 / 10$ in Table 3.4. The $\mathrm{EBF}_{6}$ closure performs poorly throughout the entire gap height, whereas the $\mathrm{IBF}_{6}$ closure performs relatively well as seen by the percentage error in reconstruction.


Figure 3.10: Selected tensor components of $a_{i j}$ for Center-Gated Disk flow with $C_{I}=$ $10^{-2}$ at $\frac{z}{b}=\frac{3}{10}$.


Figure 3.11: Selected tensor components of $a_{i j}$ for Center-Gated Disk flow with $C_{I}=$ $10^{-2}$ at $\frac{z}{b}=\frac{7}{10}$.


Figure 3.12: Transient error for Center-Gated Disk flow with $C_{I}=10^{-2}$ at $\frac{z}{b}=\frac{3}{10}$.


Figure 3.13: Transient error for Center-Gated Disk flow with $C_{I}=10^{-2}$ at $\frac{z}{b}=\frac{7}{10}$.

Table 3.4: Comparison of $\overline{E R R}_{2}, \overline{E R R}_{4}$ and $\overline{E R R}_{6}$ to $\overline{E R R}_{6}^{\mathrm{EBF}_{6}}$ and $\overline{E R R}_{6}{ }^{\mathrm{IBF}}{ }_{6}$ throughout the gap-height $z / b$ for a center-gated disk, $C_{I}=10^{-2}$.

| $z / b$ | $\overline{E R R}_{2}$ | $\overline{E R R}_{4}$ | $\overline{E R R}_{6}$ | $\overline{E R R}_{6}^{\mathrm{EBF}_{6}}$ | $\overline{E R R}_{6}^{\mathrm{IBF}_{6}}$ | $\% \overline{E R R}_{6}^{\mathrm{EBF}_{6}}$ | $\% \overline{E R R}_{6}^{\mathrm{IBF}_{6}}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| $1 / 10$ | $1.63 \times 10^{-1}$ | $8.58 \times 10^{-2}$ | $4.28 \times 10^{-2}$ | $1.37 \times 10^{-1}$ | $4.61 \times 10^{-2}$ | 220.1 | 7.79 |
| $2 / 10$ | $1.40 \times 10^{-1}$ | $6.88 \times 10^{-2}$ | $3.17 \times 10^{-2}$ | $6.85 \times 10^{-2}$ | $3.37 \times 10^{-2}$ | 116.0 | 6.34 |
| $3 / 10$ | $1.34 \times 10^{-1}$ | $6.38 \times 10^{-2}$ | $2.82 \times 10^{-2}$ | $5.08 \times 10^{-2}$ | $3.00 \times 10^{-2}$ | 80.12 | 6.22 |
| $4 / 10$ | $1.33 \times 10^{-1}$ | $6.23 \times 10^{-2}$ | $2.70 \times 10^{-2}$ | $3.64 \times 10^{-2}$ | $2.87 \times 10^{-2}$ | 34.84 | 6.24 |
| $5 / 10$ | $1.35 \times 10^{-1}$ | $6.25 \times 10^{-2}$ | $2.67 \times 10^{-2}$ | $3.24 \times 10^{-2}$ | $2.85 \times 10^{-2}$ | 21.27 | 6.42 |
| $6 / 10$ | $1.38 \times 10^{-1}$ | $6.38 \times 10^{-2}$ | $2.71 \times 10^{-2}$ | $3.10 \times 10^{-2}$ | $2.89 \times 10^{-2}$ | 14.27 | 6.60 |
| $7 / 10$ | $1.42 \times 10^{-1}$ | $6.55 \times 10^{-2}$ | $2.78 \times 10^{-2}$ | $3.13 \times 10^{-2}$ | $2.91 \times 10^{-2}$ | 12.69 | 4.66 |
| $8 / 10$ | $1.48 \times 10^{-1}$ | $6.85 \times 10^{-2}$ | $2.90 \times 10^{-2}$ | $3.42 \times 10^{-2}$ | $3.10 \times 10^{-2}$ | 17.92 | 6.97 |
| $9 / 10$ | $1.56 \times 10^{-1}$ | $7.18 \times 10^{-2}$ | $3.02 \times 10^{-2}$ | $3.54 \times 10^{-2}$ | $3.25 \times 10^{-2}$ | 17.02 | 7.33 |

### 3.1.5 Discussion of Sixth-Order Closures from $a_{i j}$

Both of the sixth-order fitted closures $\mathrm{EBF}_{6}$ and $\mathrm{IBF}_{6}$ exceed the fourth-order reconstruction truncation limit $E R R_{4}$, and for many orientation states both closures approach the sixth-order truncation limit $E R R_{6}$. By exceeding the fourth-order truncation limit, one can conclude that the sixth-order fitted closures surpass the accuracy in representing the orientation distribution of fibers than is possible by any fourthorder closure. Of the two fitted closures, clearly the $\mathrm{IBF}_{6}$ performs better than the $\mathrm{EBF}_{6}$ closure. For all the orientation states investigated, the error in reconstruction from the $\mathrm{IBF}_{6}$ closure approaches the sixth-order reconstruction limit, whereas the error in reconstruction from the $\mathrm{EBF}_{6}$ closure is not as reliable in representing the sixth-order truncation limit.

There exist several possible reasons for the noticeable difference in accuracy between the $\mathrm{IBF}_{6}$ and the $\mathrm{EBF}_{6}$ closures. One possible difference may be due to the fact that the $\mathrm{EBF}_{6}$ closure explicitly assumes that certain components of the sixth-order orientation tensor are zero, which may or may not be the actual case. Secondly, the
$\mathrm{EBF}_{6}$ closure requires the rotation of the sixth-order orientation tensor. This rotation is a function of the second-order orientation tensor, which may not contain sufficient higher-order information. To perform a more accurate rotation, it may be necessary to investigate the rotation brought about from the eigentensors of the fourth-order orientation tensor (see e.g. [93]).

In these results, there is a significant difference between the number of fitted parameters used in the $\mathrm{EBF}_{6}$ closure (24) and the $\mathrm{IBF}_{6}$ closure (70). Note, however, that the $\mathrm{EBF}_{6}$ closure was fit to a third-, fourth- and fifth-order polynomial (not given here) of the eigenvalues of the second-order orientation tensor without significant accuracy increases to warrant the increased order of the fitted polynomial. It is worthwhile to also note that the cost function for the $\mathrm{EBF}_{6}$ closure was changed to the cost function of the $\mathrm{IBF}_{6}$ closure, introducing a rotation into the calculation of the cost function. The new cost function yielded significantly less accurate results for the $\mathrm{EBF}_{6}$ closure, ergo the cost function in the principal frame of $a_{i j}$ as presented above was employed.

The $\mathrm{EBF}_{6}$ and $\mathrm{IBF}_{6}$ closures are worthwhile to approximate the sixth-order orientation tensor from second-order orientation tensor components. These sixth-order closures are demonstrated to more accurately model the distribution function of fibers than the exact fourth-order reconstruction, and reveals a degree of accuracy unobtainable from even the most elegant fourth-order closure. Of the two closures presented, the invariant based $\mathrm{IBF}_{6}$ closure yields a more accurate representation of the distribution of fibers than does the eigenvalue based $\mathrm{EBF}_{6}$. Unfortunately, both closures are only effective in accurately representing the distribution function of fibers when the second-order orientation tensor is already known by some means. When either the
$\mathrm{IBF}_{6}$ or the $\mathrm{EBF}_{6}$ are employed in actual flow simulations of the second- or fourthorder orientation tensor, solutions were seen to rapidly diverge into the non-physical regime outside of the eigenspace triangle in Figure 3.1. This issue is resolved with the more sophisticated $\mathrm{INV}_{6}$ closure discussed in the following section.

### 3.2 Sixth-order Invariant Based Closure INV ${ }_{6}$

The sixth-order fitted closure $\mathrm{INV}_{6}$ is based on the invariants of the fourth-order fiber orientation tensor and is shown to more accurately represent the fiber orientation distribution than existing closures, including the two aforementioned sixth-order fitted closures. $\mathrm{INV}_{6}$ is introduced, along with a discussion of fourth-order tensor invariants. The fitting procedure is then described, followed by an investigation into results that demonstrate the increased accuracy of the $\mathrm{INV}_{6}$ closure. The effect of the new sixthorder fitted closure on the prediction of mechanical properties and computational expense is also discussed.

As described previously, several fitted closures have been developed for $a_{i j k l}$ (e.g. $[27,30,33,35,53-56]$ ), however there is no fitted closure to represent $a_{i j l k m n}$ as a function of $a_{i j k l}$. One possible method for forming a fitted sixth-order closure is to employ a procedure analogous to that used to define the fourth-order eigenvalue based closures $[27,33,53]$. This, however, would require that the sixth-order tensor be rotated into the principal frame of the second-order tensor, or perhaps the planes of material symmetry for the fourth-order orientation tensor which introduces computationally expensive rotations. Additionally, the rotation may require the costly computation of the eigentensors (not eigenvectors) of a fourth-order tensor [63, 93].

To avoid costly tensor rotations, a sixth-order closure form is considered that more
closely follows the fourth-order natural closure of Dupret et al. [55] and the fourthorder invariant based orthotropic fitted closure of Chung and Kwon [56]. In the proposed sixth-order closure, it is assumed that the material planes of symmetry of the fourth-order orientation tensor define those of the sixth-order orientation tensor. This assumption is analogous to that of the natural and the invariant-based fitted closures in which the principal directions of the second-order orientation tensor define the material planes of symmetry for the fourth-order orientation tensor.

### 3.2.1 Functional Form of $\mathrm{INV}_{6}$

The sixth-order invariant based closure (identified here as $\mathrm{INV}_{6}$ ) is formed from a general expression for a fully symmetric sixth-order tensor $a_{i j k l m n}$ written in terms of the fourth-order tensor $a_{i j k l}$ and the unit tensor $\delta_{i j}$. $\mathrm{INV}_{6}$ is defined in terms of all possible combinations of the fourth-order orientation tensor to form a symmetric sixth-order orientation tensor as

$$
\begin{align*}
a_{i j k l m n}^{\mathrm{INV}}= & \beta_{1} S\left(\delta_{i j} \delta_{k l} \delta_{m n}\right)+\beta_{2} S\left(\delta_{i j} \delta_{k l} a_{m n p p}\right)+\beta_{3} S\left(\delta_{i j} \delta_{k l} a_{p p m r} a_{r n q q}\right) \\
& +\beta_{4} S\left(\delta_{i j} a_{k l p p} a_{m n q q}\right)+\beta_{5} S\left(\delta_{i j} a_{k l p p} a_{q q m s} a_{s n r r}\right)+\beta_{6} S\left(\delta_{i j} a_{p p k t} a_{t l q q} a_{r r m u} a_{u n s s}\right) \\
& +\beta_{7} S\left(a_{i j p p} a_{k l q q} a_{m n r r}\right)+\beta_{8} S\left(a_{i j p p} a_{k l q q} a_{r r m t} a_{t n s s}\right) \\
& +\beta_{9} S\left(a_{i j p p} a_{q q k u} a_{u l r r} a_{s s m v} a_{v n t t}\right)+\beta_{10} S\left(a_{\text {ppiv }} a_{v j q q} a_{r r k w} a_{w l s s} a_{t t m x} a_{\text {xnuu }}\right) \\
& +\beta_{11} S\left(\delta_{i j} a_{k l m n}\right)+\beta_{12} S\left(a_{i j p p} a_{k l m n}\right)+\beta_{13} S\left(a_{p p i r} a_{r j q q} a_{k l m n}\right) \tag{3.15}
\end{align*}
$$

where the terms $\beta_{i}, i=1,2, \ldots 13$ are functions of the fourth-order orientation tensor defined as a linear polynomial of the five independent invariants $\mathcal{I}_{i}, i=2,3, \ldots, 6$ of $a_{i j k l}$ written as

$$
\begin{align*}
\beta_{i} & =\mathcal{B}_{i 1}+\mathcal{B}_{i 2} \mathcal{I}_{2}+\mathcal{B}_{i 3} \mathcal{I}_{3}+\mathcal{B}_{i 4} \mathcal{I}_{4}+\mathcal{B}_{i 5} \mathcal{I}_{5}+\mathcal{B}_{i 6} \mathcal{I}_{6} \\
i & =1,2, \ldots, 13 \tag{3.16}
\end{align*}
$$

where the parameters $\mathcal{B}_{i j}$ will be computed in the fitting procedure discussed below.
The symmetric operator $S$ in Equation (3.15) represents the symmetric part of its argument which is expressed for a general sixth-order tensor $B_{i j k l m n}$ as

$$
\begin{equation*}
S\left(B_{i j k l m n}\right)=\frac{1}{720}\left(B_{i j k l m n}+B_{k l i j m n}+B_{j i k l m n}+\cdots(720 \text { total terms })\right) \tag{3.17}
\end{equation*}
$$

For example, the symmetric operator of the sixth-order tensor $\delta_{i j} \delta_{k l} \delta_{m n}$ may be expressed as

$$
\begin{align*}
S\left(\delta_{i j} \delta_{k l} \delta_{m n}\right)= & \frac{1}{720}\left(\delta_{i j} \delta_{k l} \delta_{m n}+\delta_{k l} \delta_{i j} \delta_{m n}+\delta_{j i} \delta_{k l} \delta_{m n}+\cdots(720 \text { total terms })\right) \\
= & \frac{1}{15}\left(\delta_{k l} \delta_{j m} \delta_{i n}+\delta_{j l} \delta_{k m} \delta_{i n}+\delta_{j k} \delta_{l m} \delta_{i n}+\delta_{k l} \delta_{i m} \delta_{j n}+\delta_{i l} \delta_{k m} \delta_{j n}\right. \\
& +\delta_{i k} \delta_{l m} \delta_{j n}+\delta_{j l} \delta_{i m} \delta_{k n}+\delta_{i l} \delta_{j m} \delta_{k n}+\delta_{i j} \delta_{l m} \delta_{k n}+\delta_{j k} \delta_{i m} \delta_{l n} \\
& \left.+\delta_{i k} \delta_{j m} \delta_{l n}+\delta_{i j} \delta_{k m} \delta_{l n}+\delta_{j k} \delta_{i l} \delta_{m n}+\delta_{i k} \delta_{j l} \delta_{m n}+\delta_{i j} \delta_{k l} \delta_{m n}\right) \tag{3.18}
\end{align*}
$$

The $\mathrm{INV}_{6}$ closure in Equation (3.15) may be simplified using Equation (2.15) as

$$
\begin{align*}
a_{i j k l m n}^{\mathrm{INV}_{6}}= & \beta_{1} S\left(\delta_{i j} \delta_{k l} \delta_{m n}\right)+\beta_{2} S\left(\delta_{i j} \delta_{k l} a_{m n}\right)+\beta_{3} S\left(\delta_{i j} \delta_{k l} a_{m p} a_{p n}\right)+\beta_{4} S\left(\delta_{i j} a_{k l} a_{m n}\right) \\
& +\beta_{5} S\left(\delta_{i j} a_{k l} a_{m p} a_{p n}\right)+\beta_{6} S\left(\delta_{i j} a_{k p} a_{p l} a_{m q} a_{q n}\right)+\beta_{7} S\left(a_{i j} a_{k l} a_{m n}\right) \\
& +\beta_{8} S\left(a_{i j} a_{k l} a_{m p} a_{p n}\right)+\beta_{9} S\left(a_{i j} a_{k p} a_{p l} a_{m q} a_{q n}\right)+\beta_{10} S\left(a_{i p} a_{p j} a_{k q} a_{q l} a_{m r} a_{r n}\right) \\
& +\beta_{11} S\left(\delta_{i j} a_{k l m n}\right)+\beta_{12} S\left(a_{i j} a_{k l m n}\right)+\beta_{13} S\left(a_{i p} a_{p j} a_{k l m n}\right) \tag{3.19}
\end{align*}
$$

which expresses the dependence of $\mathrm{INV}_{6}$ on $a_{i j}$ and $a_{i j k l}$.
In the principal frame of $a_{i j}$, the symmetric operator multiplying the terms $\beta_{1}$ through $\beta_{10}$ in Equation (3.19) can be reduced to a transversely isotropic fourth-order orientation tensor (see e.g. Jack and Smith [7]). When $\beta_{11}, \beta_{12}$ and $\beta_{13}$ are set to zero, the $\mathrm{IBF}_{6}$ closure in Equation (3.3) is obtained. However the terms multiplying $\beta_{11}$, $\beta_{12}$ and $\beta_{13}$ may yield a fully populated (non-zero) fourth-order orientation tensor, thereby allowing a representation of $a_{i j k l}$ that has fewer planes of material symmetry
than transversely isotropic. For example consider the symmetric operator multiplying $\beta_{11}$ in Equation (3.19) where the terms associated with the $a_{1112 k k}$ component of the $\mathrm{INV}_{6}$ closure are given as

$$
\begin{equation*}
S\left(\delta_{11} a_{12 k k}\right)=\frac{1}{15}\left(a_{1112} \delta_{k k}+6 a_{112 k} \delta_{1 k}+3 a_{12 k k} \delta_{11}+2 a_{111 k} \delta_{2 k}+3 a_{11 k k} \delta_{12}\right) \tag{3.20}
\end{equation*}
$$

Recognizing that $\delta_{k k}=3, a_{112 k} \delta_{1 k}=a_{1112}, a_{12 k k} \delta_{11}=a_{12 k k}=a_{12}$, and $a_{11 k k} \delta_{12}=0$, Equation (3.20) simplifies to

$$
\begin{equation*}
S\left(\delta_{11} a_{12 k k}\right)=\frac{1}{15}\left(11 a_{1112}+3 a_{12}\right) \tag{3.21}
\end{equation*}
$$

Note that $a_{12}$ is zero in the principal frame, but $a_{1112}$ may be non-zero even during simple flow conditions. This is of particular importance here since all existing symmetric fourth-order closures implicitly or explicitly set $a_{1112}$ to zero in the principal frame as will be discussed in the following chapter. The preceding argument can be repeated for the terms $a_{1113}, a_{1123}, a_{1222}, a_{1223}, a_{1233}, a_{2223}$, and $a_{2333}$.

The sixth-order linear closure $\hat{a}_{i j k l m n}$ from Advani and Tucker [6] is represented by Equation (3.19) when $\beta_{1}=15 / 693, \beta_{2}=45 / 99$, and $\beta_{11}=15 / 11$ with all other $\beta_{i}=0$. The sixth-order quadratic closure of Doi [48] (see, e.g. Equation (2.40)) composes one of the terms in the symmetric operator $S\left(a_{i j} a_{k l m n}\right)$ from Equation (3.19), but cannot be computed from $\mathrm{INV}_{6}$ since the quadratic closure does not exhibit the symmetries of an orientation tensor, i.e. $\tilde{a}_{i j k l m n} \neq \tilde{a}_{i j m n k l} \forall i, j, k, l, m, n$. In addition, since the sixth-order hybrid closure [6] is formed from the quadratic closure, it also may not be represented by $\mathrm{INV}_{6}$.

### 3.2.2 Fourth-order Orientation Tensor Invariants

The invariant based sixth-order closure $\mathrm{INV}_{6}$ is a function of the invariants of $a_{i j k l}$ as shown in Equation (3.15). The invariants of a second-order tensor are commonly
found in mechanics texts (see e.g. Malvern [9] and Jones [10]), however the invariants of the fourth-order orientation tensor are not prevalent in the literature. The eigenproblem for the fourth-order tensor $\mathcal{D}$ is expressed as [93]

$$
\begin{equation*}
\mathcal{D}: \mathbf{M}=\lambda \mathbf{M} \tag{3.22}
\end{equation*}
$$

where $\mathbf{M}$ is the $3 \times 3$ eigentensor of $\mathcal{D}$ and $\lambda$ is the eigenvalue associated with the eigentensor. A double contraction of the fourth-order tensor $\mathcal{D}$ with a second-order tensor A may be defined as ${ }^{1}$

$$
\begin{equation*}
\mathcal{D}: \mathbf{A}=\mathcal{D}_{i j k l} A_{j k} \mathbf{g}_{i} \otimes \mathbf{g}_{l} \tag{3.23}
\end{equation*}
$$

where $\otimes$ is the dyadic product and $\mathbf{g}$ is a basis vector. Equation (3.22) may be rewritten as

$$
\begin{equation*}
(\mathcal{D}-\lambda \mathcal{I}): \mathbf{M}=\mathbf{0} \tag{3.24}
\end{equation*}
$$

where $\boldsymbol{\mathcal { I }}$ is the fourth-order identity tensor defined as $\boldsymbol{\mathcal { I }}=\mathbf{g}_{i} \otimes \mathbf{g}_{i} \otimes \mathbf{g}_{j} \otimes \mathbf{g}_{j}$, where the usual summation is employed. Assuming the nontrivial solution $\mathbf{M} \neq \mathbf{0}$, nine eigenvalues are obtained from Equation (3.24) from the determinant

$$
\begin{equation*}
\operatorname{det}[\mathcal{D}-\lambda \boldsymbol{\mathcal { I }}]=0 \tag{3.25}
\end{equation*}
$$

which may be evaluated in the usual manner once the components of its argument $\mathcal{D}-\lambda \mathcal{I}$ are represented as a $9 \times 9$ matrix [93]. The invariants $\mathcal{I}_{i}, i=1,2, \ldots 9$ of a

[^0]fourth-order tensor $\mathcal{D}$ are computed from the eigenvalues as [93]
\[

$$
\begin{align*}
\mathcal{I}_{1} & =\lambda_{1}+\lambda_{2}+\cdots+\lambda_{9} \\
\mathcal{I}_{2} & =\lambda_{1} \lambda_{2}+\lambda_{1} \lambda_{3}+\cdots+\lambda_{8} \lambda_{9} \\
\mathcal{I}_{3} & =\lambda_{1} \lambda_{2} \lambda_{3}+\lambda_{1} \lambda_{2} \lambda_{4}+\cdots+\lambda_{7} \lambda_{8} \lambda_{9} \\
& \vdots \\
\mathcal{I}_{9} & =\lambda_{1} \lambda_{2} \cdots \lambda_{9} \tag{3.26}
\end{align*}
$$
\]

The invariants of the fourth-order orientation tensor $a_{i j k l}$ evaluated from Equation (3.26) are included in Equation (3.16) to compute the $\mathrm{INV}_{6}$ closure in Equation (3.15). Using the techniques outlined by Itskov [93] the calculation of the invariants can be performed, but at a substantial computational burden due to the calculation of the fourth-order tensor eigenvalues. A simplified computation may be derived considering that orientation tensors are super-symmetric. A super-symmetric fourthorder tensor is defined as a fourth-order tensor in which any pair of indices may be interchanged without changing its form (e.g. $\mathcal{D}_{i j k l}=\mathcal{D}_{k l i j}=\mathcal{D}_{j i k l}=\mathcal{D}_{i l k j}=\cdots$ ). For a super-symmetric fourth-order tensor the number of non-trivial eigenvalues reduces to six [93], which may be solved by writing the lowest order polynomial of the principal traces of the fourth-order tensor $\mathcal{D}$. Beginning with Equation (3.26) and following mathematical manipulations, the invariants of a general super-symmetric fourth-order tensor $\mathcal{D}$ may be evaluated as

$$
\begin{aligned}
& \mathcal{I}_{1}=\operatorname{Tr}[\mathcal{D}] \\
& \mathcal{I}_{2}=\frac{1}{2} \operatorname{Tr}[\mathcal{D}]^{2}-\frac{1}{2} \operatorname{Tr}\left[\mathcal{D}^{2}\right] \\
& \mathcal{I}_{3}=\frac{1}{6} \operatorname{Tr}[\mathcal{D}]^{3}-\frac{1}{3} \operatorname{Tr}\left[\mathcal{D}^{3}\right]-\frac{1}{2} \operatorname{Tr}\left[\mathcal{D}^{2}\right] \operatorname{Tr}[\mathcal{D}] \\
& \mathcal{I}_{4}=\frac{1}{24} \operatorname{Tr}[\mathcal{D}]^{4}-\frac{1}{4} \operatorname{Tr}\left[\mathcal{D}^{2}\right] \operatorname{Tr}[\mathcal{D}]^{2}+\frac{1}{3} \operatorname{Tr}\left[\mathcal{D}^{3}\right] \operatorname{Tr}[\mathcal{D}]+\frac{1}{8} \operatorname{Tr}\left[\mathcal{D}^{2}\right]^{2}-\frac{1}{4} \operatorname{Tr}\left[\mathcal{D}^{4}\right]
\end{aligned}
$$

$$
\begin{align*}
\mathcal{I}_{5}= & \frac{1}{120} \operatorname{Tr}[\mathcal{D}]^{5}-\frac{1}{12} \operatorname{Tr}\left[\mathcal{D}^{2}\right] \operatorname{Tr}[\mathcal{D}]^{3}+\frac{1}{8} \operatorname{Tr}\left[\mathcal{D}^{2}\right]^{2} \operatorname{Tr}[\mathcal{D}]+\frac{1}{6} \operatorname{Tr}\left[\mathcal{D}^{3}\right] \operatorname{Tr}[\mathcal{D}]^{2} \\
& -\frac{1}{6} \operatorname{Tr}\left[\mathcal{D}^{3}\right] \operatorname{Tr}\left[\mathcal{D}^{2}\right]-\frac{1}{4} \operatorname{Tr}\left[\mathcal{D}^{4}\right] \operatorname{Tr}[\mathcal{D}]+\frac{1}{5} \operatorname{Tr}\left[\mathcal{D}^{5}\right] \\
\mathcal{I}_{6}= & \frac{1}{720} \operatorname{Tr}[\mathcal{D}]^{6}-\frac{1}{48} \operatorname{Tr}[\mathcal{D}]^{4} \operatorname{Tr}\left[\mathcal{D}^{2}\right]+\frac{1}{16} \operatorname{Tr}\left[\mathcal{D}^{2}\right]^{2} \operatorname{Tr}[\mathcal{D}]^{2}+\frac{1}{18} \operatorname{Tr}\left[\mathcal{D}^{3}\right] \operatorname{Tr}[\mathcal{D}]^{3} \\
& -\frac{1}{8} \operatorname{Tr}\left[\mathcal{D}^{4}\right] \operatorname{Tr}[\mathcal{D}]^{2}-\frac{1}{6} \operatorname{Tr}\left[\mathcal{D}^{3}\right] \operatorname{Tr}\left[\mathcal{D}^{2}\right] \operatorname{Tr}[\mathcal{D}]-\frac{1}{48} \operatorname{Tr}\left[\mathcal{D}^{2}\right]^{3}+\frac{1}{18} \operatorname{Tr}\left[\mathcal{D}^{3}\right]^{2} \\
& +\frac{1}{8} \operatorname{Tr}\left[\mathcal{D}^{4}\right] \operatorname{Tr}\left[\mathcal{D}^{2}\right]+\frac{1}{5} \operatorname{Tr}\left[\mathcal{D}^{5}\right] \operatorname{Tr}[\mathcal{D}]-\frac{1}{6} \operatorname{Tr}\left[\mathcal{D}^{6}\right] \tag{3.27}
\end{align*}
$$

The above results agree with Itskov [93] who presents $\mathcal{I}_{1}-\mathcal{I}_{4}$. The remaining two invariants $\mathcal{I}_{5}$ and $\mathcal{I}_{6}$ may be shown to agree with the expression in Equation (3.26) when $\mathcal{D}$ is written in its principal frame of reference. The trace of a fourth-order tensor $\operatorname{Tr}[\mathcal{D}]$ in Equation (3.27) is defined as [93]

$$
\begin{equation*}
\operatorname{Tr}[\mathcal{D}]=\mathcal{D}:: \mathcal{I}=\mathcal{I}:: \mathcal{D}=\mathcal{D}_{i i j j} \tag{3.28}
\end{equation*}
$$

and the operation $\mathcal{D}^{2}$ in Equation (3.27) represents the double contraction of a fourth-order tensor $\mathcal{D}$ with itself as

$$
\begin{equation*}
\mathcal{D}^{2}=\mathcal{D}: \mathcal{D}=\mathcal{D}_{i j k l} \mathcal{D}_{j n r k} \mathbf{g}_{i} \otimes \mathbf{g}_{n} \otimes \mathbf{g}_{r} \otimes \mathbf{g}_{l} \tag{3.29}
\end{equation*}
$$

In a similar manner, $\mathcal{D}^{3}$ is the double contraction of the fourth-order tensor $\mathcal{D}$, with $\mathcal{D}^{2}$ written as

$$
\begin{equation*}
\mathcal{D}^{3}=\mathcal{D}:(\mathcal{D}: \mathcal{D}) \tag{3.30}
\end{equation*}
$$

which obeys the associative law of tensor multiplication. Similar expressions follow for $\mathcal{D}^{4}, \mathcal{D}^{5}$ and $\mathcal{D}^{6}$.

A further simplification for the invariant calculations in Equation (3.27) is realized through the normalization property for orientation tensors in Equations (2.13) and (2.15). For example, the first invariant of the fourth-order orientation tensor is shown
to be unity through Equations (2.13) and (2.15), i.e.,

$$
\begin{align*}
\mathcal{I}_{1}=\operatorname{Tr}\left[a_{i j k l}\right] & =a_{i i j j}=a_{1111}+a_{1122}+a_{1133}+a_{2211}+a_{2222}+a_{2233}+a_{3311}+a_{3322}+a_{3333} \\
& =a_{11}+a_{22}+a_{33}=1 \tag{3.31}
\end{align*}
$$

With $\mathcal{I}_{1}=1$ the non-trivial invariants in Equation (3.27) reduce to the five invariants $\mathcal{I}_{2}$ and $\mathcal{I}_{6}$ that form the $\mathrm{INV}_{6}$ closure in Equation (3.19).

### 3.2.3 Computed Results

The unknown coefficients $\mathcal{B}_{i j}$ in Equation (3.16) are determined through a fitting procedure that minimizes the difference between components of the exact sixth-order orientation tensor $a_{i j k l m n}$ computed from Equation (2.11), and the same tensor evaluated with the $\mathrm{INV}_{6}$ closure from Equation (3.15). During the fitting, $a_{i j k l m n}$ from the $\mathrm{INV}_{6}$ closure is computed from $a_{i j k l}$ obtained by first evaluating $\psi(\theta, \phi, t)$ with Equation (2.25) for various flow simulations. The $a_{i j k l m n}$ values used as the reference points are evaluated from the solution of $\psi(\theta, \phi, t)$ in Equation (2.25) as well.

Equation (3.16) requires the fitting of the $13 \times 6=78$ parameters $\mathcal{B}_{i j}$. Note that this number of fitted parameters is slightly more than that of the orthotropic fourthorder closure of VerWeyst et al. [53] which contains 45 fitted parameters and the fourth-order invariant based closure of Chung and Kwon [56] which fits 63 parameters. The first order polynomial of the five independent invariants of the fourth-order orientation tensor in Equation (3.16) is selected to reduce the number of fitted parameters while retaining the higher-order behavior of the fourth-order orientation tensor. A second-order polynomial, for example, would require 21 coefficients for each value $\beta_{i}$, yielding $13 \times 21=273$ parameters. Although this number is within computational feasibility, it will be demonstrated that a linear function is sufficient.

The fourth-order natural closure [55] and invariant based closure [56] both employ the condition $a_{i j k k}=a_{i j}$ to simplify the number of parameters required in the fitting procedure. For the natural closure this method produced singularities for select orientation states which were corrected by the invariant based closure. Note that a closed form expression using $a_{i j k l m m}=a_{i j k l}$ to simplify the number of fitted parameters has yet to be developed that does not present singularities for select orientation states. Therefore Equation (2.15) is not used to simplify $\mathrm{INV}_{6}$.

The fitting procedure employed in this study is similar to that used to develop the aforementioned preliminary fitted closures. The same fourteen flows presented in Section 3.1.3 are selected to encompass the eigenspace shown in Figure 3.1 for two interaction coefficients $C_{I}=10^{-3}$ and $C_{I}=10^{-2}$ to allow for a broad range of fiber interactions. Each fiber orientation calculation using Equation (2.25) begins with a completely random orientation, such that $a_{(1)}=a_{(2)}=a_{(3)}=1 / 3$, and the distribution function $\psi(\theta, \phi, t)$ is computed by solving Equation (2.25) for each flow condition until steady state is achieved using the finite difference technique of Bay [23].

The fourth-order orientation tensor components from each of the different orientation states are substituted into the $\mathrm{INV}_{6}$ closure of Equation (3.19). The 28 independent components of the sixth-order orientation tensor $a_{i j k l m n}^{\mathrm{INV}_{6}}$ are then compared with the sixth-order orientation tensor obtained from the actual distribution $a_{i j k l m n}$ using Equation (2.11). The fitted components and the components of the true sixth-order orientation tensor define the cost function of the minimization as

$$
\begin{equation*}
\chi^{2}=\sum_{N=1}^{n p t s} \sum_{i=1}^{3} \sum_{j=i}^{3} \sum_{k=j}^{3} \sum_{l=k}^{3} \sum_{m=l}^{3} \sum_{n=m}^{3}\left(a_{i j k l m n}^{N}-a_{i j k l m n}^{N \operatorname{INV}_{6}}\right)^{2} \tag{3.32}
\end{equation*}
$$

where npts is the number of orientation states included in the minimization calculation. Notice that the nonstandard summations (i.e. $j=i, \ldots, 3$ not $j=1, \ldots, 3$ )
limits the summation to be performed over the 28 independent components of the sixth-order orientation tensor. The minimization of Equation (3.32) was performed using the software VisualDOC 4.0 [92], where the 78 parameters from Equation (3.16) serve as design variables in the unconstrained optimization.

Previous fitted closures were defined based on orientation states sampled at uniform time increments throughout the orientation evolution $[27,33,56]$ which may tend to more heavily weight the steady state solution while placing less emphasis on the initial transient response. To prevent over emphasizing a particular alignment state in the calculations, the eigenspace triangle of Figure 3.1 is divided into sub-regions, each of which contributes only one orientation state to the fitting procedure so as to avoid weighting results toward a particular flow or orientation condition. A similar approach was employed by VerWeyst [91] in the formation of the eigenvalue based fourth-order ORT closure. In the fitting procedure, optimizations were performed using several values of npts ranging from 250 to 5,000 . It was found that the coefficients $\mathcal{B}_{i j}$ showed little change for npts $>750$. As a result, npts $=1,100$ was selected since little additional improvements were seen by adding more orientation states. The optimally fitted coefficients $\mathcal{B}_{i j}$ in Equation (3.16) for $n p t s=1,100$ orientation states (as illustrated in Figure 3.14) appear in Table 3.5. It is noted that other fitted coefficients $\mathcal{B}_{i j}$ may yield a lower value of $\chi^{2}$ in Equation (3.32) for a set of orientation states that are different from those defined above. However, the demonstration problems considered below illustrate that the set of $\mathcal{B}_{i j}$ in Table 3.5 yield good results for flow conditions employed in the fitting procedure as well as other, more general, conditions.


Figure 3.14: The 1,100 representative orientation states used in the fitting of $\mathrm{INV}_{6}$.

### 3.2.4 Investigation of Results

To demonstrate the effectiveness of the $\mathrm{INV}_{6}$ closure, two types of flows are investigated. The first investigation employs homogeneous flows where the components of the vorticity tensor $\omega_{i j}$ and the rate of deformation tensor $\dot{\gamma}_{i j}$ are constant. Recall that the $\mathrm{INV}_{6}$ closure is derived entirely from homogeneous flows, and that no direct consideration was made for nonhomogeneous flow. To assess the accuracy of $\mathrm{INV}_{6}$ on flows not included in the fitting procedure, consideration is made for the nonhomogeneous flow of a polymer melt in a center-gated disk, which is representative of flow near an injection molding pin gate. For the center-gated disk example, $\omega_{i j}$ and $\dot{\gamma}_{i j}$ vary with both the radial distance from the gate, and the gap height under consideration. All results are compared to the solutions obtained from Equation (2.25)

Table 3.5: The 78 coefficients $\mathcal{B}_{i j}$ for the $\mathrm{INV}_{6}$ closure from Equation (3.16).

| $\mathcal{B}_{i j}$ | $j=1$ | $j=2$ | $j=3$ | $j=4$ | $j=5$ | $j=6$ |
| ---: | :---: | :---: | :---: | :---: | :---: | :---: |
| $i=1$ | $-6.71528 \times 10^{-4}$ | $2.64957 \times 10^{-4}$ | $3.14372 \times 10^{-2}$ | $-1.93532 \times 10^{-1}$ | $-2.79700 \times 10^{0}$ | $-1.18032 \times 10^{-1}$ |
| $i=2$ | $1.07799 \times 10^{-1}$ | $-7.02833 \times 10^{-1}$ | $2.74216 \times 10^{0}$ | $-2.03454 \times 10^{0}$ | $-7.19463 \times 10^{-1}$ | $-3.03564 \times 10^{-2}$ |
| $i=3$ | $-1.02912 \times 10^{-1}$ | $6.10351 \times 10^{-1}$ | $-1.86199 \times 10^{0}$ | $-2.66728 \times 10^{-1}$ | $-2.03282 \times 10^{-1}$ | $-7.42565 \times 10^{-3}$ |
| $i=4$ | $-9.64877 \times 10^{-1}$ | $-1.67110 \times 10^{0}$ | $-1.32979 \times 10^{0}$ | $5.10429 \times 10^{-2}$ | $-1.73186 \times 10^{-1}$ | $-7.39697 \times 10^{-3}$ |
| $i=5$ | $1.84254 \times 10^{-1}$ | $4.68462 \times 10^{0}$ | $-1.41952 \times 10^{0}$ | $4.67754 \times 10^{-1}$ | $-5.64326 \times 10^{-2}$ | $-1.40796 \times 10^{-3}$ |
| $i=6$ | $3.21562 \times 10^{-1}$ | $-2.63075 \times 10^{0}$ | $-2.96517 \times 10^{0}$ | $3.79441 \times 10^{-1}$ | $-3.34489 \times 10^{-2}$ | $-1.10832 \times 10^{-3}$ |
| $i=7$ | $-2.38593 \times 10^{0}$ | $1.67553 \times 10^{0}$ | $7.95162 \times 10^{-1}$ | $6.43177 \times 10^{-1}$ | $-3.67029 \times 10^{-2}$ | $-1.88557 \times 10^{-3}$ |
| $i=8$ | $1.31397 \times 10^{0}$ | $3.32342 \times 10^{0}$ | $7.60810 \times 10^{-1}$ | $6.69170 \times 10^{-1}$ | $-1.65931 \times 10^{-2}$ | $-1.87309 \times 10^{-3}$ |
| $i=9$ | $-2.10217 \times 10^{0}$ | $1.74331 \times 10^{-2}$ | $-5.74857 \times 10^{-2}$ | $5.96671 \times 10^{-1}$ | $-1.21391 \times 10^{-2}$ | $-1.15321 \times 10^{-4}$ |
| $i=10$ | $1.98698 \times 10^{0}$ | $-1.25858 \times 10^{0}$ | $-5.77328 \times 10^{-1}$ | $5.46301 \times 10^{-1}$ | $-1.10324 \times 10^{-2}$ | $-2.09103 \times 10^{-4}$ |
| $i=11$ | $4.57917 \times 10^{-1}$ | $3.21522 \times 10^{-1}$ | $1.96795 \times 10^{0}$ | $-1.27631 \times 10^{-1}$ | $-3.36087 \times 10^{-1}$ | $-1.51742 \times 10^{-2}$ |
| $i=12$ | $5.28705 \times 10^{0}$ | $-5.36365 \times 10^{0}$ | $-2.87411 \times 10^{-1}$ | $4.86932 \times 10^{-1}$ | $-7.13534 \times 10^{-2}$ | $-2.43975 \times 10^{-3}$ |
| $i=13$ | $-3.10267 \times 10^{0}$ | $1.32892 \times 10^{0}$ | $7.30233 \times 10^{-1}$ | $6.63385 \times 10^{-1}$ | $-2.14327 \times 10^{-2}$ | $-1.30109 \times 10^{-3}$ |

for the distribution function $\psi(\theta, \phi, t)$ [23] which will be labeled DFE, for Distribution Function Evolution, in the following evaluations. The results from these DFE calculations provide a benchmark on which to evaluate closure approximations (see e.g. $[27,35,56]$ ). Flow evolutions of $a_{i j k l}$ are evaluated with Equation (2.36) employing the sixth-order closure, $\mathrm{INV}_{6}$, developed above. For comparison, flow evolutions of $a_{i j}$ using Equation (2.35) are also considered here which employ the fourth-order closure ORT (see e.g. [32,53, 91]). Other fourth-order closures are omitted in the comparison for conciseness since the ORT closure has been shown to be among the best performing closures used to solve Equation (2.35). The evolution of the fourth-order tensor is evaluated with Equation (2.36) using the sixth-order quadratic closure Quad ${ }_{6}$ of Altan et al. [61]. The Quad ${ }_{6}$ closure is presented here to provide a comparison between the $\mathrm{INV}_{6}$ and an analytical sixth-order closure. For many flow conditions, the sixth-order hybrid closure $\operatorname{Hybrid}_{6}$ of Advani and Tucker [6] yields similar results to the Quad $_{6}$ closure, therefore the $\mathrm{Hybrid}_{6}$ results are only discussed briefly for flows
with high shearing and stretching where the Quad $_{6}$ and the Hybrid ${ }_{6}$ behave differently. Note here that stable results are obtained for $\mathrm{INV}_{6}$ with $a_{i j k l}$ evolutions, unlike the issues encountered with $\mathrm{EBF}_{6}$ and $\mathrm{IBF}_{6}$.

### 3.2.4.1 Homogeneous Flows

Fiber orientation calculations were performed for several homogeneous flow conditions where a comparison of the second-order tensor components and the error in reconstructing the distribution function described above are used to asses the $\mathrm{INV}_{6}$ closure. Each flow condition begins from an initially random state where the second-order tensor components $a_{i j}=0, i \neq j$ and $a_{11}=a_{22}=a_{33}={ }^{1} / 3$. The orientation state is then evaluated with the evolution of Equation (2.25) and Equations (2.35) or (2.36). Then the errors in reconstruction are computed with Equations (2.71) and (3.11) using the distribution function reconstruction technique introduced in Equation (2.70).

The first homogeneous flow example is for simple-shear flow where $v_{1}=G x_{3}, v_{2}=$ $v_{3}=0$ with an interaction coefficient $C_{I}=10^{-2}$ (i.e. flow condition $\# 6$ used in the fitting procedure above). The principal second-order tensor components are presented in Figure 3.15 for the orientation tensors computed from the DFE and those resulting from solving Equation (2.35) and (2.36) using the ORT, Quad ${ }_{6}$, and $\mathrm{INV}_{6}$ closures. Notice the solid line representing the $a_{11}$ component obtained from the DFE calculation exhibits a rapid initial increase and stabilizes at a relatively large value. As $a_{11}$ approaches unity, the orientation of the fibers tends to align along the $x_{1}$ axis. Observe how the ORT and $\mathrm{INV}_{6}$ closure results also appearing in Figure 3.15 closely follow the DFE solution, whereas the Quad $_{6}$ closure tends to overpredict the actual fiber alignment. It is interesting to note that $a_{11}$ from the DFE reaches its largest


Figure 3.15: Selected tensor components of $a_{i j}$ for Simple-Shear for $C_{I}=10^{-2}$.
value of 0.766 at $G t=6.7$ and diminishes to a value of 0.727 for $G t>19.7$. This reduction in $a_{11}$ for the ORT closure is nearly imperceptible where $a_{11}$ reduces to 0.773 at the same final time. Alternatively, the $a_{11}$ component from the $\mathrm{INV}_{6}$ simulation is reduced to 0.740 at the end of the flow simulations and thus exhibits a response that is similar to the DFE $a_{11}$ values. Figure 3.15 illustrates that all of the $a_{i j}$ components are more accurate using the $\mathrm{INV}_{6}$ than compared to any of the closures considered here including the sixth-order closure Quad $_{6}$.

The second-order tensor components are useful when visualizing the orientation behavior in the flow, however the ability of a closure to represent the distribution of fibers is better demonstrated with the reconstruction error described previously. Using the DFE results, $E R R_{2}, E R R_{4}$ and $E R R_{6}$ are evaluated, respectively, from the exact second-, fourth-, and sixth- order orientation tensors obtained with Equations


Figure 3.16: Transient error for Simple-Shear for $C_{I}=10^{-2}$.
(2.11) and (2.71). These error measures represent the truncation limits of secondorder, fourth-order, and sixth-order reconstructions, respectively. Figure 3.16 illustrates the truncation limits where it is noted that all three limits attain their maximum value during the state of highest alignment near $G t=6.7$ as shown in Figure 3.15. As expected, $E R R_{N}$ decreases as $N$ increases. In these results, the error in reconstruction from the ORT closure ( $E R R_{\mathrm{ORT}}$ ) approaches the fourth-order truncation limit $E R R_{4}$ and the error in reconstruction formed from the $I N V_{6}$ closure $\left(E R R_{\mathrm{INV}_{6}}\right)$ is nearly indistinguishable from the sixth-order truncation limit $E R R_{6}$. On the other hand, the error in reconstruction from the Quad $_{6}$ closure $\left(E R R_{\text {Quad }_{6}}\right)$ is significantly greater than even the second-order truncation limit $E R R_{2}$ for the entire flow history. The results for the sixth-order hybrid closure Hybrid $_{6}$ are nearly identical to the results of the Quad $_{6}$ and are therefore omitted for clarity.

Simple shear flow with an interaction coefficient of $C_{I}=10^{-3}$ (i.e., flow condition \#1 in Section 3.1.3) results in a more highly aligned state than the previous example. Selected second-order tensor components computed with $C_{I}=10^{-3}$ appear in Figure 3.17 where the overshoot in $a_{11}$ is still evident. The fourth-order ORT closure fails to capture the reduction in $a_{11}$ following its peak value, which is possible with $\mathrm{INV}_{6}$. The $a_{22}$ component is well represented using the $\mathrm{INV}_{6}$ closure, whereas the ORT closure noticeably underpredicts the actual $a_{22}$ component. On the other hand, Quad ${ }_{6}$ again fails to predict the degree of alignment as can be seen in the overprediction of the $a_{11}$ component and the underprediction of the $a_{22}$ component. The errors in reconstruction from the DFE, ORT, Quad $_{6}$ and $\mathrm{INV}_{6}$ are plotted in Figure 3.18. Notice, as in the previous example, the error in reconstruction from the ORT closure approaches the fourth-order truncation limit and the error in reconstruction from the $\mathrm{INV}_{6}$ closure approaches the sixth-order truncation limit. In this example the quadratic closure performs better than in the simulation above, but still fails to approach the sixth-order truncation limit. Notice that $E R R_{\text {Quad }_{6}}$ exhibits a greater accuracy than the fourth-order truncation limit during the highly aligned state near $G t=10$. However, as fiber alignment decreases, the accuracy of the Quad $_{6}$ closure $^{\text {che }}$ does so as well. The results from the $\operatorname{Hybrid}_{6}$ closure are graphically indistinguishable from the results obtained through the $\mathrm{Quad}_{6}$, and are therefore omitted as before.

The final homogeneous flow considered in detail is Shear-Stretch C flow with $v_{1}=-G x_{1}+3.75 G x_{2}, v_{2}=-G x_{2}, v_{3}=2 G x_{3}$ and $C_{I}=10^{-2}$ (i.e. condition $\# 7$ described above). The high degree of alignment is illustrated with the $a_{33}$ component in Figure 3.19 that approaches a value of unity in steady state, which corresponds to a nearly perfect alignment of fibers along the $x_{3}$ axis. Notice that as fibers become


Figure 3.17: Selected tensor components of $a_{i j}$ for Simple-Shear for $C_{I}=10^{-3}$.


Figure 3.18: Transient error for Simple-Shear for $C_{I}=10^{-3}$.


Figure 3.19: Selected tensor components of $a_{i j}$ for Shear Stretch C for $C_{I}=10^{-2}$.
aligned, (i.e. $a_{33} \rightarrow 1$ ) the Quad ${ }_{6}$ closure yields the most accurate representation. On the other hand, the ORT closure underpredicts the fiber alignment, whereas the $\mathrm{INV}_{6}$ closure has a slight overprediction of fiber alignment. In the previous simple shear flow examples the $\mathrm{Hybrid}_{6}$ closure gave nearly identical results to the $\mathrm{Quad}_{6}$ closure $^{\text {. }}$ However, when Shear/Stretch C flow is imposed, the solution from the Hybrid ${ }_{6}$ closure rapidly diverges from the actual solution, yielding non-physical results.

In each of the previous examples, $\mathrm{INV}_{6}$ results approach the sixth-order truncation limit, surpassing the accuracy of the ORT closure and the fourth-order truncation limit. Additionally, $\mathrm{INV}_{6}$ results surpass the sixth-order quadratic closure in representing the fiber orientation distribution function for all flow conditions considered.

To better illustrate the use of fourth- and sixth-order closures, all 14 flows used in the fitting process of the $\mathrm{INV}_{6}$ closure were analyzed by computing the average percent error of reconstruction $\% \overline{E R R}_{N}$ appearing in Equation (3.12). Table 3.6
summarizes these results and presents the average percent error of reconstruction for the ORT closure $\% \overline{E R R}_{\text {ORT }}$, the Quad $_{6}$ closure $\% \overline{E R R}_{\text {Quad }_{6}}$, the Hybrid ${ }_{6}$ closure $\% \overline{E R R}_{\text {Hybrid }_{6}}$, and the $\mathrm{INV}_{6}$ closure $\% \overline{E R R}_{\text {INV }_{6}}$. In most cases the percent error from the $\mathrm{INV}_{6}$ closure is less than $1 \%$. In two cases the percent error is just over $10 \%$, but in comparison to the ORT closure with a percent error over $150 \%$ for the same cases, the $\mathrm{INV}_{6}$ is significantly better. Alternatively, the Quad $_{6}$ and Hybrid ${ }_{6}$ perform poorly for all flows with low degrees of alignment while showing some improvement for flows with high alignment states. It is important to note that solutions of Equation (2.36) were not possible using the $\operatorname{Hybrid}_{6}$ for flows where shearing is large relative to the elongational velocity component (i.e. Shear Stretch A, Shear Stretch C, and Shear Planar A). In these flows, the solution rapidly diverges and eventually yields nonphysical solutions (see e.g. Figure 3.19). For all flow cases, the percent error in reconstruction is substantially lower for the $\mathrm{INV}_{6}$ closure than that seen using any other fourth- or sixth-order closure. Note that the $\% \overline{E R R}_{\text {closure }}$ results measure the reconstruction error against the sixth-order truncation limit. A similar calculation comparing the ORT results to the fourth-order truncation limit would perhaps be more appropriate if the interest were in how the ORT performs relative to $\overline{E R R}_{4}$.

Fiber orientation calculations using the $\mathrm{INV}_{6}$ closure are expected to require more computational effort than the ORT closure since the former evaluates 14 independent components of $a_{i j k l}$ using Equation (2.36) whereas the latter solves for 5 independent components of $a_{i j}$ with Equation (2.35). This alone is expected to yield an approximately $3 \times$ increase when computing fiber orientations by evolving $a_{i j k l}$ instead of just $a_{i j}$. In addition, the form of the closure itself affects computational costs. For example, the $\mathrm{INV}_{6}$ computations in Equation (3.15) include the costly evaluations of the

Table 3.6: Comparison of $\% \overline{E R R}_{\mathrm{ORT}}, \% \overline{E R R}_{\mathrm{Quad}_{6}}, \% \overline{E R R}_{\text {Hybrid }_{6}}$, and $\% \overline{E R R}_{\mathrm{INV}_{6}}$ over the 14 flows used in the fitting procedure.

| Flow | $\% \overline{E R R}_{\text {ORT }}$ | $\% \overline{E R R}_{\text {Quad }_{6}}$ | $\% \overline{E R R}_{\text {Hybrid }_{6}}$ | $\% \overline{E R R}_{\text {INV }_{6}}$ |
| :---: | :--- | :--- | :--- | :--- |
| 1. | 32.54 | 35.41 | 25.27 | 1.310 |
| 2. | 16.88 | 537.7 | 525.8 | 0.002 |
| 3. | 4.197 | 1.034 | 0.890 | 0.248 |
| 4. | 16.03 | 15.55 | $* *$ | 0.824 |
| 5. | 4.101 | 0.708 | 0.242 | 0.001 |
| 6. | 158.1 | 1236. | 946.0 | 10.21 |
| 7. | 60.77 | 23.37 | $* *$ | 0.226 |
| 8. | 42.94 | 9.706 | 4.600 | 0.014 |
| 9. | 67.65 | 2007. | 2074. | 1.748 |
| 10 | 74.49 | 1708. | 1964. | 0.958 |
| 11. | 62.25 | 2012. | 1987. | 1.243 |
| 12. | 150.3 | 4460. | $* *$ | 11.63 |
| 13. | 52.77 | 22.24 | 17.23 | 1.341 |
| 14. | 53.53 | 21.68 | 19.73 | 0.077 |
|  | ${ }^{* *}$ Nonphysical solution reached before steady state attained. |  |  |  |

symmetric operator, whereas the ORT closure requires the rotation of the fourth-order orientation tensor into the principal frame of $a_{i j}$. In both cases, the computational effort can be reduced by taking advantage of the symmetries possessed by all fiber orientation tensors, thus eliminating redundant calculations. The computation time required to perform each of the flow evolutions used in the $\mathrm{INV}_{6}$ fitting process above was evaluated when solving Equation (2.35) with the ORT closure and solving Equation (2.36) with the $\mathrm{INV}_{6}$ closure. The more accurate $\mathrm{INV}_{6}$ evaluations were found to take $2.91 \pm 0.07$ times longer than computations performed with the ORT closure.

### 3.2.4.2 Center-Gated Disk

Since all of the results presented above involve flows employed in the fitting of $\mathrm{INV}_{6}$, it is desired to evaluate the performance of the new closure in a more general flow condition that is not included in the fitting procedure for the $\mathrm{INV}_{6}$ closure. The
nonhomogeneous flow field represented by a center gated disk discussed in Equations (3.13) and (3.14) is selected. Unlike the homogeneous flow fields used in the fitting of the $\mathrm{INV}_{6}$ closure, the velocity gradients of center-gated disk flow change with radial position and the height within the gap between the mold walls.

The first example considers fibers suspended at a gap height of $z / b=2 / 10$ where much of the initial flow history is dominated by out of plane stretching. Initially random fibers begin to orient out-of-the plane of the flow which is illustrated by the rapidly increasing $a_{22}$ component in Figure 3.20. Both the ORT and the $\mathrm{INV}_{6}$ closures accurately follow this rapid increase in out-of-plane alignment, whereas both the Quad $_{6}$ and the Hybrid $_{6}$ closures over predict the actual alignment state, as expected. As the radial location increases and the flow becomes dominated by the shearing in the radial direction, the $a_{11}$ component increases and the $a_{22}$ component decreases signifying an increased alignment in the radial direction. Again, both the ORT and the $\mathrm{INV}_{6}$ results illustrate this behavior, while the Quad $_{6}$ and the Hybrid ${ }_{6}$ over predict the alignment of the fibers in the radial direction. From the second-order tensor components in Figure 3.20 it is clear that earlier sixth-order closures $\left(_{\text {Quad }}^{6}\right.$ and Hybrid $_{6}$ ) yield poor results, whereas the ORT and INV $_{6}$ perform quite well. Similar trends are identified in the error reconstruction plots appearing in Figure 3.21. The Quad $_{6}$ and the Hybrid $_{6}$ both yield a poor representation of the fiber distribution function whereas the ORT and the $\mathrm{INV}_{6}$ reconstructions approach the fourth-order truncation limit $E R R_{4}$ and the sixth-order truncation limit $E R R_{6}$, respectively.

The second-example considers the orientation state at a gap height of $z / b=5 / 10$. As shown in Figure 3.22, tensor computations employing either the ORT or the INV $_{6}$ closures represent the actual second-order tensor components quite well throughout


Figure 3.20: Selected tensor components of $a_{i j}$ for Center-Gated Disk flow with $C_{I}=$ $10^{-2}$ at $\frac{z}{b}=\frac{2}{10}$.


Figure 3.21: Transient error for Center-Gated Disk flow with $C_{I}=10^{-2}$ at $\frac{z}{b}=\frac{2}{10}$.

Table 3.7: Comparison of $\% \overline{E R R}_{\mathrm{ORT}}, \% \overline{E R R}_{\mathrm{Quad}_{6}}, \% \overline{E R R}_{\text {Hybrid }_{6}}$, and $\% \overline{E R R}_{\mathrm{INV}_{6}}$ throughout the gap height for center-gated disk flow $C_{I}=10^{-2}$.

| Flow | $\% \overline{E R R}_{\text {ORT }}$ | $\% \overline{E R R}_{\text {Quad }_{6}}$ | $\% \overline{E R R}_{\text {Hybrid }_{6}}$ | $\% \overline{E R R}_{\text {INV }_{6}}$ |
| :---: | :--- | :--- | :--- | :--- |
| $1 / 10$ | 114.77 | 1405.27 | 1144.86 | 8.64 |
| $2 / 10$ | 138.30 | 1837.14 | 1491.62 | 10.74 |
| $3 / 10$ | 152.19 | 2027.64 | 1633.25 | 13.05 |
| $4 / 10$ | 160.82 | 2109.92 | 1689.95 | 14.76 |
| $5 / 10$ | 166.30 | 2145.03 | 1736.77 | 15.87 |
| $6 / 10$ | 169.95 | 2103.82 | 1737.32 | 16.54 |
| $7 / 10$ | 172.28 | 1968.37 | 1618.70 | 16.88 |
| $8 / 10$ | 173.83 | 1773.25 | 1447.08 | 16.98 |
| $9 / 10$ | 175.82 | 1611.73 | 1304.35 | 17.09 |

the entire flow history, but as the radial location increases tensor evaluations using the $\mathrm{INV}_{6}$ closure represents $a_{i j}$ components slightly better than those employing the ORT closure. The error in reconstruction of the distribution function appears in Figure 3.23 where it is shown that the accuracy in representing the distribution function from the $\mathrm{INV}_{6}$ simulations far surpasses the fourth-order truncation limit $E R R_{4}$ and nearly attains the sixth-order truncation limit $E R R_{6}$.

The average percent error in reconstruction is presented in Table 3.7 for gap heights of $z / b=1 / 10,{ }^{2} / 10, \ldots{ }^{9} / 10$. Throughout the range of gap heights, the average percent error in reconstruction from the $\mathrm{INV}_{6}$ closure is much less than the average percent error of any of the other closures considered.

### 3.2.4.3 Effect on Material Property Calculations

Perhaps the most important use of fiber orientation predictions is the evaluation of the mechanical properties (i.e. Young's modulus and Poisson's ratios) of the short fiber composite. Mechanical properties have been computed from the distribution function (see e.g. [29,52, 64]), therefore, it is expected that a more accurate representation


Figure 3.22: Selected tensor components of $a_{i j}$ for Center-Gated Disk flow with $C_{I}=$ $10^{-2}$ at $\frac{z}{b}=\frac{5}{10}$.


Figure 3.23: Transient error for Center-Gated Disk flow with $C_{I}=10^{-2}$ at $\frac{z}{b}=\frac{5}{10}$.
of $\psi(\theta, \phi)$ as obtained using $\mathrm{INV}_{6}$ is expected to yield more accurate mechanical properties than those computed when fourth-order closures are employed. A common practice is to evaluate the components of the stiffness or compliance tensor via the procedure described by Advani and Tucker [6], and outlined in the following chapters, which is based on the fourth- and second-order orientation tensors and the underlying unidirectional stiffness tensor obtained from the constituent materials (see e.g., [80]). Therefore, the effect of solving Equation (2.36) using $\mathrm{INV}_{6}$ on predicted mechanical properties may be exposed, in part, by considering the components of $a_{i j k l}$. To better illustrate the influence of using $\mathrm{INV}_{6}$ on computed mechanical properties, the evaluation of stiffness tensor components in the principal frame of $a_{i j k l}$ (which are assumed to be defined by the eignevectors of $\left.a_{i j}[27]\right)$ are considered. Here the analysis is restricted to the principal components of the fourth-order orientation tensor as $a_{(i j k l)}$ (where () designate the tensor components given in the principal frame) since these values represent the effect of fiber orientation on material properties as given by Advani and Tucker [6].

For the center-gated disk flow example discussed previously with a gap height of $z / b=5 / 10$, selected principal components are plotted in Figure 3.24. Observe the $\mathrm{INV}_{6}$ closure represents the fourth-order orientation tensor components more accurately than that of the fourth-order ORT closure, therefore the $\mathrm{INV}_{6}$ will represent the material stiffness tensor more accurately than the ORT closure. Note also that $a_{(1123)}$ and $a_{(2223)}$ are non-zero in center-gated disk flow, and as discussed previously the functional form of the $\mathrm{INV}_{6}$ allows the calculation of these relatively small but non-zero terms, whereas the ORT closure explicitly sets $a_{(1123)}$ and $a_{(2223)}$ to zero.


Figure 3.24: Selected tensor components of $a_{(i j k l)}$ for Center-Gated Disk flow with $C_{I}=10^{-2}$ at $\frac{z}{b}=\frac{5}{10}$.

### 3.2.5 Discussion of INV $_{6}$ Closure

An invariant based sixth-order fitted closure $\mathrm{INV}_{6}$ is defined from a general expression for a fully symmetric sixth-order tensor. This new sixth-order closure is written as a function of the fourth-order orientation tensor, where the components of the $\mathrm{INV}_{6}$ closure are fit to a linear polynomial of the fourth-order orientation tensor invariants. The sixth-order $\mathrm{INV}_{6}$ closure is demonstrated to surpass the accuracy of existing fourth-order closures in the representation of the second-order orientation tensor components in all flow simulations investigated. More importantly, reconstruction of the fiber orientation distribution function using $\mathrm{INV}_{6}$ exceeds the fourth-order reconstruction of the distribution function limit $E R R_{4}$ for all flow conditions investigated. By exceeding the fourth-order truncation limit, the $\mathrm{INV}_{6}$ closure surpasses
the accuracy in representing the orientation distribution of fibers than that which is possible using any fourth-order closure. The $\mathrm{INV}_{6}$ closure is nearly as accurate in representing the fiber orientation state as the exact sixth-order reconstruction of the distribution function.

### 3.3 Sixth-Order Fitted Closures

The stated goal into the investigation of sixth-order closures was to develop a closure that provides a more accurate representation of the fiber orientation distribution function than is possible with any fourth-order closure. Clearly each of the presented closures satisfy this goal, but only the $\mathrm{INV}_{6}$ closure may be utilized in actual short-fiber flow simulations. The sixth-order invariant based closure $\mathrm{INV}_{6}$ results demonstrate the increased accuracy of the $\mathrm{INV}_{6}$ closure with only minor additional computational costs. Clearly, the $\mathrm{INV}_{6}$ closure provides a more accurate description of fiber orientation kinematics than the existing closures, and far surpasses the usefulness of the sixth-order fitted models, $\mathrm{EBF}_{6}$ and $\mathrm{IBF}_{6}$. Flow simulations utilizing a sixth-order closure should employ the $\mathrm{INV}_{6}$ over other sixth-order fitted closures to properly evaluate the orientation state of fibers.

## CHAPTER 4

## ORIENTATION CLOSURE APPROXIMATION EFFECTS ON MECHANICAL PROPERTY PREDICTIONS

Current approaches used to simulate the flow of short-fiber suspensions employ fiber orientation tensors where the dependence on higher-order orientation tensors is alleviated through the use of a closure. Unfortunately, the effect that a given closure has on material property predictions has received little attention in the literature. Accordingly this investigation, also appearing in Jack and Smith [7], demonstrates that current objective fourth-order closures assume an orthotropic form, whereas existing sixth-order closures, such as the $\mathrm{INV}_{6}$, are shown to provide a material representation with fewer symmetries than orthotropic. Numerical calculations demonstrate the existence of shear-extensional and shear-shear coupling in simple homogeneous flow and for a center-gated disk. Although these components are minor in comparison to principal components of the stiffness tensor, these components can be obtained via the fitted sixth-order $\mathrm{INV}_{6}$ closure.

### 4.1 Motivation for Current Investigation

The current investigation into closures is undertaken to provide a better understanding related to the linear elastic material behavior when a closure is employed in polymer composite molding simulations. The closure used in a flow simulation plays a key role since fourth-order elasticity tensors describing the mechanical properties of the composite structure are computed from fourth-order orientation tensors $[6,27-29,35-37,56,64,78,79]$ as are thermo-elastic properties (e.g. Camacho et
al. [71]). The inclusion of sixth-order orientation tensor closures in the present investigation is motivated by Altan et al. [52] who state that lower- (e.g. fourth-) order approximations may result in "...errors for complex flow fields, where both shear and elongational velocity gradients exist in all three different planes. Therefore, higherorder (e.g. sixth-order) approximations may be required for the accurate description of suspension mechanics."

This chapter investigates the effect of closure approximations on material property behavior for injection molding predictions of short-fiber composites. This is only one factor that must be taken into consideration when selecting an appropriate closure for numerical predictions, but may be a deciding factor. By construction, the class of fourth-order orthotropic closures (see e.g. [27]) assume an orthotropic form. Cintra and Tucker [27] state that all objective fourth-order closures must be orthotropic, however, the form of many current fourth-order closures is not obvious. Furthermore, the orthotropic issue has not been addressed for sixth-order closures. It will be demonstrated that current fourth-order closures employed in short fiber composite process simulations either directly or indirectly assume an orthotropic form. In addition, an investigation into the sixth-order invariant based fitted closure of Jack and Smith [38] (see e.g. Equation (3.15)) will demonstrate that sixth-order closures provide a means for representing a material state that is more general than orthotropic.

Fiber orientations in several homogeneous flows are considered below to explore conditions where fourth- and/or sixth-order closures are appropriate. In addition, the inhomogeneous flow of a center-gated disk is included to relate the current work to flows found in the injection molding process. These simulations, all of which are based on the Folgar-Tucker model for fiber orientation [11], demonstrate that although
fourth-order closures are constrained by orthotropic assumptions, non-orthotropic components of the resulting elasticity tensor are small in comparison to the orthotropic components.

### 4.2 Material Behavior from Closures

The symmetries of $a_{i j k l}$ make it possible to define a contracted notation for fourthorder tensors where indices of 11,22 , and 33 are replaced with 1,2 , and 3 respectively, indices 23 and 32 are replaced by 4 , indices of 13 and 31 are replaced by 5 , and indices of 12 and 21 are replaced by 6 . Contracted notation may be used to represent the fourth-order orientation tensor $a_{i j k l}$ as the $6 \times 6$ matrix $A_{m n}$ in the usual manner as

$$
A_{m n}=\left[\begin{array}{llllll}
A_{11} & A_{12} & A_{13} & A_{14} & A_{15} & A_{16}  \tag{4.1}\\
A_{12} & A_{22} & A_{23} & A_{24} & A_{25} & A_{26} \\
A_{13} & A_{23} & A_{33} & A_{34} & A_{35} & A_{36} \\
A_{14} & A_{24} & A_{34} & A_{44} & A_{45} & A_{46} \\
A_{15} & A_{25} & A_{35} & A_{45} & A_{55} & A_{56} \\
A_{16} & A_{26} & A_{36} & A_{46} & A_{56} & A_{66}
\end{array}\right]=\left[\begin{array}{llllll}
a_{1111} & a_{1122} & a_{1133} & a_{1123} & a_{1113} & a_{1112} \\
a_{1122} & a_{2222} & a_{2233} & a_{2223} & a_{2213} & a_{2212} \\
a_{1133} & a_{2233} & a_{3333} & a_{3323} & a_{3313} & a_{3312} \\
a_{1123} & a_{2223} & a_{3323} & a_{2323} & a_{2313} & a_{2312} \\
a_{1113} & a_{2213} & a_{3313} & a_{2313} & a_{1313} & a_{1312} \\
a_{1112} & a_{2212} & a_{3312} & a_{2312} & a_{1312} & a_{1212}
\end{array}\right]
$$

Employing the symmetries of the orientation tensors from Equation (2.11), the general fourth-order orientation tensor shown in Equation (4.1) has at most 15 independent components (14 recognizing that $a_{i i j j}=a_{i i}=1$ ).

The generalized Hooke's Law relates strain $\varepsilon_{i j}$ to stress $\sigma_{i j}$ through the compliance tensor as shown in Equation (2.43). This expression may be written in contracted notation, recognizing that the compliance tensor is symmetric as

$$
\left\{\begin{array}{l}
\varepsilon_{1}  \tag{4.2}\\
\varepsilon_{2} \\
\varepsilon_{3} \\
\varepsilon_{4} \\
\varepsilon_{5} \\
\varepsilon_{6}
\end{array}\right\}=\left[\begin{array}{cccccc}
S_{11} & S_{12} & S_{13} & S_{14} & S_{15} & S_{16} \\
S_{12} & S_{22} & S_{23} & S_{24} & S_{25} & S_{26} \\
S_{13} & S_{23} & S_{33} & S_{34} & S_{35} & S_{36} \\
S_{14} & S_{24} & S_{34} & S_{44} & S_{45} & S_{46} \\
S_{15} & S_{25} & S_{35} & S_{45} & S_{55} & S_{56} \\
S_{16} & S_{26} & S_{36} & S_{46} & S_{56} & S_{66}
\end{array}\right]\left\{\begin{array}{l}
\sigma_{1} \\
\sigma_{2} \\
\sigma_{3} \\
\sigma_{4} \\
\sigma_{5} \\
\sigma_{6}
\end{array}\right\}
$$

For a general anisotropic material experiencing no planes or directions of material symmetry, there exist 21 independent components of $S_{i j k l}[10]$. As will be shown in
the following chapter, and presented in Jack and Smith [7], all current fourth-order closures assume an orthotropic form, and neglect shear-extensional and shear-shear coupling terms in the stiffness tensor shown in Equation (2.44). The shear-extensional terms, $S_{14}, S_{15}, S_{16}, S_{24}, S_{25}, S_{26}, S_{34}, S_{35}$, and $S_{36}$, represent the response of the normal strains to applied shear stresses, and the shear-shear coupling terms, $S_{45}, S_{46}$, and $S_{56}$, represent the effects of an applied shear stress in one plane to the shear strain response in a different plane. Many applications of engineering materials only account for extension $\left(S_{11}, S_{22}\right.$ and $\left.S_{33}\right)$, shear $\left(S_{44}, S_{55}\right.$ and $\left.S_{66}\right)$, and extension-extension coupling ( $S_{12}, S_{13}$ and $S_{23}$ ) behavior that compose the orthotropic components of the compliance tensor. Polymer composites experience additional coupling phenomena such as shear-shear and shear-extensional coupling which will be referred to as the anisotropic compliance tensor components. An effective closure for polymer process use would not only be capable of predicting the orthotropic terms, but would also effectively represent the other anisotropic coefficients.

Advani and Tucker [6] compute the volume-averaged stiffness tensor $\left\langle C_{i j k l}\right\rangle$, which relates to the compliance tensor $S_{i j k l}$ through the inverse, as

$$
\begin{align*}
\left\langle C_{i j k l}\right\rangle & =B_{1}\left(a_{i j k l}\right)+B_{2}\left(a_{i j} \delta_{k l}+a_{k l} \delta_{i j}\right)+B_{3}\left(a_{i k} \delta_{j l}+a_{i l} \delta_{j k}+a_{j l} \delta_{i k}+a_{j k} \delta_{i l}\right) \\
& +B_{4}\left(\delta_{i j} \delta_{k l}\right)+B_{5}\left(\delta_{i k} \delta_{j l}+\delta_{i l} \delta_{j k}\right) \tag{4.3}
\end{align*}
$$

where the five coefficients $B_{i}$ are obtained from the five independent components of the unidirectional stiffness tensor $\bar{C}_{i j k l}$ discussed in Equation (2.44). A complete derivation of this form for the stiffness tensor is derived in the following chapter. It is sufficient here to use the Advani and Tucker form in Equation (4.3) which has been widely accepted within the literature $[6,27-29,35-37,56,64]$. As discussed in Chapter 2 , the prediction of the material stiffness tensor requires a fundamental understanding
of the stiffness tensor associated with a unidirectional fiber distribution. Tucker and Liang [80] discuss several methods for computing material properties for the unidirectional stiffness tensor $\bar{C}_{i j k l}$ from the individual properties of the fiber and the matrix which include Eshelby's Equivalent Inclusion model [81], the Mori-Tanaka model [82], the Halpin-Tsai equations [70], and several others. Tucker and Liang conclude that the Halpin-Tsai equations as given in Table 2.1 yield reasonable results for the stiffness tensor of short fiber polymer composites.

A fourth-order stiffness tensor lacking any plane of material symmetry is called anisotropic (or triclinic). In the principal reference frame the anisotropic stiffness tensor would have 21 independent non-zero components [10] having the same form as the tensor in Equation (4.2). Conversely, a monoclinic stiffness tensor exhibits a single plane of material symmetry with 13 independent constants, and an orthotropic stiffness tensor with three mutually orthogonal planes of material symmetry has 9 independent components. A transversely isotropic stiffness tensor, with a plane in which properties are identical in all directions, requires only 5 independent components. A complete discussion of material symmetries can be found in Malvern [9] or Jones [10]. Regardless, since $a_{i j k k}=a_{i j}$ and $a_{i i j j}=1$ from Equations (2.13) and (2.15), the volume-averaged values of the material stiffness tensor in Equation (4.3) will clearly experience the same planes of symmetry as those of the fourth-order orientation tensor. Therefore a stiffness tensor computed from Equation (4.3) can never be anisotropic since $a_{i j k l}$ is not anisotropic.

### 4.2.1 Analytical Closures

The fourth-order quadratic closure is the simplest fourth-order closure, and is exact for highly aligned distributions [48] where the fourth-order orientation tensor is
approximated as $a_{i j k l} \simeq a_{i j} a_{k l}$. The quadratic closure is frequently used to form Advani and Tucker's hybrid closure [6] which is written as a linear combination of the quadratic closure and the linear closure of Hand [94]. Unfortunately, the quadratic closure (as well as the hybrid closure [6]) does not obey the previously mentioned symmetry requirements for a fourth-order orientation tensor, i.e. $a_{i j k l} \neq a_{i k j l} \forall i, j, k, l$. Even with that being known, the hybrid closure is still regularly used in many process simulations of short-fibers (see e.g. [36, 37]). The quadratic and hybrid closures will not be discussed further in this chapter since they do not obey the symmetries of an orientation tensor. More recently, the orthotropic smooth closure of Cintra and Tucker [27] was shown to give exact results for uniaxial, random in-space and random in-plane orientations, but experiences poor behavior in flow simulations. Dupret and Verleye [30] present an analytical 2-D Natural closure, but have yet to present a comparable analytical form for their 3-D closure. It is noted here, that although the neural-network closure of Schache [57] and Smith et al. [58] is not analytic, it is not considered in the present work because the closure is not objective since it is unclear whether the principal axis of the fourth-order orientation tensor correspond with those of the second-order orientation tensor (see e.g. [27]). As discussed by Schache, further work may be undertaken that will allow the neural-network closure to satisfy the objective criterion, at which point it will be worthwhile to also include in the following analysis.

### 4.2.2 Eigenvalue-Based Closures

There exist several orthotropic closures of the fourth-order orientation tensor $[27,33,35,53]$. These closures are orthotropic in nature and assume that the principal directions of the second-order orientation tensor define the planes of material
symmetry of the fourth-order orientation tensor. In the principal frame of the secondorder orientation tensor, the fourth-order orientation tensor is explicitly assumed to be orthotropic and is represented as

$$
A_{(m n)}=\left[\begin{array}{cccccc}
A_{(11)} & A_{(12)} & A_{(13)} & 0 & 0 & 0  \tag{4.4}\\
A_{(12)} & A_{(22)} & A_{(23)} & 0 & 0 & 0 \\
A_{(13)} & A_{(23)} & A_{(33)} & 0 & 0 & 0 \\
0 & 0 & 0 & A_{(44)} & 0 & 0 \\
0 & 0 & 0 & 0 & A_{(55)} & 0 \\
0 & 0 & 0 & 0 & 0 & A_{(66)}
\end{array}\right]
$$

The subscripted $(\cdots)$ notation is employed to designate that a tensor is in the principal reference frame of $a_{i j}$, and note that all of the zero components of the matrix in Equation (4.4) may be written in tensor form as $a_{(i i i j)}$ or $a_{(i i j k)}$ (no sum on $i, i \neq j$, $i \neq k$, and $j \neq k)$. For example $A_{(14)}=a_{(1123)}=0, A_{(15)}=a_{(1113)}=0$, etc.

Equation (4.4) may be rewritten as a function of six independent components by applying the symmetries of the fourth-order orientation tensor, $A_{(12)}=A_{(66)}$, $A_{(13)}=A_{(55)}$, and $A_{(23)}=A_{(44)}$. Then using the normalization property in Equation (2.13), $A_{(66)}$ is solved in terms of the other components as $A_{(66)}=1-A_{(11)}-2 A_{(55)}-$ $2 A_{(44)}-A_{(22)}-A_{(33)}$. Therefore, an orientation tensor satisfying Equation (4.4) can be written as a function of five independent components (c.f. Cintra and Tucker [27]), the same number of components as that of a transversely isotropic stiffness tensor. Orthotropic closures (e.g. [27] and [33]) satisfying Equation (4.4) explicitly neglect the orientation tensor components that contribute to the shear-extensional coupling terms by setting $A_{(14)}, A_{(15)}, A_{(16)}, A_{(24)}, A_{(25)}, A_{(26)}, A_{(34)}, A_{(35)}$, and $A_{(36)}$ each equal to zero. Orthotropic closures also explicitly neglect the components of $a_{(i j k l)}$ that contribute to the shear-shear coupling terms by setting the components $A_{(45)}$, $A_{(46)}$, and $A_{(56)}$ to zero.

### 4.2.3 Invariant Based Closures

Existing invariant based closures of $a_{i j k l}$ are all written as a general expression of a fully symmetric fourth-order orientation tensor in terms of $a_{i j}$ and the unit tensor $\delta_{i j}[6,30,31,56]$ as

$$
\begin{align*}
a_{i j k l}= & \beta_{1} S\left(\delta_{i j} \delta_{k l}\right)+\beta_{2} S\left(\delta_{i j} a_{k l}\right)+\beta_{3} S\left(a_{i j} a_{k l}\right)+\beta_{4} S\left(\delta_{i j} a_{k m} a_{m l}\right) \\
& +\beta_{5} S\left(a_{i j} a_{k m} a_{m l}\right)+\beta_{6} S\left(a_{i m} a_{m j} a_{k n} a_{n l}\right) \tag{4.5}
\end{align*}
$$

where $\beta_{i}, i=1,2, \ldots, 6$ are functions of the invariants of the second-order orientation tensor $a_{i j}$. The symmetric operator $S\left(T_{i j k l}\right)$ is employed to form invariant closures where $S\left(T_{i j k l}\right)$ is the symmetric part of its argument

$$
\begin{equation*}
S\left(T_{i j k l}\right)=\frac{1}{24}\left(T_{i j k l}+T_{j i k l}+\cdots(24 \text { total terms })\right) \tag{4.6}
\end{equation*}
$$

Equation (4.5) describes the linear closure [6], the natural closure [30], the fully symmetric (FSQ) [31] and the invariant based orthotropic fitted (IBOF) [56] closures, with the differences occurring in the formation of $\beta_{i}$. For example, the linear closure is formed by setting $\beta_{1}=-3 / 35, \beta_{2}=6 / 7$ and $\beta_{3}=\beta_{4}=\beta_{5}=\beta_{6}=0$. The natural, FSQ and IBOF closures each define $\beta_{1}$ through $\beta_{6}$ as functions of the invariants of the second-order tensor $a_{i j}$ (see e.g. [30,31,56]). The more recent IBOF [56] closure yields an accurate description of the second-order orientation tensor in typical flow conditions over a range of fiber interaction coefficients, and removes the singularity issues inherent in the natural closure [56, 60], whereas the FSQ has been shown to yield realizable behavior in Couette flow [95].

To explore material symmetries for invariant closures, the frame of reference is taken to be the principal frame of the second-order orientation tensor where the only three non-zero terms of $a_{i j}$ are the principal values, designated here as $a_{(11)}, a_{(22)}$,
and $a_{(33)}$, where $(\cdots)$ designates the second-order tensor components are given in the principal reference frame of $a_{i j}$. Note that if the components $a_{i i i j}$ and $a_{i i j k}$ (no sum on $i, i \neq j, i \neq k, j \neq k)$ obtained from Equation (4.5) can be shown to equate to zero in the principal frame of $a_{i j}$, then the invariant based closures will also yield orthotropic material properties as do the orthotropic closures. Chung and Kwon [56] recognize the IBOF satisfies the orthotropic assumption, however, the following discussion is undertaken to clarify the material symmetries of closures, both current and future.

To prove all fourth-order invariant based closures are orthotropic, consider any set of second-order tensors $b_{i j}, c_{i j}$ viewed in an appropriate reference frame such that $b_{i j}=0, c_{i j}=0 \quad \forall i \neq j$, where $b_{i j}$ and $c_{i j}$ may be nonzero when $i=j$. Notice, for example, the second-order tensors $a_{i j}$ and $\delta_{i j}$ are of this form. Then $b_{i k} c_{k j}=0, \forall i \neq j$ (sum on $k$ ) since for each $k=1,2$ or $3, b_{i k} c_{k j}=0\left(\right.$ no sum on $k$ ) where either $b_{i k}=0$ or $c_{k j}=0$. Using mathematical induction [96], assume that $b_{i k} c_{k l} \cdots c_{\alpha \beta} c_{\beta \gamma}=0, \forall$ $\gamma \neq i$ (no sum on $\gamma$, sum on $k, l, \ldots, \alpha, \beta$ ). Then take $b_{i k} c_{k l} \cdots c_{\alpha \beta} c_{\beta \gamma} c_{\gamma j}$. When $\gamma \neq i$ this expression is zero by the preceding inductive step, and when $\gamma=i$ then $c_{\gamma j}=c_{i j}=0$ by the above hypothesis. Therefore, by induction $b_{i k} c_{k l} \cdots c_{\alpha \beta} c_{\beta \gamma} c_{\gamma j}=0$. Similarly this will hold for any product of second-order tensors satisfying the condition imposed on $b_{i j}$ and $c_{i j}$ above. Therefore, each of the terms $a_{i i i j}$ and $a_{i i j k}$ (no sum on $i, i \neq j, i \neq k, j \neq k)$ obtained from the invariant based closure of Equation (4.5) in the principal frame of the second-order orientation tensor are zero since each term obtained from the symmetric operators can be written as a product of secondorder tensors which satisfy the property $\delta_{i j}, a_{i j}=0, \forall i \neq j$ in the principal frame of $a_{i j}$. Note that if higher-order terms formed as products of $a_{i j}$ were included in Equation (4.5) (i.e., terms such as $\left.S\left(\left(a_{i \alpha} \cdots a_{\beta j}\right)\left(a_{k \gamma} \cdots a_{\eta l}\right)\right)\right)$, they too will have
zero contribution, in the principal frame, to the orientation tensor components $a_{i i i j}$ and $a_{i i j k}$ (no sum on $i, i \neq j, i \neq k, j \neq k$ ) by the same argument. Therefore, all current and/or future fourth-order closures which can be written in the form of Equation (4.5), or as an expansion of any of the products in Equation (4.5), will yield $a_{i i i j}=a_{i i j k}=0($ no sum on $i, i \neq j, i \neq k, j \neq k)$ in the principal frame of $a_{i j}$, thereby yielding an orthotropic tensor (cf. Equation (4.4)).

### 4.2.4 Sixth-Order Closures

Only a few works related to sixth-order closures appear in the literature $[6,38,39,52$, 61]. Altan et al. [61] employed a sixth-order quadratic closure proposed by Advani and Tucker [6] for dilute suspensions of fibers. Unfortunately, the quadratic closure has since been shown to overestimate fiber alignment in flows characteristic of industrial applications [60]. For aligned states that significantly differ from random in space, the sixth-order linear closure fails to remain within the physical range (see e.g. [39] and the preceding chapter). The sixth-order hybrid closure of Advani and Tucker [6] and the sixth-order quadratic closure overestimate the actual alignment of fibers [60], and both fail to satisfy the symmetry requirements imposed by the commutative property in the formation of the orientation tensor, e.g. Equation (2.11). Conversely, the sixth-order eigenvalue based closure $\mathrm{EBF}_{6}$ [38] from Equation (3.2), the sixth-order invariant based closure $\mathrm{IBF}_{6}$ [39] form Equation (3.3), and the sixth-order invariant based fitted closure $\mathrm{INV}_{6}[38]$ of Equation (3.15) satisfy the symmetry conditions, with the $\mathrm{INV}_{6}$ providing a fully symmetric sixth-order orientation tensor as a function of the fourth-order orientation tensor.

The $\mathrm{EBF}_{6}$ explicitly assumes an orthotropic form for the fourth-order orientation tensor (see e.g. Jack and Smith [39]) and therefore will not be considered further.

Alternatively, the $\mathrm{IBF}_{6}$ and the $\mathrm{INV}_{6}$ make no explicit assumptions about the form of $a_{i j k l}$ which is obtained from $a_{i j k l m n}$ using the normalization condition $a_{i j k l}=a_{i j k l m m}$. The sixth-order linear closure, along with the $\mathrm{IBF}_{6}$ and $\mathrm{INV}_{6}$ closures given in Equations (3.3) and (3.15), respectively, are all contained within the function

$$
\begin{align*}
a_{i j k l m n} & =\beta_{1} S\left(\delta_{i j} \delta_{k l} \delta_{m n}\right)+\beta_{2} S\left(\delta_{i j} \delta_{k l} a_{m n}\right)+\beta_{3} S\left(\delta_{i j} \delta_{k l} a_{m p} a_{p n}\right)+\beta_{4} S\left(\delta_{i j} a_{k l} a_{m n}\right) \\
& +\beta_{5} S\left(\delta_{i j} a_{k l} a_{m p} a_{p n}\right)+\beta_{6} S\left(\delta_{i j} a_{k p} a_{p l} a_{m q} a_{q n}\right)+\beta_{7} S\left(a_{i j} a_{k l} a_{m n}\right) \\
& +\beta_{8} S\left(a_{i j} a_{k l} a_{m p} a_{p n}\right)+\beta_{9} S\left(a_{i j} a_{k p} a_{p l} a_{m q} a_{q n}\right)+\beta_{10} S\left(a_{i p} a_{p j} a_{k q} a_{q l} a_{m r} a_{r n}\right) \\
& +\beta_{11} S\left(\delta_{i j} a_{k l m n}\right)+\beta_{12} S\left(a_{i j} a_{k l m n}\right)+\beta_{13} S\left(a_{i p} a_{p j} a_{k l m n}\right) \tag{4.7}
\end{align*}
$$

where the operator $S$ represents the symmetric part of its argument discussed in Equations (3.17) and (3.18). For the $\mathrm{IBF}_{6}, \beta_{1}$ through $\beta_{10}$ are functions of the secondorder orientation tensor invariants, and $\beta_{11}=\beta_{12}=\beta_{13}=0$. For $\mathrm{INV}_{6}, \beta_{1}$ through $\beta_{13}$ are functions of the fourth-order orientation tensor invariants, and in general are non-zero (see e.g. Jack and Smith [39]). Observe, the $\mathrm{IBF}_{6}$ is a sixth-order closure that is a function of the second-order tensor recognizing that each $a_{i j}$ in Equation (4.7) can be replaced by $a_{i j k k}$ (sum on $k$ ) from Equation (2.13), whereas the $\mathrm{INV}_{6}$ is a function of the fourth-order orientation tensor.

It follows from the normalization condition in Equation (2.13) that the coefficients of $\beta_{1}$ through $\beta_{10}$ that contribute to the shear-extensional coupling terms $a_{(i i i j)}$ (no sum on $i, i \neq j$ ) in Equation (4.7) are each zero in the principal frame from arguments provided above. Similarly it can be shown that all $a_{(i i j k)}$ (no sum on $i, i \neq j, i \neq k$, $j \neq k)$ obtained from the $\mathrm{IBF}_{6}$ closure, through the application of Equation (2.13), are also zero. Therefore, $\mathrm{IBF}_{6}$ yields a material representation no more general than orthotropic. Alternatively, the tensor obtained from the symmetric operator that multiplies $\beta_{11}$ in Equation (4.7) yields the shear-extensional coupling components
$a_{1112 k k}($ sum on $k$ ) as

$$
\begin{equation*}
S\left(\delta_{11} a_{12 k k}\right)=\frac{1}{15}\left(a_{1112} \delta_{k k}+6 a_{112 k} \delta_{1 k}+3 a_{12 k k} \delta_{11}+2 a_{111 k} \delta_{2 k}+3 a_{11 k k} \delta_{12}\right) \tag{4.8}
\end{equation*}
$$

Since $\delta_{k k}=3, a_{112 k} \delta_{1 k}=a_{1112}$, and $a_{12 k k} \delta_{11}=a_{12}$, Equation (4.8) may be simplified to the following form

$$
\begin{equation*}
S\left(\delta_{11} a_{12 k k}\right)=\frac{1}{15}\left(11 a_{1112}+3 a_{12}\right) \tag{4.9}
\end{equation*}
$$

Considering this result in the principal frame where $a_{(12)}=0$ it is possible to have $a_{(1112)}$ non-zero, even during simple flow conditions. Similarly, other values of $a_{(i i i j m m)}=a_{(i i i j)}$ and $a_{(i i j k m m)}=a_{(i i j k)}$ (no sum on $i, i \neq j, i \neq k$, and $j \neq k$ ) obtained from the symmetric operator associated with $\beta_{11}$ can be shown to yield non-zero results in the principal frame. Similar arguments demonstrate that the symmetric operator multiplying $\beta_{12}$ and $\beta_{13}$ in Equation (4.7) yield shear-extensional and shear-shear coupling terms that are not necessarily zero. Therefore, the $I N V_{6}$ closure yields fourth-order orientation tensors that are not limited by the orthotropic assumption of other sixth-order closures and all objective fourth-order closures.

### 4.3 Numerical Examples of Mechanical Property Predictions

To illustrate the effect of material symmetries imposed by a tensor closure on computed mechanical properties, a homogeneous flow is first considered starting from an isotropic fiber orientation state. The properties to represent a typical fiber reinforced thermoplastic are given as [80]

$$
\begin{align*}
E_{f} & =30 \times 10^{9} \mathrm{~Pa} & \nu_{f} & =0.20 \\
E_{m} & =1 \times 10^{9} \mathrm{~Pa} & \nu_{m} & =0.38  \tag{4.10}\\
a_{r} & =10 & V_{f} & =0.2
\end{align*}
$$

where $V_{f}$ is the volume fraction of fibers. In this analysis, the material stiffness tensor $\left\langle C_{i j k l}\right\rangle$ is computed from Equation (4.3) as discussed in [6] and [71], with the unidirectional stiffness tensor $\bar{C}_{i j k l}$ computed from the Halpin-Tsai equations [70] in Table 2.1.

The first flow considered is uniaxial elongational flow with $v_{1}=2 G x_{1}, v_{2}=-G x_{2}$, and $v_{3}=-G x_{3}$. For uniaxial elongational flow beginning from an initially isotropic orientation state, components such as $a_{i j}(i \neq j)$ and $a_{i i i j}$ and $a_{i i j k}$ (no sum on $i$, $i \neq j, i \neq k, j \neq k)$ that begin at zero will remain zero. Therefore the stiffness tensor from Equation (4.3) will begin as transversely isotropic (actually isotropic, a subset of transversely isotropic) and remain transversely isotropic. Therefore, in predicting material properties both the orthotropic and invariant based fourth-order and sixth-order closures will yield stiffness tensors with identical material symmetries.

The second example considered is simple shear flow with velocity components $v_{1}=G x_{3}$ and $v_{2}=v_{3}=0$. In this analysis, the distribution function evolution (DFE) for $\psi(\theta, \phi, t)$ is computed with the control volume method of Bay [23] with an interaction coefficient of $C_{I}=10^{-2}$. This example is the same as that used in Cintra and Tucker [27] who presented the simple shear case to demonstrate that fourth-order closures cannot "... reproduce this non-orthotropic part of the fourth-order tensor and still be objective."

Selected non-orthotropic stiffness tensor components for simple shear flow are plotted in Figure 4.1 from DFE results, and orientation tensor flow evolutions using the fourth-order orthotropic closure (ORT) of VerWeyst et al. [53], the invariant based closure (IBOF) of Chung and Kwon [56], and the sixth-order invariant based closure $\mathrm{INV}_{6}$ of Jack and Smith [75]. Results for the stiffness tensor are given in contracted
notation. Clearly in the principal frame the shear-extensional coupling terms $C_{(25)}$ and $C_{(35)}$ and the shear-shear coupling term $C_{(46)}$ for this flow are non-zero. Observe the fourth-order closures set each of these terms to zero, whereas the sixth-order closure is able to reasonably represent the non-orthotropic material behavior. It must be noted that the non-orthotropic terms are relatively small in comparison to the orthotropic terms of the stiffness tensor, several of which are given in Figure 4.2. Observe that $C_{(66)}$ is nearly 5 times larger than $C_{(15)}$, whereas $C_{(11)}$ is nearly 400 times larger than $C_{(15)}$ at steady state. It is worthwhile to also note that the sixth-order closure is able to more accurately represent the stiffness tensor components throughout the flow history than the two fourth-order closures investigated. Percentage error results for the orthotropic components are given in Figure 4.3 and demonstrate the increased accuracy of the sixth-order closure. The percentage error remains equal to or less than $1 \%$ throughout the flow history, whereas the ORT closure has more than a $4 \%$ error in the shearing component, $C_{(66)}$ for much of the flow history.

The third example is that of flow near a pin gate $[27,56]$. Unlike homogeneous flows, velocity components in this flow are a function of radial position and gap height as discussed in Equations (3.13) and (3.14). For small radii the flow is dominated by out-of-plane stretching, and as $r$ increases the flow is dominated by shearing which causes the fibers to orient in the radial direction. Beginning from an initially isotropic fiber distribution, the orientation distribution function $\psi(\theta, \phi, t)$ is computed for a gap height of $\frac{z}{b}=\frac{5}{10}$ with an interaction coefficient of $C_{I}=10^{-2}$. Selected shear-shear and shear-extensional coupling components of the stiffness tensor are plotted in Figure 4.4. Observe that as in the simple-shear example, the orthotropic closures again set these terms to zero, whereas the sixth-order closure captures the non-orthotropic


Figure 4.1: Selected non-orthotropic tensor components of $C_{(i j)}$ for Simple Shear flow with $C_{I}=10^{-2}$.


Figure 4.2: Selected orthotropic tensor components of $C_{(i j)}$ for Simple Shear flow with $C_{I}=10^{-2}$ 。
behavior. In comparison to the orthotropic terms given in Figure 4.5, these shearshear and shear-extensional terms appear small with the $C_{(15)}$ term being nearly 5 times smaller than the $C_{(66)}$ component at steady state. Again, the sixth-order closure better represents the orthotropic components than the fourth-order closures. Figure 4.6 provides the percentage error of the ORT and the $\mathrm{INV}_{6}$ closure in representing the orthotropic stiffness tensor components. Observe, as with the simple shear example, the $\mathrm{INV}_{6}$ has less than half the percentage error as does the ORT closure.

### 4.4 Remarks on Closure Effects on Mechanical Properties

Current objective fourth-order closures are limited, by their construction, to an orthotropic tensor representation and neglect shear-extensional coupling and shearshear coupling effects. This limitation will prevent short-fiber polymer composite models, polymer crystalline models and other models which employ similar fourthorder closure techniques from representing material behavior more complex than orthotropic. The significance of this limitation may become more pronounced based upon the initial orientation and should be investigated further. Different conclusions may also be reached if a diffusion model other than that provided by the FolgarTucker model were used. To avoid the orthotropic limitation it was demonstrated that sixth-order closures can represent shear-extensional and shear-shear coupling behavior. Both the fourth-order ORT and the sixth-order INV $_{6}$ closure predict the extensional, shear, and extension-extension coupling terms of the stiffness tensor with reasonable accuracy. The $\mathrm{INV}_{6}$ yields more accurate results by more than a factor of 2 throughout much of the flow history presented for the orthotropic components


Figure 4.3: Percent Error in Select Components of Stiffness Tensor $C_{(i j)}$ for Simple Shear Flow with $C_{I}=10^{-2}$.


Figure 4.4: Selected non-orthotropic tensor components of $C_{(i j)}$ for center-gated disk flow with $C_{I}=10^{-2}$ at $\frac{z}{b}=\frac{5}{10}$.


Figure 4.5: Selected orthotropic tensor components of $C_{(i j)}$ for center-gated disk flow with $C_{I}=10^{-2}$ at $\frac{z}{b}=\frac{5}{10}$.


Figure 4.6: Percent Error in Select Components of Stiffness Tensor $C_{(i j)}$ for centergated disk flow with $C_{I}=10^{-2}$ at $\frac{z}{b}=\frac{5}{10}$.
of the stiffness tensor. Additionally, the $\mathrm{INV}_{6}$ does not experience the orthotropic limitation experienced by all existing objective orthotropic closures, and presents a reasonable representation of the anisotropic components of the stiffness tensor. Although results for the anisotropic components are not as accurate as those for the orthotropic components, the $\mathrm{INV}_{6}$ demonstrates that sixth-order closures are able to predict, with reasonable accuracy, an anisotropic stiffness tensor.

## CHAPTER 5

## ELASTIC PROPERTIES OF SHORT-FIBER POLYMER COMPOSITES

Current methods for predicting the elastic properties of short-fiber polymer composites are derived from fiber orientation tensors. These calculations are based on the orientation average of a transversely isotropic stiffness tensor and have yet to include a quantitative measure of property variation. Recognizing the statistical nature of fiber orientations within the composite commonly defined through the fiber orientation probability distribution function, analytical expressions are developed here to predict both the expectation and variance of the material stiffness tensor from the probability distribution function of fibers. As such, the fiber distribution function is expanded through the Laplace series of complex spherical harmonics. Results presented here and in Jack and Smith [79] demonstrate that the expectation of the material stiffness tensor is a function of orientation tensors up through the fourth-order and the variance requires orientation tensors up through eighth-order.

The analytic expressions developed are validated through the method of MonteCarlo where sample sets are generated from statistically independent unidirectional samples belonging to the fiber orientation probability distribution function with the Accept-Reject Generation Algorithm. Analytic predictions for sample sets of fibers from both an analytic fiber probability distribution function and the industrially relevant case of center-gated disk flow for concentrated suspensions of fibers are developed through the Central Limit Theorem. Results from the distribution of the material stiffness tensor obtained through the Central-Limit Theorem presented here and in

Jack and Smith [78] correspond with results acquired through Monte-Carlo integrations and validate the analytic expressions for material expectation and variance.

### 5.1 Analytical Forms for Expectation and Variance of the Material Property Tensor

Due to the overwhelming computational burden for industrial applications to numerically solve the fiber orientation probability distribution function $\psi(\theta, \phi)$ for complex part geometries, the method of orientation tensors introduced by Advani and Tucker [6] is often employed. As a result, little attention has been given to evaluating short fiber composite elastic properties from the fiber distribution function itself. The Advani and Tucker method to predict material stiffness behavior from orientation tensors is based on the volume average of a transversely isotropic stiffness tensor [6]. The class of objective fourth-order closures discussed in Equation (2.37) either explicitly or implicitly assume a transversely isotropic form (see e.g. Equations (4.4) and (4.5) and Jack and Smith [7]), and therefore the Advani and Tucker model for predicting an orientation-averaged material stiffness tensor is sufficient for fourth-order closure applications. Conversely, both the sixth-order closure of Altan et al. [52] as well as the sixth-order $\mathrm{INV}_{6}$ closure [38] given in Equation (3.15) are not constrained by the transversely isotropic assumption inherent to the objective fourth-order closures [7], therefore the Advani and Tucker derivation warrants further investigation. Additionally, there exists no complete derivation of an analytic method to represent the statistical nature of the material stiffness tensor.

In this section the Laplace series of complex spherical harmonics presented in Equation (2.51) is employed to expand the fiber orientation probability distribution
function assuming only that the fiber distribution is symmetric about a single axis. This approach makes no assumptions regarding the form of the orientation tensors beyond their inherent symmetry, thereby providing a means for computing material behavior from orientation tensors, and avoids the issue of a closure selection. It will be shown that the proposed method to predict the expectation value can be written in the same form as the Advani and Tucker [6] model. Additionally, an analytic form is introduced to evaluate the variance of the stiffness tensor's distribution, which requires orientation tensors through the eighth-order.

### 5.1.1 Material Stiffness Tensor Expectation Value

The expectation value (referred to elsewhere as the mean or orientation average [6]) of the material stiffness tensor is formulated from the non-correlated aggregate of unidirectional fibers defined as the first moment of the fiber orientation probability distribution function $\psi(\theta, \phi)$ as [97]

$$
\begin{equation*}
\left\langle C_{i j k l}\right\rangle=\oint_{\mathbb{S}^{2}} Q_{p i}(\theta, \phi) Q_{q j}(\theta, \phi) Q_{r k}(\theta, \phi) Q_{s l}(\theta, \phi) \bar{C}_{p q r s} \psi(\theta, \phi) \mathrm{d} \mathbb{S} \tag{5.1}
\end{equation*}
$$

where $\left\langle C_{i j k l}\right\rangle$ is the expectation value of the stiffness tensor, $\bar{C}_{p q r s}$ is the unidirectional stiffness tensor represented by the stiffness matrix in Equation (2.44), and $Q_{p i}(\theta, \phi) Q_{q j}(\theta, \phi) Q_{r k}(\theta, \phi) Q_{s l}(\theta, \phi) \bar{C}_{p q r s}$ is the unidirectional stiffness tensor aligned along the angles $(\theta, \phi)$ given in Figure 2.1. The rotation tensor $\mathbf{Q}(\theta, \phi)$ in Equation (5.1) is defined as

$$
\mathbf{Q}(\theta, \phi)=\left[\begin{array}{ccc}
\sin \theta \cos \phi & \sin \theta \sin \phi & \cos \theta  \tag{5.2}\\
-\sin \phi & \cos \phi & 0 \\
-\cos \theta \cos \phi & -\cos \theta \sin \phi & \sin \theta
\end{array}\right]
$$

It is essential to note that Equation (5.1) does not take into account the spatial correlation of fibers with those nearby since the expectation is written in terms of
the uncorrelated single fiber properties. In other words fibers must have a significant enough separation such that they do not interact. This assumption is too stringent, therefore it is assumed that the fibers are fully mixed and have no short range order as was assumed in Advani and Tucker [6] for the development of the orientation averaging procedure.

Equation (5.1) may be used to obtain orientation averaged elastic properties for any sufficiently smooth fiber orientation distribution function $\psi(\theta, \phi)$. The distribution $\psi(\theta, \phi)$ is written in terms of the Laplace series expansion of Equation (2.51) to obtain the expectation value of the stiffness tensor in Equation (5.1) as the series

$$
\begin{align*}
\left\langle C_{i j k l}\right\rangle & =\oint_{\mathbb{S}^{2}} Q_{p i}(\theta, \phi) Q_{q j}(\theta, \phi) Q_{r k}(\theta, \phi) Q_{s l}(\theta, \phi) \bar{C}_{p q r s} \sum_{l=0}^{\infty} \sum_{m=0}^{l} \beta_{l}^{m}(\theta, \phi) \mathrm{dS} \\
& =\sum_{l=0}^{\infty} \oint_{\mathbb{S}^{2}} Q_{p i}(\theta, \phi) Q_{q j}(\theta, \phi) Q_{r k}(\theta, \phi) Q_{s l}(\theta, \phi) \bar{C}_{p q r s} \sum_{m=0}^{l} \beta_{l}^{m}(\theta, \phi) \mathrm{dS} \\
& =\left\langle C_{i j k l}\right\rangle_{0}+\left\langle C_{i j k l}\right\rangle_{2}+\cdots+\left\langle C_{i j k l}\right\rangle_{2 N}+\cdots \tag{5.3}
\end{align*}
$$

where $N \in\{0\} \cup \mathbb{N}$, and for each $N,\left\langle C_{i j k l}\right\rangle_{2 N}$ is defined as

$$
\begin{equation*}
\left\langle C_{i j k l}\right\rangle_{2 N} \equiv \oint_{\mathbb{S}^{2}} Q_{p i}(\theta, \phi) Q_{q j}(\theta, \phi) Q_{r k}(\theta, \phi) Q_{s l}(\theta, \phi) \bar{C}_{p q r s}\left(\sum_{m=0}^{2 N} \beta_{2 N}^{m}(\theta, \phi)\right) \mathrm{d} \mathbb{S}(5 \tag{5.4}
\end{equation*}
$$

for $\beta_{2 N}^{m}(\theta, \phi)$ given in Equation (2.59). All odd-ordered terms $\left\langle C_{i j k l}\right\rangle_{2 N+1}, N \in$ $\{0\} \cup \mathbb{N}$, are zero since $\beta_{2 N+1}^{m}(\theta, \phi)=0$ for all values of $N$ for the symmetric fiber distribution function.

Consider for example the zeroth-order term, $\left\langle C_{i j k l}\right\rangle_{0}$, from the Laplace series expansion along with $\bar{C}_{p q r s}$ from Equation (2.44) and $\beta_{0}^{0}(\theta, \phi)=\frac{1}{4 \pi}$ given in Equation
(2.60). The expression for $\left\langle C_{1111}\right\rangle_{0}$ in this case is given as

$$
\begin{align*}
\left\langle C_{1111}\right\rangle_{0}= & \oint_{\mathbb{S}^{2}} Q_{p 1}(\theta, \phi) Q_{q 1}(\theta, \phi) Q_{r 1}(\theta, \phi) Q_{s 1}(\theta, \phi) \bar{C}_{p q r s}\left(\frac{1}{4 \pi}\right) \mathrm{d} \mathbb{S} \\
= & \frac{1}{4 \pi} \oint_{\mathbb{S}^{2}}\left(\cos ^{4} \phi\left(\bar{C}_{22} \cos ^{4} \theta+2\left(\bar{C}_{12}+2 \bar{C}_{55}\right) \cos ^{2} \theta \sin ^{2} \theta+\bar{C}_{11} \sin ^{4} \theta\right)\right. \\
& \left.+2 \cos ^{2} \phi\left(\bar{C}_{22} \cos ^{2} \theta+\left(\bar{C}_{12}+2 \bar{C}_{55}\right) \sin ^{2} \theta\right) \sin ^{2} \phi+\bar{C}_{22} \sin ^{4} \phi\right) \mathrm{d} \mathbb{S} \\
= & \frac{1}{15}\left(3 \bar{C}_{11}+4 \bar{C}_{12}+8 \bar{C}_{22}+8 \bar{C}_{55}\right) \tag{5.5}
\end{align*}
$$

The remaining components for $\left\langle C_{i j k l}\right\rangle_{0}$ are derived in a similar manner and after some simplification are written in contracted notation as

$$
\left[\langle C\rangle_{0}\right]=\left[\begin{array}{cccccc}
\xi_{0} & \eta_{0} & \eta_{0} & 0 & 0 & 0  \tag{5.6}\\
\eta_{0} & \xi_{0} & \eta_{0} & 0 & 0 & 0 \\
\eta_{0} & \eta_{0} & \xi_{0} & 0 & 0 & 0 \\
0 & 0 & 0 & \frac{\xi_{0}-\eta_{0}}{2} & 0 & 0 \\
0 & 0 & 0 & 0 & \frac{\xi_{0}-\eta_{0}}{2} & 0 \\
0 & 0 & 0 & 0 & 0 & \frac{\xi_{0}-\eta_{0}}{2}
\end{array}\right]
$$

where $\left[\langle C\rangle_{0}\right]$ represents the tensor $\left\langle C_{i j k l}\right\rangle_{0}$ in matrix form and the coefficients $\xi_{0}$ and $\eta_{0}$ are given as

$$
\begin{align*}
& \xi_{0}=\frac{1}{15}\left(3 \bar{C}_{11}+4 \bar{C}_{12}+8 \bar{C}_{22}+8 \bar{C}_{55}\right) \\
& \eta_{0}=\frac{1}{15}\left(\bar{C}_{11}+8 \bar{C}_{12}+\bar{C}_{22}+5 \bar{C}_{23}-4 \bar{C}_{55}\right) \tag{5.7}
\end{align*}
$$

As in Equation (2.44), $\bar{C}_{m n}, m, n \in\{1,2, \ldots, 6\}$, is the $(m, n)$ component of the underlying unidirectional stiffness matrix $[C]$. Equation (5.6) is an isotropic stiffness tensor (see e.g. Jones [10]) as one would expect from an isotropic fiber orientation distribution. This fact is obvious recognizing the first term $\beta_{0}^{0}(\theta, \phi)$ in the Laplace series of Equation (2.51) is the full expansion for an isotropic fiber orientation distribution.

The second-order function from the Laplace series expansion of the stiffness tensor expectation $\left\langle C_{i j k l}\right\rangle_{2}$ can be derived in a similar manner from $\sum_{m=0}^{2} \beta_{2}^{m}(\theta, \phi)$ where the functions $\beta_{2}^{m}(\theta, \phi)$ for each $m \in\{0,1,2\}$ are given in Equations (2.61)-(2.63).

After some simplification, the second-order component from the expansion of the material tensor expectation $\left\langle C_{i j k l}\right\rangle_{2}$ in contracted form is given as

$$
\begin{align*}
& {\left[\langle C\rangle_{2}\right]=} \\
& {\left[\begin{array}{cccccc}
\left(3 a_{11}-1\right) \tau_{2} & \left(3 a_{11}+3 a_{22}-2\right) \epsilon_{2} & \left(1-3 a_{22}\right) \epsilon_{2} & 3 a_{23} \epsilon_{2} & a_{13} \xi_{2} & a_{12} \xi_{2} \\
\left(3 a_{11}+3 a_{22}-2\right) \epsilon_{2} & \left(3 a_{22}-1\right) \tau_{2} & \left(1-3 a_{11}\right) \epsilon_{2} & a_{23} \xi_{2} & 3 a_{13} \epsilon_{2} & a_{12} \xi_{2} \\
\left(1-3 a_{22}\right) \epsilon_{2} & \left(1-3 a_{11}\right) \epsilon_{2} & \left(2-3 a_{11}-3 a_{22}\right) \tau_{2} & a_{23} \xi_{2} & a_{13} \xi_{2} & 3 a_{12} \epsilon_{2} \\
3 a_{23} \epsilon_{2} & a_{23} \xi_{2} & a_{23} \xi_{2} & \left(1-3 a_{11}\right) \eta_{2} & 3 a_{12} \eta_{2} & 3 a_{13} \eta_{2} \\
a_{13} \xi_{2} & 3 a_{13} \epsilon_{2} & a_{13} \xi_{2} & 3 a_{12} \eta_{2} & \left(1-3 a_{22}\right) \eta_{2} & 3 a_{23} \eta_{2} \\
a_{12} \xi_{2} & a_{12} \xi_{2} & 3 a_{12} \epsilon_{2} & 3 a_{13} \eta_{2} & 3 a_{23} \eta_{2}\left(3 a_{11}+3 a_{22}-2\right) \eta_{2}
\end{array}\right]} \tag{5.8}
\end{align*}
$$

where the coefficients $\xi_{2}, \eta_{2}, \tau_{2}$, and $\epsilon_{2}$ are defined as

$$
\begin{align*}
& \xi_{2}=\frac{6}{42}\left(3 \bar{C}_{11}+\bar{C}_{12}-4 \bar{C}_{22}+2 \bar{C}_{55}\right) \\
& \eta_{2}=\frac{1}{42}\left(2 \bar{C}_{11}-4 \bar{C}_{12}-5 \bar{C}_{22}+7 \bar{C}_{23}+6 \bar{C}_{55}\right) \\
& \tau_{2}=\frac{2 \xi_{2}}{3} \\
& \epsilon_{2}=\frac{1}{3}\left(\xi_{2}-6 \eta_{2}\right) \tag{5.9}
\end{align*}
$$

Note that when the second-order orientation tensor $a_{i j}$ is viewed in its principal reference frame, $\left[\langle C\rangle_{0}\right]+\left[\langle C\rangle_{2}\right]$ from Equations (5.6) and (5.8) yields a transversely isotropic stiffness tensor (see e.g. Jones [10]). For example in the principal frame of $a_{i j}$, the term $\left[\langle C\rangle_{0}\right]_{15}+\left[\langle C\rangle_{2}\right]_{15}=a_{13} \xi_{2}$ is zero since $a_{13}=0$.

The fourth-order contribution from the Laplace series expansion of the stiffness tensor $\left\langle C_{i j k l}\right\rangle_{4}$ is derived using Equation (5.3) with the fourth-order function of spherical harmonics $\sum_{m=0}^{4} \beta_{4}^{m}(\theta, \phi)$ as given in Equations (2.65)-(2.69). After much simplification, this component of the stiffness tensor expectation value can be written concisely as

$$
\begin{align*}
\left\langle C_{i j k l}\right\rangle_{4}= & \xi_{4}\left(a_{i j k l}-\frac{1}{7}\left(a_{i j} \delta_{k l}+a_{k l} \delta_{i j}+a_{i k} \delta_{j l}+a_{i l} \delta_{j k}+a_{j k} \delta_{i l}+a_{j l} \delta_{i k}\right)\right. \\
& \left.+\frac{1}{35}\left(\delta_{i j} \delta_{k l}+\delta_{i k} \delta_{j l}+\delta_{i l} \delta_{j k}\right)\right) \tag{5.10}
\end{align*}
$$

where

$$
\begin{equation*}
\xi_{4}=\bar{C}_{11}-2 \bar{C}_{12}+\bar{C}_{22}-4 \bar{C}_{55} \tag{5.11}
\end{equation*}
$$

In the principal frame of the second-order orientation tensor, components such as $\left\langle C_{i i i j}\right\rangle_{4}$ and $\left\langle C_{i i j k}\right\rangle_{4}$ (no sum on $i, i \neq j, i \neq k$, and $j \neq k$ ) are, in general, non-zero since the fourth-order orientation tensor components $a_{i i i j}$ and $a_{i i j k}$ (no sum on $i, i \neq j$, $i \neq k$, and $j \neq k)$ yield, in general, non-zero components in the principal frame of the second-order orientation tensor (see e.g. Equation (4.9) and Jack and Smith [7]). Therefore, the expectation of the stiffness tensor component $\left\langle C_{i j k l}\right\rangle_{4}$ may yield a stiffness tensor with fewer planes of material symmetry than a transversely isotropic material and will experience the same planes of symmetry as does the fourth-order expansion of $\psi(\theta, \phi)$.

The Laplace series expansion in Equation (5.3) is an infinite series yielding the terms $\left\langle C_{i j k l}\right\rangle_{2 N}$ for $\{N: N \in \mathbb{N}, N \geq 3\}$. Taking any of the functions $\sum_{m=0}^{2 N} \beta_{2 N}^{m}(\theta, \phi)$ for $N \geq 3$, the order of $\beta_{2 N}^{m}(\theta, \phi)$ composed of the spherical function $Y_{2 N}^{m}(\theta, \phi)$ given in Equation (2.53) is greater in order than any component of the product $Q_{p i}(\theta, \phi) Q_{q j}(\theta, \phi) Q_{r k}(\theta, \phi) Q_{s l}(\theta, \phi)$ which is at most fourth-order, therefore by the orthogonality condition from Equation (2.55), there exist no higher order terms in the expansion for the stiffness expectation value from Equation (5.3) than fourth. Ergo, assuming non-interacting fibers, Equation (5.3) may be written succinctly as

$$
\begin{equation*}
\left\langle C_{i j k l}\right\rangle=\left\langle C_{i j k l}\right\rangle_{0}+\left\langle C_{i j k l}\right\rangle_{2}+\left\langle C_{i j k l}\right\rangle_{4} \tag{5.12}
\end{equation*}
$$

where each of the tensors $\left\langle C_{i j k l}\right\rangle_{0},\left\langle C_{i j k l}\right\rangle_{2}$ and $\left\langle C_{i j k l}\right\rangle_{4}$ are given in Equations (5.6), (5.8) and (5.10), respectively.

The above expression for the stiffness tensor expectation value can be rewritten in the form introduced by Advani and Tucker [6] shown in Equation (4.3) by substituting
the following in Equations (5.6), (5.8) and (5.10)

$$
\left\{\begin{array}{l}
\xi_{0}  \tag{5.13}\\
\eta_{0} \\
\xi_{2} \\
\eta_{2} \\
\xi_{4}
\end{array}\right\}=\left[\begin{array}{ccccc}
\frac{1}{5} & \frac{2}{3} & \frac{4}{3} & 1 & 2 \\
\frac{1}{15} & \frac{2}{3} & 0 & 1 & 0 \\
\frac{3}{7} & 1 & 2 & 0 & 0 \\
\frac{1}{21} & 0 & \frac{1}{3} & 0 & 0 \\
1 & 0 & 0 & 0 & 0
\end{array}\right]\left\{\begin{array}{c}
B_{1} \\
B_{2} \\
B_{3} \\
B_{4} \\
B_{5}
\end{array}\right\}
$$

The form of Equation (5.12) and the Advani and Tucker form in Equation (4.3) for the stiffness tensor expectation value predictions will yield the identical expressions after simplification. For isotropic distributions, the stiffness tensor can be written as $\left\langle C_{i j k l}\right\rangle=\left\langle C_{i j k l}\right\rangle_{0}$, and for distributions that can be written with solely the second-order Laplace series expansion, the expectation of the stiffness tensor is simply $\left\langle C_{i j k l}\right\rangle=\left\langle C_{i j k l}\right\rangle_{0}+\left\langle C_{i j k l}\right\rangle_{2}$.

### 5.1.2 Material Stiffness Tensor Variance

Recognizing the statistical nature of the fiber orientation probability distribution function, there is a need to develop a quantitative measure of material property variation for a given fiber distribution. Continuing in a similar fashion as the above formulation for the expectation value, the assumption is continued whereby fiber interaction only occurs during processing through the fiber distribution evolution equation (see e.g. [11,45]). As such the fiber distribution function $\psi(\theta, \phi)$ is composed of uncorrelated variables $\theta$ and $\phi$ and the variance of the stiffness tensor $\sigma_{i j k l}^{2}$ may be written as the second moment of $\psi(\theta, \phi)$ about the expectation $\left\langle C_{i j k l}\right\rangle[97,98]$ as

$$
\begin{equation*}
\sigma_{i j k l}^{2}=\oint_{\mathbb{S}^{2}}\left(Q_{p i}(\theta, \phi) Q_{q j}(\theta, \phi) Q_{r k}(\theta, \phi) Q_{s l}(\theta, \phi) \bar{C}_{p q r s}-\left\langle C_{i j k l}\right\rangle\right)^{2} \psi(\theta, \phi) \mathrm{d} \mathbb{S} \tag{5.14}
\end{equation*}
$$

Equation (5.14) may be viewed as the variance of the stiffness tensors each computed from an individual fiber in the matrix. In this case, each fiber is defined by the angles $(\theta, \phi)$ taken within a region of a part where the fiber distribution $\psi(\theta, \phi)$ is assumed
continuous. The form of Equation (5.14) implies uncorrelated variables $\theta$ and $\phi$, and remains valid until the density of fibers is such that individual fibers cause changes in either the fiber angle or stress field of neighboring fibers. Correlation will cause, in the laboratory setting, a variance that differs from Equation (5.14) where positive correlations increase the measured variance and negative correlations diminish the measured variance relative to Equation (5.14) (see e.g. [98]).

In Equation (5.14), $\left\langle C_{i j k l}\right\rangle$ is independent of $\theta$ and $\phi$ and may be brought out of the integral simplifying the above equation as

$$
\begin{align*}
\sigma_{i j k l}^{2}= & \oint_{\mathbb{S}^{2}}\left(Q_{p i}(\theta, \phi) Q_{q j}(\theta, \phi) Q_{r k}(\theta, \phi) Q_{s l}(\theta, \phi) \bar{C}_{p q r s}\right)^{2} \psi(\theta, \phi) \mathrm{d} \mathbb{S} \\
& -2\left\langle C_{i j k l}\right\rangle \oint_{\mathbb{S}^{2}} Q_{p i}(\theta, \phi) Q_{q j}(\theta, \phi) Q_{r k}(\theta, \phi) Q_{s l}(\theta, \phi) \bar{C}_{p q r s} \psi(\theta, \phi) \mathrm{d} \mathbb{S} \\
& +\left\langle C_{i j k l}\right\rangle^{2} \oint_{\mathbb{S}^{2}} \psi(\theta, \phi) \mathrm{d} \mathbb{S} \tag{5.15}
\end{align*}
$$

Noting that $\oint_{\mathbb{S}^{2}} Q_{p i}(\theta, \phi) Q_{q j}(\theta, \phi) Q_{r k}(\theta, \phi) Q_{s l}(\theta, \phi) \bar{C}_{p q r s} \psi(\theta, \phi) \mathrm{d} \mathbb{S}=\left\langle C_{i j k l}\right\rangle$ from Equation (5.1) and imposing the normalization property, $\oint_{\mathbb{S}^{2}} \psi(\theta, \phi) \mathrm{d} \mathbb{S}=1$ yields

$$
\begin{equation*}
\sigma_{i j k l}^{2}=\oint_{\mathbb{S}^{2}}\left(Q_{p i}(\theta, \phi) Q_{q j}(\theta, \phi) Q_{r k}(\theta, \phi) Q_{s l}(\theta, \phi) \bar{C}_{p q r s}\right)^{2} \psi(\theta, \phi) \mathrm{d} \mathbb{S}-\left\langle C_{i j k l}\right\rangle^{2} \tag{5.16}
\end{equation*}
$$

This form for the variance of the stiffness tensor $\sigma_{i j k l}^{2}$ requires an eighth-order Laplace series expansion for $\psi(\theta, \phi)$ to fully evaluate the integrand since the square of the products of the rotation tensors will be eighth-order. The terms higher than eighthorder will integrate to zero due to the orthogonality condition of Equation (2.55).

The variance of the components of the stiffness tensor $\sigma_{i j k l}^{2}$ may be written in terms of the orientation tensors using the Laplace series expansion of Equation (2.51) as described below. For example, the variance of the extensional components (see e.g.

Jones [10]) are

$$
\begin{align*}
\sigma_{i i i i}^{2}= & \xi_{4}^{2} a_{i i i i i i i i}+\left(4 \xi_{2} \xi_{4}-\frac{12}{7} \xi_{4}^{2}\right) a_{i i i i i i}+\left(4 \xi_{2}^{2}+2 \xi_{0} \xi_{4}-\frac{100}{21} \xi_{2} \xi_{4}+\frac{222}{245} \xi_{4}^{2}\right) a_{i i i i} \\
& +\left(4 \xi_{0} \xi_{2}-\frac{8}{3} \xi_{2}^{2}-\frac{12}{7} \xi_{0} \xi_{4}+\frac{52}{35} \xi_{2} \xi_{4}-\frac{36}{245} \xi_{4}^{2}\right) a_{i i} \\
& +\frac{\left(105 \xi_{0}-70 \xi_{2}+9 \xi_{4}\right)^{2}}{11025}-\left\langle C_{i i i i}\right\rangle^{2} \tag{5.17}
\end{align*}
$$

where the summation convention on $i$, and similarly for $j$ and $k$, is suspended in Equation (5.17) for $i, j, k \in\{1,2,3\}$, and for the remainder of this section. The form of Equation (5.17) provides the variance of the three stiffness tensor components $\sigma_{1111}^{2}$, $\sigma_{2222}^{2}$ and $\sigma_{3333}^{2}$. Similarly, the variance of the shearing components of the stiffness tensor, $\sigma_{1212}^{2}, \sigma_{1221}^{2}, \sigma_{2121}^{2}, \sigma_{1313}^{2}, \ldots$ may be derived from Equation (5.16) as

$$
\begin{align*}
\sigma_{i j i j}^{2}= & \xi_{4}^{2} a_{i i i i j j j j}+\left(6 \eta_{2} \xi_{4}-\frac{2}{7} \xi_{4}^{2}\right)\left(a_{i i i i j j}+a_{i i j j j j}\right) \\
& +\left(\xi_{0} \xi_{4}-\eta_{0} \xi_{4}-4 \eta_{2} \xi_{4}+\frac{2}{35} \xi_{4}^{2}\right) a_{i i j j}+\left(3 \eta_{2}-\frac{1}{7} \xi_{4}\right)^{2}\left(a_{i i i i}+2 a_{i i j j}+a_{j j j j}\right) \\
& -\frac{1}{245}\left(35\left(\eta_{0}+4 \eta_{2}-\xi_{0}\right)-2 \xi_{4}\right)\left(21 \eta_{2}-\xi_{4}\right)\left(a_{i i}+a_{j j}\right) \\
& +\left(-\frac{1}{2} \xi_{0}+\frac{1}{2} \eta_{0}+2 \eta_{2}-\frac{1}{35} \xi_{4}\right)^{2}-\left\langle C_{i j i j}\right\rangle^{2} \tag{5.18}
\end{align*}
$$

$\forall i, j \in\{1,2,3\}, i \neq j$. The variance of the extension-extension coupling terms of the stiffness tensor are

$$
\begin{align*}
\sigma_{i i j j}^{2}= & \xi_{4}^{2} a_{i i i i j j j j}+\left(2 \xi_{2} \xi_{4}-12 \eta_{2} \xi_{4}-\frac{2}{7} \xi_{4}^{2}\right)\left(a_{i i i i j j}+a_{i i j j j j}\right) \\
& +\left(2 \eta_{0} \xi_{4}+2 \xi_{2}^{2}-24 \xi_{2} \eta_{2}+72 \eta_{2}^{2}-\frac{40}{21} \xi_{2} \xi_{4}+\frac{80}{7} \eta_{2} \xi_{4}+\frac{24}{245} \xi_{4}^{2}\right) a_{i i j j} \\
& +\left(\xi_{2}-6 \eta_{2}-\frac{1}{7} \xi_{4}\right)^{2}\left(a_{i i i i}+a_{j j j j}\right) \\
& +2\left(\xi_{2}-6 \eta_{2}-\frac{1}{7} \xi_{4}\right)\left(\eta_{0}-\frac{2}{3} \xi_{2}+4 \eta_{2}+\frac{1}{35} \xi_{4}\right)\left(a_{i i}+a_{j j}\right) \\
& +\left(\eta_{0}-\frac{2}{3} \xi_{2}+4 \eta_{2}+\frac{1}{35} \xi_{4}\right)^{2}-\left\langle C_{i i j j}\right\rangle^{2} \tag{5.19}
\end{align*}
$$

$\forall i, j \in\{1,2,3\}, i \neq j$. The variance of the stiffness tensor for the shear-extension coupling terms may be expressed as

$$
\begin{align*}
& \sigma_{i i i j}^{2}=\xi_{4}^{2} a_{i i i i i i j j}+\left(2 \xi_{2} \xi_{4}-\frac{6}{7} \xi_{4}^{2}\right) a_{i i i i j j}+\left(\xi_{2}-\frac{3}{7} \xi_{4}\right)^{2} a_{i i j j}-\left\langle C_{i i i j}\right\rangle^{2}  \tag{5.20}\\
& \sigma_{i i j k}^{2}=\xi_{4}^{2} a_{i i i i j j k k}+\left(2 \xi_{2} \xi_{4}-12 \eta_{2} \xi_{4}-\frac{2}{7} \xi_{4}^{2}\right) a_{i i j j k k}+\left(\xi_{2}-6 \eta_{2}-\frac{1}{7} \xi_{4}\right)^{2} a_{j j k k}-\left\langle C_{i i j k}\right\rangle^{2}(5.21)
\end{align*}
$$

where $i, j, k \in\{1,2,3\}, i \neq j, i \neq k$, and $j \neq k$ in Equations (5.20) and (5.21). Lastly, the variance of the stiffness tensor for the shear-shear coupling terms are written as

$$
\begin{equation*}
\sigma_{i j i k}^{2}=\xi_{4}^{2} a_{i i i i j j k k}+\left(6 \eta_{2} \xi_{4}-\frac{2}{7} \xi_{4}^{2}\right) a_{i i j j k k}+\left(3 \eta_{2}-\frac{1}{7} \xi_{4}\right)^{2} a_{j j k k}-\left\langle C_{i j i k}\right\rangle^{2} \tag{5.22}
\end{equation*}
$$

Note the orientation tensors up through eighth-order appear in Equations (5.17)(5.22) for the variance of the stiffness tensor. This requirement may pose computational difficulty since accepted closure approximations have only been developed for orientation tensors up through the sixth-order (see e.g. [6, 27, 38, 56]). For industrial applications where only the orientation tensors up to fourth- or sixth-order are known, it may become necessary to develop higher-order orientation tensor approximations in order to use Equations (5.17)-(5.22). At present one might consider using a simple eighth-order quadratic approximation $a_{i j k l m n o p} \simeq a_{i j k l m n} a_{o p}$ in conjunction with any one of the sixth-order closures. It is speculated a simple approximation for $a_{i j k l m n o p}$ similar to the fourth-order linear closure of Hand [94], the sixth-order quadratic closure used by Altan [61], or the sixth-order hybrid closure introduced by Advani [6] may provide a sufficient approximation to the eighth-order orientation tensor. In fiber orientation analysis, the analytic closures have been demonstrated to provide somewhat reasonable results in approximating the higher-order orientation tensors, even though they yield unacceptable errors in the flow equations for the orientation tensors (see e.g. Jack [59] and Jack and Smith [60]).

### 5.1.3 Example Analytic Distributions

Several analytical distribution functions are selected to demonstrate the material property expressions for expectation and variance of the short-fiber composite stiffness tensor presented above. In the following section and in Jack and Smith [78], numerical results are obtained for various fiber distribution functions $\psi(\theta, \phi)$ using the method of Monte-Carlo to verify the analytic forms. It is worthwhile to note in the following examples, conditions where the second- and/or the fourth-order expansion for the stiffness tensor $[\langle C\rangle]$ of Equation (5.12) are zero, such as the case for an isotropic fiber distribution $[\langle C\rangle]_{2}=[\langle C\rangle]_{4}=[\mathbf{0}]$ and for a second-order fiber distribution (i.e., from Equation (2.51), $\alpha_{l}(\theta, \phi)=0$ for $\left.\{l: 2<l, l \in \mathbb{N}\}\right)[\langle C\rangle]_{4}=[\mathbf{0}]$.

### 5.1.3.1 Analytic Fiber Orientation Distribution

The first example considered here is of the analytic fiber orientation probability distribution function

$$
\begin{equation*}
\psi(\theta, \phi)=c \sin ^{2 n} \theta \cos ^{2 n} \phi \tag{5.23}
\end{equation*}
$$

where $c$ is a constant chosen to satisfy the normalization condition on the distribution function $\left(\oint_{\mathbb{S}^{2}} \psi(\theta, \phi) d \mathbb{S}=1\right), \theta$ and $\phi$ are the angles defined in Figure 2.1, and $n \in$ $\{0\} \cup \mathbb{N}$. For $n=0$ the probability of a fiber being oriented near any given angle pair $\left(\theta^{\prime}, \phi^{\prime}\right) \in \mathbb{S}^{2}$ is identical to the probability of a fiber being oriented near any other angle pair as shown in Figure 5.1. As the coefficient $n$ increases, the distribution tends toward alignment along the $x_{1}$ axis corresponding to $(\theta, \phi)=\left(\frac{\pi}{2}, 0\right)$ and $(\theta, \phi)=$ $\left(\frac{\pi}{2}, \pi\right)$. For the case with $n=1$ the distribution function is shown in Figure 5.2, and as $n$ increases the peaks of the distribution become sharper as shown in Figure 5.3 for the case with $n=6$.


Figure 5.1: Isotropic fiber orientation distribution $\psi(\theta, \phi)=\frac{1}{4 \pi}$.


Figure 5.2: Transversely isotropic second-order fiber orientation distribution $\psi(\theta, \phi)=\frac{3}{4 \pi} \sin ^{2} \theta \cos ^{2} \phi$.


Figure 5.3: Transversely isotropic sixth-order fiber orientation distribution $\psi(\theta, \phi)=$ $\frac{13}{4 \pi} \sin ^{12} \theta \cos ^{12} \phi$.

The zeroth-order component $\left\langle C_{i j k l}\right\rangle_{0}$ does not depend on $\psi(\theta, \phi)$ as shown in Equation (5.6). However, the higher-order components of the stiffness tensor expectation are functions of the orientation tensors $a_{i j}$ and $a_{i j k l}$. Analytic expressions for the second-order orientation tensor $a_{i j}$ as a function of $n$ are included in Equation (5.8) to obtain the second-order stiffness tensor component $\left\langle C_{i j k l}\right\rangle_{2}$. After some simplification, $\left\langle C_{i j k l}\right\rangle_{2}$ may be written in contracted form as a function of $n$ as

$$
\left[\langle C\rangle_{2}(n)\right]=\frac{2 n}{3+2 n}\left[\begin{array}{cccccc}
2 \tau_{2} & \epsilon_{2} & \epsilon_{2} & 0 & 0 & 0  \tag{5.24}\\
\epsilon_{2} & -\tau_{2} & -2 \epsilon_{2} & 0 & 0 & 0 \\
\epsilon_{2} & -2 \epsilon_{2} & -\tau_{2} & 0 & 0 & 0 \\
0 & 0 & 0 & -2 \eta_{2} & 0 & 0 \\
0 & 0 & 0 & 0 & \eta_{2} & 0 \\
0 & 0 & 0 & 0 & 0 & \eta_{2}
\end{array}\right]
$$

Similarly, analytic expressions as a function of $n$ for the fourth-order orientation tensor $a_{i j k l}$ are substituted in Equation (5.10) to simplify the fourth-order stiffness tensor component $\left\langle C_{i j k l}\right\rangle_{4}$ as

$$
\left[\langle C\rangle_{4}(n)\right]=\frac{4 n(n-1)}{35\left(15+16 n+4 n^{2}\right)} \xi_{4}\left[\begin{array}{cccccc}
8 & -4 & -4 & 0 & 0 & 0  \tag{5.25}\\
-4 & 3 & 1 & 0 & 0 & 0 \\
-4 & 1 & 3 & 0 & 0 & 0 \\
0 & 0 & 0 & 1 & 0 & 0 \\
0 & 0 & 0 & 0 & -4 & 0 \\
0 & 0 & 0 & 0 & 0 & -4
\end{array}\right]
$$

Notice for the isotropic case (i.e., $n=0$ ) where $\psi(\theta, \phi)$ is given in Figure 5.1, all components of $\left[\langle C\rangle_{2}(0)\right]$ and $\left[\langle C\rangle_{4}(0)\right]$ are 0 . For the case $n=1$ shown in Figure 5.2, the second-order component of the stiffness tensor $\left[\langle C\rangle_{2}(1)\right]$ is non-zero, whereas the fourth-order component of the stiffness tensor $\left[\langle C\rangle_{4}(1)\right]$ remains $[\mathbf{0}]$. Note however, that for $n>1$ fourth-order stiffness tensor components contribute to the expectation of the stiffness tensor.

The variance of the stiffness tensor does not lend itself to concise expressions as seen in the preceding results for the material expectation tensor. Analytic expressions
for the orientation tensors as a function of increasing $n$ through eighth-order developed using the mathematical principle of induction (see e.g. [96]). These analytic functions are then introduced into Equations (5.17)-(5.22) to yield expressions for the variance of the stiffness tensor as a function of $n$ for the given fiber orientation probability distribution function in Equation (5.23). The variance of the extensional components $\sigma_{1111}^{2}, \sigma_{2222}^{2}$, and $\sigma_{3333}^{2}$ from Equation (5.17) are expressed after simplification as

$$
\begin{align*}
\sigma_{1111}^{2}(n)= & -\frac{16 n^{2}}{(3+2 n)^{2}} \tau_{2}^{2}+\frac{8 n \xi_{0}\left(2 \xi_{2}-3 \tau_{2}\right)}{9+6 n}+\frac{16\left(3+4 n+4 n^{2}\right) \xi_{2}^{2}}{9\left(15+16 n+4 n^{2}\right)} \\
& -\frac{256(n-1) n^{2}}{35(3+2 n)\left(15+16 n+4 n^{2}\right)} \tau_{2} \xi_{4}+\frac{256 n\left(5+2 n+2 n^{2}\right)}{105(7+2 n)\left(15+16 n+4 n^{2}\right)} \xi_{2} \xi_{4} \\
& +\frac{64\left(63+192 n+244 n^{2}+256 n^{3}+64 n^{4}\right)}{49\left(15+16 n+4 n^{2}\right)^{2}\left(63+32 n+4 n^{2}\right)} \xi_{4}^{2} \\
\sigma_{2222}^{2}(n)= & -\frac{4 n^{2}}{(3+2 n)^{2}} \tau_{2}^{2}+\frac{4 n \xi_{0}\left(3 \tau_{2}-2 \xi_{2}\right)}{9+6 n}+\frac{16\left(3+n+n^{2}\right) \xi_{2}^{2}}{9\left(15+16 n+4 n^{2}\right)} \\
& +\frac{48(n-1) n^{2}}{35(3+2 n)\left(15+16 n+4 n^{2}\right)} \tau_{2} \xi_{4}-\frac{32 n\left(10+n+n^{2}\right)}{35(7+2 n)\left(15+16 n+4 n^{2}\right)} \xi_{2} \xi_{4} \\
& +\frac{96\left(42+79 n+79 n^{2}+54 n^{3}+12 n^{4}\right)}{49\left(15+16 n+4 n^{2}\right)^{2}\left(63+32 n+4 n^{2}\right)} \xi_{4}^{2} \\
\sigma_{3333}^{2}(n)= & \sigma_{2222}^{2}(n) \tag{5.26}
\end{align*}
$$

where the final relation between $\sigma_{2222}^{2}(n)$ and $\sigma_{3333}^{2}(n)$ is realized due to the symmetry of the fiber distribution function of Equation (5.23) in the $x_{2}-x_{3}$ plane. The two independent shearing components of the stiffness tensor variance are given as

$$
\begin{align*}
\sigma_{2323}^{2}(n)= & \frac{36(1+2 n)}{(3+2 n)\left(15+16 n+4 n^{2}\right)} \eta_{2}^{2}-\frac{48 n(1+2 n)}{7\left(15+16 n+4 n^{2}\right)(3+2 n)(7+2 n)} \eta_{2} \xi_{4} \\
& +\frac{4\left(567+636 n+208 n^{2}+64 n^{3}+16 n^{4}\right)}{49\left(15+16 n+4 n^{2}\right)^{2}\left(63+32 n+4 n^{2}\right)} \xi_{4}^{2} \\
\sigma_{1212}^{2}(n)= & \frac{36(1+n)}{(3+2 n)^{2}(5+2 n)} \eta_{2}^{2}+\frac{24 n(5-2 n)}{7(3+2 n)^{2}\left(35+24 n+4 n^{2}\right)} \eta_{2} \xi_{4} \\
& +\frac{4\left(567+1644 n+2176 n^{2}+1744 n^{3}+400 n^{4}\right)}{49\left(15+16 n+4 n^{2}\right)^{2}\left(63+32 n+4 n^{2}\right)} \xi_{4}^{2} \tag{5.27}
\end{align*}
$$

The remaining shearing components are expressed as one of the two components of

Equation (5.27). Due to the symmetry of the stiffness tensor from the principal of incremental work (see e.g. Jones [10]) index pairs may be swapped, i.e. $\sigma_{2323}^{2}(n)=$ $\sigma_{3223}^{2}(n)=\sigma_{3232}^{2}(n)$ and $\sigma_{1212}^{2}(n)=\sigma_{2112}^{2}(n)=\sigma_{2121}^{2}(n)$. Due to symmetry in the $x_{2}-$ $x_{3}$ plane of the selected fiber distribution function the variance of the $C_{1313}$ component is identical to the variance of the $C_{1212}$ component, i.e. $\sigma_{1313}^{2}(n)=\sigma_{1212}^{2}(n)=$ $\sigma_{3113}^{2}(n)=\sigma_{3131}^{2}(n)$.

For the simple analytic distribution of Equation (5.23) the stiffness tensor variance for the extension-extension coupling components may be expressed as

$$
\begin{align*}
\sigma_{2233}^{2}(n)=- & \frac{16 n^{2}}{(3+2 n)^{2}} \epsilon_{2}^{2}+\frac{8 n \eta_{0}\left(3 \epsilon_{2}+6 \eta_{2}-\xi_{2}\right)}{9+6 n}+\frac{4\left(3+4 n+4 n^{2}\right)\left(\xi_{2}-6 \eta_{2}\right)^{2}}{9\left(15+16 n+4 n^{2}\right)} \\
& +\frac{32(n-1) n^{2}}{35(3+2 n)\left(15+16 n+4 n^{2}\right)} \epsilon_{2} \xi_{4}+\frac{16 n\left(5+2 n+2 n^{2}\right)\left(6 \eta_{2}-\xi_{2}\right)}{105(7+2 n)\left(15+16 n+4 n^{2}\right)} \xi_{4} \\
& +\frac{4\left(567+636 n+208 n^{2}+64 n^{3}+16 n^{4}\right)}{49\left(15+16 n+4 n^{2}\right)^{2}\left(63+32 n+4 n^{2}\right)} \xi_{4}^{2} \\
\sigma_{1122}^{2}(n)=- & \frac{4 n^{2}}{(3+2 n)^{2}} \epsilon_{2}^{2}-\frac{4 n \eta_{0}\left(3 \epsilon_{2}+6 \eta_{2}-\xi_{2}\right)}{9+6 n}+\frac{4\left(3+n+n^{2}\right)\left(\xi_{2}-6 \eta_{2}\right)^{2}}{9\left(15+16 n+4 n^{2}\right)} \\
& +\frac{64(n-1) n^{2}}{35(3+2 n)\left(15+16 n+4 n^{2}\right)} \epsilon_{2} \xi_{4}+\frac{8 n\left(8 n^{2}+8 n-25\right)\left(6 \eta_{2}-\xi_{2}\right)}{105(7+2 n)\left(15+16 n+4 n^{2}\right)} \xi_{4} \\
& +\frac{4\left(567+1644 n+2176 n^{2}+1744 n^{3}+400 n^{4}\right)}{49\left(15+16 n+4 n^{2}\right)^{2}\left(63+32 n+4 n^{2}\right)} \xi_{4}^{2} \tag{5.28}
\end{align*}
$$

The remaining extension-extension components are given as $\sigma_{2233}^{2}(n)=\sigma_{3322}^{2}(n)$ or $\sigma_{1122}^{2}(n)=\sigma_{2211}^{2}(n)=\sigma_{1133}^{2}(n)=\sigma_{3311}^{2}(n)$ from the principal of incremental work or due to the inherent symmetry of the fiber distribution function.

The independent components for the stiffness tensor variance of the shearextension coupling terms are expressed as

$$
\begin{align*}
\sigma_{1112}^{2}(n)= & \frac{1+2 n}{15+16 n+4 n^{2}} \xi_{2}^{2}+\frac{16 n(1+2 n)}{7\left(15+16 n+4 n^{2}\right)(7+2 n)} \xi_{2} \xi_{4} \\
& +\frac{8(1+2 n)\left(21+8 n+8 n^{2}\right)}{49\left(15+16 n+4 n^{2}\right)\left(63+32 n+4 n^{2}\right)} \xi_{4}^{2} \\
\sigma_{2223}^{2}(n)= & \frac{1}{15+16 n+4 n^{2}} \xi_{2}^{2}-\frac{12 n}{7\left(15+16 n+4 n^{2}\right)(7+2 n)} \xi_{2} \xi_{4} \\
& +\frac{12\left(14+3 n+3 n^{2}\right)}{49\left(15+16 n+4 n^{2}\right)\left(63+32 n+4 n^{2}\right)} \xi_{4}^{2} \\
\sigma_{1222}^{2}(n)= & \frac{1+2 n}{15+16 n+4 n^{2}} \xi_{2}^{2}-\frac{12 n(1+2 n)}{7\left(15+16 n+4 n^{2}\right)(7+2 n)} \xi_{2} \xi_{4} \\
& +\frac{12(1+2 n)\left(14+3 n+3 n^{2}\right)}{49\left(15+16 n+4 n^{2}\right)\left(63+32 n+4 n^{2}\right)} \xi_{4}^{2} \tag{5.29}
\end{align*}
$$

and

$$
\begin{align*}
\sigma_{1123}^{2}(n)= & \frac{\left(6 \eta_{2}-\xi_{2}\right)^{2}}{15+16 n+4 n^{2}}+\frac{24 n\left(\xi_{2}-6 \eta_{2}\right) \xi_{4}}{7\left(15+16 n+4 n^{2}\right)(7+2 n)} \\
& +\frac{12\left(7+12 n+12 n^{2}\right)}{49\left(15+16 n+4 n^{2}\right)\left(63+32 n+4 n^{2}\right)} \xi_{4}^{2} \\
\sigma_{2213}^{2}(n)= & \frac{(1+2 n)\left(6 \eta_{2}-\xi_{2}\right)^{2}}{15+16 n+4 n^{2}}+\frac{4 n(1+2 n)\left(6 \eta_{2}-\xi_{2}\right) \xi_{4}}{7\left(15+16 n+4 n^{2}\right)(7+2 n)} \\
& +\frac{4(1+2 n)\left(21+n+n^{2}\right)}{49\left(15+16 n+4 n^{2}\right)\left(63+32 n+4 n^{2}\right)} \xi_{4}^{2} \tag{5.30}
\end{align*}
$$

where the unwritten shear-extensional coupling variance components may be expressed as one of components given in Equations (5.29) and (5.30), i.e. $\sigma_{1121}^{2}(n)=$ $\sigma_{1112}^{2}(n), \sigma_{2333}^{2}(n)=\sigma_{2223}^{2}(n)$, etc.

Lastly, the independent stiffness tensor variance components of shear-shear coupling simplify to

$$
\begin{align*}
\sigma_{2312}^{2}(n)= & \frac{9(1+2 n)}{15+16 n+4 n^{2}} \eta_{2}^{2}-\frac{12 n(1+2 n)}{7\left(15+16 n+4 n^{2}\right)(7+2 n)} \eta_{2} \xi_{4} \\
& +\frac{4(1+2 n)\left(21+n+n^{2}\right)}{49\left(15+16 n+4 n^{2}\right)\left(63+32 n+4 n^{2}\right)} \xi_{4}^{2} \\
\sigma_{1312}^{2}(n)= & \frac{9}{15+16 n+4 n^{2}} \eta_{2}^{2}+\frac{72 n}{7\left(15+16 n+4 n^{2}\right)(7+2 n)} \eta_{2} \xi_{4} \\
& +\frac{12\left(7+12 n+12 n^{2}\right)}{49\left(15+16 n+4 n^{2}\right)\left(63+32 n+4 n^{2}\right)} \xi_{4}^{2} \tag{5.31}
\end{align*}
$$

with the remaining shear-shear components expressed in terms of either $\sigma_{2312}^{2}(n)$ or $\sigma_{1312}^{2}(n)$ from Equation (5.31), i.e. $\sigma_{1223}^{2}=\sigma_{1332}^{2}=\sigma_{1232}^{2}=\cdots=\sigma_{2312}^{2}$ and $\sigma_{1213}^{2}(n)=\sigma_{1231}^{2}(n)=\cdots=\sigma_{1312}^{2}(n)$.

The $C_{1111}$ component of the stiffness tensor in industrial parts is often of greatest interest and may be as much as two orders of magnitude greater than other components of the stiffness tensor in the principle reference frame (see e.g. [7] and Figures 4.2 and 4.5). For comparison purposes, the second component considered in further detail is the variance of the $C_{1112}$ component. The expectation of this particular component is often zero (or set to zero by the objective fourth-order closures) in the principal reference frame of $a_{i j}$, and for $\psi(\theta, \phi)$ in Equation (5.23), is explicitly zero for the selected distribution function. For the case of an isotropic distribution function (i.e. $n=0$ in Equation (5.23) and shown in Figure 5.1) the variance of the components $C_{1111}$ and $C_{1112}$ are, respectively,

$$
\begin{align*}
& \sigma_{1111}^{2}=\frac{16\left(245 \xi_{2}^{2}+4 \xi_{4}^{2}\right)}{11025}  \tag{5.32}\\
& \sigma_{1112}^{2}=\frac{5\left(147 \xi_{2}^{2}+8 \xi_{4}^{2}\right)}{11025} \tag{5.33}
\end{align*}
$$

For $n=1$, the distribution function in Equation (5.23) (see e.g., Figure 5.2) is transversely isotropic such that the second-order term of the stiffness tensor expectation
$[\langle C\rangle]_{2}$ appearing in Equation (5.8) is non-zero, while the fourth-order component from Equation (5.25) remains zero. The variance components $\sigma_{1111}^{2}$ and $\sigma_{1112}^{2}$ are given as

$$
\begin{align*}
\sigma_{1111}^{2} & =\frac{8}{848925}\left(59290 \xi_{2}^{2}+7392 \xi_{2} \xi_{4}+936 \xi_{4}^{2}+11319\left(5 \xi_{0}\left(2 \xi_{2}-3 \tau_{2}\right)-6 \tau_{2}^{2}\right)\right)  \tag{5.34}\\
\sigma_{1112}^{2} & =\frac{15}{56595}\left(4851 \xi^{2}+1232 \xi_{2} \xi_{4}+296 \xi_{4}^{2}\right) \tag{5.35}
\end{align*}
$$

As in the isotropic example, the stiffness tensor variance of the $C_{1112}$ component is of a similar magnitude as $C_{1111}$ since each of the terms $\xi_{0}, \xi_{2}, \xi_{4}$ and $\tau_{2}$ are similar in magnitude. For the highly aligned distribution of Figure 5.3, corresponding to the case $n=6$ in Equation (5.23), the variance components $\sigma_{1111}^{2}$ and $\sigma_{1112}^{2}$ are given as

$$
\begin{align*}
\sigma_{1111}^{2}= & \frac{16}{20179425}\left(1503565 \xi_{2}^{2}+338912 \xi_{2} \xi_{4}+9412 \xi_{4}^{2}\right. \\
& \left.+1345295 \xi_{0}\left(2 \xi_{2}-3 \tau_{2}\right)-434112 \xi_{4} \tau_{2}-3228708 \tau_{2}^{2}\right)  \tag{5.36}\\
\sigma_{1112}^{2}= & \frac{1105}{20179425}\left(931 \xi^{2}+672 \xi_{2} \xi_{4}+136 \xi_{4}^{2}\right) \tag{5.37}
\end{align*}
$$

It is worthwhile to note that in this example, the variance of the stiffness tensor component $C_{1112}$ is greater than the variance of $C_{1111}$ for each selected value of $n$ in Equation (5.23). Recall, that the $C_{1112}$ component is often neglected in processing simulations whereas great attention is provided the $C_{1111}$ component.

### 5.1.3.2 Orthotropic Fiber Orientation Distribution

The final analytical example considered here is that of the fiber orientation distribution appearing in Figure 5.4 which is given by

$$
\begin{equation*}
\psi(\theta, \phi)=\frac{1}{2 \pi} \frac{\left(1+\cot ^{4} \theta \csc ^{4} \phi\right)\left(1-\cos ^{2} \phi \sin ^{2} \theta\right)}{\left(1+\cot ^{2} \theta \csc ^{2} \phi\right)^{2}} \tag{5.38}
\end{equation*}
$$

Equation (5.38) is obtained from the orthotropic distribution function $\psi(\theta, \phi)=$ $\frac{1}{2 \pi} \sin ^{2} \theta\left(\cos ^{4} \phi+\sin ^{4} \phi\right)$ rotated into a frame such that the fibers tend to align with


Figure 5.4: Orthotropic fiber orientation distribution $\psi(\theta, \phi)=$ $\frac{1}{2 \pi} \frac{\left(1+\cot ^{4} \theta \csc ^{4} \phi\right)\left(1-\cos ^{2} \phi \sin ^{2} \theta\right)}{\left(1+\cot ^{2} \theta \csc ^{2} \phi\right)^{2}}$.
equal probability along both the $x_{2}$ and $x_{3}$ axis. This distribution cannot be expressed with just the second-order Laplace series expansion and is not transversely isotropic, even though all objective fourth-order closures would predict such behavior (recall Equations (4.4) and (4.5) along with Jack and Smith [7]). The zeroth-order contribution to the expectation value of the stiffness matrix is given in Equation (5.6). The second-order contribution to the expectation value of the stiffness tensor is evaluated from Equation (5.8) as

$$
\left[\langle C\rangle_{2}\right]=\frac{1}{5}\left[\begin{array}{cccccc}
-2 \tau_{2} & -\epsilon_{2} & -\epsilon_{2} & 0 & 0 & 0  \tag{5.39}\\
-\epsilon_{2} & \tau_{2} & 2 \epsilon_{2} & 0 & 0 & 0 \\
-\epsilon_{2} & 2 \epsilon_{2} & \tau_{2} & 0 & 0 & 0 \\
0 & 0 & 0 & 2 \eta_{2} & 0 & 0 \\
0 & 0 & 0 & 0 & -\eta_{2} & 0 \\
0 & 0 & 0 & 0 & 0 & -\eta_{2}
\end{array}\right]
$$

Since the distribution function for this orthotropic example requires a Laplace series of order greater than two, there will be non-zero fourth-order contributions to the
stiffness tensor expectation value evaluated from Equation (5.10) as

$$
\left[\langle C\rangle_{4}\right]=\frac{1}{70}\left[\begin{array}{cccccc}
0 & 0 & 0 & 0 & 0 & 0  \tag{5.40}\\
0 & \xi_{4} & -\xi_{4} & 0 & 0 & 0 \\
0 & -\xi_{4} & \xi_{4} & 0 & 0 & 0 \\
0 & 0 & 0 & -\xi_{4} & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0
\end{array}\right]
$$

After some simplification, the variance of $C_{1111}$ for $\psi(\theta, \phi)$ in Equation (5.38) is obtained with Equation (5.17) as

$$
\begin{equation*}
\sigma_{1111}^{2}=-\frac{1}{35} \xi_{0}\left(28 \xi_{2}+\xi_{4}\right)+\frac{267344 \xi_{2}^{2}-43736 \xi_{2} \xi_{4}+4633 \xi_{4}^{2}}{1131900} \tag{5.41}
\end{equation*}
$$

and the variance for $C_{2222}$ and $C_{3333}$ are evaluated as

$$
\begin{equation*}
\sigma_{2222}^{2}=\sigma_{3333}^{2}=-\frac{78}{175} \xi_{2}^{2}+\frac{16}{735} \xi_{2} \xi_{4}+\frac{4651}{679140} \xi_{4}^{2} \tag{5.42}
\end{equation*}
$$

The remaining components of the variance may be similarly developed from Equations (5.18)-(5.22) and are omitted here for conciseness.

### 5.2 Validation of Analytical Forms of Elastic Properties with the Method of Monte-Carlo

In the previous section and in Jack and Smith [79] an analytic form was presented to derive the expectation of the material stiffness tensor and its variance directly from the orientation tensors. This section presents statistical results obtained from the method of Monte-Carlo [97] to obtain the sample mean and sample variance of material stiffness tensor components and compare computed results with those obtained from the analytically derived expectation and variance. The results are studied in depth for a simple analytic function to demonstrate the effectiveness of the proposed method, and for the industrially relevant results obtained from numerical distribution function simulations for center-gated disk flow.

For simplicity, results throughout this section will be formulated with the HalpinTsai equations outlined in Table 2.1 to obtain the unidirectional stiffness tensor $\bar{C}_{i j k l}$, with fibers aligned along the $x_{1}$-axis given in Figure 2.1. Once the unidirectional material stiffness tensor $\bar{C}_{i j k l}$ and fiber orientation tensors are known, the expectation of the material stiffness tensor $\left\langle C_{i j k l}\right\rangle$ and variance of the material stiffness tensor $\sigma_{i j k l}^{2}$ may be analytically obtained through Equations (5.12) and (5.17)-(5.22), respectively. Recall that the form for $\left\langle C_{i j k l}\right\rangle$ was identical to the form of Advani and Tucker [6] and has been employed elsewhere to compute the mean properties of discrete sample sets (see e.g. [6, 27-29, 35-37,56,64]). The variance $\sigma_{i j k l}^{2}$ has yet to be employed or verified in any practical applications, but will be further investigated below.

### 5.2.1 Material Stiffness Prediction

One approach to obtain material stiffness values is to compute the expectation value from the aggregate of unidirectional fibers using the fiber distribution function $\psi(\theta, \phi)$ (see e.g. Jack and Smith [75]). Given a set of $N$ angle pairs $\left\{\left(\theta_{n}, \phi_{n}\right): 1 \leq n \leq N, n \in \mathbb{N}, N \in \mathbb{N}\right\}$ defining an aggregate of unidirectional fibers, where $\mathbb{N}$ is the set of positive integer numbers $\mathbb{N}=\{1,2, \ldots\}$, the sample mean $m_{i j k l}$ for the stiffness tensor from the corresponding fiber stress field is

$$
\begin{equation*}
m_{i j k l}=\frac{1}{N} \sum_{n=1}^{N}\left(Q_{q i}\left(\theta_{n}, \phi_{n}\right) Q_{r j}\left(\theta_{n}, \phi_{n}\right) Q_{s k}\left(\theta_{n}, \phi_{n}\right) Q_{t l}\left(\theta_{n}, \phi_{n}\right) \bar{C}_{q r s t}\right) \tag{5.43}
\end{equation*}
$$

where $i, j, k, l, q, r, s, t \in\{1,2,3\}, \bar{C}_{q r s t}$ is the unidirectional stiffness tensor in Equation (2.44), and $\mathbf{Q}(\theta, \phi)$ is the rotation tensor

$$
\mathbf{Q}\left(\theta_{n}, \phi_{n}\right)=\left[\begin{array}{ccc}
\sin \theta_{n} \cos \phi_{n} & \sin \theta_{n} \sin \phi_{n} & \cos \theta_{n}  \tag{5.44}\\
-\sin \phi_{n} & \cos \phi_{n} & 0 \\
-\cos \theta_{n} \cos \phi_{n} & -\cos \theta_{n} \sin \phi_{n} & \sin \theta_{n}
\end{array}\right]
$$

Note in Equation (5.43), the summation convention is in effect (except on $n$ ) where repeated indices imply summation. The sample mean $m_{i j k l}$ is an unbiased estimator
(see e.g. [97]) of the population mean of a distribution having a probability density function $P_{i j k l}(x)$ where $x$ belongs to the range of values from the sample set.

If the given set of angles $\left\{\left(\theta_{n}, \phi_{n}\right): 1 \leq n \leq N, n \in \mathbb{N}, N \in \mathbb{N}\right\}$ are sampled such that as $N \rightarrow \infty$ the distribution of representative angles approach the fiber orientation probability distribution function $\psi(\theta, \phi)$, then the sample mean $m_{i j k l}$ approaches the expectation value $\left\langle C_{i j k l}\right\rangle$ (otherwise known as the orientation average or the population mean) and may be expressed as the integrand from Equation (5.1). Equations (5.1) and (5.43) do not take into account the spatial interaction of fibers, this is assumed to be satisfied through the formation of $\psi(\theta, \phi)$.

The sample variance $s_{i j k l}^{2}$ (also labeled as ${ }_{2} m_{i j k l}$ ) is the second sample moment about $m_{i j k l}$ defined as

$$
\begin{equation*}
s_{i j k l}^{2}=\frac{1}{N} \sum_{n=1}^{N}\left(Q_{p i}\left(\theta_{n}, \phi_{n}\right) Q_{q j}\left(\theta_{n}, \phi_{n}\right) Q_{r k}\left(\theta_{n}, \phi_{n}\right) Q_{s l}\left(\theta_{n}, \phi_{n}\right) \bar{C}_{p q r s}-m_{i j k l}\right)^{2} \tag{5.45}
\end{equation*}
$$

This is a biased estimator of the population variance $\sigma_{i j k l}^{2}$. To provide an unbiased estimator, the $k$-statistic $k_{i j k l}^{2}$ is defined by

$$
\begin{equation*}
k_{i j k l}^{2}=s_{i j k l}^{2} \frac{N}{N-1} \tag{5.46}
\end{equation*}
$$

### 5.2.2 Computing Properties with Statistical Sampling

The sample mean and variance of Equations (5.43) and (5.45) may be computed from experimental data sets of discrete fibers within a sample to form an approximate fiber distribution. This chapter considers the case where the fiber orientation probability distribution is known and the method of Monte-Carlo is employed to generate statistical data to analyze both the sample mean and the sample variance of the material stiffness tensor which are then compared to results obtained analytically.

### 5.2.2.1 The Monte-Carlo Simulation Method

The Monte-Carlo simulation procedure has seen extensive use in recent history for numerical integration of multi-dimensional integrals with complicated boundaries and functions without local strong peaks (see e.g. [97-100]). The Monte-Carlo simulation procedure simply simulates synthetic data sets drawn randomly from an appropriate probability distribution. Analogous to physical experiments where random samples are drawn from experimental data, Monte-Carlo simulations are performed on a computer to randomly generate the variables $(\Theta, \Phi)$ from the fiber orientation probability distribution function to capture the statistical characteristics of the design function, in this case the material stiffness tensor.

In the following examples, the integration approximated is the material stiffness tensor in Equation (5.1) and variance in Equation (5.45) which are each evaluated as a sum of independent data sets drawn from the fiber orientation probability distribution function. The selection of random data sets from the fiber probability distribution function $\psi(\theta, \phi)$ is strongly dependant on the appropriate selection of the random angle pairs $\left(\Theta_{n}, \Phi_{n}\right)$.

### 5.2.2.2 Accept Reject Generation Algorithm

An appropriate set of angle pairs is selected in this study using the Accept-Reject Generation Algorithm (ARGA) (see e.g [97]). The ARGA numerically generates a sample set for any given probability distribution function, such as $\psi(\theta, \phi) \in \mathbb{S}^{2}$ using a uniform random number generator that provides a distribution on $(0,1) \in \mathbb{R}^{1}$. The Accept-Reject Generation Algorithm can be used when it is relatively easy to generate a random variable, such as the random variable $U$ with a uniform $(0,1)$
distribution. In one dimension the ARGA may be used to develop a continuous random variable $X$ whose probability distribution function is $f(x)$ from the probability distribution function $g(x)$ where the random variable $Y$, an observation from $g(x)$, is sufficiently easy to generate. The ARGA may be applied if there exists some constant $K \in \mathbb{R}^{+}$, such that $\forall x \in(-\infty, \infty)$ the probability distribution functions satisfy the relationship $f(x) \leq K g(x)$. The ARGA is demonstrated in Hogg et al. [97], where the following algorithm is shown to generate a random variable $X$ that has the probability distribution function $f(x)$.

- Numerically generate the random variables $Y$ and $U$
- If $U \leq \frac{f(Y)}{K g(Y)}$, then set the random variable $X$ equal to $Y$
- The random variable $X$ will have the probability distribution function $f(x)$

The proof of this algorithm is omitted, and the interested reader is directed to Hogg et al. [97]. In the current study, it was found numerically that the most effective choice of constant $K$ was to set $K=\left(\max _{x \in \mathbb{R}^{1}} f(x)\right)(1+\epsilon)$ where $\epsilon$ is a small number greater than zero. The random variable $Y$ can belong to any probability distribution function and for ease of computation this can be taken to be a uniform or normal distribution. After the selection of $K$ and random variable $Y$, moving from one dimension, $\mathbb{R}^{1}$, to the surface of the sphere, $\mathbb{S}^{2}$, is trivially accomplished with ARGA for the random variable pair $\Theta$ and $\Phi$ belonging to the probability distribution $\psi(\theta, \phi) \sin \theta$. For examples considered here, the following procedure is followed to generate a sample set of $\Theta$ and $\Phi$ values on the sphere at discrete points $(\theta, \phi)=\left(\frac{i-1}{N_{\theta}-1} \pi, \frac{j-1}{N_{\phi}} \pi\right)$ for $i \in\left\{1,2, \ldots, N_{\theta}\right\}$ and $j \in\left\{1,2, \ldots, N_{\phi}\right\}$, and $N_{\theta}$ and $N_{\phi}$ are the number of steps in $\theta$ and $\phi$, respectively (distribution function symmetry is assumed).

1. Select $K=\left(\max _{(\theta, \phi) \in \mathbb{S}^{2}} \psi(\theta, \phi)\right)(1+\epsilon)$ where $\epsilon \sim 10^{-2}$
2. Select a value for $(\theta, \phi)$ at which the observation will be made
3. Generate a random observation $U$ belonging to the uniform distribution
4. If $K U<\psi(\theta, \phi) \sin \theta$, then set $\Theta=\theta$ and $\Phi=\phi$.
5. Step two through four are repeated at each point on the sphere

### 5.2.2.3 Central Limit Theorem

The Central-Limit Theorem (see e.g. [97, 100]) states that a probability distribution composed of the summation of the mean of many smaller random deviations will, for non-correlated deviations, converge to a normal distribution. The Central-Limit Theorem is elegantly stated and proved by Hogg et al. [97] and is paraphrased here by taking the observations $X_{1}, X_{2}, \ldots, X_{n}$ of a random sample from the distribution $f(x)$ having a distinct and finite mean $\mu$ and variance $\sigma$ where $f(x)$ is not necessarily Gaussian. Given $\bar{X}_{n}$ as the mean of a set of random samples, the random variable

$$
\begin{equation*}
Y_{n}=\sqrt{n} \frac{\bar{X}_{n}-\mu}{\sigma} \tag{5.47}
\end{equation*}
$$

will, for sufficiently large $n$, converge in distribution to a random variable with a standard normal distribution (i.e., a mean of zero with variance of 1 ). This particular theorem allows easy statistical representations of distributions which are poorly approximated by the normal distribution. Additionally, the Central-Limit Theorem allows trivial mathematical representations of data sets to establish approximate probabilities on the mean $\bar{X}$ (see e.g. [97, 99]).

### 5.2.3 Monte-Carlo Predictions of Material Stiffness

Examples are selected to demonstrate the Monte-Carlo procedure described above while validating the analytical expressions for the stiffness tensor mean and variance described in Section 5.1. The first example illustrates the Monte-Carlo procedure through a comparison with values published in Advani and Tucker [6]. The second example is for a simple analytical fiber distribution function to validate the analytical material stiffness tensor mean and variance formulas presented in Section 5.1. The third example focuses on applications with the Central Limit Theorem for the same analytic fiber probability distribution function from the second example. The final example considers fiber orientation distributions obtained through numerical simulations of center-gated disk flow. In this work, results from the analytic derivation of the material stiffness tensor expectation and variance given in Equations (5.12) and (5.17)-(5.22) are compared and validated with results developed through the Monte-Carlo method of integration. Note that results for correlated sample angle pairs will yield a variance different than those predicted in Equations (5.17)-(5.22), where positive correlations cause the sample variance to increase and negative correlations cause the sample variance to decrease (see e.g. Bevington and Robinson [98]). As such, $N$ random uncorrelated angle pairs $\left\{\left(\Theta_{n}, \Phi_{n}\right): 1 \leq n \leq N, n \in \mathbb{N}, N \in \mathbb{N}\right\}$ are chosen from the fiber orientation probability distribution function $\psi(\theta, \phi)$ using the Accept-Reject Generation Algorithm discussed previously. Once the $N$ random angle pairs $\left\{\left(\Theta_{n}, \Phi_{n}\right): 1 \leq n \leq N, n \in \mathbb{N}, N \in \mathbb{N}\right\}$, from the distribution $\psi(\theta, \phi)$, are formed using ARGA, Equations (5.43) and (5.45) are used to form the sample mean and variance, respectively.

Table 5.1: Comparison between select components published in Advani and Tucker (1987) for material parameters with the sample mean obtained through the method of Monte-Carlo for the distribution $\psi(\theta, \phi)=\frac{61}{4 \pi} \sin ^{60} \theta \cos ^{60} \phi$.

| Material | Advani-Tucker | Sample Mean |
| :---: | :---: | :---: |
| Properties | Properties $[6]$ | $\left(N=2 \times 10^{9}\right)$ |
| $\left\langle E_{1}\right\rangle\left(10^{6} \mathrm{psi}\right)$ | 2.220 | 2.224 |
| $\left\langle E_{2}\right\rangle\left(10^{6} \mathrm{psi}\right)$ | 0.813 | 0.814 |
| $\left\langle E_{3}\right\rangle\left(10^{6} \mathrm{psi}\right)$ | 0.813 | 0.813 |
| $\left\langle\nu_{12}\right\rangle$ | 0.333 | 0.330 |
| $\left\langle G_{12}\right\rangle\left(10^{6} \mathrm{psi}\right)$ | 0.293 | 0.292 |
| $\left\langle G_{23}\right\rangle\left(10^{6} \mathrm{psi}\right)$ | 0.254 | 0.254 |

### 5.2.3.1 Comparison Between Proposed Method and Published Results

The first example considers the simple analytic distribution function of Equation (5.23) with $n=60$ where the material properties of the constitutive materials are published in Advani and Tucker [6]. The expected value of the material stiffness tensor $\left\langle C_{i j k l}\right\rangle$ is computed analytically from Equation (5.12) with orientation tensors calculated through Equation (2.11) for the distribution function in Equation (5.23). Next, the material parameters $E_{1}, E_{2}, E_{3}, \nu_{12}, G_{12}$, and $G_{23}$ are computed with Equation (2.44) to compare with the results published in Advani and Tucker [6]. The proposed method for the expectation of the stiffness tensor yields identical results and the results are given in the second column of Table 5.1. The method of MonteCarlo is utilized along with Equation (5.43) to predict the expectation of the stiffness tensor $\left\langle C_{i j k l}\right\rangle$ for $2 \times 10^{9}$ simulations for the same underlying material properties. The results also appear in Table 5.1. Notice that the values from Advani and Tucker [6] obtained through orientation tensors are nearly identical to those obtained from the Monte-Carlo results using Equation (5.43).

Table 5.2: Fiber and matrix material parameters employed in example problems.

| $E_{f}$ | $=30 \times 10^{9} \mathrm{~Pa}$ | $\nu_{f}$ | $=0.20$ | $E_{m}=1 \times 10^{9} \mathrm{~Pa}$ |
| :--- | :--- | :--- | :--- | :--- |
| $\nu_{m}$ | $=0.38$ | $L$ | $=100 \mu \mathrm{~m}$ | $a_{r}=10$ |
| $V_{f}$ | $=0.1$ | $V_{c}$ | $=8 \times 10^{6} \mu^{3}$ |  |

### 5.2.3.2 Stiffness Tensor Distribution Results

Material properties throughout the remainder of this chapter for the underlying fiber and matrix appear in Table 5.2 and are taken from those presented in Tucker and Liang [80] for typical fiber-reinforced engineering thermoplastics. The distribution function in Equation (5.23) is chosen here for simplicity in integration and because it provides a degree of alignment that is commonly found in typical injection molded short-fiber polymer composites with fiber volume fractions in the range of 10-30\% (see e.g. $[23,60]$ ). The calculations to follow are for $n=4$ which results in $c=\frac{9}{4 \pi}$. Figure 5.5 shows the fiber probability distribution function in Equation (5.23) with $n=4$. A sample set of fibers with $V_{f}=10 \%$ is shown in Figure 5.6 having the same $\psi(\theta, \phi)$. Observe, the fibers tend to orient along the $x_{1}$ axis, but by no means experience unidirectional alignment.

The analytic form of the material stiffness tensor expectation $\left\langle C_{i j k l}\right\rangle$, is evaluated from Equation (5.12) for $\psi(\theta, \phi)$ given in Equation (5.23) for $n=4$, and the material stiffness tensor variance $\sigma_{i j k l}^{2}$ is computed from Equations (5.17)-(5.22). Three random sample sets of $\left\{\left(\Theta_{n}, \Phi_{n}\right): 1 \leq n \leq N, n \in \mathbb{N}, N \in \mathbb{N}\right\}$ for $N=10^{2}, N=10^{5}$ and $N=10^{8}$ are chosen from $\psi(\theta, \phi)$ using ARGA for the Monte-Carlo simulations to develop both the sample mean $m_{i j k l}$ in Equation (5.43) and the $k$-statistic $k_{i j k l}^{2}$ in Equation (5.46). The analytic results for select components of $\left\langle C_{i j k l}\right\rangle$ are presented


Figure 5.5: Fiber probability distribution function $\psi(\theta, \phi)=\frac{9}{4 \pi} \sin ^{8} \theta \cos ^{8} \phi$
in Table 5.3, and the corresponding variance of the stiffness tensor $\sigma_{i j k l}^{2}$ is presented in Table 5.4. Results appearing in Tables 5.3 and 5.4 are evaluated from orientation tensors through fourth-order for the mean and through eighth-order for the variance. In all of these calculations, orientation tensors are computed directly from the fiber probability distribution function $\psi(\theta, \phi)$ through Equation (2.11). After only $10^{2}$ Monte-Carlo simulations the sample mean is predicted with reasonable accuracy for most of the components, whereas for $N=10^{5}$ and $N=10^{8}$ the Monte-Carlo results and the analytic results are in nearly perfect agreement. Conversely, for $N=10^{2}$ the Monte-Carlo simulations provide a poor representation of the analytic solution for the variance of the stiffness tensor as shown in Table 5.4. Several of the selected components have a $10 \%-15 \%$ difference between the analytic variance solution and the Monte-Carlo results, and the $\left\langle C_{2323}\right\rangle$ component has a difference of nearly $22 \%$. As the number of Monte-Carlo simulations increases to $N=10^{5}$, the difference in the variance is reduced significantly, with most of the components experiencing a


Figure 5.6: Sample fiber distribution sampled from $\psi(\theta, \phi)=\frac{9}{4 \pi} \sin ^{8} \theta \cos ^{8} \phi$ for $V_{f}=10 \%$.
difference of $1 \%-7 \%$. For $N=10^{8}$ simulations, the difference in the predicted variance drops to less than $0.1 \%$.

The analytical expectation values of the stiffness tensor $\left\langle C_{i j k l}\right\rangle$ appearing in Table 5.3 are identical to those obtained using the Advani and Tucker [6] analytical approach. This is expected since it was shown in Equation (5.13) that the method for computing the expectation of the stiffness tensor can be written in the form of the Advani and Tucker model.

To further illustrate the statistical nature of $C_{i j k l}$, frequency plots of three selected components $C_{1111}, C_{1122}$ and $C_{1123}$ appear in Figures 5.7-5.9, respectively, for $N=10^{2}$ and $N=10^{6}$ Monte-Carlo simulations. The maximum for $C_{1111}$ and $C_{1122}$ (i.e., $C_{1111}^{\max }=3.34 \mathrm{GPa}$ and $C_{1122}^{\max }=1.73 \mathrm{GPa}$, respectively) and their minimum values (i.e., $C_{1111}^{\min }=2.41 \mathrm{GPa}$ and $C_{1122}^{\min }=1.45 \mathrm{GPa}$, respectively), provide bounds on the Monte-Carlo results. Note that the upper bound on $C_{1111}$ corresponds to a

Table 5.3: Comparison between select components from analytic results for the expectation of the stiffness tensor $\left\langle C_{i j k l}\right\rangle$ with the sample mean $m_{i j k l}$ obtained through the method of Monte-Carlo for the distribution $\psi(\theta, \phi)=\frac{9}{4 \pi} \sin ^{8} \theta \cos ^{8} \phi$.

| $C_{i j k l}$ <br> component | $\left\langle C_{i j k l}\right\rangle(\mathrm{GPa})$ | $m_{i j k l}(\mathrm{GPa})$ <br> $\left(N=10^{2}\right)$ | $m_{i j k l}(\mathrm{GPa})$ <br> $\left(N=10^{5}\right)$ | $m_{i j k l}(\mathrm{GPa})$ <br> $\left(N=10^{8}\right)$ |
| :---: | :---: | :---: | :---: | :---: |
| 1111 | 3.0318 | 3.0234 | 3.0310 | 3.0318 |
| 2222 | 2.4271 | 2.4268 | 2.4273 | 2.4271 |
| 3333 | 2.4271 | 2.4290 | 2.4270 | 2.4271 |
| 1122 | 1.5273 | 1.5309 | 1.5274 | 1.5273 |
| 1133 | 1.5273 | 1.5263 | 1.5274 | 1.5273 |
| 1123 | 0.0000 | -0.0292 | -0.0002 | 0.0000 |
| 2323 | 0.4361 | 0.4373 | 0.4357 | 0.4356 |

Table 5.4: Comparison between select components from analytic results for the stiffness tensor variance $\sigma_{i j k l}^{2}$ with the sample variance obtained through the method of Monte-Carlo for the distribution $\psi(\theta, \phi)=\frac{9}{4 \pi} \sin ^{8} \theta \cos ^{8} \phi$.

| $C_{i j k l}$ <br> component | $\sigma_{i j k l}^{2}\left(\mathrm{~Pa}^{2}\right)$ | $k_{i j k l}^{2}\left(\mathrm{~Pa}^{2}\right)$ <br> $\left(N=10^{2}\right)$ | $k_{i j k l}^{2}\left(\mathrm{~Pa}^{2}\right)$ <br> $\left(N=10^{5}\right)$ | $k_{i j k l}^{2}\left(\mathrm{~Pa}^{2}\right)$ <br> $\left(N=10^{8}\right)$ |
| :---: | :---: | :---: | :---: | :---: |
| 1111 | $4.856 \times 10^{16}$ | $5.570 \times 10^{16}$ | $4.881 \times 10^{16}$ | $4.856 \times 10^{16}$ |
| 2222 | $1.276 \times 10^{15}$ | $1.115 \times 10^{15}$ | $1.362 \times 10^{15}$ | $1.275 \times 10^{15}$ |
| 3333 | $1.276 \times 10^{15}$ | $1.121 \times 10^{15}$ | $1.253 \times 10^{15}$ | $1.276 \times 10^{15}$ |
| 1122 | $5.013 \times 10^{15}$ | $4.816 \times 10^{15}$ | $5.021 \times 10^{15}$ | $5.013 \times 10^{15}$ |
| 1133 | $5.013 \times 10^{15}$ | $5.630 \times 10^{15}$ | $4.990 \times 10^{15}$ | $5.014 \times 10^{15}$ |
| 1123 | $2.332 \times 10^{15}$ | $2.624 \times 10^{15}$ | $2.350 \times 10^{15}$ | $2.333 \times 10^{15}$ |
| 2323 | $2.933 \times 10^{14}$ | $3.566 \times 10^{14}$ | $3.021 \times 10^{14}$ | $2.933 \times 10^{14}$ |

unidirectional fiber distribution along the $x_{1}$ axis, and the lower bound is located at a point $(\theta, \phi)$ near the $x_{2}-x_{3}$ plane. Conversely, the minimum value of $C_{1122}$ corresponds to a unidirectional fiber distribution along the $x_{1}$ axis and the maxima occurs when the fibers align in the $x_{2}-x_{3}$ plane. The bounds for the $C_{1123}$ component are similarly found, and provide definitive limits for the stiffness tensor distribution. It is clear that $C_{1111}, C_{1122}$ and $C_{1123}$ are not Normally distributed, but the trends are not unexpected in any of the plots. For the high fiber alignment along the $x_{1}$ axis $C_{1111}$
tends toward the unidirectional orientation state given by $C_{1111}^{\max }$, and it is expected that as the alignment increases along the $x_{1}$ direction $C_{1122}$ will diminish. Since very few fibers are perpendicular to the $x_{1}$ axis, there is a smaller probability of a $C_{1111}$ that approaches $C_{1111}^{\min }$ and the inverse is true of $C_{1122}$. Similarly, the expectation of the $C_{1123}$ component will approach a value of zero for a large enough sample set for $\psi(\theta, \phi)$ given in Equation (5.23), but clearly from Figure 5.9 there is a significant probability that the effective material behavior will not be zero.

### 5.2.3.3 Applications with the Central Limit Theorem

Multiple independent sample sets of fiber distributions are available from multiple short-fiber composite parts produced in an industrial manufacturing process. Each part exhibits its own unique stiffness tensor, such that a set of parts has a mean and variance that can be evaluated as shown below. This process is duplicated computationally with the Monte-Carlo method through the central limit theorem by taking the $N$ random sample angle pairs $\left\{\left(\Theta_{n}, \Phi_{n}\right): 1 \leq n \leq N\right\}$ obtained from the probability distribution $\psi(\theta, \phi)$ where the $N$ random sample pairs compose a single set with its own sample mean and variance. Taking $M$ unique and independent sets of the $N$ random angle pairs there will be $M$ unique sample means $m_{i}$ and variances $s_{i}^{2}$ for $\{i: i \in \mathbb{N}, 1 \leq i \leq M\}$. The Central-Limit Theorem in Equation (5.47) is used to compute the statistical properties of the random variable $Y_{i}$ for sufficiently large $N$, which converges in distribution to a random variable with a normal distribution.

The parameter $N$ is chosen to be related to the number of fibers for a given sample set as the integer part of $V_{f} \frac{V_{c}}{V_{\text {fiber }}}$, where $V_{\text {fiber }}$ is the volume of an individual fiber and $V_{c}$ is the volume of the representative sample cube. This may be considered to represent a given region within a part with volume $V_{c}$ that is selected small enough


Figure 5.7: Frequency of Monte-Carlo results for $C_{1111}$ for data samples taken from $\psi(\theta, \phi)=\frac{9}{4 \pi} \sin ^{8} \theta \cos ^{8} \phi$. (a) $N=10^{2}$ (b) $N=10^{6}$.


Figure 5.8: Frequency of Monte-Carlo results for $C_{1122}$ for data samples taken from $\psi(\theta, \phi)=\frac{9}{4 \pi} \sin ^{8} \theta \cos ^{8} \phi$. (a) $N=10^{2}$ (b) $N=10^{6}$.


Figure 5.9: Frequency of Monte-Carlo results for $C_{1123}$ for data samples taken from $\psi(\theta, \phi)=\frac{9}{4 \pi} \sin ^{8} \theta \cos ^{8} \phi$. (a) $N=10^{2}$ (b) $N=10^{6}$.
such that the fiber distribution is assumed constant, and the fibers within this sample region form the angles of the sample set. The Central Limit Theorem provides a method to predict the mean and the variance of the stiffness tensor at the same region in each of the parts for a given fiber probability distribution function.

For each sample cube $i$, $\{i: i \in \mathbb{N}, M \in \mathbb{N}, 1 \leq i \leq M\}, N$ random sample angle pairs $\left\{\left(\Theta_{n}, \Phi_{n}\right): 1 \leq n \leq N\right\}$ are generated from the fiber probability distribution function $\psi(\theta, \phi)$. The sample mean of the stiffness tensor $m_{i j k l}$ for each of the $M$ sample sets is computed from Equation (5.43). For select volume fractions, the mean of the stiffness tensor is normalized as $m_{i j k l} / N$ for sample means from $M=10^{2}$, $M=10^{3}$, and $M=10^{6}$ sample sets from the fiber orientation probability distribution function $\psi(\theta, \phi)$ given in Equation (5.23). Volume fractions of fibers within the polymer matrix of $10 \%, 20 \%$ and $30 \%$ are selected and yield, respectively, $N=$ $V_{f} \frac{V_{c}}{V_{\text {fiber }}}=101, N=203$ and $N=305$ fibers. Histograms of the Monte-Carlo results for the $C_{1111}$ component are given in Figures 5.10-5.12, simulations for the $C_{2222}$ component are given in Figures 5.13-5.15, and simulations for the $C_{1123}$ component are given in Figures 5.16-5.18 for, respectively, volume fractions of $V_{f}=10 \%, V_{f}=$ $20 \%$ and $V_{f}=30 \%$. Additionally, Figures 5.10-5.18 contain the normal distribution $P_{C_{i j k l}}(x)$ expressed as

$$
\begin{equation*}
P_{C_{i j k l}}(x)=\frac{\sqrt{N}}{\sqrt{2 \pi \sigma_{i j k l}^{2}}} \mathrm{e}^{-\left(x-\left\langle C_{i j k l}\right\rangle\right)^{2} /\left(\frac{2 \sigma_{i j k l}^{2}}{N}\right)} \tag{5.48}
\end{equation*}
$$

where the analytic values of $\left\langle C_{i j k l}\right\rangle$ and $\sigma_{i j k l}^{2}$ are evaluated, respectively, from Equations (5.12) and (5.17)-(5.22).

In Figures 5.10-5.18, by increasing the number of $M$ sample sets, the frequency plots $m_{i j k l} / N$ approach the analytically determined Normal distribution $P_{C_{i j k l}}(x)$ of Equation (5.48). For as few as $N=101$, corresponding to the case of $V_{f}=10 \%$, the


Figure 5.10: Comparison between normal distribution $P_{C_{1111}}(x)$ obtained analytically and the frequency plot $m_{1111} / N$ for Monte-Carlo results of $M$ sample sets with $N=$ 101 angle pairs for data samples taken from $\psi(\theta, \phi)=\frac{9}{4 \pi} \sin ^{8} \theta \cos ^{8} \phi$. (a) $M=10^{2}$ (b) $M=10^{3}$ (c) $M=10^{6}$.


Figure 5.11: Comparison between normal distribution $P_{C_{1111}}(x)$ obtained analytically and the frequency plot $m_{1111} / N$ for Monte-Carlo results of $M$ sample sets with $N=$ 203 angle pairs for data samples taken from $\psi(\theta, \phi)=\frac{9}{4 \pi} \sin ^{8} \theta \cos ^{8} \phi$. (a) $M=10^{2}$ (b) $M=10^{3}$ (c) $M=10^{6}$.


Figure 5.12: Comparison between normal distribution $P_{C_{1111}}(x)$ obtained analytically and the frequency plot $m_{1111} / N$ for Monte-Carlo results of $M$ sample sets with $N=$ 305 angle pairs for data samples taken from $\psi(\theta, \phi)=\frac{9}{4 \pi} \sin ^{8} \theta \cos ^{8} \phi$. (a) $M=10^{2}$ (b) $M=10^{3}$ (c) $M=10^{6}$.


Figure 5.13: Comparison between normal distribution $P_{C_{2222}}(x)$ obtained analytically and the frequency plot $m_{2222} / N$ for Monte-Carlo results of $M$ sample sets with $N=$ 101 angle pairs for data samples taken from $\psi(\theta, \phi)=\frac{9}{4 \pi} \sin ^{8} \theta \cos ^{8} \phi$. (a) $M=10^{2}$ (b) $M=10^{3}$ (c) $M=10^{6}$.


Figure 5.14: Comparison between normal distribution $P_{C_{2222}}(x)$ obtained analytically and the frequency plot $m_{2222} / N$ for Monte-Carlo results of $M$ sample sets with $N=$ 203 angle pairs for data samples taken from $\psi(\theta, \phi)=\frac{9}{4 \pi} \sin ^{8} \theta \cos ^{8} \phi$. (a) $M=10^{2}$ (b) $M=10^{3}$ (c) $M=10^{6}$.


Figure 5.15: Comparison between normal distribution $P_{C_{2222}}(x)$ obtained analytically and the frequency plot $m_{2222} / N$ for Monte-Carlo results of $M$ sample sets with $N=$ 305 angle pairs for data samples taken from $\psi(\theta, \phi)=\frac{9}{4 \pi} \sin ^{8} \theta \cos ^{8} \phi$. (a) $M=10^{2}$ (b) $M=10^{3}$ (c) $M=10^{6}$.


Figure 5.16: Comparison between normal distribution $P_{C_{1123}}(x)$ obtained analytically and the frequency plot $m_{1123} / N$ for Monte-Carlo results of $M$ sample sets with $N=$ 101 angle pairs for data samples taken from $\psi(\theta, \phi)=\frac{9}{4 \pi} \sin ^{8} \theta \cos ^{8} \phi$. (a) $M=10^{2}$ (b) $M=10^{3}$ (c) $M=10^{6}$.


Figure 5.17: Comparison between normal distribution $P_{C_{1123}}(x)$ obtained analytically and the frequency plot $m_{1123} / N$ for Monte-Carlo results of $M$ sample sets with $N=$ 203 angle pairs for data samples taken from $\psi(\theta, \phi)=\frac{9}{4 \pi} \sin ^{8} \theta \cos ^{8} \phi$. (a) $M=10^{2}$ (b) $M=10^{3}$ (c) $M=10^{6}$.


Figure 5.18: Comparison between normal distribution $P_{C_{1123}}(x)$ obtained analytically and the frequency plot $m_{1123} / N$ for Monte-Carlo results of $M$ sample sets with $N=$ 305 angle pairs for data samples taken from $\psi(\theta, \phi)=\frac{9}{4 \pi} \sin ^{8} \theta \cos ^{8} \phi$. (a) $M=10^{2}$ (b) $M=10^{3}$ (c) $M=10^{6}$.
frequency plots for the $C_{1111}$ and the $C_{1123}$ components are nearly indistinguishable for $M=10^{6}$ sample sets as that of the Normal distribution predicted from the analytic results for the expectation and variance. This trend continues as the volume fraction is increased to $V_{f}=20 \%$ and $V_{f}=30 \%$ for both the $C_{1111}$ and the $C_{1123}$ component. Conversely, for the $C_{2222}$ component with a volume fraction of $V_{f}=10 \%$, there is a clear difference between the normal distribution and the frequency plots. This discrepancy is still visible for $V_{f}=20 \%$, but as $N$ increases with $V_{f}=30 \%$ the frequency plots approach the predicted normal distribution obtained through the Central Limit Theorem. This behavior is not unexpected, as the Central Limit Theorem assumes the limiting case as $N \rightarrow \infty$, and as demonstrated in the figures. The Central Limit Theorem provides reasonable results for the probability distribution of the stiffness tensor for samples as low as $N=101$.

### 5.2.3.4 Center-Gated Disk Results

The final example is based on the flow near a pin gate $[27,56]$ as illustrated in Figure 5.19. The polymer melt is assumed to enter the mold through a pin gate, then flow radially outward where the velocity components of the flow are a function of radial position $r$ and gap height $z$. Assuming a Newtonian fluid for simplicity, the velocity components in a local Cartesian coordinate system with coordinates $\left(x_{1}, x_{2}, x_{3}\right)$, that correspond to $(r, \theta, z)$, are given in Equation (3.14). For small radii the flow is dominated by out-of-plane stretching, and as the radial location $r$ increases, the flow tends toward shearing in the direction of flow. In this work, the solution of $\psi(\theta, \phi, t)$ from Equation (2.23) is computed numerically using the control volume method of Bay [23] with an interaction coefficient of $C_{I}=10^{-2}$ and a volume fraction near $V_{f}=20 \%$ (see e.g. [1]). Orientation tensors calculated from the numerical solution of


Figure 5.19: Schematic for center-gated disk flow depicting selected radial locations in the example results.
$\psi(\theta, \phi, t)$ using Equation (2.11) are used to evaluate $\left\langle C_{i j k l}\right\rangle$ and $\sigma_{i j k l}^{2}$ analytically with Equations (5.12) and (5.17)-(5.22), respectively. For a volume fraction of $20 \%$, there are $N=V_{f} \frac{V_{c}}{V_{\text {fiber }}}=203$ fibers in each of the $M$ sample sets with the fiber and matrix constituent properties that appear in Table 5.2. Sample distributions obtained from the fiber orientation flow simulations are computed at each of the points $A, B, C$, and $D$ from Figure 5.19 which corresponds to the radial locations $r / b=2,5,10$, and 40 , respectively. The initial fiber distribution is assumed to be isotropic at $r / b=1$.

Monte-Carlo simulations for $M=10^{2}$ and $M=10^{6}$ are used to produce a statistical representation of the stiffness tensor. Results for the normalized frequency distribution $m_{i j k l} / N$ of the sample means $m_{i j k l}$ along with the analytical probability distribution function $P_{C_{i j k l}}(x)$ from the analytic expectation and variance for the $C_{1111}$ and the $C_{1122}$ stiffness tensor component are presented in Figures 5.20 and 5.21, respectively. Note the change as a function of radial position in the expectation and variance for both stiffness tensor components. This change is directly attributed to the change in the alignment of the fiber probability distribution function along the radial direction. This changing behavior in both the expectation and the variance is seen throughout the entire gap height $0 \leq z / b \leq 1$. To demonstrate the change in material behavior, the expectation $\left\langle C_{i j k l}\right\rangle$ is normalized by the isotropic value of
the stiffness tensor component $C_{i j k l}^{\text {iso }}$ which appears in Figures 5.22 and 5.23 for the $C_{1111}$ and the $C_{1122}$ components, respectively, for radial locations $1 \leq \frac{r}{b} \leq 40$. The error bars are provided by $\sigma_{i j k l}^{2}$ obtained analytically from $\psi(\theta, \phi)$. Observe the fiber probability distribution function does not reach steady state for all gap heights, and exhibits a clear change in the expectation of the material stiffness tensor as well as the variance of the material stiffness tensor.

Figure 5.22 illustrates that the variance of $C_{1111}$ for the gap height $z=\frac{1}{10} b$, at various radial locations, is small in comparison to the variance at a gap height of $z=\frac{9}{10} b$. Conversely, the variance of $C_{1122}$ in Figures 5.23 appears to be relatively constant. This observation is quantified by the coefficient of variation which describes the normalized variability (see e.g. [100]), defined as $\delta_{i j k l}=\sigma_{i j k l} /\left\langle C_{i j k l}\right\rangle$ (no sum on $i, j, k$, or $l$ ) which is plotted in Figures 5.24 and 5.25 for the $C_{1111}$ and the $C_{1122}$ components, respectively. Note that the coefficient of variation is less than $1 \%$ throughout the entire flow history. The small value for the coefficient of variation implies that few Monte-Carlo simulations are necessary to accurately capture the statistical behavior of the stiffness tensor expectation value. In a similar study by Jack and Smith [39], stiffness tensor components were investigated as a function of volume fraction for the simple analytical fiber probability distribution function from Equation (5.23). Results revealed that the material stiffness variability remained relatively small ( $\delta \approx 1 \%$ ) for the entire range of volume fractions investigated.

### 5.3 Material Property Prediction Remarks

An analytical method is presented to compute the expectation value and variance of the material stiffness tensor obtained from a fiber orientation probability distribution.


Figure 5.20: Comparison between normal distribution $P_{C_{1111}}(x)$ obtained analytically and the frequency plot $m_{1111} / N$ for Monte-Carlo results of $M$ sample sets with $N=$ 203 angle pairs for data samples taken from center-gated disk flow for a gap height of $z=\frac{5 b}{10}$. (a) $M=10^{2}$ (b) $M=10^{6}$


Figure 5.21: Comparison between normal distribution $P_{C_{1122}}(x)$ obtained analytically and the frequency plot $m_{1122} / N$ for Monte-Carlo results of $M$ sample sets with $N=$ 203 angle pairs for data samples taken from center-gated disk flow for a gap height of $z=\frac{5 b}{10}$. (a) $M=10^{2}$ (b) $M=10^{6}$


Figure 5.22: Normalized mean of the expectation value component $\left\langle C_{1111}\right\rangle / C_{1111}^{\text {iso }}$ for select gap heights throughout the flow history for center-gated disk flow, $C_{I}=10^{-2}$.


Figure 5.23: Normalized mean of the expectation value component $\left\langle C_{1122}\right\rangle / C_{1122}^{\text {iso }}$ for select gap heights throughout the flow history for center-gated disk flow, $C_{I}=10^{-2}$.


Figure 5.24: Coefficient of variation $\delta_{1111}$ for select gap heights for center-gated disk flow, $C_{I}=10^{-2}$.


Figure 5.25: Coefficient of variation $\delta_{1122}$ for select gap heights for center-gated disk flow, $C_{I}=10^{-2}$.

The elastic material constitutive behavior for short fiber composites presented here is based on the Laplace series reconstruction written in terms of complex spherical harmonic functions. By employing the Laplace series reconstruction, an expression is derived for the expectation value of the stiffness tensor using second- and fourth-order orientation tensors. The relationship between the current approach and that of Advani and Tucker [6] is presented. The current approach only requires the assumption of a single axis of symmetry characterized by a monoclinic material (see e.g. [7, 10]) through the symmetry of the distribution function of fibers, $\psi(\boldsymbol{p})=\psi(-\boldsymbol{p})$. An analytic method for computing the variance of the stiffness tensor is also presented which is shown to be a function of the orientation tensors up to eighth-order.

The analytical method to evaluate the expectation and variance for the material stiffness tensor from fiber orientation tensors is validated through the method of Monte-Carlo. The proposed analytic method and the Monte-Carlo simulations yield the same expectation as results previously published in the literature. These new results also provide values of the variance of the stiffness tensor previously unavailable in the literature. The Monte-Carlo results are shown to agree extremely well with analytic results as the number of sample sets increases. Using the Central-Limit Theorem, normal probability distributions obtained from the analytic expectation and variance of the material stiffness tensor corresponded directly with the results for the normalized frequency of the sample sets for finite angle sets corresponding to discrete fiber orientation angles.

## CHAPTER 6

## DIRECTIONAL DIFFUSION FOR FIBER ORIENTATION FLOW ANALYSIS

The Folgar and Tucker model [11] for fiber interaction behavior due to the surrounding flow kinematics has found widespread industrial acceptance, and is considered to be the standard that new or proposed models are often compared to. Recently, limitations during the transient analysis of the Folgar and Tucker model have been identified and new methods to accurately represent the orientation state during the filling stage of the injection molding process are necessary. The Folgar and Tucker model predicts fiber orientation aligns at a rate much faster than that seen in the laboratory $[2,26,42,44,46]$, and any proposed model should better predict these experimental results.

Folgar and Tucker [11] observed that an individual fiber follows the Jeffery orbits [12] for fibers in a dilute suspension as given in Equation (2.19) for short periods of time. However, the fiber will at random, reorient to another angle, then resume following a Jeffery orbit. These reorientations are attributed to fiber interactions tending to randomize the orientation. Folgar and Tucker account for collisions through rotary diffusion and scaled the diffusion with the rate of deformation of the fluid. They speculate that rotary diffusion could be constructed with a directional bias to account for differing behavior between random and aligned suspensions, but chose not to include such effects.

This chapter extends the directional diffusion model introduced by Jack [45] through an objective inclusion of local fiber collisions. The directional diffusion model
seeks to qualitatively represent the behavior of fiber orientation in semi-dilute and concentrated suspensions through scalable parameters that allow refinement of the fiber alignment rate and steady state fiber orientation. Results demonstrate the directional diffusion model yields steady state solutions to within $0.1 \%$ of those computed with the Folgar and Tucker model for the flows considered while providing a significant delay in the rate of fiber alignment.

### 6.1 A Model for Directional Diffusion

This new directional diffusion model incorporates two effects, (1) local directionally dependant effects directly proportional to the probability of collision between two fibers, and (2) large scale volume averaged diffusion behavior analogous to shear rate dependant Brownian motion. All motion of a constitutive equation must be invariant with respect to rigid body motion (see e.g. [9, 101, 102]). For a constitutive fiber motion model, an observer in a co-rotational frame that translates and rotates with each fiber views the same behavior as an observer in a co-deformational reference frame that translates, rotates, and deforms with the flowing fibers. An objective model states that any constitutive model must be independent of rigid body motion (also referred to as the principal of material reference frame indifference [9]).

The following diffusion model looks at the relative motion of two fibers represented by the unit vectors $\boldsymbol{p}$ and $\boldsymbol{\rho}$ shown in Figure 6.1. The model assumes that local directionally dependant effects are proportional to the probability of a collision between any two fibers $\boldsymbol{p}$ and $\boldsymbol{\rho}$. Jeffery's model for the motion of the fiber $\boldsymbol{\rho}$ is expressed without rigid body effects to represent the relative fiber motion of $\boldsymbol{\rho}$ with


Figure 6.1: Coordinate system describing the path through which the fiber $\boldsymbol{\rho}$ passes into and $\boldsymbol{p}$.
respect to $\boldsymbol{p}$ as

$$
\begin{equation*}
\dot{\tilde{\boldsymbol{\rho}}}^{h}=\dot{\boldsymbol{\rho}}^{h}+\frac{1}{2} \boldsymbol{\omega} \cdot \boldsymbol{\rho}=\frac{1}{2} \lambda(\dot{\boldsymbol{\gamma}} \cdot \boldsymbol{\rho}-\dot{\boldsymbol{\gamma}} \boldsymbol{\rho} \boldsymbol{\rho} \boldsymbol{\rho}) \tag{6.1}
\end{equation*}
$$

where rigid body effects due to vorticity $\boldsymbol{\omega}$ are removed from the Jeffery motion.
During the infinitesimal period of time $\Delta t$, the fiber $\boldsymbol{\rho}$ rotates by an angle relative to $\boldsymbol{p}$ proportional to $\dot{\tilde{\boldsymbol{\rho}}}^{h}$. During the time period $\Delta t$, the area through which $\boldsymbol{\rho}$ rotates is proportional to the product of the magnitude of $\boldsymbol{\rho} \times \dot{\tilde{\boldsymbol{\rho}}}^{h}$ and $\Delta t$ (assumed small). Notice that the resultant vector $\boldsymbol{\rho} \times \dot{\tilde{\boldsymbol{\rho}}}^{h}$ is normal to the plane containing the fiber. The probability of a hit between $\boldsymbol{p}$ and $\boldsymbol{\rho}$ will be zero when $\boldsymbol{p}$ is parallel to the plane containing $\boldsymbol{\rho}$ and $\dot{\tilde{\boldsymbol{\rho}}}^{h}$, and will be greatest when $\boldsymbol{p}$ is parallel to $\boldsymbol{\rho} \times \dot{\tilde{\boldsymbol{\rho}}}^{h}$. The probability of a collision $P_{\boldsymbol{\rho}}$ hit $\boldsymbol{p}$ is defined as being proportional to the scalar triple product (see e.g. [103]) of $\boldsymbol{p}$ with the vector $\boldsymbol{\rho} \times \dot{\tilde{\boldsymbol{\rho}}}^{h}$ as

$$
\begin{equation*}
P_{\boldsymbol{\rho} \text { hit } \boldsymbol{p}}=C_{1}\left|\boldsymbol{p} \cdot\left(\boldsymbol{\rho} \times \dot{\tilde{\boldsymbol{\rho}}}^{h}\right)\right| \tag{6.2}
\end{equation*}
$$

where the constant $C_{1}$ may be a function of the fiber aspect ratio $a_{r}$ and the volume fraction of fibers $V_{f}$.

Note that the magnitude of the scalar triple product for any set of vectors $\mathbf{a}, \mathbf{b}, \mathbf{c} \in \mathbb{R}^{3}$ satisfies the relationship $|\mathbf{a} \cdot(\mathbf{b} \times \mathbf{c})|=|\mathbf{b} \cdot(\mathbf{c} \times \mathbf{a})|=|\mathbf{c} \cdot(\mathbf{a} \times \mathbf{b})|=$ $|\mathbf{a} \cdot(\mathbf{c} \times \mathbf{b})|=\cdots[104]$. Therefore, the probability of $\boldsymbol{\rho}$ hitting $\boldsymbol{p}$ is proportionate to the scalar triple product as

$$
\begin{equation*}
P_{\boldsymbol{\rho} \text { hit } \boldsymbol{p}}=C_{1}\left|\boldsymbol{\rho} \cdot\left(\boldsymbol{p} \times \dot{\tilde{\boldsymbol{\rho}}}^{h}\right)\right| \tag{6.3}
\end{equation*}
$$

During the time $\Delta t$, the relative motion of fiber $\boldsymbol{\rho}$ to fiber $\boldsymbol{p}$ will experience a given motion $\dot{\tilde{\boldsymbol{p}}}^{h}$ (c.f., Equation (6.1)) so that the probability of $\boldsymbol{\rho}$ passing through $\boldsymbol{p}$ is analogous to the probability of $\boldsymbol{p}$ passing through $\boldsymbol{\rho}$. Therefore it may be similarly shown that the probability of the fiber $\boldsymbol{p}$ hitting $\boldsymbol{\rho}$ is

$$
\begin{equation*}
P_{\boldsymbol{p} \text { hit } \boldsymbol{\rho}}=C_{1}\left|\boldsymbol{p} \cdot\left(\boldsymbol{\rho} \times \dot{\tilde{\boldsymbol{p}}}^{h}\right)\right| \tag{6.4}
\end{equation*}
$$

where $C_{1}$ is assumed to be the same function of volume fraction and fiber aspect ratio given in Equation (6.3) since the collision of $\boldsymbol{\rho}$ into $\boldsymbol{p}$ is indistinguishable from the collision of $\boldsymbol{p}$ into $\boldsymbol{\rho}$.

The process described above is repeated for each fiber near the fiber $\boldsymbol{p}$ to account for other fiber collisions. Each of the fibers $\boldsymbol{\rho}$ belong to the fiber orientation probability distribution function $\psi(\boldsymbol{\rho})$, so that the expectation of a collision with a given fiber $\boldsymbol{p}$ may be represented by the integral over $\boldsymbol{\rho} \in \mathbb{S}^{2}$. The resulting rotary diffusion due to local fiber collisions is

$$
\begin{equation*}
\left.D_{r}(\theta, \phi)\right|_{\text {Local Collisions }}=\oint_{\boldsymbol{\rho} \in \mathbb{S}^{2}}\left(C_{1}\left|\boldsymbol{\rho} \cdot\left(\boldsymbol{p} \times \dot{\tilde{\boldsymbol{\rho}}}^{h}\right)\right|+C_{1}\left|\boldsymbol{p} \cdot\left(\boldsymbol{\rho} \times \dot{\tilde{\boldsymbol{p}}}^{h}\right)\right|\right) \psi(\boldsymbol{\rho}) \mathrm{d} \mathbb{S} \tag{6.5}
\end{equation*}
$$

The volume averaged effects are assumed to satisfy the Brownian-type behavior observed by Folgar and Tucker [11], and will be modeled as

$$
\begin{equation*}
\left.D_{r}(\theta, \phi)\right|_{\text {Brownian }}=\oint_{\boldsymbol{\rho} \in \mathbb{S}^{2}} C_{2}\|\dot{\gamma}\| \psi(\boldsymbol{\rho}) \mathrm{d} \mathbb{S} \tag{6.6}
\end{equation*}
$$

Incorporating the Brownian-type effects produces the desired results, and is similar to previous fiber interaction models.

The proposed model incorporates the diffusion as the sum of the local collision effects and Brownian like motion to express the rotary diffusion $D_{r}$ appearing in Equation (2.23) as

$$
\begin{aligned}
D_{r}(\theta, \phi) & =\left.D_{r}(\theta, \phi)\right|_{\text {Brownian }}+\left.D_{r}(\theta, \phi)\right|_{\text {Local Collisions }} \\
& =\oint_{\boldsymbol{\rho} \in \mathbb{S}^{2}}\left(C_{1}\left|\boldsymbol{\rho} \cdot\left(\boldsymbol{p} \times \dot{\tilde{\boldsymbol{\rho}}}^{h}\right)\right|+C_{1}\left|\boldsymbol{p} \cdot\left(\boldsymbol{\rho} \times \dot{\tilde{\boldsymbol{p}}}^{h}\right)\right|\right) \psi(\boldsymbol{\rho}) \mathrm{d} \mathbb{S}+\oint_{\boldsymbol{\rho} \in \mathbb{S}^{2}} C_{2}\|\dot{\boldsymbol{\gamma}}\| \psi(\boldsymbol{\rho}) \mathrm{d} \mathbb{S}(6.7)
\end{aligned}
$$

Each of the functions $\boldsymbol{p}, \boldsymbol{\rho}, \dot{\tilde{\boldsymbol{p}}}^{h}, \dot{\tilde{\boldsymbol{\rho}}}^{h}$ and $\|\dot{\boldsymbol{\gamma}}\|$ are assumed continuous and finite, therefore the integration in Equation (6.7) exists and is finite, and may be written as

$$
D_{r}(\theta, \phi)=C_{1} \oint_{\boldsymbol{\rho} \in \mathbb{S}^{2}}\left|\boldsymbol{\rho} \cdot\left(\boldsymbol{p} \times \dot{\tilde{\boldsymbol{\rho}}}^{h}\right)\right| \psi(\boldsymbol{\rho}) \mathrm{d} \mathbb{S}+C_{\boldsymbol{\rho}} \oint_{\boldsymbol{\rho} \in \mathbb{S}^{2}}\left|\boldsymbol{p} \cdot\left(\boldsymbol{\rho} \times \dot{\tilde{\boldsymbol{p}}}^{h}\right)\right| \psi(\boldsymbol{\rho}) \mathrm{d} \mathbb{S}+C_{\boldsymbol{\rho}} \oint_{\boldsymbol{\rho} \in \mathbb{S}^{2}}\|\dot{\boldsymbol{\gamma}}\| \psi(\boldsymbol{\rho}) \mathrm{d} \mathbb{S}(6.8)
$$

The absolute values within the integration make it impossible to obtain analytical forms for $D_{r}(\theta, \phi)$ expressed in terms of the orientation tensors $a_{i j}, a_{i j k l}$, etc. Therefore the function $f(x)=|x|$ is approximated by $g(x)=x^{2}$ for $x \in[-1,1]$. Observe from Figure 6.2 that this approximation under predicts the function $f(x)$ except when $x \in\{-1,0,1\}$. Therefore, applying this approximation within the integrations of Equation (6.8) is expected to under estimate the actual number of collisions.

The approximation which replaces $|x|$ with $x^{2}$ implies that $x$ remain within the range $(-1,1)$. By definition, the components of the unit vectors $\boldsymbol{p}$ and $\boldsymbol{\rho}$ in the scalar triple products given in Equation (6.8) will have magnitudes less than or equal to one. Unfortunately, this is not the case for the terms $\dot{\tilde{\boldsymbol{p}}}^{h}$ and $\dot{\tilde{\boldsymbol{\rho}}}^{h}$ since they experience no such bounds. Recall from Equation (6.1) that each component of $\dot{\tilde{\boldsymbol{p}}}^{h}$ (or $\dot{\tilde{\boldsymbol{\rho}}}^{h}$ ) is a product of $\boldsymbol{p}$ (or $\boldsymbol{\rho}$ ) with the rate of deformation tensor $\dot{\boldsymbol{\gamma}}$. Each component of $\boldsymbol{p}$ (or


Figure 6.2: Comparison between $f(x)=|x|$ and $g(x)=x^{2}$ for $x \in[-1,1]$.
$\boldsymbol{\rho})$ is less than or equal to one in magnitude, but $\dot{\gamma}$ deserves further consideration. The scalar magnitude of the rate of deformation tensor, $\|\dot{\gamma}\|$, relates to the magnitude of the $i, j$ component of $\dot{\gamma}$ as

$$
\begin{equation*}
\|\dot{\gamma}\|=\sqrt{\frac{1}{2} \dot{\gamma}_{k l} \dot{\gamma}_{l k}}=\frac{1}{\sqrt{2}} \sqrt{\dot{\gamma}_{k l} \dot{\gamma}_{k l}} \geq \frac{1}{\sqrt{2}} \sqrt{\dot{\gamma}_{i j}^{2}}=\frac{1}{\sqrt{2}}\left|\dot{\gamma}_{i j}\right| \tag{6.9}
\end{equation*}
$$

where the repeated indices $k$ and $l$ imply summation, and the symmetry of the rate of deformation tensor presented in Equation (2.21) is utilized along with the recognition that the sum of the squares of each tensor component is greater than or equal to the square of the individual $i, j$ tensor component. Therefore from Equation (6.9) each expression $\left|\boldsymbol{\rho} \cdot\left(\boldsymbol{p} \times \frac{\dot{\tilde{\boldsymbol{\rho}}}^{h}}{\sqrt{2}\|\dot{\boldsymbol{\gamma}}\|}\right)\right|$ and $\left|\boldsymbol{p} \cdot\left(\boldsymbol{\rho} \times \frac{\dot{\tilde{\boldsymbol{p}}}^{h}}{\sqrt{2}\|\dot{\boldsymbol{\gamma}}\|}\right)\right|$ are less than unity for any velocity field. With this modification and the approximation in Figure 6.2, Equation (6.8) is expressed as

$$
\begin{align*}
D_{r}(\theta, \phi) \approx & C_{1}\|\dot{\gamma}\| \oint_{\boldsymbol{\rho} \in \mathbb{S}^{2}}\left(\boldsymbol{\rho} \cdot\left(\boldsymbol{p} \times \frac{\dot{\tilde{\boldsymbol{\rho}}}^{h}}{\|\dot{\boldsymbol{\gamma}}\|}\right)\right)^{2} \psi(\boldsymbol{\rho}) \mathrm{d} \mathbb{S} \\
& +C_{1}\|\dot{\gamma}\| \oint_{\boldsymbol{\rho} \in \mathbb{S}^{2}}\left(\boldsymbol{p} \cdot\left(\boldsymbol{\rho} \times \frac{\dot{\tilde{\boldsymbol{p}}}^{h}}{\|\dot{\boldsymbol{\gamma}}\|}\right)\right)^{2} \psi(\boldsymbol{\rho}) \mathrm{d} \mathbb{S}+C_{2}\|\dot{\boldsymbol{\gamma}}\| \oint_{\boldsymbol{\rho} \in \mathbb{S}^{2}} \psi(\boldsymbol{\rho}) \mathrm{d} \mathbb{S} \tag{6.10}
\end{align*}
$$

where the factor $\sqrt{2}$ is absorbed into the constant $C_{1}$.

The scalar triple product for the vectors $\mathbf{a}, \mathbf{b}$, and $\mathbf{c}$ in index notation is expressed as $\mathbf{a} \cdot(\mathbf{b} \times \mathbf{c})=\epsilon_{i j k} a_{i} b_{j} c_{k}$ where $\epsilon_{i j k}$ is the permutation symbol defined as [105]

$$
\epsilon_{i j k}=\left\{\begin{array}{rl}
0 & i=j,, j=k, \text { or } k=i  \tag{6.11}\\
+1 & (i, j, k) \in\{(1,2,3),(2,3,1),(3,1,2)\} \\
-1 & (i, j, k) \in\{(1,3,2),(3,2,1),(2,1,3)\}
\end{array}\right.
$$

The last term of Equation (6.10) is simplified as $C_{2}\|\dot{\gamma}\|$ from normalization $\oint_{\boldsymbol{\rho} \in \mathbb{S}^{2}} \psi(\boldsymbol{\rho}) \mathrm{d} \mathbb{S}=1$ in Equation (2.5). Therefore, Equation (6.10) is expressed as

$$
\begin{equation*}
D_{r}(\theta, \phi)=\frac{C_{1}}{\|\dot{\gamma}\|}\left(\oint_{\boldsymbol{\rho} \in \mathbb{S}^{2}}\left(\epsilon_{i j k} \rho_{i} p_{j} \dot{\tilde{\rho}}_{k}^{h}\right)^{2} \psi(\boldsymbol{\rho}) \mathrm{d} \mathbb{S}+\oint_{\boldsymbol{\rho} \in \mathbb{S}^{2}}\left(\epsilon_{i j k} p_{i} \rho_{j} \dot{\tilde{p}}_{k}^{h}\right)^{2} \psi(\boldsymbol{\rho}) \mathrm{d} \mathbb{S}\right)+C_{2}\|\dot{\gamma}\| \tag{6.12}
\end{equation*}
$$

It is worthwhile to note that the Folgar and Tucker model for diffusion is readily obtained by setting $C_{1}=0$ and $C_{2}=C_{I}$, and conveniently allows comparisons between the directional diffusion model of Equation (6.12) and the Folgar and Tucker model results. In Equation (6.12) the diffusion is a function of the velocity gradients through the rate of deformation tensor and the fiber orientation through the unit vector $\boldsymbol{p}$, which is independent of the integration for $\boldsymbol{\rho} \in \mathbb{S}^{2}$. Concern with the directional diffusion model may arise with the expression $1 /\|\dot{\gamma}\|$ as the deformation of the fluid goes to zero. This concern is alleviated since each component of $\left(\epsilon_{i j k} \rho_{i} p_{j} \dot{\tilde{\rho}}_{k}^{h}\right)^{2}$ and $\left(\epsilon_{i j k} p_{i} \rho_{j} \dot{\tilde{p}}_{k}^{h}\right)^{2}$ contain $\dot{\gamma}_{i j}^{2}$ which goes to zero as quickly or quicker than does the scalar magnitude of the rate of deformation. Therefore, when there is no deformation of the surrounding fluid, the diffusion effects go to zero as do the relative fiber motion effects implying there is no change in the fiber orientation.

### 6.1.1 Characteristics of the Directional Diffusion Model

To better understand the directional diffusion model for $D_{r}(\theta, \phi)$, typical orientation states are taken from results obtained with the Folgar and Tucker model for two flows, uniaxial elongation and simple shear. Note that the directional diffusion model will
alter the fiber orientation path. However, orientation states obtained with the Folgar and Tucker model are used to investigate the qualitative behavior of the directional diffusion model at physically meaningful orientation states. To remove volume averaged effects from the directional diffusion model while retaining the behavior resulting from local fiber collisions, a normalized diffusion $\hat{D}_{r}(\theta, \phi)$ is defined as

$$
\begin{equation*}
\hat{D}_{r}(\theta, \phi)=\left(D_{r}(\theta, \phi)-C_{2}\|\dot{\gamma}\|\right)\left(\max _{\substack{t \in[0, \infty) \\(\theta, \phi) \in \mathbb{S}^{2}}}\left(D_{r}(\theta, \phi, t)-C_{2}\|\dot{\gamma}\|\right)\right)^{-1} \tag{6.13}
\end{equation*}
$$

where any absolute values are not necessary since $D_{r}(\theta, \phi)-C_{2}\|\dot{\gamma}\|$ is always greater than or equal to zero.

The first flow considered is that of uniaxial elongational flow with $v_{1}=2 G x_{1}$, $v_{2}=-G x_{2} v_{3}=-G x_{3}$, where the fiber orientation distribution $\psi(\theta, \phi)$ is calculated at the points $G t=\frac{1}{10}, G t=1$ and $G t \rightarrow \infty$ using the control volume method from Bay [23] for an initially isotropic orientation state with diffusion coefficients of $\left(C_{1}, C_{2}\right)=$ $\left(0, C_{I}\right)=\left(0,10^{-2}\right)$. The normalized directional diffusion $\hat{D}_{r}(\theta, \phi)$ in Equation (6.13) is computed with $D_{r}(\theta, \phi)$ from Equation (6.8) for $\left(C_{1}, C_{2}\right)=\left(1,10^{-2}\right)$ and presented graphically at $G t=\frac{1}{10}, G t=1$ and $G t \rightarrow \infty$ in Figures 6.3(a), 6.3(c) and 6.3(e), respectively. Recall for uniaxial elongational flow; fibers tend to align symmetrically along the $x_{1}$ axis, and as such, diffusion effects from Equation (6.8) on any fiber along this axis will be small. This characteristic is observed by the sharp valley in Figures $6.3(\mathrm{a}), 6.3(\mathrm{c})$ and $6.3(\mathrm{e})$ where the $x_{1}$ axis corresponds to the angles $(\theta, \phi)=(\pi / 2,0)$ and $(\theta, \phi)=(\pi / 2, \pi)$. Notice that diffusion effects for a fiber not aligned near the $x_{1}$ axis will exhibit a higher value of $\hat{D}_{r}(\theta, \phi)$ as shown. The normalized diffusion has a local minima along the plane $\phi=\pi / 2$ which is caused by the low probability of a fiber sampled from the distribution $\psi(\theta, \phi)$ even existing in the $\phi=\pi / 2$ plane. Notice that
as the fiber orientation goes from isotropic to aligned, the effects of diffusion decrease. This is attributed to the small probability of a fiber existing at orientations away from the $x_{1}$ axis since the fiber alignment is nearly uniaxial for large values of $G t$.

The directional diffusion $D_{r}(\theta, \phi)$ with the approximation $|x| \sim(x)^{2}$ appearing in Equation (6.12) is evaluated at the same three values of time in Figures 6.3(b), $6.3(\mathrm{~d})$ and $6.3(\mathrm{f})$ for uniaxial elongational flow. Note that results are similar to those in Figures 6.3(a), 6.3(c) and 6.3(e) except that the approximate model smooths the diffusion function and is most noticeable along the sharply peaked valley that occurs along the $x_{1}$ axis, as expected. Regardless, the effects between the two models share similar trends justifying the approximation procedure.

Simple shear flow with $v_{1}=G x_{3}, v_{2}=v_{3}=0$ and diffusion coefficients of $\left(C_{1}, C_{2}\right)=\left(0, C_{I}\right)=\left(0,10^{-2}\right)$ provides a second view of the diffusion model characteristics. Unlike the uniaxial elongation case, the fibers are not symmetric about a single axis. Instead, due to shearing along $x_{3}$, the fiber alignment is not symmetric when viewed along the axis associated with the principal direction. In this example right after the initial isotropic distribution the peak of alignment is near the $x_{1}$ axis, but shifted toward the $x_{3}$ axis. As time increases, the peak of the distribution shifts toward the $x_{1}$ axis but never attains a large peak in the distribution as in the uniaxial case. As in the preceding example, Equation (6.8) is normalized with Equation (6.13) for values of $\left(C_{1}, C_{2}\right)=\left(1, C_{I}\right)=\left(1,10^{-2}\right)$ for the diffusion coefficients at $G t=1$, $G t=10$ and $G t \rightarrow \infty$. Results are presented in Figures 6.4(a), 6.4(c) and 6.4(e), respectively. For $G t=1$, the directional diffusion has two minima in the $\phi=0$ plane. The first minima near $\theta=\pi / 3$ corresponds with the peak of the fiber probability distribution $\psi(\theta, \phi)$ where a fiber aligned with the distribution of fibers will experience


Figure 6.3: Normalized directional diffusion $\hat{D}_{r}(\theta, \phi)$ for $C_{1}=1, C_{2}=C_{I}=10^{-2}$ for the fiber orientation distribution predicted from the Folgar and Tucker model for uniaxial elongation flow, $v_{1}=2 G x_{1}, v_{2}=-G x_{2} v_{3}=-G x_{3}$ at (a) $G t=\frac{1}{10}$, full diffusion (b) $G t=\frac{1}{10}$, approximate diffusion (c) $G t=1$, full diffusion (d) $G t=1$, approximate diffusion (e) $G t=\rightarrow \infty$, full diffusion (f) $G t=\rightarrow \infty$, approximate diffusion
very little diffusion. The second minima near $\theta=2 \pi / 3$ is due to the low probability of any fiber existing near that direction. The peaks for the normalized diffusion represent fiber angle pairs with a significant probability of occurance away from the peak of the probability of alignment. The valley between the two peaks in the normalized diffusion function exists because of the relatively small probability of a fiber lying near the plane $(\theta, \phi)=(\theta, \pi / 2)$ in this flow. As time increases, the probability of a fiber near the $x_{3}$ axis, corresponding to $\theta=0$ and $\theta=\pi$ decreases, correspondingly the diffusion near the $x_{3}$ axis diminishs with increasing time. The results for the approximate directional diffusion model given in Equation (6.12) are plotted at the same locations in Figures 6.4(b), 6.4(d) and 6.4(f). Observe the characteristics are very similar between the full model and the approximate model. The approximate form does tend to smooth the peaks and valleys of the diffusion, and is a reasonable approximation to capture the effects of the full model.

### 6.1.2 Applications with the Directional Diffusion Model

Directional diffusion was computed in the previous section using Equations (6.8) and (6.12) for a given $\psi(\theta, \phi)$ and known flow conditions. Unfortunately, the directional diffusion model will significantly increase the computational cost in evaluating Equation (2.23) for $\psi(\theta, \phi)$ due to the integration at every point on the sphere to obtain $D_{r}(\theta, \phi)$ during the transient solution. To resolve this issue, the diffusion model is recast in an appropriate form to alleviate some of the computational burden. Every term from Equation (6.12) containing $\left(\epsilon_{i j k} \rho_{i} p_{j} \dot{\tilde{\rho}}_{k}^{h}\right)^{2}$ may be expressed as a product of components $\rho_{i} \rho_{i} \dot{\tilde{\rho}}_{k}^{h} \dot{\tilde{\rho}}_{k}^{h}$ (no sum on $i$ of $k$ ) multiplied by products of the vector $\boldsymbol{p}$, and each term of Equation (6.12) containing $\left(\epsilon_{i j k} p_{i} \rho_{j} \dot{\hat{p}}_{k}^{h}\right)^{2}$ is expressed as a product of $p_{i} p_{i} \dot{\tilde{p}}_{k}^{h} \dot{\hat{p}}_{k}^{h}$ (no sum on $i$ or $k$ ) multiplied by products of the vector $\boldsymbol{\rho}$. The product,


Figure 6.4: Normalized directional diffusion $\hat{D}_{r}(\theta, \phi)$ for $C_{1}=1, C_{2}=C_{I}=10^{-2}$ for the fiber orientation distribution predicted from the Folgar and Tucker model for simple shear flow, $v_{1}=G x_{3}, v_{2}=v_{3}=0$ at (a) $G t=1$, full diffusion (b) $G t=1$, approximate diffusion (c) $G t=10$, full diffusion (d) $G t=10$, approximate diffusion (e) $G t=\rightarrow \infty$, full diffusion (f) $G t=\rightarrow \infty$, approximate diffusion
$\rho_{i} \rho_{i} \dot{\tilde{\rho}}_{k}^{h} \dot{\tilde{\rho}}_{k}^{h}$ defines the tensor $W_{i j k l}(\boldsymbol{\rho})$ and is expressed as

$$
\begin{align*}
W_{i j k l}(\boldsymbol{\rho})= & \rho_{i}(\theta, \phi) \rho_{j}(\theta, \phi) \dot{\tilde{p}}_{k}^{h}(\theta, \phi) \dot{\tilde{p}}_{l}^{h}(\theta, \phi) \\
= & \frac{1}{4} \lambda^{2} \dot{\gamma}_{k m} \dot{\gamma}_{l n} \rho_{i} \rho_{j} \rho_{m} \rho_{n}-\frac{1}{4} \lambda^{2} \dot{\gamma}_{k m} \dot{\gamma}_{n o} \rho_{i} \rho_{j} \rho_{l} \rho_{m} \rho_{n} \rho_{o}-\frac{1}{4} \lambda^{2} \dot{\gamma}_{l m} \dot{\gamma}_{n o} \rho_{i} \rho_{j} \rho_{k} \rho_{m} \rho_{n} \rho_{o} \\
& +\frac{1}{4} \lambda^{2} \dot{\gamma}_{m n} \dot{\gamma}_{o q} \rho_{i} \rho_{j} \rho_{k} \rho_{l} \rho_{m} \rho_{n} \rho_{o} \rho_{q} \tag{6.14}
\end{align*}
$$

The tensor $W_{i j k l}(\boldsymbol{p})$ experiences several symmetries, for example the first pair (or last pair) of indices, $i, j$ (or $k, l$ ) may be interchanged, i.e.,

$$
\begin{equation*}
W_{i j k l}=W_{j i k l}=W_{i j l k}=W_{j i l k} \tag{6.15}
\end{equation*}
$$

but the first pair $i, j$ may not, in general, be interchanged with the last pair $k, l$, i.e.,

$$
\begin{equation*}
W_{i j k l} \neq W_{k j i l} \quad W_{i j k l} \neq W_{k l i j} \tag{6.16}
\end{equation*}
$$

The tensor $W_{i j k l}(\boldsymbol{p})$ is identical to $W_{i j k l}(\boldsymbol{\rho})$ given in Equation (6.14) with the vector components $\rho_{i}$ replaced by $p_{i}$. It is useful to define the orientation average of the fourth-order tensor $W_{i j k l}(\boldsymbol{\rho})$ as the tensor $T_{i j k l}$ expressed as

$$
\begin{align*}
T_{i j k l} & =\oint_{\mathbb{S}^{2}} W_{i j k l}(\theta, \phi) \psi(\theta, \phi) \mathrm{d} \mathbb{S} \\
& =\frac{1}{4} \lambda^{2}\left(\dot{\gamma}_{k m} \dot{\gamma}_{l n} a_{i j m n}-\dot{\gamma}_{k m} \dot{\gamma}_{n o} a_{i j l m n o}-\dot{\gamma}_{l m} \dot{\gamma}_{n o} a_{i j k m n o}+\dot{\gamma}_{m n} \dot{\gamma}_{o p} a_{i j k l m n o p}\right) \tag{6.17}
\end{align*}
$$

The tensor $T_{i j k l}$ experiences the same symmetries as $W_{i j k l}(\boldsymbol{\rho})$ while not being fully symmetric as is the fourth-order orientation tensor $a_{i j k l}$. Note that $T_{i j k l}$ is a function of the orientation tensors up to the eighth-order.

After simplification, the directional diffusion model from Equation (6.12) may be
fully expanded as the expression

$$
\begin{aligned}
& D_{r}(\theta, \phi)=C_{2}\|\dot{\gamma}\|+\frac{C_{1}}{\|\dot{\gamma}\|}\left(p_{1}^{2}\left(T_{2233}+T_{3322}-2 T_{2323}\right)+p_{2}^{2}\left(T_{1133}+T_{3311}-2 T_{1313}\right)\right. \\
& \quad+p_{3}^{2}\left(T_{1122}+T_{2211}-2 T_{1212}\right)+2 p_{1} p_{2}\left(T_{1323}+T_{2313}-T_{3312}-T_{1233}\right) \\
& \quad+2 p_{1} p_{3}\left(T_{1232}+T_{3212}-T_{2213}-T_{1322}\right)+2 p_{2} p_{3}\left(T_{2131}+T_{3121}-T_{1123}-T_{2311}\right) \\
& \quad+a_{11}\left(W_{2233}+W_{3322}-2 W_{2323}\right)+a_{22}\left(W_{1133}+W_{3311}-2 W_{1313}\right) \\
& \quad+a_{33}\left(W_{1122}+W_{2211}-2 W_{1212}\right)+2 a_{12}\left(W_{1323}+W_{2313}-W_{3312}-W_{1233}\right) \\
& \left.\quad+2 a_{13}\left(W_{1232}+W_{3212}-W_{2213}-W_{1322}\right)+2 a_{23}\left(W_{2131}+W_{3121}-W_{1123}-W_{2311}\right)\right)(6.18)
\end{aligned}
$$

where the dependance of $p_{i}$ and $W_{i j k l}$ on $(\theta, \phi)$ is omitted for conciseness in the expression. It is noted that $T_{i j k l}$ is independent of $(\theta, \phi)$ but depends on the orientation tensors up to eighth-order. Observe that only 21 of the 81 components of the fourthorder tensors $T_{i j k l}$ and $W_{i j k l}(\theta, \phi)$ must be evaluated at each point in the evaluation process to solve the distribution function evaluation. There are several facts that may be utilized to streamline the evaluation procedure. For example, the fourth- and sixth-order orientation tensors are contained within the eighth-order orientation tensor, e.g. Equation (2.15), and may alleviate redundant multiplications. Many of the flow conditions considered throughout this work are for constant velocity gradients, therefore operations within the parentheses of Equations (6.14) and (6.17) need be evaluated only once if the computer's memory permits storage of the products. The same can be said of the unit vector $p_{i}$ which is a constant function independent of fiber orientation and flow conditions. The form of Equation (6.18) allows rapid computations of the diffusion function $D_{r}(\theta, \phi)$ once the orientation tensors are known, and has been seen to reduce the computational time to express the diffusion function expressed in Equation (6.12) by a factor of more than 400.

The distribution function evaluation procedures with control volumes as discussed in Bay [23] requires hours to days to evaluate the Folgar and Tucker model for diffusion, which is constant with respect to $(\theta, \phi)$. The new directional diffusion model requires significantly more evaluation time since at each time step it is necessary to evaluate the integral over the unit sphere, imposing a significant increase in the number of operations at a given time step by more than a factor of $45 \times N_{\theta} \times N_{\phi}$ where $N_{\theta}$ and $N_{\phi}$ are the number of unique grid points on the unit sphere used to solve the fiber orientation distribution function, and 45 is the number of independent components of the eighth-order orientation tensor. It was observed that computations with the Folgar and Tucker model require an order of magnitude less computation time than results with the directional diffusion function expressed in Equation (6.18).

### 6.1.3 Numerical Solution of the Orientation Distribution

The continuity equation given for $\psi(\theta, \phi)$ in Equation (2.23) with the directional diffusion model $D_{r}(\theta, \phi)$ in Equation (6.12) may be solved numerically in a manner similar to Advani [3], but the control volume method of Bay [23] is selected since it ensures global conservation. The following solution of $\psi(\theta, \phi)$ is developed following the procedure of Bay with only minor modifications to incorporate the directional dependance of the diffusion model. Unfortunately due to computational limitations, results with the directional diffusion model are not available at the present time and will be left to future endeavors. The following section is intended to facilitate future developments of distribution function simulations.

Solving the equations of motion with the method of control volumes begins with Jeffery's form of Equation (2.19) for the motion of a single fiber. Jeffery's equation is premultiplied by the Kronecker Delta tensor of Equation (2.8), and after simplification
may be expressed as

$$
\begin{align*}
\frac{d \boldsymbol{p}^{h}}{d t} & =\left(\lambda^{-} \boldsymbol{\kappa}^{T}: \boldsymbol{p} \boldsymbol{\theta}+\lambda^{+} \boldsymbol{\kappa}: \boldsymbol{p} \boldsymbol{\theta}\right) \boldsymbol{\theta}+\left(\lambda^{-} \boldsymbol{\kappa}^{T}: \boldsymbol{p} \boldsymbol{\phi}+\lambda^{+} \boldsymbol{\kappa}: \boldsymbol{p} \boldsymbol{\phi}\right) \boldsymbol{\phi} \\
& =\dot{\theta}^{h}(\theta, \phi) \boldsymbol{\theta}+\dot{\phi}^{h}(\theta, \phi) \boldsymbol{\phi} \tag{6.19}
\end{align*}
$$

where $\lambda^{-}=\frac{\lambda-1}{2}$ and $\lambda^{+}=\frac{\lambda+1}{2}$. The quantities

$$
\begin{align*}
& \dot{\theta}^{h}(\theta, \phi)=\left(\lambda^{-} \boldsymbol{\kappa}^{T}: \boldsymbol{p} \boldsymbol{\theta}+\lambda^{+} \boldsymbol{\kappa}: \boldsymbol{p} \boldsymbol{\theta}\right) \\
& \dot{\phi}^{h}(\theta, \phi)=\left(\lambda^{-} \boldsymbol{\kappa}^{T}: \boldsymbol{p} \phi+\lambda^{+} \boldsymbol{\kappa}: \boldsymbol{p} \phi\right) \tag{6.20}
\end{align*}
$$

are introduced for simplicity and represent the Jeffery motion of the fiber $\boldsymbol{p}$ in the $\boldsymbol{\theta}$ and $\boldsymbol{\phi}$ directions, respectively. The equation of motion for a single fiber with directional diffusion may be cast as the sum of the Jeffery effects and the directional diffusion effects as (see e.g. Bird [25])

$$
\begin{align*}
\frac{d \boldsymbol{p}}{d t}= & \frac{d \boldsymbol{p}^{h}}{d t}-\frac{1}{\psi(\theta, \phi)} \nabla\left(D_{r}(\theta, \phi) \psi(\theta, \phi)\right) \\
= & \left(\dot{\theta}^{h}(\theta, \phi)-\frac{1}{\psi(\theta, \phi)} \frac{\partial}{\partial \theta}\left(D_{r}(\theta, \phi) \psi(\theta, \phi)\right)\right) \boldsymbol{\theta} \\
& +\left(\dot{\phi}^{h}(\theta, \phi)-\frac{1}{\psi(\theta, \phi)} \frac{\partial}{\sin \theta \partial \phi}\left(D_{r}(\theta, \phi) \psi(\theta, \phi)\right)\right) \boldsymbol{\phi} \\
= & \dot{\theta}(\theta, \phi) \boldsymbol{\theta}+\dot{\phi}(\theta, \phi) \boldsymbol{\phi} \tag{6.21}
\end{align*}
$$

where

$$
\begin{align*}
& \dot{\theta}(\theta, \phi)=\left(\dot{\theta}^{h}(\theta, \phi)-\frac{1}{\psi(\theta, \phi)} \frac{\partial}{\partial \theta}\left(D_{r}(\theta, \phi) \psi(\theta, \phi)\right)\right) \\
& \dot{\phi}(\theta, \phi)=\left(\dot{\phi}^{h}(\theta, \phi)-\frac{1}{\psi(\theta, \phi)} \frac{\partial}{\sin \theta \partial \phi}\left(D_{r}(\theta, \phi) \psi(\theta, \phi)\right)\right) \tag{6.22}
\end{align*}
$$

represent the fiber motion in the $\boldsymbol{\theta}$ and $\boldsymbol{\phi}$ directions, respectively, due to hydrodynamic effects and diffusion.

The control volume method (see e.g. [106]) is employed here to solve the fiber distribution function $\psi(\theta, \phi)$ on the sphere $\mathbb{S}^{2}$ as discussed in Bay [23]. In the control


Figure 6.5: Control volume employed for the flux balance of the fiber orientation distribution.
volume method, the sphere $\mathbb{S}^{2}$ is meshed with a uniform grid of control volumes at the points $\left(\theta_{i}, \phi_{j}\right)$ for $i \in\left\{1,2, \ldots, N_{\theta}\right\}$ and $j \in\left\{1,2, \ldots, N_{\phi}\right\}$. Each control volume such as that shown in Figure 6.5 has an incremental area $\Delta A$ equal to the product of the sides, $\Delta \theta$ and $\Delta \phi \sin \theta$. The quantity within the control volume is given by $\psi(\theta, \phi) \sin \theta \Delta \theta \Delta \phi$ which has a time rate of change within the control volume [23]

$$
\begin{equation*}
\frac{d}{d t}\left(\psi\left(\theta_{i}, \phi_{j}\right) \Delta A\right) \simeq \frac{\psi^{t+1}\left(\theta_{i}, \phi_{j}\right)-\psi^{t}\left(\theta_{i}, \phi_{j}\right)}{\Delta t} \Delta A \tag{6.23}
\end{equation*}
$$

This rate of change of $\psi(\theta, \phi) \Delta A$ is equal to the balance of the fluxes passing through the sides of the control volume in Figure 6.5. The flux from the West face is equal to the length of the side multiplied by the velocity, $\dot{\theta}(\theta, \phi)$ at the West face as

$$
\begin{align*}
& \left.\operatorname{Flux}_{W}\left(\theta_{i}, \phi_{j}\right)=\left.\psi(\theta, \phi) \dot{\theta}(\theta, \phi) \sin \theta \Delta \phi\right|_{(\theta, \phi)=\left(\theta_{i-\frac{1}{2}}, \phi_{j}\right.}\right) \\
& =\left.\psi(\theta, \phi)\left(\dot{\theta}^{h}(\theta, \phi)-\frac{\partial}{\partial \theta}\left(D_{r}(\theta, \phi) \psi(\theta, \phi)\right)\right) \sin \theta \Delta \phi\right|_{(\theta, \phi)=\left(\theta_{i-\frac{1}{2}, \phi_{j}}\right)}(6 \tag{6.24}
\end{align*}
$$

Similarly, the flux on the East face is

$$
\begin{align*}
& \operatorname{Flux}_{E}\left(\theta_{i}, \phi_{j}\right)=\left.\psi(\theta, \phi) \dot{\theta}(\theta, \phi) \sin \theta \Delta \phi\right|_{(\theta, \phi)=\left(\theta_{i+\frac{1}{2}}, \phi_{j}\right)} \\
& =\left.\psi(\theta, \phi)\left(\dot{\theta}^{h}(\theta, \phi)-\frac{\partial}{\partial \theta}\left(D_{r}(\theta, \phi) \psi(\theta, \phi)\right)\right) \sin \theta \Delta \phi\right|_{(\theta, \phi)=\left(\theta_{i+\frac{1}{2}}, \phi_{j}\right)} \tag{6.25}
\end{align*}
$$

The flux on the North and South faces are the products of the velocity at their respective faces multiplied by the length of the corresponding side expressed as

$$
\begin{align*}
& \operatorname{Flux}_{S}\left(\theta_{i}, \phi_{j}\right)=\left.\psi(\theta, \phi) \dot{\phi}(\theta, \phi) \Delta \theta\right|_{(\theta, \phi)=\left(\theta_{i}, \phi_{j-\frac{1}{2}}\right)} \\
& \qquad=\left.\psi(\theta, \phi)\left(\dot{\phi}^{h}(\theta, \phi)-\frac{1}{\sin \theta} \frac{\partial}{\partial \phi}\left(D_{r}(\theta, \phi) \psi(\theta, \phi)\right)\right) \Delta \theta\right|_{(\theta, \phi)=\left(\theta_{i}, \phi_{j-\frac{1}{2}}\right)}  \tag{6.26}\\
& \text { Flux }_{N}\left(\theta_{i}, \phi_{j}\right)=\left.\psi(\theta, \phi) \dot{\phi}(\theta, \phi) \Delta \theta\right|_{(\theta, \phi)=\left(\theta_{i}, \phi_{j+\frac{1}{2}}\right)} \\
& \quad=\left.\psi(\theta, \phi)\left(\dot{\phi}^{h}(\theta, \phi)-\frac{1}{\sin \theta} \frac{\partial}{\partial \phi}\left(D_{r}(\theta, \phi) \psi(\theta, \phi)\right)\right) \Delta \theta\right|_{(\theta, \phi)=\left(\theta_{i}, \phi_{j+\frac{1}{2}}\right)} \tag{6.27}
\end{align*}
$$

The balance from Equation (6.23) is solved by the sum of the fluxes expressed as

$$
\begin{align*}
\frac{\psi^{t+1}\left(\theta_{i}, \phi_{j}\right)-\psi^{t}\left(\theta_{i}, \phi_{j}\right)}{\Delta t} \Delta A & =\operatorname{Flux}_{W}\left(\theta_{i}, \phi_{j}\right)-\operatorname{Flux}_{E}\left(\theta_{i}, \phi_{j}\right) \\
& +\operatorname{Flux}_{S}\left(\theta_{i}, \phi_{j}\right)-\operatorname{Flux}_{N}\left(\theta_{i}, \phi_{j}\right) \tag{6.28}
\end{align*}
$$

In the above, each of the terms $\dot{\theta}^{h}(\theta, \phi)$ and $\dot{\phi}^{h}(\theta, \phi)$ are analytic functions which are evaluated numerically for all values of $(\theta, \phi) \in \mathbb{S}^{2}$. Conversely, the distribution function is only known at the grid points $\left(\theta_{i}, \phi_{j}\right)$, therefore the distribution function on a given face of the control volume is approximated as

$$
\begin{align*}
\psi\left(\theta_{i \pm \frac{1}{2}}, \phi_{j}\right) & \simeq \frac{\psi\left(\theta_{i \pm 1}, \phi_{j}\right)+\psi\left(\theta_{i}, \phi_{j}\right)}{2}  \tag{6.29}\\
\psi\left(\theta_{i}, \phi_{j \pm \frac{1}{2}}\right) & \simeq \frac{\psi\left(\theta_{i}, \phi_{j \pm 1}\right)+\psi\left(\theta_{i}, \phi_{j}\right)}{2} \tag{6.30}
\end{align*}
$$

The derivative with respect to $\theta$ of the product of the functions $\psi(\theta, \phi) D_{r}(\theta, \phi)$
on the West face is approximated using the chain rule as

$$
\begin{align*}
\frac{\partial}{\partial \theta} & \left.\left.\left(\psi(\theta, \phi) D_{r}(\theta, \phi)\right)\right|_{(\theta, \phi)=\left(\theta_{i-\frac{1}{2}} \phi_{j}\right.}\right) \\
& =\left.\left(\frac{\partial \psi(\theta, \phi)}{\partial \theta} D_{r}(\theta, \phi)+\psi(\theta, \phi) \frac{\partial D_{r}(\theta, \phi)}{\partial \theta}\right)\right|_{(\theta, \phi)=\left(\theta_{i-\frac{1}{2}} \phi_{j}\right)} \\
& \simeq \frac{\psi\left(\theta_{i}, \phi_{j}\right)-\psi\left(\theta_{i-1}, \phi_{j}\right)}{\Delta \theta} \frac{D_{r}\left(\theta_{i}, \phi_{j}\right)+D_{r}\left(\theta_{i-1}, \phi_{j}\right)}{2} \\
& +\frac{\psi\left(\theta_{i}, \phi_{j}\right)+\psi\left(\theta_{i-1}, \phi_{j}\right)}{2} \frac{D_{r}\left(\theta_{i}, \phi_{j}\right)-D_{r}\left(\theta_{i-1}, \phi_{j}\right)}{\Delta \theta} \\
& =\frac{\psi\left(\theta_{i}, \phi_{j}\right) D_{r}\left(\theta_{i}, \phi_{j}\right)-\psi\left(\theta_{i-1}, \phi_{j}\right) D_{r}\left(\theta_{i-1}, \phi_{j}\right)}{\Delta \theta} \tag{6.31}
\end{align*}
$$

The remaining derivatives are similarly obtained, and after much simplification the balance equation in Equation (6.28) may be expressed as

$$
\begin{align*}
& \frac{\psi^{t+1}\left(\theta_{i}, \phi_{j}\right)-\psi^{t}\left(\theta_{i}, \phi_{j}\right)}{\Delta t} \Delta A= \\
& \psi\left(\theta_{i-1}, \phi_{j}\right) \Delta \phi \sin \theta_{i-\frac{1}{2}}\left(\frac{\dot{\theta}\left(\theta_{i-\frac{1}{2}}, \phi_{j}\right)}{2}+\frac{D_{r}\left(\theta_{i-1}, \phi_{j}\right)}{\Delta \theta}\right) \\
& -\psi\left(\theta_{i+1}, \phi_{j}\right) \Delta \phi \sin \theta_{i+\frac{1}{2}}\left(\frac{\dot{\theta}\left(\theta_{i+\frac{1}{2}}, \phi_{j}\right)}{2}-\frac{D_{r}\left(\theta_{i+1}, \phi_{j}\right)}{\Delta \theta}\right) \\
& \quad+\psi\left(\theta_{i}, \phi_{j-1}\right) \Delta \theta\left(\frac{\dot{\phi}\left(\theta_{i}, \phi_{j-\frac{1}{2}}\right)}{2}+\frac{D_{r}\left(\theta_{i}, \phi_{j-1}\right)}{\Delta \theta}\right) \\
& \quad+\psi\left(\theta_{i}, \phi_{j+1}\right) \Delta \theta\left(\frac{\dot{\phi}\left(\theta_{i}, \phi_{j+\frac{1}{2}}\right)}{2}-\frac{D_{r}\left(\theta_{i}, \phi_{j+1}\right)}{\Delta \theta}\right) \\
& \quad+\psi\left(\theta_{i}, \phi_{j}\right)\left(\Delta \phi \sin \theta_{i-\frac{1}{2}}\left(\frac{\dot{\theta}\left(\theta_{i-\frac{1}{2}}, \phi_{j}\right)}{2}-\frac{D_{r}\left(\theta_{i}, \phi_{j}\right)}{\Delta \theta}\right)\right. \\
& \quad-\Delta \phi \sin \theta_{i+\frac{1}{2}}\left(\frac{\dot{\theta}\left(\theta_{i+\frac{1}{2}}, \phi_{j}\right)}{2}+\frac{D_{r}\left(\theta_{i}, \phi_{j}\right)}{\Delta \theta}\right) \\
& \left.\quad+\Delta \theta\left(\frac{\dot{\phi}\left(\theta_{i}, \phi_{j-\frac{1}{2}}\right)}{2}-\frac{D_{r}\left(\theta_{i}, \phi_{j}\right)}{\Delta \theta}\right)-\Delta \theta\left(\frac{\dot{\phi}\left(\theta_{i}, \phi_{j+\frac{1}{2}}\right)}{2}+\frac{D_{r}\left(\theta_{i}, \phi_{j}\right)}{\Delta \theta}\right)\right)(6 \tag{6.32}
\end{align*}
$$

This form expresses the dependance of the change in the fiber orientation distribution function $\psi(\theta, \phi)$ on the diffusion $D_{r}(\theta, \phi)$ with an explicit angular dependance. In Equation (6.32), the expression will reduce to that given in Bay [23] for a diffusion model with no angular dependance, i.e. $D_{r}(\theta, \phi)=C_{I}\|\dot{\gamma}\|$.

The periodic boundary conditions expressed in Bay [23] ensure global conservation and are given as

$$
\begin{align*}
& \psi(\theta, \phi)=\psi(\pi-\theta, \phi+\pi) \rightarrow \psi\left(\theta_{i}, \phi_{0}\right)=\psi\left(\theta_{N_{\theta}+1-i}, \phi_{N_{\phi}}\right), \psi\left(\theta_{i}, \phi_{N_{\phi}+1}\right)=\psi\left(\theta_{N_{\theta}+1-i}, \phi_{1}\right) \\
& \psi(\theta, \phi)=\psi(\theta+\pi, \phi) \rightarrow \psi\left(\theta_{0}, \phi_{j}\right)=\psi\left(\theta_{N_{\theta}}, \phi_{j}\right), \psi\left(\theta_{N_{\theta}+1}, \phi_{j}\right)=\psi\left(\theta_{1}, \phi_{j}\right) \tag{6.33}
\end{align*}
$$

Equation (6.32) may be solved on a single hemisphere of the unit sphere once an appropriate choice of step size and grid points are chosen. Bay [23] outlines a procedure to select the appropriate step size and grid points for the diffusion model of Folgar and Tucker [11]. Unfortunately to select a time step and mesh size assumes a constant diffusion term, and as such the procedure of Bay can only serve as a guideline when $D_{r}(\theta, \phi)$ in Equation (6.12) is employed.

### 6.1.4 Directional Diffusion for Orientation Tensor Solutions

Due to the computational expenses required to solve $\psi(\theta, \phi)$, it is necessary to express the directional diffusion effects on fiber orientation through the second-order orientation tensor flow equations of Equation (2.34). The expression for the secondorder orientation tensor given in Equation (2.34) is solved with the diffusion model of Equation (6.12), expressed in component form given in Equation (6.18), and after
simplification may be expressed as (see Jack [45] for the full derivation)

$$
\begin{align*}
& \oint_{\mathbb{S}^{2}} D_{r} \psi\left(2 \delta_{i j}-6 p_{i} p_{j}\right) \mathrm{d} \mathbb{S}=C_{2}\|\dot{\gamma}\|\left(2 \delta_{i j}-6 a_{i j}\right)+\frac{C_{1}}{\|\dot{\gamma}\|}( \\
& \quad+\left(4 \delta_{i j} a_{11}-6 a_{i j 11}\right)\left(T_{2233}+T_{3322}-2 T_{2323}\right)+\left(8 \delta_{i j} a_{12}-12 a_{i j 12}\right)\left(T_{1323}+T_{2313}-T_{3312}-T_{1233}\right) \\
& +\left(4 \delta_{i j} a_{22}-6 a_{i j 22}\right)\left(T_{1133}+T_{3311}-2 T_{1313}\right)+\left(8 \delta_{i j} a_{13}-12 a_{i j 13}\right)\left(T_{1232}+T_{3212}-T_{2213}-T_{1322}\right) \\
& +\left(4 \delta_{i j} a_{33}-6 a_{i j 33}\right)\left(T_{1122}+T_{2211}-2 T_{1212}\right)+\left(8 \delta_{i j} a_{23}-12 a_{i j 23}\right)\left(T_{2131}+T_{3121}-T_{1123}-T_{2311}\right) \\
& -6 a_{11}\left(Z_{i j 2233}+Z_{i j 3322}-2 Z_{i j 2323}\right)+12 a_{12}\left(Z_{i j 1323}+Z_{i j 2313}-Z_{i j 3312}-Z_{i j 1233}\right) \\
& \quad-6 a_{22}\left(Z_{i j 1133}+Z_{i j 3311}-2 Z_{i j 1313}\right)+12 a_{13}\left(Z_{i j 1232}+Z_{i j 3212}-Z_{i j 2213}-Z_{i j 1322}\right) \\
& \left.-6 a_{33}\left(Z_{i j 1122}+Z_{i j 2211}-2 Z_{i j 1212}\right)+12 a_{23}\left(Z_{i j 2131}+Z_{i j 3121}-Z_{i j 1123}-Z_{i j 2311}\right)\right) \tag{6.34}
\end{align*}
$$

The fourth-order tensor $T_{i j k l}$ is given in Equation (6.17) and the sixth-order tensor $Z_{i j k l m n}$ is defined as the integration of the products $p_{i} p_{j}$ with the function $W_{k l m n}(\theta, \phi)$ given in Equation (6.14), and is expressed as

$$
\begin{align*}
Z_{i j k l m n} & =\oint_{\mathbb{S}^{2}} p_{i}(\theta, \phi) p_{j}(\theta, \phi) p_{k}(\theta, \phi) p_{l}(\theta, \phi) \dot{\tilde{p}}_{m}^{h}(\theta, \phi) \dot{\tilde{p}}_{n}^{h}(\theta, \phi) \psi(\theta, \phi) \mathrm{dS} \\
& =\frac{1}{4} \lambda^{2}\left(\dot{\gamma}_{m o} \dot{\gamma}_{n p} a_{i j k l o p}-\dot{\gamma}_{m o} \dot{\gamma}_{p q} a_{i j k l n o p q}-\dot{\gamma}_{n o} \dot{\gamma}_{p q} a_{i j k l m o p q}+\dot{\gamma}_{o p} \dot{\gamma}_{q r} a_{i j k l m n o p q r}\right) \tag{6.35}
\end{align*}
$$

Equation (6.34) simplifies to the Folgar and Tucker model when the coefficient pair $\left(C_{1}, C_{2}\right)$ set to $\left(0, C_{I}\right)$, but more importantly the directional diffusion expression in Equation (6.34) provides a means to incorporate the directional dependance for diffusion when computing $a_{i j}$. It was observed that solutions of Equation (6.34) from the directional diffusion model required nearly 5 times the computational expense as solutions for the Folgar and Tucker model where $C_{1}=0$.

Observe that orientation tensors up to tenth-order appear in Equation (6.34) through each of the expressions $Z_{i j k l m n}$ given in Equation (6.35), and introduces complications for solutions of the orientation tensor evolution equation. The selection
of an approximation for the higher order information is necessary to obtain a solution of the second-order orientation tensor. The analytical hybrid closure was investigated in [45], but it has been shown to over predict the fiber alignment state in industrial simulations [59, 107]. Therefore the fourth-order ORT closure used by VerWeyst and Tucker [15] is employed in this work to approximate the fourth-order orientation tensor from the second-order orientation tensor. The sixth-order $\mathrm{INV}_{6}$ closure of Jack and Smith [38] given in Equation (3.15) is selected to compute the sixth-order orientation tensor once the fourth- and second-order orientation tensors are known. Unfortunately, there exists little mention in the literature of higher order closures for tensors beyond $a_{i j k l m n}$. An eighth-order approximation similar in form to the sixth-order quadratic closure of Doi [48] is introduced as

$$
\begin{equation*}
a_{i j k l m n o q} \approx a_{i j k l m n} a_{o q} \tag{6.36}
\end{equation*}
$$

Similarly, a tenth-order approximation as a function of the eighth-order orientation tensor is proposed as

$$
\begin{equation*}
a_{i j k l m n o q r s} \approx a_{i j k l m n o q} a_{r s} \tag{6.37}
\end{equation*}
$$

where for Equations (6.36) and (6.37) $i, j, k, l, m, n, o, q, r, s \in\{1,2,3\}$. In this case, note that the combination of the eighth- and tenth-order approximations yield $a_{i j k l m n o q r s} \approx a_{i j k l m n} a_{o q} a_{r s}$. Both of these eighth- and tenth-order tensor approximations do not satisfy the symmetry conditions in Equation (2.12), but Equations (6.36) and (6.37) exactly solve the higher-order orientation tensors for unidirectional alignment states while tending to over predict the degree of alignment for all other orientation states. An accurate analysis with the directional diffusion model must account for the correct form of the orientation tensors, and it is worthwhile to pursue in future research.

### 6.2 Flow Examples with Directional Diffusion

In this section the qualitative behavior of the directional diffusion model is investigated where refinements/verification of a directional diffusion model with experimental data are left to future endeavors. Calculated results are compared to those obtained with the Folgar and Tucker [11] model for diffusion. This model is used as the benchmark here since it was developed with an interaction coefficient $C_{I}$ to fit the numerical results to experimental data, often at steady state $[1,3,14,19,20,23,33,55,56,65,108-110]$. Therefore the steady state results for the directionally dependant diffusion model in Equation (6.12) will be required to be similar to the predicted steady state results from the Folgar and Tucker model.

The second-order orientation tensor flow equation of Equation (2.35) is solved numerically with the directional diffusion representation given in Equation (6.34) with a fourth-order Runge-Kutta solution procedure (see e.g. [111]). In these simulations, the fourth-order orientation tensor $a_{i j k l}$ is approximated with the orthotropic closure ORT given by Wetzel [32] and used by VerWeyst and Tucker [15]. The sixth-order orientation tensor $a_{i j k l m n}$ is approximated from $a_{i j k l}$ with the $\mathrm{INV}_{6}$ closure of Jack and Smith [38], and the eighth- and tenth-order orientation tensors are approximated with Equations (6.36) and (6.37), respectively. The orientation state is initially isotropic $\psi(\theta, \phi)=\frac{1}{4 \pi}$ in each of the following examples (see e.g. Figure 5.1) which is represented in terms of the second-order orientation tensor given in Equation (2.17). It has been shown that flows indicative of those found in industrial applications are well represented using interaction coefficients of $10^{-4} \leq C_{I} \leq 10^{-1}[1,19,56]$ in the Folgar and Tucker model. Therefore examples shown below are for an interaction coefficient of $10^{-3}$ to predict the desired steady state results.

### 6.2.1 Uniaxial Elongation Results

Elongational flow occurs in injection molding processes when the cavity containing the polymer fluid expands rapidly. In the following examples, the velocity field is characterized by $v_{1}=2 G x_{1}, v_{2}=-G x_{2}$ and $v_{3}=-G x_{3}$ where $G$ is a scaling parameter. In the absence of experimental data, the coefficient pair $\left(C_{1}, C_{2}\right)$ will be selected such that steady state results from the Folgar and Tucker model are nearly attained. Recall, that when $\left(C_{1}, C_{2}\right)=\left(0, C_{I}\right)$ in Equation (6.34) the directional diffusion model simplifies to the Folgar and Tucker results. As such all results for the steady state from the second-order orientation tensor equation of motion with the directional diffusion model will be compared to steady state results from the Folgar and Tucker model for an interaction coefficient of $C_{I}=10^{-3}$. The procedure to select a $C_{2}$ for a fixed $C_{1}$ is quantified by minimizing the parameter $\chi^{2}\left(C_{2}\right)$ defined as

$$
\begin{equation*}
\chi^{2}\left(C_{2}\right)=\left(a_{i j}^{S S}\left(0,10^{-3}\right)-a_{i j}^{S S}\left(C_{1}, C_{2}\right)\right)^{2} \tag{6.38}
\end{equation*}
$$

where $a_{i j}^{S S}\left(C_{1}, C_{2}\right)$ is the steady state second-order orientation tensor from the directional diffusion model with the coefficient pair $\left(C_{1}, C_{2}\right)$ and $a_{i j}^{S S}\left(0,10^{-3}\right)$ is the steady state second-order orientation tensor obtained without directional diffusion. The coefficient $\chi^{2}$ is minimized by changing $C_{2}$ for a given $C_{1}$ for uniaxial elongation, and results shown in Figure 6.6 indicate the directional diffusion model yields the same steady state results as the Folgar and Tucker model for $C_{1} \in(0,7)$ with very little change in $C_{2}$. The optimized results for $C_{2}$ as a function of $C_{1}$ show a nearly linear response between $C_{2}$ and $C_{1}$ with the linear fit between $C_{2}$ and $C_{1}$ given as

$$
\begin{equation*}
C_{2}=-5 \times 10^{-6} C_{1}+1 \times 10^{-3} \tag{6.39}
\end{equation*}
$$

with an $R^{2}$ value of 0.9999 . The slope of the line is $5 \times 10^{-6}$ which is several orders


Figure 6.6: Diffusion coefficient $C_{2}$ as a function of $C_{1}$ for uniaxial elongation flow, $v_{1}=2 G x_{1}, v_{2}=-G x_{2}, v_{3}=-G x_{3}$ to yield the same steady state as Folgar and Tucker model results.
of magnitude less than the $C_{2}$-axis intercept point $10^{-3}$. Since the range of applicable $C_{1}$ coefficients is less than $10^{0}$, it is assumed in the following results that $C_{2}$ is a constant equal to $C_{2}=10^{-3}$ for $C_{1} \in(0,7)$.

The results for select components of the second-order orientation tensor from the evolution of $a_{i j}$ in uniaxial elongational flow from an initial isotropic orientation state are given in Figure 6.7. The results without directional diffusion, i.e. $\left(C_{1}, C_{2}\right)=$ $\left(0,10^{-3}\right)$, are represented by the solid line for select components of the second-order orientation tensor where the steady state solution is nearly attained when $G t \simeq 2$. The directional diffusion model results for uniaxial flow from the coefficient pairs $\left(C_{1}, C_{2}\right)=\left(1,10^{-3}\right),\left(C_{1}, C_{2}\right)=\left(3,10^{-3}\right),\left(C_{1}, C_{2}\right)=\left(6,10^{-3}\right),\left(C_{1}, C_{2}\right)=\left(7,10^{-3}\right)$ and $\left(C_{1}, C_{2}\right)=\left(8,10^{-3}\right)$ are also shown in Figure 6.7. Notice how the rate at which the fibers align as viewed through the $a_{11}$ component is diminished as the value for the


Figure 6.7: Transient Solution for $a_{i j}$ from the directional diffusion model in uniaxial elongation flow, $v_{1}=2 G x_{1}, v_{2}=-G x_{2}, v_{3}=-G x_{3}$ starting from an isotropic orientation state, $\psi(\theta, \phi)=\frac{1}{4 \pi}$.
coefficient $C_{1}$ increases. For values of $C_{1} \in(0,7)$, the coefficient $C_{2}$ may be set to $10^{-3}$ and still produce the same steady state value as obtained for $\left(C_{1}, C_{2}\right)=\left(0,10^{-3}\right)$ to within $0.1 \%$ for each of the components of $a_{i j}$. As $C_{1}$ increases beyond 7 , the value for the coefficient $C_{2}$ must be significantly reduced to offset the increased diffusion. It was observed that $C_{2}$ could not be reduced below 0 without the orientation state becoming non-physical, and will be discussed further in the simple shear flow example. Recall that the directional model for diffusion is constructed such that as fibers approach unidirectional alignment, the local fiber collision effects go to zero and only the volume averaged Brownian-type effects remain. Since alignment is rarely at unidirectional, local fiber collision effects will rarely go to zero. As such the directional diffusion model will tend to delay alignment due to over predicting the actual diffusion at
steady state, and the parameter $C_{2}$ will decrease as $C_{1}$ increases. For uniaxial, these decreases were minimal and are able to be neglected.

To ensure correct steady state values, there is a limit to the degree of applied directional diffusion. Nearly all coefficient pairs in Figure 6.7 yield a steady state of approximately $a_{11}=0.995$ and $a_{22}=a_{33}=0.0025$ from an initially isotropic fiber orientation. Note however, that the coefficient pair $\left(C_{1}, C_{2}\right)=(8,0)$ produces a steady state near $a_{11}=0.6$ and $a_{22}=a_{33}=0.2$ from the same initial isotropic orientation. For $C_{1}=8$, the coefficient $C_{2}$ is decreased toward zero, but never yields a steady state alignment above $a_{11}=0.6$ and $a_{22}=a_{33}=0.2$. The directional diffusion pair $\left(C_{1}, C_{2}\right)=(8,0)$ are not irrelevant, and present some interesting characteristics. For example, given two different initial orientation states with the alignments $a_{11}=$ $0.7, a_{22}=a_{33}=0.15$ and $a_{11}=0.75, a_{22}=a_{33}=0.125$, two different steady state orientations are predicted, as shown in Figure 6.8. The orientation state $a_{11}=$ $0.7, a_{22}=a_{33}=0.15$ yields a lower orientation state than highly aligned, whereas the orientation state $a_{11}=0.75, a_{22}=a_{33}=0.125$ evolves to the unidirectional orientation state similar to that appearing in Figure 6.7. This is expected since as alignment approaches unidirectional with $C_{2}=0$, local fiber interactions go to zero and fiber motion simply follows Jeffery's equation.

The directional diffusion model is developed to affect the transient solution with minimal affects on the steady state orientation. As viewed in Figure 6.7 steady state orientation remains the same (within $\sim 0.1 \%$ ) as results from the Folgar and Tucker model, even though the transient solution is significantly altered. A quantitative metric to assess when steady state is attained is defined as

$$
\begin{equation*}
\Delta a_{i j}(t)=\frac{a_{i j}^{S S}-a_{i j}(t)}{a_{i j}^{S S}} \times 100 \% \tag{6.40}
\end{equation*}
$$



Figure 6.8: Transient Solution for $a_{i j}$ from the directional diffusion model in uniaxial elongation flow.
where $a_{i j}^{S S}$ is the true steady state value for the $a_{i j}$ component. Steady state is considered to be nearly attained when $\left|\Delta a_{11}\right|$ remains less than $1 \%$. This metric is presented in Figure 6.9 for select diffusion coefficient pairs in uniaxial elongational flow, and the $\pm 1 \%$ difference is indicated by the solid horizontal lines. As the coefficient $C_{1}$ increases, the point in time where $\left|\Delta a_{11}\right|$ remains below $1 \%$ occurs at a later point in time. This occurs for the coefficient pair $\left(C_{1}, C_{2}\right)=\left(7,10^{-3}\right)$ at a time more than three times that of $\left(C_{1}, C_{2}\right)=\left(0,10^{-3}\right)$.

The directional diffusion model in uniaxial elongational flow significantly alters the rate at which the fiber distribution becomes aligned. This effect can be viewed in Figures 6.10 and 6.11 where, respectively, the magnitude of the derivative of the $a_{11}$ and $a_{22}$ component are presented. For time $G t \in(0, \sim 0.8)$ the slope for both $a_{11}$ and $a_{22}$ decreases with increasing $C_{1}$, but as time increases the directional diffusion


Figure 6.9: Percentage difference between steady state orientation and transient orientation state for $a_{11}$ in uniaxial elongation flow starting from an isotropic orientation.
model has a higher rate of alignment for both $a_{11}$ and $a_{22}$. It is unreasonable to view the slope as a function of time since the second-order orientation tensor is nearly at steady state for $G t>2$ when $\left(C_{1}, C_{2}\right)=\left(0,10^{-3}\right)$, as viewed in Figure 6.7. This is equivalent to stating the derivative of $a_{i j}$ goes to zero for $G t$ greater than 2 without directional diffusion.

A more reasonable comparison is to observe the rate of change of the second-order orientation tensor as a function of orientation during the flow history. The principal eigenvalue of the second-order orientation tensor $a_{(1)}$ is selected as the parameter to indicate the degree of orientation. Figures 6.12 and 6.13 contain results for the magnitude of the slope as a function of alignment for $a_{11}$ and $a_{22}$, respectively. The parameter $a_{(1)}$ has a value of $\frac{1}{3}$ for the initial isotropic orientation and increases to a value approaching 1 when fibers are nearly aligned. Both Figures 6.12 and 6.13


Figure 6.10: Magnitude of the slope for $a_{11}(t)$ in uniaxial elongation flow, starting from an isotropic orientation state.


Figure 6.11: Magnitude of the slope for $a_{22}(t)$ in uniaxial elongation flow, starting from an isotropic orientation state.
demonstrate that the rate of alignment decreases with increasing $C_{1}$ throughout much of the flow history. The reverse behavior near isotropic orientation states is expected since the directional diffusion function $D_{r}(\theta, \phi)$ of Equation (6.12) is quite large for random distributions, and decreases as alignment increases (see e.g., Figure 6.3).

### 6.2.2 Simple Shear

The second example considered is that of simple shear flow which has a non-zero velocity in the $x_{1}$ direction and a constant shear rate in the $x_{3}$ direction given by the velocity components $v_{1}=G x_{3}, v_{2}=v_{3}=0$. As discussed in Section 3.1.4.1, fibers will tend to orient along the $x_{1}$ axis, and in the absence of fiber collisions, fibers approach a unidirectional orientation with time. As in the preceding example, coefficient pairs $\left(C_{1}, C_{2}\right)$ are selected to match the steady state results predicted without directional diffusion, i.e., $\left(C_{1}, C_{2}\right)=\left(0,10^{-3}\right)$. The parameter $\chi^{2}\left(C_{2}\right)$ from Equation (6.38) is minimized with $a_{i j}^{S S}\left(0,10^{-3}\right)$ being the steady state second-order orientation tensor without directional diffusion. As above, the coefficient $\chi^{2}\left(C_{2}\right)$ is minimized by changing $C_{2}$ for a given $C_{1}$ where the results for $C_{1} \in(0,0.06)$ are shown in Figure 6.14. The values for $C_{2}$ as a function of $C_{1}$ for $C_{1} \in(0,0.06)$ are nearly a perfect linear fit with $R^{2}=0.99997$ for the least squares best fit curve

$$
\begin{equation*}
C_{2}=-1.62 \times 10-2 C_{1}+1 \times 10^{-3} \tag{6.41}
\end{equation*}
$$

Unlike the uniaxial elongational flow example, the diffusion coefficient $C_{2}$ is strongly dependant on the choice of $C_{1}$. Additionally, the range for $C_{1}$ in simple shear is much smaller than the uniaxial elongation results, $C_{1} \in(0,0.06)$ versus $C_{1} \in(0,7)$. Although results for $C_{1} \geq 0.06$ diminish the rate of fiber alignment, the corresponding value for $C_{2}$ to ensure the correct steady state in simple shear flow is negative. A


Figure 6.12: Magnitude of slope for $a_{11}(t)$ in uniaxial elongation flow as a function of alignment parameter $a_{(1)}$.


Figure 6.13: Magnitude of slope for $a_{22}(t)$ in uniaxial elongation flow as a function of alignment parameter $a_{(1)}$.


Figure 6.14: Diffusion coefficient $C_{2}$ as a function of $C_{1}$ for simple shear flow, $v_{1}=$ $2 G x_{1}, v_{2}=-G x_{2}, v_{3}=-G x_{3}$ to yield the same steady state as Folgar and Tucker model results.
negative value for $C_{2}$ must be discarded since in select orientation states $D_{r}(\theta, \phi)$ as given in Equation (6.12) will yield a negative diffusion implying an increase in the energy of the system. Take, for example, a uniaxial alignment state, i.e., $a_{i j}=$ $a_{i j k l}=a_{i j k l m n}=a_{i j k l m n o p}=a_{i j k l m n o p q r}=0, \forall i, j, k, l, m, n, o, p, q, r \in\{1,2,3\}$ except $a_{11}=a_{1111}=a_{111111}=a_{11111111}=a_{1111111111}=1$, the expression multiplied by $C_{1}$ in Equation (6.12) is zero and all that remains is the component independent of orientation, $C_{2}\|\dot{\gamma}\|$. If $C_{2}$ were negative, it would force regions of the distribution function $\psi(\theta, \phi)$ to be less than zero which is impossible since the probability of finding a fiber in any given region of $(\theta, \phi)$ must be equal to or greater than 0 .

The second-order orientation tensor evolution of Equation (2.35) with the directional diffusion model $D_{r}(\theta, \phi)$ of Equation (6.12) is solved for an initially isotropic fiber distribution for three select coefficient pairs $\left(C_{1}, C_{2}\right)=\left(0,10^{-3}\right),\left(C_{1}, C_{2}\right)=$


Figure 6.15: Transient Solution for $a_{i j}$ from the directionally diffusion model in simple shear flow, $v_{1}=G x_{3}, v_{2}=v_{3}=0$ starting from an isotropic orientation state, $\psi(\theta, \phi)=\frac{1}{4 \pi}$.
$\left(3 \times 10^{-2}, 5.1 \times 10^{-4}\right)$, and $\left(C_{1}, C_{2}\right)=\left(6 \times 10^{-2}, 2.9 \times 10^{-5}\right)$. The results for select second-order orientation tensor components are presented in Figure 6.15. For each coefficient pair, the steady state results are the same as those predicted with the coefficient pair $\left(C_{1}, C_{2}\right)=\left(0,10^{-3}\right)$ which corresponds to the Folgar and Tucker model with $C_{I}=10^{-3}$. The directional diffusion model results diminish the rate of alignment, and delay when steady state is attained.

By increasing $C_{1}$, greater priority is given to local fiber collision effects yielding a steady state that occurs later in time. This characteristic is quantified in Figure 6.16 using Equation (6.40) where fiber alignment is considered to have attained steady state when $a_{11}$ is within $1 \%$ of its steady state value. Note that as the coefficient $C_{1}$ is increased, steady state is attained at a later period in time. The alignment is


Figure 6.16: Percentage difference between steady state orientation and transient orientation state for $a_{11}$ in simple shear flow starting from an isotropic orientation.
within $1 \%$ of steady state at $G t=14.5, G t=15.5, G t=20$, and $G t=39$ for the coefficient pairs $\left(C_{1}, C_{2}\right)=\left(0,10^{-3}\right),\left(C_{1}, C_{2}\right)=\left(1 \times 10^{-2}, 8.4 \times 10^{-4}\right),\left(C_{1}, C_{2}\right)=$ $\left(3 \times 10^{-2}, 5.1 \times 10^{-4}\right)$, and $\left(C_{1}, C_{2}\right)=\left(6 \times 10^{-2}, 2.9 \times 10^{-5}\right)$, respectively. For $C_{1}=$ $6 \times 10^{-2}$, steady state is attained at a time nearly 2.5 times higher than as predicted with $\left(C_{1}, C_{2}\right)=\left(0,10^{-3}\right)$.

The important characteristic to take note of in Figure 6.15 is the time at which the steady state orientation is attained. Directional diffusion causes the fiber distribution to align at a slower rate than experienced without directionality of the diffusion. As in the previous example, this characteristic is quantified by the magnitude of the slope for the $a_{11}$ and $a_{13}$ components of the second-order orientation tensor presented in Figures 6.17 and 6.18 , respectively, from select $\left(C_{1}, C_{2}\right)$ pairs. On close inspection, as $C_{1}$ is increased the rate of change in the $a_{11}$ component decreases throughout nearly
the entire flow history. Similarly, the slope of the $a_{13}$ component also decreases as $C_{1}$ increases throughout much of the flow history. The downward spike in Figure 6.18 for $D a_{13} / D t$ is attributed to the peak in the $a_{13}$ component occurring near $G t=2$ as viewed in Figure 6.15.

The directional diffusion model is designed to slow the rate of alignment as a function of fiber orientation. As in the previous example, the principal eigenvalue of the second-order orientation tensor $a_{(1)}$ is selected as the parameter of alignment. In Figures 6.19 and 6.20 it is evident that the directional diffusion model results diminish the rate of alignment as a function of alignment with the greatest change in alignment rate occurring for relatively high alignment states with $a_{(1)}>0.7$.

It is important to ensure robustness of the transient flow solutions with respect to the initial choice of alignment. As shown in the previous example, when $\left(C_{1}, C_{2}\right)=$ $\left(8,10^{-3}\right)$ in uniaxial elongational flow, $a_{i j}$ may come to rest at two different steady state solutions. To test robustness in simple shear, a highly aligned initial orientation with $a_{11}=0.995$ and $a_{22}=a_{33}=0.0025$ is selected and results from the transient solution for the second-order orientation tensor with $\left(C_{1}, C_{2}\right)=\left(3 \times 10^{-2}, 5.1 \times 10^{-4}\right)$ and $\left(C_{1}, C_{2}\right)=\left(6 \times 10^{-2}, 2.9 \times 10^{-5}\right)$ are presented in Figure 6.21 along with the Folgar and Tucker solution from an initial isotropic distribution. As desired, steady state solutions for the initial high alignment state are identical to those obtained from the initial isotropic orientation state.

The relationship is quite different between $C_{1}$ and $C_{2}$ given in Equations (6.39) and (6.41) for, respectively, uniaxial elongational flow and simple shear flow. A coefficient pair developed for simple shear flow has little effect on the orientation results for uniaxial elongational flow. This effect is viewed in Figure 6.22 where the equation


Figure 6.17: Magnitude of the slope for $a_{11}(t)$ in simple shear flow, starting from an isotropic orientation state.


Figure 6.18: Magnitude of the slope for $a_{13}(t)$ in simple shear flow, starting from an isotropic orientation state.


Figure 6.19: Magnitude of the slope for $a_{11}(t)$ in simple shear flow as a function of alignment parameter $a_{(1)}$.


Figure 6.20: Magnitude of the slope for $a_{13}(t)$ in simple shear flow as a function of alignment parameter $a_{(1)}$.


Figure 6.21: Transient Solution for $a_{i j}$ from the directionally diffusion model in simple shear flow starting from a highly aligned orientation state along the $x_{1}$ axis, compared to the Folgar and Tucker model results with an initial isotropic orientation state.
of motion for $a_{i j}$ is solved with the coefficient pair $\left(C_{1}, C_{2}\right)=\left(6 \times 10^{-2}, 2.9 \times 10^{-5}\right)$. Differences between the solution without any directional diffusion are indistinguishable from the solution obtained through the simple shear coefficient pair $\left(C_{1}, C_{2}\right)=$ $\left(6 \times 10^{-2}, 2.9 \times 10^{-5}\right)$. Conversely, a coefficient pair developed from uniaxial elongational flow will significantly alter steady state results for simple shear flow as viewed in Figure 6.23 for the uniaxial coefficient pair $\left(C_{1}, C_{2}\right)=\left(1,1 \times 10^{-3}\right)$ and $\left(C_{1}, C_{2}\right)=\left(3,1 \times 10^{-3}\right)$. Recall, the coefficient $C_{1}$ for simple shear could only be increased up to 0.06 with no effects on the steady state solution. Since $C_{1}$ is relatively large and $C_{2}$ is greater than zero, the diffusion coefficient pair from uniaxial elongational flow will be so great in simple sharing flow that fibers are not able to remain in a highly aligned orientation state. The converse is true for the simple shear
coefficients in uniaxial elongational flow, since the coefficient $C_{1}=0.06$ is much less than 1 where nominal effects were seen throughout the orientation history.

### 6.2.3 Simple Plaque Flow

The final example is that of simple plaque flow of a Newtonian fluid as shown in the schematic in Figure 6.24, where flow is in the $x_{1}$ direction with shearing in the $x_{3}$ direction. It may be shown for a Newtonian fluid the velocity is simply [112]

$$
\begin{equation*}
v_{1}=-\frac{d P}{d x_{1}} \frac{1}{\mu} h^{2}\left(1-\frac{x_{3}^{2}}{h^{2}}\right) \quad v_{2}=v_{3}=0 \tag{6.42}
\end{equation*}
$$

where $P$ is the pressure, a linear function of $x_{1}$ (see e.g. [112]), $\mu$ is the viscosity, and $h$ is half the gap thickness. The velocity gradients are zero for all components except $\frac{\partial v_{1}}{\partial x_{3}}$ which is simply

$$
\begin{equation*}
\frac{\partial v_{1}}{\partial x_{3}}=2 \frac{d P}{d x_{1}} \frac{1}{\mu} x_{3}=K x_{3} \tag{6.43}
\end{equation*}
$$

This is simple shear flow as discussed in Section 3.1.4.1 where the scaling parameter $G$ is defined for each $x_{3}$ as $G=K x_{3}$. Therefore the previous simple shear flow results (c.f. Section 6.2.2) may be appropriately scaled by setting

$$
\begin{equation*}
G t=\frac{K x_{3} x_{1}}{h^{2}\left(1-\frac{x_{3}^{2}}{h^{2}}\right)} \tag{6.44}
\end{equation*}
$$

The plaque flow results are presented in Figures 6.25(a)-(d) for the $a_{11}$ orientation tensor component through the thickness at $\frac{x_{1}}{h}=1, \frac{x_{1}}{h}=4, \frac{x_{1}}{h}=8$, and $\frac{x_{1}}{h}=20$, respectively. Note that results for $a_{11}$ from the directional diffusion model are always less than results obtained by neglecting directional diffusion. Along the center of the channel, the directional diffusion appears to have little effect, and only along the edges can a noticeable difference be viewed with the directional diffusion results. Along the


Figure 6.22: Transient Solution for $a_{i j}$ from the directionally diffusion model in uniaxial elongation flow with simple shear coefficient pair.


Figure 6.23: Transient Solution for $a_{i j}$ from the directionally diffusion model in simple shear flow with uniaxial elongation coefficient pair.


Figure 6.24: Schematic for simple plaque flow.
wall edge the flow experiences large shear rates and the fibers are rapidly aligned, whereas in the center of the channel the shear rate is small causing the fibers to align at a much slower rate. As discussed in the preceding section, the directional diffusion model in shearing flow has little effect for alignment states near isotropic relative to the Folgar and Tucker model. It is not until the fiber distribution becomes aligned indicated by $a_{11}>0.6$ that differences between the two models are apparent.

### 6.3 Directional Diffusion Model Conclusions

The directional diffusion model combines the effects from local fiber collisions with the volume averaged effects proposed by Folgar and Tucker [11] and is shown to alter the transient orientation solution while not affecting the steady state orientation. The new model is shown to significantly reduce the time when steady state is attained for elongational and shearing flows by a factor of more than two.

There are two limitations to the directional diffusion model. The first is given by the upper bound on the rate of diffusion, therefore if the transient solution must be delayed more than that shown here, the model must undergo additional developments. It would be desired that a phenomenological model would provide the capability to reduce the rate of alignment in shearing flows for orientation states near isotropic. The present model in shearing flows only had a significant effect once the fibers began


Figure 6.25: Plaque flow orientation at select values of $x_{1}$ through the thickness for (a) $a_{11}$ at $\frac{x_{1}}{h}=1$, (b) $a_{11}$ at $\frac{x_{1}}{h}=8$ (c) $a_{22}$ at $\frac{x_{1}}{h}=1$, (d) $a_{22}$ at $\frac{x_{1}}{h}=8$ (e) $a_{33}$ at $\frac{x_{1}}{h}=1$, (f) $a_{33}$ at $\frac{x_{1}}{h}=8$.
to align. Incorporating a model to effect the rate of alignment for isotropic orientation states will allow better adjustments for alignments in the center of the channel for simple plaque flow. The second limitation is the varied choice of parameters for elongational flows and shearing flows. Since discussion in the literature is only for shearing flows, the limitations of the Folgar and Tucker model in elongational flows are uncertain. If there are no limitations in elongational flows, it would appear that the selection of diffusion coefficients from shearing flows may be sufficient since their use has little effect in elongational flows. It is desired that future diffusion models effect alignment rates near isotropic with adjustable parameters that apply to both shearing and elongational flows. It is possible that the incorporation of the relative motion between the center of mass between the two fibers $\boldsymbol{p}$ and $\boldsymbol{\rho}$, which is neglected in the derivation, may provide the key to properly analyze the directional nature of fiber collisions.

## CHAPTER 7

## CONCLUSIONS AND RECOMMENDATIONS

Processing conditions of a short fiber composite have a strong impact on the fiber orientation within the final part, and as such a thorough understanding of the flow kinematics on fiber orientation is essential for accurate design. Representing the fiber orientation through the fiber distribution function is computationally cumbersome and not practical for industrial applications. Therefore fiber orientation tensors are typically employed to provide a concise representation of the fiber alignment state. Currently accepted closures are of the fourth-order orientation tensor and their effects on material property representations are not fully understood. Material characteristics of the processed composite part contribute significantly to the design process, and as a result, expressions for the material stiffness relationship from the fiber orientation distribution will be of considerable use in the design setting. To fully represent fiber orientation throughout the fabrication process an accurate representation of the orientation state during processing is necessary, where current theories for fiber flow kinematics tend to predict aligned orientation states sooner than observed physically.

The significant contributions of this work to assist in solving the preceding issues are briefly summarized in the following list.

- Three sixth-order fitted closures are presented to approximate the sixth-order orientation tensor from lower-ordered orientation tensor components. The sixthorder closures more accurately model the distribution function of fibers than even the exact fourth-order reconstruction and thus provide a degree of accuracy unobtainable from even the most advanced fourth-order closure. The invariant
based sixth-order fitted closure $\mathrm{INV}_{6}$ is defined from a general expression for a fully symmetric sixth-order tensor, where the components of $\mathrm{INV}_{6}$ are fit to a linear polynomial of the fourth-order orientation tensor invariants. The $\mathrm{INV}_{6}$ closure is demonstrated to surpass the accuracy of existing fourth-order closures in the representation of the fiber orientation distribution in all flow simulations investigated, and is nearly as accurate in representing the fiber orientation state as the sixth-order expansion of the distribution function.
- It is demonstrated that existing fourth-order closures are limited, by their construction, to an orthotropic tensor representation and neglect shear-extensional coupling and shear-shear coupling effects. This limitation will prevent shortfiber polymer composite models, polymer crystalline models and other models which employ similar fourth-order closure techniques from representing material behavior more complex than orthotropic. The significance of this limitation may become more pronounced based upon the initial orientation. To avoid the orthotropic limitation it was demonstrated that sixth-order closures, in particular the $\mathrm{INV}_{6}$, can represent shear-extensional and shear-shear coupling behavior.
- Analytical expressions are developed to compute the expectation value and variance of the material stiffness tensor obtained from the fiber orientation probability distribution function. The elastic material constitutive behavior for short fiber composites presented here is based on the Laplace series reconstruction written in terms of complex spherical harmonic functions. As part of this analysis, a method to complete the series expansion of Onat [86,87] is introduced by defining previously undefined higher-order functions in Onat's series expansion using the spherical harmonic functions. By employing the Laplace series
reconstruction, an expression is derived for the expectation value of the stiffness tensor using second- and fourth-order orientation tensors. The relationship between the current approach and that of Advani and Tucker [6] is presented where the current approach only requires the assumption of a single axis of symmetry characterized by a monoclinic material through the symmetry of the distribution function of fibers, $\psi(\boldsymbol{p})=\psi(-\boldsymbol{p})$. An analytic method for computing the variance of the stiffness tensor is presented and is shown to be a function of the orientation tensors up through eighth-order. The analytical method to evaluate both the expectation and the variance for the material stiffness tensor from the fiber orientation tensors is validated through the method of Monte-Carlo. The Monte-Carlo results are shown to agree extremely well with analytic results as the number of sample sets is increased. With the central-limit theorem, normal probability distributions obtained from the analytic expectation and variance of the material stiffness tensor are shown to correspond directly with the results for the normalized frequency of the sample sets of discrete fiber orientation angles.
- The directional diffusion model investigated incorporates contributions from local fiber collisions and volume averaged effects. The diffusion model is demonstrated to significantly delay the rate of alignment for elongational and shearing flows by a factor of more than 2. As observed in center-gated disk flow, the initial out-of-plane stretching effects on the rate of orientation is significantly reduced and alters the orientation path taken toward steady state.

There is still much work to be undertaken in the area of fiber orientation analysis, and several thoughts for the direction of future research are presented below.

- The sixth-order closures although accurate, are cumbersome, and efficient
means for improving the calculation speeds are desired. This is a question of programming prowess, and efficient methods of computation can be of significant use. The sixth-order fitted closure is limited to a small range of fiber interactions, and a more general fitting procedure is of use. It would be worthwhile to develop a sixth-order fitted closure that was not directly dependant on specific flow conditions, but is fit to fiber distributions encompassing the orientation space of the fourth-order orientation tensor. This orientation space will be in five dimensions providing a significant technical hurdle beyond the fourth-order closures which are fit in a two-dimensional space. It has been speculated that the full fiber orientation distribution may be represented by Bingham distributions [113] (see e.g. Chaubal and Leal [49] for the application to the fiber distribution). A similar approach may be quite useful and may save a significant amount of computational expense.
- Although sixth-order closures provide a means to compute shear-extensional and shear-shear material behavior, it is unclear as to whether this feature is of much industrial use. There has not been an exhaustive study to conclusively state whether or not this behavior may be utilized or even measured experimentally.
- With the advent of the directional diffusion model, there is an increased concern with the need for higher-order information. Therefore it is worthwhile to investigate a different basis other than the spherical harmonics. Wavelet basis (see e.g. Frazier [114]) may provide a mathematically rigorous means to capture higher order information without the full lower-order reconstructions.
- It is understood that the assumption in the derivation of the analytical expressions for expectation and variance was for statistically independent fibers under a constant stress field. That assumption loses its validity as the density of fibers increases into the concentrated regime, and it is unclear what additional effects may need to be incorporated for concentrated fiber suspensions. Hine et al. [115] generate statistical distributions of fibers from Bingham distributions and solve, through finite elements, the resultant material stiffness behavior of the composite. It would be worthwhile to compare the analytical results for material property expectation and variance developed in this work to those found through finite elements for fiber distributions determined through the flow evolution equations. Limits could then be placed on the validity of the analytic functions as a function of volume fraction and fiber aspect ratio.
- Future work must investigate fiber interaction models for the fiber kinematics. A complete understanding of the flow on the fiber orientation can provide accurate methods for predicting fiber alignment. Since there are limitations with the Folgar and Tucker model for fiber interaction and the directional diffusion model, an accurate model to represent the fiber orientation kinematics based on a strong physical foundation must be found. There are several aspects of fiber collisions that may be investigated, but the most fundamental is to work and develop a new form for the Jeffery equation [12] that directly incorporates fiber collisions. This work will require a fundamental understanding of the physical phenomena as two fibers pass near each other, and future work should incorporate experimental observations to grasp the full nature of the
fiber orientation during the flow history and move beyond a qualitative representation of fiber orientation kinematics. It is desired that the final model for fiber interactions will have adjustable parameters which can scale both the rate of alignment throughout the transient solution and the steady state orientation while retaining a reasonable physical foundation such as the investigated model.


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## VITA

David Abram Jack was born on September 10, 1977 in Niceville, Florida and was home schooled by his loving parents during most of his preliminary education. While attending the Colorado School of Mines he proposed to, and married, his best friend, the former Trisha Joy Nadeau of Kittredge, Colorado. David graduated first in his class from the Colorado School of Mines with B.S. degrees in Mechanical Engineering and in Engineering Physics. David followed his advisor Douglas E. Smith from Mines to the University of Missouri - Columbia where he has received an M.S. in Mechanical and Aerospace Engineering. He defended his M.S. thesis in Applied Mathematics and his Ph.D. in Mechanical and Aerospace Engineering in the Fall of 2006. David is a prolific writer and is the lead author for four published articles in internationally recognized journals as prestigious as the Journal of Rheology and Composites, Part A and currently has several articles under review. He is the lead author of nine different conference proceedings, and served as co-author for two others. During the evenings and weekends of his graduate education, David served as the Scoutmaster for Boy Scout Troop 52 of Columbia, Missouri, and is currently serving as the Assistant Scoutmaster in the same troop.


[^0]:    ${ }^{1}$ Note that the double contraction given is only one of three possible forms. Each form would lead to its own matrix representation and correspondingly to its own eigenvalue problem (see e.g. Itskov [93]).

