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Spin diffusion in doped semiconductors: The role of Coulomb interactions

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Abstract. – We examine the effect of Coulomb interaction on the mobility and diffusion of spin packets in doped semiconductors. We find that the diffusion constant is *reduced*, relative to its non-interacting value, by the combined effect of Coulomb-enhanced spin susceptibility and spin Coulomb drag. In ferromagnetic semiconductors, the spin diffusion constant vanishes at the ferromagnetic transition temperature.

The ability to control inhomogeneous distributions of electrons and holes in semiconductors is essential to the operation of modern electronic devices. Unlike electron packets in metals, which spread out very quickly under the action of their own electric field, electron-hole packets in semiconductors are charge-neutral objects and can therefore be long-lived. The time evolution of such packets is controlled by a drift-diffusion equation with mobility and diffusion constants μ and D , as was verified in detail in the classic Haynes-Shockley experiment [1].

Recently, a broader category of possible disturbances, involving inhomogeneous *spin* distributions, has come into sharp focus in the context of the emerging field of “spintronics” [2]. Consider, for example, a spin packet consisting of excess up-spin electrons compensated by a deficiency of down-spin electrons. Such a disturbance can occur in the conduction band of a metal or of a doped semiconductor [3]. Like an ordinary electron-hole packet this is a charge-neutral object and can therefore be extremely long-lived (recent experiments measure a spin relaxation time τ_s of the order of 10 ns [3, 4]). Unlike an electron-hole packet, however, the disturbance involves carriers of a single polarity —electrons— and therefore evolves with the mobility and diffusion constants of the conduction band, which are usually larger than those of the valence band. Indeed, large values of the spin drift mobility $\mu_s \sim 3 \times 10^3$ cm²/V s and spin diffusion constant $D_s \sim 10^3$ cm²/s have recently been observed in experiments on *n*-doped GaAs [4].

The qualitative difference between unipolar and bipolar disturbances in semiconductors has recently been emphasized by Flatté and Byers [5] within the frame of a simple model in which the electron-electron interaction is treated in the Hartree approximation. In this letter, we refine their analysis by examining the more subtle effects of exchange and Coulomb correlation on the mobility and diffusion constant of a spin packet. It will be shown that these many-body effects have a serious impact on the value of the spin diffusion constant and can be quantitatively probed in a Haynes-Shockley-type experiment that measures independently the mobility and the diffusion constant of a spin packet. The two key physical effects are i) the reduction of the spin stiffness (inverse of the spin susceptibility) due to (mainly) exchange interactions and ii) the spin Coulomb drag [6] working like friction against the relative motion of up-spin and down-spin electrons. Both effects tend to reduce the diffusion constant. By contrast, the spin-packet mobility turns out to be essentially unaffected by interactions.

Under certain conditions, the electron (hole) gas in doped semiconductors may undergo a ferromagnetic transition. The phenomenon occurs either at very low densities, due to the Coulomb interaction [7] or, for instance, in GaAs [8,9] under heavy doping with magnetic Mn impurities. As the ferromagnetic transition temperature T_c is approached the longitudinal spin stiffness vanishes and so does the spin diffusion constant, which thus exhibits a critical behavior.

We begin our analysis by assuming, as usual, a linear relationship between the number current densities $\vec{J}_\alpha(\vec{r})$ ($\alpha = \uparrow$ or \downarrow) and the gradient of the local electro-chemical potentials $\psi_\alpha(\vec{r}) = \phi(\vec{r}) - (1/e)\partial f(n_\uparrow, n_\downarrow, T)/\partial n_\alpha$, where $\phi(\vec{r})$ is the electrostatic potential, e is the absolute value of the electron charge, and $f(n_\uparrow, n_\downarrow, T)$ is the free energy per unit volume of a homogeneous interacting electron gas at the local spin densities $n_\alpha(\vec{r})$ and temperature T [10]. This leads to the equation

$$e\vec{J}_\alpha(\vec{r}) = \sum_\beta \left(\sigma_{\alpha\beta}(\vec{r})\vec{\nabla}\phi(\vec{r}) - eD_{\alpha\beta}(\vec{r})\vec{\nabla}n_\beta(\vec{r}) \right), \quad (1)$$

where $\sigma_{\alpha\beta}$ is the homogeneous conductivity matrix, calculable from the Kubo formula, and the diffusion matrix $D_{\alpha\beta}$ is given by the generalized Einstein relation

$$e^2D_{\alpha\beta} = \sum_\gamma \sigma_{\alpha\gamma}S_{\gamma\beta}, \quad (2)$$

where

$$S_{\alpha\beta} = \frac{\partial^2 f(n_\uparrow, n_\downarrow, T)}{\partial n_\alpha \partial n_\beta} \quad (3)$$

is the static spin-stiffness matrix —minus the inverse of the spin susceptibility matrix.

On a formal level the main effect of the Coulomb interaction is the appearance of non-vanishing off-diagonal elements of the conductivity and spin-stiffness matrices. These off-diagonal matrix elements have a simple physical interpretation. $\sigma_{\uparrow\downarrow} \neq 0$ implies that an electric field acting only on the up-spin electrons must necessarily drag along a current of down-spin electrons. Similarly, $S_{\uparrow\downarrow} \neq 0$ means that the chemical potential of up-spins $\partial f(n_\uparrow, n_\downarrow, T)/\partial n_\uparrow$ is a function of *both* up and down spin densities. In addition the Coulomb interaction significantly modifies the values of the diagonal elements of these matrices, as we shall see momentarily.

Let us apply eq. (1) to the calculation of the time evolution of a spin packet obtained by injecting an *excess* spin density $\Delta m(\vec{r}, 0) = \Delta M\delta(\vec{r})$ near the origin at time $t = 0$. We denote by $m(\vec{r}, t) = n_\uparrow(\vec{r}, t) - n_\downarrow(\vec{r}, t)$ the net spin density at point \vec{r} and time t , by $m^{(0)} =$

$n_{\uparrow}^{(0)} - n_{\downarrow}^{(0)}$ the uniform value of the spin density at thermodynamic equilibrium, and by $\Delta m(\vec{r}, t) \equiv m(\vec{r}, t) - m^{(0)}$ the excess spin density following spin injection. Similarly, we denote by $n(\vec{r}, t) = n_{\uparrow}(\vec{r}, t) + n_{\downarrow}(\vec{r}, t)$ the total electron density and by $n^{(0)}$ and by $\Delta n(\vec{r}, t)$ the equilibrium and the excess density, respectively.

The solution of the problem requires as additional inputs the continuity equations for the number and spin densities:

$$\frac{\partial \Delta n(\vec{r}, t)}{\partial t} = -\vec{\nabla} \cdot \vec{J}(\vec{r}, t), \quad (4)$$

$$\frac{\partial \Delta m(\vec{r}, t)}{\partial t} = -\frac{\Delta m(\vec{r}, t)}{\tau_s} - \vec{\nabla} \cdot \vec{J}_m(\vec{r}, t), \quad (5)$$

where $\vec{J}_m = \vec{J}_{\uparrow} - \vec{J}_{\downarrow}$ is the spin current density and τ_s is the spin relaxation time, which is very long [3, 4].

In practice, following the procedure familiar in the theory of bipolar carrier packets [11], we first combine eqs. (4), (5) and (1) to eliminate the $\vec{\nabla} \cdot \vec{E}$ term related to the Poisson equation, and *then* impose the local charge neutrality constraint [12]

$$\Delta n_{\uparrow}(\vec{r}, t) = -\Delta n_{\downarrow}(\vec{r}, t). \quad (6)$$

This yields the result

$$\begin{aligned} \frac{\partial \Delta m(\vec{r}, t)}{\partial t} = & -\frac{\Delta m(\vec{r}, t)}{\tau_s} + D_s \nabla^2 \Delta m(\vec{r}, t) + \\ & + \mu_s \vec{E} \cdot \vec{\nabla} \Delta m(\vec{r}, t), \end{aligned} \quad (7)$$

where

$$\mu_s = \frac{(n_{\uparrow} + n_{\downarrow}) \tilde{\sigma}_{\uparrow} \tilde{\sigma}_{\downarrow}}{n_{\uparrow} n_{\downarrow} (\tilde{\sigma}_{\uparrow} + \tilde{\sigma}_{\downarrow})} \quad (8)$$

and

$$D_s = \frac{\tilde{\sigma}_{\uparrow} \tilde{D}_{\downarrow} + \tilde{\sigma}_{\downarrow} \tilde{D}_{\uparrow}}{\tilde{\sigma}_{\uparrow} + \tilde{\sigma}_{\downarrow}} \quad (9)$$

are the effective mobility and diffusion constants [13], and \vec{E} is an externally applied electric field. Equations (8) and (9) reduce to the expressions presented in [5] in the non-interacting case. The constants $\tilde{\sigma}_{\alpha}$ and \tilde{D}_{α} are presently given by

$$\begin{aligned} \tilde{\sigma}_{\alpha} &= \sigma_{\alpha\alpha} + \sigma_{\alpha\bar{\alpha}}, \\ \tilde{D}_{\alpha} &= D_{\alpha\alpha} - D_{\alpha\bar{\alpha}}. \end{aligned} \quad (10)$$

The fact that the conductivities enter a spin symmetric combination while the diffusion constants are in a spin antisymmetric combination reflects the fact that the electrostatic field has the same sign for both spin components, while the density gradients have opposite signs (see (6)).

The solution of eq. (7) in a homogeneous and isotropic liquid is

$$\Delta m(\vec{r}, t) = \frac{\Delta M e^{-t/\tau_s}}{(4\pi D_s t)^{3/2}} e^{-\frac{|\vec{r} + \mu_s \vec{E} t|^2}{4D_s t}}. \quad (11)$$

Thus, we see that a Haynes-Shockley-type experiment can in principle determine μ_s and D_s independently, provided that τ_s is sufficiently long.

Let us now proceed to the calculation of μ_s and D_s . The necessary inputs are the conductivity and the spin stiffness matrices. The conductivity matrix is best computed from its inverse, namely the resistivity matrix $\rho_{\alpha\beta}$ whose general structure is determined by the principle of Galilean invariance and Newton's third law. The explicit form of $\rho_{\alpha\beta}$, extracted from eq. (3) of ref. [6], is

$$\rho = \begin{pmatrix} \frac{m^*}{n_{\uparrow} e^2 \tau_{\uparrow}} - \frac{n_{\downarrow}}{n_{\uparrow}} \rho_{\uparrow\downarrow} & \rho_{\uparrow\downarrow} \\ \rho_{\uparrow\downarrow} & \frac{m^*}{n_{\downarrow} e^2 \tau_{\downarrow}} - \frac{n_{\uparrow}}{n_{\downarrow}} \rho_{\uparrow\downarrow} \end{pmatrix}. \quad (12)$$

Here τ_{α} are the combined electron-impurity and electron-phonon scattering times, usually of the order of 10^{-3} - 10^{-4} ns, m^* is the effective mass of the carriers, and $\rho_{\uparrow\downarrow}$ is the spin drag transresistivity calculated in ref. [6]. After computing $\sigma_{\alpha\beta} = [\rho^{-1}]_{\alpha\beta}$ the diffusion matrix is straightforwardly obtained from eq. (2), and μ_s , D_s are calculated from eqs. (8) and (9). The algebra is greatly simplified by the reasonable assumption that the scattering times for the two spin components are not too different, *i.e.*, $\tau_{\uparrow} = \tau_{\downarrow} = \tau_D$ [14]. Under this condition, the result is

$$\mu_s = \frac{e\tau_D}{m^*} \quad (13)$$

and

$$D_s = \frac{\mu_s k_B T}{e} \frac{S}{S_c} \frac{1}{1 - \rho_{\uparrow\downarrow}/\rho_D}, \quad (14)$$

where $S = \partial^2 f(n, m, T)/\partial m^2$ is the spin stiffness, $S_c = k_B T n / 4n_{\uparrow} n_{\downarrow}$ is the Curie spin stiffness of an ideal classical gas, and $\rho_D = m^* / n e^2 \tau_D$ is the Drude resistivity.

Equation (13) tells us that the mobility of the packet is not *explicitly* modified by the Coulomb interaction and in fact coincides with the ordinary homogeneous mobility. Strictly speaking, this result is only valid under the assumption that up-spin and down-spin electrons have equal mobilities and thus drift at the same speed in an applied electric field. Coulomb interactions, being Galilean-invariant, cannot change the total momentum of such a uniformly drifting electron gas.

The situation is completely different for the diffusion constant. As the spin packet spreads out, the up- and down-spin currents are directed in opposite directions and friction arises: for this reason the expression for D_s contains the spin-drag resistivity as a factor that reduces the numerical value of D_s . In addition, the Coulomb interaction together with the Pauli exclusion principle reduces the energy cost of spin-density fluctuations (*i.e.*, the spin stiffness) further decreasing the rate of diffusion of a spin packet.

Figure 1 presents the necessary ingredients to calculate D_s . Figure 1a shows that $\rho_{\uparrow\downarrow}$ — a negative number — vanishes at low temperature as $(T/T_F)^2$ peaking at a temperature of the order of the Fermi temperature T_F . As the inset illustrates, the prefactor $1/(1 - \rho_{\uparrow\downarrow}/\rho_D)$ (see eq. (14)) displays a marked dependence on the sample mobility, increasing with the latter. Figure 1b shows S rescaled by its non-interacting value S_{ni} and its behavior at the onset of the ferromagnetic instability. We evaluated S numerically starting from the work of Tanaka and Ichimaru [15] where the free energy density is calculated as a function of temperature, density, and spin polarization, by means of a self-consistent integral equation approach that satisfies the thermodynamic sum rules.

In fig. 2 we plot D_s/D_c , where $D_c = \mu_s k_B T / e$ is the classical non-interacting diffusion constant, for n -doped GaAs in a range of densities that are relevant to the experiments of ref. [4]. The solid line corresponds to our full-interacting calculation, while the dashed line to the non-interacting case. We see that the interaction correction is quite significant, and reduces the value of D_s as expected. Despite this reduction, D_s is still considerably larger

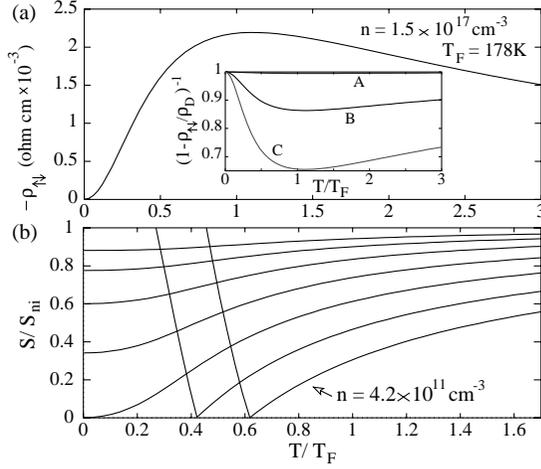


Fig. 1 – (a) Transresistivity $\rho_{\uparrow\downarrow}$ vs. the reduced temperature T/T_F for typical semiconductor parameters. The inset shows the behavior of the factor $1/(1 - \rho_{\uparrow\downarrow}/\rho_D)$ for three different mobilities: $\mu = 10^2 \text{ cm}^2/\text{Vs}$ (A), $\mu = 3 \times 10^3 \text{ cm}^2/\text{Vs}$ (B), $\mu = 10^4 \text{ cm}^2/\text{Vs}$ (C). (b) Spin stiffness S vs. T/T_F . The density is $n = 4.2 \times 10^{11} \text{ cm}^{-3}$ for the lower curve and increases by a factor 10 for each line starting from the bottom. The cusps represent the onset of ferromagnetism.

than D_c , consistently with experimental observations. These results show that the effect of the reduced spin stiffness dominates at low and intermediate temperature, while the spin drag contribution dominates at high temperature (see inset of fig. 2).

In the non-degenerate limit $T \gg T_F(n)$ we find that D_s/D_c approaches 1. In the non-interacting theory [5] this limit is approached from above because the leading correction to the spin stiffness coming from the quantum kinetic energy is positive: $S_{ni} = S_c [1 + \lambda_T^3 n / 2\sqrt{2}]$, where λ_T is the de Broglie thermal wavelength at temperature T . In the interacting theory

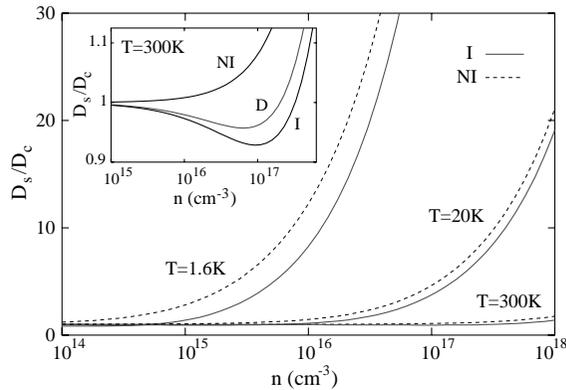


Fig. 2 – The interacting diffusion constant of a spin-packet (I) and its non-interacting approximation (NI) (rescaled by D_c) vs. density for different temperatures. The inset shows the comparison with the value obtained considering interactions only through the spin Coulomb drag effect (D). In all the calculations the dielectric constant of the semiconductor is $\epsilon = 12$ and the mobility is $\mu = 3 \times 10^3 \text{ cm}^2/\text{Vs}$.

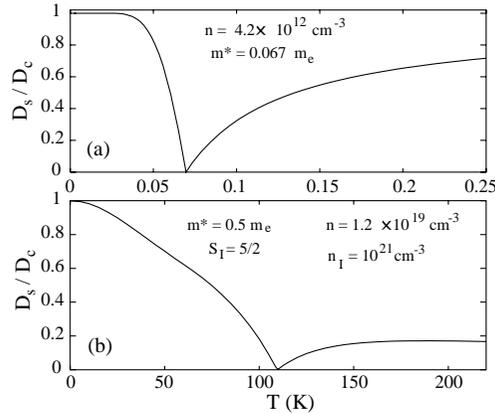


Fig. 3 – (a) The diffusion constant of a spin packet *vs.* temperature in a low-density electron gas with no magnetic impurities. (b) Same as (a) for an electron gas of typical density $n = 1.2 \times 10^{19} \text{ cm}^{-3}$ in a semiconductor doped with magnetic impurities (n_I and S_I are the impurity concentration and spin, respectively).

instead, the leading correction to D_s is due to the spin Coulomb drag term and it is negative. In fact, for $T \gg T_F(n)$, $\rho_{\uparrow\downarrow}/\rho_D \sim [n/(k_B T)^{3/2}] \ln(n/(k_B T)^2)$, and the logarithmic term dominates over corrections entering the spin stiffness in both the $n \rightarrow 0$ and $T \rightarrow \infty$ limits. Due to interactions, $D_s/D_c \rightarrow 1$ from *below* always.

A very interesting feature of eq. (14) is the possibility of large variations in D_s when the electron gas undergoes a ferromagnetic transition. From the curves in fig. 1b and fig. 3 we see that S (interpreted as *longitudinal* spin stiffness in the ferromagnetic phase) and D_s vanish at the transition temperature T_c . For $T < T_c$, D_s/D_c increases at first, due to the sharp increase in spin stiffness (see fig. 1), but then begins to saturate and tends to 1 as full polarization sets in [16].

In an ordinary electron liquid, the ferromagnetic transition is predicted to occur only at extremely low densities [7]. There is, however, an interesting variant: semiconductors doped with magnetic impurities (Mn) can undergo a ferromagnetic transition at rather high carrier densities [9, 17], $n \sim 10^{20} \text{ cm}^{-3}$ for (Ga, Mn)As, and temperatures as large as 110 K [9]. Our theory can be extended to such materials, but, for the sake of clarity, we shall present this extension in a longer paper. Here we simply remark that in a simple mean field theory, such as that of ref. [18], the calculated value of D_s/D_c for realistic values of the parameters has the form shown in fig. 3b, where, as in the intrinsic ferromagnetic case, D_s vanishes at T_c [19].

In conclusion, we have demonstrated that a Haynes-Shockley experiment measuring D_s and μ_s for a unipolar spin packet would be a sensitive probe of many-body effects such as the spin-Coulomb drag and the Coulomb enhancement of the spin susceptibility, and would provide a strong signature of a ferromagnetic ordering transition. Conversely, many-body effects must be taken into account in a quantitative theory of spin diffusion.

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