

# Non- $V$ -representability of currents in time-dependent many-particle systems

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We argue that an arbitrarily chosen time-dependent current density is generically non- $V$ -representable in a many-particle system; i.e., it cannot be obtained by applying only a time-dependent scalar potential to the system. Furthermore, we show by a concrete example that even a current that is  $V$ -representable in an interacting many-particle system may (and in general will) turn out to be non- $V$ -representable when the interaction between the particles is turned off.

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## I. INTRODUCTION

Since the beginning of density functional theory (DFT),<sup>1-3</sup> the problem of answering the following questions has been recognized to be of fundamental importance:

(1) Given a (positive) particle density  $n(\vec{r})$  in an  $N$ -particle system, is there a local potential  $V(\vec{r})$  that produces this density in the ground state of the system?

(2) If a certain particle density  $n(\vec{r})$  arises in the ground state of an  $N$ -particle system subjected to a local potential  $V(\vec{r})$ , is there a local potential  $V_s(\vec{r})$  that produces the same density in the ground state of the same system with the particle-particle interactions turned off?

The first question is known as the *V-representability question*, and a given density is said to be *V-representable* if the answer is affirmative. The original formulation of DFT by Hohenberg and Kohn<sup>1</sup> made heavy use of the assumption that “all reasonable densities are  $V$ -representable.” Subsequent work<sup>4</sup> has shown that this assumption is necessary only when one tries to prove the existence of the functional derivative of the energy functional. The second question lies at the very heart of the Kohn-Sham formalism.<sup>2</sup> Recall that within this formalism, one tries to obtain the ground-state density of an interacting many-particle system by applying a local potential to a noninteracting version of the same system. Obviously, it is vital to the success of this theory that the target density, which is by assumption  $V$ -representable, be also noninteracting  $V$ -representable; i.e., the answer to question (2) must be affirmative.

Even though mathematically rigorous answers to the two  $V$ -representability questions are not known, DFT has been widely applied to the calculation of the electronic structure of matter. In these calculations it is tacitly assumed that the set of  $V$ -representable densities in both interacting and noninteracting systems is dense enough to approximate to an arbitrary level of accuracy any physical ground-state density (these beliefs are supported by mathematical work on lattice system<sup>5</sup>). These assumptions have been automatically transplanted to the relatively younger field of time-dependent density functional theory (TDDFT),<sup>6-8</sup> wherein the questions are whether a given *time-dependent* particle density  $n(\vec{r}, t)$  evolving from a given initial state  $\Psi$  can be produced by a local time-dependent scalar potential  $V(\vec{r}, t)$  and, in the affirmative case, whether the same density can also be produced

by a time-dependent scalar potential  $V_s(\vec{r}, t)$  starting from an initial state  $\Psi_s$ , now with interactions turned off.

In this paper we are not going to challenge the wisdom of the standard  $V$ -representability assumptions in DFT or TD-DFT, but rather will examine whether such assumptions can be plausibly extended to the *particle current density*  $\vec{j}(\vec{r}, t)$ . There are good reasons to undertake this study. During the past ten years we have seen many indications that the time-dependent current density,<sup>9-13</sup> together with the initial state of the system (and hence the initial density) may provide a more fundamental description of the dynamics. Indeed, the proof that the time-dependent current density determines the external scalar potential is the very first step in the proof of the Runge-Gross (RG) theorem—the foundation theorem of TDDFT.<sup>6</sup> However, the RG theorem does not say anything about the  $V$ -representability question for the current; i.e., whether a given time-dependent current can be produced by a local time-dependent scalar potential. Reasoning by analogy with the particle density has led (or, as we are going to show, misled) some workers to believe that the  $V$ -representability assumption for the current density is about as plausible as the corresponding assumption for the density, and that therefore any physical current density can be approximated to an arbitrary degree of precision by the current density generated by a suitably chosen *scalar* potential, in an interacting as well as in a noninteracting system.<sup>8</sup>

The purpose of this paper is to show that this is not the case. Due to the vector character of the current density it is usually impossible for an arbitrary current to be generated by a single scalar function: the potential. Even in those special (but physically very relevant) cases in which this can be done, the current will not be simultaneously representable in the noninteracting system.<sup>17</sup> A more general theory that makes use of an effective *vector* potential to generate the current is therefore needed: such a theory exists and it is known as time-dependent current density functional theory (TDCDFT).<sup>9,13</sup>

## II. NON- $V$ -REPRESENTABILITY OF GENERIC CURRENT DENSITIES

In this section we present our main argument against  $V$ -representability of the current density. Recall that the vec-

tor field  $\vec{j}(\vec{r}, t)$ , like any other vector field, can be written as the sum of a longitudinal component  $\vec{j}_L(\vec{r}, t)$ , a transversal component  $\vec{j}_T(\vec{r}, t)$ , and a constant that can be assumed to vanish if the full current has to vanish at infinity:

$$\vec{j}(\vec{r}, t) = \vec{j}_L(\vec{r}, t) + \vec{j}_T(\vec{r}, t). \quad (1)$$

The longitudinal current density  $\vec{j}_L(\vec{r}, t)$  is curl-free and can therefore be represented as the gradient of a scalar field, while the transversal current  $\vec{j}_T(\vec{r}, t)$  is divergence-free and can therefore be represented as the curl of a vector field. The spatial Fourier components of  $\vec{j}_L$  and  $\vec{j}_T$ , denoted by  $\vec{j}_L(\vec{q}, t)$  and  $\vec{j}_T(\vec{q}, t)$ , respectively, are obtained by projecting the Fourier component of  $\vec{j}(\vec{q}, t)$  along directions parallel and perpendicular to the unit vector  $\hat{q}$ , as

$$\vec{j}_L(\vec{q}, t) = [\vec{j}(\vec{q}, t) \cdot \hat{q}] \hat{q}, \quad (2)$$

and

$$\vec{j}_T(\vec{q}, t) = \vec{j}(\vec{q}, t) - \vec{j}_L(\vec{q}, t). \quad (3)$$

Notice that the particle density is completely determined by the longitudinal current density since, according to the continuity equation, one has

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

with initial condition  $n(\vec{r}, t) = n_0(\vec{r})$ .

We begin our argument by assuming that a certain current density  $\vec{j}(\vec{r}, t)$  is  $V$ -representable, and let  $\vec{j}_L(\vec{r}, t)$  and  $\vec{j}_T(\vec{r}, t)$  denote its longitudinal and transverse components, respectively. According to the RG theorem, the potential  $V(\vec{r}, t)$  that produces  $\vec{j}(\vec{r}, t)$  is unique up to an arbitrary function of time. Consider now a second current density  $\vec{j}'(\vec{r}, t) = \vec{j}_L(\vec{r}, t) + \vec{j}'_T(\vec{r}, t)$ ; i.e.,  $\vec{j}'(\vec{r}, t)$  differs from  $\vec{j}(\vec{r}, t)$  only because its transverse component,  $\vec{j}'_T(\vec{r}, t)$  differs from  $\vec{j}_T(\vec{r}, t)$ . We claim that  $\vec{j}'(\vec{r}, t)$  is not  $V$ -representable. Indeed, if it were  $V$ -representable, then there would be a potential  $V'(\vec{r}, t) \neq V(\vec{r}, t)$  that generates it (here and in the following, the  $\neq$  sign means that two potentials differ by more than a mere function of time). However, this is impossible, since, according to Eq. (4), these two different potentials would give the same particle density, in contradiction with the Runge-Gross theorem.<sup>6</sup> Thus, for a given longitudinal current density  $\vec{j}_L(\vec{r}, t)$ , there is at most one transverse current density  $\vec{j}_T(\vec{r}, t)$  such that the full current density  $\vec{j}_L(\vec{r}, t) + \vec{j}_T(\vec{r}, t)$  is  $V$ -representable.

The ease with which, given a  $V$ -representable current density, we were able to construct infinitely many non- $V$ -representable ones is a strong indication that  $V$ -representable currents are a rather exceptional occurrence in the space of all possible currents. To strengthen the argument let us make the plausible assumption that the mapping from potentials to  $V$ -representable currents, via the solution of the time-dependent Schrödinger equation, is not only invertible (RG theorem), but also continuous. This implies

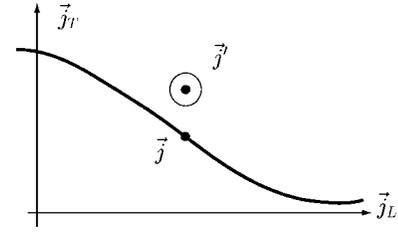


FIG. 1. A simple cartoon of the sparsity of the  $V$ -representable current densities. The  $V$ -representable current density  $\vec{j}$  lies on a continuous hypersurface (here schematized as a continuous curve) in current density space. Due to the continuity of the mapping between  $\vec{j}_L$  and  $\vec{j}_T$  a sufficiently small neighborhood of the non- $V$ -representable current density  $\vec{j}'$  contains only non- $V$ -representable current densities.

that, within the subset of  $V$ -representable current densities,  $\vec{j}_T(\vec{r}, t)$  is a continuous functional of  $\vec{j}_L(\vec{r}, t)$ . Let  $\vec{j}(\vec{r}, t) = \vec{j}_L(\vec{r}, t) + \vec{j}_T(\vec{r}, t)$  be a  $V$ -representable current density. Consider then a small “neighborhood” of the non- $V$ -representable current density  $\vec{j}'(\vec{r}, t) = \vec{j}_L(\vec{r}, t) + \vec{j}'_T(\vec{r}, t)$  and let  $\vec{j}_1(\vec{r}, t)$  be a (hypothetical)  $V$ -representable current density in this neighborhood. Since, by choice, the longitudinal component of  $\vec{j}_1, \vec{j}_{1L}$ , is close to  $\vec{j}_L$ , the continuity of the mapping from  $\vec{j}_L$  to  $\vec{j}_T$  for  $V$ -representable currents implies that the transverse component of  $\vec{j}_1, \vec{j}_{1T}$ , is close to  $\vec{j}_T$ . However, this cannot be true for a sufficiently small neighborhood of  $\vec{j}'(\vec{r}, t)$  if the difference between  $\vec{j}_T$  and  $\vec{j}'_T$  is finite (see Fig. 1). We conclude that every non- $V$ -representable current density is surrounded by a neighborhood that contains only non- $V$ -representable current densities: the set of  $V$ -representable current densities is not “dense” in the space of all possible current densities.

### III. INTERACTING VERSUS NONINTERACTING $V$ -REPRESENTABILITY

Undeterred by the above arguments one might insist that, after all, the task of the Kohn-Sham theory is to approximate  $V$ -representable current densities by noninteracting  $V$ -representable ones. We know that the set of  $V$ -representable current densities is characterized by a certain  $\vec{j}_T$  versus  $\vec{j}_L$  relation: it is the presence of this constraint that makes the set so “sparse.” Similarly, in the noninteracting system, the set of the  $V$ -representable current densities is characterized by another  $\vec{j}_T$  versus  $\vec{j}_L$  relation. It would be nice if these two relations happened to be the same relation, so that a Kohn-Sham potential yielding the correct density would also automatically yield the correct current density.

This conjecture has found its way in the literature,<sup>8</sup> so that it is important to examine it carefully. In this section we construct an example of a  $V$ -representable current density, which is definitely non- $V$ -representable in the noninteracting (Kohn-Sham) system. Thus, in this example, the  $\vec{j}_T$  versus  $\vec{j}_L$  relation of the interacting system turns out to be incompatible with the  $\vec{j}_T$  versus  $\vec{j}_L$  relation of the corresponding non-

interacting one. We will argue (without proof) that this state of affairs is quite generic for currents generated by scalar potentials in anisotropic interacting systems.

The system we consider is a two-dimensional interacting electron liquid that is initially in the ground state with inhomogeneous density

$$n_0(\vec{r}) = \bar{n}(1 + 2\gamma \cos \vec{G} \cdot \vec{r}), \quad (5)$$

where  $\vec{G}$  is a two-dimensional vector parallel to the  $x$  axis,  $\bar{n}$  is the average density, and  $\bar{n}\gamma$  the amplitude of the density modulation, with  $\gamma \ll 1$ . This density is produced by the application of the static potential

$$V_0(\vec{r}) = \frac{2\bar{n}\gamma}{\chi^h(\vec{G})} \cos \vec{G} \cdot \vec{r} \quad (6)$$

to an initially homogeneous electron liquid of density  $\bar{n}$ .<sup>18</sup> Here  $\chi^h(\vec{G})$  is the static density susceptibility of the homogeneous electron gas of density  $\bar{n}$  at wave vector  $\vec{G}$ .<sup>19</sup> We now apply to this system a time-dependent, periodic scalar potential of the form

$$V(\vec{r}, t) = V e^{i(\vec{q} \cdot \vec{r} - \omega t)} + \text{c.c.}, \quad (7)$$

where  $\vec{q}$  is a two-dimensional wave vector, which we assume to be much smaller in magnitude than both  $G$  and  $k_F$  ( $k_F$  being the Fermi wave vector corresponding to the average density  $\bar{n}$ ). The frequency  $\omega$  is assumed to be larger than both  $v_F q$  and  $v_F G$ , where  $v_F$  is the Fermi velocity associated with  $k_F$ .

It is well known that the time-dependent potential of Eq. (7) can be recast as a longitudinal vector potential

$$\vec{A}(\vec{r}, t) = \frac{\vec{q}V}{\omega} e^{i(\vec{q} \cdot \vec{r} - \omega t)} + \text{c.c.}, \quad (8)$$

so that we can say that the induced current density is

$$j_\alpha(\vec{r}, t) = \sum_{l, \beta} \chi_{\alpha\beta}(\vec{q} + l\vec{G}, \vec{q}, \omega) \frac{q_\beta V}{\omega} e^{i[(\vec{q} + l\vec{G}) \cdot \vec{r} - \omega t]} + \text{c.c.}, \quad (9)$$

where  $\chi_{\alpha\beta}(\vec{q} + l\vec{G}, \vec{q}, \omega)$  is the dynamical linear response function of the inhomogeneous liquid, which connects the cartesian  $\beta$  component of the vector potential amplitude at wave vector  $\vec{q}$  to the Cartesian  $\alpha$  component of the current density amplitude at wave vector  $\vec{q} + l\vec{G}$ , where  $l$  is an integer. It should be noted that this response function coincides with the homogeneous response function  $\chi_{\alpha\beta}^h(\vec{q}, \omega)$  up to corrections of order  $\gamma^2$  when  $l=0$ , and is of first order in  $\gamma$  when  $l = \pm 1$ . The components with  $|l| \geq 2$  are of order  $\gamma^2$  at least.

We now want to show that the exact current of Eq. (9) cannot be obtained in a non-interacting system subjected only to scalar potentials that yield the exact density. We first notice that the ground-state density  $n_0(\vec{r})$  is enforced in a noninteracting electron gas by the scalar potential

$$V_{0,s}(\vec{r}) = \frac{2\bar{n}\gamma}{\chi_0^h(\vec{G})} \cos \vec{G} \cdot \vec{r}, \quad (10)$$

where  $\chi_0^h(\vec{G})$  is the static density susceptibility of the noninteracting electron gas of density  $\bar{n}$  at wave vector  $\vec{G}$ . We know from the invertibility of the mapping between vector potentials and currents that the exact current density of Eq. (9) can be generated in the noninteracting electron gas by one and only one time-dependent vector potential,

$$A_{s,\alpha}(\vec{r}, t) = A_\alpha(\vec{r}, t) + \sum_{l, l', \beta, \delta} f_{Hxc,\alpha\beta}(\vec{q} + l\vec{G}, \vec{q} + l'\vec{G}, \omega) \times \chi_{\beta\delta}(\vec{q} + l'\vec{G}, \omega) \frac{q_\delta V}{\omega} e^{i[(\vec{q} + l\vec{G}) \cdot \vec{r} - \omega t]} + \text{c.c.}, \quad (11)$$

where  $f_{Hxc,\alpha\beta}(\vec{q} + l\vec{G}, \vec{q} + l'\vec{G}, \omega)$  is the sum of the so-called exchange-correlation kernel  $f_{xc,\alpha\beta}(\vec{q} + l\vec{G}, \vec{q} + l'\vec{G}, \omega)$  and the Hartree kernel  $f_{H,\alpha\beta}(\vec{q} + l\vec{G}, \vec{q} + l'\vec{G}, \omega) \equiv (2\pi e^2 / |\vec{q} + l\vec{G}|) [(q_\alpha + lG_\alpha)(q_\beta + lG_\beta) / \omega^2] \delta_{ll'}$ , for our system. Thus, in order to prove that the current of Eq. (9) cannot be generated in the noninteracting system by a scalar potential, we only need to show that the vector potential  $\vec{A}_s(\vec{r}, t)$  has a finite transversal component. If this is the case, then the uniqueness of  $A_s$  guarantees that the current density cannot be produced by a purely longitudinal vector potential, and hence not by a simple scalar potential.

In order to establish the existence of a transversal component of  $\vec{A}_s$  we focus on the Fourier component at wave vector  $\vec{q}$  and we discard both  $\vec{A}$  and the contribution of the Hartree kernel because they are purely longitudinal fields. The quantity of interest is thus the exchange-correlation vector potential

$$A_{xc,\alpha}(\vec{q}, \omega) = \sum_{l, \beta, \delta} f_{xc,\alpha\beta}(\vec{q}, \vec{q} + l\vec{G}, \omega) \chi_{\beta\delta}(\vec{q} + l\vec{G}, \vec{q}, \omega) \frac{q_\delta V}{\omega}. \quad (12)$$

The zeroth order in  $\gamma$  of this expression is purely longitudinal and can therefore be discarded. The next nonvanishing contribution is of order  $\gamma^2$  and is given by

$$A_{xc,\alpha}(\vec{q}, \omega) \approx \sum_{\beta, \delta} \{ f_{xc,\alpha\beta}^h(\vec{q}, \omega) \chi_{\beta\delta}^{(2)}(\vec{q}, \vec{q}, \omega) + f_{xc,\alpha\beta}^{(2)}(\vec{q}, \vec{q}, \omega) \times \chi_{\beta\delta}^h(\vec{q}, \omega) + f_{xc,\alpha\beta}^{(1)}(\vec{q}, \vec{q} + \vec{G}, \omega) \chi_{\beta\delta}^{(1)}(\vec{q} + \vec{G}, \vec{q}, \omega) + f_{xc,\alpha\beta}^{(1)}(\vec{q}, \vec{q} - \vec{G}, \omega) \chi_{\beta\delta}^{(1)}(\vec{q} - \vec{G}, \vec{q}, \omega) \} \frac{q_\delta V}{\omega}, \quad (13)$$

where the quantities with superscripts (1) and (2) refer to the inhomogeneous system of density  $n_0(\vec{r})$  and are first order and second order in  $\gamma$ , respectively.

To proceed, we now make use of certain exact identities, which can be obtained starting from identities that were derived in Ref. 14 from a careful consideration of the behavior of the current response function and its associated  $xc$  kernel under transformation to an accelerated reference frame.

These ‘‘acceleration identities’’ are summarized in the following four equations:

$$\chi_{\alpha\beta}^{(1)}(\vec{G}, \vec{0}, \omega) = \frac{\bar{n}\gamma}{m} \left[ \delta_{\alpha\beta} - \frac{\chi^h(G, \omega)}{\chi^h(G)} P_{\alpha\beta}^L(\vec{G}) \right], \quad (14)$$

$$\chi_{\alpha\beta}^{(2)}(\vec{0}, \vec{0}, \omega) = -2 \left( \frac{\gamma \bar{m} G}{m\omega} \right)^2 \frac{1}{\chi^h(G)} \left[ 1 - \frac{\chi^h(G, \omega)}{\chi^h(G)} \right] P_{\alpha\beta}^L(\vec{G}), \quad (15)$$

$$f_{xc, \alpha\beta}^{(1)}(\vec{G}, \vec{0}, \omega) = -\frac{\gamma G^2}{\omega^2} \{ [f_{xcL}^h(G, \omega) - f_{xcL}^h(G, 0)] P_{\alpha\beta}^L(\vec{G}) + f_{xcT}^h(G, \omega) P_{\alpha\beta}^T(\vec{G}) \}, \quad (16)$$

$$f_{xc, \alpha\beta}^{(2)}(\vec{0}, \vec{0}, \omega) = -2\gamma f_{xc, \alpha\beta}^{(1)}(\vec{G}, \vec{0}, \omega), \quad (17)$$

where  $f_{xcL}^h(G, \omega)$  and  $f_{xcT}^h(G, \omega)$  are the longitudinal and transverse  $xc$  kernels of the homogeneous electron liquid at density  $\bar{n}$  and wave vector  $\vec{G}$ , while  $P_{\alpha\beta}^L(\vec{G}) \equiv G_\alpha G_\beta / G^2$  and  $P_{\alpha\beta}^T(\vec{G}) \equiv \delta_{\alpha\beta} - P_{\alpha\beta}^L(\vec{G})$  are the projectors parallel and perpendicular to the direction of  $\vec{G}$ , respectively. The derivation of these identities is briefly presented in Appendix A.

What makes these identities relevant to the evaluation of  $\vec{A}_{xc}(\vec{q}, \omega)$  is the fact that in our model,  $q$  is much smaller than  $G$  or  $k_F$ ; therefore, the quantities appearing in Eq. (13) can be evaluated in the  $q \rightarrow 0$  limit, where they reduce precisely to the quantities that appear in Eqs. (14)–(17). Underlying the calculation is of course the assumption that the  $q \rightarrow 0$  limits of the current response functions and  $xc$  kernels are regular—an assumption we have presently no reason to doubt.

By making use of Eqs. (14)–(17) and of the limiting form<sup>15</sup>

$$\lim_{q \rightarrow 0} \chi_{\alpha\beta}^h(\vec{q}, \omega) = \frac{\bar{n}}{m} \delta_{\alpha\beta} \quad (18)$$

in Eq. (13), and discarding all but the leading-order terms in  $q$ , we arrive, after some algebra, at

$$\vec{A}_{xc}(\vec{q}, \omega) = 2\gamma^2 \frac{\bar{n}\vec{G}(\vec{G} \cdot \vec{q})}{m\omega^2} \frac{\chi^h(G, \omega)}{\chi^h(G)} \times [f_{xcL}^h(G, \omega) - f_{xcL}^h(G, 0)] \frac{V}{\omega}. \quad (19)$$

This vector has a component perpendicular to  $\vec{q}$ , unless  $\vec{q}$  happens to be either parallel or perpendicular to  $\vec{G}$ : choosing  $\vec{q}$  in any other direction provides the desired example of non- $V$ -representability. Although our derivation has made use of Eq. (18), which is valid only in two dimensions, it is possible to show that the transverse part of Eq. (19) is unchanged in three dimensions.<sup>20</sup>

It is also interesting to ask what is the behavior of the Fourier components of  $\vec{A}_{xc}$  at wave vectors  $\vec{q} \pm \vec{G}$ . These components are first order in  $\gamma$  and one might think that they lead more directly to the desired result. Remarkably, this is not

the case: A calculation very similar to the one described in the previous paragraphs reveals that these components are purely longitudinal; i.e., parallel to  $\vec{q} \pm \vec{G}$ . One needs to go to at least second order in  $\gamma$  to see a transverse component of  $\vec{A}_{xc}$ .

#### IV. DISCUSSION

The example worked out in the previous section shows that a perfectly legitimate  $V$ -representable current density can turn out to be non- $V$ -representable in the noninteracting system. We believe that this state of affairs is generic. Only in exceptional cases will the current density be  $V$ -representable in both the interacting and the noninteracting versions of the same system. Hence, in general, the Kohn-Sham equation does not give the correct value of the transverse current density.

We may now ask, how big an error does one make if one insists on calculating the transverse current by means of the Kohn-Sham theory? Going back to our model system it is not difficult to see that the Fourier components of the Kohn-Sham potential are given by

$$V_{KS}(\vec{q} + l\vec{G}, \omega) = \frac{\omega}{|\vec{q} + l\vec{G}|} |\vec{A}_{s,L}(\vec{q} + l\vec{G}, \omega)|, \quad (20)$$

i.e., the Kohn-Sham potential is simply the scalar representation of the longitudinal part of the vector potential  $\vec{A}_s$  calculated in the previous section ( $\vec{A}_{s,L}$  and  $\vec{A}_{s,T}$  are the longitudinal and transversal component of  $\vec{A}_s$ , respectively). The above equation is accurate up to corrections of order  $\gamma^3$ . Indeed, because  $\vec{A}_{s,T}$  is of order  $\gamma^2$  its influence on the longitudinal current begins at order  $\gamma^3$ , implying that  $\vec{A}_{s,L}$  alone fully accounts for the longitudinal current density up to corrections of order  $\gamma^3$ . On the other hand, because  $V_{KS}$  is equivalent to  $\vec{A}_{s,L}$  it clearly fails to produce the part of the transverse current that is due to  $\vec{A}_{s,T}$ . This is of order  $\gamma^3$  for the Fourier components at wave vector  $\vec{q} + l\vec{G}$  with  $l \neq 0$  and of order  $\gamma^2$  for the Fourier component at wave vector  $\vec{q}$ . We conclude that the error on the transverse current is overall of order  $\gamma^2$ : this may partly explain the difficulty of finding examples in which the Kohn-Sham current density differs significantly from the exact one.

Where does this leave us with regard to the application of the time-dependent Kohn-Sham theory to the calculation of current densities? From a fundamental standpoint it is clear that only the time-dependent CDFT can provide the right answer. In TDCDFT one does not need  $V$ -representability, but only the much weaker  $\vec{A}$ -representability assumption. We know that this assumption holds true in linear response theory, and it is highly reasonable to assume that the set of  $\vec{A}$ -representable current densities is dense in the space of all current densities. On the other hand, we have also found that the error entailed by the use of the ordinary Kohn-Sham equation of TDDFT is of second order in the parameter that measures the strength of the density nonuniformity, and may

perhaps be reasonably neglected in practical implementations of the theory that are based on the local density approximation.

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### APPENDIX A: ACCELERATION IDENTITIES FOR THE CURRENT RESPONSE FUNCTIONS OF A WEAKLY INHOMOGENEOUS ELECTRON LIQUID

Our starting point is the acceleration identity in real space

$$\sum_{\delta} \int \chi_{\alpha\delta}(\vec{r}, \vec{r}', \omega) \left[ \delta_{\delta\beta} + \frac{\nabla'_{\delta} \nabla'_{\beta} V_0(\vec{r}')}{m(i\omega)^2} \right] d\vec{r}' = \frac{n_0(\vec{r})}{m} \delta_{\alpha\beta}, \quad (\text{A1})$$

which was derived in Ref. 14, beginning on p. 205. Both sides of this identity can be expanded in a series of  $\gamma$ . The Fourier transform of the coefficients of this expansion will give us the identities (14) and (15). The first order in  $\gamma$  evaluated at wave vector  $\vec{G}$  gives

$$\chi_{\alpha\beta}^{(1)}(\vec{G}, \vec{0}, \omega) + \sum_{\delta} \chi_{\alpha\delta}^h(\vec{G}, \omega) \frac{G_{\delta} G_{\beta}}{m\omega^2} V_0(\vec{G}) = \frac{\bar{n}\gamma}{m} \delta_{\alpha\beta}. \quad (\text{A2})$$

Inserting

$$V_0(\vec{G}) = \frac{\bar{n}\gamma}{\chi^h(\vec{G})} \quad (\text{A3})$$

from Eq. (6), and recalling that

$$\sum_{\delta} \chi_{\alpha\delta}^h(\vec{G}, \omega) G_{\delta} G_{\beta} = \chi^h(\vec{G}, \omega) \omega^2 P_{\alpha\beta}^L(\vec{G}), \quad (\text{A4})$$

where  $\chi^h(\vec{G}, \omega)$  is the density-density response function, we arrive at Eq. (14).

To derive Eq. (15) we take the  $q=0$  component of both sides of Eq. (A1) of the second order in  $\gamma$ . Since  $V_0(\vec{r})$  and  $n_0(\vec{r})$  are given by Eqs. (5) and (6) up to corrections of order  $\gamma^3$  we readily obtain

$$\begin{aligned} \chi_{\alpha\beta}^{(2)}(\vec{0}, \vec{0}, \omega) = & - \sum_{\delta} [\chi_{\alpha\delta}^{(1)}(\vec{0}, \vec{G}, \omega) + \chi_{\alpha\delta}^{(1)}(\vec{0}, -\vec{G}, \omega)] \\ & \times \frac{G_{\delta} G_{\beta}}{m\omega^2} V_0(\vec{G}). \end{aligned} \quad (\text{A5})$$

Inserting Eq. (14) for  $\chi_{\alpha\delta}^{(1)}(\vec{0}, \vec{G}, \omega)$  and Eq. (A3), we immediately arrive at Eq. (15).

We proceed similarly for the last two identities, Eqs. (16) and (17). The starting point in this case is Eq. (27) of Ref. 14:

$$\int f_{xc,\alpha\beta}(\vec{r}, \vec{r}', \omega) n_0(\vec{r}') d\vec{r}' = - \frac{\nabla_{\alpha} \nabla_{\beta} V_{s,xc}(\vec{r})}{\omega^2}, \quad (\text{A6})$$

where, to the required accuracy<sup>18</sup>

$$V_{s,xc}(\vec{r}) = 2\bar{n}\gamma f_{xcL}^h(\vec{G}, 0) \cos \vec{G} \cdot \vec{r} \quad (\text{A7})$$

is the exchange-correlation part of the static Kohn-Sham potential  $V_s(\vec{r})$ . Notice that  $f_{xcL}^h(\vec{G}, 0)$  is the *scalar* exchange-correlation kernel of the homogeneous electron liquid at zero frequency, quite different from the tensorial and frequency-dependent exchange-correlation kernel of the inhomogeneous system, which appears on the left-hand side of Eq. (A6). Taking the Fourier component at wave vector  $\vec{G}$  of both sides of Eq. (A6) to first order in  $\gamma$ , and recalling that

$$f_{xc,\alpha\beta}^h(\vec{G}, \omega) = \frac{G^2}{\omega^2} [f_{xcL}^h(\vec{G}, \omega) P_{\alpha\beta}^L(\vec{G}) + f_{xcT}^h(\vec{G}, \omega) P_{\alpha\beta}^T(\vec{G})], \quad (\text{A8})$$

we arrive at Eq. (16). Taking the Fourier component of both sides of Eq. (A6) at wave vector 0 to second order in  $\gamma$ , we finally arrive at Eq. (17).

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<sup>1</sup>P. Hohenberg and W. Kohn, Phys. Rev. **136**, B864 (1964).

<sup>2</sup>W. Kohn and L. J. Sham, Phys. Rev. **140**, A1133 (1965).

<sup>3</sup>R. M. Dreizler and E. K. U. Gross, *Density Functional Theory* (Springer-Verlag, Heidelberg, 1990).

<sup>4</sup>M. Levy, Proc. Natl. Acad. Sci. U.S.A. **76**, 6062 (1979).

<sup>5</sup>J. T. Chayes, L. Chayes, and M. B. Ruskai, J. Stat. Phys. **38**, 497 (1985).

<sup>6</sup>E. Runge and E. K. U. Gross, Phys. Rev. Lett. **52**, 997 (1984).

<sup>7</sup>*Density Functional Theory of Many-Fermion Systems*, edited by S. B. Trickey, Advances in Quantum Chemistry Vol. 21 (Academic, San Diego, 1990).

<sup>8</sup>E. K. U. Gross, J. F. Dobson, and M. Petersilka, in *Density Func-*

*tional Theory II*, edited by R. F. Nalewajski Topics in Current Chemistry Vol. 181 (Springer, Berlin, 1996), p. 81.

<sup>9</sup>S. K. Ghosh and A. K. Dhara, Phys. Rev. A **38**, 1149 (1988).

<sup>10</sup>G. Vignale and W. Kohn, Phys. Rev. Lett. **77**, 2037 (1997).

<sup>11</sup>C. A. Ullrich and G. Vignale, Phys. Rev. B **65**, 245102 (2002).

<sup>12</sup>N. T. Maitra, I. Souza, and K. Burke, Phys. Rev. B **68**, 045109 (2003).

<sup>13</sup>G. Vignale, Phys. Rev. B **70**, 201102(R) (2004).

<sup>14</sup>G. Vignale and W. Kohn, in *Electronic Density Functional Theory: Recent Progress and New Directions*, edited by J. F. Dobson, G. Vignale, and M. P. Das (Plenum, New York, 1996), p. 199.

<sup>15</sup>G. F. Giuliani and G. Vignale, *Quantum Theory of the Electron*

*Liquid* (Cambridge University Press, Cambridge, UK, 2005).

<sup>16</sup>For a review of some of the rigorous results on the  $V$ -representability in DFT, see R. van Leeuwen, *Adv. Quantum Chem.* **43**, 25 (2003).

<sup>17</sup>The possibility that a  $V$ -representable current might be non- $V$ -representable in a noninteracting system was noted by N. T. Maitra, K. Burke, H. Appel, E. K. U. Gross, and R. van Leeuwen, in *Reviews in Modern Quantum Chemistry: A celebration of the contributions of Robert Parr*, edited by K. Sen (World Scientific, Singapore, 2002), p. 1186.

<sup>18</sup>In this form of the potential some higher order harmonics, e.g., the ones with wave vectors  $\pm 2\vec{G}$ , have been neglected. However, these higher harmonics, being themselves of order  $\gamma^2$ , produce only higher order corrections (at least  $\gamma^3$ ) in the subsequent cal-

culations. For this reason they have been neglected.

<sup>19</sup>Here and in the following, the quantities with the superscript  $h$  refer to the *homogeneous* system of density  $\bar{n}$ . Their Fourier transforms have a single wave vector argument.

<sup>20</sup>In three dimensions, one has  $\chi_{\alpha\beta}^h(\vec{q}, \omega) = \chi_L^h(\vec{q}, \omega) P_{\alpha\beta}^L(\hat{q}) + \chi_T^h(\vec{q}, \omega) P_{\alpha\beta}^T(\hat{q})$ , where (see Ref. 15, Chap. 5)  $\lim_{q \rightarrow 0} \chi_L^h(\vec{q}, \omega) = [\bar{n}/m(1 - \omega_p^2/\omega^2)]$ ,  $\lim_{q \rightarrow 0} \chi_T^h(\vec{q}, \omega) = \bar{n}/m$ , and  $\omega_p = \sqrt{4\pi\bar{n}e^2/m}$  is the plasmon frequency in three dimensions at  $q=0$ . However, these modifications contribute only to the longitudinal part of  $\vec{A}_{xc}$ : the transverse part of Eq. (19) remains unchanged. Notice that in two dimensions, one has  $\omega_p=0$  (at  $q=0$ ), and thus the longitudinal and transverse parts of the current-current response function coincide in the limit  $q \rightarrow 0$ , as seen in Eq. (18).