Solving the Ultranonlocality Problem in Time-Dependent Spin-Density-Functional Theory

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It has been known for some time that the exchange-correlation potential in time-dependent density-functional theory is an intrinsically nonlocal functional of the density as soon as one goes beyond the adiabatic approximation. In this paper we show that a much more severe nonlocality problem, with a completely different physical origin, plagues the exchange-correlation potentials in time-dependent *spin*-density functional theory. We show how the use of the *spin current density* as a basic variable solves this problem, and we provide an explicit local expression for the exchange-correlation fields as functionals of the spin currents.

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For many years the local density approximation (LDA) has provided the much needed handle on the difficult problem of approximating the density dependence of the exchange-correlation (xc) potential—the single particle potential that incorporates the many-body effects in the Kohn-Sham equation for the ground state density [1]. In LDA, the xc potential $V_{\rm xc}(\vec{r})$ is simply a function of the local density $n(\vec{r})$. This approximation is not unreasonable as long as the functional derivative of $V_{\rm xc}(\vec{r})$ with respect to $n(\vec{r}')$—the so-called *exchange-correlation kernel* $f_{\rm xc}(\vec{r}, \vec{r}') \equiv [\delta V_{\rm xc}(\vec{r})/\delta n(\vec{r}')]$—is a sufficiently short-ranged function of the distance $|\vec{r} - \vec{r}'|$ [2].

However, much recent work [3–7] has demonstrated that the requirement of short rangedness is not always fulfilled in physical systems, and when this happens the local density approximation is flawed. This does not mean that a local description of exchange and correlation is absolutely impossible, only that such a description cannot be achieved in terms of the particle density.

For example, in the density-functional theory of crystalline insulators it has been found [4–6] that the xc potential has an "ultranonlocal" dependence on the density, due to the fact that the Fourier transform of the xc kernel $f_{\rm xc}(\vec{k}, \vec{k})$ diverges as $1/k^2$ for $k \to 0$. But, the ultranonlocality disappears if one reformulates the theory in terms of the electric polarization $\vec{P}(\vec{r})$ and the exchange-correlation electric field $\vec{E}_{\rm xc}(\vec{r})$ associated with it.

Another instance of the ultranonlocality problem was discovered in the time-dependent density-functional theory (TDDFT) [8] following the realization that the frequency-dependent LDA [9] fails to satisfy Kohn's theorem [10,11]. The pathology was traced to a singularity of the form $\frac{\vec{k}\cdot\vec{k}'}{k^2}$ in the xc kernel $f_{\rm xc}(\vec{k}, \vec{k}', \omega)$ for $k \to 0$ at finite $\vec{k}'$ and ω. The ensuing nonlocality problem was solved by upgrading to time-dependent *current*-density functional theory (TDCDFT), where the basic variable is the current density, and its conjugate field is a vector potential[3]. TDCDFT has since been applied to the calculation of the optical spectra of solids [12] and the polarizability of long polymer chains [13] with considerable success.

In this Letter we show that the ultranonlocality problem occurs in an *aggravated form* in the time-dependent spin-density functional theory or, more generally, in the time-dependent DFT of any multi-component system. The novel features of the spin-dependent problem stem from the fact that the xc kernel presents a divergence even in the homogeneous electron liquid. More precisely, it can be shown that the Fourier transform of the spin-dependent exchange-correlation kernel $f_{{\rm xc},\sigma\sigma'}(r - r', t - t') \equiv [\delta V_{{\rm xc},\sigma}(\vec{r}, t)/\delta n_{\sigma'}(\vec{r}', t')]$ in a homogeneous electron liquid has the long-wavelength expansion

$$f_{{\rm xc},\sigma\sigma'}(k, \omega) \xrightarrow{k\to 0} \frac{A(\omega)}{k^2}\frac{\sigma\sigma' n^2}{4n_\sigma n_{\sigma'}} + B_{\sigma\sigma'}(\omega) + O(k^2), \quad (1)$$

where $A(\omega)$ and $B_{\sigma\sigma'}(\omega)$ are complex functions of frequency, n_σ is the density of σ-spin electrons ($\sigma = +1$ for $\uparrow$-spin and $\sigma = -1$ for $\downarrow$-spin), and $n = n_\uparrow + n_\downarrow$ is the total density. Since the xc potential created by a small density variation $\delta n_\sigma(\vec{k}, \omega)$ is given by the formula

$$V_{{\rm xc},\sigma}(\vec{k}, \omega) = \sum_{\sigma'} f_{{\rm xc},\sigma\sigma'}(k, \omega)\delta n_{\sigma'}(\vec{k}, \omega), \quad (2)$$

we see that Eq. (1) rules out the possibility of a local connection between $V_{{\rm xc},\sigma}(\vec{r}, t)$ and $\delta n_{\sigma'}(\vec{r}', t')$.

The existence of the long-wavelength singularity in $f_{{\rm xc},\sigma\sigma'}(k, \omega)$ has been known for some time. It was first pointed out by Goodman and Sjölander [14] that the third-moment sum rule for the spin-density response function implies such a singularity. Approximate formulae for $f_{{\rm xc},-}(k, \omega) = f_{{\rm xc},\uparrow\uparrow}(k, \omega) - f_{xc,\uparrow\downarrow}(k, \omega)$ exhibiting the singularity were proposed in [15] and, for imaginary frequencies, in [16]. More recently, D'Amico and Vignale [17] have shown that, at low frequency and finite temperature, the singularity is related to the friction that arises between up- and down-spin currents when they

have different average velocities (the so-called spin-drag effect).

By contrast, the implications of Eq. (1) for spin-density functional theory have not been explored so far. This is understandable, since the singularity (1) arises only at finite frequency [$A(0) = 0$] and therefore does not affect the *static* spin DFT. Furthermore, the singularity does not show up in the density response of spin-compensated systems, since the relevant combination of xc kernels, in that case, is $\sum_{\sigma\sigma'} n_\sigma n_{\sigma'} f_{\mathrm{xc},\sigma\sigma'}$, which is nonsingular. It is only in the time-dependent spin DFT [18] that the issue of the singularity becomes really critical not only to the calculation of the spin-response, but even to the calculation of just the density response[19].

In this Letter we propose a resolution of the ultranonlocality problem based on the use of the spin components of the current density $\vec{j}_\uparrow(\vec{r}, \omega)$ and $\vec{j}_\downarrow(\vec{r}, \omega)$ as basic variables. We provide an explicit expression for the spin-dependent exchange-correlation field $\vec{E}_{\mathrm{xc},\sigma}(\vec{r}, \omega)$ as a local linear functional of the currents $\vec{j}_\sigma$.

The general method for upgrading from the density to the current-density formulation is described in detail in Ref. [20], so we mention only the essential steps here. We introduce a spin-dependent xc vector potential $\vec{A}_{\mathrm{xc},\sigma}(\vec{k}, \omega)$ [whose time derivative, $i\omega\vec{A}_{\mathrm{xc},\sigma}(\vec{k}, \omega) = \vec{E}_{\mathrm{xc}}(\vec{k}, \omega)$, is the xc electric field], and notice that this is linearly related to the currents in the following manner

$$A^\alpha_{\mathrm{xc},\sigma}(\vec{k}, \omega) = \frac{k^2}{\omega^2} \sum_{\sigma'} f^\alpha_{\mathrm{xc},\sigma\sigma'}(\vec{k}, \omega) j^\alpha_{\sigma'}(\vec{k}, \omega), \qquad (3)$$

where the superscript α denotes the longitudinal ($\alpha = L$) or transverse ($\alpha = T$) component of a vector relative to the direction of $\vec{k}$. It is not difficult to see that the *longitudinal* xc kernel defined in this manner coincides with the xc kernel of Eq. (1). The extra factor k^2/ω^2 in Eq. (3) exactly cancels the small-k singularity of f_{xc}, and leads to a theory that admits a local approximation. The imaginary part of the current xc kernel $f^\alpha_{\mathrm{xc},\sigma\sigma'}(k, \omega)$ is expressed in terms of a causal response function as follows:

$$\mathrm{Im} f^\alpha_{\mathrm{xc},\sigma\sigma'}(k, \omega) = \frac{1}{V n_\sigma n_{\sigma'} k^2} \mathrm{Im} \langle\langle \hat{F}^\alpha_\sigma(\vec{k}); \hat{F}^\alpha_{\sigma'}(-\vec{k}) \rangle\rangle_\omega, \qquad (4)$$

where $\langle\langle \hat{A}; \hat{B} \rangle\rangle_\omega \equiv -\frac{i}{\hbar} \int_0^\infty \langle [\hat{A}(t), \hat{B}] \rangle e^{i\omega t} dt$ is the linear response function associated with the operators $\hat{A}$ and $\hat{B}$; $\hat{F}^\alpha_\sigma(\vec{k}) = -\frac{im}{\hbar}[\hat{H}, \hat{j}^\alpha_\sigma(\vec{k})]$ is the time derivative of the Fourier transform of the current-density operator $\hat{\vec{j}}_\sigma(\vec{k})$, $\hat{H}$ is the Hamiltonian, and V is the volume.

Once the imaginary part of $f^\alpha_{\mathrm{xc},\sigma\sigma'}(k, \omega)$ is known, its real part is determined by the Kramers-Krönig dispersion relation

$$\mathrm{Re} f^\alpha_{\mathrm{xc},\sigma\sigma'}(k, \omega) = f^\alpha_{\mathrm{xc},\sigma\sigma'}(k, \infty) - \frac{2}{\pi} \mathcal{P} \int_0^\infty d\omega' \frac{\omega' \mathrm{Im} f^\alpha_{\mathrm{xc},\sigma\sigma'}(k, \omega')}{\omega^2 - \omega'^2}, \qquad (5)$$

where $\mathcal{P}$ denotes the principal part integral, and the infinite frequency limit of $f_{\mathrm{xc},\sigma\sigma'}$ is determined by the *third moment sum rule*. In a three-dimensional electron liquid, this sum rule gives

$$f^\alpha_{\mathrm{xc},\sigma\sigma'}(k, \infty) \xrightarrow{k\to 0} -\frac{4\pi e^2}{3k^2} \frac{n_\uparrow n_\downarrow}{n_\sigma n_{\sigma'}} [g_{\uparrow\downarrow}(0) - 1] \sigma\sigma' + a^\alpha \frac{t_{c\sigma}}{n_\sigma} \delta_{\sigma\sigma'} + \frac{b^\alpha}{2} \int d\vec{r} \frac{e^2}{r} [g_{\sigma\sigma'}(r) - 1], \qquad (6)$$

where $a^L = 2$, $a^T = 2/3$, $b^L = 4/15$, and $b^T = -2/15$. Here $g_{\sigma\sigma'}(r)$ is the spin-resolved pair correlation function and $t_{c\sigma}$ is the average correlation kinetic energy of the σ-spin component. Note that the result for the longitudinal case was first obtained in Ref. [14].

It is evident from the above equations that both the longitudinal and the transverse kernels exhibit $1/k^2$ singularities, which are "cured" by the k^2/ω^2 factor of Eq. (3). In particular, substituting the small-k expansion $\hat{F}^\alpha_\sigma(\vec{k}) = \hat{F}^\alpha_\sigma(0) + O(\vec{k})$ in Eq. (4), where $\hat{F}^\alpha_\sigma(0)$ is the operator of the total force acting on σ-spin electrons, and noting that terms of first order in $\vec{k}$ vanish by inversion symmetry, we see that the xc kernels have the small-k expansion

$$f^\alpha_{\mathrm{xc},\sigma\sigma'}(k, \omega) \xrightarrow{k\to 0} \frac{A(\omega)}{k^2} \frac{\sigma\sigma' n^2}{4 n_\sigma n_{\sigma'}} + B^\alpha_{\sigma\sigma'}(\omega) + O(k^2), \qquad (7)$$

where

$$\mathrm{Im} A(\omega) = -\frac{4}{V n^2} \mathrm{Im} \langle\langle \hat{F}^\alpha_\uparrow; \hat{F}^\alpha_\downarrow \rangle\rangle_\omega \qquad (8)$$

and

$$\mathrm{Re} A(\omega) = -\frac{16\pi e^2}{3} [g_{\uparrow\downarrow}(0) - 1] - \frac{2}{\pi} \mathcal{P} \int_0^\infty d\omega' \frac{\omega' \mathrm{Im} A(\omega')}{\omega^2 - \omega'^2}. \qquad (9)$$

The factor $\sigma\sigma'$ in Eq. (7) arises from the fact that the total force $\hat{F}_\uparrow + \hat{F}_\downarrow$ vanishes, due to translational invariance, so that $\langle\langle \hat{F}_\sigma; \hat{F}_{\sigma'} \rangle\rangle_\omega = -\sigma\sigma' \langle\langle \hat{F}_\uparrow; \hat{F}_\downarrow \rangle\rangle_\omega$. Notice also that $A(\omega)$ is independent of the direction α—longitudinal or transverse. The microscopic expression for $B^\alpha_{\sigma\sigma'}$ is more complicated: a simple approximation for this quantity will be presented below.

Substituting the expansion (7) into Eq. (3), calculations similar to those described in [20] lead us to the following *local* approximation for the xc field in terms of the spin currents

$$-e\vec{E}_{\mathrm{xc},\sigma}(\omega) = -\vec{\nabla}V^{\mathrm{LDA}}_{\mathrm{xc},\sigma} + \frac{1}{n_\sigma}\vec{\nabla}\cdot\vec{\sigma}_{\mathrm{xc},\sigma}(\omega) + \frac{in^2A(\omega)}{4\omega}\sum_{\sigma'}\frac{\sigma\sigma'}{n_\sigma n_{\sigma'}}\vec{j}_{\sigma'}. \quad (10)$$

Here the $\vec{r}$ dependence has been left implicit, and the xc stress tensor $\vec{\sigma}_{\mathrm{xc}}(\omega)$, as well as $A(\omega)$, is a function of the local spin densities, as discussed below.

Equation (10) is the central result of this paper. The first two terms on the right are well known: they are, respectively, the adiabatic LDA contribution and the viscoelastic force term, where the stress tensor $\sigma_{\mathrm{xc},\sigma}(\omega)$ is related to $B_{\mathrm{xc},\sigma\sigma'}$ by obvious extensions of the formulas reported in [20]. The expression for the xc stress tensor is

$$\sigma_{\mathrm{xc},\sigma,ij} = \sum_{\sigma'}\Big[\eta_{xc,\sigma\sigma'}\Big(\frac{\partial u_{\sigma',i}}{\partial r_j} + \frac{\partial u_{\sigma',j}}{\partial r_i} - \frac{2}{3}\vec{\nabla}\cdot\vec{u}_{\sigma'}\delta_{ij}\Big) + \zeta_{\mathrm{xc},\sigma\sigma'}\vec{\nabla}\cdot\vec{u}_{\sigma'}\delta_{ij}\Big], \quad (11)$$

where $\vec{u}_\sigma = \vec{j}_\sigma/n_\sigma$, and

$$\eta_{\mathrm{xc},\sigma\sigma'} = -\frac{n_\sigma n_{\sigma'}}{i\omega}B^T_{\sigma\sigma'}(\omega), \quad (12)$$

$$\zeta_{\mathrm{xc},\sigma\sigma'} = -\frac{n_\sigma n_{\sigma'}}{i\omega}\Big[B^L_{\sigma\sigma'}(\omega) - \frac{4}{3}B^T_{\sigma\sigma'}(\omega) - \epsilon''_{\mathrm{xc},\sigma\sigma'}\Big], \quad (13)$$

where $\epsilon''_{\mathrm{xc},\sigma\sigma'} = (\partial^2\epsilon_{\mathrm{xc}}/\partial n_\sigma\partial n_{\sigma'})$. The last term in Eq. (10) is new, and comes directly from the $1/k^2$ singularity of Eq. (7). The essential feature of the new term is that it produces damping of the spin-current proportional to the relative velocity between up- and down-spin electrons. This makes it readily distinguishable from the usual viscous friction contained in the second term, which is proportional to the *derivatives* of the velocity field. The physical reason for the difference is that, whenever up and down-spin currents travel with different average velocities, they exert *friction* on each other: the "spin drag coefficient" is $\gamma(\omega) = [in^3A(\omega)/4\omega m n_\uparrow n_\downarrow]$. Of course, like all the quantities considered here, $\gamma(\omega)$ is complex and frequency dependent, and, in the limit of zero frequency, its real part can be shown to be related to the spin diffusion constant D_s by the Einstein relation $D_s = [n/m\chi_s\gamma(0)]$, where χ_s is the static, macroscopic spin susceptibility.

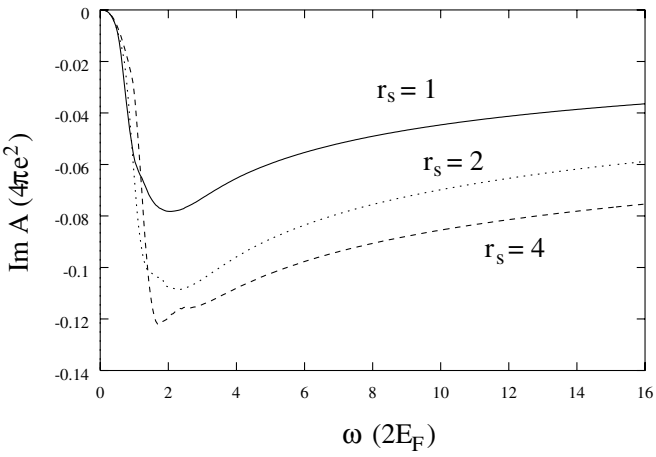

FIG. 1. Imaginary part of $A(\omega)$ evaluated from Eq. (14) with the correction factor given in Eq. (16). The values of $a(r_s, 0)$ are 1.92, 3.36, and 7.49 at $r_s = 1$, 2, and 4, respectively.

Unfortunately, an exact calculation of $A(\omega)$ from the microscopic expressions (8) and (9) is beyond the reach of present-day many-body techniques. However, we can obtain a rather good approximation with the help of the following exact results: (i) For $\omega \to 0$, $\mathrm{Im}A(\omega) \propto \omega^3$ and $\mathrm{Re}A(\omega) \propto \omega^2$; (ii) For large ω, $\mathrm{Im}A(\omega) \to -(16\pi e^2/3) \times (n_\uparrow n_\downarrow/n^2)(\alpha r_s/\sqrt{\bar{\omega}})[1/(1+\zeta)^{1/3}]$ and $\mathrm{Re}A(\omega) \to -(16\pi e^2/3)(n_\uparrow n_\downarrow/n^2)[g_{\uparrow\downarrow}(0) - 1]$. Here $\bar{\omega} = (\omega/2E_{F\uparrow})$, where $E_{F\uparrow}$ is the Fermi energy for majority spin electrons and $\zeta = (n_\uparrow - n_\downarrow)/n$ measures the degree of spin polarization, and $\alpha = (4/9\pi)^{1/3}$ [21]. Note that $g_{\uparrow\downarrow}(0)$ is accurately known from the work of Gori-Giorgi and Perdew [22]. The high and low frequency limits of $\mathrm{Re}A(\omega)$ are both obtained from the third-moment sum rule. In particular, the vanishing of $\mathrm{Re}A(0)$ follows from the fact that $\frac{2}{\pi}\int_0^\infty[\mathrm{Im}A(\omega')/\omega']$ is equal to (minus) the first moment of the current-current response function, which, by gauge invariance and the continuity equation, coincides with the third-moment of the density-density response function, i.e., $-A(\infty)$.

The ω^3 behavior of $\mathrm{Im}A(\omega)$ at low frequency is easily obtained from the approximate zero-temperature formula [17]

$$\mathrm{Im}A(\omega) \simeq -\frac{4}{3n^2V}\sum_{\vec{q}} v^2_{\vec{q}}q^2\int_0^\omega\frac{d\omega'}{\pi}[\mathrm{Im}\chi_{\uparrow\uparrow}(q,\omega-\omega')\mathrm{Im}\chi_{\downarrow\downarrow}(q,\omega') - \mathrm{Im}\chi_{\uparrow\downarrow}(q,\omega-\omega')\mathrm{Im}\chi_{\downarrow\uparrow}(q,\omega')], \quad (14)$$

which is exact in the limits of high density and high frequency. Here $v_{\vec{q}} = \frac{4\pi e^2}{q^2}$ and $\chi_{\sigma\sigma'}(q,\omega)$ are the spin-density response functions of the homogeneous liquid.

We have evaluated $\chi_{\sigma\sigma'}$ in the generalized random phase approximation

$$\chi^{-1}_{\sigma\sigma'}(q,\omega) = [\chi^{(0)}_\sigma]^{-1}(q,\omega)\delta_{\sigma\sigma'} - v_{\vec{q}}[1 - G_{\sigma\sigma'}(q)], \quad (15)$$

where $\chi^{(0)}_\sigma(q,\omega)$ is the Lindhard function and $G_{\sigma\sigma'}(q)$ are local field corrections [23]. At typical metallic densities we multiply $\mathrm{Im}A(\omega)$ by an empirical factor

$$g(\omega) = \frac{1+\sqrt{\bar{\omega}}}{a(r_s,\zeta)+\sqrt{\bar{\omega}}}, \quad (16)$$

designed to satisfy the condition $\mathrm{Re}A(0) = 0$ without

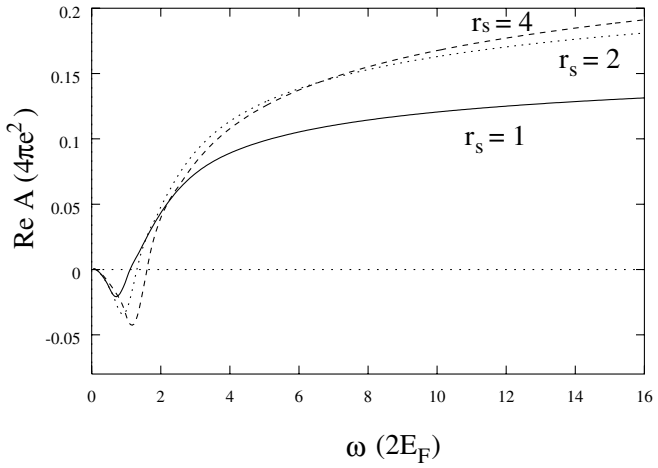

FIG. 2. Real part of $A(\omega)$ obtained from Eq. (9).

altering the high-frequency behavior. Notice that $a(r_s, \zeta) \to 1$ for $r_s \to 0$. The results evaluated with this procedure are shown in Figs. 1 and 2.

Finally, we briefly remark on the calculation of the regular part of the xc kernel. The spin symmetric combinations $B^{\alpha}_{+} \equiv B^{\alpha}_{\uparrow\uparrow} + B^{\alpha}_{\uparrow\downarrow}$ with $\alpha = L$ or T have been approximately calculated, *in the paramagnetic state* ($\zeta = 0$), by two of us [24]. For the antisymmetric combinations $B^{\alpha}_{-} \equiv B^{\alpha}_{\uparrow\uparrow} - B^{\alpha}_{\uparrow\downarrow}$ a provisional solution is offered by the time-honored Gross-Kohn interpolation formula [9], with parameters suitably tuned to satisfy the exact identity $\lim_{\omega\to 0}\{[B^{L}_{\sigma\sigma'}(\omega) - \frac{4}{3}B^{T}_{\sigma\sigma'}(\omega) - \epsilon''_{\mathrm{xc},\sigma\sigma'}]/\omega\} = 0$ [25], the third-moment sum rule, and the limiting form $B^{\alpha}_{-}(\omega) \to \omega^{-3/2}$ for $\omega \to \infty$. Finally, for general polarization, we propose to use the formula $B^{\alpha}_{\sigma\sigma'}(\zeta) = P_{\sigma\sigma'}(\zeta) B^{\alpha}_{\sigma\sigma'}(\zeta = 0)$ where $P_{\sigma\sigma'}(\zeta) = [\epsilon''_{\mathrm{xc},\sigma\sigma'}(\zeta)/\epsilon''_{\mathrm{xc},\sigma\sigma'}(0)]$, so that the above conditions are satisfied for all ζ. This completes the construction of the input for Eq. (10). We hope that the new expression for the spin-current dependent xc field will stimulate applications of CDFT to the calculation of spin excitations in spin-polarized systems.

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