Spin-Density Functionals from Current-Density Functional Theory and Vice Versa: A Road towards New Approximations

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It is shown that the exchange-correlation functional of spin-density functional theory is identical, on a certain set of densities, with the exchange-correlation functional of current-density functional theory. This rigorous connection is used to construct new approximations of the exchange-correlation functionals. These include a conceptually new generalized-gradient spin-density functional and a nonlocal current-density functional. [S0031-9007(97)02428-9]

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Density-functional theory (DFT) has been widely and very successfully applied in solid-state physics and quantum chemistry [1]. The power of the method lies in its numerical simplicity which follows from mapping the interacting many-body system of interest onto an auxiliary system (the so-called Kohn-Sham system) of noninteracting particles moving in an effective single-particle potential. The latter consists of the external potential, the Hartree term, and the so-called exchange-correlation (xc) potential which contains all the many-body aspects of the original interacting system. In practice, this quantity has to be approximated. To deal with the presence of external magnetic fields, two generalizations of DFT have been developed; spin-density functional theory (SDFT) [2] and current-density functional theory (CDFT) [3]. In SDFT only the Zeeman term is taken into account, while in CDFT the coupling of the magnetic field to the orbital currents is also included, thus allowing for the treatment of systems in strong magnetic fields. CDFT has been used successfully to describe the magnetic behavior of atoms, molecules [4], and extended systems [5]. However, to date, there exist only a few approximations of the xc functional of CDFT: The local density approximation (LDA) of the exchange energy is known exactly [6], and the LDA of the correlation energy has been calculated [7] within the RPA. In SDFT, on the other hand, a large variety of approximate xc functionals is available. These include the LDA (beyond RPA), gradient dependent approximations (GDA), as well as nonlocal schemes [1]. The purpose of the present Letter is to establish an exact connection between SDFT and CDFT which is then used to construct new approximate functionals for both SDFT and CDFT.

The Hamiltonian of SDFT is given by

$$\hat{H}^S = \hat{T} + \hat{V} + \hat{W} - \int d^3r\, \hat{\mathbf{m}}(\mathbf{r}) \cdot \mathbf{B}(\mathbf{r}), \qquad (1)$$

where, as usual, $\hat{T}$, $\hat{V}$, and $\hat{W}$ stand for the operators of kinetic energy, external potential, and particle-particle interaction, respectively. The Hamiltonian of CDFT in turn is

$$\hat{H}^{C1} = \hat{T} + \hat{V} + \hat{W} - \int d^3r\, \hat{\mathbf{m}}(\mathbf{r}) \cdot \mathbf{B}(\mathbf{r}) - \frac{q}{c} \int d^3r\, \hat{\mathbf{j}}_p(\mathbf{r}) \cdot \mathbf{A}(\mathbf{r}) + \frac{q^2}{2mc^2} \int d^3r\, \hat{n}(\mathbf{r}) \mathbf{A}(\mathbf{r})^2, \qquad (2)$$

where q denotes the charge of particles involved. The density is defined in terms of field operators as

$$n(\mathbf{r}) = \langle \hat{n}(\mathbf{r}) \rangle = \sum_\sigma \langle \hat{\Psi}^\dagger_\sigma(\mathbf{r}) \hat{\Psi}_\sigma(\mathbf{r}) \rangle . \qquad (3)$$

The physical current density is given by $\mathbf{j}(\mathbf{r}) = \mathbf{j}_p(\mathbf{r}) + \mathbf{j}_d(\mathbf{r}) + \mathbf{j}_s(\mathbf{r})$, where

$$\mathbf{j}_p(\mathbf{r}) = \langle \hat{\mathbf{j}}_p(\mathbf{r}) \rangle = \frac{\hbar}{2mi} \langle [\hat{\Psi}^\dagger(\mathbf{r}) \nabla \hat{\Psi}(\mathbf{r}) - \nabla \hat{\Psi}^\dagger(\mathbf{r}) \hat{\Psi}(\mathbf{r})] \rangle \qquad (4)$$

is the paramagnetic current density,

$$\mathbf{j}_d(\mathbf{r}) = -\frac{q}{mc} n(\mathbf{r}) \mathbf{A}(\mathbf{r}) \qquad (5)$$

is the diamagnetic contribution, and

$$\mathbf{j}_s(\mathbf{r}) = \frac{c}{q} \nabla \times \mathbf{m}(\mathbf{r}) \qquad (6)$$

is the spin-current density. The spin magnetization $\mathbf{m}(\mathbf{r})$ is finally given by

$$\mathbf{m}(\mathbf{r}) = \langle \hat{\mathbf{m}}(\mathbf{r}) \rangle = \mu_0 \sum_{\alpha\beta} \langle \hat{\Psi}^\dagger_\alpha(\mathbf{r}) \boldsymbol{\sigma}_{\alpha\beta} \hat{\Psi}_\beta(\mathbf{r}) \rangle \qquad (7)$$

with the Bohr magneton $\mu_0 = q\hbar/2mc$ and the vector of the Pauli matrices $\boldsymbol{\sigma}$. In the following, the physical (i.e., gauge invariant) orbital current will be denoted as $\mathbf{j}_{\mathrm{orb}}(\mathbf{r}) := \mathbf{j}_p(\mathbf{r}) + \mathbf{j}_d(\mathbf{r})$, while the sum of the paramagnetic and spin currents will be abbreviated as $\mathbf{j}_m(\mathbf{r}) := \mathbf{j}_p(\mathbf{r}) + \mathbf{j}_s(\mathbf{r})$.

There exist several formulations of CDFT which differ by the choice of the current-density variable: From the form of the Hamiltonian H^{C1} it appears most natural to use $\mathbf{j}_p$ together with n and $\mathbf{m}$. This choice was adopted by Vignale and Rasolt in the original formulation [3] of CDFT, with the (commonly applied) additional

 0031-9007/97/78(10)/1872(4)$10.00

assumption of $\mathbf{m}$ having a fixed direction in space. Alternatively, one can insert $\mathbf{B} = \nabla \times \mathbf{A}$ in (2) and then perform a partial integration on the Zeeman term, leading to $-\frac{q}{c}\int d^3r\,\mathbf{j}_s \cdot \mathbf{A}$ plus a surface term which vanishes for finite systems. This yields the Hamiltonian

$$\hat{H}^{C2} = \hat{T} + \hat{V} + \hat{W} - \frac{q}{c}\int d^3r\,\hat{\mathbf{j}}_m(\mathbf{r})\cdot\mathbf{A}(\mathbf{r}) + \frac{q^2}{2mc^2}\int d^3r\,\hat{n}(\mathbf{r})\mathbf{A}(\mathbf{r})^2, \quad (8)$$

suggesting the use of the densities n and $\mathbf{j}_m$. For each of the three Hamiltonians, (1), (2), and (8), HK- and KS-type theorems can be formulated in the usual way, for all v-representable densities $(n,\mathbf{m})$ $(n,\mathbf{m},\mathbf{j}_p)$, and $(n,\mathbf{j}_m)$, respectively. In particular, the many-body ground states of (1), (2), and (8) are uniquely determined by the respective densities, i.e., $\Psi^S = \Psi^S[n,\mathbf{m}]$, $\Psi^{C1} = \Psi^{C1}[n,\mathbf{m},\mathbf{j}_p]$, and $\Psi^{C2} = \Psi^{C2}[n,\mathbf{j}_m]$. Likewise, the corresponding KS Slater determinants are uniquely fixed by these densities, i.e., $\Phi^S = \Phi^S[n,\mathbf{m}]$, $\Phi^{C1} = \Phi^{C1}[n,\mathbf{m},\mathbf{j}_p]$, and $\Phi^{C2} = \Phi^{C2}[n,\mathbf{j}_m]$.

For *finite* systems, H^{C1} (with $\mathbf{B} = \nabla \times \mathbf{A}$) and H^{C2} are identical. Hence the corresponding many-body wave functions must be identical as well, i.e., $\Psi^{C1}[n,\mathbf{m},\mathbf{j}_p] = \Psi^{C2}[n,\mathbf{j}_m = \mathbf{j}_p + \frac{c}{q}\nabla\times\mathbf{m}]$. As an immediate consequence, we find

$$\begin{aligned} F^{C1}[n,\mathbf{m},\mathbf{j}_p] &= \langle\Psi^{C1}|\hat{T} + \hat{W}|\Psi^{C1}\rangle \\ &= \langle\Psi^{C2}|\hat{T} + \hat{W}|\Psi^{C2}\rangle \\ &= F^{C2}\Big[n,\mathbf{j}_m = \mathbf{j}_p\frac{c}{q}\nabla\times\mathbf{m}\Big]. \end{aligned} \quad (9)$$

According to the Levy-Lieb constrained-search formulation [8] of DFT and its recent extension [9] to CDFT, the KS ground state $\Phi^{C1}[n,\mathbf{m},\mathbf{j}_p]$ is the determinant which minimizes $\langle\hat{T}\rangle$ and simultaneously yields $(n,\mathbf{m},\mathbf{j}_p)$. Likewise, $\Phi^{C2}[n,\mathbf{j}_m]$ is the determinant which minimizes $\langle\hat{T}\rangle$ and simultaneously yields $(n,\mathbf{j}_m)$. However, since Φ^{C1} also yields $\mathbf{j}_m = \mathbf{j}_p + \frac{c}{q}\nabla\times\mathbf{m}$, we conclude that $\Phi^{C1}[n,\mathbf{m},\mathbf{j}_p] = \Phi^{C2}[n,\mathbf{j}_m = \mathbf{j}_p + \frac{c}{q}\nabla\times\mathbf{m}]$. Consequently, the noninteracting kinetic energy functionals are identical:

$$\begin{aligned} T_s^{C1}[n,\mathbf{m},\mathbf{j}_p] &= \langle\Phi^{C1}|\hat{T}|\Phi^{C1}\rangle = \langle\Phi^{C2}|\hat{T}|\Phi^{C2}\rangle \\ &= T_s^{C2}\Big[n,\mathbf{j}_m = \mathbf{j}_p + \frac{c}{q}\nabla\times\mathbf{m}\Big]. \end{aligned} \quad (10)$$

For each of the three Hamiltonians, (1), (2), and (8), the xc energy is defined as $E_{\rm xc} = F - T_s - \frac{1}{2}\int d^3r d^3r' n(\mathbf{r})w(\mathbf{r},\mathbf{r}')n(\mathbf{r}')$. Therefore, as an immediate consequence of (9) and (10), we obtain

$$E_{\rm xc}^{C1}[n,\mathbf{m},\mathbf{j}_p] = E_{\rm xc}^{C2}\Big[n,\mathbf{j}_m = \mathbf{j}_p + \frac{c}{q}\nabla\times\mathbf{m}\Big]. \quad (11)$$

Using this identity, we find for the xc potentials $v_{\rm xc}$, $\mathbf{B}_{\rm xc}$, $\mathbf{A}_{\rm xc}^{C1}$, and $\mathbf{A}_{\rm xc}^{C2}$, which are functional derivatives of the xc energy with respect to n, $\mathbf{m}$, $\mathbf{j}_p$, and $\mathbf{j}_m$, respectively,

$$v_{xc}^{C1}[n,\mathbf{m},\mathbf{j}_p] = v_{xc}^{C2}\Big[n,\mathbf{j}_p + \frac{c}{q}\nabla\times m\Big], \quad (12)$$

$$\mathbf{A}_{\rm xc}^{C1}[n,\mathbf{m},\mathbf{j}_p] = \mathbf{A}_{\rm xc}^{C2}\Big[n,\mathbf{j}_p + \frac{c}{q}\nabla\times m\Big], \quad (13)$$

$$\mathbf{B}_{\rm xc}^{C1}[n,\mathbf{m},\mathbf{j}_p] = \nabla\times\mathbf{A}_{\rm xc}^{C2}\Big[n,\mathbf{j}_p + \frac{c}{q}\nabla\times m\Big]. \quad (14)$$

In CDFT the true densities $(n,\mathbf{m},\mathbf{j}_p)$ can all be obtained from the corresponding CDFT KS orbitals. In SDFT, on the other hand, only the densities $(n,\mathbf{m})$ are reproduced by the SDFT KS orbitals, while $\mathbf{j}_p$ is a functional of $(n,\mathbf{m})$:

$$\mathbf{j}_p[n,\mathbf{m}] = \langle\Psi^S[n,\mathbf{m}]\,|\hat{\mathbf{j}}_p|\,\Psi^S[n,\mathbf{m}]\rangle. \quad (15)$$

This quantity will, in general, be different from the current resulting from the SDFT KS orbitals:

$$\begin{aligned} \mathbf{j}_p^{\rm KS}[n,\mathbf{m}] &= \langle\Phi^S[n,\mathbf{m}]\,|\hat{\mathbf{j}}_p|\,\Phi^S[n,\mathbf{m}]\rangle \\ &= \frac{\hbar}{2im}\sum_k^N(\varphi_k^{S*}\nabla\varphi_k^S - \varphi_k^S\nabla\varphi_k^{S*}). \end{aligned} \quad (16)$$

In the following, we specialize to situations where $\mathbf{B} = \mathbf{A} = 0$. In this case, $\mathbf{j}_p^{\rm KS}[n,\mathbf{m}] \equiv 0$ implies $\mathbf{j}_p[n,\mathbf{m}] \equiv 0$ and vice versa. Furthermore, $H^S = H^{C1} = H^{C2}$ and, consequently, $\Psi^S = \Psi^{C1} = \Psi^{C2}$. From this, we readily conclude that the three versions of the functional $F = \langle\Psi|\hat{T} + \hat{W}|\Psi\rangle$ coincide:

$$\begin{aligned} F^S[n,\mathbf{m}] &= F^{C1}[n,\mathbf{m},\mathbf{j}_p[n,\mathbf{m}]] \\ &= F^{C2}\Big[n,\mathbf{j}_m = \mathbf{j}_p[n,\mathbf{m}] + \frac{c}{q}\nabla\times\mathbf{m}\Big], \end{aligned} \quad (17)$$

if $\mathbf{B}[n,\mathbf{m}] = 0$. The functional $\mathbf{B}[n,\mathbf{m}]$ represents the magnetic field that, within SDFT, corresponds to the densities $(n,\mathbf{m})$. We emphasize that even for a vanishing external magnetic field the spin magnetization $\mathbf{m}$ need not be zero. All atoms and molecules with an odd number of electrons, as well as ferromagnetic solids, are examples of this situation. As a consequence, the KS Hamiltonian of SDFT does not reduce to the KS Hamiltonian of ordinary DFT in these cases. Likewise, $\mathbf{j}_{\rm orb}$ need not be zero if $\mathbf{B}[n,\mathbf{m}] = 0$. For example, an atom with a single p electron outside a closed shell, prepared in the $m_l = \pm 1$ state, will have a nonvanishing orbital current.

In the following, we further specialize to situations of vanishing orbital current, i.e., we shall only consider densities that lie in the following set:

$$\mathcal{M} = \{(n,\mathbf{m}) : \mathbf{B}[n,\mathbf{m}] = 0, \quad \mathbf{j}_p[n,\mathbf{m}] = 0\}. \quad (18)$$

Using again the constrained-search characterization of the KS determinants, we readily conclude that

$$\Phi^S[n,\mathbf{m}] = \Phi^{C1}[n,\mathbf{m},0] = \Phi^{C2}\Big[n,\frac{c}{q}\nabla\times\mathbf{m}\Big] \quad (19)$$

for $(n, \mathbf{m}) \in \mathcal{M}$. This immediately yields

$$T_s^S[n, \mathbf{m}] = T_s^{C1}[n, \mathbf{m}, 0] = T_s^{C2}\left[n, \frac{c}{q} \nabla \times \mathbf{m}\right], \quad (20)$$

and, by virtue of (17),

$$E_{\mathrm{xc}}^S[n, \mathbf{m}] = E_{\mathrm{xc}}^{C1}[n, \mathbf{m}, 0] = E_{\mathrm{xc}}^{C2}\left[n, \frac{c}{q} \nabla \times \mathbf{m}\right] \quad (21)$$

for $(n, \mathbf{m}) \in \mathcal{M}$. Taking the functional derivatives of this equation with respect to n and $\mathbf{m}$ on the set $\mathcal{M}$ we obtain

$$v_{\mathrm{xc}}^S[n, \mathbf{m}] = v_{\mathrm{xc}}^{C1}[n, \mathbf{m}, 0] = v_{\mathrm{xc}}^{C2}\left[n, \frac{c}{q} \nabla \times \mathbf{m}\right] \quad (22)$$

and

$$B_{\mathrm{xc}}^S[n, \mathbf{m}] = B_{\mathrm{xc}}^{C1}[n, \mathbf{m}, 0] = \nabla \times \mathbf{A}_{\mathrm{xc}}^{C2}\left[n, \frac{c}{q} \nabla \times \mathbf{m}\right] \quad (23)$$

for $(n, \mathbf{m}) \in \mathcal{M}$. A very important property of E_{xc}^{C1}, first pointed out by Vignale and Rasolt [3], is that gauge invariance forces the functional to depend on the current *only* through the vorticity $\boldsymbol{\nu}_p = \nabla \times (\mathbf{j}_p/n)$, i.e.,

$$E_{\mathrm{xc}}^{C1}[n, \mathbf{m}, \mathbf{j}_p] = \overline{E}_{\mathrm{xc}}^{C1}[n, \mathbf{m}, \boldsymbol{\nu}_p]. \quad (24)$$

This property carries over directly to the $\mathbf{j}_m$ formulation of CDFT, i.e.,

$$E_{\mathrm{xc}}^{C2}[n, \mathbf{j}_m] = \overline{E}_{\mathrm{xc}}^{C2}[n, \boldsymbol{\nu}_m], \quad (25)$$

where $\boldsymbol{\nu}_m$ stands for the vorticity $\boldsymbol{\nu}_m = \nabla \times (\mathbf{j}_m/n)$. Hence we obtain from (21) that

$$E_{\mathrm{xc}}^S[n, \mathbf{m}] = \overline{E}_{\mathrm{xc}}^{C2}\left[n, \frac{c}{q} \nabla \times \frac{\nabla \times \mathbf{m}}{n}\right] \quad (26)$$

for $(n, \mathbf{m}) \in \mathcal{M}$. We observe that for $(n, \mathbf{m}) \in \mathcal{M}$ the xc-energy functional of SDFT depends on the magnetization only through the combination $\nabla \times ((\nabla \times \mathbf{m})/n)$. This fact by itself is an interesting and previously unknown property of E_{xc}^S. It applies to all *finite* [10] systems with vanishing orbital currents and no $\mathbf{B}$ fields. This, in fact, is exactly the situation where SDFT is usually employed.

In the remainder of this Letter we show how (26) can be used as a tool for the construction of approximate functionals for finite systems. First, we assume that we are given an approximate CDFT functional, $\overline{E}_{\mathrm{xc}}^{C2}$. Inserting this approximate functional on the right-hand side, the equality (26) immediately yields an approximation for the SDFT functional $E_{\mathrm{xc}}^S[n, \mathbf{m}]$. If, for example, an LDA-type approximation is used for $\overline{E}_{\mathrm{xc}}^{C2}[n, \boldsymbol{\nu}_m]$, we obtain a new GDA for $E_{\mathrm{xc}}^S[n, \mathbf{m}]$.

Next, we demonstrate that (26) can also be used to obtain CDFT functionals from SDFT. We assume that we are given an approximate SDFT functional of the magnetization on the left-hand side of (26). In general, this will not depend explicitly on $\mathbf{m}$ through $\boldsymbol{\nu}_m$, although the right-hand side of (26) must do so. To construct a CDFT functional from a given SDFT functional via (26), we thus have to find a way to write a functional $Q[\mathbf{m}]$ as a functional $\tilde{Q}[\boldsymbol{\nu}_m|_{\mathbf{j}_p=0}]$, which can then be used on the left-hand side of (26).

To this end we employ the Helmholtz theorem (HT) of vector analysis, which states that a vector field can be decomposed in a part which depends only on the sources and another that depends only on the curl of the field:

$$\mathbf{m}(\mathbf{r}) = \mathbf{m}_0 + \mathbf{f}_1[\nabla \cdot \mathbf{m}(\mathbf{r})] + \mathbf{f}_2[\nabla \times \mathbf{m}(\mathbf{r})]. \quad (27)$$

In the second term we rewrite the argument by dividing and multiplying with the density and then use the HT again, this time for the vector field $\nabla \times \mathbf{m}(\mathbf{r})/n(\mathbf{r})$. This yields

$$\mathbf{m}(\mathbf{r}) = \mathbf{m}_0 + \mathbf{f}_1[\nabla \cdot \mathbf{m}(\mathbf{r})] + \mathbf{f}_2\left[n(\mathbf{r})\left(\mathbf{f}_1\left[\nabla \cdot \frac{\nabla \times \mathbf{m}(\mathbf{r})}{n(\mathbf{r})}\right] + \mathbf{f}_2\left[\nabla \times \frac{\nabla \times \mathbf{m}(\mathbf{r})}{n(\mathbf{r})}\right]\right)\right] \quad (28)$$

with a constant $\mathbf{m}_0$. Inserting this result on the left-hand side of Eq. (26) we obtain another rigorous identity. The latter can be simplified by using the fact that the exact xc functional depends on the magnetization *only* through the vorticity $\boldsymbol{\nu}_m$. Since the vorticity depends only on the curl of $\mathbf{m}(\mathbf{r})$, the constant $\mathbf{m}_0$ drops out and one can add any gradient of a scalar function to the magnetization without changing the functional. The same argument holds for the combination $\nabla \times \mathbf{m}(\mathbf{r})/n(\mathbf{r})$. The vorticity depends on this quantity only through its curl, so that the addition of any gradient of a scalar field to $\nabla \times \mathbf{m}(\mathbf{r})/n(\mathbf{r})$ does not change the functional. Now we use this freedom by adding gradient terms in such a way that the second and third terms in Eq. (28) vanish identically. Explicitly, for the second term this means to substitute $\mathbf{m}(\mathbf{r}) + \nabla\phi^{(1)}$ for $\mathbf{m}(\mathbf{r})$, which does not change the vorticity, and to choose $\phi^{(1)}$ such that $\nabla \cdot \mathbf{m}(\mathbf{r}) + \triangle\phi^{(1)} = 0$. For the third term we add $\nabla\phi^{(2)}$ to $\nabla \times \mathbf{m}(\mathbf{r})/n(\mathbf{r})$ and proceed in the same manner. The corresponding part of the fourth term is not changed by these additions. It can now be identified with the vorticity for $\mathbf{j}_p(\mathbf{r}) = 0$. This leads to

$$\overline{E}_{\mathrm{xc}}^{C2}[n, \boldsymbol{\nu}_m|_{\mathbf{j}_p=0}] = E_{\mathrm{xc}}^S\left[n, \mathbf{f}_2\left[n\mathbf{f}_2\left[\frac{q}{c}\boldsymbol{\nu}_m|_{\mathbf{j}_p=0}\right]\right]\right] \quad (29)$$

for $(n, \mathbf{m}) \in \mathcal{M}$. Equation (29) constitutes an explicit prescription for obtaining $\overline{E}_{\mathrm{xc}}^{C2}[n, \boldsymbol{\nu}_m|_{\mathbf{j}_p=0}]$, given $E_{\mathrm{xc}}^S[n, \mathbf{m}]$.

The prescription (29) is far more general than it appears at first sight. The crucial point is that, although (29) was derived from (26) which was valid only for systems with vanishing paramagnetic current, (29) can also be employed to construct CDFT functionals for current-carrying systems (which constitute the prime application of CDFT by its very nature). This is due to the simple fact that any functional identity $f[x] = g[x]$ implies $f[x + y] = g[x + y]$ provided that $x + y$ lies within the domain of the respective functionals. In our case, this means that the *complete* functional dependence of $\overline{E}_{\mathrm{xc}}^{C2}$ on the vorticity is

determined by (29), even if we drop the requirement $\mathbf{j}_p = 0$. The only change is that $\boldsymbol{\nu}_m|_{\mathbf{j}_p=0} = \frac{c}{q}\nabla \times ((\nabla \times \mathbf{m})/n)$ is replaced by $\boldsymbol{\nu}_m = \boldsymbol{\nu}_m|_{\mathbf{j}_p=0} + \nabla \times (\mathbf{j}_p/n)$. This is an advantage of the $\mathbf{j}_m$ formulation of CDFT as compared to the $\mathbf{j}_p$ formulation: Once the dependence on $\boldsymbol{\nu}_m|_{\mathbf{j}_p=0}$ is known, the dependence on $\boldsymbol{\nu}_m|_{\mathbf{j}_p\neq 0}$, and thus on $\mathbf{j}_p$, is automatically known as well. Thus we finally have

$$\overline{E}_{\mathrm{xc}}^{C2}[n, \boldsymbol{\nu}_m] = E_{\mathrm{xc}}^{S}\left[n, \mathbf{f}_2\left[n\mathbf{f}_2\left[\frac{q}{c}\,\boldsymbol{\nu}_m\right]\right]\right]. \tag{30}$$

Through (30) every approximation for a functional of the magnetization generates a functional of the current density $\mathbf{j}_m$ which can be used in CDFT.

The explicit form of the functionals $\mathbf{f}_1$ and $\mathbf{f}_2$ is [11] $\mathbf{f}_1[a] = \int d^3r' a(\mathbf{r}')\mathbf{G}(\mathbf{r}, \mathbf{r}')$ for scalar functions $a(\mathbf{r})$ and $\mathbf{f}_2[\mathbf{b}] = \int d^3r' \mathbf{b}(\mathbf{r}') \times \mathbf{G}(\mathbf{r}, \mathbf{r}')$ for vectorial functions $\mathbf{b}(\mathbf{r})$, with $\mathbf{G}(\mathbf{r}, \mathbf{r}') = \frac{1}{4\pi}\nabla_{r'}(1/|\mathbf{r} - \mathbf{r}'|)$. These forms are correct whenever the field to be decomposed vanishes faster than $1/r^2$ as r approaches infinity. While this condition is satisfied by $\mathbf{m}$ for all finite systems, it is generally not satisfied by $\nabla \times \mathbf{m}(\mathbf{r})/n(\mathbf{r})$. Various generalizations of $\mathbf{G}(\mathbf{r}, \mathbf{r}')$ for more slowly vanishing or even weakly diverging fields exist, see, e.g., Ref. [12]. Our Eqs. (29) and (30) hold for these cases as well. Instead of using these generalizations, however, one can also explicitly subtract out the asymptotic functional $L[n, \mathbf{m}] = \lim_{r\to\infty} \nabla \times \mathbf{m}(\mathbf{r})/n(\mathbf{r})$ and apply the HT decomposition only to the remainder.

Given approximate CDFT potentials, v_{xc}^{C} and $\mathbf{A}_{\mathrm{xc}}^{C}$ Eqs. (22) and (23) can readily be used to deduce approximate SDFT potentials v_{xc}^{S} and $\mathbf{B}_{\mathrm{xc}}^{S}$. Conversely, applying the HT once more, we can use Eq. (22) to construct an approximate CDFT xc potential from a given SDFT potential as

$$v_{\mathrm{xc}}^{C2}[n, \mathbf{j}_m] = v_{\mathrm{xc}}^{S}\left[n, \mathbf{f}_2\left[\frac{q}{c}\,\mathbf{j}_m\right]\right]. \tag{31}$$

The xc vector potential of CDFT is given by [3]

$$-\frac{q}{c}\,\mathbf{A}_{\mathrm{xc}}^{C2} = \frac{\delta E_{\mathrm{xc}}^{C2}}{\delta \mathbf{j}_m} = -\frac{1}{n}\nabla \times \frac{\delta \overline{E}_{\mathrm{xc}}^{C2}}{\delta \boldsymbol{\nu}_m}. \tag{32}$$

Inserting (30) on the right-hand side of (32), we obtain

$$\mathbf{A}_{\mathrm{xc}}^{C2}(\mathbf{r}) = \mathbf{f}_2[\mathbf{B}_{\mathrm{xc}}^{S}](\mathbf{r}). \tag{33}$$

It is a notorious fact that the construction of approximate CDFT functionals is an extremely demanding task: Even the simplest possible CDFT functional, the exchange-only LDA, is a very complicated functional [6] which is rather hard to implement in practice. In view of the large variety of simple and reliable SDFT functionals, we expect the approximate CDFT functionals resulting from (30), (31), and (33) to be very useful in practical applications. To demonstrate this with an explicit example we employ the local spin density (LSD) approximation on the right-hand side of Eq. (33). Within this approximation, only the z component of $\mathbf{B}_{\mathrm{xc}}^{S}$ has a nonvanishing value, which is given by $B_{\mathrm{xc}}^{\mathrm{LSD}}(\mathbf{r}) = -[v_{\mathrm{xc}\uparrow}^{\mathrm{LSD}}(\mathbf{r}) - v_{\mathrm{xc}\downarrow}^{\mathrm{LSD}}(\mathbf{r})]/2\mu_0$. Here $v_{\mathrm{xc}\uparrow}^{\mathrm{LSD}}$ and $v_{\mathrm{xc}\downarrow}^{\mathrm{LSD}}$ denote the ordinary xc potentials of SDFT within the LSD approximation. Together with the explicit form of the functional $\mathbf{f}_2$ given above, Eq. (33) leads to the simple approximation

$$\mathbf{A}_{\mathrm{xc}}^{C2} = -\int \frac{d^3r'}{8\pi\mu_0}[0, 0, v_{\mathrm{xc}\uparrow}^{\mathrm{LSD}}(\mathbf{r}') - v_{\mathrm{xc}\downarrow}^{\mathrm{LSD}}(\mathbf{r}')] \times \nabla_{r'}\frac{1}{|\mathbf{r} - \mathbf{r}'|} \tag{34}$$

for the vector potential of CDFT[13].

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