Density matrices and density functionals in strong magnetic fields

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(Received 26 August 1997)

The equation of motion for the first-order density matrix (1DM) is constructed for interacting electrons moving under the influence of given external scalar and vector potentials. The 1DM is coupled there to the 2DM by means of the electron-electron interaction. This equation is then employed to obtain the differential virial equation for interacting electrons moving in a magnetic field of arbitrary strength. Suitable integration leads back to the virial theorem derived recently by Erhard and Gross. The exchange-correlation scalar potential of the current-density functional theory of Vignale and Rasolt is derived in two forms, in terms of 1DMs and 2DMs and their noninteracting-system counterparts, involving also (in a linear way) the vector potentials: external and exchange-correlation (xc) ones in the first form, and the xc one in the second form. An equation is obtained also for determining the corresponding xc vector potential in terms of the same DMs and the external vector potential. Approximate exchange-only scalar and vector potentials are proposed in terms of noninteracting 1DM. Finally the Hartree-Fock 1DM for atoms and molecules in magnetic fields is shown to satisfy the same equation of motion as the fully interacting 1DM. [S1050-2947(97)06312-9]

PACS number(s): 31.15.Ew, 31.15.Md, 71.15.Mb

I. INTRODUCTION

There is considerable current interest in the problem of atoms and molecules subjected to intense external magnetic fields (see, e.g., [1,2]). Numerical studies, using both quantum Monte Carlo simulation for the H_2 molecule [3] and the Hartree-Fock approximation for some atoms [4], have been reported very recently. Our purpose in the present work is to provide the basic underlying theory for a density-matrix (DM) approach to the problem of molecules in magnetic fields of arbitrary strength. This will, in essence, provide a generalization of our earlier studies [5–7], made for zero magnetic field, of the exchange-correlation potential $v_{\rm xc}(\mathbf{r})$ of the density-functional theory in terms of first- and second-order density matrices (1DMs, 2DMs). In particular, we give in Sec. II below the equation of motion for 1DM and also the differential virial equation, which both played a central role in our earlier works [5,6], in the presence of a magnetic field of arbitrary strength. However, in contrast to the procedure adopted in [5], it will be valuable here to approach the differential virial equation via the equation of motion for the 1DM. The integral virial theorem in a magnetic field has recently been given by Erhard and Gross [8] and their result will be shown to follow also from our present derivation of the differential virial theorem.

Section III will extend the results of Sec. II to include spin-dependent potentials. Direct contact will be made with the work of Vignale and Rasolt [9] on the current-density functional theory. We will deal specifically with the forms of the exchange-correlation potentials $v_{{\rm xc}\sigma}(\mathbf{r})$ and $\mathbf{A}_{{\rm xc}\sigma}(\mathbf{r})$ in the presence of an applied magnetic field. Approximations for exchange-only potentials will also be proposed. By way of illustration of the exact theory, Sec. IV will derive the Hartree-Fock single-determinantal approximation, but now in the density-matrix form, from the equation of motion of Sec. III. Section V constitutes a summary. The gauge invariance of the obtained equations is discussed in the Appendix.

II. ORBITAL MOTION IN MAGNETIC FIELD IN TERMS OF DENSITY MATRICES

A. The Hamiltonian

When a vector potential $\mathbf{A}$ is imposed on an atom or a molecule, through the application of a strong magnetic field $\mathbf{B}$, related to $\mathbf{A}$ by

$$\mathbf{B}(\mathbf{r})=\boldsymbol{\nabla}\times\mathbf{A}(\mathbf{r}), \tag{2.1}$$

then it is well known that $\mathbf{A}$ is ill defined to the extent of addition of the gradient of any scalar function. The effect of $\mathbf{A}$ is to change the kinetic energy $[1/(2m)]\mathbf{p}^2$ of an electron to $[1/(2m)]\{\mathbf{p}+(e/c)\mathbf{A}\}^2$ (note $e>0$). It is useful to define an operator $\hat{w}$ by

$$\hat{w}(\mathbf{r};[\mathbf{A}])=\frac{1}{2m}\left(\mathbf{p}(\mathbf{r})+\frac{e}{c}\mathbf{A}(\mathbf{r})\right)^2-\frac{1}{2m}\{\mathbf{p}(\mathbf{r})\}^2$$
$$=\frac{e^2}{2mc^2}\mathbf{A}^2(\mathbf{r})-i\frac{\hbar e}{2mc}\{[\boldsymbol{\nabla}\cdot\mathbf{A}(\mathbf{r})]+2\mathbf{A}(\mathbf{r})\cdot\boldsymbol{\nabla}(\mathbf{r})\}, \tag{2.2}$$

using $\mathbf{p}(\mathbf{r})=-i\hbar\boldsymbol{\nabla}(\mathbf{r})=-i\hbar\partial/\partial\mathbf{r}$. In terms of this operator $\hat{w}$ we next construct the one-body Hamiltonian $\hat{h}$ defined by

$$\hat{h}(\mathbf{r};[v,\mathbf{A}])=\hat{t}(\mathbf{r})+\hat{w}(\mathbf{r};[\mathbf{A}])+v(\mathbf{r}), \tag{2.3}$$

where

$$\hat{t}(\mathbf{r})=-\frac{\hbar^2}{2m}\nabla^2(\mathbf{r}), \tag{2.4}$$

and $v(\mathbf{r})$ denotes the external scalar potential. In order to focus on the electron current due to the presence of $\mathbf{A}(\mathbf{r})$, we disregarded in the Hamiltonian (2.3) any dependence on the spin variable, leaving this subject to be taken into account in the remaining sections.

The N-electron nonrelativistic Hamiltonian may then be written

$$\hat{\mathcal{H}}[v,\mathbf{A}]=\sum_{j=1}^{N}\hat{h}(\mathbf{r}_j;[v,\mathbf{A}])+\hat{\mathcal{U}}=\hat{\mathcal{T}}+\hat{\mathcal{W}}+\hat{\mathcal{V}}+\hat{\mathcal{U}}, \quad (2.5)$$

where

$$\hat{\mathcal{U}}=\sum_{1\leqslant i<j\leqslant N}u(\mathbf{r}_i,\mathbf{r}_j), \quad u(\mathbf{r}_i,\mathbf{r}_j)=\frac{e^2}{|\mathbf{r}_i-\mathbf{r}_j|}, \quad (2.6)$$

and

$$\hat{\mathcal{T}}=\sum_{j=1}^{N}\hat{t}(\mathbf{r}_j), \quad \hat{\mathcal{W}}=\sum_{j=1}^{N}\hat{w}(\mathbf{r}_j;[\mathbf{A}]), \quad \hat{\mathcal{V}}=\sum_{j=1}^{N}v(\mathbf{r}_j). \quad (2.7)$$

B. Equation of motion for the first-order density matrix γ_1

With the usual definitions of DMs from the N-electron wave function Ψ_E (see, e.g., Davidson [10], Parr and Yang [11], and also below), the equation of motion (EOM) for γ_1, in terms of the commutator of $\hat{\gamma}_1$ and $\hat{\mathcal{H}}$, takes the form

$$\langle\Psi_E|[\hat{\gamma}_1(\mathbf{1};\mathbf{1}'),\hat{\mathcal{H}}]_-|\Psi_E\rangle=0, \quad (2.8a)$$

where Ψ_E satisfies

$$\hat{\mathcal{H}}|\Psi_E\rangle=E|\Psi_E\rangle, \quad \langle\Psi_E|\Psi_E\rangle=1. \quad (2.8b)$$

In the coordinate representation, the evaluation of Eq. (2.8a) can be performed in a straightforward way as

$$N\int d^4 2\cdots d^4 N[\Psi_E^*(\mathbf{1}'\mathbf{23}\cdots\mathbf{N})\{\hat{\mathcal{H}}\Psi_E(\mathbf{123}\cdots\mathbf{N})\} -\{\hat{\mathcal{H}}\Psi_E(\mathbf{1}'\mathbf{23}\cdots\mathbf{N})\}^*\Psi_E(\mathbf{123}\cdots\mathbf{N})]=0. \quad (2.9)$$

Thus dealing with the spin-independent Hamiltonian (2.5), we arrive at the following EOM for the 1DM γ_1:

$$\{\hat{h}(1;[v,\mathbf{A}])-\hat{h}^*(1';[v,\mathbf{A}])\}\gamma_1(\mathbf{1};\mathbf{1}') +2\int d^4 2\{u(12)-u(1'2)\}\gamma_2(\mathbf{12};\mathbf{1}'\mathbf{2}) =0, \quad (2.10)$$

where

$$\gamma_2(\mathbf{12};\mathbf{1}'\mathbf{2}')=\frac{N(N-1)}{2}\times\int d^4 3\cdots d^4 N\Psi_E(\mathbf{123}\cdots\mathbf{N})\times\Psi_E^*(\mathbf{1}'\mathbf{2}'\mathbf{3}\cdots\mathbf{N}) \quad (2.11)$$

and

$$\gamma_1(\mathbf{1};\mathbf{1}')=\frac{2}{(N-1)}\int d^4 2\,\gamma_2(\mathbf{12};\mathbf{1}'\mathbf{2}) \quad (2.12)$$

are DMs generated from the eigenfunction Ψ_E. Here $\mathbf{1}\equiv\mathbf{x}_1\equiv\{\mathbf{r}_1 s_1\}$ denotes space and spin coordinates of an electron, $\int d^4 2$ means $\Sigma_{s_2}\int d^3 r_2$, and $u(1'2)$ means $u(\mathbf{r}_1',\mathbf{r}_2)$. An analogous EOM in the absence of a magnetic field is already well known, see, e.g., Dawson and March [12] and Ziesche [13], and was used extensively in our previous investigation [6]. The EOM (2.10) plays a quite central role in the present study. In particular, we shall immediately utilize it to derive the differential virial equation for interacting electrons in a molecule subjected to an intense magnetic field (similarly as done in [6] for the case when such a field is absent). The gauge invariance of Eq. (2.10) is discussed in the Appendix.

C. Differential virial equation (DVE)

By acting with the operator $\frac{1}{2}[\boldsymbol{\nabla}(1)-\boldsymbol{\nabla}(1')]$ on the EOM (2.10), setting thereafter $\mathbf{1}'=\mathbf{1}$, and taking a sum over s_1 and s_2, we can rewrite the resulting DVE in terms of spinless matrices defined by

$$\rho_1(\mathbf{r}_1;\mathbf{r}_1')=\sum_{s_1}\gamma_1(\mathbf{r}_1 s_1;\mathbf{r}_1' s_1), \quad n(1)\equiv n_1(1)=\rho_1(1;1), \quad (2.13)$$

and

$$n_2(\mathbf{r}_1,\mathbf{r}_2)=\sum_{s_1,s_2}\gamma_2(\mathbf{r}_1 s_1,\mathbf{r}_2 s_2;\mathbf{r}_1 s_1,\mathbf{r}_2 s_2). \quad (2.14)$$

The ''diagonals,'' the particle density $n(1)\equiv n_1(1)$ and the pair density $n_2(12)$, are crucial ingredients in expressing the DVE in a form useful for our present purposes:

$$\mathbf{z}(1;[\rho_1])-\frac{\hbar^2}{4m}\boldsymbol{\nabla}\nabla^2 n(1) +n(1)\boldsymbol{\nabla}\left(v(1)+\frac{e^2}{2mc^2}A^2(1)\right)+\mathbf{k}(1;[\mathbf{j}_{\mathrm{p}}[\rho_1],\mathbf{A}]) +2\int d^3 2\{\boldsymbol{\nabla}(1)u(12)\}n_2(12)=0, \quad (2.15)$$

where

$$z_\alpha(1;[\rho_1])=2\sum_\beta\nabla_\beta(1)t_{\alpha\beta}(1;[\rho_1]), \quad (2.16)$$

in terms of the kinetic-energy-density tensor

$$t_{\alpha\beta}(1;[\rho_1])=\frac{\hbar^2}{4m}\{\nabla_\alpha(1')\nabla_\beta(1'') +\nabla_\beta(1')\nabla_\alpha(1'')\}\rho_1(1+1';1+1'')|_{1'=1''=0}, \quad (2.17)$$

and

$$k_\alpha(1;[\mathbf{j}_\mathrm{p},\mathbf{A}])=\sum_\beta \frac{e}{c}[\{\nabla_\alpha(1)A_\beta(1)\}j_{\mathrm{p}\beta}(1) + \nabla_\beta(1)\{A_\beta(1)j_{\mathrm{p}\alpha}(1)\}], \tag{2.18}$$

in terms of the paramagnetic-current-density vector $\mathbf{j}_p$, defined also as a functional of ρ_1 by

$$\begin{aligned}\mathbf{j}_\mathrm{p}(1;[\rho_1]) &= \frac{1}{2m}\{\mathbf{p}(1')+\mathbf{p}^*(1'')\}\rho_1(1+1';1+1'')\big|_{1'=1''=0} \\ &= -\frac{i\hbar}{2m}\{\boldsymbol{\nabla}(1')-\boldsymbol{\nabla}(1'')\}\rho_1(1+1';1+1'')\big|_{1'=1''=0} \\ &= \frac{\hbar}{m}\boldsymbol{\nabla}(1')\mathrm{Im}\rho_1(1+1';1)\big|_{1'=0}.\end{aligned} \tag{2.19}$$

Note that all terms in Eq. (2.15) and also $t_{\alpha\beta}$ and $\mathbf{j}_\mathrm{p}$, Eqs. (2.17) and (2.19), are real-value quantities.

The above DVE (2.15) leads immediately to the integral virial equation (VE). One applies the operation $\int d^3r_1\mathbf{r}_1\cdot$ to the DVE to obtain

$$2T[\rho_1]+E_{\mathrm{ee}}[n_2]=\int d^3r n(\mathbf{r})\mathbf{r}\cdot\boldsymbol{\nabla}\left(v(\mathbf{r})+\frac{e^2}{2mc^2}A^2(\mathbf{r})\right) + \int d^3r\mathbf{j}_\mathrm{p}(\mathbf{r};[\rho_1])\{\mathbf{r}\cdot\boldsymbol{\nabla}-1\}\frac{e}{c}\mathbf{A}(\mathbf{r}), \tag{2.20}$$

where the global kinetic energy T is given explicitly by

$$\begin{aligned}T[\rho_1] &= \int d^3r_1\hat{t}(\mathbf{r}_1)\rho_1(\mathbf{r}_1;\mathbf{r}_1')\big|_{\mathbf{r}_1'=\mathbf{r}_1} \\ &= \int d^3r\sum_\alpha t_{\alpha\alpha}(\mathbf{r};[\rho_1]),\end{aligned} \tag{2.21}$$

while the electron-electron interaction energy E_{ee} is

$$E_{\mathrm{ee}}[n_2]=\int d^31d^32u(12)n_2(12), \tag{2.22}$$

the Coulombic form (2.6) of $u(12)$ being used to obtain the VE (2.20). The above VE (2.20) is precisely Eq. (36) of Erhard and Gross [8], obtained by them in a completely different way.

The VE (2.20) is an example belonging to a vast family of so-called hypervirial equations, which can be easily generated by applying prefactors, different than $\mathbf{r}_1$, prior to the integration of the DVE (2.15). In this way generalizations may be obtained, for interacting many-electron systems in a magnetic field, of our previous results [14] on hypervirial equations, concerning noninteracting systems in the absence of such a field.

D. Interpretation of DVE as a force-balance equation

We define the external force $\mathbf{f}(\mathbf{r})$ as minus the gradient of the external potential $v(\mathbf{r})$. Then we readily obtain from Eq. (2.15) the result

$$\begin{aligned}\mathbf{f}(\mathbf{r}) &= n^{-1}(\mathbf{r})\left(\mathbf{z}(\mathbf{r};[\rho_1])-\frac{\hbar^2}{4m}\boldsymbol{\nabla}\nabla^2 n(\mathbf{r}) + 2\int d^3r'[\boldsymbol{\nabla}(\mathbf{r})u(\mathbf{r},\mathbf{r}')]n_2(\mathbf{r},\mathbf{r}')\right) \\ &\quad + \left(n^{-1}(\mathbf{r})\mathbf{k}(\mathbf{r};[\mathbf{j}_\mathrm{p}[\rho_1],\mathbf{A}])+\frac{e^2}{2mc^2}\boldsymbol{\nabla}[A^2(\mathbf{r})]\right) \\ &= \mathbf{f}^{\mathrm{nmg}}(\mathbf{r})+\mathbf{f}^{\mathrm{mag}}(\mathbf{r}).\end{aligned} \tag{2.23}$$

In making the separation of $\mathbf{f}(\mathbf{r})$ shown in Eq. (2.23) into the sum of two parts, what is to be emphasized is that the magnetic part $\mathbf{f}^{\mathrm{mag}}(\mathbf{r})$ vanishes as the magnetic vector potential $\mathbf{A}$ is switched off. While the nonmagnetic part $\mathbf{f}^{\mathrm{nmg}}(\mathbf{r})$ depends on the magnetic field through the fact that the DMs correspond to the eigenfunction calculated in the presence of the vector potential $\mathbf{A}$, it has a nonzero limit as $\mathbf{A}\to\mathbf{0}$, when our earlier result [5] in zero magnetic field is of course recovered.

As to the physical significance of $\mathbf{f}^{\mathrm{mag}}(\mathbf{r})$, it can again be decomposed into the sum of two terms, the first being the well-known Lorentz force of electromagnetism, namely [see Eq. (2.1)],

$$\begin{aligned}f_\alpha^{\mathrm{Lor}}(\mathbf{r}) &= n^{-1}(\mathbf{r})\left(\mathbf{j}(\mathbf{r})\times\frac{e}{c}\mathbf{B}(\mathbf{r})\right)_\alpha \\ &= n^{-1}(\mathbf{r})\frac{e}{c}\sum_\beta\{j_\beta(\mathbf{r})\nabla_\alpha A_\beta(\mathbf{r})-j_\beta(\mathbf{r})\nabla_\beta A_\alpha(\mathbf{r})\},\end{aligned} \tag{2.24}$$

expressed in terms of the gauge-invariant *physical* current $\mathbf{j}(\mathbf{r})$, defined as a sum of the *paramagnetic* current $\mathbf{j}_\mathrm{p}(\mathbf{r})$ and a term due to the vector potential $\mathbf{A}(\mathbf{r})$,

$$\mathbf{j}(\mathbf{r})=\mathbf{j}_\mathrm{p}(\mathbf{r})+\frac{e}{mc}n(\mathbf{r})\mathbf{A}(\mathbf{r}). \tag{2.25}$$

The continuity equation for the stationary $n(\mathbf{r})$, i.e., satisfying $\partial n(\mathbf{r})/\partial t=0$ (as corresponding to an eigenfunction), reads

$$\boldsymbol{\nabla}\cdot\mathbf{j}(\mathbf{r})=0. \tag{2.26}$$

Writing then

$$\mathbf{f}^{\mathrm{mag}}(\mathbf{r})=\mathbf{f}^{\mathrm{Lor}}(\mathbf{r})+\mathbf{f}^{\mathrm{inh}}(\mathbf{r}), \tag{2.27}$$

and invoking Eq. (2.26), the second force is obtained in the form

$$f_\alpha^{\mathrm{inh}}(\mathbf{r})=n^{-1}(\mathbf{r})\sum_\beta\nabla_\beta D_{\alpha\beta}(\mathbf{r}), \tag{2.28}$$

where the tensor $D_{\alpha\beta}=D_{\beta\alpha}$ is defined by

$$D_{\alpha\beta}(\mathbf{r}) = j_\alpha(\mathbf{r})\frac{e}{c}A_\beta(\mathbf{r}) + j_\beta(\mathbf{r})\frac{e}{c}A_\alpha(\mathbf{r}) - \frac{e^2}{mc^2}n(\mathbf{r})A_\alpha(\mathbf{r})A_\beta(\mathbf{r}) = j_{\mathrm{p}\alpha}(\mathbf{r})\frac{e}{c}A_\beta(\mathbf{r}) + j_{\mathrm{p}\beta}(\mathbf{r})\frac{e}{c}A_\alpha(\mathbf{r}) + \frac{e^2}{mc^2}n(\mathbf{r})A_\alpha(\mathbf{r})A_\beta(\mathbf{r}). \tag{2.29}$$

So f_α^{inh} arises from inhomogeneity in the tensor $D_{\alpha\beta}(\mathbf{r})$ — a combination of the current density, particle density, and the vector potential. It is worth noting that the structure of the force (2.28) is similar to the structure of the contribution in Eq. (2.23) due to inhomogeneity of the kinetic-energy-density tensor $t_{\alpha\beta}=t_{\beta\alpha}$:

$$n^{-1}(\mathbf{r})\mathbf{z}(\mathbf{r}) = 2n^{-1}(\mathbf{r})\sum_\beta \nabla_\beta t_{\alpha\beta}(\mathbf{r}). \tag{2.30}$$

See the Appendix for comments concerning the gauge invariance of various force terms.

The above force-balance equation (2.23) is to be compared with the one-electron result of Amovilli and March [15], namely,

$$-\nabla v = \nabla\left(\frac{\hbar^2}{8m}n^{-2}(\nabla n)^2 - \frac{\hbar^2}{4m}n^{-1}\nabla^2 n + \frac{m}{2}n^{-2}j^2\right). \tag{2.31}$$

Equation (2.31) is readily obtained from the general result (2.23) by setting $n_2=0$ and $\rho_1(\mathbf{r};\mathbf{r}')=\phi(\mathbf{r})\phi^*(\mathbf{r}')$, with $\phi(\mathbf{r})$ now the one-electron wave function. Again in Eq. (2.31) the continuity equation (2.26) has been invoked.

Before relating the above more directly to the current-density functional theory as set out by Vignale and Rasolt [9], it is important to turn next to the case when the electron spin is included.

III. INCLUSION OF ELECTRON SPIN AND DIRECT CONTACT WITH CURRENT-DENSITY-FUNCTIONAL THEORY

A. Equation of motion and differential virial equation

In this and the remaining sections we shall restrict our considerations to the case when the external magnetic field $\mathbf{B}(\mathbf{r})$ has a constant direction. Choosing the z axis as the field direction, then $\mathbf{B}(\mathbf{r})$ is defined by its magnitude and the unit vector $\hat{\mathbf{z}}$:

$$\mathbf{B}(\mathbf{r}) = B(\mathbf{r})\hat{\mathbf{z}} = |\nabla\times\mathbf{A}(\mathbf{r})|\hat{\mathbf{z}}. \tag{3.1}$$

Now one must extend the one-body Hamiltonian by adding a term $[\hbar e/(mc)]B(\mathbf{r})\hat{s}_z(\sigma)$ [where $\hat{s}_z(\sigma)$ is a spin operator with the eigenvalues $\pm\frac{1}{2}$]. Having in mind the current-density functional application (see Vignale and Rasolt [9]) of our result, we admit an even more general one-body (magnetic) Hamiltonian, allowing it to be σ dependent [see Eq. (2.3)]:

$$\hat{h}_\sigma(\mathbf{r}) = \hat{h}(\mathbf{r};[v_\sigma,\mathbf{A}_\sigma]) = \hat{h}_{\mathrm{m}}(\mathbf{r}\sigma) \equiv \hat{h}_{\mathrm{m}}(\mathbf{x}), \tag{3.2}$$

where both scalar and vector potentials are σ dependent and arbitrary. This Hamiltonian acts on a function $\varphi(\mathbf{x})$ as a local or differential operator with respect to spatial coordinate, and as a diagonal matrix, having diagonal elements $\hat{h}_{\mathrm{m}}(\mathbf{r}\uparrow)$ and $\hat{h}_{\mathrm{m}}(\mathbf{r}\downarrow)$, with respect to spin coordinate, so $(\hat{h}_{\mathrm{m}}\varphi)(\mathbf{r}\sigma)=\hat{h}_{\mathrm{m}}(\mathbf{r}\sigma)\varphi(\mathbf{r}\sigma)$, without any summation over the spin coordinate σ. In application to real systems, one imposes $v_\uparrow(\mathbf{r})+v_\downarrow(\mathbf{r})=2v(\mathbf{r})$, where $v(\mathbf{r})$ is the physical external scalar potential, $v_\uparrow(\mathbf{r})-v_\downarrow(\mathbf{r})=2[\hbar e/(2mc)]B(\mathbf{r})$, where $B(\mathbf{r})$ is the physical external magnetic field, and sets $\mathbf{A}_\uparrow(\mathbf{r})=\mathbf{A}_\downarrow(\mathbf{r})=\mathbf{A}(\mathbf{r})=$ the physical external vector potential.

Using the general $\hat{h}_{\mathrm{m}}(\mathbf{x})$, Eq. (3.2), we have for the interacting system the magnetic Hamiltonian [compare Eq. (2.5)]

$$\hat{\mathcal{H}}_{\mathrm{m}} = \sum_{j=1}^{N}\hat{h}_{\mathrm{m}}(\mathbf{x}_j) + \hat{\mathcal{U}} = \hat{\mathcal{T}} + \hat{\mathcal{W}}_{\mathrm{m}} + \hat{\mathcal{V}}_{\mathrm{m}} + \hat{\mathcal{U}}. \tag{3.3}$$

Evaluating Eq. (2.9) with $\hat{\mathcal{H}}$ replaced by $\hat{\mathcal{H}}_{\mathrm{m}}$, we arrive at the EOM for γ_1, similar to Eq. (2.10):

$$\{\hat{h}_{\mathrm{m}}(\mathbf{1}) - \hat{h}_{\mathrm{m}}^*(\mathbf{1}')\}\gamma_1(\mathbf{1};\mathbf{1}') + 2\int d^4 2\{u(12)-u(1'2)\}\gamma_2(\mathbf{12};\mathbf{1'2}) = 0. \tag{3.4a}$$

Since the Hamiltonian $\hat{\mathcal{H}}_{\mathrm{m}}$, Eq. (3.3), commutes with the operator of the total-spin z component, the DM elements $\gamma_1(\mathbf{r}_1\uparrow;\mathbf{r}_1'\downarrow)$ and $\gamma_1(\mathbf{r}_1\downarrow;\mathbf{r}_1'\uparrow)$ vanish (see Davidson [10]). Therefore the EOMs are obtained from Eq. (3.4a) for the remaining two elements of the 1DM, for $\sigma=\uparrow,\downarrow$:

$$\{\hat{h}(1;[v_\sigma,\mathbf{A}_\sigma]) - \hat{h}^*(1';[v_\sigma,\mathbf{A}_\sigma])\}\rho_{1\sigma}(1;1') + 2\int d^3 2\{u(12)-u(1'2)\}\rho_{2\sigma}(12;1'2) = 0, \tag{3.4b}$$

where

$$\rho_{1\sigma}(\mathbf{r}_1;\mathbf{r}_1') = \gamma_1(\mathbf{r}_1\sigma;\mathbf{r}_1'\sigma) \tag{3.5a}$$

and

$$\rho_{2\sigma}(\mathbf{r}_1,\mathbf{r}_2;\mathbf{r}_1',\mathbf{r}_2') = \sum_{s_2}\gamma_2(\mathbf{r}_1\sigma,\mathbf{r}_2 s_2;\mathbf{r}_1'\sigma,\mathbf{r}_2' s_2). \tag{3.5b}$$

There are then two DVEs, namely

$$\mathbf{z}(1;[\rho_{1\sigma}]) - \frac{\hbar^2}{4m}\nabla\nabla^2 n_\sigma(1) + n_\sigma(1)\nabla\left(v_\sigma(1) + \frac{e^2}{2mc^2}A_\sigma^2(1)\right) + \mathbf{k}(1;[\mathbf{j}_{\mathrm{p}}[\rho_{1\sigma}],\mathbf{A}_\sigma]) + 2\int d^3 2\{\nabla(1)u(12)\}n_{2\sigma}(12) = 0, \tag{3.6}$$

where

$$n_\sigma(1)\equiv n_{1\sigma}(1)=\rho_{1\sigma}(1;1) \tag{3.7a}$$

and

$$n_{2\sigma}(12)=\rho_{2\sigma}(12;12). \tag{3.7b}$$

One can, as a consequence, write two force-balance equations and two integral virial equations.

Let us investigate the EOM (3.4b) on the diagonal. By performing the limit $1'\to 1$ we transform this equation to the form

$$-i\hbar\boldsymbol{\nabla}\left(\mathbf{j}_{\mathrm{p}}(1;[\rho_{1\sigma}])+\frac{\hbar e}{mc}n_\sigma(1)\mathbf{A}_\sigma(1)\right)=0. \tag{3.8a}$$

All real terms involving $\rho_{1\sigma}$ and the whole term involving $\rho_{2\sigma}$ vanished, because of Hermicity of DMs. The obtained Eq. (3.8a) is, in fact, the static continuity equation, because the σ component of the physical current density is known to be

$$\mathbf{j}_\sigma(1)=\mathbf{j}_{\mathrm{p}}(1;[\rho_{1\sigma}])+\frac{\hbar e}{mc}n_\sigma(1)\mathbf{A}_\sigma(1) \tag{3.8b}$$

[compare Eqs. (2.25) and (2.26) in the spinless form]. In the absence of a magnetic field, this diagonal limit of the EOM as the continuity equation was pointed out by Ziesche [13].

B. Application to current-density-functional theory of Vignale and Rasolt

A reference *noninteracting* system is introduced (its DMs are marked out with the superscript 's'), such that the particle-number spin densities and the paramagnetic-current spin densities are the same as in the original interacting system:

$$n^{\mathrm{s}}_\sigma(\mathbf{r})\equiv\rho^{\mathrm{s}}_{1\sigma}(\mathbf{r};\mathbf{r})=n_\sigma(\mathbf{r})\equiv\rho_{1\sigma}(\mathbf{r};\mathbf{r}), \tag{3.9a}$$

$$\mathbf{j}^{\mathrm{s}}_{\mathrm{p}\sigma}(\mathbf{r})\equiv\mathbf{j}_{\mathrm{p}}(\mathbf{r};[\rho^{\mathrm{s}}_{1\sigma}])=\mathbf{j}_{\mathrm{p}\sigma}(\mathbf{r})\equiv\mathbf{j}_{\mathrm{p}}(\mathbf{r};[\rho_{1\sigma}]) \tag{3.9b}$$

[see Eq. (2.19)]. The particles now move in effective potential fields: $v^{\mathrm{eff}}_\sigma(\mathbf{r})$, $\mathbf{A}^{\mathrm{eff}}_\sigma(\mathbf{r})$. The corresponding one-body Hamiltonian is therefore [compare Eq. (3.2)]

$$\hat{h}^{\mathrm{eff}}_\sigma(1)=\hat{h}(1;[v^{\mathrm{eff}}_\sigma,\mathbf{A}^{\mathrm{eff}}_\sigma]). \tag{3.10}$$

The EOM for the reference system [Eq. (3.4b) with $u=0$] is

$$\{\hat{h}^{\mathrm{eff}}_\sigma(1)-\hat{h}^{\mathrm{eff}*}_\sigma(1')\}\rho^{\mathrm{s}}_{1\sigma}(1;1')=0, \tag{3.11}$$

where, assuming a nondegenerate ground state and, therefore, a single-determinantal wave function, the 1DMs are given by

$$\rho^{\mathrm{s}}_{1\sigma}(1;1')=\sum_{a=1}^{N_\sigma}\phi_{a\sigma}(1)\phi^*_{a\sigma}(1'), \tag{3.12}$$

with $\phi_{a\sigma}$, satisfying the Kohn-Sham equations

$$\hat{h}^{\mathrm{eff}}_\sigma(1)\phi_{a\sigma}(1)=\epsilon_{a\sigma}\phi_{a\sigma}(1),\quad \epsilon_{a\sigma}\leqslant\epsilon_{a+1,\sigma} \tag{3.13a}$$

and the number N_σ, known from the interacting system to be

$$N_\sigma=\int d^31\, n_\sigma(1),\quad N_\uparrow+N_\downarrow=N. \tag{3.13b}$$

The physical current in the reference system [compare Eq. (3.8b)]

$$\mathbf{j}^{\mathrm{s}}_\sigma(1)=\mathbf{j}_{\mathrm{p}\sigma}(1)+\frac{\hbar e}{mc}n_\sigma(1)\mathbf{A}^{\mathrm{eff}}_\sigma(1) \tag{3.14}$$

satisfies the continuity equation $\boldsymbol{\nabla}\cdot\mathbf{j}^{\mathrm{s}}_\sigma=0$.

The DVE in the reference system reads

$$\mathbf{z}(1;[\rho^{\mathrm{s}}_{1\sigma}])-\frac{\hbar^2}{4m}\boldsymbol{\nabla}\nabla^2 n_\sigma(1)+\mathbf{k}(1;[\mathbf{j}_{\mathrm{p}\sigma},\mathbf{A}^{\mathrm{eff}}_\sigma])+n_\sigma(1)\boldsymbol{\nabla}\left(v^{\mathrm{eff}}_\sigma(1)+\frac{e^2}{2mc^2}[\mathbf{A}^{\mathrm{eff}}_\sigma(1)]^2\right)=0. \tag{3.15}$$

C. Exchange-correlation scalar potential

Let us partition the one-body Hamiltonian, Eq. (2.3) with Eq. (2.2), into its differential-operator (dop) and local (loc) terms:

$$\hat{h}(\mathbf{r};[v,\mathbf{A}])=\hat{h}_{\mathrm{dop}}(\mathbf{r};[\mathbf{A}])+h_{\mathrm{loc}}(\mathbf{r};[v,\mathbf{A}]), \tag{3.16a}$$

$$\hat{h}_{\mathrm{dop}}(\mathbf{r};[\mathbf{A}])=\hat{t}(\mathbf{r})-i\frac{\hbar e}{mc}\mathbf{A}(\mathbf{r})\boldsymbol{\nabla}(\mathbf{r}), \tag{3.16b}$$

$$h_{\mathrm{loc}}(\mathbf{r};[v,\mathbf{A}])=v(\mathbf{r})+\frac{e^2}{2mc^2}\mathbf{A}^2(\mathbf{r})-i\frac{\hbar e}{2mc}[\boldsymbol{\nabla}\cdot\mathbf{A}(\mathbf{r})]. \tag{3.16c}$$

By dividing the EOM (3.4b) of the interacting system by $\rho_{1\sigma}(1;1')$ and that, Eq. (3.11), of the reference system by $\rho^{\mathrm{s}}_{1\sigma}(1;1')$, we can separate out the local terms

$$h_{\mathrm{loc}}(1;[v_\sigma,\mathbf{A}])-h^*_{\mathrm{loc}}(1';[v_\sigma,\mathbf{A}])=-W_{\mathrm{dop}}(11';[\mathbf{A},\rho_{1\sigma}])-W_{\mathrm{ee}}(11';[u,\rho_{1\sigma},\rho_{2\sigma}]), \tag{3.17a}$$

where

$$W_{\mathrm{dop}}(11';[\mathbf{A},\rho_1])=\frac{\{\hat{h}_{\mathrm{dop}}(1;[\mathbf{A}])-\hat{h}^*_{\mathrm{dop}}(1';[\mathbf{A}])\}\rho_1(1;1')}{\rho_1(1;1')}, \tag{3.17b}$$

$$W_{\mathrm{ee}}(11';[u,\rho_1,\rho_2])=\frac{2\int d^32\{u(12)-u(1'2)\}\rho_2(12;1'2)}{\rho_1(1;1')}, \tag{3.17c}$$

and

$$h_{\rm loc}(1;[v_\sigma^{\rm eff},\mathbf{A}_\sigma^{\rm eff}])-h_{\rm loc}^*(1';[v_\sigma^{\rm eff},\mathbf{A}_\sigma^{\rm eff}]) = -W_{\rm dop}(11';[\mathbf{A}_\sigma^{\rm eff},\rho_{1\sigma}^{\rm s}]). \quad (3.18)$$

The replacement $\mathbf{A}_\sigma \to \mathbf{A}$ was performed in Eq. (3.17a) in order to have results pertaining to the real system.

This Eq. (3.17a) can be immediately rearranged to the form $v_\sigma(\mathbf{r}) = v_\sigma(\mathbf{r}') + \{\text{terms depending on } \rho_{1\sigma},\ \rho_{2\sigma},\ u,\ \mathbf{A}\}$. It demonstrates that the external scalar potential can be reconstructed from the known DMs $\rho_{1\sigma},\rho_{2\sigma}$, the interaction u, and the external vector potential $\mathbf{A}$. For that reason one must choose and fix some reference point $\mathbf{r}'$ and an arbitrary constant $v_\sigma(\mathbf{r}')$. This last freedom reflects the fact that scalar potentials which differ by a constant are equivalent. A similar statement concerns Eq. (3.18).

As shown by Vignale and Rasolt [9], the effective potentials can be written in the forms

$$\mathbf{A}_\sigma^{\rm eff}(1) = \mathbf{A}(1) + \mathbf{A}_{{\rm xc}\sigma}(1), \quad (3.19)$$

$$v_\sigma^{\rm eff}(1) = v_\sigma(1) + v_{\rm es}(1) + v_{{\rm xc}\sigma}(1) + \frac{e^2}{2mc^2}\{[\mathbf{A}(1)]^2 - [\mathbf{A}_\sigma^{\rm eff}(1)]^2\}, \quad (3.20)$$

with

$$v_{\rm es}(1) = v_{\rm es}(1;[n]) = \int d^3 2\, u(12) n(2), \quad (3.21)$$

$$n(2) = n_\uparrow(2) + n_\downarrow(2), \quad (3.22)$$

$$v_{{\rm xc}\sigma}(1;[n_\uparrow,n_\downarrow,\mathbf{j}_{{\rm p}\uparrow},\mathbf{j}_{{\rm p}\downarrow}]) = \frac{\tilde{\delta} E_{\rm xc}[n_\uparrow,n_\downarrow,\mathbf{j}_{{\rm p}\uparrow},\mathbf{j}_{{\rm p}\downarrow}]}{\tilde{\delta} n_\sigma(1)}, \quad (3.23)$$

$$\frac{e}{c}\mathbf{A}_{{\rm xc}\sigma}(1;[n_\uparrow,n_\downarrow,\mathbf{j}_{{\rm p}\uparrow},\mathbf{j}_{{\rm p}\downarrow}]) = \frac{\tilde{\delta} E_{\rm xc}[n_\uparrow,n_\downarrow,\mathbf{j}_{{\rm p}\uparrow},\mathbf{j}_{{\rm p}\downarrow}]}{\tilde{\delta} \mathbf{j}_{p\sigma}(1)}, \quad (3.24)$$

$E_{\rm xc}$ being the exchange-correlation energy, see [9]. Here $\tilde{\delta}$ indicates the *partial* functional differentiation.

After inserting the potentials $v_\sigma^{\rm eff}$ and $\mathbf{A}_\sigma^{\rm eff}$, Eqs. (3.20), (3.19), into Eq. (3.18), and then subtracting from it Eq. (3.17a), one obtains from the real part of the result an expression for the scalar exchange-correlation potentials' difference

$$v_{{\rm xc}\sigma}(1) - v_{{\rm xc}\sigma}(1') = {\rm Re}\{W_{\rm dop}(11';[\mathbf{A},\rho_{1\sigma}]) - W_{\rm dop}(11';[\mathbf{A}+\mathbf{A}_{{\rm xc}\sigma},\rho_{1\sigma}^{\rm s}]) + W_{\rm ee}(11';[u,\rho_{1\sigma},\rho_{2\sigma}]) - v_{\rm es}(1;[n]) + v_{\rm es}(1';[n])\}, \quad (3.25)$$

and from the imaginary part — an expression for the sum of the corresponding vector potential divergences

$$-\frac{\hbar e}{2mc}\{\nabla\cdot\mathbf{A}_{{\rm xc}\sigma}(1) + \nabla\cdot\mathbf{A}_{{\rm xc}\sigma}(1')\} = {\rm Im}\{W_{\rm dop}(11';[\mathbf{A},\rho_{1\sigma}]) - W_{\rm dop}(11';[\mathbf{A}+\mathbf{A}_{{\rm xc}_\sigma},\rho_{1\sigma}^{\rm s}]) + W_{\rm ee}(11';[u,\rho_{1\sigma},\rho_{2\sigma}])\}. \quad (3.26)$$

The structure of $\hat{h}_{\rm dop}$, Eq. (3.16b), induces the following splitting of $W_{\rm dop}$, Eq. (3.17b):

$$W_{\rm dop}(11';[\mathbf{A},\rho_1]) = W_{\rm kin}(11';[\rho_1]) - i\frac{\hbar e}{mc}\{\mathbf{A}(1)\mathbf{W}_{\rm gr}(11';[\rho_1]) + \mathbf{A}(1')\mathbf{W}_{\rm gr}^*(1'1;[\rho_1])\}, \quad (3.27a)$$

where the kinetic energy operator contribution is

$$W_{\rm kin}(11';[\rho_1]) = \frac{\{\hat{t}(1)-\hat{t}(1')\}\rho_1(1;1')}{\rho_1(1;1')}, \quad (3.27b)$$

and the reduced gradient of 1DM is

$$\mathbf{W}_{\rm gr}(11';[\rho_1]) = \frac{\nabla(1)\rho_1(1;1')}{\rho_1(1;1')}. \quad (3.27c)$$

So the final expression for $v_{{\rm xc}\sigma}$ is

$$v_{{\rm xc}\sigma}(1) = v_{{\rm xc}\sigma}(1') + {\rm Re}\{W_{\rm kin}(11';[\rho_{1\sigma}]) - W_{\rm kin}(11';[\rho_{1\sigma}^{\rm s}]) + W_{\rm ee}(11';[u,\rho_{1\sigma},\rho_{2\sigma}]) - v_{\rm es}(1;[n]) + v_{\rm es}(1';[n])\} + \frac{\hbar e}{mc}{\rm Im}\{(\mathbf{A}(1)\{\mathbf{W}_{\rm gr}(11';[\rho_{1\sigma}]) - \mathbf{W}_{\rm gr}(11';[\rho_{1\sigma}^{\rm s}])\} - \mathbf{A}_{{\rm xc}\sigma}(1)\mathbf{W}_{\rm gr}(11';[\rho_{1\sigma}^{\rm s}]))+(1 \overset{\rightarrow}{\leftarrow} 1')^*\}. \quad (3.28)$$

As seen from the above, the term preceded by Im vanishes with vanishing $\mathbf{A}$ and $\mathbf{A}_{{\rm xc}\sigma}$, while that preceded by Re survives. In this way our earlier result [6] in the absence of a magnetic field is recovered.

Equation (3.28) demonstrates that the scalar exchange-correlation potential can be reconstructed from the known DMs $\rho_{1\sigma}$, $\rho_{2\sigma}$ of the interacting system, and $\rho_{1\sigma}^{\rm s}$ of the reference system, the interaction u, and the vector potentials: external $\mathbf{A}$ and exchange-correlation $\mathbf{A}_{{\rm xc}\sigma}$. The reference point $1'$, chosen arbitrarily, is kept fixed. The presence of an arbitrary constant $v_{{\rm xc}\sigma}(1')$ reflects the freedom of the gauge transformation for the static scalar potential $v_\sigma^{\rm eff}$ of the reference system: DMs obtained with this potential and with the transformed potential (i.e., shifted by a constant), are the same, therefore that constant cannot be determined from the knowledge of DMs.

The difference of the DVEs (3.15) and (3.6) yields

$$\mathbf{z}(1;[\rho^{\mathrm{s}}_{1\sigma}-\rho_{1\sigma}])+\mathbf{k}(1;[\mathbf{j}_{\mathrm{p}\sigma},\mathbf{A}^{\mathrm{eff}}_{\sigma}-\mathbf{A}_{\sigma}]) + n_{\sigma}(1)\boldsymbol{\nabla}\Big(v^{\mathrm{eff}}_{\sigma}(1)-v_{\sigma}(1) + \frac{e^2}{2mc^2}\{[\mathbf{A}^{\mathrm{eff}}_{\sigma}(1)]^2-\mathbf{A}^2_{\sigma}(1)\}\Big) - 2\int d^3 2\{\boldsymbol{\nabla}(1)u(12)\}n_{2\sigma}(12)=0. \tag{3.29}$$

It is essential that $n_{\sigma}(1)$ and $\mathbf{j}_{\mathrm{p}\sigma}(1)$ are the same for the reference and the original systems. After inserting the Vignale and Rasolt effective potentials, Eqs. (3.19) and (3.20), we obtain from Eq. (3.29)

$$\begin{aligned}&\mathbf{f}_{\mathrm{xc}\sigma}(1)\\ &\quad=-\boldsymbol{\nabla}(1)v_{\mathrm{xc}\sigma}(1)\\ &\quad=n^{-1}_{\sigma}(1)\{\mathbf{z}(1;[\rho^{\mathrm{s}}_{1\sigma}-\rho_{1\sigma}])+\mathbf{k}(1;[\mathbf{j}_{\mathrm{p}}[\rho^{\mathrm{s}}_{1\sigma}],\mathbf{A}_{\mathrm{xc}\sigma}])\}\\ &\quad-2\int d^3 2\{\boldsymbol{\nabla}(1)u(12)\}\{n^{-1}_{\sigma}(1)n_{2\sigma}(12)-\tfrac{1}{2}n(2)\}.\end{aligned} \tag{3.30a}$$

It gives the force arising from the scalar exchange-correlation potential $v_{\mathrm{xc}\sigma}(1)$ in terms of $\rho_{1\sigma}$, $n_{2\sigma}$, $\rho^{\mathrm{s}}_{1\sigma}$, u, and $\mathbf{A}_{\mathrm{xc}\sigma}$. The force field $\mathbf{f}_{\mathrm{xc}\sigma}(\mathbf{r})$ is conservative because it stems from the potential $v_{\mathrm{xc}\sigma}(\mathbf{r})$ [see the first line of Eq. (3.30a)], therefore, similarly as in [5], the potential can be evaluated as a line integral

$$v_{\mathrm{xc}\sigma}(\mathbf{r}_0)=v_{\mathrm{xc}\sigma}(\mathbf{r}'_0)-\int_{\mathbf{r}'_0}^{\mathbf{r}_0} d\mathbf{r}\cdot\mathbf{f}_{\mathrm{xc}\sigma}(\mathbf{r}), \tag{3.30b}$$

which is independent of the particular path chosen for integration. Again, the reference point $\mathbf{r}'_0$ and the constant $v_{\mathrm{xc}\sigma}(\mathbf{r}'_0)$ are arbitrary.

Equations (3.28) and (3.30)—two alternative explicit expressions for the exchange-correlation scalar potential of the current-density functional theory—are the main results of the present subsection.

D. Exchange-correlation vector potential

In order to simplify Eq. (3.26) let us note that the difference of the continuity equations in terms of the physical current in the reference system, Eq. (3.14), and the original system, Eq. (3.8b), gives [see Eq. (4.9) of [9]]

$$0=\boldsymbol{\nabla}\{\mathbf{j}^{\mathrm{s}}_{\sigma}-\mathbf{j}_{\sigma}\}=\boldsymbol{\nabla}\left(\frac{\hbar e}{mc}n_{\sigma}\mathbf{A}_{\mathrm{xc}\sigma}\right) \tag{3.31}$$

[Eqs. (3.9) and (3.19) have been used]. So the divergence of the exchange-correlation vector potential can be written from Eq. (3.31) as

$$\boldsymbol{\nabla}\cdot\mathbf{A}_{\mathrm{xc}\sigma}(1)=-\mathbf{A}_{\mathrm{xc}\sigma}(1)\mathbf{g}(1;[n_{\sigma}]), \tag{3.32a}$$

in terms of the reduced density gradient

$$\mathbf{g}(1;[n])=\frac{\boldsymbol{\nabla}n(1)}{n(1)}. \tag{3.32b}$$

Finally, using Eqs. (3.27) and (3.32), Eq. (3.26) can be rewritten as

$$\mathbf{A}_{\mathrm{xc}\sigma}(1)\mathbf{G}(11')+\mathbf{A}_{\mathrm{xc}\sigma}(1')\mathbf{G}(1'1)=H(11'), \tag{3.33a}$$

with

$$\begin{aligned}\mathbf{G}(11')&=\frac{\hbar e}{mc}\{\tfrac{1}{2}\mathbf{g}(1;[n_{\sigma}])-\mathrm{Re}\mathbf{W}_{\mathrm{gr}}(11';[\rho^{\mathrm{s}}_{1\sigma}])\}\\ &=\frac{\hbar e}{mc}\,\mathrm{Re}\{\mathbf{W}_{\mathrm{gr}}(11'';[\rho^{\mathrm{s}}_{1\sigma}])\big|_{1''=1}-\mathbf{W}_{\mathrm{gr}}(11';[\rho^{\mathrm{s}}_{1\sigma}])\}\\ &=\mathbf{G}(11';[\rho^{\mathrm{s}}_{1\sigma}]),\end{aligned} \tag{3.33b}$$

$$\begin{aligned}H(11')&=H(1'1)=H(11';[\mathbf{A},u,\rho^{\mathrm{s}}_{1\sigma},\rho_{1\sigma},\rho_{2\sigma}])\\ &=\Big(\frac{\hbar e}{mc}\mathbf{A}(1)\ \mathrm{Re}\{\mathbf{W}_{\mathrm{gr}}(11';[\rho^{\mathrm{s}}_{1\sigma}])\\ &\quad-\mathbf{W}_{\mathrm{gr}}(11';[\rho_{1\sigma}])\}\Big)+\{1\ \rightleftarrows\ 1'\}\\ &\quad+\mathrm{Im}\{W_{\mathrm{kin}}(11';[\rho_{1\sigma}])-W_{\mathrm{kin}}(11';[\rho^{\mathrm{s}}_{1\sigma}])\\ &\quad+W_{\mathrm{ee}}(11';[u,\rho_{1\sigma},\rho_{2\sigma}])\},\end{aligned} \tag{3.33c}$$

having diagonals

$$\mathbf{G}(11)=\mathbf{0},\quad H(11)=0. \tag{3.33d}$$

Thus Eq. (3.33a) relates, at any point pair $\{1,1'\}$, the exchange-correlation vector potential field with the external one, and with some simple functionals of DMs $\rho^{\mathrm{s}}_{1\sigma}$, $\rho_{1\sigma}$, $\rho_{2\sigma}$, and the interaction u. Since our aim is to reconstruct $\mathbf{A}_{\mathrm{xc}\sigma}$ from the known DMs with the help of Eq. (3.33), let us investigate first if some freedom is left by the gauge transformation of $\mathbf{A}^{\mathrm{eff}}_{\sigma}$, Eq. (A1). We see from Eq. (A4) that a constant Λ only does not change DMs, but it means no transformation at all, Eq. (A1). So, opposite to the case of the scalar potential, the vector potential $\mathbf{A}_{\mathrm{xc}\sigma}$ must follow from the DMs in a unique way.

We sketch now a procedure for calculation of the field $\mathbf{A}_{\mathrm{xc}\sigma}(\mathbf{r})$ from Eq. (3.33) by applying a discretization of the problem. The vectors $\mathbf{A}_{\mathrm{xc}\sigma}(\mathbf{r})$ will be determined on a grid of (large number) M points. Since the derivatives of DMs are undefined at nuclear positions (because of ''cusps'' there, see, e.g., [10]), these points should be avoided on the grid. Equation (3.33a) is symmetric in $\{1,1'\}$, and its diagonal $\{1,1\}$ represents a trivial identity [see Eq. (3.33d)]. Therefore the discretization of Eq. (3.33a) leads to a system of $(M-1)M/2$ linear inhomogeneous equations in $3M$ unknowns—the components of $\mathbf{A}_{\mathrm{xc}\sigma}(j)$, $j=1,\ldots,M$. This system, although overdetermined, cannot be contradictory, because it stems from exact relations satisfied by DMs. So, a subsystem of $3M$ equations, having a nonsingular coefficient matrix, can be chosen, and then solved. It should be noted that the DVE (3.29) does not provide a separate equation for

determination of $\mathbf{A}_{\mathrm{xc}\sigma}(\mathbf{r})$ [to be an analog of the derived from EOM Eq. (3.26)], because it is lacking the imaginary part.

The determined field $\mathbf{A}_{\mathrm{xc}\sigma}(\mathbf{r})$ can be inserted into Eq. (3.28) or (3.30), which give $v_{\mathrm{xc}\sigma}(\mathbf{r})$. In this way the construction of both vector and scalar exchange-correlation potentials is possible from the following input: $\rho^{\mathrm{s}}_{1\sigma}$, $\rho_{1\sigma}$, $\rho_{2\sigma}$, u, and $\mathbf{A}$.

E. Approximate exchange-only potentials

In our investigations [5,6] of finite electron systems in the absence of magnetic fields, we proposed approximations for the exchange-only potential, derived from the exact expressions for the exchange-correlation potential by means of replacing there the interacting-system DMs by their noninteracting counterparts. We were guided by the fact that such replacement in the exact expression (in terms of DMs) for E_{xc} resulted in the exact exchange energy. Applying this replacement now to Eq. (3.28) we obtain an approximate expression for the exchange scalar potential from the EOM approach, free of the kinetic energy operator terms W_{kin} and of the external vector potential $\mathbf{A}$. The exchange-correlation vector potential remaining there should be replaced by its exchange-only part, to be consistent with the applied approximation. The result is

$$\begin{aligned} v^{\mathrm{EOM}}_{\mathrm{x}\sigma}(1) = v^{\mathrm{EOM}}_{\mathrm{x}\sigma}(1') &- \frac{\hbar e}{mc}\,\mathrm{Im}(\{\mathbf{A}_{\mathrm{x}\sigma}(1)\mathbf{W}_{\mathrm{gr}}(11';[\rho^{\mathrm{s}}_{1\sigma}])\} \\ &+\{1 \;\overrightarrow{\leftarrow}\; 1'\}^{*}) \\ &- \int d^3 2\{u(12)-u(1'2)\} \\ &\times \mathrm{Re}\left(\frac{\rho^{\mathrm{s}}_{1\sigma}(1;2)\rho^{\mathrm{s}}_{1\sigma}(2;1')}{\rho^{\mathrm{s}}_{1\sigma}(1;1')}\right). \end{aligned} \tag{3.34}$$

The form of its last term results from expressing the 2DM γ^{s}_2 in terms of the 1DM γ^{s}_1 as

$$\gamma^{\mathrm{s}}_2(12;1'2') = \tfrac{1}{2}\{\gamma^{\mathrm{s}}_1(1;1')\gamma^{\mathrm{s}}_1(2;2') - \gamma^{\mathrm{s}}_1(1;2')\gamma^{\mathrm{s}}_1(2;1')\}, \tag{3.35}$$

valid, in general, for DMs derived from a single-determinantal wave function [10], and from expressing the 1DM as [see Eq. (3.12)]

$$\begin{aligned} \gamma^{\mathrm{s}}_1(\mathbf{r}_1 s_1;\mathbf{r}'_1 s'_1) = &\rho^{\mathrm{s}}_{1\uparrow}(\mathbf{r}_1;\mathbf{r}'_1)\alpha(s_1)\alpha(s'_1) \\ &+\rho^{\mathrm{s}}_{1\downarrow}(\mathbf{r}_1;\mathbf{r}'_1)\beta(s_1)\beta(s'_1), \end{aligned} \tag{3.36}$$

because the noninteracting-system Hamiltonian commutes with the total-spin z component operator [10], and applying next the definition of $\rho^{\mathrm{s}}_{2\sigma}$ according to Eq. (3.5b). While the exact $v_{\mathrm{xc}\sigma}(\mathbf{r})$, determined from Eq. (3.28), must be independent of the choice of $\mathbf{r}'$ for the reference point [up to arbitrary additive constant $v_{\mathrm{xc}\sigma}(\mathbf{r}')$], this property may be lost, due to approximations, by $v^{\mathrm{EOM}}_{x\sigma}(\mathbf{r})$, Eq. (3.34), although we anticipate this potential dependence on $\mathbf{r}'$ will be weak.

By applying the replacements $\rho_{k\sigma}\rightarrow\rho^{\mathrm{s}}_{k\sigma}$ in Eq. (3.33), we obtain an equation for the approximate exchange-only vector potential

$$\mathbf{A}^{\mathrm{EOM}}_{\mathrm{x}\sigma}(1)\mathbf{G}(11') + \mathbf{A}^{\mathrm{EOM}}_{\mathrm{x}\sigma}(1')\mathbf{G}(1'1) = H^{\mathrm{EOM}}(11'), \tag{3.37a}$$

with

$$\begin{aligned} H^{\mathrm{EOM}}(11') &= \mathrm{Im}W_{\mathrm{ee}}(11';[u,\rho^{\mathrm{s}}_{1\sigma},\rho^{\mathrm{s}}_{2\sigma}]) \\ &= -\int d^3 2\{u(12)-u(1'2)\} \\ &\quad\times \mathrm{Im}\left(\frac{\rho^{\mathrm{s}}_{1\sigma}(1;2)\rho^{\mathrm{s}}_{1\sigma}(2;1')}{\rho^{\mathrm{s}}_{1\sigma}(1;1')}\right), \end{aligned} \tag{3.37b}$$

free of dependence on the external potential $\mathbf{A}(\mathbf{r})$ [compare Eq. (3.34) for the W_{ee} contribution].

Determination of the field $\mathbf{A}^{\mathrm{EOM}}_{\mathrm{x}\sigma}$ from Eq. (3.37) may be performed in a similar way as in the case of $\mathbf{A}_{\mathrm{xc}\sigma}$ from Eq. (3.33). Because of approximations, the system of $(M-1)M/2$ linear inhomogeneous equations in $3M$ unknowns, corresponding to the discretized Eq. (3.37a), may be (slightly) contradictory. Therefore it should be solved as a linear least-squares problem. One ought, perhaps, to add to the minimized least-squares sum also the terms corresponding to the discretized Eq. (3.32a) (with the divergence represented via finite differences), in order to impose on the approximate $\mathbf{A}^{\mathrm{EOM}}_{\mathrm{x}\sigma}$ the constraint of satisfying the continuity equation.

By applying again the discussed replacements $\rho_{p\sigma}\rightarrow\rho^{\mathrm{s}}_{p\sigma}$ to the expression for the exchange-correlation force, Eq. (3.30a), we obtain the following approximation for the exchange-only force stemming from the DVE:

$$\begin{aligned} \mathbf{f}^{\mathrm{DVE}}_{\mathrm{x}\sigma}(1) = &n^{-1}_{\sigma}(1)\mathbf{k}(1;[\mathbf{j}_{\mathrm{p}}[\rho^{\mathrm{s}}_{1\sigma}],\mathbf{A}_{\mathrm{x}\sigma}]) \\ &+\int d^3 2\{\boldsymbol{\nabla}(1)u(12)\}n^{-1}_{\sigma}(1)|\rho^{\mathrm{s}}_{1\sigma}(12)|^2, \end{aligned} \tag{3.38a}$$

and, with the help of line integration, the corresponding potential

$$v^{\mathrm{DVE}}_{\mathrm{x}\sigma}(\mathbf{r}_0) = v^{\mathrm{DVE}}_{\mathrm{x}\sigma}(\mathbf{r}'_0) - \int_{\mathbf{r}'_0}^{\mathbf{r}_0} d\mathbf{r}\cdot\mathbf{f}^{\mathrm{DVE}}_{\mathrm{x}\sigma}(\mathbf{r}). \tag{3.38b}$$

Similarly as in the EOM case, the approximate exchange scalar potential $v^{\mathrm{DVE}}_{\mathrm{x}\sigma}$ may depend on the choice of $\mathbf{r}'_0$ for the reference point and on the integration path, because the approximate force field may contain also some (small) nonconservative component. When $\mathbf{A}^{\mathrm{EOM}}_{\mathrm{x}\sigma}$, determined from Eq. (3.37), is inserted for $\mathbf{A}_{\mathrm{x}\sigma}$ in Eq. (3.34) or (3.38), construction of the approximate exchange vector and scalar potentials becomes feasible solely in terms of $\rho^{\mathrm{s}}_{1\sigma}$ and u.

It should be noted that the result (3.38a) reduces to the Harbola-Sahni exchange-only result [16] in their work formalism,

$$\mathbf{f}_{\mathrm{x}\sigma}^{\mathrm{DVE}}(\mathbf{r}_1)\rightarrow\int d^3r_2\rho_{\mathrm{x}}(\mathbf{r}_1,\mathbf{r}_2)(\mathbf{r}_1-\mathbf{r}_2)/|\mathbf{r}_1-\mathbf{r}_2|^3,< \tag{3.39}$$

when the magnetic field is absent (and so $\mathbf{A}_{\mathrm{x}\sigma}=\mathbf{0}$), and a spin-compensated system ($\rho_{1\uparrow}=\rho_{1\downarrow}=\frac{1}{2}\rho_1$) is considered together with the Coulombic $u(\mathbf{r}_1,\mathbf{r}_2)$. Here $\rho_{\mathrm{x}}(\mathbf{r}_1,\mathbf{r}_2)$ denotes the so-called exchange hole density (see, e.g., [11]). Since all the approximations discussed above, Eqs. (3.34), (3.37), (3.38), are analogs or extensions of the Harbola-Sahni approximation, one may expect them to lead to similarly promising results as their zero-field work (see, e.g., Sahni's review [17]).

IV. HARTREE-FOCK APPROACH

As the Hartree-Fock (HF) approximation is still widely used to investigate atoms in a magnetic field (e.g., [3]), we derive below its density-matrix form. The self-consistent one-electron eigenfunctions of the HF single determinant are solutions of

$$\hat{h}_{\mathrm{HF}}(\mathbf{x})\phi_a^{\mathrm{HF}}(\mathbf{x})=\epsilon_a^{\mathrm{HF}}\phi_a^{\mathrm{HF}}(\mathbf{x}), \tag{4.1}$$

where

$$\hat{h}_{\mathrm{HF}}(\mathbf{r}\sigma)=\hat{h}(\mathbf{r};[v_\sigma,\mathbf{A}])+v_{\mathrm{es}}(\mathbf{r};[n^{\mathrm{HF}}])+\hat{v}_{\mathrm{x}}^{\mathrm{HF}}(\mathbf{r}\sigma;[\gamma_1^{\mathrm{HF}}]). \tag{4.2}$$

Here the HF 1DM

$$\gamma_1^{\mathrm{HF}}(\mathbf{x};\mathbf{x}')=\sum_{a=1}^{N}\phi_a^{\mathrm{HF}}(\mathbf{x})\phi_a^{\mathrm{HF}*}(\mathbf{x}') \tag{4.3}$$

enters the kernel

$$\tilde{v}_{\mathrm{x}}^{\mathrm{HF}}(\mathbf{12};[\gamma_1^{\mathrm{HF}}])=-u(12)\gamma_1^{\mathrm{HF}}(\mathbf{1};\mathbf{2}) \tag{4.4}$$

of the nonlocal exchange potential (integral operator)

$$\hat{v}_{\mathrm{x}}^{\mathrm{HF}}(\mathbf{1})\varphi(\mathbf{1})=\int d^4 2\,\tilde{v}_{\mathrm{x}}^{\mathrm{HF}}(\mathbf{12})\varphi(\mathbf{2}), \tag{4.5}$$

while the external potential is

$$v_\sigma(\mathbf{r})=v(\mathbf{r})\pm\frac{e\hbar}{2mc}B(\mathbf{r}),\quad \pm \text{ for } \sigma=\uparrow,\downarrow. \tag{4.6}$$

The EOM for the HF 1DM is [compare Eq. (3.11)]

$$\{\hat{h}_{\mathrm{HF}}(\mathbf{1})-\hat{h}_{\mathrm{HF}}^{*}(\mathbf{1}')\}\gamma_1^{\mathrm{HF}}(\mathbf{1};\mathbf{1}')=0. \tag{4.7}$$

Since the z component of the total spin is a good quantum number, the HF 1DM is given by Eq. (3.36) with the replacement s→HF for superscripts. Therefore Eq. (4.7) is equivalent to two equations, for $\sigma=\uparrow,\downarrow$:

$$\{\hat{h}_{\mathrm{HF}}(\mathbf{r}_1\sigma)-\hat{h}_{\mathrm{HF}}^{*}(\mathbf{r}_1'\sigma)\}\rho_{1\sigma}^{\mathrm{HF}}(\mathbf{r}_1;\mathbf{r}_1')=0. \tag{4.8}$$

The terms containing v_{es} and $\hat{v}_{\mathrm{x}}^{\mathrm{HF}}$ can be combined to give a contribution in terms of the HF 2DM γ_2^{HF}, which is a combination of HF 1DM γ_1^{HF} elements [Eq. (3.35) with s→HF], so an alternative form of the EOM looks like the exact EOM (3.4b) of the interacting system

$$\{\hat{h}(1;[v_\sigma,\mathbf{A}])-\hat{h}^{*}(1';[v_\sigma,\mathbf{A}])\}\rho_{1\sigma}^{\mathrm{HF}}(1;1')+2\int d^3 2\{u(12)-u(1'2)\}\rho_{2\sigma}^{\mathrm{HF}}(12;1'2)=0. \tag{4.9}$$

The analogous equation in the absence of a magnetic field was obtained by us earlier in [18]. The corresponding DVE reads

$$\mathbf{z}(1;[\rho_{1\sigma}^{\mathrm{HF}}])-\frac{\hbar^2}{4m}\boldsymbol{\nabla}\nabla^2 n_\sigma^{\mathrm{HF}}(1)+n_\sigma^{\mathrm{HF}}(1)\boldsymbol{\nabla}\left(v_\sigma(1)+\frac{e^2}{2mc^2}A^2(1)\right)+\mathbf{k}(1;[\mathbf{j}_{\mathrm{p}}[\rho_{1\sigma}^{\mathrm{HF}}],\mathbf{A}])+2\int d^3 2\{\nabla(1)u(12)\}n_{2\sigma}^{\mathrm{HF}}(12)=0 \tag{4.10}$$

so it looks like the exact DVE (3.6) of the interacting system.

Equations (4.9) and (4.10) open an interesting possibility of checking the accuracy of any numerical solution of the HF equations for a particular system. Since Eq. (3.4b) has exactly the same form as Eq. (4.9), Eq. (3.17a), which is derived from it, can be rewritten in terms of the HF DMs:

$$v_\sigma(\mathbf{r})+\frac{e^2}{2mc^2}A^2(\mathbf{r})-\text{const}=-\mathrm{Re}\{W_{\mathrm{dop}}(\mathbf{r},\mathbf{r}';[\mathbf{A},\rho_{1\sigma}^{\mathrm{HF}}])+W_{\mathrm{ee}}(\mathbf{r},\mathbf{r}';[u,\rho_{1\sigma}^{\mathrm{HF}},\rho_{2\sigma}^{\mathrm{HF}}])\}, \tag{4.11a}$$

$$-\frac{\hbar e}{2mc}\{\nabla\mathbf{A}(\mathbf{r})+\nabla\mathbf{A}(\mathbf{r}')\}=-\mathrm{Im}\{W_{\mathrm{dop}}(\mathbf{r},\mathbf{r}';[\mathbf{A},\rho_{1\sigma}^{\mathrm{HF}}])+W_{\mathrm{ee}}(\mathbf{r},\mathbf{r}';[u,\rho_{1\sigma}^{\mathrm{HF}},\rho_{2\sigma}^{\mathrm{HF}}])\}, \tag{4.11b}$$

the reference point $\mathbf{r}'$ is kept fixed. Similarly, from Eq. (4.10) divided by $n_\sigma^{\mathrm{HF}}(\mathbf{r})$ we have

$$\boldsymbol{\nabla}\left(v_\sigma(\mathbf{r})+\frac{e^2}{2mc^2}A^2(\mathbf{r})\right)=-\{n_\sigma^{\mathrm{HF}}(\mathbf{r})\}^{-1}\times\left(\mathbf{z}(\mathbf{r};[\rho_{1\sigma}^{\mathrm{HF}}])-\frac{\hbar^2}{4m}\boldsymbol{\nabla}\nabla^2 n_\sigma^{\mathrm{HF}}(\mathbf{r})+\mathbf{k}(\mathbf{r};[\mathbf{j}_{\mathrm{p}}[\rho_{1\sigma}^{\mathrm{HF}}],\mathbf{A}])+2\int d^3 r'\{\boldsymbol{\nabla}(\mathbf{r})u(\mathbf{r},\mathbf{r}')\}n_{2\sigma}^{\mathrm{HF}}(\mathbf{r},\mathbf{r}')\right). \tag{4.11c}$$

After evaluating the right-hand sides of Eqs. (4.11) on a grid of points $\mathbf{r}$, using $\rho_{1\sigma}^{\mathrm{HF}}$ and $\rho_{2\sigma}^{\mathrm{HF}}$ (calculated from the inaccurate HF orbitals) and the given $\mathbf{A}$, the results may be thought of as representing the (shown on the left-hand sides) combinations of some actual potentials $\{v_\sigma(\mathbf{r}),\mathbf{A}(\mathbf{r})\}$ for which the considered HF orbitals are the exact solutions. By comparing the obtained combinations with those evaluated (according to

the left-hand side expressions) from the applied external potentials, **r** regions of substantial discrepancy can be detected. This may be helpful when looking for improvements in the calculational procedure, for instance, by providing more accurate representation of orbitals in the troublesome regions.

Since Eqs. (3.4b) and (3.6) in terms of the DMs generated from the exact wave function $\Psi_E(\mathbf{1}\cdots\mathbf{N})$ look the same as Eqs. (4.9) and (4.10) in terms of the HF DMs, all the above considerations, concerning investigation of the quality of the HF numerical solutions, can be applied to an analogous study of the N-electron wave function obtained by some ''accurate'' methods (like the configuration interaction or the perturbation theory).

V. SUMMARY

The most important results established through the present density-matrix approach to the nonrelativistic current-density functional theory, applicable to finite electron systems in magnetic fields of arbitrary strength but a constant direction, are (i) the equation of motion (3.4) for the first-order density matrix; (ii) the differential virial equation (3.6); (iii) an expression for the exchange-correlation scalar potential $v_{\mathrm{xc}\sigma}(\mathbf{r})$ in terms of density matrices: (a) in a direct form, involving the vector potentials $\mathbf{A}(\mathbf{r})$, $\mathbf{A}_{\mathrm{xc}\sigma}(\mathbf{r})$, in Eq. (3.28), (b) in a line-integral form, involving $\mathbf{A}_{\mathrm{xc}\sigma}(\mathbf{r})$, in Eq. (3.30); (iv) an equation for determining the exchange-correlation vector potential $\mathbf{A}_{\mathrm{xc}\sigma}(\mathbf{r})$ in terms of density matrices and the external potential $\mathbf{A}(\mathbf{r})$, Eq. (3.33); (v) an expression for an approximate exchange-only scalar potential in terms of noninteracting-system density matrices and $\mathbf{A}_{\mathrm{x}\sigma}(\mathbf{r})$: (a) in a direct form, in Eq. (3.34), (b) in a line-integral form, in Eq. (3.38); (vi) an equation for determining an approximate exchange-only vector potential in terms of the noninteracting-system first-order density matrix, in Eq. (3.37); and (vii) the proof that the equation of motion (4.9) and the differential virial equation (4.10), satisfied by the density matrices of the Hartree-Fock approximation, are the same as corresponding equations (3.4b) and (3.6), satisfied by the exact matrices.

While $v_{\mathrm{xc}\sigma}(\mathbf{r})$ and $\mathbf{A}_{\mathrm{xc}\sigma}(\mathbf{r})$ are defined via functional differentiation, Eqs. (3.23) and (3.24), the merit of the results (iii) and (iv) is to avoid such differentiation, but the price paid is that potentials are found in terms of nondiagonal elements of density matrices, going beyond these diagonal and close-to-diagonal elements, which define $n_\sigma(\mathbf{r})$, $\mathbf{j}_{\mathrm{p}\sigma}(\mathbf{r})$, and $n_{2\sigma}(\mathbf{r}_1,\mathbf{r}_2)$. Although these results are somewhat formal, they may become useful in practice if some controlled approximations for the interacting-system density matrices (as functionals of the density) can be inserted.

As the present investigation was approaching completion, a work by Capelle and Gross [19] appeared, linking intimately two versions of the current-density functional theory (CDFT) and the spin-density functional theory (SDFT). Their result may be used to transform our equations for exact $v_{\mathrm{xc}\sigma}$, $\mathbf{A}_{\mathrm{xc}\sigma}$ and approximate $v_{\mathrm{x}\sigma}$, $\mathbf{A}_{\mathrm{x}\sigma}$, pertaining to the Vignale and Rasolt CDFT1, to corresponding potentials of CDFT2 and SDFT, on a certain set of densities. Their work [19] motivates also future extension of our investigation via the density-matrix approach, from the present CDFT1 to the CDFT2 and SDFT, including the extension to an arbitrary direction of a magnetic field.

ACKNOWLEDGMENTS

We wish to acknowledge that the collaboration which led to the present work (in particular, the visit of A.H. to England) was partially supported by the Leverhulme Trust for research on density matrices and density functional theory. N.H.M. also wishes to acknowledge partial financial support for work on atoms and molecules in intense applied fields under an ONR Contract. Dr. P. Schmidt is thanked for valuable discussions and his continuing support.

APPENDIX: GAUGE INVARIANCE

For time-independent problems, if we transform the vector potential, occurring in the Hamiltonian $\hat{\mathcal{H}}[v,\mathbf{A}]$, Eq. (2.5), according to

$$\mathbf{A}(\mathbf{r})\rightarrow\mathbf{A}_{\mathrm{new}}(\mathbf{r})=\mathbf{A}(\mathbf{r})-8\boldsymbol{\nabla}\Lambda(\mathbf{r}), \tag{A1}$$

where $\Lambda(\mathbf{r})$ is an arbitrary scalar function, and at the same time modify the wave function according to

$$\Psi(\mathbf{x}_1,\ldots,\mathbf{x}_N)\rightarrow\Psi_{\mathrm{new}}(\mathbf{x}_1,\ldots,\mathbf{x}_N) = \exp\left(i\frac{e}{\hbar c}\{\Lambda(\mathbf{r}_1)+\cdots+\Lambda(\mathbf{r}_N)\}\right)\Psi(\mathbf{x}_1,\ldots,\mathbf{x}_N), \tag{A2}$$

then the form of the original Schrödinger equation (2.8b) remains unchanged,

$$\hat{\mathcal{H}}[v,\mathbf{A}_{\mathrm{new}}]\Psi_{E,\mathrm{new}}=E\Psi_{E,\mathrm{new}}. \tag{A3}$$

This is called the gauge invariance of the Schrödinger equation. The transformation (A2) induces the following transformation of DMs, Eqs. (2.11), (2.12):

$$\gamma_{2,\mathrm{new}}(\mathbf{12};\mathbf{1'2})=\exp\left(i\frac{e}{\hbar c}\{\Lambda(1)-\Lambda(1')\}\right)\gamma_2(\mathbf{12};\mathbf{1'2}), \tag{A4a}$$

$$\gamma_{1,\mathrm{new}}(\mathbf{1};\mathbf{1'})=\exp\left(i\frac{e}{\hbar c}\{\Lambda(1)-\Lambda(1')\}\right)\gamma_1(\mathbf{1};\mathbf{1'}). \tag{A4b}$$

Since the EOM (2.10) follows directly from Eq. (2.9), its gauge invariance is dictated by Eq. (A3), which means that Eq. (2.10) remains unchanged if transformations $\mathbf{A}\rightarrow\mathbf{A}_{\mathrm{new}}$, Eq. (A1), and $\gamma_k\rightarrow\gamma_{k,\mathrm{new}}$, Eq. (A4), are performed simultaneously. The gauge invariance of the DVE (2.15) means the same, because this equation is obtained from Eq. (2.10) by differentiation. It should be noted that the diagonal elements of DMs, Eqs. (2.13) and (2.14), are invariant, $n_{\mathrm{new}}=n$, $n_{2,\mathrm{new}}=n_2$, as it follows from Eq. (A4). The gauge invariance of the physical current, Eq. (2.25), means that

$$\mathbf{j}_{\mathrm{p}}[\rho_{1,\mathrm{new}}]+\frac{e}{mc}n\mathbf{A}_{\mathrm{new}}=\mathbf{j}_{\mathrm{p}}[\rho_1]+\frac{e}{mc}n\mathbf{A}. \tag{A5}$$

Concerning the force-balance equation (2.23), we note invariance of the following terms: in $\mathbf{f}^{\mathrm{nmg}}$ the term $\boldsymbol{\nabla}\nabla^2 n$ and the term involving n_2, in $\mathbf{f}^{\mathrm{mag}}$ the Lorentz force $\mathbf{f}^{\mathrm{Lor}}$, and, finally, the sum $(\mathbf{f}^{\mathrm{inh}}+n^{-1}\mathbf{z})$.

When extending all above considerations to include the electron spin, in the manner introduced in Sec. III A, it is sufficient to replace in Eqs. (A1)–(A4) the fields $\mathbf{A}(\mathbf{r}_j)$ and $\Lambda(\mathbf{r}_j)$ by $\mathbf{A}_{\sigma_j}(\mathbf{r}_j)$ and $\Lambda_{\sigma_j}(\mathbf{r}_j)$ (here the jth electron coordinate is $\mathbf{x}_j=\{\mathbf{r}_j\sigma_j\}$). Then the gauge invariance is shown separately for $\sigma=\uparrow,\downarrow$, in the EOM and DVE, (3.4b), (3.6), similarly in two physical currents (3.8b), and in two continuity equations $\boldsymbol{\nabla}\cdot\mathbf{j}_\sigma=0$.

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