Density-Functional Theory in Strong Magnetic Fields

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We formulate the current-density–functional theory for systems in arbitrarily strong magnetic fields. A set of self-consistent equations comparable to the Kohn-Sham equations for ordinary density-functional theory is derived, and proved to be gauge invariant and to satisfy the continuity equation. We prove that the exchange-correlation energy functional $E_{xc}[n,\mathbf{j}_p]$ [$n(\mathbf{r})$ is the density and $\mathbf{j}_p(\mathbf{r})$ is the "paramagnetic" current density] depends on the current via the combination $\boldsymbol{\nu}(\mathbf{r})=\nabla\times[\mathbf{j}_p(\mathbf{r})/n(\mathbf{r})]$. An explicit formula for E_{xc} is derived, which is local in $\boldsymbol{\nu}(\mathbf{r})$.

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Since the seminal papers of Hohenberg and Kohn[1] and Kohn and Sham,[2] density-functional theory (DFT) has developed into an important tool for the treatment of many-body problems in condensed-matter physics. Its practical success has prompted much theoretical work aimed at the extension of the applicability of the theory to systems more general than those considered in the original papers.[3,4]

The interest of this Letter is the formulation of the *current-density–functional theory* for systems in the presence of an external magnetic field. Many recent exciting phenomena[5,6] involving electrons in large magnetic fields are a strong motivation for the need of such a formulation.

Magnetic fields have been incorporated in the DFT only insofar as they cause spin polarization.[3,7–9] The fact that also orbital currents are induced has long been recognized.[7] Here we incorporate this effect into the self-consistent formulation of one-particle equations which lies at the heart of a practical implementation of DFT. The basic variables are the particle density $n(\mathbf{r})$ and the "paramagnetic" current density $\mathbf{j}_p(\mathbf{r})$ [see Eq. (2)]. The latter must be used rather than the physical current density [see Eq. (3)] because it uniquely determines the vector potential and the ground-state wave function. Furthermore, since in the variational principle the external vector potential is kept constant, the minimization of the energy functional will have to be with respect to $\mathbf{j}_p$. Consequently, a key difficulty that we had to overcome is that a one-particle–equations formulation does not *a priori* satisfy the physical requirements of gauge invariance and the continuity equation.

In the following we demonstrate that both requirements can be actually satisfied because of an exact transformation property of the exchange-correlation energy functional, which we derive here. As a consequence of this transformation, the exchange-correlation energy functional $E_{xc}[n,\mathbf{j}_p]$, which is now a functional of *both* particle density and "paramagnetic" current density, takes the form

$$E_{xc}[n(\mathbf{r}),\mathbf{j}_p(\mathbf{r})]=\bar{E}_{xc}[n(\mathbf{r}),\nabla\times[\mathbf{j}_p(\mathbf{r})/n(\mathbf{r})]] \qquad (1)$$

{$\bar{E}_{xc}$ is a functional of both $n(\mathbf{r})$ and $\nabla\times[\mathbf{j}_p(\mathbf{r})/n(\mathbf{r})]$}. This is the first key result of the Letter, and it is this which allows the formulation of single-particle equations in the presence of a magnetic field. A second result is an explicit calculation of $\bar{E}_{xc}$ in the limit of a slowly varying magnetic field.

We start with a brief discussion of the uniqueness and variational properties of the energy, now in the presence of an external magnetic field. The nonrelativistic Hamiltonian for a system of N electrons in external scalar and vector potentials $v(\mathbf{r})$ and $\mathbf{A}(\mathbf{r})$ is

$$H=T+U+V+W.$$

The definitions for the various terms are (e is the absolute value of the charge) as follows:

$$T=\int d^3r\,\psi^\dagger(\mathbf{r})[-(\hbar^2/2m)\nabla^2]\psi(\mathbf{r}),\quad U=\tfrac{1}{2}\int d^3r\int d^3r'\,\psi^\dagger(\mathbf{r})\psi^\dagger(\mathbf{r}')u(\mathbf{r},\mathbf{r}')\psi(\mathbf{r}')\psi(\mathbf{r}),$$

$$V=\int d^3r\,v(\mathbf{r})n^{op}(\mathbf{r}),\quad W=\frac{e}{c}\int d^3r\,\mathbf{j}_p^{op}(\mathbf{r})\cdot\mathbf{A}(\mathbf{r})+\frac{e^2}{2mc^2}\int d^3r\,n^{op}(\mathbf{r})A^2(\mathbf{r}).$$

The density operator is defined as $n^{op}(\mathbf{r})\equiv\psi^\dagger(\mathbf{r})\psi(\mathbf{r})$, and the paramagnetic-current-density operator is

$$\mathbf{j}_p^{op}(\mathbf{r})=\frac{\hbar}{2mi}\{\psi^\dagger(\mathbf{r})\nabla\psi(\mathbf{r})-[\nabla\psi^\dagger(\mathbf{r})]\psi(r)\}. \qquad (2)$$

The physical-current-density operator is given by

$$\mathbf{j}^{\mathrm{op}}(\mathbf{r}) = \mathbf{j}_p^{\mathrm{op}}(\mathbf{r}) + (e/mc) n^{\mathrm{op}}(\mathbf{r}) \mathbf{A}(\mathbf{r}), \tag{3}$$

and satisfies the continuity equation

$$\nabla \cdot \mathbf{j}^{\mathrm{op}}(\mathbf{r};t) + \partial n^{\mathrm{op}}(\mathbf{r};t)/\partial t = 0.$$

In the above equations we have disregarded spin in order to concentrate on the novel physical feature of the orbital currents. Inclusion of spin is straightforward and will be considered elsewhere.

We now run through the basic theorems of density-functional theory in magnetic fields. Let $\mathbf{j}_p(\mathbf{r})$, $\mathbf{j}(\mathbf{r})$, and $n(\mathbf{r})$ denote the ground-state averages of the corresponding operators: Then the potentials $v(\mathbf{r})$ and $\mathbf{A}(\mathbf{r})$, and hence the ground-state wave function ψ, are uniquely determined (apart from an additive constant in the scalar potential) by the knowledge of the density distributions $n(\mathbf{r})$ and $\mathbf{j}_p(\mathbf{r})$.[10] For, suppose that there are two sets of fields $v(\mathbf{r}),\mathbf{A}(\mathbf{r})$ and $v'(\mathbf{r}),\mathbf{A}'(\mathbf{r})$ giving the same ground-state distributions $n(\mathbf{r})$ and $\mathbf{j}_p(\mathbf{r})$. Let $|\psi\rangle$ and $|\psi'\rangle$ be the two different ground states corresponding to the two sets of fields. Let H and H' be the two corresponding Hamiltonians and E and E' the two ground-state energies. Then from the variational principle for the ground state of H, we obtain the inequality

$$E = \langle\psi|H|\psi\rangle < \langle\psi'|H|\psi'\rangle = E' + \int d^3r\, n(\mathbf{r})[v(\mathbf{r}) - v'(\mathbf{r})] + \frac{e}{c}\int d^3r\, \mathbf{j}_p(\mathbf{r})\cdot[\mathbf{A}(\mathbf{r}) - \mathbf{A}'(\mathbf{r})] + \frac{e^2}{2mc^2}\int d^3r\, n(\mathbf{r})[A^2(\mathbf{r}) - A'^2(\mathbf{r})].$$

Another inequality is obtained by our interchanging the primed and the unprimed variables, and summing the two inequalities we get the contradiction

$$E + E' < E + E',$$

which proves the theorem.

For the variational principle, let

$$F[n',\mathbf{j}_p'] \equiv \langle\psi[n',\mathbf{j}_p']\,|\,(T+U)\,|\,\psi[n',\mathbf{j}_p']\rangle, \tag{4}$$

where $\psi[n',\mathbf{j}_p']$ is the ground-state wave function corresponding to n' and $\mathbf{j}_p'$. Then the functional

$$E_{v,\mathbf{A}}[n',\mathbf{j}_p'] = F[n',\mathbf{j}_p'] + \int d^3r'\, n'(\mathbf{r})v(\mathbf{r}) + \frac{e}{c}\int d^3r\, \mathbf{j}_p'(\mathbf{r})\cdot\mathbf{A}(\mathbf{r}) + \frac{e^2}{2mc^2}\int d^3r\, n'(\mathbf{r})A^2(\mathbf{r}) \tag{5}$$

has a minimum when n' and $\mathbf{j}_p'$ take the actual values corresponding to the potentials v and $\mathbf{A}$. This follows from the variational principle for the ground state of H, since

$$E_{v,\mathbf{A}}[n',\mathbf{j}_p'] = \langle\psi[n'\mathbf{j}_p']\,|\,H\,|\,\psi[n',\mathbf{j}_p']\rangle \geq \langle\psi[n,\mathbf{j}_p]\,|\,H\,|\,\psi[n,\mathbf{j}_p]\rangle = E_{v,\mathbf{A}}[n,\mathbf{j}_p],$$

which proves the theorem.

We now turn to the formulation of the one-particle equations. We first define the exchange-correlation energy functional as follows:

$$F[n,\mathbf{j}_p] = T_s[n,\mathbf{j}_p] + \tfrac{1}{2}\int d^3r \int d^3r'\, n(\mathbf{r})u(\mathbf{r},\mathbf{r}')n(\mathbf{r}') + E_{\mathrm{xc}}[n,\mathbf{j}_p], \tag{6}$$

where

$$T_s[n,\mathbf{j}_p] \equiv \langle\psi_0[n,\mathbf{j}_p]\,|\,T\,|\,\psi_0[n,\mathbf{j}_p]\rangle, \tag{7}$$

and $\psi_0[n,\mathbf{j}_p]$ is the ground-state wave function corresponding to n and $\mathbf{j}_p$ in a noninteracting version of the system.[11] $\psi_0[n,\mathbf{j}_p]$ is a Slater determinant of one-electron orbitals ψ_i which satisfy a one-particle Schrödinger equation with some, as yet undetermined, effective potentials. The functional T_s is also expressed in terms of the ψ_i's. Putting this representation of T_s in Eqs. (6) and (5) and carrying out the minimization of $E_{v,\mathbf{A}}$ determines the effective potentials. We get the following self-consistent formulation for the ground-state density and current:

$$n(\mathbf{r}) = \sum_{i=1}^{N} |\psi_i(\mathbf{r})|^2, \quad \mathbf{j}_p(\mathbf{r}) = \frac{\hbar}{2mi}\sum_{i=1}^{N}\{\psi_i^*(\mathbf{r})\nabla\psi_i(\mathbf{r}) - [\nabla\psi_i^*(\mathbf{r})]\psi_i(\mathbf{r})\},$$

$$\left[\frac{1}{2m}\left(-i\hbar\nabla + \frac{e}{c}[\mathbf{A}(\mathbf{r}) + \mathbf{A}_{\mathrm{xc}}(\mathbf{r})]\right)^2 + \frac{e^2}{2mc^2}\{\mathbf{A}^2(\mathbf{r}) - [\mathbf{A}(\mathbf{r}) + \mathbf{A}_{\mathrm{xc}}(\mathbf{r})]^2\} + v(\mathbf{r}) + \int d^3r'\, u(\mathbf{r},\mathbf{r}')n(\mathbf{r}') + v_{\mathrm{xc}}(\mathbf{r})\right]\psi_i(\mathbf{r}) = \epsilon_i\psi_i(\mathbf{r}), \tag{8}$$

$$v_{\mathrm{xc}}(\mathbf{r}) = \delta E_{\mathrm{xc}}[n,\mathbf{j}_p]/\delta n(\mathbf{r})\,|_{\mathbf{j}_p}, \tag{9}$$

$$(e/c)\mathbf{A}_{\mathrm{xc}}(\mathbf{r}) = \delta E_{\mathrm{xc}}[n,\mathbf{j}_p]/\delta\mathbf{j}_p(\mathbf{r})\,|_n. \tag{10}$$

The ground-state energy is given by

$$E=\sum_{i=1}^{N}\epsilon_i-\tfrac{1}{2}\int d^3r\int d^3r'\,n(\mathbf{r})u(\mathbf{r},\mathbf{r}')n(\mathbf{r}')-\int d^3r\,n(\mathbf{r})v_{\rm xc}(\mathbf{r})-\frac{e}{c}\int d^3r\,\mathbf{j}_p(\mathbf{r})\cdot\mathbf{A}_{\rm xc}(\mathbf{r})+E_{\rm xc}[n,\mathbf{j}_p]. \tag{11}$$

Notice that the effective vector potential $\mathbf{A}+\mathbf{A}_{\rm xc}$ enters the Schrödinger-type equation *linearly*.

We now prove the important result, Eq. (1), for the form of $E_{\rm xc}$.

Proof.—Consider the transformation

$$\mathbf{j}_p(\mathbf{r})\rightarrow\mathbf{j}_p(\mathbf{r})+(e/mc)n(\mathbf{r})\nabla\Lambda(\mathbf{r})\equiv\mathbf{j}_p'(\mathbf{r}), \tag{12}$$

where $\Lambda(\mathbf{r})$ is an arbitrary function. Clearly, the transformation of the wave function is

$$\psi[n,\mathbf{j}_p']=\psi[n,\mathbf{j}_p]\exp\left[i\frac{e}{\hbar c}\sum_{i=1}^{N}\Lambda(\mathbf{r}_i)\right].$$

The transformation of F is easily obtained from the definition (4):

$$F[n,\mathbf{j}_p']=F[n,\mathbf{j}_p]+\frac{e}{c}\int d^3r\,\mathbf{j}_p(\mathbf{r})\cdot\nabla\Lambda(r)+\frac{e^2}{2mc^2}\int d^3r\,n(\mathbf{r})\,|\nabla\Lambda(\mathbf{r})|^2.$$

The crucial point above this equation is that the transformation depends only on n and $\mathbf{j}_p$, not on the wave function. Therefore, the same transformation applies also to $T_s[n,\mathbf{j}_p]$, defined in Eq. (7). Putting these results together in Eq. (6), we discover

$$E_{\rm xc}[n,\mathbf{j}_p+(e/mc)n\nabla\Lambda]=E_{\rm xc}[n,\mathbf{j}_p]. \tag{13}$$

This is an *exact* property of the $E_{\rm xc}$ functional. Another way of expressing this property is that $E_{\rm xc}$ can only depend on the combination $\mathbf{v}(\mathbf{r})\equiv\nabla\times[\mathbf{j}_p(\mathbf{r})/n(\mathbf{r})]$:

$$E_{\rm xc}[n,\mathbf{j}_p]=\bar{E}_{\rm xc}[n,v]. \tag{14}$$

We next prove that this form, in conjunction with Eq. (8), implies $\nabla\cdot\mathbf{j}=0$; i.e., the static continuity equation is satisfied.

Proof.—Taking the functional derivatives of Eq. (14), we find

$$\frac{e}{c}\mathbf{A}_{\rm xc}(\mathbf{r})=-\frac{1}{n(\mathbf{r})}\nabla\times\frac{\delta\bar{E}_{\rm xc}[n,\mathbf{v}]}{\delta\mathbf{v}(\mathbf{r})}\bigg|_n, \tag{15}$$

$$v_{\rm xc}(\mathbf{r})=\frac{\delta\bar{E}_{\rm xc}[n,\mathbf{v}]}{\delta n(\mathbf{r})}\bigg|_{\mathbf{v}}-\frac{e}{c}\mathbf{A}_{\rm xc}(\mathbf{r})\cdot\frac{\mathbf{j}_p(\mathbf{r})}{n(\mathbf{r})}. \tag{16}$$

Equation (15) implies that $\nabla\cdot[n(\mathbf{r})\mathbf{A}_{\rm xc}(\mathbf{r})]=0$. Since the Schrödinger-type equation (8) already guarantees

$$\nabla\cdot\{\mathbf{j}_p(\mathbf{r})+(e/mc)n(\mathbf{r})[\mathbf{A}(\mathbf{r})+\mathbf{A}_{\rm xc}(\mathbf{r})]\}=0,$$

it follows that $\nabla\cdot[\mathbf{j}_p+(e/mc)n\mathbf{A}]\equiv\nabla\cdot\mathbf{j}=0$.

Finally, we prove that Eqs. (15) and (16) also ensure the gauge invariance of the formulation.

Proof.—Let ψ_i be the self-consistent orbitals corresponding to the vector potential $\mathbf{A}$. If $\mathbf{A}\rightarrow\mathbf{A}-\nabla\Lambda\equiv\mathbf{A}_{\rm new}$, let us multiply all the ψ_i's by a phase factor $\exp[(ie/\hbar c)\Lambda(\mathbf{r})]$. As a consequence, $\mathbf{j}_p$ transforms according to Eq. (12), while n is invariant. From Eqs. (15) and (16), one sees that $\mathbf{A}_{\rm xc}$ is invariant, while $V_{\rm xc}$ changes by $-(e^2/mc^2)\mathbf{A}_{\rm xc}\cdot\nabla\Lambda$. Putting these transformations in Eq. (8), one easily verifies that the new ψ_i's are indeed the solutions of the new self-consistent problem. Similarly, one verifies that the ground-state energy, given by Eq. (11), is gauge invariant.

Equation (14) is a strong constraint on the admissible form of any approximate $E_{\rm xc}$. For example, one immediately sees that a local approximation for the current density does not exist. On the other hand, a local approximation in $\mathbf{v}(\mathbf{r})$ is possible. We now present a local form for $E_{\rm xc}$, which is exact in the high-density limit and to order v^2. For an almost uniform electron gas, the $E_{\rm xc}$ functional can be written as

$$E_{\rm xc}[n,\mathbf{j}_p]=E_{\rm xc}^0[n]-\tfrac{1}{2}\textstyle\sum_{\mathbf{q}}\mathbf{j}_p(\mathbf{q})\cdot[\mathbf{P}^{-1}(q)-\mathbf{P}_0^{-1}(q)]\cdot\mathbf{j}_p(-\mathbf{q}),$$

where $E_{\rm xc}^0$ is the usual functional in the absence of currents and $\mathbf{P}(\mathbf{q})$ is the paramagnetic response tensor of the uniform electron gas[12] [$\mathbf{P}_0(\mathbf{q})$ is the same function for the noninteracting-electron gas]. The longitudinal part of $\mathbf{P}$ ($\mathbf{P}^0$) is equal to $-n/m$ and cancels in the difference. The transverse part, for small q, has the form $P(q)=-n/m-(c^2\chi_L/e^2)q^2$, where χ_L is the diamagnetic susceptibility (for the noninteracting gas $c^2\chi_L^0/e^2=-v_{\rm F}/12\pi^2\hbar$, where $v_{\rm F}$ is the Fermi velocity). The local form corresponds to a slowly varying current. We obtain, therefore,

$$E_{\rm xc}[n,\mathbf{j}_p]\cong E_{\rm xc}^0[n]+\int d^3r\,\frac{mk_{\rm F}}{24\pi^2}\left(\frac{\chi_L}{\chi_L^0}-1\right)\left|\nabla\times\left(\frac{\mathbf{j}_p(\mathbf{r})}{n}\right)\right|^2 \tag{17}$$

($k_{\rm F}$ is the Fermi momentum). We have calculated the diamagnetic susceptibility of the interacting-electron gas in the high-density limit using the random-phase approximation (RPA). The calculation is very lengthy and will be presented

elsewhere. It parallels Ma and Bruckner's calculation[13] of the exchange-correlation energy of a weakly inhomogeneous electron gas, but here is applied to the current-current correlation function. We found the exact result[14]

$$\chi_L/\chi_L^0 = 1 + 0.02764\, r_s \ln r_s + 0.01407\, r_s + O(r_s^2 \ln r_s) \tag{18}$$

(r_s is the usual electron-gas parameter). We have also extended the RPA calculation to metallic densities, where the high-density expansion is no longer applicable.[15] The transition to the inhomogeneous system is made by letting $n \rightarrow n(\mathbf{r})$ in Eq. (17). The Fermi momentum and the diamagnetic susceptibilities are now those corresponding to the local density. The condition (14) removes the ambiguity in the position of $n(r)$ relative to the ∇ operator.

Equation (17) is the first explicit construction of a current-dependent E_{xc}. It is valid at high densities and for magnetic fields B satisfying the condition $2\mu_B B(\mathbf{r}) \ll E_F(\mathbf{r})$ [μ_B is the Bohr magneton, and $E_F(\mathbf{r})$ is the local Fermi energy]. For systems with $E_F \cong 1$ eV, this condition is satisfied by all practically attainable field strengths.

In conclusion, we have formulated the current-density-functional theory for systems in arbitrarily strong magnetic fields. We have constructed self-consistent one-electron equations which are gauge invariant and satisfy the continuity equation. An important property of the exchange-correlation energy functional has been discovered, and used as a guide to determine the form of the functional in a slowly varying magnetic field. With such an explicit form for E_{xc}, the exchange-correlation potentials v_{xc} and $\mathbf{A}_{xc}$ can be calculated and the self-consistent equations (8)–(10) can be solved.[16] It is therefore no longer necessary to ignore the important effect of the magnetic field on the form of the exchange-correlation potential, as was the case, for example, in the paper by Wang, Grempel, and Prange.[17] We have now a rigorous foundation for incorporating the orbital effects of strong magnetic fields within the framework of DFT. Further extension of Eq. (18) to lower dimensionalities will be presented elsewhere.

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