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Current-density-functional theory for a non-relativistic electron gas in a strong magnetic field

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Abstract. A current-density-functional theory is formulated in terms of the physical gauge-invariant current density instead of the 'paramagnetic' or canonical current density used previously by Vignale and Rasolt. The appropriate energy functional is obtained by a procedure of 'constrained search'. An equivalent Hohenberg–Kohn theorem is also proved and Kohn–Sham equations are derived. The possibility of local approximations is discussed. As a by-product a generalized Ritz inequality for systems in a magnetic field is found.

1. Introduction

Density-functional theory based on the fundamental papers of Hohenberg and Kohn (1964) and Kohn and Sham (1965) has developed into a powerful tool for the study of electronic structure in condensed matter physics and quantum chemistry. The extension to the fully relavistic case was originally formulated by Rajagopal and Callaway (1973) (cf also Eschrig *et al* 1985). In this relativistic theory the external potential Φ and density n are replaced by analogous 4-vectors, the 4-potential $(\Phi/c, \boldsymbol{A})$ and 4-current density $(nc, \boldsymbol{j})$, respectively. However, the dependence on the current density $\boldsymbol{j}$ must be conserved also in the limiting case of a non-relativistic electron gas if a strong magnetic field $\boldsymbol{B} = \operatorname{curl} \boldsymbol{A}$ is applied. A corresponding current-density-functional theory was developed by Vignale and Rasolt (1987, 1988). A strange feature of their theory consists in the fact that they use a non-physical 'paramagnetic' current density $\boldsymbol{j}_{\mathrm{P}}$ which is not gauge invariant whereas in the relativistic formulation the physical gauge-invariant quantity $\boldsymbol{j}$ occurs in a natural way. In the Vignale–Rasolt theory gauge invariance must be ensured by a restricting condition on the choice of the exchange-correlation energy.

The aim of this paper is to present an alternative current-density-functional theory for non-relativistic particles in a magnetic field using the gauge-invariant current density $\boldsymbol{j}$. For the sake of simplicity the considerations start with a system of spinless particles obeying a many-particle Schrödinger equation. An appropriate energy functional is constructed in section 2 and the corresponding variational principle is established. A by-product of these developments is a modified Ritz principle for the ground state of a system in a magnetic field outlined in section 3. It consists in an inequality for the ground state energy which is stronger than the usual one. This new principle enables us to derive a Hohenberg–Kohn theorem for our system in

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presence of a magnetic field according to which the density n and current density j of the ground state uniquely determine the external fields E and B (the potentials Φ and A being determined up to an arbitrary gauge transformation). This is done in section 4. Section 5 sketches one of the possible ways to include the electron spin in the formalism by introducing a total current density comprising both orbital and spin contributions. Another approach, not developed in this paper, would be to maintain an orbital current density and to describe the influence of the spin with the help of spin densities (and associated spin current densities) as did Vignale and Rasolt in their paper. Finally, in section 6, Kohn–Sham equations are derived from the variational principle. They differ from those found in the formalism of Vignale and Rasolt by a simpler form of the effective potentials. A discussion of possible local approximations completes the paper.

2. Energy functional

Let us start with the Hamiltonian for a system of N spinless particles in an external magnetic field

$$H = \sum_{k=1}^{N}\left[\frac{1}{2m}\left(\frac{\hbar}{\mathrm{i}}\frac{\partial}{\partial r_k} + eA(r_k)\right)^2 - e\Phi(r_k)\right] + U \tag{1}$$

where A and Φ are external potentials and U denotes the interaction energy. The charge of the particles is $-e$. An arbitrary state of the many-body system is described by an antisymmetric wavefunction $\Psi(r_1 \ldots r_N)$. The corresponding particle density is

$$n = N\int \mathrm{d}^3 r_2 \cdots \mathrm{d}^3 r_N \, |\Psi|^2 \tag{2}$$

and the 'paramagnetic' or 'canonical' current density is performed with the operator of the canonical momentum

$$j_{\mathrm{P}} = \frac{N}{m}\mathrm{Re}\int \mathrm{d}^3 r_2 \cdots \mathrm{d}^3 r_N \, \Psi^* \frac{\hbar}{\mathrm{i}}\frac{\partial}{\partial r_1}\Psi. \tag{3}$$

As long as the vector potential is not fixed, the physical current density j belonging to a given state Ψ is completely undetermined. An arbitrary j can be obtained by an appropriate choice of the vector potential a

$$j = j_{\mathrm{P}}[\Psi] + \frac{en}{m}a. \tag{4}$$

This equation associates a vector potential $a[\Psi, j]$ to a given state Ψ and to a required current density j. In the following one has to distinguish between this trial a and the actual vector potential A. For stationary states the continuity equation requires

$$\partial j / \partial r = 0. \tag{5}$$

Using definitions (3) and (4) one can write down the identity

$$\langle\Psi|H|\Psi\rangle = \langle\Psi|\frac{1}{2m}\sum_k\left(-\hbar^2\frac{\partial^2}{\partial r_k^2} - e^2a^2(r_k)\right) + U|\Psi\rangle$$
$$+ e\int d^3r\ (jA - n\Phi) + \int d^3r\ \frac{e^2n}{2m}(A-a)^2. \tag{6}$$

The first term on the rhs depends on the state Ψ and the assumed current density j, but is independent of the actual fields A and Φ. Adopting the procedure of 'constrained search' of Levy (1979) one can define a new functional by minimizing this term with respect to Ψ for fixed $n(r)$ and $j(r)$

$$F[n,j] = \inf_{n,j}\langle\Psi|H_a|\Psi\rangle$$
$$H_a := -\frac{1}{2m}\sum_k\left(\hbar^2\frac{\partial^2}{\partial r^2{}_k} + e^2a^2(r_k)\right) + U. \tag{7}$$

Note that a depends on the state Ψ via (4). In the usual density-functional theory the Levy functional suffers from non-convexity. Lieb (1983) overcame this difficulty by admitting in the minimization not only pure states, but also mixed states described by a density operator ϱ. Similarly, one could also modify definition (7) by minimizing with respect to mixed states

$$F'[n,j] = \inf_{n,j}\mathrm{Tr}(H_a\varrho) \leqslant F[n,j]$$
$$j = j_P[\varrho] + \frac{en}{m}a. \tag{8}$$

For the sake of simplicity, however, we use the functional (7). Furthermore, we assume that there is a minimum state Ψ_M with an associated vector potential $a_M[r;n,j]$ such that

$$F[n,j] = \langle\Psi_M|H_{a_M}|\Psi_M\rangle. \tag{9}$$

It is shown in Appendix A that Ψ_M and a_M are not unique. In fact, one can carry out a gauge transformation of Ψ_M and a_M without changing the value of F provided that j satisfies the continuity condition (5). If (5) would not hold, any result for the expectation value in (7) could be generated by a simple gauge transformation and, therefore, a minimum would not exist. Thus the functional is only defined for current densities obeying equation (5).

Inserting the functional F in the identity (6) leads to the relation

$$E[n,j;\Phi,A] := F[n,j] + e\int d^3r\ [jA - n\Phi] \leqslant \langle\Psi|H|\Psi\rangle - \int d^3r\ \frac{ne^2}{2m}(A-a)^2. \tag{10}$$

This inequality holds for any Ψ possessing the given n. Note that the vector potential a depends on Ψ and is determined by (4). In the next step let us choose j such that the vector potential a_M in F equals the external potential A

$$a_M[n,j] = A \to j = j[n,A] = j_A. \tag{11}$$

The resulting current density is denoted by j_A and may be found from the stationarity condition

$$\delta E = E[j_A + \delta j] - E[j_A] = 0 \tag{12}$$

as shown in appendix B. Note that this condition does not provide us an absolute minimum of the functional E, as also discussed in appendix B . In fact, the functional E tends to minus infinity for infinitly growing $|j|$. Inserting the special choice (11) in the lhs of (10) and defining

$$\Psi_M \to \Psi[n, A] = \Psi_A \qquad \text{for } a_M \to A \tag{13}$$

one obtains

$$E[n, \Phi, A] = \langle \Psi_A | H_A | \Psi_A \rangle + e \int d^3r \, [j_A A - n\Phi] = \langle \Psi_A | H | \Psi_A \rangle. \tag{14}$$

In the last equality identity (6) is used with $a = A$. Thus, (10) becomes

$$\langle \Psi_A | H | \Psi_A \rangle \leqslant \langle \Psi | H | \Psi \rangle - \int d^3r \, \frac{ne^2}{2m} (A - a)^2 \tag{15}$$

where Ψ and Ψ_A have the same density n and a is now determined by

$$a = \frac{m}{ne} (j_A[n, A] - j_P[\Psi]). \tag{16}$$

Further, if one minimizes the lhs of (15) with respect to n

$$E_o[\Phi, A] := \min \langle \Psi_A | H | \Psi_A \rangle \tag{17}$$

and omits the negative term on the right-hand side of (15), one obtains

$$E_0 \leqslant \langle \Psi | H | \Psi \rangle \tag{18}$$

which holds for all Ψ. Thus, E_0 represents the ground-state energy, and the corresponding density n_0, current density $j_0 = j_A[n_0, A]$ and wave function $\Psi_0 = \Psi_A[n_0, A]$ also describe the ground state of the system. Summarizing the above considerations, one can conclude that the ground-state density n_0 and current density j_0 for a given 4-potential Φ, A satisfy the variational principle

$$\delta E[n, j; \Phi, A] = \delta \{ F[n, j] + e \int d^3r \, (jA - n\Phi) \} = 0. \tag{19}$$

Let us keep in mind, however, that according to the remark following (12) the ground state does not correspond to an absolute minimum of the functional E.

3. Generalized Ritz inequality

Now we consider relation (15) especially for the ground-state density

$$E_0 = \langle \Psi_0 | H | \Psi_0 \rangle \leqslant \langle \Psi | H | \Psi \rangle - \int \mathrm{d}^3 r \, \frac{e^2 n}{2m} (\boldsymbol{A} - \boldsymbol{a})^2. \tag{20}$$

The inequality holds for any Ψ possessing the same density $n = n_0$ as the ground state where $\boldsymbol{a}$ is connected with the ground state current $\boldsymbol{j}_0$ by

$$\boldsymbol{a} = \frac{m}{e n_0} (\boldsymbol{j}_0 - \boldsymbol{j}_{\mathrm{P}}[\Psi]) \qquad (n = n_0). \tag{21}$$

Moreover, it follows from (15) and (17) that (20) is also valid for an arbitrary Ψ with density $n \neq n_0$ and with $\boldsymbol{a}$ determined by

$$\boldsymbol{a} = \frac{m}{e n} (\boldsymbol{j}_{\mathrm{A}}[n, \boldsymbol{A}] - \boldsymbol{j}_{\mathrm{P}}[\Psi]) \qquad (n \neq n_0). \tag{22}$$

The inequality (20) is stronger than the ordinary Ritz principle

$$E_0 = \langle \Psi_0 | H | \Psi_0 \rangle \leqslant \langle \Psi | H | \Psi \rangle. \tag{23}$$

An obstacle for the application of (20), (22) is the fact that, in general, the functional $\boldsymbol{j}_{\mathrm{A}}$ will be unknown. Equality in (20) holds if Ψ and Ψ_0 differ only by the gauge

$$\Psi = \Psi_0 \exp\left(-\frac{\mathrm{i}e}{\hbar} \sum_k \chi(\boldsymbol{r}_k) \right). \tag{24}$$

Then

$$\begin{aligned} &\langle \Psi | \frac{1}{2m} \sum_k \left(\frac{\hbar}{\mathrm{i}} \frac{\partial}{\partial \boldsymbol{r}_k} + e\boldsymbol{A}(\boldsymbol{r}_k) \right)^2 | \Psi \rangle \\ &\quad = \langle \Psi_0 | \frac{1}{2m} \sum_k \left(\frac{\hbar}{\mathrm{i}} \frac{\partial}{\partial \boldsymbol{r}_k} + e\boldsymbol{A}(\boldsymbol{r}_k) - e\frac{\partial \chi}{\partial \boldsymbol{r}_k} \right)^2 | \Psi_0 \rangle \\ &\quad = \langle \Psi_0 | \frac{1}{2m} \sum_k \left(\frac{\hbar}{\mathrm{i}} \frac{\partial}{\partial \boldsymbol{r}_k} + e\boldsymbol{A}(\boldsymbol{r}_k) \right)^2 | \Psi_0 \rangle \\ &\qquad \times - e \int \mathrm{d}^3 r \, \boldsymbol{j}_0 \frac{\partial \chi}{\partial \boldsymbol{r}} + \int \mathrm{d}^3 r \, \frac{e^2 n_0}{2m} \left(\frac{\partial \chi}{\partial \boldsymbol{r}} \right)^2. \end{aligned} \tag{25}$$

The first term in the last line vanishes because

$$\int \mathrm{d}^3 r \, \boldsymbol{j}_0 \frac{\partial \chi}{\partial \boldsymbol{r}} = - \int \mathrm{d}^3 r \, \chi \frac{\partial \boldsymbol{j}_0}{\partial \boldsymbol{r}} = 0. \tag{26}$$

Therefore

$$\langle \Psi | H | \Psi \rangle = \langle \Psi_0 | H | \Psi_0 \rangle + \int \mathrm{d}^3 r \frac{e^2 n_0}{2m} \left(\frac{\partial \chi}{\partial \boldsymbol{r}} \right)^2 > E_0. \tag{27}$$

On the other hand, one has

$$j_P[\Psi] = j_P[\Psi_0] - \frac{en_0}{m}\frac{\partial \chi}{\partial r} = j_0 - \frac{en_0}{m}\left(A + \frac{\partial \chi}{\partial r}\right). \tag{28}$$

Insertion in (21) yields

$$a = A + \frac{\partial \chi}{\partial r}. \tag{29}$$

This relation together with (25) leads to

$$\langle \Psi | H | \Psi \rangle - \int d^3r \, \frac{e^2 n_0}{2m}(A - a)^2 = \langle \Psi_0 | H | \Psi_0 \rangle = E_0. \tag{30}$$

Thus, contrarily to the ordinary Ritz principle (23) the stronger inequality (20) reduces to an equality for all wave functions that differ from the ground state only by the gauge.

4. Hohenberg–Kohn theorem

The generalized Ritz principle enables us to prove a Hohenberg–Kohn theorem for a system subject to a magnetic field. The theorem asserts that the ground state density n_0 and current density j_0 uniquely determine the external fields E and B. The potentials Φ and A are determined up to the gauge. The proof can be given along the same lines as argued Vignale and Rasolt (1988). Suppose there were two different ground states Ψ_0, Ψ_0' belonging to different potentials Φ, A and Φ', A', respectively, but giving the same density n_0 and current density j_0. The states are assumed to differ by more than a gauge transformation. Then (20) becomes

$$E_0 = \langle \Psi_0 | H | \Psi_0 \rangle < \langle \Psi_0' | H | \Psi_0' \rangle - \int d^3r \, \frac{e^2 n_0}{2m}(A - A')^2. \tag{31}$$

The expectation value on the right-hand side may be transformed into

$$\langle \Psi_0' | H | \Psi_0' \rangle = \langle \Psi_0' | H' | \Psi_0' \rangle + e \int d^3r \, j_0(A - A') + \int d^3r n_0 \left[\frac{e^2}{2m}(A - A')^2 - e(\Phi - \Phi')\right]. \tag{32}$$

With the notation $E_0' = \langle \Psi_0' | H' | \Psi_0' \rangle$, equation (31) becomes

$$E_0 < E_0' + e \int d^3r \, [j_0(A - A') - n_0(\Phi - \Phi')]. \tag{33}$$

Interchanging the primed and unprimed quantities and summing the two inequalities, one obtains

$$E_0 + E_0' < E_0' + E_0. \tag{34}$$

This contradiction shows that the assumption at the beginning was wrong. It is excluded that physically different external fields lead to the same ground state density n_0 and current density j_0. Observe that the theorem is now proven for the gauge-invariant physical current density j_0 whereas Vignale and Rasolt derived it for the canonical current density j_P.

5. Particles with spin

The above considerations can easily be extended to particles with spin described by the Pauli equation. One has to add the term

$$\sum_{k=1}^{N} \frac{e\hbar}{2m} \sigma_k B \tag{35}$$

to the Hamiltonian (1). The wavefunction becomes a two-component quantity for each particle. Consequently, in the definitions (2) and (3) of n and j_P, Ψ^* has to be replaced by Ψ^+. Due to the spin magnetization M there is an additional contribution to the current density (4)

$$j = j_P + \frac{\partial}{\partial r} \times M + \frac{en}{m} a \tag{36}$$

with

$$M = \frac{N\hbar}{2m} \int d^3 r_2 \cdots d^3 r_N \; \Psi^+ \sigma_1 \Psi. \tag{37}$$

For stationary states the total current density satisfies again (5). The supplementary term with M does not contribute to the divergence. The identity (6) remains unchanged. The additional term occurring in the expectation value of H on the left-hand side

$$\langle \Psi | \sum_k \frac{e\hbar}{2m} \sigma_k B | \Psi \rangle = \int d^3 r \, eMB = e \int d^3 r \, M \left(\frac{\partial}{\partial r} \times A \right) = e \int d^3 r \, A \left(\frac{\partial}{\partial r} \times M \right) \tag{38}$$

is compensated by the spin contribution in the integral $e \int d^3 r \, jA$ on the right-hand side. All further considerations are based on the identity (6) and, therefore, remain valid. The only modification consists in the replacement

$$j_P[\Psi] \to j_{PM}[\Psi] := j_P + \frac{\partial}{\partial r} \times M. \tag{39}$$

Thus, the resulting energy functional $E[n, j, \Phi, A]$ (10) and the Hohenberg–Kohn theorem now involve the total current density (36) comprising both orbital and spin effects. However, despite the principal existence of this formalism, its practical applicability is questionable. The problem is to find sufficiently simple expressions for the energy functional.

An alternative and perhaps more promising way to incorporate spin effects in the formulation could be the use of spin-polarized densities or, more precisely, a density matrix $n_{ss'}$ and a corresponding current matrix $j_{ss'}$ defined analogously to (2) and (3). The extension of the above considerations to this case is straightforward and will not be written down here.

6. Kohn–Sham equations

In a manner that it analogous to the usual Kohn–Sham theory, the variational principle (19) can be transformed into effective one-particle equations. The functional F (7) has to be decomposed into

$$F[n,j] = F_0[n,j] + E_H[n] + E_{xc}[n,j]$$
$$F_0[n,j] = -\min_{n,j} \frac{1}{2m} \sum_k \langle \phi_k | \hbar^2 \frac{\partial^2}{\partial r^2} + e^2 a^2 | \phi_k \rangle \tag{40}$$

$$E_H = -\frac{e}{2} \int d^3r\, n\Phi_H = \frac{e^2}{8\pi\varepsilon_0} \int d^3r\, d^3r' \frac{nn'}{|r-r'|} \qquad \varepsilon_0 \Delta \Phi_H = en$$

where F_0 represents the functional of a non-interacting system expressed through one-particle states ϕ_k. The so-called Hartree energy E_H is the electrostatic energy of the mean charge distribution and E_{xc} comprises all correlation and exchange effects. The minimization in F_0 has to be carried out under the constraints

$$n = \sum_k \phi_k^+ \phi_k$$
$$j = \frac{1}{m} \mathrm{Re} \sum_k \phi_k^+ \frac{\hbar}{i} \frac{\partial}{\partial r} \phi_k + \frac{\partial}{\partial r} \times \frac{\hbar}{2m} \sum_k \phi_k^+ \sigma \phi_k + \frac{e}{m} an. \tag{41}$$

The last line shows that, due to the fixed j, the variation of ϕ_k is in general connected with a variation of a. In (41) the spin contribution is included according to section 5. The theory of spinless or spin- polarized particles is easily recovered by omitting all terms involving the spin operator.

Instead of minimizing F_0 under the constraints (41) and then varying n and j in the energy functional E according to (19), one can abbreviate the procedure by varying E with respect to ϕ_k and a without constraints (except normalization). Thus

$$\delta \left[E - \sum_k \varepsilon_k \langle \phi_k | \phi_k \rangle \right] = 0$$
$$E = F_0 + E_H + E_{xc} + e \int d^3r\, [jA - n\Phi] \tag{42}$$

for arbitrary variations of ϕ_k and a. The resulting expressions are

$$\delta F_0 = -\frac{1}{m} \mathrm{Re} \sum_k \int d^3r \left\{ \delta \phi_k^+ \left(\hbar^2 \frac{\partial^2}{\partial r^2} + e^2 a^2 \right) \phi_k - \frac{e^2 n}{m} \delta a a \right\}$$
$$\delta E_H = -\frac{e}{2} \int d^3r\, (\delta n \Phi_H + n \delta \Phi_H) = -e \int d^3r\, \delta n \Phi_H$$
$$\delta E_{xc} = \int d^3r \left(\delta n \frac{\delta E_{xc}}{\delta n} + \delta j \frac{\delta E_{xc}}{\delta j} \right) \tag{43}$$
$$\delta n = 2\mathrm{Re} \sum_k \delta \phi_k^+ \phi_k$$
$$\delta j = \frac{1}{m} \mathrm{Re} \sum_k \left(\delta \phi_k^+ \frac{\hbar}{i} \frac{\partial \phi_k}{\partial r} - \frac{\hbar}{i} \frac{\partial \delta \phi_k^+}{\partial r} \phi_k \right)$$
$$+ \frac{\partial}{\partial r} \times \frac{\hbar}{m} \mathrm{Re} \sum_k \delta \phi_k^+ \sigma \phi_k + \frac{en}{m} \delta a + \frac{e}{m} \delta n a.$$

Inserting these expressions in (42), performing some integrations by parts and then comparing all terms with $\delta\phi_k^+$ and with δa, respectively, one finds

$$\left\{\frac{1}{2m}\left(\frac{\hbar}{\mathrm{i}}\frac{\partial}{\partial \boldsymbol{r}}+e\boldsymbol{A}_{\mathrm{eff}}\right)^2+\frac{e\hbar}{2m}\sigma\boldsymbol{B}_{\mathrm{eff}}-e\Phi_{\mathrm{eff}}\right\}\phi_k=\varepsilon_k\phi_k$$

$$\boldsymbol{a}=\boldsymbol{A}_{\mathrm{eff}}:=\boldsymbol{A}+\boldsymbol{A}_{\mathrm{xc}} \qquad e\boldsymbol{A}_{\mathrm{xc}}=\frac{\delta E_{\mathrm{xc}}}{\delta \boldsymbol{j}}$$

$$\boldsymbol{B}_{\mathrm{eff}}:=\frac{\partial}{\partial \boldsymbol{r}}\times\boldsymbol{A}_{\mathrm{eff}}$$

$$\Phi_{\mathrm{eff}}:=\Phi+\Phi_{\mathrm{H}}+\Phi_{\mathrm{xc}} \qquad e\Phi_{\mathrm{xc}}=-\frac{\delta E_{\mathrm{xc}}}{\delta n}. \tag{44}$$

Formally, these Kohn–Sham equations look a little simpler than those obtained by Vignale and Rasolt, where $\boldsymbol{A}$ und $\boldsymbol{A}_{\mathrm{xc}}$ are also involved in the effective- potential term. A detailed comparison of both formalisms is difficult, however, because the definitions of the E_{xc} energies do not coincide completely. In (44) spin and orbital currents couple to the same effective magnetic field $\boldsymbol{A}_{\mathrm{eff}}$. Note, however, that in the present formulation only n and $\boldsymbol{j}$ have a physical meaning. The ϕ_k and, consequently, their spin and orbital magnetizations are only auxiliary functions without direct physical interpretation. This would be different in a formulation treating separately spin polarization and orbital currents.

7. Discussion

The above considerations show that in the case of the non-relativistic electron gas there exists a current-density formalism based on the physical current density $\boldsymbol{j}$ instead of the canonical or paramagnetic current density $\boldsymbol{j}_{\mathrm{P}}$. The question whether to use an energy functional depending on $\boldsymbol{j}$ or on $\boldsymbol{j}_{\mathrm{P}}$ is more a practical than a principal one. The usefulness of each of the different formalisms depends on the possibility of finding simple and reasonable approximations for the exchange-correlation energy E_{xc}. Nevertheless it is important to know that there is no principal discrepancy between a fully relativistic theory usually formulated in terms of the gauge-invariant current j^μ and the non-relativistic limit discussed in this paper. For practical purposes a local approximation of the energy functional E_{xc} is desirable. In order to find an appropriate energy density e_{xc},one has to consider the limiting case of a homogeneous electron gas at rest. Then e_{xc} is completely determined as a function of the density n and the magnetic field $\boldsymbol{B}$. By definition, this field is related to the current densities $\boldsymbol{j}$ and $\boldsymbol{j}_{\mathrm{P}}$ via the equations

$$\boldsymbol{j}=\boldsymbol{j}_{\mathrm{P}}+\frac{e}{m}\boldsymbol{A}n \qquad \frac{\partial}{\partial \boldsymbol{r}}\times\frac{\boldsymbol{j}}{n}=\frac{\partial}{\partial \boldsymbol{r}}\times\frac{\boldsymbol{j}_{\mathrm{P}}}{n}+\frac{e}{m}\boldsymbol{B}. \tag{45}$$

A local expression for the energy density using the current $\boldsymbol{j}$ or its derivatives is not conceivable in the homogeneous limit, since $\boldsymbol{j}$ then vanishes inside the system. Then (45) implies that $\boldsymbol{B}$ can be expressed by the canonical current $\boldsymbol{j}_{\mathrm{P}}$

$$\boldsymbol{B}=-\frac{m}{e}\frac{\partial}{\partial \boldsymbol{r}}\times\frac{\boldsymbol{j}_{\mathrm{P}}}{n}. \tag{46}$$

Now, if we consider a moving electron gas and want to desribe it by the same energy density $e_{xc}(n,B)$ as the gas at rest, we have to choose a local coordinate system moving together with the electron gas and, therefore, rotating with the angular velocity

$$\omega = \frac{1}{2}\frac{\partial}{\partial r} \times v = \frac{1}{2}\frac{\partial}{\partial r} \times \frac{j}{n}. \tag{47}$$

In a rotating system, however, there is an additional force, the Coriolis force. It has the same form as the Lorentz force and can therefore be taken into account in e_{xc} by the simple replacement

$$B \to B' = B - \frac{2m}{e}\omega = B - \frac{m}{e}\frac{\partial}{\partial r} \times \frac{j}{n} = -\frac{m}{e}\frac{\partial}{\partial r} \times \frac{j_P}{n}. \tag{48}$$

These considerations are in favour of the formalism developed by Vignale and Rasolt (1987, 1988). In many practical cases, however, the difference between j and j_P can expected to be small and the use of them to be equivalent. This will be true, in particular, for rapidly moving electrons which need a relativistic treatment.

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Appendix A. Gauge invariance of F

The functional F defined in (7) and (9) is invariant with respect to the gauge transformation

$$\Psi_M \to \Psi'_M = \Psi_M \exp\left\{-\frac{ie}{\hbar}\sum_k \chi(r_k)\right\} \qquad a_M \to a'_M = a_M + \frac{\partial \chi}{\partial r} \tag{A1}$$

where χ is an arbitrary function. Inserting the replacement (A1) in expression (9) yields

$$\begin{aligned}
&\langle \Psi'_M | \frac{1}{2m}\sum_k \left(-\hbar^2 \frac{\partial^2}{\partial r_k^2} - e^2 a'^2_M\right) | \Psi'_M \rangle \\
&\quad = \langle \Psi_M | \frac{1}{2m}\sum_k \left(-\hbar^2 \left(\frac{\partial}{\partial r_k} - \frac{ie}{\hbar}\frac{\partial \chi(r_k)}{\partial r_k}\right)^2\right. \\
&\qquad \left. - e^2\left(a_M(r_k) + \frac{\partial \chi(r_k)}{\partial r_k}\right)^2\right) | \Psi_M \rangle \\
&\quad = \langle \Psi_M | \frac{1}{2m}\sum_k \left\{\left(-\hbar^2 \frac{\partial^2}{\partial r_k^2} - e^2 a_M^2\right)\right. \\
&\qquad \left. - e\left[\frac{\hbar}{i}\frac{\partial}{\partial r_k} + e a_M(r_k), \frac{\partial \chi}{\partial r_k}\right]_+\right\} | \Psi_M \rangle \\
&\quad = \langle \Psi_M | \frac{1}{2m}\sum_k \left(-\hbar^2 \frac{\partial^2}{\partial r_k^2} - e^2 a_M^2\right) | \Psi_M \rangle - e\int d^3 r\, j(r)\frac{\partial \chi}{\partial r}.
\end{aligned} \tag{A2}$$

Integration by parts in the last term leads to

$$-\int \mathrm{d}^3r\, j\frac{\partial\chi}{\partial r} = \int \mathrm{d}^3r\, \chi\frac{\partial j}{\partial r} = 0. \tag{A3}$$

Thus, F is gauge invariant for all j satisfying the continuity equation (5). In the opposite case any value of (A2) and, consequently of the expectation value in F can be obtained by an appropriate choice of χ and a minimum does not exist.

Appendix B. Variation of F

F is obtained from a minimization of $\langle\Psi|H_a|\Psi\rangle$ under the constraints of given n and $j = j_P[\Psi] + en/\,ma$. The minimizing functions Ψ_M and a_M obey the variational condition

$$\delta\{\langle\Psi_M|H_{a_M}|\Psi_M\rangle - \int \mathrm{d}^3r\,[\mu(r)n(r) + \kappa(r)j(r)]\} = 0 \tag{B1}$$

where μ and κ are Lagrange parameters. The resulting equations are

$$\kappa = -ea_M$$
$$\left\{\frac{1}{2m}\sum_k\left[\left(\frac{\hbar}{\mathrm{i}}\frac{\partial}{\partial r_k} - \kappa(r_k)\right)^2 - \mu(r_k)\right] + U\right\}\Psi_M = 0. \tag{B2}$$

Here, μ and κ must be chosen such that n and j are obtained. Now the variation of F reads

$$\begin{aligned}\delta F &= \delta\langle\Psi_M|H_{a_M}|\Psi_M\rangle \\ &= 2\mathrm{Re}\langle\delta\Psi_M|H_{a_M}|\Psi_M\rangle - \int \mathrm{d}^3r\,\frac{e^2n}{m}a_M\delta a_M.\end{aligned} \tag{B3}$$

Inserting equation (B2) for Ψ_M leads to

$$\begin{aligned}\delta F &= 2\mathrm{Re}\langle\delta\Psi_M|\sum_k\left\{\mu(r_k) - \frac{1}{2m}\left[\frac{\hbar}{\mathrm{i}}\frac{\partial}{\partial r_k} + ea_M(r_k), ea_M(r_k)\right]_+\right\}|\Psi_M\rangle \\ &\quad - \int \mathrm{d}^3r\,\frac{e^2n}{m}a_M\delta a_M \\ &= \int \mathrm{d}^3r\,(\mu\delta n - ea_M\delta j).\end{aligned} \tag{B4}$$

For fixed n the variation (12) of E gives

$$\delta E = \int \mathrm{d}^3r\; e(A - a_M)\delta j. \tag{B5}$$

The variation of j is restricted by the continuity equation (5). Therefore, the stationarity condition leads to

$$\delta E = 0 \rightarrow A - a_M = \frac{\partial\chi}{\partial r} \tag{B6}$$

with an arbitrary χ. According to appendix A one can carry out an appropriate gauge transformation of Ψ_M and a_M to obtain $a_M = A$. The stationarity condition (12) does not correspond to an absolute minimum of E. This can easily be seen from (10) in connection with (4)

$$E[n,j,\Phi,A] \leqslant \langle\Psi|H|\Psi\rangle - \int d^3r\, \frac{m}{2n}\left(j - j_P[\Psi] - \frac{en}{m}A\right)^2. \qquad (B7)$$

For fixed Ψ the right-hand side tends to minus infinity if $|j| \to \infty$. The same must hold for the left-hand side. Thus, there is no absolute minimum of $E[n,j]$.

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