

Hartree-Fock theory of a current-carrying two-dimensional homogeneous electron gasH. Mera,^{1,2} P. Bokes,^{1,3} and R. W. Godby¹¹*Department of Physics, University of York, Heslington, York YO10 5DD, United Kingdom*²*Niels Bohr Institute and Nano-Science Center, Universitetsparken 5, DK-2100 Copenhagen, Denmark*³*Department of Physics, Faculty of Electrical Engineering and Information Technology, Slovak University of Technology, Ilkovičova 3, 812 19 Bratislava, Slovakia*

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State-of-the-art simulation tools for nonequilibrium quantum transport systems typically take the current-carrier occupations to be described in terms of equilibrium distribution functions characterized by two different electrochemical potentials, while for the description of electronic exchange and correlation, the local density approximation (LDA) to density functional theory is generally used. However, this involves an inconsistency because the LDA is based on the homogeneous electron gas *in equilibrium*, while the system is not in equilibrium and may be far from it. In this paper, we analyze this inconsistency by studying the interplay between nonequilibrium occupancies obtained from a maximum entropy approach and the Hartree-Fock exchange energy, single-particle spectrum and exchange hole, for the case of a two-dimensional homogeneous electron gas. The current dependence of the local exchange potential is also discussed. It is found that the single-particle spectrum and exchange hole have a significant dependence on the current, which has not been taken into account in practical calculations since it is not captured by the commonly used functionals. The exchange energy and the local exchange potential, however, are shown to change very little with respect to their equilibrium counterparts. The weak dependence of these quantities on the current is explained in terms of the symmetries of the exchange hole.

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I. INTRODUCTION

One of the uncontrollable approximations introduced in *ab initio* calculations of the transport properties of nanoscale conductors consists in the application of density functional theory (DFT), a ground state theory, outside the equilibrium regime. An immediate consequence of this approximation is that these properties are typically calculated at the level of the local density approximation (LDA), which is derived from the case of a homogeneous electron gas in equilibrium. The extent to which these approximations might affect the calculated electronic structure of the nonequilibrium systems remains largely unknown, and thus, a comparison between electronic properties calculated *exactly* for an admittedly highly idealized nonequilibrium system and those of the same system in equilibrium constitutes a particularly simple way of approaching and illustrating this problem.

In order to put these ideas into practice, we will consider a two-dimensional electron gas in equilibrium and in a *model* nonequilibrium state. To model a homogeneous electron gas outside equilibrium, we will assume that the nonequilibrium steady state of the two-dimensional electron gas can be characterized by the average total energy of the electron gas and by *different* average numbers of left- and right-moving electrons, and that the nonequilibrium steady state is given by the density matrix that maximizes the entropy of the electron gas with constraints on the above-mentioned averages.

Such an assumption leads in the noninteracting case to a momentum distribution characterized by *two Fermi hemispheres of different radii*; we take a pragmatic approach here and ignore the problems associated with the discontinuous character of this momentum distribution for the time being since we are interested in the question of how these current-

inducing constraints affect the electronic properties of the two-dimensional electron gas. Note that this type of momentum distribution is precisely of the form used in Landauer-Büttiker type of approaches and thus is familiar to the *ab initio* quantum transport community,¹⁻⁴ which constantly makes use of it. Similar momentum distributions are predicted by semiclassical transport theories in two-dimensional quantum point contacts.⁵ Alternatively, and perhaps also more physically, a current constraint may be used instead of the above-mentioned constraint to search for the nonequilibrium maximum entropy density matrix.⁶⁻⁹

To summarize, we will maximize the entropy of a two-dimensional homogeneous electron gas with constraints on the average numbers of left- and right-moving electrons to obtain a description of a steady state at the Hartree-Fock level of approximation, which can then be used to obtain the electronic structure of the gas in the presence of a current and to compare it with the usual approximations. The rest of the paper is organized as follows: In the next section, we discuss our theoretical approach to the problem and its numerical implementation. In Sec. III, we discuss the current dependence of the Hartree-Fock pair probability distribution, single-particle spectrum, total energy, and local exchange potential. We conclude with a discussion of the relevance of our work for practical calculations.

II. THEORY

In order to proceed, let us consider the entropy per unit area of the two-dimensional electron gas to be a functional of the electronic occupancies given by¹⁰

$$S[f(\mathbf{k})] = - \int_{\mathfrak{R}^2} \frac{d^2\mathbf{k}}{2\pi^2} \{f(\mathbf{k}) \ln f(\mathbf{k}) + [1 - f(\mathbf{k})] \ln [1 - f(\mathbf{k})]\}. \quad (1)$$

The electronic occupancies are written as

$$f(\mathbf{k}) = \begin{cases} f_L(\mathbf{k}) & \text{if } k_x < 0 \\ f_R(\mathbf{k}) & \text{if } k_x > 0, \end{cases}$$

where $\mathbf{k}=(k_x, k_y)$ and $f_{L/R}$ are the occupation functions to be varied independently in order to maximize Eq. (1) with constraints on the average total energy per unit area and different average numbers of left- and right-moving particles per unit area. In our model, the two-dimensional electron gas is assumed to be embedded between two reservoirs characterized by two different electrochemical potentials. The reservoirs effectively constrain the numbers of left- and right-moving states to be different in the gas, and hence, we add this constraint to our maximization procedure. We represent our knowledge about the system's constituents and their interactions by the many-electron interacting Hamiltonian together with the Hartree-Fock (i.e., exchange-only) approximation to the many-body wave function. In such a model, at zero or nonzero temperatures, the expectation value of the Hamiltonian is given by

$$\langle E \rangle = 2 \int_{\mathfrak{R}^2} \frac{d^2\mathbf{k}}{(2\pi)^2} f(\mathbf{k}) \frac{k^2}{2} - \int_{\mathfrak{R}^2} \frac{d^2\mathbf{k}'}{(2\pi)^2} \int_{\mathfrak{R}^2} \frac{d^2\mathbf{k}}{(2\pi)^2} f(\mathbf{k}) f(\mathbf{k}') v(\mathbf{k}, \mathbf{k}'), \quad (2)$$

where $v(\mathbf{k}, \mathbf{k}') = 2\pi/|\mathbf{k} - \mathbf{k}'|$ is the Fourier transform of the Coulomb interaction in two dimensions. The number of left- and right-moving electrons per unit area can be written as

$$n_{L(R)} = \frac{2}{(2\pi)^2} \int_{k_x < (>) 0} d^2\mathbf{k} f_{L(R)}(\mathbf{k}). \quad (3)$$

In order to maximize the entropy functional with respect to $f_{L/R}$ subject to the above-mentioned constraints, we use the method of Lagrange multipliers and consider the auxiliary functional

$$\mathcal{L}[f(\mathbf{k})] = S - \beta(\langle E \rangle - \mu_L n_L - \mu_R n_R), \quad (4)$$

together with the extremal condition

$$\frac{\delta \mathcal{L}}{\delta f_{L/R}} = 0. \quad (5)$$

A straightforward calculation shows that the occupation functions that maximize the entropy functional with constraints in the above-mentioned averages are given by

$$f_{L,R}(\mathbf{k}) = \frac{1}{1 + \exp\{\beta[k^2/2 + \epsilon_x(\mathbf{k}) - \mu_{L,R}]\}}, \quad (6)$$

where

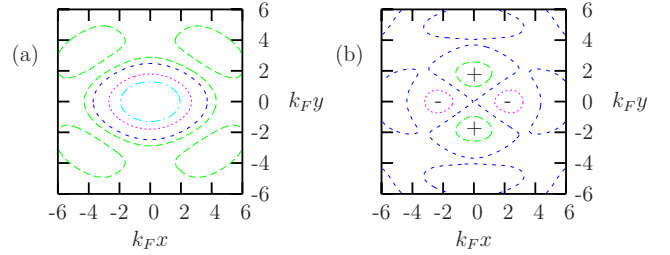


FIG. 1. (Color online) (a) Pair distribution function for like spins in the nonequilibrium regime ($n_L/n_R=0.5$). In the nonequilibrium regime, the exchange hole is elongated along the direction of the current. The contours are at $g=0.5, 0.75, 0.9$, and 0.95 . (b) Difference between the equilibrium and nonequilibrium holes, Δg (see text). The contours are drawn at 0.1 (dashed), -0.1 (dotted), and 0 (dotted). Δg is oscillating, integrates to zero, and has a marked antisymmetric character. Thus, the current dependence of the local exchange potential and exchange energy is expected to be weak.

$$\epsilon_x(\mathbf{k}) = - \frac{1}{(2\pi)^2} \left[\int_{k_x < 0} d^2\mathbf{k}' f_L(\mathbf{k}') v(\mathbf{k}, \mathbf{k}') + \int_{k_x > 0} d^2\mathbf{k}' f_R(\mathbf{k}') v(\mathbf{k}, \mathbf{k}') \right], \quad (7)$$

i.e., the occupations that maximize the entropy are similar to those of the Landauer-Büttiker approach but with a modified exchange part of the spectrum. In the calculation, we fix the ratio n_L/n_R that together with the charge neutrality condition $n_L + n_R = 1/\pi r_s^2$ completely determines both n_L and n_R . With the equilibrium spectrum as a trial $\epsilon_x(\mathbf{k})$, we solve Eq. (3) for μ_L and μ_R . With these values of $\mu_{L,R}$, a new spectrum is constructed using Eq. (7), and the iteration is completed and subsequently repeated until the input and output spectra are identical to each other within the desired tolerance. All the results presented here are obtained in the $\beta \rightarrow \infty$ limit, where our approach is equivalent to that of Hershfield¹¹ in the Hartree-Fock approximation.¹⁴ Once the self-consistent spectrum and occupation factors are obtained, other quantities such as the exchange energy and exchange hole can be easily obtained. From these, we can study how the local exchange potential of the electron gas depends on the current density.¹⁵

III. RESULTS

A. Hartree-Fock pair distribution function

Let us begin by discussing the current dependence of the Hartree-Fock pair distribution function for spinlike electrons, which is given by

$$g(\mathbf{r}, \mathbf{r}') = 1 - \left| \frac{1}{n} \int \frac{d^2\mathbf{k}}{(2\pi)^2} \exp[-i\mathbf{k} \cdot (\mathbf{r} - \mathbf{r}')] f(\mathbf{k}) \right|^2 \quad (8)$$

and shown for $n_L/n_R=0.5$ in Fig. 1(a). For $n_L=n_R$, g is spherically symmetric, while for $n_L \neq n_R$ is elongated in the direction of the current. Similar phenomenology has been reported previously by Skudlarski and Vignale for the three-

dimensional electron gas in the presence of a magnetic field,¹² where the exchange hole is elongated in the direction of the field. In Ref. 12, the elongation arises from the change of occupancies associated with the Zeeman splitting due to the externally applied magnetic field. In the present case, the elongation of the hole can be understood in terms of the change in the electronic occupancies that results from our constrained maximization of the entropy functional. In both cases, the elongation of the hole is the result of the change in the polarizability induced by the change in the occupancies.¹² As argued in Ref. 12, when correlations are included the isotropy of the Coulomb interaction will bring the shape of the pair-probability distribution back to a more spherical form and, thus, closer to equilibrium.

Note that the difference between the equilibrium and non-equilibrium exchange holes, $\Delta g = g_{eq} - g_{neq}$, shown in Fig. 1(b), has a strong antisymmetric character; i.e., defining $\mathbf{R} = \mathbf{r} - \mathbf{r}' = (X, Y)$, then $\Delta g(X, Y) \sim -\Delta g(Y, X)$. We shall return to this point later when discussing the weak dependence of the exchange energy on the current density.

B. Single-particle spectrum

Figure 2 shows the self-consistent single-particle energy spectrum. Figure 2(a) shows the total (kinetic+exchange) spectrum, while in Fig. 2(b), we plotted only its exchange part on the $k_y=0$ line as given by Eq. (7) for $n_L=n_R$ and $n_L/n_R=0.5$.

The combined effect of the constraints and the exchange interaction shifts the spectrum toward higher values of k_x . Note also that, when compared to the equilibrium spectrum, the minimum of the nonequilibrium spectrum is less negative. Hence, we expect the total nonequilibrium exchange energy to increase with respect to the equilibrium one. Note that the constraints alter the total kinetic energy of the system, but do not change the kinetic contribution to the single-particle spectrum, since this contribution does not depend on the electronic occupancies. Hence, the changes in the single-particle spectrum are entirely due to the exchange interaction, which raises (lowers) the single-particle energy of electrons with $k_x < 0$ ($k_x > 0$). The anomalous behavior in the $k_x=0$ plane inherited from the discontinuous character of the maximum entropy momentum distribution can be seen clearly in Fig. 2(a), between μ_L and μ_R .

The interplay between nonequilibrium occupancies and the single-particle spectrum observed here is just a consequence of the orbital dependence of the Fock operator and will also be seen in any practical calculation that combines a nonequilibrium theory such as the Landauer-Büttiker approach or the Keldysh-nonequilibrium Green's function (NEGF) formalism, with an *orbital-dependent* description of the interactions between the electrons, such as the Hartree-Fock approximation.

We would like to point out that practical implementations of NEGF formalism typically take the electronic structure of the leads to be that of the equilibrium system (see Ref. 13 and references therein),¹⁶ and hence, the dependence of the single-particle spectrum on the nonequilibrium current (and vice versa) is commonly ignored. The validity of this ap-

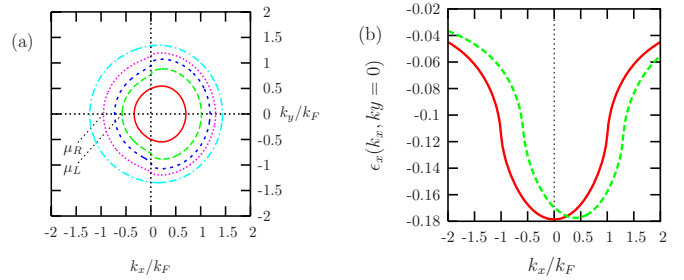


FIG. 2. (Color online) (a) Contour plots of the total single-particle energy spectrum of the model nonequilibrium electron gas for $n_L/n_R=0.25$ and $r_s=4$. The contours corresponding to $\mu_L=-1.110^{-2}$ a.u. and -9.210^{-2} a.u. are labeled. The other contours shown correspond to $(\mu_L+\mu_R)/2$ (short dashes), $\mu_L-0.510^{-2}$ (a.u.) (solid), and $\mu_R+0.510^{-2}$ (a.u.) (dot-dashed). (b) Exchange contribution to the single-particle energy spectrum, $\epsilon_x(\mathbf{k})$, evaluated on the $k_y=0$ line calculated for $n_L=n_R$ (solid) and $n_L/n_R=0.25$ (dashed). The main effect of the nonequilibrium constraints used in our variational approach is to shift the exchange part of the single-particle spectrum toward higher values of k_x .

proximation is geometry dependent: it works in quantum point contact geometries, while it does not in planar electrode geometries at high currents. As a consequence, under the “noninteracting equilibrium lead approximation,” the distribution of incoming electrons would be current independent, while, as this example shows,¹⁷ the unavoidable presence of interactions in the leads induces a current dependence in the nonequilibrium occupancies through the exchange part of the single-particle spectrum. Unless the geometry is adequately chosen, the distribution of incoming electrons will be that of a *nonequilibrium* lead such as ours.

In order to capture this subtle effect within the NEGF type of approaches, the coupling self-energy needs to be obtained from the lead's Green's function, which includes the necessary current dependence of the Fock operator. This can be achieved within a self-consistent scheme where not only the central region but also the leads' Green's functions are calculated self-consistently, as we have discussed in Ref. 13, with the nonlocal Fock operator being used within the whole system (electrodes and the central region).

C. Total energy

Once the self-consistent single-particle spectrum is calculated, the total exchange energy E_x can be obtained from the second term in the right hand side of Eq. (2). Figure 3 shows the dependence of the r_s -invariant quantity $-E_x/E_x^{eq}$ on $(1-n_L/n_R)$. For $n_L/n_R=0.25$, the exchange energy deviates by about 1%–2% from its equilibrium value. We also see that, even though the non-self-consistent results provide a good estimate to the self-consistent ones, full self-consistency is needed in the nonequilibrium case, even for a homogeneous gas. The error bars in the self-consistent results are estimated by comparing the exact exchange energy in equilibrium with the exchange energy obtained from our code for $n_L=n_R$ and different values of r_s . Therefore, the exchange energy depends on the current density, but this dependence is extremely weak in our model system. One

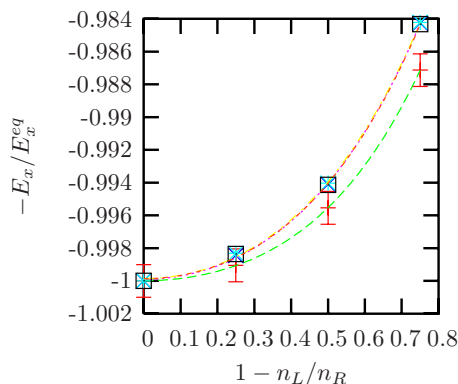


FIG. 3. (Color online) Exchange energy (in units of the equilibrium exchange energy) versus $1 - n_L/n_R$. In equilibrium, $1 - n_L/n_R = 0$. The dashed line shows the self-consistent results with estimated error bars. The non-self-consistent results are also shown with points calculated for different values of r_s , showing that the exchange energy scales with r_s as $1/r_s$. The lines are fits to parabolic functions.

could now proceed to calculate this current density explicitly and work out a current dependent local density approximation from the dependence of E_x on the current density. However, the weak dependence of the exchange energy on the current density deduced from Fig. 3 means that the current dependence of the local exchange functional is also very weak, and the changes it will induce in the associated LDA–Kohn-Sham effective potential will be well within the error bar of the LDA itself.

D. Local exchange potential

The weak dependence of the local exchange potential on the current density can be seen clearly in terms of the symmetries of the exchange hole. Consider the expression for Slater’s exchange potential, v_x^s , in terms of the Hartree-Fock pair distribution function:

$$v_x^s(\mathbf{r}) = \int d^2\mathbf{r}' \frac{[g(\mathbf{r} - \mathbf{r}') - 1]}{|\mathbf{r} - \mathbf{r}'|} n(\mathbf{r}'), \quad (9)$$

where $n(\mathbf{r}')$ is the electron density and $g(\mathbf{r} - \mathbf{r}')$ is the exchange hole. Then, the difference between equilibrium and nonequilibrium exchange potentials is, for our homogeneous system, given by

$$\Delta v_x^s \int d^2\mathbf{R} \frac{\Delta g(\mathbf{R})}{|\mathbf{R}|}, \quad (10)$$

where \mathbf{R} and Δg are defined as above. From Eq. (10), it follows that

$$\Delta g(X, Y) = -\Delta g(Y, X) \Rightarrow \Delta v_x^s = 0, \quad (11)$$

and hence, only the symmetric part of $\Delta g(X, Y)$ contributes to the deviation of exchange potential with respect to its equilibrium value. Note that $\Delta g(X, Y)$ is an oscillatory function that integrates to zero, which also has a marked antisymmetric character shown in Fig. 1(b). This explains the weak dependence of E_x and v_x on the current density.

IV. CONCLUSIONS

In conclusion, we have maximized the entropy of a two-dimensional homogeneous electron gas with constraints on the average total energy and average numbers of left- and right-moving electrons to obtain a simplified description of the steady state within the Hartree-Fock approximation. Our results show that both the single-particle spectrum and the exchange hole depend significantly on the current density, while averaged quantities such as the local exchange potential or the exchange energy do not. Our results provide support to the idea that within a static-DFT approach to quantum transport problems, nonequilibrium corrections to the exchange-correlation functional—at least at the exchange-only level—may be neglected without substantial loss of accuracy.

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¹⁴That is, to find the Slater determinant that minimizes the expectation value of the effective Hamiltonian $\hat{F} = \hat{H}_{HF} - \mu_L \hat{N}_R - \mu_L \hat{N}_L$.

¹⁵For two Fermi hemispheres of radii k_L and k_R , the noninteracting electronic current is related to these densities by the expression $j = \frac{2}{3\sqrt{2}\pi}(n_L + n_R)(n_R^3 - n_L^3)$.

¹⁶In particular, Eq. (22) of Ref. 13 applies also to our considerations here, where $\delta H_{L/R}$ contains contributions from the Fock operator that do not vanish for any finite length of the central region.

¹⁷We may view our two-dimensional electron gas as a simple model of one of the leads to which the nanoscale conductor is attached.