Two-Dimensional Electron Gas: Correlation Energy versus Density and Spin Polarization

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ABSTRACT: We propose a simple analytic representation of the correlation energy ϵ_c for the two-dimensional electron gas, as a function of the density parameter r_s and the spin polarization ζ. This new parametrization includes most of the known high- and low-density limits and fits our new fixed-node diffusion Monte Carlo simulations, performed for a wide range of electron densities ($1 \leq r_s \leq 40$) and spin-polarization states ($0 \leq \zeta \leq 1$). In this way we provide a reliable local-spin-density energy functional for two-dimensional systems. The corresponding correlation potential is discussed and compared with previous models.   © 2002 Wiley Periodicals, Inc. Int J Quantum Chem 91: 126–130, 2003

Key words: correlation energy; electron density; Monte Carlo simulation; spin polarization; two-dimensional electron gas

Introduction

The ideal two-dimensional electron gas (2DEG) is a simple model in which N strictly 2D electrons are confined in a square of surface S (periodically repeated in space) and interact via a $1/r$ potential within a uniform, rigid neutralizing background. When studying this model, one is usually interested in its macroscopic properties, that is, the thermodynamic limit ($N, S \rightarrow \infty$ keeping $n = N/S$ constant) of its extensive physical quantities per particle. Two parameters are enough to define the zero-temperature phase diagram of the 2D electron gas, namely, the density parameter $r_s = 1/\sqrt{\pi n}a_B$ (where n is the density and a_B the Bohr radius) and the spin polarization $\zeta = (n_\uparrow - n_\downarrow)/n$, where $n_{\uparrow(\downarrow)}$ is the density of spin-up (-down) electrons.

The model itself is interesting, because it can provide information about electrons confined in two dimensions realized in semiconductor heterostructures [1]. Moreover, just like the three-dimen-

sional case, the correlation energy of the 2D electron gas as a function of density r_s and spin polarization ζ provides the local spin density (LSD) energy functional for density functional calculations of 2D systems. Currently, 2D LSD functionals are based on parametrized diffusion quantum Monte Carlo (DMC) data [2] at $\zeta = 0$ and $\zeta = 1$ [3–6]. At intermediate spin polarizations, $0 < \zeta < 1$, an exchange-like interpolation is often used [3].

We have recently presented new DMC simulations for a wide range of electron densities r_s and spin polarizations ζ [7]. The direct DMC calculation of the ζ dependence is new and provides a reliable basis for building an LSD energy functional for 2D systems. In this work we present and discuss an accurate parametrization of these new data as a function of r_s and ζ. This new parametrization accurately reproduces the ζ dependence of the DMC data and includes most of the known high- and low-density limits. We also compare the corresponding correlation potential to previous approximations, finding significant discrepancies at $\zeta \neq 0$.

Hartree atomic units are used throughout this work.

Diffusion Monte Carlo Data

Our calculations use standard fixed-node diffusion Monte Carlo (FN-DMC) [8], which projects the lowest-energy eigenstate Φ of the many-body Hamiltonian with the boundary condition that Φ vanishes at the nodes of a trial function Ψ. Details of the simulation are similar to those of Ref. [9]; further details can be found in Ref. [7]. For each of the densities corresponding to $r_s = 1, 2, 5, 10$ we have considered about 20 values of N and 10–12 polarizations ζ. For the densities $r_s = 20$ and 30 we have used the data of Ref. [9]. We have also computed the energy at $r_s = 40$ for $\zeta = 1$. To estimate the difference Δ between the energy $\epsilon_N(r_s, \zeta)$ of the finite system and its thermodynamic limit $\epsilon(r_s, \zeta)$ we adopted a new strategy. Rather than a separate size extrapolation for each density based on variational energies [2, 10, 11], we performed a global fit directly based on FN-DMC energies, which exploits two physically motivated ingredients: (1) the Fermi-liquid–like size correction [12]

$$\Delta(r_s, \zeta, N) = \epsilon_N(r_s, \zeta) - \epsilon(r_s, \zeta)$$
$$= \delta(1 + \lambda\zeta^2)[t_N(r_s, \zeta) - t_s(r_s, \zeta)]$$
$$- (\eta + \gamma\zeta^2)/N \quad (1)$$

(t_N and t_s are the Fermi energies of N and ∞ particles, respectively, and δ, λ, η, and γ are r_s-dependent parameters); (2) an analytic expression for $\epsilon(r_s, \zeta)$, detailed in the next section, which involves 12 more free parameters.

The only uncontrolled source of error, the fixed-node approximation, depends on the nodal structure of Ψ. We choose a Slater–Jastrow trial function with plane waves (PW) as single orbitals. However, within the fixed-node approximation, better results are obtained with backflow (BF) correlations in the wavefunction [11]. Because simulations with the BF wavefunction are considerably more demanding than with PW determinants, we calculated BF energies only for $\zeta = 0$, $N = 58$ and $\zeta = 1$, $N = 57$ for each density. For other values of N and ζ, the effect of BF is estimated as a quadratic interpolation in ζ and appended to PW energies, under the further assumption that the size dependence be the same for BF and PW [7].

Analytic Model for the Correlation Energy

In this section we present our parametrization of the correlation energy of the 2D gas as a function of r_s and ζ. We first discuss the ζ dependence at a given fixed density, then our choice for the r_s dependence is presented, and finally we impose the exact high- and low-density limits to our functional form.

SPIN-POLARIZATION DEPENDENCE

We first noticed that, for $r_s \gtrsim 5$, the ζ dependence of the exchange-correlation energy $\epsilon_{xc} = \epsilon - t_s$ of our DMC data is accurately described by a biquadratic form, $c_0(r_s) + c_1(r_s)\zeta^2 + c_2(r_s)\zeta^4$ (see also Ref. [9]). On the other hand, the known high-density limit [13],

$$\epsilon_{xc}(r_s \to 0, \zeta) = \epsilon_x(r_s, \zeta) + a_0(\zeta)$$
$$+ b_0(\zeta)r_s \ln r_s + O(r_s), \quad (2)$$

contains nonnegligible contributions from higher powers of ζ: the dominating exchange term ϵ_x goes like $(1 + \zeta)^{3/2} + (1 - \zeta)^{3/2}$, and the constant term $a_0(\zeta)$ is well fitted by an eighth-degree polynomial function of ζ [13]. Because we want to interpolate the energy between high and low density, we choose a functional form that quenches the contri-

butions to ϵ_x beyond fourth order in ζ as r_s increases,

$$\epsilon_c(r_s\zeta) = (e^{-\beta r_s} - 1)\epsilon_x^{(6)}(r_s, \zeta) + \alpha_0(r_s)$$
$$+ \alpha_1(r_s)\zeta^2 + \alpha_2(r_s)\zeta^4, \quad (3)$$

where

$$\epsilon_x^{(6)}(r_s, \zeta) = \epsilon_x(r_s, \zeta) - (2 + \tfrac{3}{4}\zeta^2 + \tfrac{3}{64}\zeta^4)\epsilon_x(r_s, 0)/2$$

is the Taylor expansion of ϵ_x beyond fourth order in ζ. Because the first term on the right side of Eq. (3) contains power 6 and higher of ζ, it immediately identifies the function $\alpha_0(r_s)$ as the correlation energy at zero polarization,

$$\alpha_0(r_s) = \epsilon_c(r_s, 0).$$

Furthermore,

$$\alpha_1(r_s) = 2 \frac{\partial^2}{\partial \zeta^2} \epsilon_c(r_s, \zeta)\big|_{\zeta=0}$$

gives the spin stiffness, and

$$\alpha_2(r_s) = 24 \frac{\partial^4}{\partial \zeta^4} \epsilon_c(r_s, \zeta)\big|_{\zeta=0}.$$

DENSITY DEPENDENCE

We have now to fix the r_s dependence of the functions α_i. We generalize the Perdew and Wang [14] form (designed for the three-dimensional gas) to the 2D case as follows

$$\alpha_i(r_s) = A_i + (B_i r_s + C_i r_s^2 + D_i r_s^3)$$
$$\times \ln\left(1 + \frac{1}{E_i r_s + F_i r_s^{3/2} + G_i r_s^2 + H_i r_s^3}\right). \quad (4)$$

This form possesses the small- and large-r_s expansions:

$$\alpha_i(r_s \to 0) = A_i - B_i r_s \ln r_s + O(r_s) \quad (5)$$

$$\alpha_i(r_s \to \infty) = A_i + \frac{D_i}{H_i} + \left(\frac{C_i}{H_i} - \frac{D_i G_i}{H_i^2}\right)$$
$$\times \frac{1}{r_s} - \frac{D_i F_i}{H_i^2} \frac{1}{r_s^{3/2}} + O\left(\frac{1}{r_s^2}\right), \quad (6)$$

and it thus has the correct high- and low-density behavior [13, 15], provided that the constraint $A_i + D_i/H_i = 0$ is imposed.

EXACT LIMITS

Our $\epsilon_c(r_s, \zeta)$ has the correct functional form for small and large r_s; it is now straightforward to impose most of the known quantitative constraints. We constrain our $\epsilon_c(r_s, \zeta)$ to fulfill: (1) the requirement that the exact values [13, 16] of $a_0(\zeta)$ and $b_0(\zeta)$ at $\zeta = 0$ and $\zeta = 1$ in the small-r_s expansion of Eq. (2) are recovered, which implies

$$A_0 = -0.1925 \quad (7)$$

$$B_0 = \frac{\sqrt{2}}{3\pi}(10 - 3\pi) \quad (8)$$

$$A_0 + A_1 + A_2 + a_x \beta \mathscr{F}(1) = -0.039075 \quad (9)$$

$$B_0 + B_1 + B_2 = \frac{10 - 3\pi}{12\pi}, \quad (10)$$

where

$$\mathscr{F}(\zeta) = (1 + \zeta)^{3/2} + (1 - \zeta)^{3/2} - (2 + \tfrac{3}{4}\zeta^2 + \tfrac{3}{64}\zeta^4)$$
$$(11)$$

$$a_x = \frac{4}{3\pi\sqrt{2}}; \quad (12)$$

(2) the requirement that the total energy $\epsilon(r_s, \zeta)$ be independent of ζ for $r_s \to \infty$ up to order $O(r_s^{-2})$, thus recovering the low-density behavior $\epsilon \to -m/r_s + n/r_s^{3/2} + O(r_s^{-2})$ [15] with positive m and n independent of ζ. We thus have

$$A_i + \frac{D_i}{H_i} = 0 \quad (13)$$

$$\frac{C_1}{H_1} - \frac{D_1 G_1}{H_1^2} = \frac{3}{4} a_x \quad (14)$$

$$\frac{C_2}{H_2} - \frac{D_2 G_2}{H_2^2} = \frac{3}{64} a_x \quad (15)$$

$$F_1 = F_2 = 0. \quad (16)$$

TABLE I
Optimal-fit parameters for the correlation energy, as parametrized in Eqs. (3) and (4).

	$i = 0$	$i = 1$	$i = 2$
A_i	-0.1925[a]	0.117331[a]	0.0234188[a]
B_i	0.0863136[a]	-3.394×10^{-2}	-0.037093[a]
C_i	0.057234	-7.66765×10^{-3} [a]	0.0163618[a]
E_i	1.0022	0.4133	1.424301
F_i	-0.02069	0[a]	0[a]
G_i	0.340	6.68467×10^{-2}	0[a]
H_i	1.747×10^{-2}	7.799×10^{-4}	1.163099
β	1.3386		

[a] Values obtained from exact conditions. The parameter $D_i = -A_i H_i$ is not listed (see text).

We also fixed A_1 according to the high-density limit of the spin susceptibility [13, 16], and $G_2 = 0$ because it turned out to be an irrelevant parameter in our fitting procedure. In this way, we have built an analytic model that interpolates between the exact high- and low-density limits and has 12 free parameters to be fixed by a best fit to our diffusion Monte Carlo data. We then perform a global fit (r_s, ζ, N), which also includes the infinite-size extrapolation of Eq. (1), to our data set (122 data for $1 \leq r_s \leq 40$, $0 \leq \zeta \leq 1$, and $21 \leq N \leq 114$). In this way we fix the values of 36 free parameters, 24 of which disappear from the final analytic expression of ϵ_c because they concern only the $N \rightarrow \infty$ extrapolation. This fit yields a reduced χ^2 of 3.8. The optimal values of the parameters that yield the model for ϵ_c of the infinite system are reported in Table I.

LSD Correlation Potential

The 2D LSD correlation potential μ_c^σ for electrons of spin σ is given by

$$\mu_c^\sigma(r_s, \zeta) = \frac{\partial[n\epsilon_c(r_s, \zeta)]}{\partial n_\sigma}$$

$$= \epsilon_c(r_s, \zeta) - \frac{r_s}{2} \frac{\partial \epsilon_c(r_s, \zeta)}{\partial r_s}$$

$$- (\zeta - \text{sgn } \sigma) \frac{\partial \epsilon_c(r_s, \zeta)}{\partial \zeta}, \quad (17)$$

where sgn σ is $+1$ for spin-$\uparrow$ electrons and -1 for spin-$\downarrow$ electrons. From our parametrization of $\epsilon_c(r_s, \zeta)$ we get:

$$\frac{\partial \epsilon_c(r_s, \zeta)}{\partial r_s} = a_x \mathscr{F}(\zeta) \frac{[e^{-\beta r_s}(1 + \beta r_s) - 1]}{r_s^2}$$

$$+ \alpha_0'(r_s) + \alpha_1'(r_s)\zeta^2 + \alpha_2'(r_s)\zeta^4, \quad (18)$$

where $\mathscr{F}(\zeta)$ and the constant a_x are given by Eqs. (11) and (12), respectively, and

$$\alpha_i'(r_s) = \frac{d\alpha_i}{dr_s} = (B_i + 2C_i r_s + 3D_i r_s^2)$$

$$\times \ln\left[1 + \frac{1}{f_i(r_s)}\right] - \frac{(B_i + C_i r_s^2 + D_i r_s^3) f_i'(r_s)}{f_i(r_s)[f_i(r_s) + 1]} \quad (19)$$

$$f_i(r_s) = E_i r_s + F_i r_s^{3/2} + G_i r_s^2 + H_i r_s^3 \quad (20)$$

$$f_i'(r_s) = E_i + \frac{3}{2} F_i r_s^{1/2} + 2G_i r_s + 3H_i r_s^2. \quad (21)$$

The derivative with respect to ζ is simply

$$\frac{\partial \epsilon_c(r_s, \zeta)}{\partial \zeta} = \frac{a_x}{r_s} (1 - e^{-\beta r_s})\mathscr{F}'(\zeta)$$

$$+ 2\alpha_1(r_s)\zeta + 4\alpha_2(r_s)\zeta^3 \quad (22)$$

$$\mathscr{F}'(\zeta) = \frac{3}{2}(\sqrt{1 + \zeta} - \sqrt{1 - \zeta}) - \frac{3}{2}\zeta - \frac{3}{16}\zeta^3. \quad (23)$$

It is interesting to compare our correlation potential with the approximations used in previous LSD calculations in two dimensions. The most used 2D LSD functional is the one given by Tanatar and Ceperley [2], who performed diffusion Monte Carlo simulations at $\zeta = 0$ and $\zeta = 1$ and gave an analytic fit of the corresponding correlation energies. For the ζ dependence, many authors [3] used the exchange-like approximation

$$\epsilon_c(r_s, \zeta) = \epsilon_c(r_s, 0) + \frac{[(1 + \zeta)^{3/2} + (1 - \zeta)^{3/2} - 2]}{2^{3/2} - 2}$$

$$\times [\epsilon_c(r_s, 1) - \epsilon_c(r_s, 0)]. \quad (24)$$

In Fig. 1 we compare our correlation potential (as a function of r_s and for three different values of the spin polarization ζ) with this widely used Tanatar–Ceperley plus exchangelike correlation potential. One can see that, while for $\zeta = 0$ the two potentials are almost indistinguishable, for $\zeta \neq 0$ there are significant discrepancies: at $\zeta = 1$, the difference between the two potentials is ~30% at $r_s = 1$; for lower densities this difference is lower, being 15% at $r_s = 4$ and 7% at $r_s = 10$. At $\zeta = 1$ the discrepancies do not in fact depend on the exchangelike
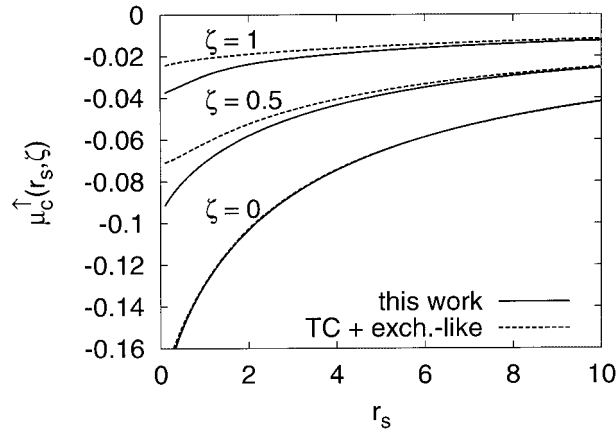
FIGURE 1. Correlation potential for spin-up electrons as a function of the density parameter r_s and for three different values of the spin polarization ζ. The present result is compared with the exchangelike interpolation [3] applied to the Tanatar and Ceperley data [2].

choice for the ζ dependence: they are exclusively due to the corresponding correlation energy of Tanatar and Ceperley, which differs from ours between 35% and 4% for $r_s \in [0, 10]$. To test the intrinsic quality of the exchangelike interpolation against our new ζ-interpolation scheme, we plugged into Eq. (24) our new correlation energies at $\zeta = 0$ and $\zeta = 1$. As shown in Figure 1 of Ref. [7], the ζ dependence of QMC data is rather different, especially at lower densities: at the density of the transition to the fully polarized gas, $r_s \sim 26$, the exchangelike interpolation predicts an energy barrier between the $\zeta = 0$ and the $\zeta = 1$ phases that is more than an order of magnitude higher than the QMC result.

Our correlation energy, which at $\zeta = 1$ is, as said, quite different from the Tanatar–Ceperley [2] value, should be much closer to the true one because (1) we included the effect of BF on the nodes, (2) we imposed the exact high-density limit, and (3) the infinite-size extrapolation is directly performed on a DMC data set.

Summary and Conclusions

We have presented a new, reliable, LSD functional for 2D systems, based on a new set of DMC data for a wide range of electron densities and spin polarizations, and on an analytic form that efficiently reproduces these data and includes most of the known high- and low-density limits. A comparison of the corresponding correlation potential with previous approximations shows, for $\zeta \neq 0$, differences up to 30% for $r_s \in [0, 10]$.

Fortran subroutines available at axtmt2.phys. uniroma1.it/PGG/elegas.html.

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