Correlation energy of finite two-dimensional systems: Toward nonempirical and universal modeling

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The capability of density-functional theory to deal with the ground state of strongly correlated low-dimensional systems, such as semiconductor quantum dots, depends on the accuracy of functionals developed for the exchange and correlation energies. Here we extend a successful approximation for the correlation energy of the three-dimensional inhomogeneous electron gas, originally introduced by Becke [J. Chem. Phys. **88**, 1053 (1988)], to the two-dimensional case. The approach is based on nonempirical modeling of the correlation-hole functions satisfying a set of exact properties. Furthermore, the electron current and spin are explicitly taken into account. As a result, good performance is obtained in comparison with numerically exact data for quantum dots with varying external magnetic field, and for the homogeneous two-dimensional electron gas, respectively.

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I. INTRODUCTION

Density-functional theory[1] (DFT) maps the complicated many-particle problem onto a simple one of noninteracting electrons moving in an effective local potential, the Kohn-Sham (KS) potential. The latter is constructed in such a way that the ground-state density of the noninteracting particles reproduces the ground-state density of the interacting system. Practical success of the approach depends on finding good approximations for the exchange-correlation (xc) energy which, through functional derivation with respect to the particle density, defines the xc part of the KS potential. Most of the approximations developed so far have focused on three-dimensional (3D) systems, i.e., atoms, molecules, and solids. Many advances have been made beyond the commonly used local (spin) density approximation [L(S)DA] by means of, e.g., generalized gradient approximations, orbital functionals, and hybrid functionals.[2] Such efforts for two-dimensional (2D) systems have been relatively scarce despite the rapidly increasing experimental and theoretical interest in quasi-2D structures such as semiconductor layers and surfaces, quantum-Hall systems, graphene, and various types of quantum dots[3] (QDs).

When using DFT, QDs are most commonly treated using the 2D-LSDA exchange[4] combined with the 2D-LSDA correlation parametrized first by Tanatar and Ceperley[5] and later, with more satisfactory spin dependence, by Attaccalite *et al.*[6] In many cases, the LSDA (prefix "2D" omitted below) performs relatively well compared, e.g., with quantum Monte Carlo calculations.[7] Nevertheless, there is a lack of 2D functionals to deal with diverse few-electron QD systems, especially in the strong-correlation regime. Only very recently, a local correlation functional was developed in 2D within the Colle-Salvetti approach, which was found to outperform the LSDA.[8] In its current form, however, this local functional applies only to closed-shell systems with zero spin and zero current. In addition to the correlation-energy functional, exchange-energy functionals have been developed for finite 2D systems in our foregoing works.[9–11]

In this paper we develop a correlation-energy functional in 2D. In the derivation, along the lines of the work of Becke[12] for the 3D case, we discuss a model for spin-dependent correlation-hole functions satisfying a set of exact properties in 2D. As a result, we find a spin- and current-dependent approximation for the correlation energy. In comparison with numerically exact results, the obtained accuracy is found to be superior to the LSDA. However, we also find, and elucidate, that further modeling of the dependency on the average electron density of those parameters describing the size of the correlation-hole functions in terms of the size of the exchange-hole (x-hole) functions would be required. The applications of the functional to a set of few-electron QDs with various relative amounts of correlation, ground-state spins, electron currents, and external magnetic fields confirm the overall usefulness of the approach.

II. THEORY

Within the KS method of spin-DFT,[13] the ground-state energy and spin densities $\rho_\uparrow(\mathbf{r})$ and $\rho_\downarrow(\mathbf{r})$ of a system of $N=N_\uparrow+N_\downarrow$ interacting electrons are determined. The total energy of the interacting system is written as a functional of the spin densities[14]

$$E_v[\rho_\uparrow,\rho_\downarrow]=T_s[\rho_\uparrow,\rho_\downarrow]+\int d\mathbf{r}v(\mathbf{r})\rho(\mathbf{r})+E_H[\rho]+E_{xc}[\rho_\uparrow,\rho_\downarrow], \tag{1}$$

where $T_s[\rho_\uparrow,\rho_\downarrow]$ is the kinetic-energy functional of noninteracting electrons with spin densities $\rho_\uparrow$, $\rho_\downarrow$. v is (at vanishing external magnetic field) the external (local) scalar potential acting upon the interacting system, $E_H[\rho]$ is the classical electrostatic or Hartree energy of the total charge density $\rho=\rho_\uparrow+\rho_\downarrow$, and $E_{\mathrm{xc}}[\rho_\uparrow,\rho_\downarrow]$ is the xc energy functional. E_{xc} may be further decomposed into the exchange energy, E_{x}, and correlation energy E_{c}. We have already considered E_{x} in

Refs. 9–11, and thus we here focus on E_c. Our starting point is the formal expression for E_c in terms of the correlation-hole (c-hole) function,

$$E_c[\rho_\uparrow,\rho_\downarrow] = \frac{1}{2}\sum_{\sigma\sigma'}\int d\mathbf{r}_1 \int d\mathbf{r}_2 \frac{\rho_\sigma(\mathbf{r}_1)}{|\mathbf{r}_1-\mathbf{r}_2|} h_c^{\sigma\sigma'}(\mathbf{r}_1,\mathbf{r}_2), \quad (2)$$

where

$$h_c^{\sigma\sigma'}(\mathbf{r}_1,\mathbf{r}_2) = \int_0^1 d\lambda h_{c,\lambda}^{\sigma\sigma'}(\mathbf{r}_1,\mathbf{r}_2) = \int_0^1 d\lambda h_\lambda^{\sigma\sigma'}(\mathbf{r}_1,\mathbf{r}_2) - h_x^\sigma(\mathbf{r}_1,\mathbf{r}_2)\delta_{\sigma\sigma'} \quad (3)$$

is the c-hole function. Here $\{\sigma\sigma'\}=\{\uparrow\uparrow,\downarrow\downarrow,\uparrow\downarrow,\downarrow\uparrow\}$ and the parameter $\lambda\in[0,1]$ is the electronic coupling strength.[1] In the above expression, the c-hole corresponds to the *full* hole function $h_\lambda^{\sigma\sigma'}$ subtracted by the x-hole function defined as

$$h_x^\sigma(\mathbf{r}_1,\mathbf{r}_2) = -\frac{\left|\sum_{k=1}^{N_\sigma}\psi_{k,\sigma}^*(\mathbf{r}_1)\psi_{k,\sigma}(\mathbf{r}_2)\right|^2}{\rho_\sigma(\mathbf{r}_1)}. \quad (4)$$

Note that here h_x^σ is defined to have a negative sign in contrast to the standard definition [see, e.g., Eq. (4) in Ref. 9]. The λ-dependent hole function is given by

$$h_\lambda^{\sigma\sigma'}(\mathbf{r}_1,\mathbf{r}_2) = \frac{P_{2,\lambda}^{\sigma\sigma'}(\mathbf{r}_1,\mathbf{r}_2)}{\rho_\sigma(\mathbf{r}_1)} - \rho_{\sigma'}(\mathbf{r}_2), \quad (5)$$

where $P_{2,\lambda}^{\sigma\sigma'}$ is the two-body reduced density matrix (2BRDM) (Ref. 12),

$$P_{2,\lambda}^{\sigma\sigma'}(\mathbf{r}_1,\mathbf{r}_2) = N(N-1)\int d3\int d4\ldots\int dN \times \Psi_\lambda^*(1,2,\ldots,N)\Psi_\lambda(1,2,\ldots,N). \quad (6)$$

Here, $\Psi_\lambda(1,2,\ldots,N)$ stands for the ground-state many-body wave function, which is the exact solution of the electronic system with a Coulomb coupling strength λ, $\int dN$ denotes the spatial integration and spin summation over the Nth spatial spin coordinates $(\mathbf{r}_N,\sigma_N)$, and we have identified $\sigma_1=\sigma$, $\sigma_2=\sigma'$. Note that the spin densities in Eq. (5) are the same as in the actual fully interacting system.

As it is well known, in determining the correlation energy it is sufficient to know the angular average of the c-hole function. In 2D it is natural to consider the *cylindrical* average, given by

$$\bar{h}_{c,\lambda}^{\sigma\sigma'}(\mathbf{r},s) = \frac{1}{2\pi}\int_0^{2\pi} d\phi h_{c,\lambda}^{\sigma\sigma'}(\mathbf{r},\mathbf{r}+\mathbf{s}), \quad (7)$$

where $\mathbf{r}_1=\mathbf{r}$, $\mathbf{r}_2=\mathbf{r}+\mathbf{s}$, and ϕ is the angle between $\mathbf{r}$ and $\mathbf{s}$. The modeling of the c-hole functions is based on satisfying the following conditions:

(i) sum rule

$$\int_0^\infty ds s\bar{h}_{c,\lambda}^{\sigma\sigma'}(\mathbf{r},s) = 0; \quad (8)$$

(ii) correct short-range behavior for $s\to 0$;

(iii) a proper decay in the limit $s\to\infty$;

(iv) characteristic size assumed to be proportional to the characteristic size of the corresponding angular averaged x-hole.

The short-range behavior of the c-hole can be worked out by considering the electronic cusp conditions for the 2D electronic wave function.[15] The angular average of the 2BRDM in Eq. (6) is then given by

$$\bar{P}_{2,\lambda}^{\sigma\sigma}(\mathbf{r},s\to 0) \approx A_{\sigma\sigma}(\mathbf{r})s^2\left(1+\frac{2}{3}\lambda s\right), \quad (9)$$

and

$$\bar{P}_{2,\lambda}^{\sigma\bar{\sigma}}(\mathbf{r},s\to 0) \approx A_{\sigma\bar{\sigma}}(\mathbf{r})(1+2\lambda s), \quad (10)$$

for the same-spin and opposite-spin elements, respectively. Note that the symbol $\bar{\sigma}$ always denotes spin opposite to σ, whereas σ', when used, can be equal to either σ or $\bar{\sigma}$. Here the coefficients $A_{\sigma\sigma'}(\mathbf{r})$ have a spatial dependence. Although the cusp condition as developed in Ref. 15 pertains to the center of mass and relative coordinate, it can be shown, as noted by Becke,[12] that at the order considered above the same results apply also for the coordinate system of $\mathbf{r}$ and $\mathbf{s}$.

The short-range behavior of the 2D x-hole is known.[9,16] Thus, it is possible to consider the following model for the same-spin and opposite-spin c-hole functions, respectively:

$$\bar{h}_{c,\lambda}^{\sigma\sigma}(\mathbf{r},s) = \left[B_{\sigma\sigma}(\mathbf{r}) - D_\sigma(\mathbf{r}) + \frac{2}{3}\lambda B_{\sigma\sigma}(\mathbf{r})s\right]s^2 F[\gamma_{\sigma\sigma}(\mathbf{r})s], \quad (11)$$

and

$$\bar{h}_{c,\lambda}^{\sigma\bar{\sigma}}(\mathbf{r},s) = [B_{\sigma\bar{\sigma}}(\mathbf{r}) - \rho_{\bar{\sigma}}(\mathbf{r}) + 2\lambda B_{\sigma\bar{\sigma}}(\mathbf{r})s]\,F[\gamma_{\sigma\bar{\sigma}}(\mathbf{r})s]. \quad (12)$$

Here $B_{\sigma\sigma}:=A_{\sigma\sigma}/\rho_\sigma$ and $B_{\sigma\bar{\sigma}}:=A_{\sigma\bar{\sigma}}/\rho_\sigma$ are coefficients to be determined, and

$$D_\sigma := \frac{1}{2}\left(\tau_\sigma - \frac{1}{4}\frac{(\nabla\rho_\sigma)^2}{\rho_\sigma} - \frac{\mathbf{j}_{p,\sigma}^2}{\rho_\sigma}\right); \quad (13)$$

where $\tau_\sigma=\sum_{k=1}^{N_\sigma}|\nabla\psi_{k,\sigma}|^2$ is the (double of the) kinetic-energy density and $\mathbf{j}_{p,\sigma}=\frac{1}{2i}\sum_{k=1}^{N_\sigma}[\psi_{k,\sigma}^*(\nabla\psi_{k,\sigma})-(\nabla\psi_{k,\sigma}^*)\psi_{k,\sigma}]$ is the spin-dependent paramagnetic current density. In Eqs. (11) and (12), the functions $F[\gamma_{\sigma\sigma'}(\mathbf{r})s]$ are introduced to ensure the decay of the c-holes in the limit $s\to\infty$. We choose them to have the form $F(x)=\exp(-x^2)$, which seems appropriate in the case of finite 2D systems. The parameters $\gamma_{\sigma\sigma'}(\mathbf{r})$ are determined by the zero-integral constraint of Eq. (8) (see below).

Next, we introduce the characteristic sizes of the c-holes, $z_{\sigma\sigma}$ and $z_{\sigma\bar{\sigma}}$, for which the corresponding c-hole function vanishes, i.e.,

$$\bar{h}_{c,\lambda}^{\sigma\sigma}(\mathbf{r},z_{\sigma\sigma}) = 0; \quad \bar{h}_{c,\lambda}^{\sigma\bar{\sigma}}(\mathbf{r},z_{\sigma\bar{\sigma}}) = 0. \quad (14)$$

Implying these conditions, the coefficients $B_{\sigma\sigma}$ and $B_{\sigma\bar{\sigma}}$ in Eqs. (11) and (12) can be written as

$$B_{\sigma\sigma} = \frac{D_\sigma}{1 + \frac{2}{3}\lambda z_{\sigma\sigma}}, \quad B_{\sigma\bar{\sigma}} = \frac{\rho_{\bar{\sigma}}}{1 + 2\lambda z_{\sigma\bar{\sigma}}}. \tag{15}$$

It should be noted, however, that the size parameters $z_{\sigma\sigma}$ and $z_{\sigma\bar{\sigma}}$ are functions of $\mathbf{r}$. As for the 3D functional, we assume the size of the c-hole to be proportional to the size of the x-hole. Thus, we set

$$z_{\sigma\sigma}(\mathbf{r}) := c_{\sigma\sigma}[|U_{\mathrm{x}}^{\sigma}(\mathbf{r})|^{-1} + |U_{\mathrm{x}}^{\sigma}(\mathbf{r})|^{-1}] = 2c_{\sigma\sigma}|U_{\mathrm{x}}^{\sigma}(\mathbf{r})|^{-1}, \tag{16}$$

and

$$z_{\sigma\bar{\sigma}}(\mathbf{r}) := c_{\sigma\bar{\sigma}}[|U_{\mathrm{x}}^{\sigma}(\mathbf{r})|^{-1} + |U_{\mathrm{x}}^{\bar{\sigma}}(\mathbf{r})|^{-1}], \tag{17}$$

where U_{x}^{σ} is the x-hole potential[9,17] for spin σ, and $c_{\sigma\sigma}$ and $c_{\sigma\bar{\sigma}}$ are constants to be determined (see below). As suggested by Becke,[12] the proportionality is plausible due to the fact that each electron is surrounded by its Fermi hole, and hence the electrostatic interaction between two electrons can be expected to be screened beyond some characteristic length proportional to the average size of the x-hole. The argument is not spin related, beyond the fact that the characteristic length for the x-hole could be different for spin up than for spin down. Of course, this is only a simplification, but the physical picture is appealing, and it has led to good results in 3D atomic systems.

The λ-dependent c-hole functions can now be written as

$$\bar{h}_{\mathrm{c},\lambda}^{\sigma\sigma}(\mathbf{r},s) = \frac{2}{3}\lambda D_\sigma(\mathbf{r})\left[\frac{s - z_{\sigma\sigma}(\mathbf{r})}{1 + \frac{2}{3}\lambda z_{\sigma\sigma}(\mathbf{r})}\right] s^2 F[\gamma_{\sigma\sigma}(\mathbf{r})s], \tag{18}$$

and

$$\bar{h}_{\mathrm{c},\lambda}^{\sigma\bar{\sigma}}(\mathbf{r},s) = 2\lambda\rho_{\bar{\sigma}}(\mathbf{r})\left[\frac{s - z_{\sigma\bar{\sigma}}(\mathbf{r})}{1 + 2\lambda z_{\sigma\bar{\sigma}}(\mathbf{r})}\right] F[\gamma_{\sigma\bar{\sigma}}(\mathbf{r})s]. \tag{19}$$

Integrating over λ yields

$$\bar{h}_{\mathrm{c}}^{\sigma\sigma}(\mathbf{r},s) = \frac{D_\sigma(\mathbf{r})[s - z_{\sigma\sigma}(\mathbf{r})]s^2 F[\gamma_{\sigma\sigma}(\mathbf{r})s]}{2z_{\sigma\sigma}^2(\mathbf{r})} \times\left[2z_{\sigma\sigma}(\mathbf{r}) - 3\ln\left(\frac{2}{3}z_{\sigma\sigma}(\mathbf{r}) + 1\right)\right], \tag{20}$$

and

$$\bar{h}_{\mathrm{c}}^{\sigma\bar{\sigma}}(\mathbf{r},s) = \frac{\rho_{\bar{\sigma}}(\mathbf{r})[s - z_{\sigma\bar{\sigma}}(\mathbf{r})]F[\gamma_{\sigma\bar{\sigma}}(\mathbf{r})s]}{2z_{\sigma\bar{\sigma}}^2(\mathbf{r})} \times\{2z_{\sigma\bar{\sigma}}(\mathbf{r}) - \ln[2z_{\sigma\bar{\sigma}}(\mathbf{r}) + 1]\}. \tag{21}$$

Finally, we enforce the sum rules in Eq. (8) giving

$$\gamma_{\sigma\sigma}(\mathbf{r}) = \frac{3\sqrt{\pi}}{4z_{\sigma\sigma}(\mathbf{r})}, \tag{22}$$

and

$$\gamma_{\sigma\bar{\sigma}}(\mathbf{r}) = \frac{\sqrt{\pi}}{2z_{\sigma\bar{\sigma}}(\mathbf{r})}. \tag{23}$$

This concludes the derivation of the c-hole functions $\bar{h}^{\sigma\sigma}$ and $\bar{h}^{\sigma\bar{\sigma}}$, apart from the determination of constants $c_{\sigma\sigma}$ and $c_{\sigma\bar{\sigma}}$, respectively.

From the c-hole functions we can calculate the c-hole *potentials* as

$$U_{\mathrm{c}}^{\sigma\sigma'}(\mathbf{r}) = 2\pi\int_0^\infty ds\bar{h}_{\mathrm{c}}^{\sigma\sigma'}(\mathbf{r},s). \tag{24}$$

For the same- and opposite-spin cases of our approximation, we find respectively

$$U_{\mathrm{c}}^{\sigma\sigma}(\mathbf{r}) = \frac{16}{81\pi}(8 - 3\pi)D_\sigma(\mathbf{r})z_{\sigma\sigma}^2(\mathbf{r}) \times\left[2z_{\sigma\sigma}(\mathbf{r}) - 3\ln\left(\frac{2}{3}z_{\sigma\sigma}(\mathbf{r}) + 1\right)\right], \tag{25}$$

and

$$U_{\mathrm{c}}^{\sigma\bar{\sigma}}(\mathbf{r}) = (2 - \pi)\rho_{\bar{\sigma}}(\mathbf{r}) \times \{2z_{\sigma\bar{\sigma}}(\mathbf{r}) - \ln[2z_{\sigma\bar{\sigma}}(\mathbf{r}) + 1]\}. \tag{26}$$

The correlation energies are given by

$$E_{\mathrm{c}}^{\sigma\sigma'} = \frac{1}{2}\int d\mathbf{r}\rho_\sigma(\mathbf{r})U_{\mathrm{c}}^{\sigma\sigma'}(\mathbf{r}). \tag{27}$$

Thus

$$E_{\mathrm{c}}[\rho_\uparrow,\rho_\downarrow] = E_{\mathrm{c}}^{\uparrow\uparrow} + E_{\mathrm{c}}^{\downarrow\downarrow} + 2E_{\mathrm{c}}^{\uparrow\downarrow}, \tag{28}$$

where we have used the condition $E_{\mathrm{c}}^{\uparrow\downarrow} = E_{\mathrm{c}}^{\downarrow\uparrow}$.

Alternatively, we can compute the correlation energy directly from the c-hole functions. From Eqs. (2) and (7) we get

$$E_{\mathrm{c}}[\rho_\uparrow,\rho_\downarrow] = \pi\sum_{\sigma\sigma'}\int d\mathbf{r}\int_0^\infty ds\rho_\sigma(\mathbf{r})\bar{h}_{\mathrm{c}}^{\sigma\sigma'}(\mathbf{r},s). \tag{29}$$

We remind that $D_\sigma(\mathbf{r})$ introduced in Eq. (13) vanishes for all the single-particle ($N=1$) systems.[12] Therefore, the c-hole and thus E_c vanish as well. In other words, our approximation for the correlation energy is self-interaction free for $N=1$.[18]

III. NUMERICAL RESULTS

The first task in the numerical applications is to complete the correlation functional by finding approximations for constants $c_{\sigma\sigma}$ and $c_{\sigma\bar{\sigma}}$ in Eqs. (16) and (17). For this purpose, we consider a set of harmonically confined QDs, where the external confinement is given by $v(r) = \omega^2 r^2/2$. Reference results for the correlation energies can be obtained from

$$E_{\mathrm{c}}^{\mathrm{ref}} = E_{\mathrm{tot}}^{\mathrm{ref}} - E_{\mathrm{tot}}^{\mathrm{EXX}}, \tag{30}$$

where $E_{\mathrm{tot}}^{\mathrm{ref}}$ is the exact total energy obtained, e.g., from, an analytic accurate configuration-interaction (CI) or quantum

TABLE I. Total energies from the full configuration-interaction calculations (Ref. 21) for totally spin-polarized ($S_z=N/2$) quantum dots, exact-exchange total energies, the reference correlation energies ($E_c^{ref}=E_{tot}-E_{tot}^{EXX}$), $c_{\sigma\sigma}$ yielding $E_c^{model}=E_c^{ref}$, E_c^{model} obtained with a fixed average value $c_{\sigma\sigma}=1.32$, and the LSDA correlation energy.

N	ω	E_{tot}^{ref}	E_{tot}^{EXX}	E_c^{ref}	$c_{\sigma\sigma}$	E_c^{model}	E_c^{LSDA}
3	1/4	2.081	2.103	−0.0226	1.27	−0.0245	−0.0538
3	1/16	0.6908	0.7075	−0.0167	1.41	−0.0144	−0.0382
6	1/4	7.233	7.296	−0.0640	1.24	−0.0750	−0.1125
6	1/16	2.553	2.599	−0.0458	1.36	−0.0441	−0.0795

Monte Carlo (QMC) calculation, and EXX refers to the exact exchange. Here we have calculated the EXX energies in the Krieger-Li-Iafrate[19] (KLI) approach[19] in the octopus DFT code.[20] The self-consistent EXX result—the x-hole potential, (spin) density, kinetic-energy density, and current density—is used as input for our correlation functional.

Table I shows the results for the same-spin case. Now the QDs are completely spin polarized with $S_z=N/2$. For each QD we show that value of $c_{\sigma\sigma}$ which yields the reference correlation energy, i.e., $E_c^{model}=E_c^{ref}$. In these examples we find $c_{\sigma\sigma}=1.24,\ldots,1.41$. Thus, the variation in $c_{\sigma\sigma}$ is rather small in view of the fact that the density parameter, approximated in harmonic QDs as $r_s^{QD}=N^{-1/6}\omega^{-2/3}$,[22] varies from 1.9 to 5.3. The second last column of Table I shows the correlation energy obtained by using a fixed average value of $c_{\sigma\sigma}=1.32$. This leads to the maximum deviation of ~23% from E_c^{ref}. In comparison, the self-consistent LSDA correlation energy (last column) deviates from the reference result by up to 130%.

Table II shows the results for a set of unpolarized ($S_z=0$) QDs in the range $1.5<r_s^{QD}<5.7$. Note that for $N>2$ both same- and opposite-spin components of the correlation are present, and we have fixed $c_{\sigma\sigma}=1.32$ according to the conclusions above. Fixing $c_{\sigma\bar{\sigma}}=0.75$ yields deviations of only $\lesssim 10\%$ from E_c^{ref}, except for the highly correlated case of $N=2$ and $\omega=1/16$, which shows a deviation of 25%. The LSDA is still considerably further off the reference result than our functional, but it performs relatively better than in the polarized case discussed above. In particular, when $N=12$ the error in the LSDA correlation is only about 9%. This is in line with the well-known fact that both the L(S)DA exchange and correlation become more accurate with increasing particle number.

In Fig. 1 we consider the total energy of a more general case: A six-electron QD as a function of the magnetic field B directed perpendicular to the dot plane. Increasing the field leads to nontrivial changes in the ground-state quantum numbers (S_z,L_z) and hence to "kinks" in the ground-state total energy as a function of B. As the reference data, we use here the variational QMC results (red solid line) given in Refs. 7 and 25 for a wide range of B up to total spin polarization. The confinement strength is here $\omega=0.42168$, corresponding to a typical confinement of 5 meV when modeling QDs in GaAs.[3] Note that the *total* confinement energy, i.e., $6\Omega=6\sqrt{\omega^2+\omega_c^2/4}$, where $\omega_c=B/c$, has been subtracted from the total energies to clarify the comparison. We point out that the variational QMC method gives an upper bound for the true total energy. On the basis of previous comparisons between the variational and diffusion QMC, and exact diagonalization,[26] our reference data in Fig. 1 can be expected to overestimate the exact total energy by *at most* $0.2,\ldots,3$ meV. The maximum possible errors are smaller in the polarized regime ($B\gtrsim 5$) T.

Overall, Fig. 1 shows reasonable agreement of our functional (circles) with the QMC data through the full range of the magnetic field. However, the functional yields systematically slightly too low correlation energies, and thus too low total energies, even if the possible overestimation of the total energy given by the variational QMC is taken into account. We point out that the functional is here applied with the fixed parameters $c_{\sigma\sigma}=1.32$ and $c_{\sigma\bar{\sigma}}=0.75$ suggested by the results in Tables I and II, which correspond to considerably weaker confining potentials (smaller values of ω). Obviously, this difference implies a different average electron density, and thus a different range of the relative correlation energy. In fact, if the parameter values are reduced to $c_{\sigma\sigma}=1.1$ and

TABLE II. Similar to Table I but for unpolarized ($S_z=0$) quantum dots. The correlation energies from our functional, E_c^{model}, have been calculated using the fixed average values $c_{\sigma\sigma}=1.32$ and $c_{\sigma\bar{\sigma}}=0.75$.

N	ω	E_{tot}^{ref}	E_{tot}^{EXX}	E_c^{ref}	$c_{\sigma\bar{\sigma}}$	E_c^{model}	E_c^{LSDA}
2	1	3[a]	3.162	−0.162	0.72	−0.171	−0.199
2	1/4	0.9324[b]	1.046	−0.114	0.82	−0.102	−0.139
2	1/16	0.3031[b]	0.373	−0.070	0.96	−0.053	−0.085
6	1/4	6.995[b]	7.391	−0.396	0.73	−0.406	−0.457
12	$1/1.89^2$	25.636[c]	26.553	−0.917	0.71	−0.983	−1.000

[a]Analytic solution by Taut from Ref. 23.
[b]CI data from Ref. 21.
[c]Diffusion QMC data from Ref. 24.

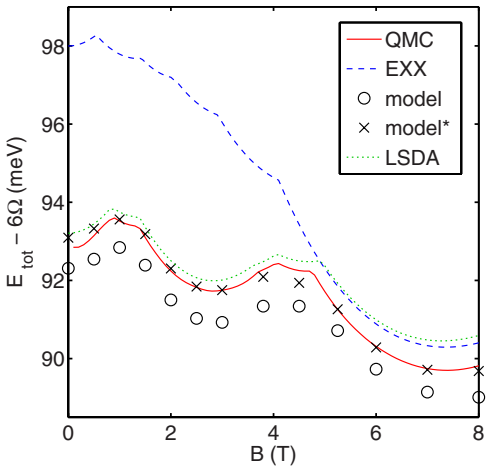

FIG. 1. (Color online) Total energies (minus the confinement energy) for a six-electron quantum dot as a function of the magnetic field. Results are shown for the reference quantum Monte Carlo calculations from Ref. 7 (red solid line), for the exact-exchange data (blue dashed line), for our functional with $c_{\sigma\sigma}=1.32$ and $c_{\sigma\bar{\sigma}}=0.75$ (circles), for our functional with $c_{\sigma\sigma}=1.1$ and $c_{\sigma\bar{\sigma}}=0.7$ (crosses), and for the local-spin-density approximation (green dotted line). The total energy is calculated from our functional as $E_{\mathrm{tot}}^{\mathrm{model}}=E_{\mathrm{tot}}^{\mathrm{EXX}}+E_{\mathrm{c}}^{\mathrm{model}}$.

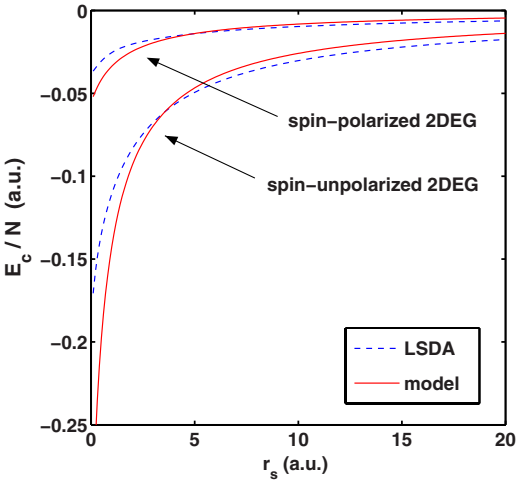

FIG. 2. (Color online) Correlation energy per electron in a homogeneous two-dimensional electron gas of full spin polarization (upper curves) and zero polarization (lower curves). The solid lines show the result from our functional with the original average parameter values $c_{\sigma\sigma}=1.32$ and $c_{\sigma\bar{\sigma}}=0.75$. The dashed lines show to the local-spin-density approximation for the correlation, corresponding to the numerically exact result in this system (Ref. 6).

$c_{\sigma\bar{\sigma}}=0.7$, excellent agreement with QMC is found (see the crosses in Fig. 1). Hence, it seems that particularly high precision of our functional would require modeling of $c_{\sigma\sigma}$ and $c_{\sigma\bar{\sigma}}$ as a function of the particle density. Most importantly, however, the systematic performance of our approximation in Fig. 1 demonstrates that the magnetic field effects, electron currents, and spin are correctly accounted for.

We note that the good accuracy of the LSDA in terms of total energies (see the dotted line in Fig. 1) is due to the compensation of respective errors in the exchange and correlation energies.[25] On the other hand, we tested our correlation functional for the important limit of the *homogeneous* 2D electron gas (2DEG), for which the LSDA correlation is exact, and found reasonable agreement as a function of $r_s=1/\sqrt{\pi\rho}$ for both zero and full spin polarization (see Fig. 2). Here we used the original average parameter values $c_{\sigma\sigma}=1.32$ and $c_{\sigma\bar{\sigma}}=0.75$. Note that according to the numerically exact results in Ref. 6, the ground state of the 2DEG is unpolarized for $0<r_s\lesssim 26$.

Finally, we point out that in principle a given functional should be evaluated with KS orbitals obtained from self-consistent calculation instead of a post-hoc manner as we have done in this work. However, the variational nature of DFT implies that if one evaluates the total energy with densities which slightly differ from the self-consistent one, the resulting change in the energy is of second order in the deviation of the densities.

IV. CONCLUSIONS

We have derived a spin- and current-dependent approximation for the correlation energy of finite two-dimensional electron systems. The core of the derivation is a model for the correlation-hole function of both same-spin and opposite-spin pairs, respectively, that satisfies a set of exact properties. The excellent results obtained for few-electron quantum dots with different spin polarization, current, external magnetic field, and covering a wide range of correlation energies strongly recommend further developments along the construction we have presented here.

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