# Dimensionality dependence of the self-interaction correction in the local-density approximation to density functional theory

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The local-density approximation (LDA) to density functional theory is not self-interaction free. Motivated by this intellectual challenge, and the possible practical importance of strong electron-correlation in a Wigner-type model, the capability of LDA is investigated for a two-dimensional electron system in the low-density limit. It is found for this essentially single-electron limit that the performance of LDA is slightly better in two dimension than in the equivalent three-dimensional problem treated earlier in Phys. Rev. B **18**, 6506 (1978). An *a priori* explanation of this fact is given in terms of the different characters of potential fields generated by normalized charge distributions in these dimensions.

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In the terminology of the review article[1] of Walter Kohn on condensed matter physics, the so-called Wigner lattice[2,3] is governed by the *radical effect* of the electron-electron interaction $v(r)=e^2/r$. The most accurate numerical calculations for such systems are performed by quantum Monte Carlo simulations[4,5] in three and two dimensions. The output energies per particle of these calculations are used to constrain an input form to the local-density approximation (LDA) for the exchange-correlation energy at low densities. A well-constrained form for this many-body term is vital in the practical *a posteriori* implementation of the Kohn-Sham orbital method as applied to various interacting many-electron systems.

While the exact density functional for the ground-state energy is self-interaction free, the practical method based on LDA is not. The elimination of the self-interaction is, therefore, an important issue.[6] In this Brief Report, we shall investigate the two-dimensional (2D) self-interaction problem for the low-density, i.e., Wigner lattice limit since this is *one* of the limits on which an interpolation procedure to design an input exchange-correlation energy per particle in LDA is based. The real advantage of using the low-density limit is that in this important limit one has an essentially single-electron problem. Due to this fact, it is easier to identify the contribution of the self-interaction error, as was explicitly pointed out earlier[7] for the three-dimensional (3D) case. Following this logic, we start by a short review of energetics at low densities. We will use Hartree atomic units, $\hbar=m_e=e^2=1$, in our comparative study.

In a classical, point charge in the continuum, Wigner-Seitz modeling we take an electron to the center of the charge-compensating (rigid) background with a certain radius $R$. There are two terms contributing to a *variational* lattice ($L$) energy $\epsilon_L(D)$ in this[8] modeling. The Coulomb interaction energy $[\epsilon_{eb}(D)]$ of the pointlike electron with the positive jellium background, and the background self-energy $[\epsilon_{bb}(D)]$ are

$$\epsilon_{eb}=-\frac{D}{D-1}\frac{R^{D-1}}{r_s^D}, \tag{1}$$

$$\epsilon_{bb}=\frac{1}{2}\frac{D}{D-1}\frac{R^{2D-1}}{r_s^{2D}}\frac{D}{(2D-1)!!}\left(\frac{\Gamma(D)}{\Gamma[(D+1)/2]}\right)^2, \tag{2}$$

where $r_s$ is the Wigner-Seitz radius.[8] After a variational procedure, we get the $R=r_s$ and $R=(\sqrt{\pi}/2)r_s$ values in 3D and 2D, respectively, at which the classical lattice-energies $\epsilon_L(D)\equiv\epsilon_{eb}(D)+\epsilon_{bb}(D)$ per particle are optimal. In a quantum-mechanical treatment, we have an oscillator ($os$) contribution of $\epsilon_{os}(D)=[D/(2r_s^{3/2})]$ to the above sum of two terms in the lattice energy.

We stress the point that, in 3D, the $R=r_s$ derived here via a variational method fits completely to the remarkable theorem established[9] by Lieb and Narnhofer. According to this theorem, the 3D lattice energy is *bounded* from below by the value, $\epsilon_L(3D)=-0.9/r_s$, obtained for it via the standard (and simple) Wigner-Seitz picture.

In order to model the strong electron-electron interaction in the fluid phase, we can use Wigner's *original* (as noted by Kirkwood)[10] picture in which a slow electron is surrounded by an electrostatically driven hole. Taking this normalized hole as a positive charge-distribution with an $r_s$ radius, as the energetically optimal limiting result due to electron-electron interaction, we get

$$\epsilon_{xc}(D)=\frac{1}{2}\left[-\frac{D}{D-1}\frac{1}{r_s}\right], \tag{3}$$

which is one-half the term given by Eq. (1) at $R=r_s$. At low densities the quantum-mechanical kinetic energy contributions ($\sim r_s^{-2}$) to $\epsilon_{xc}(D)$ become negligible.

It is important to note that in his *first* work on the 3D prototype many-body system, Wigner used the above $\epsilon_{xc}(3D)=-0.75/r_s$ expression and subtracted from it the Hartree-Fock, i.e., exchange-only, $\epsilon_x(3D)$ value in order to define a correlation energy.[1,2,10] One has

 

$$\epsilon_x(D) = -\frac{2}{\pi} p_F \frac{D}{D^2 - 1}, \tag{4}$$

in terms[8] of the Fermi wave-number $p_F$. From this, we obtain $\epsilon_x(3D) \cong -0.46/r_s$ and $\epsilon_x(2D) \cong -0.60/r_s$.

Next we illustrate the applicability of the above-outlined simple energetics via an *estimation* for transition between different[11] phases in our model system. To this estimation we use the $\epsilon_{xc}(D) = \epsilon_L(D) + [D/(2r_s^{3/2})]$ constraint, i.e., we neglect the kinetic term which is higher-order in the inverse density parameter. With the above *variational* values for the $R$ radius, we get $r_s(3D) = 100$ and $r_s(2D) = 9(2\sqrt{\pi} - 3)^{-2} \cong 30$ for the transition values. These are, within few percent, in good agreement with predictions $[r_s(3D) = 106 \pm 1$, and $r_s(2D) = 31 \pm 1]$ based on quantum Monte Carlo[4,5] simulations. However, one should keep in mind the remark of Anderson[11] about an interpolation[2,6] procedure. He pointed out the delicate nature of the commonly applied interpolation between *different* phases.

Notice, in passing, that the numerical values of the ratio of $\epsilon_L(D)/\epsilon_x(D)$ are close (from below) to two independently of $D$, as was emphasized[12] in a recent analysis on bounds in LDA. Considering the common scaling $[\sim(1/r_s)]$ of the leading terms in the energies of the many-body system at low densities, one has to use the $\epsilon_{xc}^{(LDA)}(D) = -\beta/r_s$ form to design properly a local input-construction to numerical applications.[7,13]

In the Wigner lattice, a pointlike electron sees *only* the normalized compensating charge density of the rigid background. Thus, in an LDA treatment, the spurious self-interaction term, which is one-half the integrated[6,7] electrostatic interaction of a normalized electronic charge-distribution with the potential field generated by itself, must cancel the energy-contribution calculated via a local (input) exchange-correlation term $[\epsilon_{xc}^{(LDA)}(r)]$. Earlier, insightful work gave, based on this robust global constraint, the $\beta(3D) \cong 0.675 (< 0.75 < 0.9)$ value as the optimal (*maximum*) one in the 3D case.[7]

At this point we introduce our main *a priori* statement on the dimensionality-dependence of deviations in LDA from the bounded exchange-correlations. Our statement rests on the following simple observation. The ratio $[\epsilon_{bb}(2D)/\epsilon_{bb}(3D)] > 1$ at $R = r_s$. This heralds, in the light of the above-outlined cancellation-constraint, than in 2D-LDA with arbitrary (normalized) charge distributions we will get a closer agreement with bounds on exchange-correlation than in the 3D case.

So, *how close* can we go in 2D-LDA to the established rigorous bounds in the investigated low-density limit? Our quantitative estimations are based on three forms of the electronic charge distribution. The first of them

$$n_H(r) = \frac{2\alpha^2}{\pi} e^{-2\alpha r}, \tag{5}$$

corresponds to a standard hydrogenic probability distribution function. The second is a Gaussian

$$n_G(r) = \frac{\alpha}{\pi} e^{-\alpha r^2}. \tag{6}$$

Finally, in order to allow a more tunable (via $\mu > 0$) representation, a power function is introduced

$$n_P(r) = \frac{\alpha}{\pi} \frac{\mu}{(1 + \alpha r^2)^{\mu+1}}. \tag{7}$$

The cancellation constraint to estimate an optimal $\beta(2D)$ value to 2D-LDA is formulated as follows:

$$\beta \int dr r [\pi n(r)]^{1/2} n(r) \equiv \frac{1}{2} \int dr r \varphi(r) n(r), \tag{8}$$

where $\varphi(r)$ is the potential field generated by the given electronic charge distribution $n(r)$ itself.

The calculation of this field is, however, more complicated[14] in 2D real-space than in 3D. We illustrate this with the $n_H(r)$ distribution, with which we have

$$r\varphi_H(r) = 2u^2 [I_0(u)K_1(u) - I_1(u)K_0(u)], \tag{9}$$

in terms[15] of a shorthand $u \equiv \alpha r/2$. The asymptotic form at $r \to \infty$ is $r\varphi_H(r) \to [1 + 3/(2\alpha^2 r^2)]$, which signals that the decay is *slow* in 2D. In 3D, the corresponding (hydrogenic) limit is, as is well-known from textbooks on quantum mechanics, unity to an exponential accuracy. The observed long-range differences in the fields generated by charge distributions are behind the enhanced values of the self-energy integrations, Eq. (8), in 2D.

In practice, we performed the integration on the left-hand side of Eq. (8) in real space, while the right-hand side (RH) is evaluated by using the convolution (faltung) theorem. Applying this theorem, we easily get

$$(RH) = \frac{1}{2} \int_0^\infty dq q \left[ n(q) \frac{1}{q} n(q) \right], \tag{10}$$

where $n(q)$ is the Fourier-Hankel transform of $n(r)$. In the most complicated [power $(P)$] case the result is

$$\beta_P(\mu) = \frac{\pi(3\mu + 1)}{4^{(\mu+1)} \mu^{5/2}} \frac{\Gamma(2\mu + 1/2)\Gamma(\mu + 1/2)}{[\Gamma(\mu)]^3}. \tag{11}$$

One gets $\beta_P(\mu = 1/2) = 5/(4\sqrt{2}) \simeq 0.88$. From Eq. (11) $\beta_P(\mu \to 0) = (\pi/2)^2 \sqrt{\mu}$ and $\beta_P(\mu \to \infty) = (3/4)\sqrt{\pi/2}$. This last value is equal to the one obtained in the Gaussian case, where $\beta_G(2D) = (3/4)\sqrt{\pi/2}$ also. With $n_H(r)$ the (very similar) result is $\beta_H(2D) = 27\pi/(64\sqrt{2})$.

Therefore, quite independently of the applied models, we have a $-0.94/r_s$ *maximum-type* estimation for the exchange-correlation energy per particle within 2D-LDA of density functional theory in the low-density limit. It is, therefore, the best representation for this important limit which is free from the spurious self-interaction in a local treatment. This result differs only a little from $\epsilon_{xc}(2D) = -1/r_s$ obtained for the homogeneous gas at low densities of the 2D system. The ratio of $[(-0.94/r_s)/\epsilon_L(R = r_s)]$ is, on the other hand, about 0.82 in 2D. The corresponding ratio in the 3D case[7] has an about 0.75 value, due to $\beta(3D) \cong 0.675$. This informative ratio also signals a stronger deviation in 3D.

As a future direction we propose to investigate, following an earlier insightful work[16] on the hydrogen atom in 3D, a gradient-corrected correlation functional in the Wigner limit where one is dealing with isolated electrons. In such a treatment[16] for this important limit, the correlation *potential* of the functional must cancel the $\phi(r)$ potential of Eq. (8) locally. Since the $\phi(r)$ Coulomb potential depends on the applied single-particle orbital, as a comparison of Eq. (9) with the Gaussian case

$$r\varphi_G(r) = \sqrt{2\pi z} I_0(z) e^{-z} \tag{12}$$

clearly shows $[z=\alpha r^2/2]$, the results for the correlation potential will be orbital-dependent. We note finally that the many-electron self-interaction error, beyond the one-electron self-interaction problem discussed in our comparative study, has also a broad interest in constructions of universally useful approximate functionals.[17]

In conclusion, there is, as we explained *a priori* in this short work, a better performance of the LDA in 2D than in 3D at the low-density limit, where we have an essentially *single-electron* problem. The necessary self-interaction correction on an input many-body term to LDA in DFT is, therefore, smaller in 2D at this limit. Immediate applications of the result found in our study could be a modification of the LDA-based input exchange-correlation in thickness-dependent modeling[13] of an electron layer, and in the fundamental problem of bound states[18] around a negative point charge in a 2D electron system at low densities. In the attractive, so-called interaction strength *interpolation* modeling[19,20] of exchange and correlation, the input at strong coupling rests on the details of the Wigner limit, as well.

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