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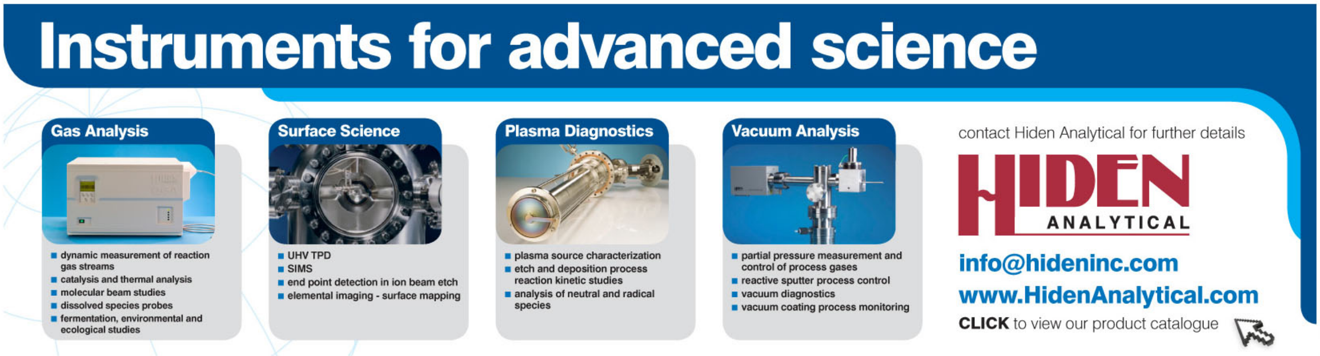

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High-density correlation energy expansion of the one-dimensional uniform electron gas

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We show that the expression of the high-density (i.e., small-r_s) correlation energy per electron for the one-dimensional uniform electron gas can be obtained by conventional perturbation theory and is of the form $\epsilon_c(r_s) = -\pi^2/360 + 0.00845\, r_s + \ldots$, where r_s is the average radius of an electron. Combining these new results with the low-density correlation energy expansion, we propose a local-density approximation correlation functional, which deviates by a maximum of 0.1 mhartree compared to the benchmark diffusion Monte Carlo calculations. © *2013 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4790613]

I. INTRODUCTION

Recently much attention has been devoted to one-dimensional (1D) systems. For example, Wagner *et al.*[1] have shown that 1D chemical systems, such as light atoms (H, He, Li, Be, ...), ions (H^-, Li^+, Be^+, ...), or diatomics (e.g., H_2), can be used as "theoretical laboratory" to study strong correlation in "real" three-dimensional chemical systems within density-functional theory (DFT).[2]

One-dimensional systems can be also experimentally realized in carbon nanotubes,[3–7] organic conductors,[8–12] transition metal oxides,[13] edge states in quantum Hall liquids,[14–16] semiconductor heterostructures,[17–21] confined atomic gases,[22–24] and atomic or semiconducting nanowires.[25,26] The uniform electron gas (UEG) paradigm which is the main "ingredient" of most of the correlation functionals and the cornerstone of the most popular DFT approximation—the local-density approximation (LDA)—is particularly well-adapted to the theoretical study of subtle effects involved by electron correlation in such systems. However, while the high-density (small-r_s) reduced (i.e., per electron) correlation energy expansions

$$\epsilon_c(r_s) = \sum_{j=0}^{\infty} \left(\lambda_j \ln r_s + \epsilon_j\right) r_s^j = \lambda_0 \ln r_s + \epsilon_0 + \lambda_1 r_s \ln r_s + \epsilon_1 r_s + \ldots \qquad (1)$$

(where r_s is the Seitz radius) of the two-dimensional (2D) and three-dimensional (3D) UEGs are quite well-known,[27–51] much less has been discovered about the 1D UEG. This lack of information is mainly due to the divergence of the Coulomb operator $1/x$ in 1D for small interelectronic distance x,[52–56] which makes conventional perturbation theory difficult to apply due to the absence of a Fourier transform for the Coulomb operator. In this article, we propose to fill this gap by reporting the values of the first few high-density coefficients (see Table I). We note that, although the bare Coulomb operator is not the natural operator in 1D (i.e., the solution of the 1D Poisson's equation does not give a Coulombic potential), in the following study we are interested in real electrons that are confined so that they can move in only one dimension of a 3D space. For this reason, it is appropriate to use the 1/x Coulomb potential.

The present system is constructed by allowing the number n of electrons in a 1D box of length L with periodic boundary conditions to approach infinity with the density

$$\rho = \frac{n}{L} = \frac{1}{2r_s} \qquad (2)$$

held constant.[2,57] Because the paramagnetic and ferromagnetic states are degenerate for strict 1D systems, we will consider only the latter (i.e., a spin-polarized electron gas).[54–56]

To avoid the divergence of the Coulomb operator, we will consider in our derivation a "soften" version of the Coulomb operator $1/\sqrt{x^2+R^2}$, where R is a parameter which removes the singularity at $x = 0$.[58,59] Then, we will carefully take the limit $R \to 0$. We will show that, unlike the 2D and 3D version of the UEG, second- and third-order perturbation theories are convergent, i.e., there is no need to use resummation techniques.[39] Combining these new results with the low-density energy expansion and the available diffusion Monte Carlo (DMC) data, we propose a new LDA functional for the reduced correlation energy of the 1D UEG. Atomic units are used throughout.

II. HIGH-DENSITY EXPANSION

A. Second-order perturbation theory

In 1D, the spinorbitals of the free electron gas are

$$\psi_k(x) = \frac{e^{ikx}}{\sqrt{L}}, \qquad (3)$$

with the energy $\kappa_k = k^2/2$, and where the periodic boundary conditions imply $k = 2\pi\, m/L$ ($m \in \mathbb{Z}$). The coefficient ϵ_0 is

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TABLE I. High-density coefficients for the paramagnetic state of the 1D, 2D, and 3D UEGs. The paramagnetic and ferromagnetic states are degenerated in 1D. β and ζ are the Dirichlet beta and Riemann zeta functions, respectively.[61]

Coefficient	Term	1D	2D	3D
λ_0	$\ln r_s$	0	0	$(1-\ln 2)/\pi^2$
ϵ_0	r_s^0	$-\pi^2/360$	$\ln 2 - 1 + \beta(2) - 8\beta(4)/\pi^2$	$-0.071099 + (\ln 2)/6 - 3\zeta(3)/(4\pi^2)$
λ_1	$r_s \ln r_s$	0	$-\sqrt{2}\,(10/(3\pi) - 1)$	$+0.009229$
ϵ_1	r_s	$+0.00845$	unknown	-0.020

given by second-order perturbation theory[60]

$$\epsilon_0(R) = \frac{1}{4n}\sum_{ab}^{\text{occ}}\sum_{rs}^{\text{virt}} \frac{|\langle ab\|rs\rangle|^2}{\kappa_a + \kappa_b - \kappa_r - \kappa_s}, \tag{4}$$

where $\langle ab\|rs\rangle = \langle ab|rs\rangle - \langle ab|sr\rangle$ and

$$\langle ab|rs\rangle = \int_{-L/2}^{L/2}\int_{-L/2}^{L/2} \frac{\psi_a^*(x_1)\psi_b^*(x_2)\psi_r(x_1)\psi_s(x_2)}{\sqrt{(x_1-x_2)^2+R^2}}dx_1dx_2. \tag{5}$$

The constant coefficient ϵ_0 is usually decomposed into a direct ("ring-diagram") term ϵ_0^{a} and an exchange term ϵ_0^{b}, which read explicitly as

$$\epsilon_0^{\text{a}}(R) = \frac{1}{2n}\sum_{ab}^{\text{occ}}\sum_{rs}^{\text{virt}} \frac{\langle ab|rs\rangle\langle rs|ab\rangle}{\kappa_a + \kappa_b - \kappa_r - \kappa_s}, \tag{6}$$

$$\epsilon_0^{\text{b}}(R) = -\frac{1}{2n}\sum_{ab}^{\text{occ}}\sum_{rs}^{\text{virt}} \frac{\langle ab|rs\rangle\langle rs|ba\rangle}{\kappa_a + \kappa_b - \kappa_r - \kappa_s}. \tag{7}$$

Using the Fourier transform of the soft Coulomb potential

$$\frac{1}{\sqrt{x^2+R^2}} = \frac{1}{\pi}\int_{-\infty}^{\infty} K_0(|k|R)e^{ikx}dk, \tag{8}$$

where K_0 is the zeroth-order modified Bessel function of the second kind,[61] the well-known relation

$$\delta(x) = \frac{1}{2\pi}\int_{-\infty}^{\infty} e^{ikx}dk, \tag{9}$$

and transforming the sums in (6) and (7) into integrals

$$\sum_k \to \frac{L}{2\pi}\int dk, \tag{10}$$

with $p_1 = a/k_{\text{F}}$, $p_2 = b/k_{\text{F}}$, $q = k/k_{\text{F}}$ and $R \leftarrow k_{\text{F}}R$, where $k_{\text{F}} = \pi\rho$ is the Fermi wave vector,[62] we eventually find[63]

$$\epsilon_0^{\text{a}}(R) = -\frac{1}{4\pi^2}\int_{-\infty}^{\infty}dq \int_{\substack{|p_1|<1\\|p_1+q|>1}} dp_1 \times \int_{\substack{|p_2|<1\\|p_2+q|>1}} dp_2 \frac{K_0(|q|R)^2}{q(p_1+p_2+q)} \tag{11}$$

and

$$\epsilon_0^{\text{b}}(R) = \frac{1}{4\pi^2}\int_{-\infty}^{\infty}dq \int_{\substack{|p_1|<1\\|p_1+q|>1}} dp_1 \int_{\substack{|p_2|<1\\|p_2+q|>1}} dp_2 \times \frac{K_0(|q|R)K_0(|p_1+p_2+q|R)}{q(p_1+p_2+q)}. \tag{12}$$

For $R > 0$, Eqs. (11) and (12) can be evaluated numerically. As shown in Fig. 1, $\epsilon_0(R)$ decreases monotonically to reach a constant at $R = 0$. However, for $R = 0$, both integral diverge at opposite rates. Thus, to find the limiting value, it is better not to split $\epsilon_0(R)$ into two contributions but to consider them together. For small R, we have

$$\epsilon_0(R) = -\frac{1}{2\pi^2}\int_0^{\infty}dq \int_{\substack{|p_1|<1\\|p_1+q|>1}} dp_1 \int_{\substack{|p_2|<1\\|p_2+q|>1}} dp_2 \times \frac{[\ln q - \ln(p_1+p_2+q)][\ln(qR/2)+\gamma]}{q(p_1+p_2+q)} + O(R^2). \tag{13}$$

The integrations over p_1 and p_2 can be performed at this stage and it yields

$$\epsilon_0(R) = -\frac{1}{4\pi^2}\int_0^{\infty}\frac{\Lambda(q)}{q}[\ln(qR/2)+\gamma]dq, \tag{14}$$

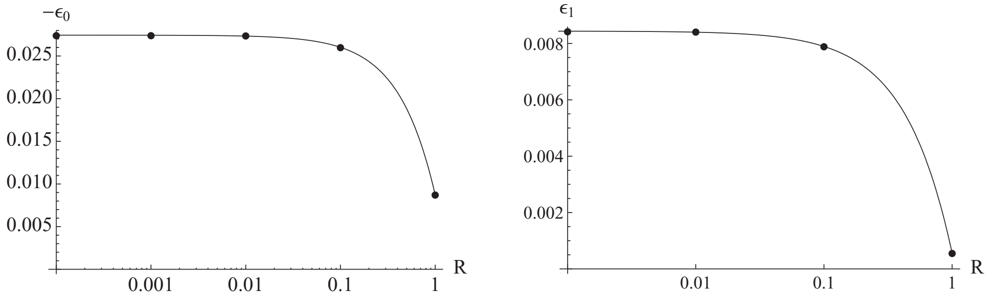

FIG. 1. Convergence of $\epsilon_0(R) = \epsilon_0^{\text{a}}(R) + \epsilon_0^{\text{b}}(R)$ (Eqs. (11) and (12)) and $\epsilon_1(R) = \pi^{-4}\sum_{k=1}^{8}\Xi_i(R)$ (explicit expressions given in Table II) with respect to R.

where γ is the Euler-Mascheroni constant[61] and

$$\begin{aligned}\Lambda(q) = {} & (2+q)[\ln(2+q)]^2 + (2-q)[\ln(2-q)]^2 \\ & - 2(1+\ln q)(2+q)\ln(2+q) \\ & - 2(1+\ln q)(2-q)\ln(2-q) \\ & - 4(\ln 2)^2 + 8\ln 2(1+\ln q)\end{aligned} \tag{15}$$

for $0 \le q \le 2$, and

$$\begin{aligned}\Lambda(q) = {} & (2+q)[\ln(2+q)]^2 + (2-q)[\ln(q-2)]^2 \\ & - 2(1+\ln q)(2+q)\ln(2+q) \\ & - 2(1+\ln q)(2-q)\ln(q-2) \\ & + 2(2+\ln q)q\ln q\end{aligned} \tag{16}$$

otherwise. Performing the last integration over the two distinct regions ($0 \le q \le 2$ and $q > 2$) gives two contributions that diverges as $\ln R$ for small R with opposite sign. Thus, the divergences cancel and we find

$$\epsilon_0 = \lim_{R\to 0} \epsilon_0(R) = -\frac{\pi^2}{360}, \tag{17}$$

which nicely reproduces the result obtained with a ring geometry.[56]

Because $\epsilon_c(r_s) = \epsilon_0 + O(r_s)$ (see below), ϵ_0 provides the *exact* value of the correlation energy at $r_s = 0$, and it is roughly -27.4 mhartree per electron. It is worth noting that, in most of the studies on 1D systems, a soft Coulomb operator is considered. For example, in Refs. 1 and 64, the authors used $R = 1$, yielding a correlation energy (-8.7 mhartree) more than three times smaller than the value obtained using the genuine Coulomb operator (i.e., $R = 0$). Moreover, using a quasi-1D model with a transverse harmonic potential, Casula *et al.* conclude that, in the high-density limit, the correlation energy vanishes quadratically with r_s.[65] This strikingly different prediction stresses the importance of employing a realistic Coulomb operator.

B. Third-order perturbation theory

Using the same approach, third-order perturbation theory gives[41]

$$\begin{aligned}\epsilon_1(R) = {} & \frac{1}{8\pi}\sum_{abcd}^{\text{occ}}\sum_{rs}^{\text{virt}} \frac{\langle ab\|rs\rangle\langle cd\|ab\rangle\langle rs\|cd\rangle}{\kappa_{a,b,r,s}\kappa_{c,d,r,s}} \\ & + \frac{1}{8\pi}\sum_{ab}^{\text{occ}}\sum_{rstu}^{\text{virt}} \frac{\langle ab\|rs\rangle\langle rs\|tu\rangle\langle tu\|ab\rangle}{\kappa_{a,b,r,s}\kappa_{a,b,t,u}} \\ & + \frac{1}{\pi}\sum_{abc}^{\text{occ}}\sum_{rst}^{\text{virt}} \frac{\langle ab\|rs\rangle\langle cs\|tb\rangle\langle rt\|ac\rangle}{\kappa_{a,b,r,s}\kappa_{a,c,r,t}} \\ & + \frac{1}{\pi}\sum_{abc}^{\text{occ}}\sum_{rst}^{\text{virt}} \frac{\langle ab\|rs\rangle\langle ar\|ct\rangle\langle rs\|ab\rangle}{\kappa_{a,b,r,s}^2},\end{aligned} \tag{18}$$

with $\kappa_{a,b,r,s} = \kappa_a + \kappa_b - \kappa_r - \kappa_s$. Equation (18) can be decomposed, using the same transformations as in (11) and (12), into eight distinct contributions

$$\epsilon_1(R) = \frac{1}{\pi^4}\sum_{k=1}^{8} \Xi_i(R). \tag{19}$$

TABLE II. Explicit expressions of the $\Xi_k(R)$'s and their regions of integration. $v_R(q) = K_0(|q|R)$ and $\int d^m p = \int \dots \int dp_1 \dots dp_m$.

$\Xi_k(R)$	Integral	Region of integration
$\Xi_1(R)$	$\int dq\, d^3p \frac{v_R(q)^3}{q(p_1+p_2+q)q(p_1+p_3+q)}$	$\lvert p_i\rvert < 1, \lvert p_i+q\rvert > 1$
$\Xi_2(R)$	$-\int dq\, d^3p \frac{v_R(q)^2 v_R(p_1-p_2)}{q(p_1+p_3+q)q(p_2+p_3+q)}$	$\lvert p_i\rvert < 1, \lvert p_i+q\rvert > 1$
$\Xi_3(R)$	$-2\int dq\, d^3p \frac{v_R(q)^2 v_R(p_1+p_2+q)}{q(p_1+p_2+q)q(p_1+p_3+q)}$	$\lvert p_i\rvert < 1, \lvert p_i+q\rvert > 1$
$\Xi_4(R)$	$-2\int dq\, d^3p \frac{v_R(q) v_R(p_1+p_2+q) v_R(p_2-p_3)}{q(p_1+p_2+q)q(p_1+p_3+q)}$	$\lvert p_i\rvert < 1, \lvert p_i+q\rvert > 1$
$\Xi_5(R)$	$-\int d^2q\, d^2p \frac{v_R(q_1)v_R(q_2)\left[v_R(q_1-q_2)-v_R(p_1-p_2)\right]}{q_1(p_1-p_2)q_2(p_1-p_2)}$	$\lvert p_1+q_1\rvert > 1, \lvert p_1+q_2\rvert > 1, \lvert p_1\rvert < 1$
		$\lvert p_2+q_1\rvert < 1, \lvert p_2+q_2\rvert < 1, \lvert p_2\rvert > 1$
$\Xi_6(R)$	$\frac{1}{2}\int d^2q\, d^2p \frac{v_R(q_1)v_R(q_2)\left[v_R(q_1-q_2)-v_R(p_1+p_2+q_1+q_2)\right]}{q_1(p_1+p_2+q_1)q_2(p_1+p_2+q_2)}$	$\lvert p_1+q_1\rvert > 1, \lvert p_1+q_2\rvert > 1, \lvert p_1\rvert < 1$
		$\lvert p_2+q_1\rvert > 1, \lvert p_2+q_2\rvert > 1, \lvert p_2\rvert < 1$
		$\lvert p_1+q_1\rvert < 1, \lvert p_1+q_2\rvert < 1, \lvert p_1\rvert > 1$
		$\lvert p_2+q_1\rvert < 1, \lvert p_2+q_2\rvert < 1, \lvert p_2\rvert > 1$
$\Xi_7(R)$	$\int dq\, d^3p \frac{v_R(q)^2\left[v_R(p_1-p_2)-v_R(p_1-p_2+q)\right]}{(q^2+q(p_1+p_3))^2}$	$\lvert p_i\rvert < 1, \lvert p_1+q\rvert > 1, \lvert p_3+q\rvert > 1$
$\Xi_8(R)$	$\int dq\, d^3p \frac{v_R(q)v_R(p_1+p_2+q)\left[v_R(p_1-p_3+q)-v_R(p_1-p_3)\right]}{(q^2+q(p_1+p_2))^2}$	$\lvert p_i\rvert < 1, \lvert p_1+q\rvert > 1, \lvert p_2+q\rvert > 1$

The explicit expressions of the $\Xi_k(R)$'s and their regions of integration are given in Table II.

Again, for $R = 0$, most of the integrals diverge. The first five terms have to be considered together, as well as the last two integrals while the sixth integral is finite. Evaluating numerically each contribution and extrapolating the result to $R = 0$ using the relation $\alpha R^\beta + \epsilon_1$ (see Fig. 1), we find

$$\epsilon_1 = \lim_{R\to 0} \epsilon_1(R) = +0.00844(7), \tag{20}$$

which is agreement with the exact numerical value (+0.008446) obtained for the ring geometry of Ref. 56. The error in (20) has been obtained by taking into account each numerical error estimate and extrapolating the overall error to $R = 0$.[66] We note that the present 1D UEG is one of the few systems where the r_s coefficient of the high-density expansion is known.[46,48]

In summary, we have shown that the high-density correlation energy expansion (1) of the 1D UEG is

$$\epsilon_c(r_s) = -\frac{\pi^2}{360} + 0.00845\, r_s + \dots . \tag{21}$$

We note that, contrary to the 2D and 3D UEGs, the expansion (21) does not contain any logarithm term up to first order in r_s, i.e., $\lambda_0 = \lambda_1 = 0$ (cf. Eq. (1)).

III. LDA FUNCTIONAL

For the 1D UEG, it is known[56,59] that the low-density (large-r_s) expansion of the correlation energy is

$$\begin{aligned}\epsilon_c(r_s) &= \frac{\eta_0}{r_s} + \frac{\eta_1}{r_s^{3/2}} + \dots\\ &= -\frac{\ln(\sqrt{2\pi}) - 3/4}{r_s} + \frac{0.359933}{r_s^{3/2}} + \dots . \end{aligned}\tag{22}$$

Using the "robust" interpolation proposed by Cioslowski[68] and the high- and low-density expansions (21) and (22), the correlation energy can be approximated by

$$\epsilon_c^{\text{LDA}}(r_s) = t^2 \sum_{j=0}^{3} c_j t^j (1-t)^{3-j}, \tag{23}$$

TABLE III. Reduced correlation energy ($-\epsilon_c(r_s)$ in mhartree) for various r_s. The DMC results are computed using the CASINO software[67] and are taken from Refs. 54 and 56. Subscripts represent the statistical errors in the last digits. The deviation with respect to the DMC result is given in parenthesis.

r_s	DMC	This work
0	…	27.416
0.2	25.91_1	25.90 (−0.01)
0.5	23.962_1	24.021 (+0.059)
1.0	$21.444\,1_2$	21.518 (+0.074)
2.0	$17.922\,02_7$	17.927 (+0.005)
5.0	$12.317\,74_2$	12.220 (−0.097)
10.0	$8.292\,096_9$	8.201 (−0.092)
15.0	$6.319\,404_4$	6.251 (−0.069)
20.0	$5.132\,504_2$	5.081 (−0.052)

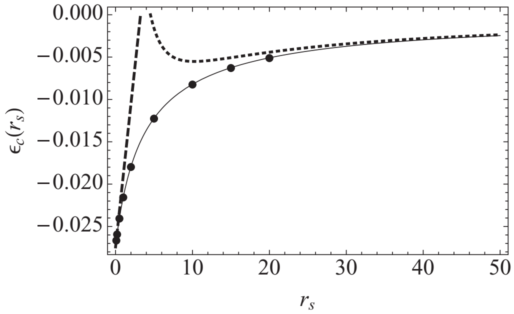

FIG. 2. $\epsilon_c(r_s)$ given by Eq. (23) as a function of r_s (solid line). DMC results are shown by black dots. The small-r_s expansion of Eq. (21) (dashed line) and large-r_s approximation of Eq. (22) (dotted line) are also shown.

with

$$t = \frac{\sqrt{1 + 4\,k\,r_s} - 1}{2\,k\,r_s} \tag{24}$$

and

$$c_0 = k\,\eta_0, \quad c_1 = 4\,k\,\eta_0 + k^{3/2}\eta_1, \tag{25}$$

$$c_2 = 5\,\epsilon_0 + \epsilon_1/k, \quad c_3 = \epsilon_1, \tag{26}$$

where $k = 0.414254$ is a scaling factor which is determined by a least-square fit of the DMC data given in Refs. 54 and 56.

We disagree with the last comment made in Ref. 68, which claims that this type of interpolation is not applicable to cases where the high- and low-density asymptotic expansions pertain to *de facto* different states, e.g., the 3D UEG. We claim that the non-applicability of such an interpolation is only due to the presence of logarithmic terms in the 2D and 3D UEGs. However, in our case, the 1D UEG does not involve any non-analytical terms. Thus, the methodology of Ref. 68 is applicable in the present case.

The results using the new correlation functional (23) are compared to the DMC calculations of Refs. 54 and 56. The results are gathered in Table III and depicted in Fig. 2. For $0.2 \le r_s \le 20$, the LDA and DMC correlation energies agree to within 0.1 mhartree, which is remarkable given the simplicity of the functional. Overall, our LDA correlation functional gives accurate estimates of the correlation energy.

IV. CONCLUSION

In this article, we have shown that the expression of the high-density correlation energy for the 1D UEG is $\epsilon_c(r_s) = -0.02742 + 0.00845 r_s + \dots$. Combining these new results with the low-density correlation energy expansion $\epsilon_c(r_s) = -[\ln(\sqrt{2\pi}) - 3/4]\, r_s^{-1} + 0.359933\, r_s^{-3/2} + \dots$ and the available DMC data, we have proposed a LDA correlation functional, which yields satisfactory estimates of the correlation energy at high, intermediate and low densities. We believe these new results will be valuable for electronic structure calculations (especially within DFT).

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