Spin resolution of the electron-gas correlation energy: Positive same spin contributions

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The negative correlation energy $\epsilon_c(r_s,\zeta)$ per particle of a uniform electron gas of density parameter r_s and spin polarization ζ is well known, but its spin resolution into $\uparrow\downarrow$, $\uparrow\uparrow$, and $\downarrow\downarrow$ contributions is not. Widely used estimates are incorrect, and hamper the development of reliable density functionals and pair distribution functions. For the spin resolution, we present interpolations between high- and low-density limits that agree with available quantum Monte Carlo data. In the low-density limit for $\zeta=0$, we find that the same-spin correlation energy is unexpectedly positive, and we explain why. We also estimate the $\uparrow$ and $\downarrow$ contributions to the kinetic energy of correlation.

I. INTRODUCTION

The uniform electron gas is a paradigm for density-functional theory,[1–3] the most widely used method for electronic structure calculations in both condensed-matter physics and quantum chemistry. The effects of exchange and correlation can be evaluated and understood in the uniform-density limit, and then transferred to realistic systems. This is done not only in the local spin-density (LSD) approximation but also beyond LSD in generalized gradient approximations (GGA's), meta-GGA's, and hybrid functionals.[3] The correlation energy $\epsilon_c(r_s,\zeta)$ per particle in a uniform gas of density parameter $r_s=(4\pi n a_0^3/3)^{-1/3}$ and spin polarization $\zeta=(n_\uparrow -n_\downarrow)/n$ (where n_σ is the density of spin-σ electrons and $n=n_\uparrow+n_\downarrow$) is well known, for example, from quantum Monte Carlo (QMC) studies[4,5] that have been parametrized[6–8] to respect known limits, but the spin resolution of ϵ_c into $\uparrow\downarrow$, $\uparrow\uparrow$, and $\downarrow\downarrow$ contributions is not known. In this work, we determine the spin resolution for all r_s and ζ as an interpolation between high- and low-density limits, consistent with $\zeta=0$ QMC data.[5]

This spin resolution is of interest in its own right, and can also be used in several ways: (i) Some beyond LSD correlation energy functionals need a missing spin resolution[9] or have been constructed[10–13] on the basis of the exchange-like ansatz of Stoll *et al.*[14]

$$E_c^{\uparrow\downarrow}[n_\uparrow,n_\downarrow] \approx E_c[n_\uparrow,n_\downarrow] - E_c[n_\uparrow,0] - E_c[0,n_\downarrow] \quad (1)$$

for the uniform gas. This assumption was shown (using QMC results) to be inaccurate for $\zeta=0$ (see Fig. 1) in Ref. 15, although the significance of this observation for density-functional theory was not fully recognized there. Our work provides a firmer basis than Eq. (1) for such constructions. (ii) Correlation energy functionals such as the local spin density[1] and generalized gradient approximations,[16] etc.,[17] can alternatively be constructed without a spin resolution, but their later spin resolution (to permit comparison or combination with correlated-wave-function results[12,18,19]) demands such a resolution for uniform densities. (iii) A sophisticated analytic model[21] is now available for the pair

distribution function[15,20,21] $g_{xc}(r_s,\zeta,u)$ of the uniform gas for all r_s and ζ. Our present work provides the missing ingredient needed to find the corresponding spin-resolved pair distribution function, which could serve as the starting point for the development of density functionals such as spin-resolved weighted density approximations.[22] (iv) An estimate can be made for the ζ dependence of the $\uparrow$ and $\downarrow$ contributions to the kinetic energy of correlation, a key ingredient for the approach to spin dynamics of Qian and Vignale[23] and also for the momentum distribution[24] of a spin-polarized electron gas.

We shall first derive exact limits at high densities ($r_s \rightarrow 0$) and extreme low densities ($r_s \rightarrow \infty$) using simple physical arguments. In the latter limit, we find that the same spin contribution to the correlation energy can be positive, and we provide an intuitive physical picture to explain this feature. While the total correlation energy must be negative, individual terms of it (e.g., the kinetic energy of correlation) can be positive. We then build up and discuss our interpolation formulas.

II. DEFINITIONS

Correlation effects arise from the Coulomb interaction, which is a two-body operator. When evaluating the energy of the system $\langle\Psi|H|\Psi\rangle$ one can split the sum over the electron spins into $\uparrow\downarrow$, $\uparrow\uparrow$, and $\downarrow\downarrow$ contributions. The corresponding splitting of the correlation energy of the uniform electron gas,

$$\epsilon_c(r_s,\zeta) = \epsilon_c^{\uparrow\downarrow}(r_s,\zeta) + \epsilon_c^{\uparrow\uparrow}(r_s,\zeta) + \epsilon_c^{\downarrow\downarrow}(r_s,\zeta) \quad (2)$$

is the object of this Rapid Communication. The real-space analysis of the spin-resolved correlation energies $\epsilon_c^{\sigma\sigma'}(r_s,\zeta)$ is provided by the correlation holes $n_{\sigma'}\bar{g}_c^{\sigma\sigma'}(r_s,\zeta,u)$ (see, e.g., Ref. 21), where $u=|\mathbf{r}_1-\mathbf{r}_2|$ is the electron-electron distance

$$\epsilon_c^{\sigma\sigma'}(r_s,\zeta) = 2\pi\frac{n_\sigma}{n}\int_0^\infty n_{\sigma'}\bar{g}_c^{\sigma\sigma'}(r_s,\zeta,u)u\,du. \quad (3)$$
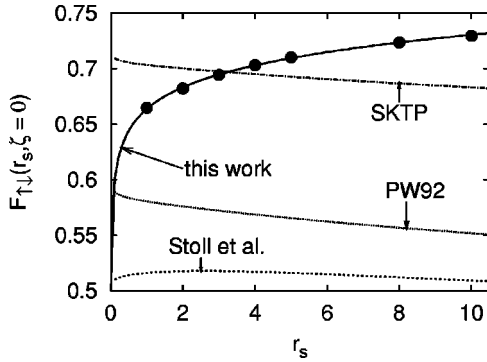
FIG. 1. Fraction of $\uparrow\downarrow$ correlation energy, $F_{\uparrow\downarrow}(r_s,\zeta)$ $=\epsilon_c^{\uparrow\downarrow}(r_s,\zeta)/\epsilon_c(r_s,\zeta)$ at $\zeta=0$. Our Eq. (9) is compared with the GSB [Gori-Giorgi, Sacchetti, and Bachelet (Ref. 15)] values extracted from QMC (Ref. 5) data ($\bullet$), and with the Stoll et al.[14] PW92 (Ref. 20), and SKTP [Schmidt, Kurth, Tao, and Perdew (Ref. 28)] scaling relations. Valence electrons have $2 \lesssim r_s \lesssim 6$.

The correlation hole $n_{\sigma'} \bar{g}_c^{\sigma\sigma'}(r_s,\zeta,u)$ describes the change (due to correlation only) of spin-σ' electron density at **u**, when a spin-σ electron is at the origin. $\bar{g}_c$ is averaged over coupling strength, while g_c is for full coupling strength. We define fractions $F_{\sigma\sigma'}(r_s,\zeta)$ such that

$$\epsilon_c^{\sigma\sigma'}(r_s,\zeta) = \epsilon_c(r_s,\zeta) F_{\sigma\sigma'}(r_s,\zeta), \tag{4}$$

and we investigate their properties. In what follows, we use Hartree atomic units, and the parametrization of $\epsilon_c(r_s,\zeta)$ and its limits from Ref. 8.

III. EXACT LIMITS

When $r_s \to 0$, the Coulomb electron-electron interaction can be treated as a perturbation to the noninteracting Fermi gas. The first-order (in the Coulomb potential) correction term gives the exchange energy $\epsilon_x = \epsilon_x^{\uparrow\uparrow} + \epsilon_x^{\downarrow\downarrow}$, where $\epsilon_x^{\uparrow\uparrow}$ $= -(3/8\pi\alpha r_s)(1+\zeta)^{4/3}$, $\epsilon_x^{\downarrow\downarrow} = -(3/8\pi\alpha r_s)(1-\zeta)^{4/3}$, and $\alpha = (9\pi/4)^{-1/3}$. As for correlation, the real-space analysis of the exchange energies is provided by the exchange holes $n_\sigma[g_x^{\sigma\sigma}(\zeta,u/r_s)-1]$, which are analytically known (see, e.g., Ref. 21).

The second-order correction to the energy of the noninteracting Fermi gas is the sum of a direct term and a second-order exchange term. Only the direct term diverges, and,

when a cutoff $\propto 1/\sqrt{r_s}$ (due to Thomas-Fermi screening effects) at small wave vectors is introduced, gives rise to a leading term in $\epsilon_c(r_s,\zeta)$, equal to $c_0(\zeta)\ln r_s$. The function $c_0(\zeta)$ is exactly known.[25] The direct term [Eq. (5.110) of Ref. 26] can be divided into $\uparrow\downarrow$, $\uparrow\uparrow$, and $\downarrow\downarrow$ excitation pairs to derive

$$F_{\uparrow\uparrow}(r_s \to 0,\zeta) \equiv F_{\uparrow\uparrow}^{HD}(\zeta) = \frac{1+\zeta}{4I(\zeta)}, \tag{5}$$

with $I(\zeta) = c_0(\zeta)/c_0(0)$, as conjectured in Ref. 25. [Since $F_{\downarrow\downarrow}(r_s,\zeta) = F_{\uparrow\uparrow}(r_s,-\zeta)$ and $F_{\uparrow\downarrow} = 1 - F_{\uparrow\uparrow} - F_{\downarrow\downarrow}$, we only report formulas for $\uparrow\uparrow$.] The Stoll et al. ansatz of Eq. (1) is thus correct for $r_s \to 0$ (and for all r_s when $|\zeta|=1$, but not otherwise).

In the opposite or strong-interaction limit, $r_s \to \infty$, the long-range Coulomb repulsion between the electrons becomes dominant with respect to the kinetic energy, and thus with respect to statistics; Coulomb repulsion suppresses electron-electron overlap so that the electrons no longer know they are fermions. In this limit, the total energy becomes independent[4,5,8,27,21] of ζ. Its leading term in the $r_s \to \infty$ expansion is equal to $-d_1/r_s$, where[8] $d_1 \simeq 0.892$, and is purely potential energy, with no kinetic energy contribution. In this limit, the total energy is thus equal to the exchange-correlation energy $\epsilon_{xc} = \epsilon_x + \epsilon_c$. Moreover, since the statistics becomes irrelevant, we expect that

$$\left(\frac{2}{1+\zeta}\right)^2 \epsilon_{xc}^{\uparrow\uparrow} = \left(\frac{2}{1-\zeta}\right)^2 \epsilon_{xc}^{\downarrow\downarrow} = \frac{2}{(1-\zeta^2)} \epsilon_{xc}^{\uparrow\downarrow} = \epsilon_{xc}, \tag{6}$$

where the prefactors take into account the available numbers of pairs. In other words, we expect that $\int_0^\infty du\, 4\pi u^2 \bar{g}_{xc}^{\sigma\sigma'}/u$ becomes independent of σ and σ', so that spin structure becomes unimportant for the exchange-correlation and total energies (although very important for the correlation energy alone). Then the $F_{\sigma\sigma'}(r_s \to \infty,\zeta) \equiv F_{\sigma\sigma'}^{LD}(\zeta)$ are given by

$$F_{\uparrow\uparrow}^{LD}(\zeta) = \frac{3(1+\zeta)^{4/3} - 2\pi\alpha(1+\zeta)^2 d_1}{3[(1+\zeta)^{4/3} + (1-\zeta)^{4/3}] - 8\pi\alpha\, d_1}. \tag{7}$$

The high and low density $F_{\sigma\sigma'}$ are displayed in Fig. 2. We see that, in the spin-unpolarized gas, the same spin ($\uparrow\uparrow$ $+ \downarrow\downarrow$) contribution to the correlation energy is 50% when $r_s \to 0$ but roughly 0 when $r_s \to \infty$. This can be understood in
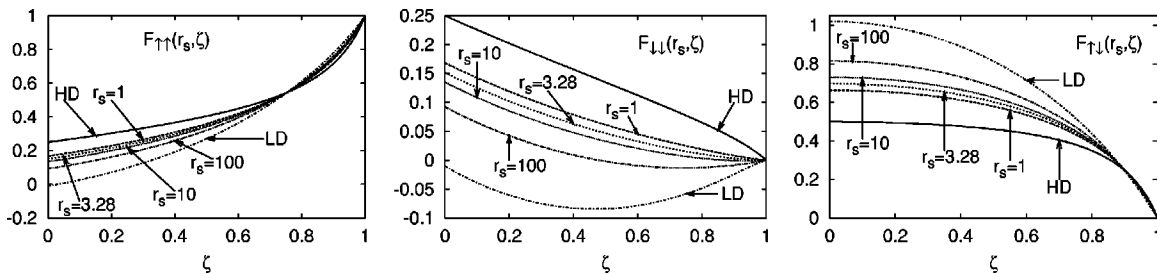

FIG. 2. Spin resolution $F_{\sigma\sigma'}(r_s,\zeta) = \epsilon_c^{\sigma\sigma'}(r_s,\zeta)/\epsilon_c(r_s,\zeta)$ as a function of ζ for different r_s. The high-density (HD) and low-density (LD) limits are given in Eqs. (5) and (7). The $r_s = 3.28$ curves correspond to the SKTP (Ref. 28) scaling relation of Eq. (8), while for other density values ($r_s = 1$, 10, and 100) our interpolation formulas of Eq. (9) have been used.
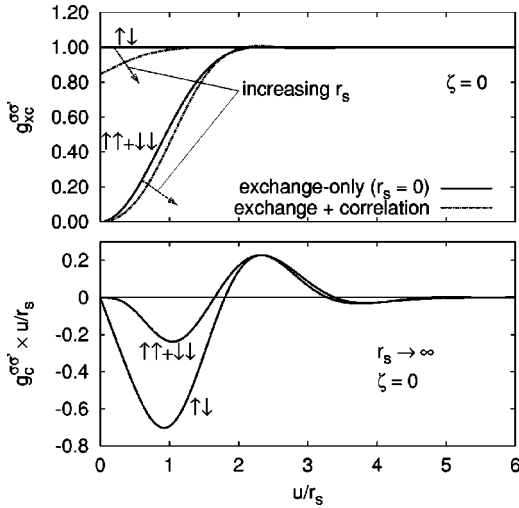
FIG. 3. Upper panel: the spin-resolved pair distribution functions for the paramagnetic gas. The dashed arrows show the trend of the holes as the coupling strength r_s is increased. Lower panel: real-space analysis of the correlation energy in the extreme low-density limit for the paramagnetic gas. The results are from the model of Ref. 21.

a simple way. The exchange hole seen by the same spin electrons is deep for electron-electron distances $u \lesssim r_s$, as shown in the upper panel of Fig. 3 (solid line, $\uparrow\uparrow + \downarrow\downarrow$). But there is a second length scale, the Thomas-Fermi screening length $\sqrt{r_s}$. For $r_s \rightarrow 0$, the important correlations, which determine the leading term ($\propto \ln r_s$) of ϵ_c, arise from this second length scale, $\sqrt{r_s} \gg r_s$, and are essentially unaffected by exchange: the electrons that participate in this correlation have no way to know if the electron at $u = 0$ is spin-$\uparrow$ or spin-$\downarrow$, so by symmetry the same spin and opposite spin correlation energies are equal. In the opposite limit $r_s \rightarrow \infty$ the antiparallel-spin correlation hole can get deep for $u \lesssim r_s$, as shown in the upper panel of Fig. 3.

As r_s increases, $g_{xc}^{\uparrow\downarrow}$ deviates more and more from its noninteracting value (equal to 1 for all u), the only constraint being its positiveness. But the same spin correlation hole is "blocked" from doing this by the exchange hole (see, again, the upper panel of Fig. 3). Thus the system minimizes its energy by focussing the correlation on opposite spin pairs. In the extreme low-density limit, a simple qualitative picture can be obtained by using the correlation-hole model of Ref. 21 (in which energetically unimportant long-range oscillations are averaged out); in the lower panel of Fig. 3, we report the corresponding real-space analysis of $\epsilon_c^{\uparrow\downarrow}$ and $\epsilon_c^{\uparrow\uparrow} + \epsilon_c^{\downarrow\downarrow}$ for $r_s \rightarrow \infty$. We see that the same spin correlation hole for $u \lesssim r_s$ cannot get as deep as the opposite spin one.

Figure 2 also shows that in the spin-unpolarized gas the same spin correlation energy is slightly positive ($F_{\sigma\sigma} < 0$) when $r_s \rightarrow \infty$. In this limit, the electrons correlate strongly, and the exchange-correlation holes show a high first-neighbor peak at $u \approx 2r_s$ (lower panel of Fig. 3). If the only effect of same spin correlation were to push same spin electrons away from the region of small u and pile them up at $u \approx 2r_s$, then [by the sum rule integral $\int du \, 4\pi u^2 n_\sigma \bar{g}_c^{\sigma\sigma}(u)$

$=0$] the same spin correlation energy [Eq. (3) with $\sigma = \sigma'$] would necessarily be negative. So, what must really happen is that the same spin electrons that accumulate in the peak at $u \approx 2r_s$ include some that have been pushed out from $u \ll 2r_s$ and some that have been pulled in from $u \gg 2r_s$. This is again illustrated in the lower panel of Fig. 3. We interpret the second zero of g_c, which appears at large u but only at large r_s, as the energetically important remnant of the long-range oscillation of g_c in a Wigner crystal.

Positive same spin correlation energy may be an exotic effect, but the blockage of negative same-spin correlation also occurs in a nonmagnetic Mott insulator, e.g., an expanded lattice of hydrogen atoms where Coulomb correlation suppresses the $(1s)^2$ configuration on a given site. The blockage of same spin correlation occurs even in a weakly correlated spin-unpolarized system when the correlation hole is spatially constrained, as for an atom.[12,18,19] In the neon atom, the true (as cited in Ref. 18) antiparallel-spin correlation energy is 65% of its LSD value, while the true parallel-spin correlation energy is only 30% of its LSD value.

IV. INTERPOLATION BETWEEN HIGH AND LOW DENSITY

We want to build up interpolation formulas for $F_{\sigma\sigma'}(r_s, \zeta)$ that include all the information available on the spin resolution of ϵ_c. Besides the high- and low-density limits, we have data for $F_{\sigma\sigma'}(r_s, 0)$, in the range $0.8 \le r_s \le 10$. These data have been obtained in Ref. 15 [Gori-Giorgi, Sacchetti, and Bachelet (GSB)] by integrating spin-resolved QMC correlation holes.[5] Moreover, Schmidt, Kurth, Tao, and Perdew[28] (SKTP), starting from nearly exact limits of the spin-resolved correlation holes, proposed a scaling relation that is in agreement with the GSB data at $r_s = 3.28$, and that, as shown in Fig. 2 (curves labeled with "$r_s = 3.28$"), lies in between the high- and the low-density limits with a very "reasonable" shape. The SKTP scaling should thus be a good "intermediate point" for our interpolation formulas. We thus define

$$F_{\uparrow\uparrow}^{\text{SKTP}}(\zeta) = \left(\frac{1+\zeta}{2}\right)^{11/6} \frac{\epsilon_c(3.28,1)}{\epsilon_c(3.28,\zeta)}, \qquad (8)$$

and we parametrize $F_{\sigma\sigma'}(r_s, \zeta)$ as

$$F_{\sigma\sigma'}(r_s, \zeta) = \frac{F_{\sigma\sigma'}^{\text{HD}}(\zeta) + A_{\sigma\sigma'}(\zeta)\sqrt{r_s} + B F_{\sigma\sigma'}^{\text{LD}}(\zeta) r_s}{1 + C\sqrt{r_s} + B r_s}. \quad (9)$$

$A_{\sigma\sigma'}(\zeta)$ is found by requiring that $F_{\sigma\sigma'}(3.28, \zeta) = F_{\sigma\sigma'}^{\text{SKTP}}(\zeta)$, i.e.,

$$A_{\sigma\sigma'}(\zeta) = \frac{F_{\sigma\sigma'}^{\text{SKTP}}(\zeta) - F_{\sigma\sigma'}^{\text{HD}}(\zeta)}{\sqrt{3.28}} + C F_{\sigma\sigma'}^{\text{SKTP}}(\zeta)$$
$$+ B\sqrt{3.28}[F_{\sigma\sigma'}^{\text{SKTP}}(\zeta) - F_{\sigma\sigma'}^{\text{LD}}(\zeta)]. \quad (10)$$

The form of Eq. (9) is motivated by the expression for the correlation energy given in Ref. 7. The parameters B and C are fixed by a best fit of $F_{\sigma\sigma'}(r_s, 0)$ to the GSB data for r_s

$\in[0.8,10]$: $B=0.178488$, $C=2.856$. In Fig. 1, our $F_{\uparrow\downarrow}(r_s,0)$ is compared with the GSB data,[15] and with the widely used Stoll et al.[14] ansatz of Eq. (1), which strongly underestimates the fraction of $\uparrow\downarrow$ correlation energy at metallic and lower densities. The results for the paramagnetic gas corresponding to other proposed scaling relations are also shown. Our interpolation formulas as functions of ζ, at $r_s=1,10$, and 100, are displayed in Fig. 2.

V. KINETIC ENERGY OF CORRELATION

Defining[29] $\epsilon_c^{\uparrow}=\epsilon_c^{\uparrow\uparrow}+\frac{1}{2}\epsilon_c^{\uparrow\downarrow}$ (with a similar equation for $\downarrow$), the adiabatic connection between the noninteracting and interacting limits for a given density suggests estimating the $\uparrow$ and $\downarrow$ contributions (from the one-particle density matrix) to the kinetic energy of correlation $t_c=t_c^{\uparrow}+t_c^{\downarrow}$ as[29]

$$t_c^{\sigma}(r_s,\zeta)\approx-\frac{\partial}{\partial r_s}[r_s\epsilon_c^{\sigma}(r_s,\zeta)], \qquad (11)$$

although as Ref. 30 points out there is only one coupling constant with a Hellmann-Feynman theorem, not one for each σ. Taking Eq. (11) as a plausible approximation, we find that the corresponding result for $t_c^{\uparrow}-t_c^{\downarrow}$ is in reasonable agreement with the scaling relation given in Eq. (29) of Ref. 23. (For $r_s\lesssim5$, the difference is less than 3.5%.) Via Eq.

(11), we also confirm that, for $1\lesssim r_s\lesssim10$, the quantity $(t_c^{\uparrow}-t_c^{\downarrow})/t_c(r_s,\zeta)$ is almost independent of r_s, as recently found in a more sophisticated calculation within the Singwi-Tosi-Land-Sjöland approximation.[31]

VI. CONCLUSIONS

In summary, we have found the spin resolution of the electron gas correlation energy, via an approach applied to but not restricted to the three-dimensional uniform electron gas. Our results can be used to understand correlation in more realistic systems, and to construct improved density functionals and pair distribution functions. We have found that the same spin correlation energy can be unexpectedly but understandably positive. We have also provided support for resolutions[23,31] of the kinetic energy of correlation into $\uparrow$ and $\downarrow$ terms. It is further possible to show that the positive spin stiffness of correlation[6,8] has positive $\uparrow\downarrow$ and negative $\uparrow\uparrow+\downarrow\downarrow$ contributions.

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