Wavevector analysis of the jellium exchange-correlation surface energy in the random-phase approximation: detailed support for nonempirical density functionals

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We report the first three-dimensional wavevector analysis of the jellium exchange-correlation (xc) surface energy in the random-phase approximation (RPA). The RPA accurately describes long-range xc effects which are challenging for semi-local approximations, since it includes the universal small-wavevector behavior derived by Langreth and Perdew. We use these rigorous RPA calculations for jellium slabs to test RPA versions of nonempirical semi-local density-functional approximations for the xc energy. The local spin density approximation (LSDA) displays cancelling errors in the small and intermediate wavevector regions. The PBE GGA improves the analysis for intermediate wavevectors, but remains too low for small wavevectors (implying too-low jellium xc surface energies). The nonempirical meta-generalized gradient approximation of Tao, Perdew, Staroverov, and Scuseria (TPSS meta-GGA) gives a realistic wavevector analysis, even for small wavevectors or long-range effects. We also study the effects of slab thickness and of short-range corrections to RPA.

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I. INTRODUCTION

Modern electronic-structure calculations for atoms, molecules, and solids usually rely upon Kohn-Sham (KS) density-functional theory (DFT), in which only $E_{xc}[\rho]$, the exchange-correlation (xc) energy as a functional of electron density, must be approximated. Semi-empirical approximations tend to be limited to systems that resemble those in the fitted data set (typically small molecules), but nonempirical ones are constructed to satisfy universal constraints and so should have a wider range of applicability. For example, it is expected that a good description of chemical reactions at a solid surface requires a good description of both the molecules and the surface.

Jellium is a simple model of a simple metal, in which the valence electrons are neutralized by a uniform positive background that extends up to a sharp planar surface. The apparent success of the simplest density functional, the local spin density approximation (LSDA), for the jellium surface energy motivated early interest in density functionals and in refinements of the LSDA such as the generalized gradient approximation (GGA).

It was therefore a matter of some concern when wavefunction-based Fermi HyperNetted-Chain (FHNC) and fixed-node Diffusion Monte Carlo (DMC) calculations for jellium slabs (and their extrapolation to infinite thickness) predicted surface energies considerably higher than those obtained in the LSDA. Indeed, DMC is usually a gold standard of accuracy. However, it encounters special difficulties for jellium slabs; furthermore, the large deviations between the available DMC and LSDA calculations have been attributed in part to inconsistency between the energy of the inhomogeneous system and that of the corresponding homogeneous electron gas. Recent approaches have all suggested that the actual jellium surface energies are only a little higher than those obtained in the LSDA. The jellium surface-energy story is presented in full detail in Ref. 16.

In this paper, we perform a detailed analysis of exchange and correlation in jellium slabs, exact at the level of the random phase approximation (RPA), to show that the most refined nonempirical density functional, the meta-generalized gradient approximation of Tao, Perdew, Staroverov, and Scuseria (TPSS meta-GGA), can account even for the most long-ranged xc effects at a jellium surface. This is a considerable achievement for a semi-local functional that is inherently more reliable for short-ranged effects than for long-ranged ones. RPA is known to be correct at long range; because it has serious deficiencies at short-range and, therefore, cannot be compared to standard versions of the semi-local functionals, we use RPA versions of these functionals in this test.

In order to separate long-range and short-range xc effects, we look at the surface contribution to the spherically-averaged real-space xc hole, averaged over the electron density of the system, and its Fourier transform (wavevector analysis). Langreth and Perdew showed that the exact xc energy of an arbitrary inhomogeneous system can be obtained from a three-dimensional (3D) Fourier transform of the spherical average of the xc hole density, which is a function of a 3D wavevector $k$. In the case of a plane-bounded electron gas, this wavevector-dependent spherical average is dominated at long wavelengths ($k \to 0$) by the zero-point energy-shift of the newly created surface collective oscillations (surface plas-
mons) and takes a simple analytical form. This known limit has been used to carry out a wavevector interpo-
lation correction to LSDA, PBE-GGA, and TPSS-
metaGGA xc surface energies. The wavevector interpo-
lration corrections to these functionals were control-
led by using the exact RPA values reported in Ref. 12 and
led to a consistent set of predicted surface energies.16

In a DFT context, the RPA is based upon the
time-dependent Hartree approximation for the density-
response function but replacing the occupied and unoc-
cupied single-particle Hartree orbitals and energies by the
the corresponding eigenfunctions and eigenvalues of the KS
Hamiltonian of DFT. Hence, it describes the exchange
energy and the long-range part of the correlation energy
correctly. Essentially exact RPA surface energies were
evaluated from single-particle LSDA orbitals and energies
in Ref. 12. These calculations provide an accurate
standard against which approximate density functionals
(in their RPA versions) can be tested and normed. The
RPA versions of LSD and GGA were reported in Refs. 18
and 19, respectively. Because RPA is not self-correlation-
free, the GGA for RPA correlation is its own meta-GGA.
The RPA version of the nonempirical TPSS-meta-GGA
was investigated in Ref. 10.

Unless stated otherwise, atomic units are used
throughout, i.e., $\varepsilon^2 = \hbar = m_e = 1$.

II. THEORETICAL FRAMEWORK

The exact xc energy, $E_{xc}[n]$, of an arbitrary inhomoge-
neous system of density $n(r)$ can be obtained from the
spherical average $\bar{n}_{xc}(r, u)$ of the coupling-constant averaged
xc hole density $\bar{n}_{xc}(r, r')$ at $r'$ around an electron at
$r$, as follows:16

$$E_{xc}[n] = \int d\mathbf{r} \, n(r) \, \varepsilon_{xc}[n](r), \tag{1}$$

where $\varepsilon_{xc}[n](r)$ represents the xc energy per particle at
point $r$:

$$\varepsilon_{xc}[n](r) = 4 \int_{0}^{\infty} dk \, \int_{0}^{\infty} du \, u^2 \frac{\sin ku}{ku} \, \bar{n}_{xc}(r, u), \tag{2}$$

with

$$\bar{n}_{xc}(r, u) = \frac{1}{4\pi} \int d\Omega \, \bar{n}_{xc}(r, r'), \tag{3}$$
d$\Omega$ being a differential solid angle around the direction
of $\mathbf{u} = r' - r$.

The xc surface energy, $\sigma_{xc}$, is obtained by subtracting
from the xc energy $E_{xc}[n]$ of a semi-infinite electron
system the corresponding energy $E_{xc}^{\text{unif}}(n)$ of a uniform
electron gas. In a jellium model, in which the electron
system is translationally invariant in the plane of the sur-
face, and assuming the surface to be normal to the $z$-axis,
one finds

$$\sigma_{xc} = \int_{0}^{\infty} d\left(\frac{k}{2k_F}\right) \gamma_{xc}(k), \tag{4}$$

where

$$\gamma_{xc}(k) = 2 \frac{k_F}{\pi} \int_{-\infty}^{+\infty} dz \, n(z) \, b_{xc}(k, z), \tag{5}$$

with $k_F = (3\pi^2 n)^{1/3}$, $n$ being the background density, and

$$b_{xc}(k, z) = 4\pi \int_{0}^{\infty} du \, u^2 \frac{\sin ku}{ku} \left[\bar{n}_{xc}(z, u) - \bar{n}_{xc}^{\text{unif}}(u)\right]. \tag{6}$$

Alternatively, one can introduce Eq. (3) into Eq. (1) to
find:

$$b_{xc}(k, z) = \frac{1}{2} \int_{-k}^{k} \frac{dk_z}{k} \int_{-\infty}^{+\infty} dz' \, e^{ik_z(z-z')} \times \bar{n}_{xc}(k; z, z') - \bar{n}_{xc}^{\text{unif}}(k), \tag{7}$$

with $k_0 = \sqrt{k^2 - k_z^2}$, and $\bar{n}_{xc}(k_0; z, z')$ and $\bar{n}_{xc}^{\text{unif}}(k)$ representing Fourier transforms of the coupling-constant averaged xc hole densities $\bar{n}_{xc}(r, r')$ and $\bar{n}_{xc}^{\text{unif}}(r, r')$, respectively. At long wavelengths ($k \rightarrow 0$), one finds the exact

$$\gamma_{xc}(k) = \frac{k_F}{4\pi} \left(\omega_s - \omega_p\right) k, \tag{8}$$

which only depends on the bulk- and surface-plasmon
energies $\omega_p = (4\pi n)^{1/2}$ and $\omega_s = \omega_p/\sqrt{2}$, and does not
depend, therefore, on the electron-density profile at the
surface.

The spherical average $\bar{n}_{xc}(z, u)$ entering Eq. (3) can be
obtained within local or semi-local density-functional
approximations (such as LSDA, PBE GGA, and TPSS
meta-GGA) from models16,19,21,22 that require knowl-
dge of the xc hole density $\bar{n}_{xc}^{\text{unif}}(u)$ of a uniform electron
gas. Alternatively, rigorous calculations of $\bar{n}_{xc}^{\text{unif}}(k)$ and
the fully nonlocal $\bar{n}_{xc}(k_0; z, z')$ entering Eq. (7) can be
carried out from knowledge of the $\lambda$-dependent density-
response functions $\chi_\lambda^{\text{unif}}(k, \omega)$ and $\chi_\lambda(k_0; \omega; z, z')$, respec-
tively, defined by adiabatically switching on the $e$-$e$
interaction via the coupling constant $\lambda$ and by adding, at the
same time, an external potential so as to maintain the
true ($\lambda = 1$) ground-state density in the presence of the
modified $e$-$e$ interaction.25,26 By using the fluctuation-
dissipation theorem25,26 one finds:

$$\bar{n}_{xc}^{\text{unif}}(k) = \frac{1}{n} \left[\frac{1}{\pi} \int_{0}^{1} d\lambda \int_{0}^{\infty} d\omega \chi_\lambda^{\text{unif}}(k, i\omega) - \bar{n}\right] \tag{9}$$

and

$$\bar{n}_{xc}(k_0; z, z') = \frac{1}{\pi n(z)} \int_{0}^{1} d\lambda \int_{0}^{\infty} d\omega \chi_\lambda(k_0; i\omega; z, z') \delta(z - z'). \tag{10}$$

With the aim of testing the performance of local and
semi-local density-functional approximations for the xc
surface energy, we compare these (local and semi-local)
calculations [obtained from Eq. (3)] to their fully nonlocal counterparts [obtained from Eq. (7) with the aid of Eqs. (10) and (11)] at the same level of approximation, which we choose to be the RPA. On the one hand, we evaluate \( \gamma_{xc}(k) \) from RPA versions (LSDA-RPA, PBE-RPA, and TPSS-RPA) of the local (or semilocal) \( n_{xc}(z, u) \) entering Eq. (4) based on the RPA xc hole density \( n_{xc}^{\text{unif}}(u) \) of a uniform electron gas. On the other hand, we evaluate \( \gamma_{xc}(k) \) from a fully nonlocal version (exact-RPA) of \( n_{xc}(k||z, z') \) entering Eq. (1) based on \( \chi^\lambda(k||\omega; z, z') \).

III. RESULTS

In the calculations presented below, we have considered a jellium slab of background thickness \( a = 2.23 \lambda_F \), \( \lambda_F \) being the Fermi wavelength \((\lambda_F = 2\pi/k_F)\), and background density \( \bar{n} = \left(4\pi \right)^{3/2}/2 \) with \( r_s = 2.07 \). This slab corresponds to about four atomic layers of Al(100).

For the LSDA-RPA calculations, we have obtained the RPA xc hole density \( n_{xc}^{\text{unif}}(u) \) of a uniform electron gas either from Eq. (4) or from a non-oscillatory parametrization. For the PBE-RPA and TPSS-RPA calculations, we have always used a non-oscillatory parametrization of the RPA xc hole density \( n_{xc}^{\text{unif}}(u) \).

For the evaluation of the fully nonlocal (exact-RPA) \( \gamma_{xc}(k) \) of Eq. (4), we follow the method described in Ref. [22]. We first assume that \( n(z) \) vanishes at a distance \( z_0 \) from either jellium edge and we expand the single-particle wave functions \( \phi_l(z) \) and the density-response function \( \chi^\lambda(k||\omega; z, z') \) in sine and double-cosine Fourier representations, respectively. We then perform the integrals over the coordinates \( z \) and \( z' \) analytically, and we find an explicit expression for \( \gamma_{xc}(k) \) [see Eqs. (A1)-(A5) of the Appendix] in terms of the single-particle energies \( \varepsilon_l \) and the Fourier coefficients \( b_{k||l} \) and \( c_{k||l} \) of the single-particle wave functions \( \phi_l(z) \) and the density-response function \( \chi^\lambda(k||\omega; z, z') \), respectively. We have taken all the single-particle wave functions \( \phi_l(z) \) and energies \( \varepsilon_l \) to be the LDA eigenfunctions and eigenvalues of the KS Hamiltonian of DFT, as obtained by using the Perdew-Wang parametrization of the Ceperley-Alder xc energy of the homogeneous electron gas. For the jellium slab with \( r_s = 2.07 \) and \( a = 2.23 \lambda_F \) considered here, the exact RPA xc surface energy is found to be \( \sigma_{xc} = 3091 \text{ erg/cm}^2 \), not far from the corresponding RPA xc surface energy of a semi-infinite jellium which is known to be \( \sigma_{xc} = 3064 \text{ erg/cm}^2 \).

In Figs. 1 and 2 we have plotted (solid lines) the exact-exchange contribution to \( \gamma_{xc}(k) \), i.e., \( \gamma_x(k) \), which we have obtained from Eqs. (A1)-(A5) with the quantities \( \chi^{0}_{\text{unif}}(k, \omega) \) and \( \chi^{0}_{\text{unif}}(k, \omega) \) replaced by their noninteracting counterparts \( \chi^{0}_{\text{unif}}(k, \omega) \) and \( \chi^{0}_{\text{unif}}(k, \omega) \), respectively. Also plotted in these figures are the LSDA, PBE, and TPSS calculations of \( \gamma_x(k) \) that we have obtained by replacing the xc hole densities \( \bar{n}_{xc}(z, u) \) and \( n_{xc}^{\text{unif}}(u) \) entering Eq. (4) by their corresponding exchange-only counterparts (dashed lines).

The LSDA \( \gamma_x(k) \) represented in Fig. 1 has been obtained by using both the actual exchange hole density \( n_{xc}^{\text{unif}}(u) \) of a uniform electron gas [dashed curve labeled \( \gamma_{x}^{\text{LSDA}} \)], which we have obtained from Eq. (4) with \( \chi^{0}_{\text{unif}}(k, \omega) \) replaced by \( \chi^{0}_{\text{unif}}(k, \omega) \), and the non-oscillatory exchange hole density \( n_{xc}^{\text{unif}}(u) \) reported in Ref. [22] [dashed curve labeled \( \gamma_{x}^{\text{LSDA}}(k) \) and \( \gamma_{x}^{\text{LSDA}}(k) \) yield, by construction of the non-oscillatory exchange hole density \( n_{xc}^{\text{unif}}(u) \), the same exchange surface energy \( \sigma_{x} \); they are also almost identical in a wide range of low wavevectors, but \( \gamma_x^{\text{LSDA}}(k) \) is considerably less accurate near \( k = 2k_F \) where the exact \( \gamma_x(k) \) has a kink. This kink is realistic for jellium-like systems, but not for atoms and molecules.

The PBE and TPSS \( \gamma_x(k) \) represented in Fig. 2 have both been obtained by using the non-oscillatory exchange hole density \( n_{xc}^{\text{unif}}(u) \) reported in Ref. [22] which yields a wrong behavior of \( \gamma_x(k) \) at large wavevectors. Nevertheless, both the actual exchange hole density \( n_{xc}^{\text{unif}}(u) \) of a uniform electron gas (not used in these calculations) and the corresponding non-oscillatory exchange hole density would yield the same exchange surface energy \( \sigma_x \) by construction, as occurs in the LSDA.

Figs. 1 and 2 show that while the LSDA \( \gamma_x(k) \) considerably overestimates the exact \( \gamma_x(k) \) at low wavevectors (see Fig. 1), leading to an exchange surface energy \( \sigma_x \) that is too large, the PBE and TPSS \( \gamma_x(k) \) are close to the ex-
act \( \gamma_x(k) \) (see Fig. 2). We note that the peaks of \( \gamma_{\text{PBE}}(k) \) and \( \gamma_{\text{TPSS}}(k) \) are close to the exact one, a fact which was used in the construction of the TPSS exchange hole \( \gamma_{\text{TPSS}} \) and that at larger wavevectors \( \gamma_{\text{PBE}}(k) \) and \( \gamma_{\text{TPSS}}(k) \) nearly coincide, as expected; at lower wavevectors, however, the TPSS meta-GGA differs from the PBE GGA, leading to a wavevector-dependent \( \gamma_x(k) \) that is closer to the exact behavior.

We have also carried out calculations of the exact \( \gamma_x(k) \) for increasing values of the background thickness \( a \), and we have found that (i) \( \gamma_x(k) \) is only sensitive to the size of the system at wavevectors below the minimum that is present in the solid lines of Figs. 1 and 2 and (ii) as \( k \to 0 \) the wavevector-dependent \( \gamma_x(k) \) approaches in the semi-infinite limit the profile-independent negative value \( \left( \gamma_x = -1.50 \times 10^4 / r_s^3 \text{erg/cm}^2 \right) \) reported in Refs. 22 and 23.

Figures 3 and 4 exhibit the results that we have obtained for the RPA \( \gamma_{\text{xc}}(k) \) from Eqs. (16), (17), (18), and (19) (solid lines) and within the LSDA-RPA, PBE-RPA, and TPSS-RPA (dashed lines). As in the case of the exchange-only contributions represented in Figs. 1 and 2 the LSDA \( \gamma_{\text{xc}}(k) \) represented in Fig. 3 has been obtained by using both the actual RPA xc hole density \( \tilde{n}_{\text{xc}}(u) \) [dashed line labeled \( \gamma_{\text{xc}}^{\text{LSDA-RPA}} \)], which we have obtained from Eq. (11), and a non-oscillatory xc hole density \( \tilde{n}_{\text{xc}}(u) \) [dashed line labeled \( \gamma_{\text{xc}}^{\text{LSDA-RPA}} \)] of the RPA xc hole density \( \tilde{n}_{\text{xc}}^{\text{LSDA-RPA}}(u) \) \( \left( < \gamma >_{\text{LSDA-RPA}} \right) \). The area under each curve represents the RPA xc surface energy: \( \sigma_{\text{xc}}^{\text{exact-RPA}} = 3034 \text{erg/cm}^2 \) and \( \sigma_{\text{xc}}^{\text{act-RPA}} = 3091 \text{erg/cm}^2 \).

FIG. 2: Wavevector analysis \( \gamma_x(k) \), versus \( k/2k_F \), of the exchange surface energy of a jellium slab of thickness \( a = 2.23 \lambda_F \) and \( r_s = 2.07 \). Solid and dashed lines represent exact and semi-local (PBE and TPSS) calculations, respectively. The semi-local PBE and TPSS calculations have been performed from the non-oscillatory parametrization of \( \tilde{n}_{\text{xc}}(u) \) reported in Ref. 22. The area under each curve represents the exchange surface energy: \( \sigma_{\text{xc}}^{\text{PBE}} = 2155 \text{erg/cm}^2 \), \( \sigma_{\text{xc}}^{\text{TPSS}} = 2247 \text{erg/cm}^2 \), and \( \sigma_{\text{xc}}^{\text{exact}} = 2348 \text{erg/cm}^2 \).
In order to investigate the impact of short-range corrections to the RPA $\gamma_{xc}(k)$, we have plotted in Fig. 7 the correlation contribution to $\gamma_{xc}(k)$, i.e., $\gamma_c(k)$, as obtained in the RPA (LSDA-RPA, TPSS-RPA, and exact-RPA) and also in standard versions of local and semi-local density-functionals (LSDA and TPSS) that use an accurate (beyond RPA) non-oscillatory parametrization of the correlation hole density $\tilde{n}^{\text{unif}}_c(u)$. The area under each curve represents the correlation surface energy: $\sigma_{\text{RPA}}^{\text{exact}} = 3091 \text{ erg/cm}^2$ and $3043 \text{ erg/cm}^2$, for $a = 2.23 \lambda_F$ and $a = 0.56 \lambda_F$, respectively.

Finally, in order to investigate the impact of short-range corrections to the RPA $\gamma_{xc}(k)$, we have plotted in Fig. 7 the correlation contribution to $\gamma_{xc}(k)$, i.e., $\gamma_c(k)$, as obtained in the RPA (LSDA-RPA, TPSS-RPA, and exact-RPA) and also in standard versions of local and semi-local density-functionals (LSDA and TPSS) that use an accurate (beyond RPA) non-oscillatory parametrization of the correlation hole density $\tilde{n}^{\text{unif}}_c(u)$ of a uniform electron gas. The area under each curve represents the correlation surface energy: $\sigma_{\text{RPA}}^{\text{exact}} = 336 \text{ erg/cm}^2$, $\sigma_{\text{TPSS-RPA}}^{\text{exact}} = 743 \text{ erg/cm}^2$, $\sigma_{\text{LSDA}}^{\text{exact}} = 290 \text{ erg/cm}^2$, and $\sigma_{\text{TPSS}}^{\text{exact}} = 756 \text{ erg/cm}^2$. The area under each curve represents the correlation surface energy: $\sigma_{\text{RPA}}^{\text{exact}} = 336 \text{ erg/cm}^2$, $\sigma_{\text{TPSS-RPA}}^{\text{exact}} = 743 \text{ erg/cm}^2$, $\sigma_{\text{LSDA}}^{\text{exact}} = 290 \text{ erg/cm}^2$, and $\sigma_{\text{TPSS}}^{\text{exact}} = 756 \text{ erg/cm}^2$.

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standard LSDA exhibit serious deficiencies, both TPSS-RPA and the more accurate standard TPSS coincide with the exact-RPA. At shorter wavelengths, the standard TPSS predicts a substantial correction to its TPSS-RPA and exact-RPA counterparts, which is first positive and then negative and leads, therefore, to a persistent cancellation of short-range correlation effects beyond the RPA similar to the cancellation that was reported in Ref. 11. The exact short-range correlation functional in the framework of time-dependent density-functional theory and a two-dimensional wavevector analysis of the correlation surface energy.

IV. CONCLUSIONS

We have reported the first 3D wavevector analysis of the jellium xc surface energy in the RPA, and we have used this fully nonlocal (essentially exact) RPA calculation to test RPA versions of nonempirical semi-local density-functional approximations for the xc energy. We have tested the first three-rungs of the Jacob’s ladder classification of nonempirical density functionals, LSDA, PBE GGA, and TPSS meta-GGA. We have found that while the LSDA displays cancelling errors in the small and intermediate wavevector regions and the PBE GGA improves the analysis for intermediates wavevectors while remaining too low for small wavevectors (implying two-low xc surface energies), the TPSS meta-GGA yields a realistic wavevector analysis even for small wavevectors or long-range effects. We have also demonstrated numerically the correctness of the LSDA at large wavevectors, where LSD-RPA, TPSS-RPA, and the exact-RPA coincide, as shown in Fig. 7 and the universal low-wavevector behavior derived by Langreth and Perdew which is nicely reproduced by the TPSS meta-GGA.

We have carried out fully nonlocal RPA calculations for increasing values of the background thickness, and we have found that the 3D wavevector analysis of the xc surface energy is remarkably insensitive to the slab thickness except at very long wavelengths ($k \to 0$) where decreasing the slab thickness reduces the universal slope that is dictated by the presence of bulk and surface collective oscillations.

Finally, we have found that the TPSS wavevector analysis of the correlation surface energy, as obtained from an accurate (beyond RPA) non-oscillatory parametrization of the xc hole density of a uniform electron gas, provides both the exact short-$k$ limit, where LDA fails badly, and the exact large-$k$ limit, where RPA is wrong. Hence, our calculations support the conclusion that the TPSS meta-GGA xc density functional accurately describes the jellium surface, including not only short-range but also long-range effects.

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APPENDIX A

Here we give an explicit expression for the wavevector-dependent contribution $\gamma_{xc}(k)$ to the xc surface energy $\sigma_{xc}$ of a jellium slab of background density $\bar{n}$ and thickness $a$, in terms of the single-particle energies $\varepsilon_l$ and the Fourier coefficients $b_{ls}$ and $\chi_{mn}^\lambda$ of the single-particle wave functions $\phi_l(z)$ and the density-response function $\chi_{\lambda}(k_z; z, z')$, respectively. From Eqs. 16, 17, 20, and 21 and performing the integrals over the coordinates $z$ and $z'$ analytically, we find:

$$
\gamma_{xc}(k) = \frac{k_F}{\pi} \int_0^k \frac{dk_z}{k} \sum_{m=0}^\infty \sum_{n=0}^\infty \alpha_{mn}(k_z) \beta_{mn}(k_{||})
$$

$$
= \bar{n} a \bar{n}_{xc}^{\text{unif}}(k),
$$

(A1)

where

$$
\alpha_{mn}(k_z) = 2k_F^2 \pi \left[ \frac{1}{k_z^2 - (m\pi/d)^2}(k_z^2 - (n\pi/d)^2) \right]
$$

(A2)

and

$$
\beta_{mn}(k_{||}) = \frac{1}{\pi} \int_0^1 d\lambda \int_0^\infty d\omega \chi_{mn}^\lambda (k_{||}, i\omega)
$$

$$
= -\frac{\mu_m \mu_n}{\pi d^2} \sum_{l=1}^{\lambda} \sum_{l'=1}^{\lambda} (E_F - \varepsilon_l) \int_{l'}^{\lambda} G_{ll'}^{mn} G_{ll'}^{mn},
$$

(A3)

with $d = a + 2z_0$,

$$
\mu_m = \begin{cases} 
1, & \text{for } m = 0, \\
2, & \text{for } m \geq 1
\end{cases}
$$

(A4)

and

$$
G_{ll'}^{mn} = \frac{1}{2} \sum_{s=1}^\infty \sum_{s'=1}^\infty b_{ls} b_{l's'} (\delta_{m,s-s'} + \delta_{m,s'-s} - \delta_{m,s+s'}).
$$

(A5)

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