Quasi-Low-Dimensional Electron Gas with One Populated Band as a Testing Ground for Time-Dependent Density-Functional Theory of Mesoscopic Systems

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We find an exact analytical solution to the exchange-only time-dependent density-functional theory (TDDFT) problem for a significant class of quasi-low-dimensional (QLD) materials: QLD electron gas with only one band filled in the direction perpendicular to the layer or wire. The theory yields the TD exchange potential as an explicit nonlocal operator of the TD spin density. The dressed interband (image states) excitation spectra of quasi-two-dimensional electron gas are obtained, while the comparison with the Kohn-Sham transitions provides insights into the qualitative and quantitative role of the many-body interactions. Important cancellations between the Hartree f_H and the exchange f_x kernels of TDDFT are found in the low-density regime, elucidating the interrelations between the Kohn-Sham and the many-body dynamics in mesoscopic systems.

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Density-functional theory (DFT) [1] and its time-dependent counterpart TDDFT [2] are presently, by far, the most popular methods to conceive the ground-state and excitation properties, respectively, of atomic, molecular, and condensed matter systems. Substituting the quantum many-body problem with the single-particle one, both DFT and TDDFT require the knowledge of the exchange-correlation (XC) potentials, $v_{\mathrm{XC}}(\mathbf{r})$ and $v_{\mathrm{XC}}(\mathbf{r},t)$, respectively, which are necessary to close the system of Kohn-Sham (KS) equations [1], which (in the TD case) read

$$i\frac{\partial\phi_i(\mathbf{r},t)}{\partial t}=\left[-\frac{1}{2}\Delta+v_{\mathrm{ext}}(\mathbf{r},t)+v_H(\mathbf{r},t)+v_{\mathrm{XC}}(\mathbf{r},t)\right]\phi_i(\mathbf{r},t). \tag{1}$$

Since the external potential $v_{\mathrm{ext}}(\mathbf{r},t)$ is set by the problem and the Hartree potential $v_H(\mathbf{r},t)=\int n(\mathbf{r}',t)/|\mathbf{r}-\mathbf{r}'|d\mathbf{r}'$ is defined by the electron density $n(\mathbf{r},t)$, Eqs. (1) with proper initial conditions can be solved, determining the orbitals $\phi_i(\mathbf{r},t)$, and from them constructing the density $n(\mathbf{r},t)=\sum_i|\phi_i(\mathbf{r},t)|^2$, the latter being the key quantity of interest in (TD)DFT. Unfortunately, although the exact XC potentials $v_{\mathrm{XC}}(\mathbf{r},t)$ exist in principle, they are never known for nontrivial systems, making us resort to approximations.

The XC potentials now overwhelmingly used in applications are local functions of the electron density or also of its spatial derivatives, the local-density approximation (LDA) [1] and the generalized-gradient approximation (GGA) [3], respectively. While simple and efficient in implementations, these approximations suffer from the well-known drawbacks. The one of our specific concern here will be the inherent dimensionality dependence of both LDA and GGA, i.e., their having distinct 3D, 2D, and 1D versions, which makes them poorly substantiated and unreliable in the case of systems of intermediate dimensionality, such as quasi-low-dimensional materials. A truly first-principles XC functional, being one and the same for all systems, must work equally well for different dimensionalities, including the intermediate ones. The exact exchange (EXX) [or optimized-effective potential [4,5] stands out in DFT as a first-principles potential not, in particular, bound to any specific dimensionality. This potential obeys a number of important requirements of the exact theory, such as the correct asymptotic behavior $-e^2/r$ for finite systems, the support of image states at surfaces and in low dimensions [6–8], it produces [9,10] the derivative discontinuity in the energy dependence on the fractional electrons number [11], and it is free from self-interaction. The time-dependent version of the EXX theory has been developed [12,13] and found to support the excitonic effect in semiconductors [14]. For all the advantages, an unfortunate drawback of the EXX theory is the extreme complexity of its implementation: It is the orbital-dependent formalism which involves the solution of the notoriously tedious optimized-effective potential integral equation [4,5]. This has prevented EXX from becoming widely used in applications, and even qualitative insights are often obscured by heavy numerical complications.

It, therefore, came recently as a surprise that, for the quasi-low-dimensional electron gas (QLDEG) with only one band populated in the transverse direction, the ground-state EXX problem has a simple explicit solution in terms of the spin density [8]. A natural question arises whether the same route can be taken to build the analytical EXX theory of many-body excitations in QLDEG. In this Letter, we give to this a positive answer by finding an explicit solution to the TD exchange potential in terms of the TD spin density for QLDEG with one band populated. For the solution to be expressible through the density, the applied perturbation must not change the symmetry of the QLDEG, as is discussed below.

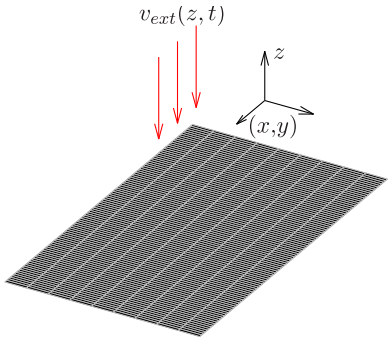

FIG. 1. Schematics of the quasi-two-dimensional electron gas (Q2DEG) under the action of a time-dependent external potential.

We start from the ground state of a Q2DEG (for 1D case, see below), uniform in the xy plane and confined in the z direction by a potential $v_{\rm ext}(z)$. The in-plane and the perpendicular variables separate in this case. We further assume that only the states $\mu_0^{\uparrow}(z)$ and $\mu_0^{\downarrow}(z)$, one for each spin orientation, are occupied in the z direction [8], leading to the orbitals of the form

$$\psi^{\sigma}_{\mathbf{k}_\parallel}(\mathbf{r}) = \frac{1}{\sqrt{\Omega}} e^{i\mathbf{k}_\parallel\cdot\mathbf{r}_\parallel}\mu_0^{\sigma}(z), \tag{2}$$

where Ω is the normalization area. To this system we apply a TD potential, which is assumed to depend on the z coordinate only (as schematized in Fig. 1) and, by this, it preserves the system's lateral uniformity during the time evolution. We will see that a wealth of many-body phenomena are preserved within these constraints, while the gain is the system admitting an analytical solution.

The main result of this Letter is that, with the above setup, the TDEXX potential is

$$v_x^{\sigma}(z,t) = -\frac{1}{n_{\rm 2D}^{\sigma}}\int \frac{F_2(k_F^{\sigma}|z-z'|)}{|z-z'|} n^{\sigma}(z',t)dz', \tag{3}$$

where $n^{\sigma}(z,t)$ is the spin density,

$$F_2(u) = 1 + \frac{L_1(2u) - I_1(2u)}{u},$$

L_1 and I_1 are the first-order modified Struve and Bessel functions [15,16], respectively, $n_{\rm 2D}^{\sigma} = \int_{-\infty}^{\infty} n^{\sigma}(z,t)dz$ is the 2D spin density, which does not change during the time evolution, and $k_F^{\sigma} = \sqrt{4\pi n_{\rm 2D}^{\sigma}}$ is the corresponding 2D Fermi radius. We derive Eq. (3) in Sec. I of the Supplemental Material [17], with the use of the adiabatic connection perturbation method [13,18]. In the linear-response regime, Eq. (3) gives immediately for the exchange kernel

$$f_x^{\sigma\sigma'}(z,z',\omega) = \frac{\delta v_x^{\sigma}(z,\omega)}{\delta n^{\sigma'}(z',\omega)} = -\frac{1}{n_{\rm 2D}^{\sigma}}\frac{F_2(k_F^{\sigma}|z-z'|)}{|z-z'|}\delta_{\sigma\sigma'}. \tag{4}$$

Notably, f_x of Eq. (4) is frequency-independent. Our point, however, is that Eqs. (3) and (4) are by no means an adiabatic approximation: A detailed derivation ([17], Sec. I) shows that they hold *exactly* within the fully dynamic TDEXX for QLDEG with one band filled, provided that the exciting field is applied perpendicularly to the layer.

We use the kernel of Eq. (4) with the basic linear-response TDDFT equality [19,20]

$$(\chi^{-1})^{\sigma\sigma'}(z,z',\omega) = (\chi_s^{-1})^{\sigma\sigma'}(z,z',\omega) - f_H(z,z') - f_x^{\sigma\sigma'}(z,z',\omega), \tag{5}$$

where χ and χ_s are the interacting-electrons and KS spin-density-response functions, respectively, the latter given in our case by

$$\chi_s^{\sigma\sigma'}(z,z',\omega) = n_{\rm 2D}^{\sigma}\mu_0^{\sigma}(z)\mu_0^{\sigma}(z')\sum_{n=1}^{\infty}\left(\frac{1}{\omega+\lambda_0^{\sigma}-\lambda_n^{\sigma}+i0_+} - \frac{1}{\omega-\lambda_0^{\sigma}+\lambda_n^{\sigma}+i0_+}\right)\mu_n^{\sigma}(z)\mu_n^{\sigma}(z')\delta_{\sigma\sigma'}, \tag{6}$$

where λ_n^{σ} and $\mu_n^{\sigma}(z)$ are the eigenenergies and the eigenfunctions of the perpendicular motion, respectively, and the summation over the states includes the integration over the continuous spectrum. A remarkable property of the KS response function of this system is that it is immediately invertible to ([17], Sec. II)

$$(\chi_s^{-1})^{\sigma\sigma'}(z,z',\omega) = \frac{\delta_{\sigma\sigma'}}{2n_{\rm 2D}^{\sigma}\mu_0^{\sigma}(z)\mu_0^{\sigma}(z')}[\omega^2 X_1^{\sigma}(z,z') - X_2^{\sigma}(z,z')], \tag{7}$$

with

$$X_1^{\sigma}(z,z') = \sum_{n=1}^{\infty}\frac{\mu_n^{\sigma}(z)\mu_n^{\sigma}(z')}{\lambda_n^{\sigma}-\lambda_0^{\sigma}} = (\hat{h}_s^{\sigma}-\lambda_0^{\sigma})^{-1}[\delta(z-z') - \mu_0^{\sigma}(z)\mu_0^{\sigma}(z')], \tag{8}$$

$$X_2^{\sigma}(z,z') = \sum_{n=1}^{\infty}(\lambda_n^{\sigma}-\lambda_0^{\sigma})\mu_n^{\sigma}(z)\mu_n^{\sigma}(z') = (\hat{h}_s^{\sigma}-\lambda_0^{\sigma})\delta(z-z'), \tag{9}$$

where $\hat{h}_s^{\sigma}$ is the static KS Hamiltonian [21]. The Hartree part of the kernel is

$$f_H^{\sigma\sigma'}(z,z') = -2\pi|z-z'|. \tag{10}$$

The many-body excitation energies ω are found from the equation

$$\sum_{\sigma'}\int (\chi^{-1})^{\sigma\sigma'}(z,z',\omega)\delta n^{\sigma'}(z',\omega)dz' = 0, \tag{11}$$

where $\delta n^{\sigma}(z,\omega)$ is the self-oscillation of the spin density.

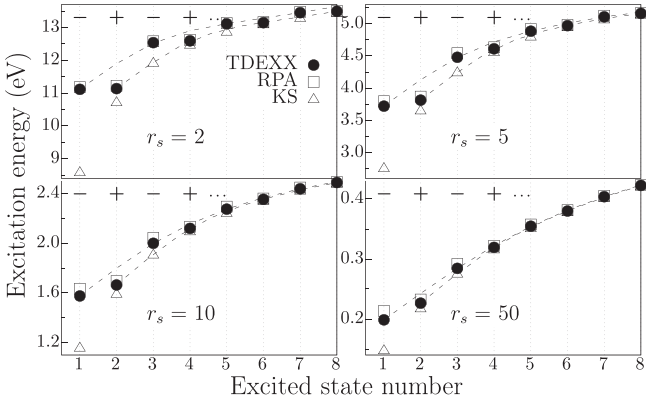

FIG. 2. Excitation energies of a spin-neutral Q2DEG with one transverse band filled. Circles, squares, and triangles are TDEXX, RPA, and KS excitation energies, respectively. Plus and minus signs mark even and odd excitations, respectively. Dashed lines connect eigenvalues of the even and odd self-oscillations, separately.

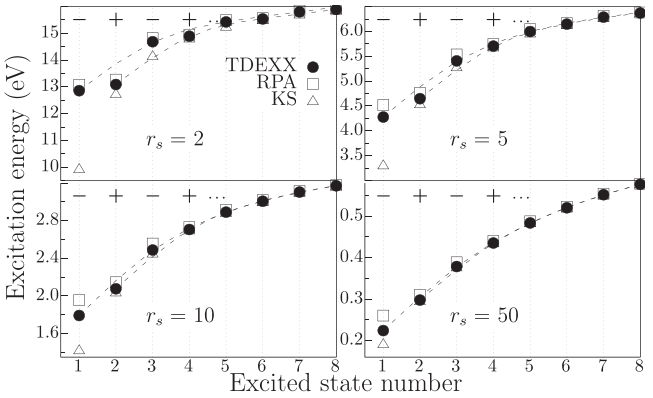

FIG. 3. The same as Fig. 2, but for the fully spin-polarized Q2DEG.

With the use of Eqs. (5) and (7)–(9), Eq. (11) can be rewritten as the following eigenvalue problem ([17], Sec. II)

$$(\hat{h}_s^\sigma - \lambda_0^\sigma)\Big[(\hat{h}_s^\sigma - \lambda_0^\sigma)y^\sigma(z) + 2n_{2D}^\sigma \times \int \mu_0^\sigma(z)\sum_{\sigma'} f_H^{\sigma\sigma'}(z,z')\mu_0^{\sigma'}(z')y^{\sigma'}(z')dz' + 2n_{2D}^\sigma \times \int \mu_0^\sigma(z) f_x^{\sigma\sigma}(z,z')\mu_0^\sigma(z')y^\sigma(z')dz'\Big] = \omega^2 y^\sigma(z), \quad (12)$$

where $y^\sigma(z) = \delta n^\sigma(z)/\mu_0^\sigma(z)$.

We have obtained the eigenvalues and eigenfunctions of Eq. (12) numerically on a z-axis grid for a number of the EG densities. The confining potential $v_{\rm ext}(z)$ was chosen to be that of the 2D positive charge background. The static KS problem was solved self-consistently with the use of the EXX potential, which is that of Eq. (3) with the ground-state density in lieu of the TD one [8]. Results for the eigenenergies of the excited states are presented in Figs. 2 and 3, for spin-neutral and fully spin-polarized Q2DEG, respectively, where TDEXX is compared to the random-phase approximation (RPA) [setting $f_x = 0$ in Eq. (12)] and with the KS transitions [setting $f_x = f_H = 0$ in Eq. (12)]. Obviously, the first excited state is affected strongly by the many-body interactions, resulting in the both TDEXX and RPA being very different from the single-particle KS transition. This effect, however, weakens for higher excited states. Second, the difference between the TDEXX and RPA increases with the growth of r_s (decrease of the density), the former moving closer to the KS values, which is more pronounced for the spin-polarized than for the spin-neutral EG. This has an elegant explanation: Expanding Eq. (4) in powers of k_F^σ, we can write at small k_F^σ

$$f_x^{\sigma\sigma'}(z,z',\omega) \approx \left(-\frac{32}{3k_F^\sigma} + 2\pi|z-z'|\right)\delta_{\sigma\sigma'}. \quad (13)$$

Noting that the first term in Eq. (13) is a constant and, consequently, it does not play a role in f_x, and comparing with Eq. (10), we conclude that, for a dilute EG, the exchange part of the kernel by a half and completely cancels the Hartree part, for the spin-neutral and fully spin-polarized EG, respectively. In the fully spin-polarized case, at low densities, this brings the many-body excitation energies back to the KS values, as can be observed in Fig. 3, right lower panel.

We note that the energies of the vertical interband excitations, i.e., those without the transfer of the in-plane wave vector, which we obtain, are purely real, which means that, within the present setup, the finite lifetime of the image states would require the inclusion of the correlations. In contrast to the KS transitions, TDEXX and RPA excitation energies split into the even and odd series, the values changing smoothly within each, while jumping across the series. In Figs. 2 and 3, the points within each series are connected with dashed lines serving as guides for the eye. In Fig. 4, we plot the self-oscillations $\delta n(z,\omega)$ themselves.

The quasi-1D electron gas admits the same treatment as the Q2DEG above, leading to the following results (cf. the static case [8]): The TD exchange potential is given by

$$v_x^\sigma(\boldsymbol{\zeta},t) = -\frac{1}{n_{1D}^\sigma}\int \frac{F_1(k_F^\sigma|\boldsymbol{\zeta}-\boldsymbol{\zeta}'|)}{|\boldsymbol{\zeta}-\boldsymbol{\zeta}'|} n^\sigma(\boldsymbol{\zeta}',t)d\boldsymbol{\zeta}', \quad (14)$$

and the corresponding exchange kernel is

$$f_x^{\sigma\sigma'}(\boldsymbol{\zeta},\boldsymbol{\zeta}',\omega) = -\frac{1}{n_{1D}^\sigma}\frac{F_1(k_F^\sigma|\boldsymbol{\zeta}-\boldsymbol{\zeta}'|)}{|\boldsymbol{\zeta}-\boldsymbol{\zeta}'|}\delta_{\sigma\sigma'}, \quad (15)$$

where $\boldsymbol{\zeta} = (x,y)$, the wire is stretched along the z axis,

$$F_1(u) = \frac{1}{2\pi}G_{2,4}^{2,2}\left[u^2\middle|\begin{matrix}\frac{1}{2},1\\ \frac{1}{2},\frac{1}{2},-\frac{1}{2},0\end{matrix}\right],$$

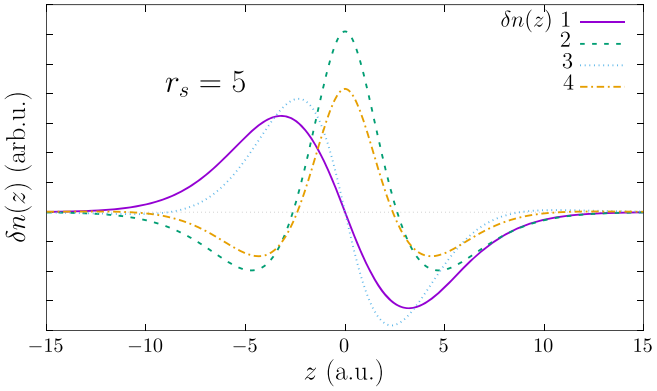

FIG. 4. Self-oscillations of the density of the spin-neutral EG of $r_s = 5$, corresponding to the transitions to the first four excited states.

and $G^{m,n}_{p,q}\left[u\middle|\begin{matrix}a_1, & \ldots, & a_p\\ b_1, & \ldots, & b_q\end{matrix}\right]$ is the Meijer G function [15,16]. The Hartree kernel in the Q1D case is

$$f_H^{\sigma\sigma'}(\boldsymbol{\zeta},\boldsymbol{\zeta}') = -2\log\left(k_F^{\sigma}|\boldsymbol{\zeta}-\boldsymbol{\zeta}'|\right). \tag{16}$$

As shown earlier [8], the assumption of the QLDEG having one spin-state occupied in the transverse direction is not very restrictive: This is a regime actually realizing at $r_s > 1.46$ and $r_s > 0.72$, for the Q2D and Q1D cases, respectively, provided the confining potential is that of the positive 2D (1D) uniform background. The second feature of our setup, that of the perturbation field being applied perpendicularly to the system's extent, is important: By this we do not study the excitation spectra of the 2D (1D) EG proper, which problem has been extensively addressed in the literature before ([20,22], and references therein) but we are concerned with the interband excitations, which are the excitations to the image states of QLDEG. The latter excitations we handle as properly dressed, i.e., accounting for the many-body dynamic interactions, doing this at the level of TDEXX. With the understanding of the above, our theory is exact.

The localized Hartree-Fock potential (LHF) [23] has recently attracted new attention as a single-particle potential providing the best possible fulfillment of the many-body TD Schrödinger equation by a Slater determinant wave function [24,25] and, in the spirit of the "direct-energy" potentials [26], yielding the total energy as a sum of KS eigenvalues. It has been recently shown [8] that for QLDEG with one populated band in its ground state, LHF potential coincides, up to a constant, with the EXX one. In Sec. III of the Supplemental Material [17], we prove that the same coincidence, up to a TD constant, holds between TDEXX and TDLHF potentials.

Technically trivial but conceptually instructive, the Q0D case can be treated similarly. The only systems with one, at most, filled orbital per each spin direction are then the singlet of two electrons and one-electron systems, for spin-neutral and spin-polarized cases, respectively. In both cases the TD exchange potential is well known to be minus half of and minus Hartree potential, respectively. We recall that in the Q2D and Q1D cases the latter property holds in the low-density limit only, while for two and one electron systems it just holds, the latter limit having, obviously, no meaning. Accordingly, the exchange kernels in these cases are minus half of and minus Hartree kernel, respectively.

In conclusion, we have identified the quasi-low-dimensional electron gas with one populated band as a unique system admitting an analytical solution of the many-body excitation problem by means of the time-dependent density-functional theory at the level of the exact exchange. The scent and feel of TDDFT was achieved by the straightforward analytical construction of the key quantities of the theory. We have applied our method to obtain the interband excitation spectra (excitations to image states) of Q2DEG. The low-lying excited states are shown to be strongly affected by the many-body interactions for the EG of higher densities. In the low-density regime, we have shown that the exchange kernel cancels the Hartree one by a half and entirely, in the case of the spin-neutral and fully spin-polarized EG, respectively. This demonstrates how qualitatively wrong and inconsistent the often used random-phase approximation (i.e., keeping the Hartree part of the kernel only) may be. For the dilute fully spin-polarized QLDEG this leads to the fundamental conclusion that the Kohn-Sham transitions can be, at the same time, the true excitation energies of a many-body system.

We, finally, argue that the concept of the quasi-low-dimensional electron gas with one populated band has the potential to continue to bring valuable results in mesoscopic physics on the one hand, and to enrich our understanding of DFT and TDDFT themselves, on the other. Among the applications envisaged, to name only the two, are the nonlinear dynamics with the use of Eq. (3), including pumping with laser fields, and the construction of advanced approximate XC functionals, such as, e.g., meta-GGA ones [27], which require tuning against exact solutions. Inclusion of electron correlations in the formalism is the next natural step. Preliminary analysis shows that this is feasible by means of the adiabatic connection perturbation theory to the second order in interaction. This work is under way.

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