Exchange-correlation energy of a multicomponent two-dimensional electron gas

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We discuss the exchange-correlation energy of a multicomponent (multi-valley) two-dimensional electron gas and show that an extension of the recent parametrisation[1] of the exchange-correlation energy by Attacalite et al. describes well also the multicomponent system. We suggest a simple mass dependence of the correlation energy and apply it to study the phase diagram of the multicomponent 2D electron (or hole) gas. The results show that even a small mass difference of the components (e.g. heavy and light holes) decreases the concentration of the lighter components already at relatively high densities.

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I. INTRODUCTION

One of the most interesting discoveries in semiconductor heterostructures was the formation of a two-dimensional (2D) electron gas[2]. For example, this has set the starting point for building nanostructures such as quantum dots or wires (for reviews see[3, 4, 5]). Many of the simplified models describing semiconductor nanostructures are based on the assumption of a homogeneous electron gas. The effects of the detailed band structure are included only by using an effective mass for the electrons and by screening the electron-electron interaction with a static dielectric constant. For the conduction electrons and by screening the electron-electron interaction are included only by using an effective mass for the electrons.

The valence band maximum in most semiconductors is degenerate, consisting of two bands corresponding to heavy and light holes. In 3D structures the hole gas will then consist of two kinds of holes. In 2D structures there will be an energy separation between the band minima due to the fact that heavy holes can be more easily localised than the light hole. Nevertheless, if the mass difference is not too large, both kinds of holes can coexist and the correlations between them have to be considered. The hole gas of most semiconductors is one example of a four-component Fermi gas. Electron layers in semiconductor heterostructures provide yet another system of multicomponent electron (or hole) gas, intensively studied theoretically[7, 8, 9, 10]. Recently, electron addition spectra of vertical quantum dots based on layered structures have also been measured[11].

In the following, we call such a normal 2D electron gas as a two-component gas, the components being the spin-up and spin-down electrons. Similarly, a polarised electron gas is called a one-component Fermi gas. (In the literature, the latter is sometimes referred to as “spinless fermions”).

Layered semiconductors, however, provide several structures where the simple picture of an ideal (one- or two-component) two-dimensional electron gas is likely to fail. In elemental semiconductors, silicon and germanium, the conduction band minimum consists of four equivalent minima at nonzero values of the k-vector and, moreover, these minima have non-isotropic effective masses. The resulting conduction electron gas has an “internal” degeneracy of eight (two from spin and four from the four minima), and is a possible example of an eight-component Fermi gas. The correlations in such an electron gas are much more complicated and have been studied in connection with the 3D electron-hole plasma in semiconductors[6].

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first discuss the general high and low density limits and the effect of mass difference between the components. We will then show that an analytic continuation of the parametrisation by Attaccalite et al. fits well the existing many-body calculations for multicomponent systems. As reference data we use the calculations of Conti and Senatore for a four-component 2D electron gas and the results of Apaja et al. for a 2D charged Bose gas which can be viewed as an upper bound for the energy of a Fermi gas with an infinite number of internal degrees of freedom.

Finally, we will study the phase diagram suggested for this energy functional and show that already a small mass difference will decrease the concentration of the lighter-mass components. When the density gets low enough, eventually all systems become one-component, as predicted by the Attaccalite et al. parametrisation for the normal 2D electron gas.

II. EXCHANGE-CORRELATION ENERGY: GENERAL DISCUSSION

We consider a homogenous gas consisting of Λ different kinds of fermions, or components, as we call them. For instance, the different spin states of the homogenous gas are treated as different components. For a normal electron gas Λ = 2 and for a fully polarised gas Λ = 1. We assume that all the components have the same charge, but they may have different masses. The total density n of the gas is

\[ n = \sum_{i=1}^{\Lambda} n_i = n \sum_{\nu=1}^{\Lambda} \nu, \]

where \( n_i \) is the number density and \( \nu_i = n_i/n \) is a dimensionless concentration of the component \( i \). Note that the concentrations are normalised as \( \sum_{i=1}^{\Lambda} \nu_i = 1 \). The density parameter \( r_s = 1/\sqrt{\pi n} \) is always related to the total density \( n \) of the gas. The kinetic energy per particle of the noninteracting gas becomes

\[ \epsilon_k(r_s, \{\nu_i\}, \{m_i\}) = \frac{\hbar^2}{2 r_s^2} \sum_{i=1}^{\Lambda} \frac{\nu_i^2}{m_i}, \]

where \( m_i \) is the mass of the component \( i \). The total exchange energy is the sum of exchange energies of the different components. The total exchange energy per particle is then

\[ \epsilon_x(r_s, \{\nu_i\}) = -\frac{e^2}{4\pi\varepsilon_0\epsilon} \frac{8}{3\pi r_s} \sum_{i=1}^{\Lambda} \nu_i^{3/2}. \]

Note that while the kinetic energy depends on the masses of the components, the exchange energy is independent of the masses. The correlation energy per particle is defined as the difference between the exact energy and the Hartree-Fock energy, \( \epsilon_c = \epsilon_{\text{tot}} - (\epsilon_k + \epsilon_x) \). In true semiconductor systems the different components can represent, for example, different band minima. The exchange energy has then also components coming from the exchange between electrons belonging to these different minima. Generally such exchange interaction is found to be small. In the spirit of the effective mass approximation we will completely neglect such effects here.

In order to study the multicomponent correlation energy we will first consider the case where all the components have the same mass. In this case the exchange correlation energy can be written as a function of the density parameter \( r_s \) and the concentrations \( \nu_i \). The symmetry requires that the energy functional is symmetric with respect of the interchange of the different concentrations \( \nu_i \), i.e., it does not depend on the indexing. We define numbers

\[ Z_{\gamma} = \sum_{i=1}^{\Lambda} \nu_i^\gamma, \]

which by construction have the desired symmetry. We notice that \( Z_1 = 1, \epsilon_x \propto Z_{3/2} \) and the kinetic energy \( \epsilon_k \propto Z_2 \) (for equal masses). In general any function \( g(\{\nu_i\}) \) with the desired symmetry property can be written as a function of the \( Z_{\gamma} \)-numbers, for example, as a series expansion

\[ g(\{\nu_i\}) = f(\{Z_{\gamma_i}\}) = a_1 Z_{\gamma_1} + a_2 Z_{\gamma_2} + a_3 Z_{\gamma_3} + \cdots , \]

where \( a_i \) are constants and \( \gamma_i \) can be noninteger. One way to approach the correlation energy is to use the suggestion of Wigner who interpolated the exchange-correlation energy of an electron gas between the two limiting cases, the pure exchange in the high density limit and the energy of the Wigner crystal in the low density limit:

\[ \epsilon_{xc}(r_s, \{\nu_i\}, \{m_i\}) = \epsilon_{WC}(r_s) + g(r_s, \{\nu_i\}, \{m_i\})[\epsilon_x(r_s, \{\nu_i\}) - \epsilon_{WC}(r_s)] \]

where \( \epsilon_{WC} \) is the classical Madelung energy of the Wigner crystal, which does not depend on the masses. The advantage of this approach is that the mass dependence appears only in the interpolation function \( g \). Furthermore, if the masses are equal, the interpolation function \( g \) can immediately be written as in Eq. (4).

In order to estimate the interpolating function \( g \) one would need results of accurate many-body calculations for systems with different numbers of components, and for several values of \( r_s \) and the concentrations \( \nu_i \). We are not in the position to perform such computations at this point but use existing data for special cases. For the two-component system Attaccalite et al. have performed Monte Carlo calculations. They fitted an accurate interpolation formula to the results which is valid for any \( r_s \) and polarisation.

For the four-component system, Conti and Senatore have presented Monte Carlo results.
for several values of \( r_s \) in the case where all the concentrations \( \nu_i \) are equal. For systems with more components there exist no accurate data. However, the limit of infinite components can be approximated by the results obtained for charged bosons. If all the concentrations are the same (\( \nu_i = 1/\Lambda \)) in the limit of infinitely many components, all the particles are at different internal state and the Pauli exclusion principle can not prevent putting all the particles in the same orbital state. The symmetric boson state is then a legitimate state for such a fermion system and provides an upper bound for the energy. However, the existing results beyond the one and two-component systems are still quite limited for an accurate interpolation of the function \( g \).

A. Mass dependence

As discussed above, the exchange energy is mass-independent and even in the multi-component electron gas only the kinetic and the correlation energies will depend on the masses. If all the components have the same mass \( M \), the kinetic energy of Eq. \( 2 \) can be written as \( \epsilon_k = \hbar^2 Z_2/(M r_s^2) \). In the general case, the kinetic energy can still be written in the same form by defining an average mass \( M \) as

\[
\frac{1}{M} = \frac{1}{Z_2} \sum_i^\Lambda \frac{\nu_i^2}{m_i}.
\]

We notice a scaling: By deviding the the kinetic and the exchange energies by \( M \), the resulting ratios will only depend on the product \( M r_s \) (\( \epsilon_k/M = \hbar^2 Z_2/(M r_s^2) \) and \( \epsilon_x/M = -8\epsilon Z_3/2/(4\pi\nu_0 3\pi M r_s) \)). Moreover, considering the atomic units in the case that all masses are the same, we notice that (in this case) also the total energy, and thus the correlation energy has the same scaling. It is then a reasonable first approximation to take the mass as an average mass and use it as a scaling factor

\[
\epsilon_x(r_s, \{\nu_i\}, \{m_i\}) = \frac{M}{m_e} \epsilon_x(M r_s, \{\nu_i\}, \{m_i = m_e\}),
\]

where \( m_e \) is the bare mass of the electron (or any suitably chosen effective mass). (Note that within this approximation, the interpolation function \( g \) in Eq. \( 6 \) would be written as \( g(M r_s, \{Z_i\}) \).

III. EXTENSION OF THE TWO-COMPONENT FUNCTION

For estimating an interpolation formula for the multi-component correlation energy, we start by considering the parametrised form of the two-component 2D electron gas by Attaccalite \textit{et al.} \cite{1}, in order to see if their function can be written in terms of the \( r_s \) and the \( Z_i \)-numbers, as discussed above. Attaccalite \textit{et al.} use the common description where the energy is written as a function of \( r_s \) and the polarisation \( \zeta = (\nu_1 - \nu_2)/n = \nu_1 - \nu_2 \) of the electron gas. They chose a correlation energy function \( \epsilon_c(r_s, \zeta) = \epsilon^{(6)}_c(r_s, \zeta) - \epsilon_x(r_s, \zeta) \) which diminishes the contributions of \( \epsilon_x \) beyond fourth order in \( \zeta \) as \( r_s \) increases. The parametrisation satisfies known high- and low-density limits: For high densities i.e. small \( r_s \) the exchange-correlation energy obeys the perturbation theory result \( \epsilon_{xc} = \epsilon_c + \alpha_0(\zeta) + b_0(\zeta) r_s \ln r_s \). For low densities the Wigner crystal limit is recovered, \( \epsilon_{xc} \sim -c_1/r_s + c_2/r_s^{1/2} \), where the constants \( c_1 \) and \( c_2 \) are independent of the number of components.

Using the concentrations \( \nu_i \), the square of the polarisation \( \zeta \) of the two-component gas becomes \( \zeta^2 = 2(\nu_1^2 + \nu_2^2) - 1 = 2Z_2 - 1 \). Now the exchange-correlation energy introduced by Attacalite \textit{et al.} \cite{1} can be written as (using their notations except for the above replacement for \( \zeta \))

\[
\epsilon^{(4)}_{xc}(r_s, \{\nu_i\}) = e^{-\beta r_s} [\epsilon^{(6)}_c - \epsilon^{(4)}_x]
\]

\[
+ \epsilon^{(6)} + \alpha_0(r_s) + \alpha_1(r_s)(2Z_2 - 1) + \alpha_2(r_s)(2Z_2 - 1)^2,
\]

where \( \epsilon^{(6)}_x = (1 + \frac{3}{2}(2Z_2 - 1) + \frac{3}{2(2Z_2 - 1)^2}) \epsilon_{xc}(r_s, \zeta = 0) \). The functions \( \alpha_i(r_s) \) are parametrised by Attacalite \textit{et al.} \cite{1} and are independent of the concentrations. This two-component exchange-correlation function of Attacalite \textit{et al.} is thus already of the desired form: It depends only on \( r_s, Z_2 \) and \( Z_{3/2} \) (through \( \epsilon_x \)) and can be directly extended to any number of components.

For a given value of \( r_s \) Attacalite \textit{et al.} \cite{1} fitted their function to several values of polarisations, \( \zeta \in [0, 1] \), which correspond to \( Z_2 \in [0.5, 1] \) and \( Z_{3/2} \in \sqrt{2}/1 \). An analytic continuation to larger number of components means an extrapolation of the \( \epsilon_{xc} \) to the region \( Z_2 \in [0, 1] \) and \( Z_{3/2} \in [0, 1] \) for a system with infinitely many components, but only to \( Z_2 \in [0.25, 1] \) and \( Z_{3/2} \in [0.5, 1] \) for a four-component system. Moreover, since \( Z_{3/2} \) only appears in the exchange energy, Eq. \( 3 \), which is exact for any number of components, errors in the extrapolation only result from the extension of the \( Z_2 \) space.

The multi-component gas must obey the low density Wigner crystal limit and the high density exchange limit; both of these requirements are satisfied by Eq. \( 4 \). By taking a “paramagnetic” gas, i.e. by setting \( \nu_i = \Lambda^{-1} \) for all the components and fixing \( r_s \), it can be seen that the exchange-correlation energy above is also a monotonous function of the number of components.

Figure 1 shows the exchange-correlation energy derived from Eq. \( 4 \) for selected values of \( r_s \) as a function of \( Z_2 \). For the interval \( Z_2 \in [0.5, 1] \) the lines correspond to the normal two-component electron gas with partial polarisation. For the interval \( Z_2 \in [0, 0.5] \) the lines are derived for the case where all the concentrations are equal, \( \nu_i = 1/\Lambda \) and consequently \( Z_2 = 1/\Lambda \). The points are results of different many-body calculations. Note that the lines from \( Z_2 = 0.5 \) to \( Z_2 = 1 \) agree exactly to the results of Attacalite \textit{et al.} \cite{1}. We can see that the extension to a
four component system ($Z_2 = .25$) is fairly accurate and the extension even to the boson case ($Z_2 = 0$) is still reasonable. In fact, the agreement of the Attaccalite et al. interpolation formula of Attaccalite et al. [25], extended to multicomponent systems. The points correspond to numerical results of many-body calculations as follows: Black dots for $Z_2 = 1$ and $Z_2 = 0.5$ are from Ref. [1]; black dots for $Z_2 = 0.25$ from Ref. [14]; black dots for $Z_2 = 0$ from Ref. [14]; open circles from Ref. [14]; black squares from Ref. [17]. Results for $r_s = 2, 10$, and 20 are shown.

IV. PHASE DIAGRAM OF THE MULTICOMPONENT 2D ELECTRON GAS

It was recently suggested by Attaccalite et al. [1] that the 2D electron gas shows a transition to a polarised gas at $r_s \approx 25[1]$. We will first study the stability of the multicomponent gas assuming all the masses to be equal. The extended exchange-correlation energy functional suggests that all multicomponent cases have a transition to a one-component phase when $r_s$ increases. Table I gives the estimated transition points. For any number of components, the transition happens nearly at the same point and directly from the multicomponent to a one-component phase. It is caused by the slightly different $r_s$-dependence of the one-component total energy as compared to the others. For $\Lambda \geq 2$ the total energy is a monotonously decreasing function of $\Lambda$ (for any $r_s$).

Next we will study the effect of the mass difference, using the approximation of Eq. [3]. We assume a four-component system and fix the masses pairwise equal so that two components are heavier than the two others, say $m_1 = m_2 = m_3$ and $m_3 = m_4 = m_5$. Note that due to the spin degeneracy of electrons (or holes) there will always be two components with the same mass, and the number of components will be even, except of the special case one. The phase diagram of the four-component electron gas obtained using Eq. (2) is shown in figure 2. We show the maximum of the concentrations ($\nu_h$) as a function of $r_s$ and the mass ratio $m = m_h/m_l$. When the masses are equal, also the concentrations remain equal ($\nu_i = 0.25$) until $r_s \approx 26.2$ at which point the system becomes a one-component gas. When the mass ratio increases, the concentrations $\nu_3$ and $\nu_4$ increase due to the reduction of the kinetic energy, and eventually $\nu_3 = \nu_4 = 0.5$ and the system has become two-component. A further increase of the mass ratio eventually changes the system to a one-component gas. We notice that as the mass is increased the polarisation occurs at smaller $r_s$. The reason is the scaling of the total energy and the dimensions with the mass.

In real semiconductor heterostructures the mass differ-
ence can arise from different constituents of the two layers in a double layer system or, in the case of holes, simply from the mass difference of the heavy and light hole. In both cases, however, there can be a constant potential energy difference from one component to another. This energy difference arises from the localisation of the particle in the direction perpendicular to the 2D-layer. For example, if we consider holes in a 1D harmonic potential, the ground state energy (the lowest perpendicular mode) has the energy $\epsilon_0(m) = \hbar \omega_0/(2 \sqrt{m/m_e})$, where $\omega_0$ is the confinement strength for $m_e$. This energy is smaller for the heavy hole and, consequently, further favours the transition to a two-component system where only heavy holes exist. In order to have a 2D electron gas the Fermi energy has to be clearly below the first excited perpendicular state, $\epsilon_1(m) = 3\hbar \omega_0/(2 \sqrt{m/m_e})$ in a harmonic well. In order for the heavy (mass $m_h$) and light holes (mass $m_l$) to coexist, the ratio $(\epsilon_0(m_l) - \epsilon_0(m_h))/\epsilon_1(m_l) - \epsilon_0(m_h)$ has to be clearly smaller than one. For example in the cases of Si, Ge and GaAs ($m_h/m_l > 5$) this ratio is larger than 0.6. Consequently, a 2D layer with both heavy and light holes is not possible. However, there are semiconductors, like AlAs, where $m_h/m_l \approx 2$ and the above energy ratio is only about 0.2, making the four-component gas possible.

V. CONCLUSIONS

We have studied the possibility to formulate a general multicomponent 2D exchange-correlation energy to be used in density functional calculations. After general considerations of the concentration and mass dependence of the correlation energy, we have demonstrated that the recent parametrisation by Attacalite et al.\cite{1} of the two-component exchange-correlation energy has functional properties which allow a direct extension to multicomponent systems. Furthermore, although the function is fitted only to the two-component electron gas, it gives fairly accurate results for four-component and infinite-component systems.

We suggest that the mass dependence of the correlation energy can be estimated with a properly chosen average mass. The application for multicomponent 2D gas shows an interesting phase diagram. For equal masses all systems transform from a multicomponent directly to a one-component gas when $r_s$ increases to about 25 · · · 27 depending on the number of components. Even a small mass difference decreases the concentration of the lighter component and it transforms first to a purely two-component and then to a one-component system when $r_s$ increases.

A more accurate interpolation formula requires extensive many-body calculations for multi-component gases. We hope that these considerations encourage such work.

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