Electronic compressibility of a graphene bilayer

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We calculate the electronic compressibility arising from electron-electron interactions for a graphene bilayer within the Hartree-Fock approximation. We show that, due to the chiral nature of the particles in this system, the compressibility is rather different from those of either the two-dimensional electron gas or ordinary semiconductors. We find that an inherent competition between the contributions coming from intra-band exchange interactions (dominant at low densities) and inter-band interactions (dominant at moderate densities) leads to a non-monotonic behavior of the compressibility as a function of carrier density.

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The recently developed experimental capability of isolating and manipulating an arbitrary number of graphene layers [1] has attracted considerable attention both for its impact on basic science [2] and for the tantalizing potential technological applications. The graphene bilayer is particularly interesting because of the possibility of opening - and controlling - a gap in the electronic spectrum by applying an external electric field [3, 4, 5, 6]. This is not possible for the single layer graphene. The bilayer, therefore, while inheriting many of the peculiar electronic characteristics of the monolayer due to its chiral Dirac fermion (though massive) spectrum, has the added virtue of being capable of acting as an electronic switch. It is thus essential to obtain a comprehensive characterization of this material. While some transport experiments are available [7], thermodynamic measurements are largely lacking. Among the thermodynamic quantities to be measured, the electronic compressibility \( \kappa \) stands out as an excellent tool to provide insight into the many-body interactions present in this material. \( \kappa \) can be obtained from the ground state energy as:

\[
\kappa^{-1} = n_e^2 \frac{\partial^2 E}{\partial n_e^2},
\]

where \( E \) is the ground state energy per unit area, and \( n_e \) is the electronic density. The electronic compressibility of a single layer graphene has been recently measured [8], and its behavior, besides being remarkably different from that of the usual two-dimensional gas (2DEG), seems to indicate that contributions from Coulomb interactions are either very weak or cancel out. Hartree-Fock [9] and Random Phase Approximation (RPA) [10] calculations predict a correction between 10% and 20% to the free theory for experimentally realized dopings. This correction increases logarithmically as the doping is lowered. It is natural then to ask what role interactions play in the bilayer. In many aspects, the bilayer graphene closely resembles the 2DEG, as described below. Hence, the bilayer system provides an opportunity to isolate the effects arising from its single layer constituents, from those occurring in an ordinary 2DEG. In particular, the issue of the chirality, which is so important for weak-localization physics [11], is the main difference between these two systems, and as we will show, plays an important role in the many-body physics of the bilayer. For small doping, the bilayer can be mapped approximately to a chiral two-dimensional massive fermionic system with parabolic bands [12, 13], in contrast with the massless, cone-like dispersion found in the monolayer. This limit is useful to compare the behavior of the bilayer (with its chirality) to that of the ordinary 2DEG, where experiments have shown that interactions play a dominant role, making the proper compressibility negative for small electron densities [14] as opposed to a positive constant given by the non-interacting model. This behavior is already present at the Hartree-Fock level. Due to the aforementioned mapping, \textit{a priori} it is reasonable to expect that the effect of electron-electron interactions would be observable in the graphene bilayer.

In this paper we calculate within the Hartree-Fock approximation the dependence of the inverse compressibility on the Fermi vector \( k_F \) using both a full, four-band (4B) model and the two-band (2B) approximation, which is valid for very small doping. We show that the most important qualitative signatures of the compressibility are already present at the 2B model level but that the 4B calculation, while more cumbersome, reveals finer features.

Throughout this paper we will refer (loosely) to the quantity \( \tilde{\kappa}^{-1} = \partial \mu / \partial n_e \) as the “inverse compressibility”. Here \( \mu \) stands for the chemical potential of the system. \( \tilde{\kappa} \) differs by a factor of \( n_e^2 \) from \( \kappa \) in (1). This is appropriate since \( \tilde{\kappa}^{-1} \) is usually the actual experimentally measured quantity. The density of electrons is given by \( n_e = g_s g_v k_F^2 / (4\pi) \), with \( g_s = 2 \), \( g_v = 2 \) being the spin and valley degeneracy, respectively. In the following we will consider the case of small doping but outside the range of ferromagnetic instability that is found at extremely low doping [13]. At the Hartree-Fock level, the ground state energy is given by \( E = K + E_{ex} \), where \( K \) stands for the kinetic energy and \( E_{ex} \) is the exchange energy per unit of area.

A graphene bilayer consists of two planes of graphene stacked as shown in Fig. (1). The kinetic term of the Hamiltonian can be written, in the nearest neighbor tight binding approximation [15] by...
expanding around the K, K' points of the Brillouin zone, as \( H_{\text{kin}} = \sum \psi_Q^\dagger \mathcal{K}(p) \psi_Q \) with \( \psi_Q = \{ (p, A_1, \sigma, a) \, | \, (p, B_1, \sigma, a) \} \) where \( \sigma \) labels the valley, \( \alpha \) the sublattice in the plane \( i = 1, 2 \). \( \sum_Q \) represents the sum over all the indices. The kinetic energy matrix is given by (we use units such that \( \hbar = 1 \):

\[
\mathcal{K}(p) = \begin{pmatrix}
0 & pe^{i\phi(p)} & -t_\perp & 0 \\
pe^{-i\phi(p)} & 0 & 0 & 0 \\
-t_\perp & 0 & 0 & pe^{-i\phi(p)} \\
0 & 0 & pe^{i\phi(p)} & 0
\end{pmatrix},
\]

where \( \tan \phi(p) = p_y/p_x \), \( t_\perp = 0.35 \text{ eV} \) is the inter-layer hopping energy and we have set \( v_F = 3a\sqrt{2} = 6.6 \text{ eV \AA} \) to unity (\( t \) is the in-layer hopping energy and \( a \) the in-plane carbon-carbon distance). The interaction is given by the 2D Fourier transform of the 3D Coulomb potential, which is \( V_{\text{op}}(k) = \frac{2\pi^2}{\epsilon_0} \frac{1}{k} \) for the interaction among electrons within the same plane and \( V_{\text{op}}(k) = \frac{2\pi^2}{\epsilon_0} \frac{e^{-kd}}{k} \) otherwise, being \( d \approx 3.35 \text{ \AA} \) the inter-plane distance.

The kinetic energy matrix \( \mathcal{K}(p) \) can be diagonalized by a unitary transformation \( S^\dagger(p) \). The resulting dispersion bands (see Fig. 1) are: \( E_1(p) = -t + E(p) \), \( E_2(p) = \tilde{t} - E(p) \), \( E_3(p) = \tilde{t} + E(p) \) and \( E_4(p) = -\tilde{t} - E(p) \); being \( E(p) = \sqrt{t^2 + p^2} \) and \( \tilde{t} = t_\perp/2 \). It is convenient to work with the symmetric and anti-symmetric combinations of the layer densities, \( \rho_\pm = \rho_1 \pm \rho_2 \), which can be expressed in the diagonal basis as \( \rho_\alpha(q) = \sum p \Phi_\alpha(p + q) \chi^\alpha(p) \Phi(p) \) with \( \Phi(p) = S^\dagger(p)\psi(p) \) and \( \alpha = \pm \). The \( 4 \times 4 \) matrices \( \chi^\alpha \) contain the information of the overlap due to the change of basis. Then the interaction Hamiltonian takes the form \( H_I = 1/(2A) \sum_{p \neq 0} \sum_\alpha \rho_\alpha(q) V_{\text{op}}(q) \rho_\alpha(q) \), and the exchange energy per unit area \( A \) can be written in the continuum as:

\[
E_{\text{ex}} = -g_s g_v \frac{1}{2} \int d^2p \int d^2q \frac{1}{(2\pi)^2} \frac{1}{(2\pi)^2} \sum_{\alpha, i, j} \chi^{\alpha}_{ij}(q, p) \chi^{\alpha}_{ji}(p, q)
\]

\[
n_i(q)n_j(q)V_{\text{op}}(q - p),
\]

where \( i, j = 1, \ldots 4; n_i(q) = \langle \Phi^\dagger_i(q) \Phi_i(q) \rangle > 0 \) and \( V_{\pm}(k) = \frac{1}{2}(V_{\text{op}}(k) \pm V_{\text{op}}(k)) \). The occupation factors are given by \( n_1(q) = \Theta(k_F - q) \) \( n_2(q) = 1 - \Theta(k_F - q) \), \( n_3(q) = 0 \) and \( n_4(q) = 1 \) in the case of electron [hole] doping. This model however requires a cutoff \( \Lambda \) of the order of the inverse of the lattice parameter.

Being simpler to work with, and widely used as a starting point for calculations in the graphene bilayer, we start our analysis with the approximate 2B model that can be constructed at low energies by performing degenerate perturbation theory \( 12 \). This results in an effective kinetic Hamiltonian:

\[
H_{\text{kin}} = \sum \tilde{\psi}_Q^\dagger \mathcal{K}(p) \tilde{\psi}_Q = \begin{pmatrix}
0 & e^{-2i\phi(p)} \\
e^{2i\phi(p)} & 0
\end{pmatrix} \tilde{\psi}_Q,
\]

with \( \tilde{\psi}_Q = (e^{i\phi(p)} (p, B_1, \sigma, a)^\dagger (p, A_2, \sigma, a) \). The result of the approximation is an effective model with opposite parabolic dispersion bands of energy \( E_a = p^2/(2\tilde{t}) \), \( E_b = -p^2/(2\tilde{t}) \) as shown in Fig. 1. The effective kinetic energy per unit area then is given by \( K = (k_F^2 - \Lambda^2)/(4\pi\tilde{t}) \) giving a kinetic contribution to the inverse compressibility of \( k_F^{-1} = \pi/(4\tilde{t}) \).

In this reduced Hilbert space Eq. (3) is still valid, but this time the \( \chi^\alpha(p, q) \) are \( 2 \times 2 \) matrices. Combining all the contributions and re-inserting the units, we find the total inverse compressibility \( k^{-1} \) in the 2B model to be given by the expression:

\[
k^{-1} = \frac{v_F}{\Lambda} \left[ \frac{\pi}{2\tilde{t}} - \frac{g}{4k_F} \int_0^\pi d\theta \int_0^\pi dp \partial \partial F \right.
\]

\[
\left. \left[ \frac{k_F}{r(p, 1, \theta)} \left( 1 + \cos 2\theta e^{-dr(p, k_F, \theta)} \right) \right. \right]
\]

Here we have defined \( r(p, q, \theta) = \sqrt{p^2 + q^2 - 2pq \cos \theta} \), \( k_F \) and \( 1/d \) are in units of \( \Lambda \), and \( g = e^2/(\epsilon_0 \alpha) \) is the graphene coupling strength. The \( \pm \) indicates the expression for electrons (+) or holes (-). The differing term however is roughly a constant and can be neglected for small doping. Therefore, in what follows we will use the results for electron doping.

Fig. 2 shows a plot of \( k_F^{-1} \) as a function of \( k_F \) for \( \tilde{t}/\Lambda = 0.026 \), and \( d\Lambda = 3.7 \) \( (\lambda = 1.06 \approx 1 \text{ \AA}^{-1}) \). As can be seen, for very small doping the compressibility changes sign, becoming negative and divergent. This behavior, as
being
\[ D_{\alpha}^{++} = \int_{0}^{2\pi} d\theta \int_{0}^{k_p} p dp \chi^{\alpha}_{11}(k_F, p) |\chi^{\alpha}_{22}(k_F, p)|^2, \]
\[ D_{\alpha}^{+-} = \int_{0}^{2\pi} d\theta \int_{0}^{\Lambda} p dp \chi^{\alpha}_{12}(k_F, p) |\chi^{\alpha}_{24}(k_F, p)|^2, \]
where \( \theta \) is the angle between \( k_F, p \). As the notation suggests, \( D_{\alpha}^{++} \) corresponds to exchange within the positive conduction band 1 while \( D_{\alpha}^{+-} \) measures the exchange between the negative filled sea and the conduction band. The calculation for hole doping is completely analogous, with the overlap elements to be considered for that case being \( |\chi^{\alpha}_{22}(k_F, p)|^2 \) and \( |\chi^{\alpha}_{24}(k_F, p)|^2 \). Since the compressibility involves only occupied states, its behavior is not symmetric with respect to particle-hole exchange. Nonetheless, the explicit calculation shows that the difference is negligible for small doping, being the same as in the 2B case. On the other hand, the kinetic contribution to the inverse compressibility is independent of the type of carrier and it is easily calculated to be \( \tilde{\kappa}^{-1} = \pi/[2E(p)] \). The final results for the four band calculation, by summing all the contributions, is shown in Fig. (3).

We see that the full model confirms the major qualitative features found within the 2B approximation. The compressibility is negative for small electronic density, diverging in the limit of \( k_F \to 0 \), and the inter-band exchange contributes to the incompressibility of the system (see Fig. (4)). The difference observed at larger values of the Fermi momentum is simply due to the difference in the kinetic term, since it is a constant in the two band

\[ 1/\kappa [-eV^2] \]

\[ k_F/\Lambda \]

\[ 0.02 \quad 0.04 \quad 0.06 \quad 0.08 \]

FIG. 2: (Color online) Inverse compressibility vs. Fermi wave vector in the 2-band approximation. Total: solid line (red), intra-band plus kinetic contribution: dashed (blue), kinetic: dash/dot (green), 2DEG: dotted (black). The inset depicts the contribution of intra-band exchange (solid line) and inter-band exchange (dashed).
FIG. 3: (Color online) Inverse compressibility as a function of the Fermi wave vector as calculated from the 4B model. Negative values of the Fermi vector indicate the result for hole doping.

FIG. 4: (Color online) Comparison between the exchange contribution for the 2B and 4B models. Total exchange (red): 4B solid line, 2B dotted; intra-band only (blue): 4B dashed, 2B dash/dot.

The 4B model seems to predict a behavior that is a hybrid between the one of a 2DEG, where the total contribution to the compressibility from exchange is negative, and the graphene monolayer, where it is positive.

In conclusion, we have studied the electronic compressibility of a graphene bilayer within the Hartree-Fock approximation and have found a behavior that is remarkably different from the two-dimensional electron gas due to the presence of inter-band transitions, and also from graphene monolayer. We have shown that the inverse compressibility is not a monotonic function of the electronic density and that the effective 2B model gives a good description of the problem only at very low densities. At intermediate densities, the four bands are important to explain the behavior of the compressibility. The non-monotonic behavior of the compressibility obtained with the 4B model is highly unusual. Generally, nonmonotonicity is associated with some external factor, such as confinement or applied magnetic fields. However here it is due solely to intrinsic electronic interactions. The implications of this remain to be understood. On the other hand, the negativity of the compressibility is understood once it is realized that the one involved is not the total compressibility but only that of the electronic gas. The total compressibility will comprise also the positive ionic background, which stabilizes the system. Nonetheless, the negative divergence of the inverse compressibility, present in both models for low enough electronic densities, could signal the eventual onset of Wigner crystallization [17]. These results, as in the case of the single layer graphene [8], can be studied via single electron transistor (SET) measurements. Our results indicate that the compressibility turns negative at density values of approximately \( n_e \approx 10^{11}/\text{cm}^2 \), which borders the current available precision [8]. However, being a Hartree-Fock calculation, this can act just as a very rough estimate. We have also neglected the trigonal warping term, which might be of importance at very low densities [12].

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