Coulomb interactions of massless Dirac fermions in graphene; pair-distribution functions and exchange-driven spin-polarized phases.

M.W.C. Dharma-wardana
National Research Council of Canada,
Ottawa, Canada. K1A 0R6

(Dated: March 23, 2022)

The quasi-2D electrons in graphene behave as massless fermions obeying a Dirac-Weyl equation in the low-energy regime near the two Fermi points. The stability of spin-polarized phases (SPP) in graphene is considered. The exchange energy is evaluated from the analytic pair-distribution functions, and the correlation energies are estimated via a closely similar four-component 2D electron fluid which has been investigated previously. SPPs appear for sufficiently high doping, when the exchange energy alone is considered. However, the inclusion of correlations is found to suppress the spin-phase transition in ideal graphene.

PACS numbers: PACS Numbers: 73.43.-f,73.50.-h,73.22.-f

Introduction—The carbon atoms in graphene form a quasi-two-dimensional (Q2D) honeycomb lattice and contribute one electron per carbon to form an unusual 2D electron system (2DES) with a massless Dirac-fermion dispersion near the Fermi points. Graphene and related materials (e.g., nanotubes, fullerenes) have become a mine of novel technologies and a new paradigm for studying various aspects of physics, including cosmological models on honeycomb branes, superconductivity on bi-partite lattices and nanotubes, Hubbard models, spin-phase transitions, and other aspects of strongly correlated electrons.

The hexagonal Brillouin zone has two inequivalent points $K=(1/3, 1/\sqrt{3})$ and $K’=(-1/3, 1/\sqrt{3})$, in units of $2\pi/a_0$, where $a_0$ is the lattice constant. The simplest tight-binding model with nearest-neighbour hopping $t$ is sufficient to describe the low-energy regime where the valence and conduction bands ($\pi$ and $\pi^*$) have linear dispersion near the $K$, $K'$ points, with zero bandgap. The graphene 2D electron system (G2DES) is nominally “half-filled”, with the $\pi^*$ band unoccupied, and has spin and valley degeneracies, with a Berry phase associated with the valley index.

The vanishing of the density of states and the effective mass of the 2D electrons near the Fermi points suggest that the Coulomb interactions of the massless fermions remain strong, unlike in the usual Fermi-liquid picture. This also means that perturbation approaches have to be treated with great caution. The Coulomb interactions may induce a gap between the $\pi$ and $\pi^*$ bands, or a lifting of the sublattice (valley) degeneracies, or stabilize spin-polarized phases (SPP) in preference to the unpolarized state. Such SPPs in GaAs/AlAs 2DES, predicted to appear at low coupling ($\sim 2 - 4$) when perturbation methods are used, get pushed to high coupling if non-perturbative approaches were used~(26), as discussed in Refs. 10, 11, 12. The two-valley 2D electron system (2v-2DES) does not show a SPP when full non-local non-perturbative calculations are used, presumably because these methods properly account for the direct interactions which are three times as many as the exchange interactions. The exchange and correlation energy $E_{xc}$ in the 2v-2DES of the Si/SiO$_2$ system was calculated from the distribution functions in Ref. 12, using the classical-map hyper-netted-chain (CHNC) technique, accurately recovering the Quantum Monte-Carlo (QMC) results even in the strong coupling regimes. CHNC provides the pair-distribution functions (PDFs) $g_{ij}(r)$ as a function of the coupling strength. Then $E_{xc}$ is evaluated via a coupling constant integration. The method has been successfully applied to a variety of problems including the 2DES14, the 2v-2DES in Si-MOSFET devices12, and the thick quasi-2DES in HIGFET structures16. However, a direct calculation of exchange and correlation in graphene involves and $8 \times 8$ matrix of two-component PDFs due to the spin and valley indices as well as the presence of $\pi, \pi^*$ bands. Hence in this study we consider the exchange energy $E_x$ via an analytic evaluation of the non-interacting PDFs, and indirectly estimate the correlation energy $E_c$ using results for the spin-polarized four-component 2v-2DES, exploiting the analogies between the two systems.

The non-interacting PDFs of G2DES, viz., $G_{ij}^0(r)$ involve two components, the first being a Bessel function as in the ordinary 2DES, while a second, associated with the cosine of the angle of e-e scattering, involves Bessel and Struve functions, as we show below. We find that there are stable SPPs in an exchange-only approach. However, including the correlation energy using the 2v-2DES data stabilizes the disorder-free G2DES in the unpolarized state.

Model for the graphene 2DES—The kinetic energy near the $K$ points is given by a Dirac-Weyl Hamiltonian of the form:

$$H_k = v_F (p_x \tau_z \sigma_x + p_y \sigma_y)$$ (1)
treat it as tunable. Assume maximized if no effective Bohr radius. The coupling constant $r$ of the G2DES. This plays the same role as the $r_1$ parameter in electron-gas theory of nonrelativistic finite-mass fermions. The usual $r_s$ is not available for G2DES since the effective mass $m^*$ is zero and there is no effective Bohr radius. The coupling constant $g^0$ is maximized if $\epsilon_0$ is unity, and consistent with this case we assume $g^0 = 2.75$ for our G2DES studies, and do not treat it as tunable.

The 4-component envelope-eigenfunctions of the kinetic energy term are made up of two-component functions $U = (b, e^{i\phi_k})$, $U' = (e^{i\phi_{k'}}, b)$ and $O = (0, 0)$ where $\phi_k$ is the angle of the vector $\vec{k}$ in the 2-D plane. Thus

$$F^{K}(r) = (2A)^{1/2} (U, O) T \chi \sigma \tag{3}$$
$$F^{K'}(r) = (2A)^{1/2} (O, U') T \chi \sigma \tag{4}$$

Here $b = \pm 1$ is a $\pi^*$, $\pi$ band index, $(\cdots)_{T}$ indicates the transpose, and $\chi_{\sigma}$ is the spin function. Then, using $v = 1, 2$ as a valley index, the Coulomb interaction may be written in the form:

$$H_I = \frac{1}{8A} \sum_{v_i, b_i, \sigma_i, \vec{k}, \vec{p}, \vec{q}} V_q \left[ b_{1}b_{2}e^{i\phi_{k} - \phi_{(k+\vec{q})}} + 1 \right]$$
$$\times \left[ b_{2}b_{3}e^{i\phi_{p} - \phi_{(p+\vec{q})}} + 1 \right] \times$$
$$\delta_{k_{1}b_{1}, \sigma_{1}a_{1}^{+}} \delta_{p_{1}b_{2}, \sigma_{2}a_{2}^{+}} \delta_{k_{1}b_{1}, \sigma_{1}} \delta_{p_{1}b_{2}, \sigma_{2}} \delta_{k_{1}b_{1}, \sigma_{1}} \delta_{p_{1}b_{2}, \sigma_{2}}$$

Here $a_{1}^{+}, a_{2}^{+}$ are electron creation and annihilation operators and $V_q = 2\pi e^2/(\epsilon_0 q)$ is the 2D Coulomb interaction. The phase factors introduce a novel $\cos(\theta)$ contribution where $\theta$ is the scattering angle, not found in the usual jellium-2DES. The resulting form of the exchange energy per Carbon is:

$$E_x/E_\sigma = -\frac{A_0 g^0}{(2\pi)^2} \frac{1}{k_c} \frac{1}{4} \sum_{b_1, b_2, \sigma} \int_{0}^{2\pi} d\theta dk dp$$
$$\times k_F \frac{1 + b_{1}b_{2} \cos(\theta)}{|k - p|} n_{b_1, \sigma}(k)n_{b_2, \sigma}(p) \tag{5}$$

Here we have introduced the intrinsic coupling constant $g^0$ and the energy unit $E_\sigma = v_F k_c$. Here $k_c = K_c/\sqrt{2} = \sqrt{(4\pi n_c)}$ is based on the electron density per spin species, $n_c = N_c/2 = 1/(2A_0)$. The above form of the exchange energy can be reduced to an evaluation of a few elliptic integrals. The normal “half-filled” G2DES can be doped with electrons or holes; but it is easy to show that symmetry enables us to limit to one type of doping. However, given a system with an areal density of $N_0$ dopant electrons per valley, with $n_d = N_d/2$ per spin, the carriers in the spin-polarized system could be electrons only, or both electrons and holes, as shown in Fig. 11 for the $\pi^*$ and $\pi$ bands at one K point. The intrinsic system with $n_d = 0$ can be an unpolarized state, as in Fig. 11a), or spin-polarized state with electrons and holes, as in Fig. 11c). Such exchange-driven systems have been studied by Peres et al. [8], while the correlation effects have not been considered. Since the correlation energy terms outnumber the exchange terms by 3:1, it is imperative to include the correlation energies. The calculation of correlation energies is always more difficult than exchange energies. Peres et al. treated $g^0$ as an externally tunable parameter for forcing SPPs. Here we evaluate the exchange energy $E_x$ at the intrinsic $g^0$, from the non-interacting PDFs, and indirectly evaluate the correlation.
energy $E_c$ from the four-component 2v-2DES with the same coupling strength ($r_s = g^0$) and spin-polarization.

Pair-distribution functions of the G2DES—Although we are dealing with a four-component system (2-valleys, 2-spin states), as seen from Fig. 1 we need to consider the redistribution of electrons and holes among the $\pi^*$ and $\pi$ bands when comparing the energy of spin-polarized states with the unpolarized state. From Fig. 1 we see that the e-e interactions at a given valley can be constructed from (i) interactions with a $\pi^*\sigma_u$ band of up-spin electrons of density $n_u$, filled to $k_u$, (ii) a $\pi^*\sigma_d$ set of electrons or a $\pi\sigma_u$ spin-down holes, of density $n_d$, filled up to $k_d$ (iii) the $\pi\sigma_d$ band, with electron density $n_e$, filled to $k_e$ and (iv) the $\pi\sigma_u$ band, density $n_e$, filled to $k_t$. There will also be similar inter-valley terms. Each term in this 4×4 matrix, denoted by $G_{ij}(r)$ where $i,j = 1\cdots 4$, will have two components associated with those in $U'$ and $U = (b,e^{i\phi_b})$. Thus $G_{ij}(r) = g_{ij}^b(r), g_{ij}^s(r)$, where the superfixes $\{b,s\}$ indicate that the noninteracting forms are Bessel-function like, and Struve-function like, respectively. The Struve form arises from the cos($\theta$) terms in the Coulomb interaction. The numbering scheme of the matrix is shown in Fig. 1(b). Thus, defining the pair-correlation functions (PCFs) $H_{ij}(r) = G_{ij}(r) - 1$, or its components $h_{ij}(r) = g_{ij}(r) - 1$, we have:

$$h_{ij}^{0,\beta}(r) = -(n_in_j)^{-1}\int_{k_0}^{k_i} \frac{dk_1}{(2\pi)^2} \int_{k_0}^{k_j} \frac{dk_2}{(2\pi)^2} e^{i(k_1-k_2)\cdot r}$$

$$= -\frac{2}{k_ir} J_1(k_ir) \frac{2}{k_nr} J_1(k_nr)$$

$$h_{ij}^{0,s}(r) = -\frac{n_in_j}{n_n} \int_0^{k_i} \frac{dk_1}{(2\pi)^2} \int_0^{k_j} \frac{dk_2}{(2\pi)^2} \cos(\theta) e^{i(k_1-k_2)\cdot r}$$

$$= -\frac{\pi}{k_ir} J_1(k_ir) \frac{\pi}{k_nr} J_1(k_nr) [J_0H_1 - J_1H_0]_d$$

Here $J_0, J_1$ are Bessel functions, while $H_0$ and $H_1$ are Struve $H$-functions. Also, in $[J_0H_1 - J_1H_0]_d$, the functions are evaluated at the argument $k_ir$. The wavevectors $k_i = \sqrt{4\pi n_i}$ are for each component $i$, of density $n_i$. We show typical noninteracting PCFs for a doped, unpolarized case as in Fig. 1(a), with the doping fraction $n_d/n_e = 0.2$. In CHNC, the exchange-hole is mapped exactly into a classical Coulomb fluid using the Lado procedure[16]. The figure shows that the exchange-hole is strongly reduced by the presence of the cos($\theta$) term which has been averaged into the Struve-like PCFs $h^\beta(r)$. When the Coulomb interaction is included, the cos($\theta$) term has a similar mitigating effect and exchange-correlation in the G2DES is actually considerably weaker than in the corresponding 2v-2DES which may be realized in a Si MOSFET. The CHNC calculation for the 2v-2DES for the conditions stipulated in Fig. 2 show that the correlation energy is only about a third of the exchange energy at $r_s = g^0$. This and other model calculations allow us to conclude that a good approximation is to ignore the $E_c$ contribution arising from the Struve-like distribution functions and estimate the $E_c$ only from the Bessel-like functions which are the only forms appearing in the 2v-2DES. This justifies our use of the 2v-2DES for the correlation energy, while the $E_x$ is exactly evaluated.

The kinetic and exchange energies.—When the doping per valley is $N_\delta = 2n_\delta$, the total number of electrons per valley is $N = N_e + N_\delta$. Also, using the $i = 1, 2, 3, 4$ notation of Fig. 1(b), we set $n_1 = n_u$, $n_2 = n_d$, $n_3 = n_4 = n_c$. Hence the spin polarization $s = n_u - n_d$, where the band index $b_d = -1$ for holes. The degree of spin-polarization $\zeta = s/N$.

The composition fractions, inclusive of the valley index $v = 1, 2$ are $x_{vi} = n_i/N_i$. We note that $k_F = \sqrt{2\pi n_\delta}$, $k_u = \sqrt{2\pi(n_\delta + s)}$, $k_d = \sqrt{2\pi(n_\delta - s)}$. The exchange energy $E_x(n_\delta, \zeta)$ can be written as:

$$E_x(n_\delta, \zeta)/N_i = (N_i/2) \int \frac{2\pi r dr}{r}$$

$$\times \sum_{ij} x_{vi} x_{vj} [G^{0}_{ij}(r) - 1]$$

It is implied that the Struve-like component in $G_{v,v',ij}$ where $v$ label the valleys, is summed with the appropriate $b_rb_j$ band ± factors. Only a sum over the components in one valley is needed in evaluating the exchange. The calculation of both exchange and correlation involves exactly the same formula, but with a further integration over a coupling constant $\lambda$ included in the PDFs $G_{v',v'\prime,ij,\lambda}$. The above formula for $E_x$ is made more explicit below. Thus, the total kinetic and exchange energy...
pressed in terms of elliptic integrals. We have done the
PDFs are replaced by their
G2DES we note that
5 of Ref. [12]. In transferring from the 2v-2DES to the
energy/per electron,
4 analogous four-component 2v-2DES. This correlation en-
ting
polarized system shown in Fig. 1(a), is obtained by set-
E
ζ
zarising from the Bessel-like
v
2
0.0025
ζ
0.1 0.2 0.3 0.4
0.005
ζ
0.1 0.2 0.3
f
d
0.005
f
d
= electron doping fraction
FIG. 3: Upper panel- the energy difference \( \Delta E_{kz} \), i.e.,
K.E+exchange, between the polarized and unpolarized
phases, in units of \( E_n = v_F k_c \), as a function of the spin polarization \( \zeta \) and the dopant fraction \( f_d \). Electron-carrier systems, Fig. 3(b) are more stable than electron-hole systems, and show stable spin-polarized states. However, addition of the correlation energy (lower panel) makes the unpolarized state the most stable phase.

predominate over exchange interactions, and the SPP is suppressed, as found in CHNC [12] or QMC [12]. Here too, we conclude that the inclusion of correlations suppresses the exchange-driven spin-phase transition in the ideal graphene system.

\* Email address: chandre.dharma-wardana@nrc.ca

[1] P.R. Wallace, Phys. Rev. 71, 622 (1947); G. Semenoff, Phys. Rev. Lett. 53, 2449 (1984); D. P. DiVincenzo and E. J. Mele, Phys. Rev. B 29, 1685 (1984)
[2] for a recent review, see T. Ando, J. Phys. Soc. Jpn. 74, 777 (2005)
[3] A. Geim, Physics Today, 59 21 (2006)
[4] J. Nagamatsu et al. Nature(London) 410, 63 (2001)
[5] N. Tit and M. W. C. Dharma-wardana, Europhys. Lett., 62, 405 (2003) J. Gonzalez, Phys. Rev. Lett. 88, 76403 (2002)
[6] S. Sorella and E. Tosatti, Europhys. Lett. 19, 699 (1996); N. M. R. Peres, M. N. A. Araujo and D. Bozil, Phys. Rev. B 70, 195122 (2004)
[7] K. S. Novoselov, A. K. Geim, S. V. Morozov, D. Jiang,
M. I. Katsnelson, I. V. Grigorieval, S. V. Dubunos, and A. A. Firsov. Nature 438, 197 (2005); Y. B. Zhang Y-W Tan, H. L. Stormer, and P. Kim Nature (London). 438, 201 (2005)

[8] N. M. R. Peres, F. Guinea and A. H. Castro Neto, Phys. Rev. 72 174406 (2005)
[9] D. V. Khveshchenko, arXiv:cond-mat/064180
[10] C. Attaccalite, S. Moroni, P. Gori-Giorgi, and G. B. Bachelet, Phys. Rev. Lett. 88, 256601 (2002)
[11] M. W. C. Dharma-wardana and F. Perrot., Phys. Rev. Lett. 90, 136601 (2003)
[12] M. W. C. Dharma-wardana and F. Perrot, Phys. Rev. B 70, 035308 (2004)
[13] S. Conti and G. Senatore, Europhys. Lett. 36, 695, (1996)
[14] François Perrot and M. W. C. Dharma-wardana, Phys. Rev. Lett. 87, 206404 (2001)
[15] M. W. C. Dharma-wardana, Phys. Rev. B 72, 125339 (2005)