Excitonic instability in two-dimensional degenerate semimetals

H. Leal and D. V. Khveshchenko
Department of Physics and Astronomy, University of North Carolina, Chapel Hill, NC 27599

We study the possibility of excitonic pairing in layered degenerate semimetals such as graphite, where the electron density of states almost vanishes at the Fermi level and, therefore, the Coulomb interactions remain essentially unscreened. By focusing on the Dirac-like low-energy electron excitations and numerically solving a non-linear gap equation for the order parameter, we obtain a critical value of the Coulomb coupling and establish the Kosterlitz-Thouless-like nature of a putative semimetal-to-excitonic insulator transition.

In layered graphite, a poor screening of the Coulomb interaction sets the stage for a novel form of excitonic instability resulting in the opening of a gap in the quasi-two-dimensional electronic spectrum and manifesting itself through the onset of an insulating charge density wave [1].

It was also argued in Ref. [1] that the pseudo-relativistic kinematics of the Dirac-like electronic excitations in a single sheet of graphite allows one to draw a formal parallel with the phenomenon of chiral symmetry breaking (CSB) that has long been studied in the context of the three-dimensional Quantum Electrodynamics (QED_{2+1}) and other relativistic fermion theories.

In the case of QED_{2+1}, a zero temperature CSB transition was predicted to occur for a sufficiently small ($N < N_c$) number of fermion species, regardless of the interaction strength [2], while the short-ranged Higgs-Yukawa (HY) four-fermion interactions can drive this transition for any N, provided that the HY coupling is strong enough [3].

The CSB scenario has recently become a common theme of several QED_{2+1}-like descriptions of the pseudogap phase in underdoped cuprates [4–6]. In this regard, some authors [5] cited the original estimates of the critical number of fermions $N_c^{(0)} \approx 3.24$ [2] to support the conjecture that the inherent spin density wave instability of the d-wave symmetrical pseudogap state can be readily described by the conventional QED_{2+1} theory with $N = 2$ species of the Dirac-like nodal quasiparticles.

However, the mounting analytical [7] and numerical [8] evidence indicating that in the conventional QED_{2+1} the actual critical number of flavors N_c may be less than two seems to suggest otherwise and calls for a need to further modify the minimal QED_{2+1} theory of the pseudogap phase (possibly, beyond recognition).

The implications of the results of Refs. [7,8] made other groups [6] emphasize a potential importance of additional four-fermion couplings and/or anisotropic quasiparticle dispersion (in high-T_c cuprates, the quasiparticle velocity is strongly dependent on the direction, $v_1/v_2 \sim 10-20$), whose systematic account has yet to be done. It has been recently argued, however, that N_c may not be affected by the dispersion anisotropy at all [9], thus implying that the four-fermion couplings (which are absent in the conventional QED_{2+1}) may indeed play a crucial role in the theory of the pseudogap-to-antiferromagnet transition in underdoped cuprates.

In the problem of the conjectured excitonic instability in graphite, the non-relativistic nature of the Coulomb interaction further invalidates any naive attempts to make use of the results pertaining solely to the relativistically invariant systems. Nevertheless, the earlier work on the subject has already provided for some analytical evidence that the excitonic transition may indeed occur for a sufficiently strong Coulomb coupling [1,10].

It was also predicted [11] that a magnetic field normal to the layers might further facilitate the formation of excitonic insulator, which would then be reminiscent of the phenomenon of magnetic catalysis introduced in the abstract field-theoretical setting [12]. This scenario was discussed [10,11] in connection with the recent reports of a magnetic field-induced insulating behavior in highly oriented pyrolytic graphite (HOPG) [13].

In the present paper, we numerically solve the gap equation for the excitonic order parameter derived in Refs. [1,10], thus putting to the test some of the predictions made in the earlier analytical work on the subject.

We start out by reviewing the derivation of the gap equation. In the vicinity of the two (in the absence of a lattice strain, exactly degenerate and labeled as $i = 1, 2$) conical K-points of the 2D Brillouin zone of graphite, the low-energy excitations of the valence and the conduction bands with a linear dispersion $E_k = \pm v_F k$ where $v_F \approx 2 \times 10^6 m/s$ can be described as two-component (Weyl) spinors $\psi_{i\sigma}$, of which there are N different species with the spin index $\sigma = 1, ..., N$ [14].

Provided that the Zeeman coupling to an external magnetic field is much weaker than the orbital (diamagnetic) term, the number of fermion species in graphite is $N = 2$, and the dimensionless Coulomb coupling $g = 2\pi e^2/\epsilon v_F$ with the dielectric constant $\epsilon \approx 2.8$ appears to be the only relevant parameter.

However, in a strong in-plane field, the Zeeman splitting between the spin-up and spin-down bands, while having no effect on the electron orbital motion, reduces the number of fermions down to $N = 1$, which prompts one to treat N as yet another (to a certain extent) adjustable parameter.

The pair of spinors $\psi_{i\sigma}$ can be further combined into a

single bi-spinor $\Psi_\sigma = (\psi_{1\sigma}, \psi_{2\sigma})$, thereby allowing one to use the Dirac-like representation for a kinetic energy of the two-dimensional electrons in a single layer of graphite

$$H_K = iv_F \sum_{\sigma=1}^{N} \int_{\mathbf{r}} \overline{\Psi}_\sigma (\hat{\gamma}_1 \nabla_x + \hat{\gamma}_2 \nabla_y) \Psi_\sigma \qquad (1)$$

where $\overline{\Psi}_\sigma = \Psi_\sigma^\dagger \hat{\gamma}_0$ and the 4×4 (reducible) representation of the γ-matrices $\hat{\gamma}_{0,1,2} = (\tau_3, i\tau_2, -i\tau_1) \otimes \tau_3$ satisfying the anticommutation relations $\{\hat{\gamma}_\mu, \hat{\gamma}_\nu\} = 2\mathrm{diag}(1,-1,-1)$ is constructed in terms of the Pauli triplet $\tau_{1,2,3}$.

Accordingly, the Coulomb interaction term in the Hamiltonian takes the form

$$H_C = \frac{1}{4\pi} \sum_{\sigma,\sigma'=1}^{N} \int_{\mathbf{r},\mathbf{r}'} \overline{\Psi}_\sigma(\mathbf{r}) \hat{\gamma}_0 \Psi_\sigma(\mathbf{r}) \frac{g}{|\mathbf{r}-\mathbf{r}'|} \overline{\Psi}_{\sigma'}(\mathbf{r}') \hat{\gamma}_0 \Psi_{\sigma'}(\mathbf{r}') \qquad (2)$$

Both Eqs.(1) and (2) remain invariant under arbitrary $U(2N)$ rotations of the $2N$-component vector $(\Psi_{L\sigma}, \Psi_{R\sigma})$ composed of the chiral (L,R) parts of the Dirac fermion Ψ_σ defined as $\Psi_{(L,R)\sigma} = \frac{1}{2}(\mathbf{1} \pm \hat{\gamma}_5)\Psi_\sigma$ where the matrix $\hat{\gamma}_5 = \mathbf{1} \otimes \tau_2$ anticommutes with any $\hat{\gamma}_\mu$.

In the relativistic theories of Refs. [2,3], the standard CSB pattern $U(2N) \to U(N) \otimes U(N)$ is signaled by the development of a singlet order parameter $\Delta_s(\mathbf{r}) =< \sum_\sigma^N \overline{\Psi}_\sigma(\mathbf{r}) \Psi_\sigma(\mathbf{r}) >$, which is the type of excitonic pairing that we focus upon below.

The standard Dyson-Schwinger equation for the Dirac propagator reads (hereafter $p_\mu = (\epsilon, \mathbf{p})$)

$$\hat{G}^{-1}(\omega,\mathbf{p}) = \hat{G}_0^{-1}(\omega,\mathbf{p}) - T\sum_{\Omega} \int \frac{d^2\mathbf{k}}{(2\pi)^2} V(\Omega-\omega, \mathbf{k}-\mathbf{p}) \hat{\gamma}_0 \hat{G}(\Omega,\mathbf{k}) \hat{\gamma}_0 \qquad (3)$$

where $\hat{G}_0(\omega,\mathbf{p}) = \hat{\gamma}_\mu p_\mu / p^2$ is the bare propagator of the gapless Dirac fermions and the renormalized Coulomb interaction

$$V(\Omega,\mathbf{q}) = \frac{g}{\mathbf{q} + Ng\chi(\Omega,\mathbf{q})} \qquad (4)$$

is modified, as compared to its bare form, by to the fermion polarization $\chi(\Omega,\mathbf{q})$.

Being primarily concerned with the possibility of a spontaneous gapping of the conical spectrum, we search for a solution to Eq.(3) in the form $\hat{G}(p) = (\hat{\gamma}_\mu p_\mu + \Delta(p))^{-1}$, while neglecting both the vertex and the fermion wave function renormalization. On the basis of the experience gained from the relativistic theories [2,15], one might expect that, albeit being capable of affecting such practically important details as the value of the critical coupling $g_c(N)$, the latter do not alter the very existence of a solution (or a lack thereof).

Proceeding along the lines of the previous numerical analyses of the finite temperature version of QED_{2+1} [15], below we focus on the momentum dependence of the static ($\omega = 0$) component of the order parameter. To this end, we neglect all but the $\Omega = 0$ harmonics of the effective interaction (4) which, as shown in Refs. [15], suffices for determining the critical conditions for the emergent CSB order. In the case of interest, this static approximation appears to be even better justified, since the Lorentz invariance is broken already at zero temperature.

The static component of the finite-temperature fermion polarization can, in turn, be approximated by the expression

$$\chi(0,\mathbf{q}) = \frac{1}{8}\left(|\mathbf{q}| + \frac{T}{C}\exp\left(-C\frac{|\mathbf{q}|}{T}\right)\right). \qquad (5)$$

which, for $C = \pi/16\ln 2$, provides an up to a few percent accurate interpolation between the two opposite limits: $v_F\mathbf{q} \gg T$ where Eq.(5) agrees with the zero-temperature result, $\chi(0,\mathbf{q}) \sim |\mathbf{q}|$, and $v_F\mathbf{q} \ll T$ where it accounts for thermal screening, $\chi(0,\mathbf{0}) \sim T$ [15].

Taking the sum over the Matsubara frequencies in Eq.(3), we then arrive at the gap equation derived in Refs. [1,10] (hereafter we use the units $v = \hbar = 1$)

$$\Delta(\mathbf{p}) = \int \frac{d^2\mathbf{k}}{8\pi^2} V(0,\mathbf{k}-\mathbf{p}) \Delta(\mathbf{k}) \frac{\tanh\frac{\sqrt{\mathbf{k}^2+\Delta(\mathbf{k})^2}}{2T}}{\sqrt{\mathbf{k}^2+\Delta(\mathbf{k})^2}} \qquad (6)$$

This form of the gap equation is more familiar in condensed matter-related applications where the gap is routinely considered to be a function of momentum but not energy, consistent with our use of the static approximation.

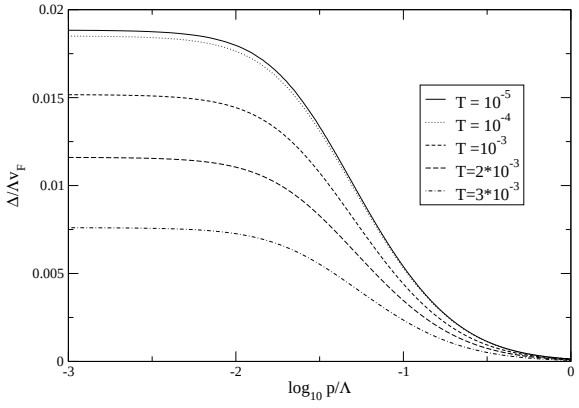

FIG. 1. Momentum dependence of the solution to the gap equation (6) for $N = 1$ and different temperatures.

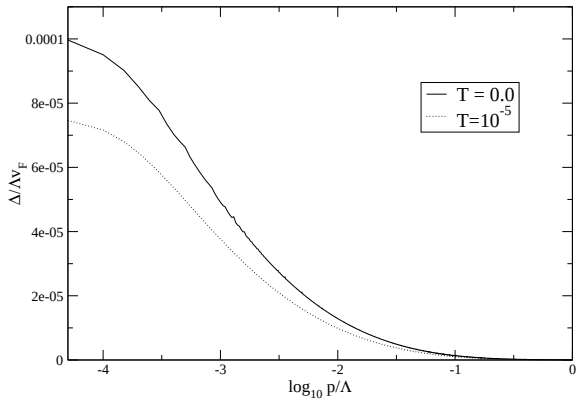

FIG. 2. Solution to the gap equation for $N = 2$.

In fact, even a partial account of the momentum and/or energy dependence of the gap function goes well beyond the customary BCS-like (constant) solution for the gap. In the case of short-ranged repulsive interactions, a BCS-like solution of the analog of Eq.(6) was recently discussed in conjunction with the conjectured excitonic instability in hexaborides [16]. By contrast, in a degenerate semimetal such as graphite the strong momentum dependence of the unscreened Coulomb interaction rules out a constant solution ($\Delta^{BCS}(\mathbf{p}) = \Delta$) altogether.

In Fig.1, we present the results of our numerical solution to Eq.(6) for $N = 1$ and several different temperatures. As a function of momentum, the gap $\Delta(\mathbf{p})$ levels off at $\mathbf{p} \sim \Delta(0)$, in accord with the approximate analytical solution of Refs. [1,10] where Eq.(6) was substituted with a differential equation complemented by the boundary conditions $d\Delta(\mathbf{p})/d\mathbf{p}|_{\mathbf{p}=0} = 0$ and $\Delta(\mathbf{p}) + \mathbf{p}d\Delta(\mathbf{p})/d\mathbf{p}|_{\mathbf{p}=\Lambda} = 0$, Λ being the upper momentum cutoff given by the maximum span of the Brillouin zone.

The functional dependence of the solution for $N = 2$ is similar to the $N = 1$ case, apart from the overall suppression by roughly two orders of magnitude (see Fig.2).

In the strong coupling zero-temperature ($g \to \infty$ and $T \to 0$) limit, the N-dependence of the zero-momentum gap shown in Fig.3 can be well fitted with the formula

$$\Delta(0) = 1.2 v_F \Lambda \exp\left(-1.7\pi/\sqrt{N_c^{\infty}/N - 1}\right) \qquad (7)$$

which manifests the Kosterlitz-Thouless nature of the excitonic transition that occurs at $T = 0$ and $N_c^{\infty} \equiv N_c(g = \infty) \approx 2.6$, in agreement with the predictions made in Refs. [1,10].

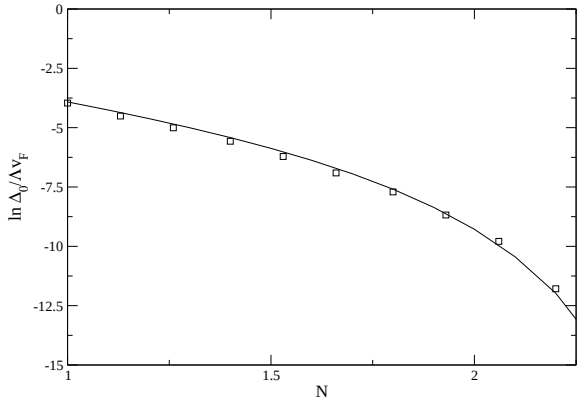

FIG. 3. Zero-momentum gap for different values of N at $T = 10^{-5}$ and $g \to \infty$. The solid line is the fit given by Eq.(7).

In Fig.4, we present the N-dependence of the critical coupling $g_c(N)$. For $N = 2$, our numerical result $g_c(N = 2) \approx 7$ differs by a factor of two from that obtained analytically in Ref. [10], although the agreement improves for smaller N.

Notably, the relevant values of g_c are rather large, which indicates that any weak-coupling approach would be utterly inadequate for the problem in question.

Even with the possible caveat that the gap equation tends to systematically underestimate the critical strength of the repulsive interactions [7,8], the above values compare favorably with the estimates $g \sim 5 - 10$ obtained for the HOPG samples of Ref. [13].

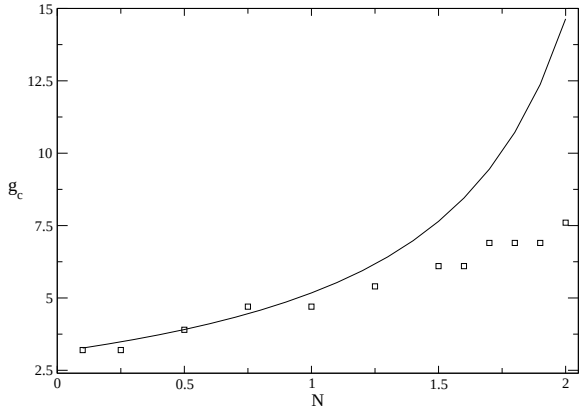

FIG. 4. The critical coupling g_c for different values of N and $T = 0$. The solid line shows the analytical result of Ref.10.

For any N, the zero-momentum gap attains its maximum value $\Delta(0) \approx 10^{-4} v_F \Lambda$ at $g \to \infty$. Using the parameters of the HOPG band structure, we estimate the maximal possible gap at $N = 2$ as $\Delta(0) \sim 30K$, which turns out to be small compared to the typical Fermi energy $E_F \sim 250K$ in HOPG [13]. The latter characterizes

the actual (as opposed to the idealized point-like) Fermi surface of graphite which represents a combined effect of inter-layer hopping, finite doping, and/or disorder.

At this point, it remains unknown to what extent the above factors can modify Eq.(6) derived for a clean two-dimensional sheet of undoped graphite. On the other hand, in a layered system the propensity towards excitonic pairing is further strengthened by the inter-layer Coulomb repulsion, which, considering the fact that $g > 1$, might be even more important than the finite E_F [1].

To this end, it is worth mentioning that the singlet excitonic order parameter is directly related to the electron density imbalance between the A and B sublattices of the bi-partite hexagonal lattice: $\Delta_s(\mathbf{p} = \mathbf{0}, \mathbf{r}) = \sum_{\sigma=1,2} < \overline{\Psi}_\sigma(\mathbf{r})\Psi_\sigma(\mathbf{r}) >= \sum_{i\sigma=1,2}(\delta_{\mathbf{r},A} < \psi^\dagger_{i\sigma}(A)\psi_{i\sigma}(A) > -\delta_{\mathbf{r},B} < \psi^\dagger_{i\sigma}(B)\psi_{i\sigma}(B) >)$.

In a multi-layer system stacked in the staggered $(A-B)$ configuration, the inter-layer Coulomb repulsion favors spontaneous depletion (respectively, pile-up) of the electron density on a sublattice formed by the carbon atoms above and below the centers (respectively, corners) of the hexagons in adjacent layers. The resulting commensurate charge density wave alternates between the layers, thereby keeping the electrons in the adjacent layers as far apart as possible and nudging the system closer to the excitonic instability.

In order to decide on the ultimate outcome of the competition between the frustrating effect of the extended Fermi surface and the strong (yet, non-singular, unlike Eq.(2)) inter-layer Coulomb repulsion, the present analysis has to be further refined by incorporating the above factors through, e.g., a non-linear fermion dispersion and effective four-fermion terms [17].

Although, thus far, we were only interested in the possibility of singlet pairing, for $N = 2$ and in the leading Coulomb approximation (small transferred momenta) the formation of a triplet order parameter appears to be just as likely. In fact, similar to the case of the short-ranged repulsion discussed in Refs. [18], the excitonic ground state possesses a degeneracy with respect to arbitrary $SU(4)$ rotations of the four-dimensional complex vector composed of the singlet Δ_s and the triplet $\vec{\Delta}_t = \sum_{\sigma,\sigma'=1,2} < \overline{\Psi}_\sigma \vec{\sigma}_{\sigma\sigma'} \Psi_{\sigma'} >$ order parameters.

This approximate degeneracy gets lifted upon including the (so far, neglected) short-ranged Coulomb exchange interactions which cause transitions between the conduction and valence bands [18]. Alongside the Zeeman coupling, the latter favor the triplet order parameter, thus enforcing the Hund's rule.

It is also conceivable that, with increasing doping, the triplet excitonic insulator can give way to an itinerant ferromagnetic metal. The interest in this scenario which had been previously discussed only in the case of the three-dimensional non-degenerate semimetals was bolstered by the discovery of a weak ($\sim 0.07\mu_B$ per carrier), yet robust ($T_c \sim 600 - 1000K$), ferromagnetism in hexaborides [16].

However, more recent experimental studies of hexaborides [19] have indicated the presence of a large spectral gap which seems to rule out the excitonic mechanism, thus putting graphite in a rather unique position of the best currently known candidate for excitonic ferromagnet. This possibility appears to be particularly intriguing in the light of the reports of a comparably weak ($\sim 0.1\mu_B$ per carrier) magnetization observed in HOPG samples at room temperatures [20].

The conditions for the emergence of excitonic ferromagnetism and the putative global phase diagram of graphite will be discussed at a greater length in our future work [17].

To summarize, in the present paper we obtained a numerical solution to the gap equation describing the conjectured excitonic transition in two-dimensional semimetals such as graphite. From our solution, we inferred the minimal necessary strength of the Coulomb coupling g_c and the largest possible number of fermion species N_c^∞ for which this Kosterlitz-Thouless-type transition may occur at zero temperature. Although this analysis was carried out for a single undoped layer, we believe that our predictions for the existence of a critical Coulomb coupling, momentum dependence of the gap function, and the nature of the excitonic transition should remain robust upon including further complicating factors such as inter-layer coupling, doping, and disorder.

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