Magnetic-Field-Induced Insulating Behavior in Highly Oriented Pyrolitic Graphite

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We propose an explanation for the apparent semimetal-insulator transition observed in highly oriented pyrolitic graphite in the presence of magnetic field perpendicular to the layers. We show that the magnetic field opens an excitonic gap in the linear spectrum of the Coulomb interacting quasiparticles, in close analogy with the phenomenon of dynamical chiral symmetry breaking in the relativistic theories of the (2 + 1)-dimensional Dirac fermions. Our strong-coupling approach allows for a nonperturbative description of the corresponding critical behavior.

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The recently discovered carbon-based materials provide a new playground for applications of the advanced methods of quantum field theory. Although the best known example is that of the one-dimensional carbon nanotubes described as the Luttinger liquid, some of the available nonperturbative techniques can also be applied to higher dimensional systems, such as layered highly oriented pyrolitic graphite (HOPG).

In a single sheet of graphite, the low-energy spectrum of the quasiparticle excitations becomes linear in the vicinity of the two conical points located at the opposite corners of the two-dimensional Brillouin zone, where the conduction and valence bands touch each other [1]. These low-energy excitations can be conveniently described in terms of a four-component Dirac spinor $\Psi_\sigma = (\psi_{1A\sigma}, \psi_{1B\sigma}, \psi_{2A\sigma}, \psi_{2B\sigma})$, combining the Bloch states $\psi_{i\sigma}(\mathbf{r})$ with spin σ which are composed of the momenta near one of the conical points ($i = 1, 2$) and propagate independently on the two different sublattices ($\mathbf{r} = A, B$) of the bipartite hexagonal lattice of the graphite sheet. In the following discussion, we will treat the number of the spin components N as a tunable parameter, the physical case corresponding to $N = 2$.

The use of the Dirac spinor representation allows one to cast the quasiparticle kinetic energy in the relativisticlike form

$$K = iv \sum_{\sigma=1}^{N} \int d^2\mathbf{r}\, \overline{\Psi}_\sigma(\hat{\gamma}_1 \nabla_x + \hat{\gamma}_2 \nabla_y)\Psi_\sigma, \qquad (1)$$

where $\overline{\Psi}_\sigma = \Psi_\sigma^\dagger \gamma_0$. The reducible representation of the 4×4 $\hat{\gamma}$ matrices $\hat{\gamma}_{0,1,2} = (\tau_3, i\tau_2, -i\tau_1) \otimes \tau_3$ satisfying the usual anticommutation relations $\{\hat{\gamma}_\mu, \hat{\gamma}_\nu\} = 2\,\mathrm{diag}(1, -1, -1)$ is given in terms of the triplet of the Pauli matrices τ_i, and the velocity $v \sim 10^6$ m/s is proportional to the width of the electronic π-orbital band.

The Lorentz invariance of the noninteracting Hamiltonian is not, however, respected by the interaction term

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

which accounts for the long-range part of the Coulomb coupling whose strength is characterized by the dimensionless parameter $g = 2\pi e^2/\epsilon_0 v$.

The earlier perturbative studies of the effects of the Coulomb interaction resulted in the prediction that, upon renormalization, the strength of the effective coupling $g(\epsilon) \sim 1/|\ln\epsilon|$ monotonically decreases with the energy ϵ, hence the paramagnetic semimetallic ground state remains stable [2]. However, this conclusion appears to contradict the recent experimental observation of a ferromagnetic bulk magnetization (inconsistent with the estimated number of magnetic impurities) in the HOPG samples showing insulating behavior of the resistivity [3].

In fact, the large value of the bare Coulomb coupling $g \gtrsim 10$ suggests that a more appropriate starting point might be the strong coupling regime, where perturbation theory fails and a more capable approach is needed. In this Letter, we propose such an approach by focusing on a recent experimental observation of the apparent magnetic field-driven semimetal-insulator transition in HOPG [4] and demonstrate that external magnetic field can trigger the instability towards excitonic insulator phase. Interestingly enough, the latter appears to have much in common with the phenomenon of chiral symmetry breaking (CSB) which has been previously studied in the relativistic theories of the interacting Dirac fermions.

In the case of graphite, the issue of CSB comes about due to the invariance of Eqs. (1) and (2) under arbitrary U($2N$) rotations of the $2N$-component vector comprised of the chiral Dirac fermions $\Psi_{(L,R)\sigma} = \frac{1}{2}(\mathbf{1} \pm \hat{\gamma}_5)\Psi_\sigma$, where the matrix $\hat{\gamma}_5 = \mathbf{1} \otimes \tau_2$ anticommutes with any $\hat{\gamma}_\mu$.

In a quantum system, strong interactions can give rise to the appearance of a fermion mass and gapping of the fermion spectrum, thereby breaking the continuous chiral symmetry U($2N$) down to its subgroup U(N) $\otimes$ U(N) which corresponds to the independent rotations of $\Psi_{L\sigma}$ and $\Psi_{R\sigma}$.

As one important (2 + 1)-dimensional example, CSB can be caused by interaction with a scalar Higgs-Yukawa (HY) bosonic mode coupled to the Dirac fermions via the mass operator $\sum_\sigma \overline{\Psi}_\sigma\Psi_\sigma$. In this case, CSB is known to occur for any number of fermion species N, provided that the strength of the (intrinsically attractive) HY coupling exceeds a certain critical value. Recently, this model was applied to the analysis of the condensed matter systems, where the scalar bosonic field describes fluctuations of a superconducting order parameter [5] or piezoelectric phonons [6].

In contrast, the repulsive Lorentz-invariant vectorlike coupling via the current operator $\sum_\sigma \overline{\Psi}_\sigma \hat{\gamma}_\mu \Psi_\sigma$ drives the Dirac fermions towards the CSB transition, regardless of the coupling strength, provided that the number of fermion species N is sufficiently small ($N < N_c$). This behavior is believed to occur in the strong-coupling infrared fixed point in the $(2 + 1)$-dimensional quantum electrodynamics (QED_3) where the zero temperature value of N_c was found to be smaller or equal to $3/2$ [7].

However, this situation changes drastically in the presence of the magnetic field which suppresses the orbital motion of the Dirac fermions and collapses their spectrum into a discrete set of the (relativistic) Landau levels. This effectively reduces the dimensionality of the problem, thus enabling CSB to occur, regardless of the coupling strength and/or the number N of fermion species.

In the Dirac picture of the quasiparticle excitations in layered graphite, the onset of CSB would be manifested by a nonzero value of the order parameter $\sum_\sigma \overline{\Psi}_\sigma \Psi_\sigma = \sum_{i\sigma}(\psi^\dagger_{iA\sigma}\psi_{iA\sigma} - \psi^\dagger_{iB\sigma}\psi_{iB\sigma})$. It determines the magnitude of the fermion gap proportional to the electron density imbalance between the A and B sublattices and corresponds to the formation of a site-centered charge density wave (CDW) in the excitonic insulating ground state.

While for a sufficiently small N the excitonic instability could develop even in a single layer of graphite [8], in the physical case $N = 2$ it is unlikely to occur in the absence of the interlayer Coulomb repulsion. Indeed, in a realistic HOPG system consisting of many layers stacked in a staggered configuration, the latter favors spontaneous depletion of the electron density on one of the two sublattices (e.g., A) formed by the carbon atoms positioned, vertically, at the centers (respectively, corners) of the hexagons in the adjacent layers. Within each layer, such a depletion (and excess occupation of the complementary sublattice, e.g., B) conforms to one of the two degenerate CDW patterns which then alternate between the layers, thereby keeping the electrons in the adjacent layers as far apart as possible and further strengthening the propensity towards the excitonic instability. Although the minimal strength of the interlayer Coulomb repulsion required for CSB to occur in HOPG remains unknown, this whole situation, too, changes when the system is exposed to magnetic field normal to the layers which promotes CSB even in the absence of the interlayer coupling.

In the presence of the magnetic field B, the Dirac fermion Green function remains diagonal in the space of the physical spin (not to be confused with the $4 \otimes 4$ space of the γ matrices representing the orbital dynamics), and, for a given σ, it reads as

$$\hat{G}(x,y) = e^{i/2(x-y)_\mu A_\mu(x+y)} \hat{\mathcal{G}}(x - y), \tag{3}$$

where the translationally noninvariant phase factor contains the vector potential of the external field $A_\mu(x) = (0, -Bx_2/2, Bx_1/2)$. Upon separating this factor out, one obtains a translationally (albeit not Lorentz-) invariant Green function $\hat{\mathcal{G}}(x)$.

The effect of the fermion interactions can be fully accounted for by introducing the gap function $\Delta(p)$ as well as the wave function (Z) and velocity (Z_v) renormalization factors into the Fourier transform of $\hat{\mathcal{G}}(x)$ given by the integral representation [hereafter, we use the units $v = e = \hbar = 1$ and the relativistic notations, such as $p_\mu = (\epsilon, \mathbf{p})$]

$$\hat{\mathcal{G}}(p) = \frac{i}{B}\int_0^\infty ds \exp\left[-\frac{s}{B}\left(\Delta^2(p) - Z^2\epsilon^2 + Z_v^2\mathbf{p}^2\frac{\tanh s}{s}\right)\right] \times \{[\Delta(p) + Z\epsilon\hat{\gamma}_0](1 - i\hat{\gamma}_1\hat{\gamma}_2\tanh s) - Z_v\hat{\gamma}\mathbf{p}(1 - \tanh^2 s)\} \tag{4}$$

which the exact Green function naturally inherits from the bare one, $\hat{\mathcal{G}}_0(p)$, given by Eq. (4) with $Z = Z_v = 1$ and $\Delta(p) = 0$.

Owing to its nonperturbative nature, the phenomenon of CSB eludes weak-coupling analysis based on perturbation theory. Nonetheless, akin to its relativistic counterpart, the occurrence of CSB in the system of the Coulomb interacting Dirac fermions can be inferred from the nonperturbative solution of the Dyson equation for the renormalized Green function

$$\hat{\mathcal{G}}^{-1}(p) - \hat{\mathcal{G}}_0^{-1}(p) = ig\int\frac{d^3k}{(2\pi)^3}Z\frac{\hat{\gamma}_0\hat{\mathcal{G}}(p + k)\hat{\gamma}_0}{|\mathbf{k}| + Ng\chi(k)}. \tag{5}$$

In Eq. (5) we made use of the Ward identity relating the vertex function $\hat{\gamma}_0 Z$ to the energy derivative of $\mathcal{G}^{-1}(p)$ and cast the effective intralayer Coulomb interaction in the form governed by the scalar (density-density) component of the fermion polarization operator

$$\chi(k) = i\,\mathrm{Tr}\int\frac{d^3p}{(2\pi)^3}Z\gamma_0\hat{G}(p + k)\gamma_0\hat{G}(p). \tag{6}$$

As a result of the broken Lorentz invariance, due to both the nonrelativistic nature of the Coulomb interaction and the presence of the magnetic field, the solution of the gap equation (5) can feature totally different dependencies on the energy and momentum variables. Apart from the Zeeman shift $\sigma\mu_B B$ of the position of the Fermi level for the spin-σ fermions, Eq. (5) remains spin degenerate.

The analysis of Eq. (5) is complicated by the fact that, unlike in the previous studies of the excitonic transition in semimetals with overlapping conduction and valence bands, the naive picture of static Debye screening

($\chi_k \approx$ const) fails to properly describe the feedback from the planar Dirac fermions on the bare Coulomb interaction. Instead, in the undoped or lightly doped graphite with a low density of carriers the zero temperature fermion polarization (6) can be expressed only in the form of a cumbersome double integral [9],

$$\chi(k) = \frac{Z\mathbf{q}^2}{2\sqrt{\pi B}} \int_0^\infty \frac{\sqrt{u}\,du}{\sinh u} \int_{-1}^{1} dv\,(\cosh uv - v\coth u \sinh uv) \times \exp\left[-\frac{u}{B}\left(\Delta^2 - \frac{1}{4}(1 - v^2)Z^2\omega^2\right) - \frac{Z_v^2\mathbf{q}^2}{2B\sinh u}(\cosh u - \cosh uv)\right]. \tag{7}$$

Nonetheless, progress towards obtaining the solution of Eq. (5) can still be made in the strong field limit where the distance between the adjacent Landau levels of the noninteracting Dirac fermions $E_n = \pm\sqrt{2|n|B}$ by far exceeds the Coulomb interaction-related energy gap Δ, and the only relevant fermion states appear to be those of the so-called lowest Landau level (LLL) with $n = 0$. In this regard, the problem at hand bears a certain resemblance to the fractional quantum Hall effect (FQHE) in the system of nonrelativistic fermions with a parabolic dispersion. In contrast to the spatially homogeneous FQHE, however, the sought solution of the gap equation corresponds to a nonuniform CDW ground state, as explained above.

In the strong field approximation $\Delta \ll \sqrt{B}$ the fermion Green function reduces to its LLL projection

$$\hat{\mathcal{G}}_{\mathrm{LLL}}(p) \approx ie^{-Z_v^2\mathbf{p}^2/B}\frac{Z\epsilon\hat{\gamma}_0 - \Delta}{\Delta^2 - Z^2\epsilon^2}(1 - i\hat{\gamma}_1\hat{\gamma}_2) \tag{8}$$

and, correspondingly, the zero temperature fermion polarization receives its main contribution from the transitions between the LLL and the first excited Landau level

$$\chi_{\mathrm{LLL}}(\omega, \mathbf{k}) \approx \sqrt{2B}\,\frac{Z\mathbf{k}^2}{B - Z^2\omega^2/2}\,e^{-Z_v^2\mathbf{k}^2/2B}. \tag{9}$$

Neglecting the wave function, velocity, and vertex renormalizations in the scalar part of Eq. (5) (see below), one readily obtains a closed equation for the gap function

$$\Delta(p) = i\int \frac{d\omega\,d\mathbf{k}}{(2\pi)^3}\frac{\Delta(k + p)}{(\epsilon + \omega + i\delta)^2 - \Delta^2(k + p)} \times \frac{ge^{-[(\mathbf{k}+\mathbf{p})^2+\mathbf{p}^2]/B}}{|\mathbf{k}| + \sqrt{B}\,gN\mathbf{k}^2e^{-\mathbf{k}^2/2B}(B - \omega^2/2)^{-1}}. \tag{10}$$

Equation (10) should be contrasted with that derived in the strong field limit in the case of QED_3 where the LLL projection eliminates all but the scalar component of the Lorentz-invariant Abelian gauge interaction, thus resulting in the gap equation (10) with the term $|\mathbf{k}|$ in the denominator of the integrand replaced by $k^2 = \mathbf{k}^2 - \omega^2$, as required by the Lorentz invariance of the bare gauge field spectrum.

It is this difference which gives rise to the nontrivial energy dependence of the solution of the QED_3 gap equation found in Ref. [9]. By contrast, in our case the solution of Eq. (10) remains independent of the energy variable as long as $\epsilon \lesssim \sqrt{B}$ and, being the sole function of the momentum, it falls off faster than $e^{-2\mathbf{p}^2/B}$ for $|\mathbf{p}| > \sqrt{B}$. It can also be shown that the approximate constancy of $\Delta(p)$ at small momenta and energies justifies our neglecting the wave function and velocity renormalization ($Z = Z_v = 1$).

The magnitude of the zero-momentum gap $\Delta = \Delta(0)$ estimated from Eq. (10),

$$\Delta \approx \frac{\sqrt{B}\ln(Ng)}{4\pi N}, \tag{11}$$

demonstrates that the strong field condition holds at large N regardless of the field strength B (which should, however, be smaller than the width of the linear part of the bare quasiparticle spectrum).

At finite temperatures the gap decreases, and its momentum dependence at $|\mathbf{p}| \lesssim (T\sqrt{B})^{1/2}$ becomes even less pronounced. Although the prohibitive form of the finite temperature fermion polarization impedes analytical calculations, its main effect can be taken into account through the modified lower cutoff in the logarithmically divergent momentum integral in Eq. (10) which now becomes the larger of $\sqrt{B}/gN$ and $(T\sqrt{B})^{1/2}$.

After being extended to finite temperatures, Eq. (10) yields the self-consistent equation for the magnitude of the gap at small momenta

$$\Delta_T \approx \frac{\sqrt{B}}{4\pi N}\ln\frac{\sqrt{B}}{\max[\sqrt{B}/gN; (T\sqrt{B})^{1/2}]}\tanh\frac{\Delta_T}{2T}. \tag{12}$$

Equation (12) possesses a nontrivial solution below the transition temperature $T_c(B)$ whose large-N estimate is given by the expression

$$T_c \approx \frac{\sqrt{B}\ln N}{16\pi N}. \tag{13}$$

Conversely, Eq. (13) determines a threshold magnetic field $B_c(T) \propto T^2$ which has to be exceeded in order for CSB to occur at a nonzero temperature T. This relation defines a critical line in the $B - T$ phase diagram, along which the gap vanishes as

$$\Delta_T(B \to B_c) \propto \sqrt{B - B_c(T)} \tag{14}$$

As far as the nature of the CSB transition is concerned, the dependence (14) is characteristic of the second order transition, whereas the conjectured zero field transition (for $N < N_c$) is of the topological (Kosterlitz-Thouless) type [8].

These predictions should be compared with the available experimental evidence obtained from the HOPG samples showing metallic behavior of the zero field resistivity. The data of Ref. [4] indicate that, albeit absent in zero field, the apparent semimetal-insulator transition can be induced by magnetic field normal to the layers. The insulating behavior was found to set in at the field-dependent characteristic temperature fitted as $T^* \propto \sqrt{B - B_0}$ which, apart from the offset field B_0, agrees with Eq. (13) and falls into the experimental range $\lesssim$100 K for the applied magnetic field $B \lesssim 0.2$ T (in order to avoid confusion we recall that in our quasirelativistic system the role of the "speed of light" is played by v). The proposed orbital (as opposed to the spin-related) nature of the observed phenomenon is also consistent with the findings of Ref. [4], according to which the in-plane magnetic field has a substantially (2 orders of magnitude) weaker effect.

In fact, the spin degeneracy between the triplet and singlet excitonic gaps (for $N = 2$) will be lifted upon including the short-range Coulomb exchange interaction omitted in Eq. (2) which involves transitions between the conduction and valence bands. Alongside the Zeeman coupling, the latter favors the triplet excitonic order parameter, in accordance with Hund's rule. In the doped system, the spin up and down states will then be occupied asymmetrically at low temperatures, resulting in the occurrence of a ferromagnetic spin polarization $\mathbf{M} = \mathrm{Tr}(\vec{\sigma}G)$ in a window of the electron chemical potential μ set by the gap Δ. The induced ferromagnetic moment will then be proportional to $\mu \sim \Delta$ as well, which compares favorably with the electron spin resonance (ESR) data in HOPG showing the presence of the magnetization $\mathbf{M} \propto \sqrt{B - B_0}$ on the insulator side of the field-induced transition [10].

Before concluding, we comment on the previous attempts to apply the alternate scenario of the magnetic field-driven CSB in the Lorentz-invariant HY model to the analysis of the quasiparticle transport in the mixed state of the planar d-wave superconductors [11]. Specifically, the authors of Ref. [11] focused on the experimentally observed kinklike feature in the magnetic-field dependence of the total (inclusive of both the normal quasiparticle and the phonon contributions) thermal conductivity, the position of the kink scaling with temperature as $B^*(T) \propto T^2$ [12] (this behavior was not seen, however, in a more recent experiment [13]).

On the theoretical side, the suggestion of using the standard HY model to describe the nodal d-wave quasiparticles in the mixed state must be taken cautiously. Apart from the default choice of the quasiparticle interaction in the form of the attractive HY coupling, the "magnetic catalysis" scenario of Refs. [11] requires that the total effective magnetic field, which the nodal quasiparticles are exposed to, has a nonzero overall flux, resulting in the formation of the Landau levels.

However, as pointed out by several authors [14], the correct picture of the d-wave quasiparticle spectrum is rather that of the extended energy bands, owing to the fact that in the mixed state the quasiparticles are actually experiencing both the external magnetic field and the solenoidal superfluid flow created by the vortices. On average, the two fluxes exactly cancel each other [14], thus rendering inapplicable the standard HY mechanism of the magnetic catalysis even in the regime of weak-to-moderate magnetic fields, where the individual vortices strongly overlap and the physical magnetic field is almost uniform. In light of the above, HOPG might be the only presently known example of a condensed matter system where the phenomenon of magnetic catalysis can indeed occur.

In summary, we study the problem of the Coulomb interaction-driven electronic instabilities in layered graphite in the presence of magnetic field. Elaborating on the relativisticlike description of the low-energy quasiparticle excitations in a single sheet of graphite, we propose a possible explanation for the recently discovered field-induced semimetal-insulator transition by showing that applied magnetic field induces the excitonic insulator phase, thus gapping up the quasiparticle spectrum and creating a site-centered CDW.

Since the phenomenon in question was observed only in perpendicular magnetic field, we believe that the experimental findings of Refs. [4,10] reveal some novel, intrinsically two-dimensional, physics whose origin is different from that of the (semi)metal-insulator transitions observed in other carbon-based materials, such as the one-dimensional [(Ru, Cs)C_{60}] as well as three-dimensional (KC_{60}) alkali doped fullerides. The latter are likely to be associated with the structural instabilities and/or accompanied by the onset of the antiferromagnetic spin-Peierls state. In order to decisively discriminate between these possibilities, such experimental techniques as ESR, NMR, x-ray diffraction, and electron photoemission can be further implemented.

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