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Coulomb-interacting Dirac fermions in disordered graphene

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We study such experimentally relevant characteristics of the Coulomb-interacting Dirac quasiparticles in disordered graphene as the quasiparticle width and density of states that can be probed by photoemission, magnetization, and tunneling measurements. We find that an interplay between the unscreened Coulomb interactions and pseudorelativistic quasiparticle kinematics can be best revealed in the ballistic regime, whereas in the diffusive limit the behavior is qualitatively similar to that of the ordinary two-dimensional electron gas with parabolic dispersion.

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The recent advances in microfabrication of graphitic monolayers[1] have made it possible to test and confirm the earlier theoretical predictions of anomalous, relativisticlike, kinematic properties of the electronic states in graphene.[2] Thus far, both experimental and theoretical studies have been primarily focusing on the unusual (magneto)transport phenomena which, for their most part, can be described in terms of noninteracting Dirac quasiparticles propagating ballistically in the bulk or in finite geometries.

However, it has long been recognized that a (nearly) degenerate two-dimensional semimetal such as graphene might provide a unique playground for studying the effects of the Coulomb interactions, because the latter are expected to remain essentially unscreened.[3]

Among the previously discussed manifestations of the Coulomb correlations is a possible opening of the interaction-induced excitonic gap at sufficiently strong Coulomb couplings.[4] Alternatively, such a gap can also be generated by a magnetic field, which phenomenon represents the Dirac counterpart of fractional Quantum Hall effect (FQHE) in the conventional two-dimensional electron gas (2DEG) with parabolic electron dispersion, as was pointed out in Ref. 5 (see also Ref. 6 for the references to the earlier studies of a related phenomenon of "magnetic catalysis" in the abstract field-theoretical setting).

Should a spectral gap develop, it would be exhibited by the conductivity and other transport characteristics. However, apart from the recent observation[7] of a complete lifting of the fourfold degeneracy of the $n=0$ Landau level (which phenomenon would indeed be consistent with the scenario of a field-induced gap opening, resulting in a spontaneous breakdown of the sublattice symmetry[5,6]), no conclusive evidence of such a behavior has yet been found.

Also, if proven to be of a genuine bulk nature (as opposed to being due to magnetic impurities, edges, and/or structural defects), the previously reported weak, albeit robust, ferromagnetism in pyrolytic graphite[8] could be indicative of a possible instability towards a (weakly) ferromagnetic excitonic state with unequal gaps for the spin-up and spin-down electrons.[4]

Obviously, a further experimental work is needed in order to accertain a real status of the scenario of a latent excitonic insulator proposed in Ref. 4, as well as contrasting it with such alternative predictions as that of the Stoner instability resulting in a fully polarized ferromagnetic state.[9]

In light of this uncertainty, in the present Rapid Communication we focus on the effects of the moderately strong Coulomb correlations which might not be powerful enough to generate a finite gap in the Dirac spectrum. As we demonstrate below, even in this case the quasiparticle properties can be affected in a number of experimentally relevant ways. To that end, we study the quasiparticle width and density of states in both the ballistic and diffusive regimes, and contrast the results against those pertaining to the ordinary 2DEG with parabolic dispersion.

An extensive experience gained in the course of the previous studies of the conventional 2DEG suggests that such transport characteristics as the longitudinal and Hall dc conductivities may not provide the best means of revealing the Coulomb correlations. A general reason is that the two-particle response functions probed by transport measurements appear to be only weakly affected by such correlations due to a routine cancellation between the (potentially, large) fermion self-energy and vertex corrections. In that regard, a greater insight into the physics of interacting Dirac fermions can be provided by various single-particle probes, including photoemission, tunneling, and magnetization measurements.

The low-energy properties of graphene are governed by the electronic states in the vicinity of one of the two inequivalent nodal points ($\alpha=1,2$). Such states can be described by the Dirac Hamiltonian[2,3]

$$H = iv_F \sum_{\alpha=1,2} \int_{\mathbf{r}} \Psi^\dagger_\alpha [\hat{\sigma}_x \nabla_x + (-1)^\alpha \hat{\sigma}_y \nabla_y] \Psi_\alpha + \frac{v_F}{4\pi} \sum_{\alpha,\beta=1,2} \times \int_{\mathbf{r}} \int_{\mathbf{r}'} \Psi^\dagger{}_\alpha(\mathbf{r}') \Psi_\alpha(\mathbf{r}') \frac{g}{|\mathbf{r}-\mathbf{r}'|} \Psi^\dagger{}_\beta(\mathbf{r}) \Psi_\beta(\mathbf{r}), \tag{1}$$

where v_F is the Fermi velocity, $g_0 = 2\pi e^2/\epsilon_0 v_F \sim 3$ is the bare value of the dimensionless Coulomb coupling, and $\hat{\sigma}_i$ is a triplet of the Pauli matrices acting in the space of (pseudo)spinors $\Psi_\alpha = (\psi_\alpha(A), \psi_\alpha(B))$ composed of the values of the electron wave function on the A and B sublattices of the bipartite hexagonal lattice of graphene.

The effects of the Coulomb interactions on the fermion propagator and interaction function are encoded in the fermion self-energy Σ and polarization operator Π,

$$\hat{G}^R_\alpha(\omega,\mathbf{p})^{-1} = (\omega+\mu)\hat{1} - v_F[\hat{\sigma}_x p_x + (-1)^\alpha \hat{\sigma}_y p_y] + \hat{\Sigma}^R(\omega,\mathbf{p}),$$
$$V^R(\omega,\mathbf{q}) = (q/g_0 + \Pi^R(\omega,\mathbf{q}))^{-1}. \tag{2}$$

In the one-loop approximation, the former is given by the expression

$$\hat{\Sigma}^R(\epsilon,\mathbf{p}) = \int \frac{d\omega}{(2\pi)} \sum_{\mathbf{q}} \left(\text{Im}\, V^A(\omega,\mathbf{q})\hat{G}^R(\epsilon+\omega,\mathbf{p}+\mathbf{q})\coth\frac{\omega}{2T} - V^A(\omega,\mathbf{q})\text{Im}\,\hat{G}^R(\epsilon+\omega,\mathbf{p}+\mathbf{q})\tanh\frac{\epsilon+\omega}{2T}\right). \quad (3)$$

The linear-in-momentum term in Re $\hat{\Sigma}^R(\epsilon,\mathbf{p})$ gives rise to a renormalization of the running Coulomb coupling $g(\omega)$ described by the renormalization-group (RG) equation derived in Ref. 3,

$$\frac{dg(\omega)}{d\ln(\Omega/\omega)} = -\frac{1}{8\pi}g^2(\omega). \quad (4)$$

The solution $g(\omega)\approx g_0/[1+(g_0/8\pi)\ln(\Omega/\omega)]$, where Ω is an upper cutoff of order the electronic bandwidth, shows that the effective Coulomb coupling slowly decreases with decreasing energy.

At a finite temperature T, chemical potential μ, or elastic quasiparticle width γ the RG flow described by Eq. (4) terminates below the energy scale $\sim\max[T,\mu,\gamma]$.

At $T>0$ and/or in the presence of disorder, a functional form of the fermion polarization operator becomes quite prohibitive. However, in the ballistic limit and near half filling ($\mu\approx 0$), it can still be approximated as follows:

$$\Pi^R(\omega,\mathbf{q}) \approx \frac{1}{4v_F}\frac{\mathbf{q}^2}{\sqrt{v_F^2\mathbf{q}^2-(\omega+i0)^2}}, \quad Q_+ \gg T \approx \frac{2T\ln 2}{\pi v_F}\left(1-\frac{\omega}{\sqrt{(\omega+i0)^2-v_F^2\mathbf{q}^2}}\right), \quad Q_+ \ll T, \quad (5)$$

where $Q_+^2=\omega^2+v_F^2\mathbf{q}^2$.

In Eq. (5), the first expression is the temporal component of the Lorentz-invariant free fermion polarization bubble computed at $T=0$, whereas the second one exhibits the (well-known in high-energy physics) phenomenon of "thermal Debye screening" and concomitant Landau damping due to thermally excited quasiparticles.

A straightforward analysis of Eq. (3) shows that the inelastic quasiparticle width defined as $\Gamma(\epsilon,\mathbf{p}) = \text{Im Tr}\hat{\Sigma}^R_{in}(\epsilon,\mathbf{p})$ exhibits a strong "light-cone" singularity, akin to that previously encountered in the studies of the normal quasiparticles in d-wave superconductors where the commonly quoted T^3 behavior of the inverse quasiparticle lifetime represents a rough estimate that is only applicable to the thermal quasiparticles with energies and momenta $\epsilon\sim vp\sim T$ (see Ref. 10).

When evaluated to the lowest order in the Coulomb coupling, $\Gamma(\epsilon,\mathbf{p})$ appears to be discontinuous at $\epsilon=v_Fp$ and singular at $\epsilon, v_Fp\to 0$:

$$\delta\Gamma(\epsilon,\mathbf{p}) \sim g^2\theta(P_-^2)P_+, \quad P_+>T \sim g^2\theta(P_-^2)\frac{T^2}{P_+}, \quad P_+<T, \quad (6)$$

where $P_\pm^2=\epsilon^2\pm v_F^2\mathbf{p}^2$, and $\theta(x)$ is the Heaviside step function.

In both regimes, the dominant contribution comes from the transferred momenta of order $\sim P_+$ due to the long-ranged nature of the Coulomb coupling [for a screened interaction, the last line in Eq. (6) would be replaced with $\sim(T^3/P_+)^{1/2}$].

At $p=T=0$ the linear energy dependence of the quasiparticle width was predicted in Refs. 3. It is worth noting, however, that in the case of bulk graphite discussed in Ref. 3 the linear dependence would only hold at the momenta higher than the inverse interlayer separation $1/d$, while for $q\lesssim 1/d$ the screened interaction potental becomes less singular, $V(\mathbf{q})\approx g_0(d/q)^{1/2}$.

By contrast, in the case of graphene Eq. (6) would indeed hold all the way down to the energies $\sim\max[T,\mu,\gamma]$, if it were not for the higher order corrections. In order to estimate their effect, we recalculate Eq. (3) with the random-phase approximation (RPA)-dressed interaction function accounting for the fermion polarization (5), which procedure yields the result

$$\Gamma(\epsilon,\mathbf{p}) \sim \theta(P_-^2)\frac{P_-^2}{P_+}\ln g, \quad P_+ \gtrsim gT \sim \theta(P_-^2)\left(g\frac{P_-^2T}{P_+}\right)^{1/2}\ln\frac{P_+}{T}, \quad T\lesssim P_+ \lesssim gT \quad (7)$$

while for $P_+\lesssim T$ the logarithmic factor (which would have been absent altogether for any interaction less singular than Coulomb) disappears from Eq. (7). In contrast to Eq. (6), the result (7) remains continuous at the threshold $\epsilon=v_Fp$.

In the presence of potential disorder, the low-energy quasiparticle width is dominated by the elastic part of the self-energy. A closed expression for the latter can be readily obtained in the case of short-range impurities with concentration n_i and scattering amplitude u,

$$\hat{\Sigma}^R_{el}(\epsilon,\mathbf{0}) = \frac{n_iu^2(\epsilon+i\gamma)\ln(\Omega/\epsilon+i\gamma)}{1-u^2(\epsilon+i\gamma)^2 ln^2(\Omega/\epsilon+i\gamma)}\hat{\mathbf{1}}. \quad (8)$$

Equation (8) allows for a self-consistent calculation of the zero-energy quasiparticle width $\gamma=\text{Im Tr}\hat{\Sigma}^R_{el}(0,0)$, ranging from the Born ($u\to 0$) to the unitarity ($u\to\infty$) limit. It is worth mentioning, however, that Eq. (8) would need to be further modified in the potentially relevant case of the Coulomb impurities.[11]

Having obtained γ, one can compute the noninteracting density of states (DOS) at the Fermi energy and the corresponding Drude conductivity

$$\nu_0 = -\frac{1}{\pi}\text{Im Tr}\sum_{\mathbf{p}}\hat{G}_0^R(0,\mathbf{p}) \approx \max\left(\frac{\gamma}{2\pi v_F^2}\ln\frac{\Omega}{\gamma},\frac{4\mu}{v_F^2\pi}\right), \quad \sigma_0 \approx \frac{e^2}{h}\max\left(\frac{4}{\pi},\frac{\mu}{\gamma}\right), \quad (9)$$

where $\hat{G}_0^R(\epsilon,\mathbf{p})$ accounts for the impurity-induced broadening, but does not include any inelastic scattering.

In order to study a crossover between the ballistic and diffusive regimes, we use the formula

$$\Pi^R(\omega,\mathbf{q}) = \frac{1}{4v_F}\frac{\mathbf{q}^2}{\sqrt{v_F^2\mathbf{q}^2-(\omega+i\gamma)^2}-\gamma} \quad (10)$$

interpolating between Eq. (5) for $Q_+\gtrsim\gamma$ and the standard diffusive expressions, such as $\Pi^R(\omega,\mathbf{q})=\sigma_0\mathbf{q}^2/(D\mathbf{q}^2-i\omega)$, for $Q_+ \lesssim \gamma$ (here $D=\sigma_0/\nu_0$ is the diffusion coefficient).

In the ballistic limit ($\epsilon, T \gg \gamma$) the total self-energy is approximately given by the sum of Eqs. (7) and (8), whereas in the opposite, diffusive, regime ($\epsilon, T \lesssim \gamma$) the inelastic width can be found from a self-consistent equation

$$\Gamma(\epsilon,T) = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi}\left[\tanh\left(\frac{\epsilon+\omega}{2T}\right) - \coth\left(\frac{\omega}{2T}\right)\right] \times \sum_{\mathbf{q}} \text{Im}\, \frac{V^R(\omega,\mathbf{q})}{Dq^2 - i\omega + \Gamma(\epsilon+\omega,T)} \qquad (11)$$

whose solution behaves as

$$\Gamma(\epsilon,T) \sim \frac{\max[\epsilon,T]}{\sigma_0} \ln \frac{\sigma_0^2 g_\gamma^2 \gamma}{\max[\epsilon,T]}, \qquad (12)$$

where $g_\gamma = g(\omega \sim \gamma)$.

The total quasiparticle width can be deduced from the angular resolved photoemission (ARPES) data.[12] Alternatively, it can be inferred from the de Haas–van Alphen (dHvA) experiments and (to the extent that the oscillating part of the resistivity is indicative of the behavior of the single-particle Green function) the Shubnikov de Haas (SdH) ones. Namely, fitting the magnetization data to the formula

$$\Delta M(B) \sim \frac{T\mu^2}{eBv_F^2} \sum_{n=1}^{\infty} \frac{n \sin(\pi n \mu^2/eB)}{\sinh(2\pi^2 nT\mu/eB)} e^{-2\pi n\mu\Gamma(\mu,T)/eB} \qquad (13)$$

can provide, apart from such a spectacular hallmark of the free Dirac kinematics as the geometric (Berry) phase π,[13] a valuable information on the energy and temperature dependence of the quasiparticle width.

In the same spirit, one can estimate the Coulomb-controled phase breaking time $\Gamma_\phi(T) \sim (T/\sigma_0)\ln \sigma_0$ whose temperature dependence (familiar from the theory of the conventional 2DEG) can be manifested by magnetoresistance associated with the localization corrections to the Drude conductivity.[14]

Yet another viable experimental probe is provided by tunneling measurements. Previous studies have been primarily concerned with the behavior of the electronic DOS in the vicinity of strong potential impurities.[15] However, despite offering a greater experimental observability of such prominent features as a resonant peak at [or close to, if the particle-hole symmetry is broken by subdominant terms in Eq. (1)] zero energy, the near-impurity DOS appears to be highly nonuniversal and therefore reveals more information about the impurity potential itself than about the Coulomb correlations in the host electronic system. Notably, a typical plot of the near-impurity DOS (Ref. 15) appears to be very similar to that obtained in the case of a d-wave superconductor (see, e.g., Ref. 16 and references therein) where the Coulomb interactions would be completely screened out by the condensate.

In view of the above, in what follows we concentrate on the bulk DOS, the first interaction correction to which is given by the expression

$$\delta\nu(\epsilon) = \frac{1}{\pi}\sum_{\mathbf{p}} \text{Im}\,\text{Tr}[\hat{G}_0^R(\epsilon,\mathbf{p})]^2 \hat{\Sigma}^R(\epsilon,\mathbf{p}) = \sim -g\epsilon \ln\frac{\Omega}{\epsilon},$$

$$\epsilon, T \gg \gamma \sim -\frac{\nu_0}{\sigma_0} \ln\frac{\gamma}{\epsilon} \ln\frac{\tilde{\gamma}}{\epsilon}, \quad \epsilon, T \lesssim \gamma, \qquad (14)$$

where $\tilde{\gamma} = \gamma\sigma_0^4 g_\gamma^4$. In the ballistic regime, the correction to the bare (linear) DOS features an additional (as compared to the case of the conventional disordered 2DEG, Ref. 17) logarithmic factor due to the aforementioned kinematic "light-cone" singularity, while in the diffusive limit one obtains the same diffusion-related (double-log) enhancement, as in the standard case.

Associated with the DOS correction (14), there are the Altshuler-Aronov-type contributions to such observables as specific heat and quasiparticle conductivity which, unlike their weak-localization counterparts,[14] cannot be readily suppressed by an external in-plane magnetic field.

Beyond the leading approximation, one finds an interference between the Coulomb interactions and disorder, which further modifies the behavior of the idealized (clean and noninteracting) Dirac fermion system. Given the large bare strength of the Coulomb interaction, the higher-order terms might contribute significantly, thus prompting one to employ an adequate nonerturbative technique.

To that end, we make use of the tunneling action method of Refs. 18. Adapting this approach to the case of graphene, we cast the tunneling DOS in the form

$$\nu(\epsilon) \approx -\frac{1}{\pi}\,\text{Im}\,\text{Tr}\int_{-\infty}^{\infty} \hat{G}_0^R(\mathbf{0},t) e^{-S(t)+i\epsilon t} dt. \qquad (15)$$

The disorder-averaged real-space and time Green function $\hat{G}_0^R(\mathbf{0},t) \propto e^{-\gamma t}/t^2$ is computed in the absence of the Coulomb interactions, while the latter are incorporated through the (imaginary part of) the action

$$S(t) = \int \frac{d\omega}{4\pi} \coth\frac{\omega}{2T} \sum_{\mathbf{q}} \text{Im}\, V(\omega,\mathbf{q}) \times \int_0^t dt_1 \int_0^t dt_2 e^{-i\omega(t_1-t_2)} \langle e^{i\mathbf{q}[\mathbf{r}(t_1)-\mathbf{r}(t_2)]}\rangle \qquad (16)$$

which describes the spreading of the excess charge associated with an act of tunneling into the graphene sample from, e.g., the surface tunneling microscope (STM) tip.

In the path-integral language, the averaging in Eq. (16) is carried out over all the quasiparticle trajectories $\mathbf{r}(t)$ contributing to the tunneling amplitude.[18] In the ballistic regime, one obtains $\langle e^{i\mathbf{q}(\mathbf{r}_1-\mathbf{r}_2)}\rangle \approx 1$, whereas in the diffusive limit $\langle e^{i\mathbf{q}(\mathbf{r}_1-\mathbf{r}_2)}\rangle \approx e^{-D\mathbf{q}^2}$.

To facilitate a direct contact with experiment, we evaluate the tunneling conductance

$$G(V,T) \sim \frac{d}{dV}\int_0^{\infty} [n(V+\epsilon) - n(\epsilon)]\nu_{FL}(V+\epsilon)\nu(\epsilon) d\epsilon \propto \int d\epsilon \frac{\nu(\epsilon)}{T\cosh^2(V+\epsilon/2T)}, \qquad (17)$$

where $\nu_{FL}(\epsilon) \approx$ const is the electron DOS of the normal (Fermi-liquid-like) STM tip biased at a voltage V.

In the ballistic regime ($V, T \gg \gamma$) Eq. (16) yields

$$S(t) \approx \frac{g_0^2}{(4\pi)^2}\ln(\Omega t), \quad g_0 \ll 1$$
$$\approx \frac{1}{\pi^2}\ln(\Omega t)\ln\left(\frac{8\pi}{e}\ln(\Omega t)\right), \quad g_0 \gg 1 \tag{18}$$

thereby resulting in the approximate power-law behavior

$$G(V,T) \propto \max[V,T]^{1+\eta}. \tag{19}$$

At weak coupling, the "zero-bias anomaly" (19) features a purely algebraic behavior with the anomalous exponent $\eta = g_0^2/(4\pi)^2$. In contrast, at strong bare coupling the energy dependence of $g(\omega)$ gives rise to an approximate power-law decay where the effective exponent $\eta(V,T)$ deviates slowly (only as $\sim \ln\ln\Omega/\max[V,T]$) from the universal value $\eta = (1/\pi^2)\ln(8\pi e) \approx 0.43$ attained at $\max[V,T] = \Omega$.

In the diffusive regime ($T, V \lesssim \gamma$), the running coupling $g(\omega)$ levels off at the value g_γ, and the counterpart of Eq. (18) reads

$$S(t) \approx \frac{1}{(4\pi)^2\sigma_0}\ln(t\tilde{\gamma})[\ln(t\gamma) + O(1)], \quad 1/\gamma < t < 1/T,$$
$$\approx \frac{1}{(4\pi)^2\sigma_0}\ln\frac{\tilde{\gamma}}{T}\left(\ln\frac{\gamma}{T} + 2Tt\right), \quad t > 1/T. \tag{20}$$

As a result, for $\mu \gg \gamma$ (or $\sigma_0 \gg 1$) there exists an interval $\sqrt{\sigma_0} < \ln(\gamma/\max[V,T]) < \sigma_0$ where one obtains the dependence similar to that of the conventional disordered 2DEG,[18]

$$G(V,T) \propto \nu_0 \exp\left[-\frac{1}{16\pi^2\sigma_0}\ln\left(\frac{\gamma}{\max[V,T]}\right)\ln\left(\frac{\tilde{\gamma}}{\max[V,T]}\right)\right]. \tag{21}$$

At still lower biases and/or temperatures ($\max[V,T] < \gamma e^{-4\pi^2\sigma_0}$) the conductance resumes a linear dependence

$$G(V,T) \propto \frac{e^{4\pi^2\sigma_0}}{\sigma_0^{1/2} g_\gamma^2}\max[V,T] \tag{22}$$

reminiscent of the noninteracting DOS, but with a completely different prefactor. In contrast to the intermediate asymptotic regime (21) that can only occur in the strongly metallic case ($\mu \gg \gamma$), the linear dependence (22) might be expected to set in at the lowest biases and temperatures for an arbitrary electron density. It is worth emphasizing, however, that, unlike in the case of the conventional 2DEG, the bare DOS of graphene is entirely due to disorder at low electron densities ($\mu \lesssim \gamma$).

In summary, we analyzed the effects of moderately strong Coulomb interactions on the Dirac quasiparticle excitations in graphene. Taken at their face values, the above results for the quasiparticle width and DOS suggest that the Dirac physics can be best revealed in the ballistic regime [see Eqs. (7) and (19)], while the diffusive dynamics of this system [see Eqs. (12), (21), and (22)] appears to be deceptively similar to that of the conventional 2DEG. These predictions of both, novel and mundane, features can be tested in future experiments on photoemission, tunneling, and magnetization measurements.

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