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Quasiparticle spectral function in doped graphene: Electron-electron interaction effects in ARPES

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We calculate the electron self-energy as well as the quasiparticle spectral function in doped graphene, taking into account electron-electron interaction in the leading order dynamically screened Coulomb coupling and electron-impurity interaction associated with quenched disorder. Our theory provides the basis for calculating all one-electron properties of extrinsic graphene. Comparison with existing angle-resolved photoemission spectroscopy measurements shows broad qualitative and semiquantitative agreement between theory and experiment, for both the momentum-distribution and energy-distribution curves in the measured spectra.

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The great deal of current activity[1] in two-dimensional (2D) graphene arises from its possible technological significance as a new 2D electronic material where carrier density can be controlled by an external gate voltage, and from its fundamental significance as a novel 2D zero-band-gap semiconductor system with chiral linear Dirac-like electron-hole band energy dispersion. In particular, the chiral linear energy dispersion with the conduction and the valence band crossing at the "Dirac point" has naturally led to an interesting analogy with QED, whereas the gate-induced tunability of graphene carrier density brings up a tantalizing analogy with Si metal-oxide-semiconductor field-effect transistors MOSFETs. An important question in this context is the extent to which the Coulomb interaction between carriers will modify or renormalize the chiral linear electron-hole band dispersion in graphene. Study of the many-body renormalization of single-particle properties due to electron-electron interactions is one of the most important topics in modern solid state physics.[2]

In this Rapid Communication we theoretically consider carrier interaction effects in *extrinsic* graphene by calculating the many-body self-energy, the quasiparticle spectral function, and the renormalized momentum distribution function of graphene in the presence of free carriers (i.e., for doped or gated graphene where carriers fill the 2D band up to the Fermi level E_F). Our results show that, as already argued in the literature,[3] although the quantitative renormalization effects of interactions on the graphene single-particle properties are substantial, extrinsic graphene remains an effective 2D Fermi liquid "metal," qualitatively preserving its noninteracting chiral linear band dispersion even in the presence of mutual Coulomb interaction. More importantly, our theoretical calculation of the interacting spectral function enables an excellent understanding of the recent experimental angle-resolved photoemission spectroscopy (ARPES) data which obtained the graphene spectral function.[4,5]

The quasiparticle spectral function $A(\mathbf{k},\omega)$ is a central quantity in the many-body physics of interacting systems, with $A(\mathbf{k},\omega)\equiv -2\,\mathrm{Im}\,G(\mathbf{k},\omega)$ where $G(\mathbf{k},\omega)$ is the single-particle (retarded) Green function for momentum $\mathbf{k}$ and energy ω (we use $\hbar=1$ throughout this paper). For the noninteracting bare system, we immediately get $A_0(\mathbf{k},\omega)=2\pi\delta(\omega-\varepsilon_{sk}+E_F)$ where $\varepsilon_{sk}=s\gamma k$ with $k\equiv|\mathbf{k}|$ is the bare graphene linear band dispersion with $s=\pm1$ denoting the conduction $(+1)$ and the valence (-1) band, and $\gamma\approx 10^6$ cm/s the band velocity. We will assume that the system is n doped with electrons filling the graphene conduction band up to a free carrier density (n) dependent chemical potential or Fermi level given by $E_F=\gamma k_F$, where the Fermi momentum $k_F=(\pi n)^{1/2}$. We have taken into account the spin and the valley degeneracy of graphene in obtaining the Fermi momentum. The noninteracting spectral function $A_0(\mathbf{k},\omega)$ being a δ function signifies that the band electron at momentum $\mathbf{k}$ has all its spectral weight precisely at the energy $\varepsilon_k=\gamma k$, i.e., the noninteracting particle exists entirely at the energy γk for a given momentum $\mathbf{k}$. In the presence of interaction effects, the many-body self-energy function $\Sigma(\mathbf{k},\omega)$ modifies the single-particle Green function, $G^{-1}(\mathbf{k},\omega)=G_0^{-1}(\mathbf{k},\omega)-\Sigma(\mathbf{k},\omega)$, and the corresponding interacting or renormalized spectral function is given by

$$A(\mathbf{k},\omega)\equiv\frac{2\,\mathrm{Im}\,\Sigma(\mathbf{k},\omega)}{[\omega-\xi_k-\mathrm{Re}\,\Sigma(\mathbf{k},\omega)]^2+[\mathrm{Im}\,\Sigma(\mathbf{k},\omega)]^2},\qquad(1)$$

where $\Sigma(\mathbf{k},\omega)=\mathrm{Re}\,\Sigma(\mathbf{k},\omega)+i\,\mathrm{Im}\,\Sigma(\mathbf{k},\omega)$ is complex, and $\xi_{sk}\equiv\varepsilon_{sk}-E_F$. In general, $A(\mathbf{k},\omega)$ could be a complicated function of $\mathbf{k}$ and ω, and there is no guarantee that it will have a δ-function peak defining a quasiparticle. We note that $\int(d\omega/2\pi)A(\mathbf{k},\omega)=1$ is a sum rule, guaranteeing that the electron at momentum $\mathbf{k}$ exists in the whole energy space, but it may exist completely incoherently spread out over the whole ω space without any coherent structure (i.e., a δ function at $k=k_F$), indicating a complete failure of the Fermi liquid picture. Thus, the Fermi liquid theory applies only when the renormalized spectral function $A(k=k_F,\omega)$ at the Fermi momentum has a δ-function peak, i.e., $A(k_F,\omega)$ can be written as $A(k_F,\omega)=2\pi Z\delta(w-\xi_k^*)+A_{\mathrm{in}}(\omega)$, where Z is the so-called renormalization factor, ξ_k^* denotes the renormailzed quasiparticle energy (measured from the chemical potential), and A_{in} is the incoherent background spectral function. If $A(k_F,\omega)$ does not have any δ-function peak at all, then the system is a non-Fermi liquid.

In Fig. 1 we show our calculated interacting quasiparticle spectral function for extrinsic graphene at a fixed carrier density $n=10^{12}$ cm^{-2}. The calculations are carried out at $T=0$,

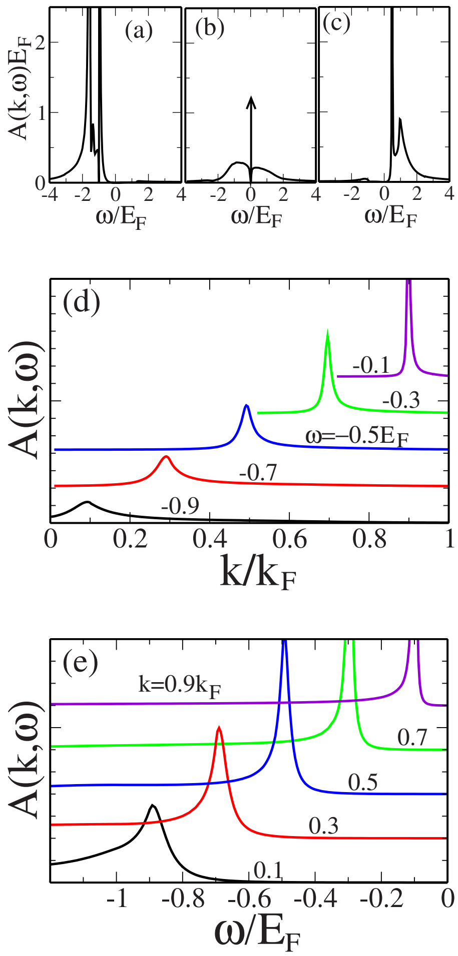

FIG. 1. (Color online) Calculated graphene spectral function for different wave vectors $k=$ (a) 0, (b) k_F, and (c) $1.5k_F$ without any disorder effects. (d) Spectral function as a function of wave vector for different energies (MDC) and (e) as a function of energy for different wave vectors (EDC). In (d) and (e) we include an impurity scattering rate of $0.5E_F$ as explained in Fig. 3(b). We show only the spectral features above the Dirac point. Here all energies are measured from the Fermi level, and the Dirac point is at $\omega=-E_F$.

but thermal effects are unimportant here since $T/T_F \ll 1$ even at room temperatures. The following salient features of graphene quasiparticle spectral function are notable in Fig. 1. (1) There is a well-defined δ-function quasiparticle peak at k_F with a rather substantial spectral weight of $Z \approx 0.85$, indicating that 15% of the bare spectral weight goes into incoherent background. (2) The quasiparticle spectral function for $k \neq k_F$ shows, in general, broadened peak structures indicating damped quasiparticles. (3) The generic broadened double-peak structure at $k \neq k_F$ indicates that, away from the Fermi surface, the renormalized graphene spectra would have two distinct energies—the second peak, which has been well studied in the literature in both 2D (Ref. 6) and 3D (Ref. 7) interacting electron systems, is often referred to as the "plasmaron" peak, indicating a coupled electron-plasmon composite excitation. (4) All spectral functions for $k \neq k_F$ have finite width corresponding to quasiparticle damping defined by the imaginary part of the many-body self-energy, $\mathrm{Im}\,\Sigma(k,\omega)$. Note that in Figs. 1(d) and 1(e) disorder contributes to combining the double-peak structure into a very broadened single peak.

Since Fig. 1 giving the interacting graphene quasiparticle spectral function is the central result being presented in this paper, we first discuss the importance and the implications of our calculated spectral function before describing the details of our theory and other results. First, extrinsic (i.e., gated or doped) graphene is a Fermi liquid with a well-defined undamped quasiparticle at k_F. Thus, the chiral Dirac-like linear dispersion of graphene band structure does not lead to any anomalous non-Fermi-liquid behavior. Second, extrinsic graphene has well-defined, but damped, quasiparticle peaks for all momenta. One direct experimental probe of the quasiparticle spectral function is tunneling spectroscopy, which has been studied extensively in GaAs-based 2D systems,[8] but has not yet been studied in graphene. The measurement that comes closest to studying $A(\mathbf{k},\omega)$ in graphene is ARPES.[4,5] Unfortunately, there are problems in directly comparing our theoretical spectral function of Fig. 1 with the experimental ARPES results. One problem is that electron-phonon interaction-induced many-body renormalization[9] also contributes to the graphene spectral function. In Figs. 1(d) and 1(e), we show the theoretical results corresponding to the experimental ARPES spectra—in particular, Figs. 1(d) and 1(e), respectively, correspond to the so-called momentum distribution curves (MDCs) and energy distribution curves (EDCs) of ARPES data. Since the actual theoretical spectral function Fig. 1 has very strong momentum (k) and energy (ω) dependence, a direct comparison with ARPES data will necessarily involve detailed instrumental issues involving resolution, the (k,ω) regime of averaging, and instrumental errors, which are all well beyond the scope of the current theoretical work. A cursory comparison between our results and the experimental ARPES data[4,5] shows reasonable qualitative agreement, but short of large-scale data fitting, one cannot make definitive quantitative statements. One important point to note here is that the plasmaron structure (i.e., the additional peak) does not really show up in the MDC and EDC spectra, since they carry small spectral weight compared with the main quasiparticle peak, particularly in the energy regime near E_F, as observed experimentally.[4] Note that the inclusion of disorder in the theory is essential in getting agreement with the measured spectra since the two-peak structure found in Figs. 1(a) and 1(c) without disorder is not experimentally observed.

We now describe the theory leading to our calculation of the interacting quasiparticle spectral function depicted in Fig. 1. The self-energy $\Sigma(\mathbf{k},\omega)$ defining the spectral function through Eq. (1) is given, in the leading order dynamically screened Coulomb interaction approximation[6,7]

$$\Sigma_s(\mathbf{k},i\omega_n) = -k_BT\sum_{s'}\sum_{\mathbf{q},i\nu_n} G_{0,s'}(\mathbf{k}+\mathbf{q},i\omega_n+i\nu_n) \times \frac{V_c(\mathbf{q})}{\epsilon(q,i\nu_n)}F_{ss'}(\mathbf{k},\mathbf{k}+\mathbf{q}), \qquad (2)$$

where $V_c(q)=2\pi e^2/\kappa q$ is the 2D Coulomb interaction with

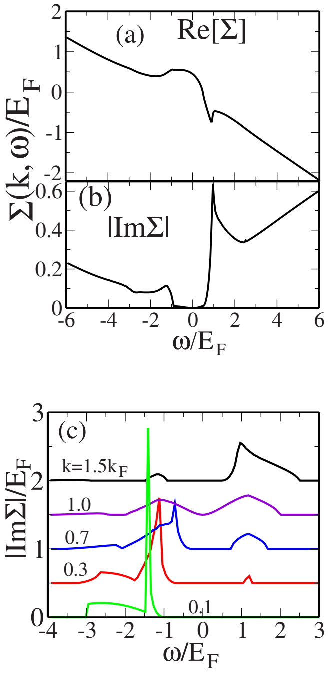

FIG. 2. (Color online) Calculated graphene correlation self energy for $k=1.5k_F$ as a function of energy: (a) Re Σ and (b) $|\mathrm{Im}\,\Sigma|$. (c) Plasmon contribution to the imaginary part of the self-energy for several wave vectors. The Dirac point is at $\omega=-E_F$.

the background lattice dielectric constant κ, $F_{ss'}(\mathbf{k},\mathbf{k}')=(1+ss'\cos\theta_{\mathbf{kk}'})/2$ is a sublattice overlap matrix element arising from the graphene band structure, and $\epsilon(q,\omega)=1+V_c(q)\Pi(q,\omega)$ is the dynamical random-phase approximation (RPA) dielectric function for graphene with the irreducible electron-hole polarizability $\Pi(q,\omega)$, which has recently been calculated.[10] After the standard procedure of analytical continuation, the self-energy can be separated into the exchange and correlation parts, $\Sigma_s(\mathbf{k},\omega)=\Sigma_s^{\mathrm{ex}}(\mathbf{k})+\Sigma_s^{\mathrm{cor}}(\mathbf{k},\omega)$.[6,7] Since $\Sigma_s^{\mathrm{ex}}(k)$ has earlier been discussed in detail,[11] we concentrate on the correlation part of the self-energy, Σ_s^{cor}. We note that the irreducible self-energy approximation used in our theory corresponds to keeping the Coulomb interaction and the infinite series of electron-hole bubble diagrams in the self-energy calculation.

An important technical point in the calculation of the self-energy in graphene is that two distinct types of field-theoretical divergence appear in the theory: infrared (small momentum) and ultraviolet (large momentum).[3] The infrared divergence arises from the $1/q$ long-range divergence of the Coulomb interaction, and is regularized by our RPA theory through screening.[6,7] The ultraviolet divergence in the graphene self-energy, which arises from the peculiar graphene band dispersion, is fixed by realizing that the linear Dirac dispersion of graphene applies only up to momenta of the order of the inverse lattice constant, and therefore all momentum integrals should have an upper cutoff $k_c\sim 1/a$, where a is the graphene lattice constant.

In Fig. 2 we show our calculated graphene self-energy for $n=10^{12}$ cm^{-2} at $k=1.5k_F$. We note that the peak in Im Σ correlates with the dip in Re Σ, and most of the spectral weight resides near the dip of Re Σ (or equivalently at the peak of Im Σ). We emphasize that, as is apparent from the detailed spectral function in Fig. 1, the various structures in the self-energy lead to specific features in $A(k,\omega)$—in particular, Re Σ and Im Σ control, respectively, the energy renormalization and the broadening (or damping) of the quasiparticle. An important issue in this context is the precise contribution of the collective plasmon mode to Im Σ, which we have evaluated explicitly as shown in Fig. 2(c). Only far away from the Fermi surface are the plasmon contributions strong, giving rise to the strong plasmaron peak in the spectral function.

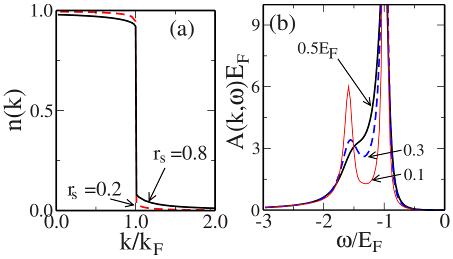

FIG. 3. (Color online) (a) Renormalized momentum distribution function as a function of momentum for $r_s=0.8$ (solid line) and 0.2 (dashed line). (b) Disorder effects on the $k=0$ spectral function for different impurity scattering rates of $(0.1,0.3,0.5)E_F$.

In Fig. 3 we show our calculated renormalized quasiparticle momentum distribution function $n(k)=\int_{-\infty}^{E_F}(d\omega/2\pi)A(k,\omega)$ for extrinsic graphene and disorder effects on the spectral function. The effect of electron-electron interaction on $n(k)$ is rather obvious in Fig. 3(a). We note that the interacting momentum distribution function has a discontinuity of relative size Z at $k=k_F$, clearly establishing the Fermi liquid behavior of extrinsic (i.e., doped or gated) graphene. The discontinuity increases as r_s decreases. This discontinuity may not be easy to directly observe experimentally, since thermal and disorder effects will broaden it.

Impurity effects are usually introduced diagrammatically into the RPA screening by including ladder impurity diagrams into the electron-hole bubble. We use a particle-conserving expression,[12] which captures the essential physics of impurity scattering effects on the electron polarizability Π. We find that disorder strongly suppresses the plasmaron peak, but not the quasiparticle peak. Thus the double-peak structure in the spectral function becomes a very broadened single peak. In the strongly disordered experimental graphene samples, therefore, we have only a broadened quasiparticle peak.

We now comment on the experimental implications, the theoretical approximations, and the connections to earlier work. As mentioned before, the graphene ARPES measurements have information on the electron spectral function, which are, however, complicated by the presence of additional interaction effects such as electron-phonon interactions.[9] If these additional interaction effects can be subtracted out (e.g., the ~200 meV structure[4] presumably arising from phonons in the experimental data), then ARPES measurements could indeed be compared with our calculated spectral function shown in Fig. 1. We note that, in agreement with the ARPES data, the quasiparticle spectral peak becomes wider due to the plasmon contribution as the energy approaches the Dirac point ($\omega=-E_F$). Including disorder effects, indeed there is good agreement between theory and experiment. In experimental ARPES data, one broadened spectral peak can be explained by the broadened double-peak spectral features (the quasiparticle peak and the plasmaron peak) due to disorder effects. The observed 200 meV structure in the ARPES arises from phonons, as has recently been explained.[9]

Our theory is based on the leading order expansion of the electron self-energy using the dynamically screened Coulomb interaction (the so-called infinite bubble diagram expansion, with each bubble being the noninteracting electron-hole polarizability[10]), which we believe to be a quantitatively accurate approximation for extrinsic graphene (i.e., at any finite carrier density) by virtue of graphene having a reasonably small (and density independent) dimensionless interaction parameter $r_s = e^2/\kappa\gamma \sim 0.8$ (0.4) for graphene on a SiO_2 (SiC) substrate. In fact, the same self-energy approximation, often referred to in the literature as the RPA or *GW* approximation, is known to work well in 3D alkali metals where $r_s \sim 3-6$,[7] and in 2D semiconductor system[6,8] where $r_s \sim 1-10$. The validity of the RPA and Fermi liquid theory in extrinsic graphene has recently been discussed in the literature.[3]

In discussing the connection between our work and earlier work, we mention that, although there has been a great deal of recent theoretical work on interaction effects in graphene, much of it has focused on intrinsic graphene,[13] and most of the work on extrinsic graphene has focused on thermodynamic properties.[14] A recent calculation of the quasiparticle spectral function in graphene due to electron-phonon interaction has appeared[9] in the literature, and within the weak-coupling theory (i.e., both electron-electron and electron-phonon interactions are weak), the total spectral function should be a sum of these two spectral functions. We do note that our calculated spectral function and self-energy in extrinsic graphene are qualitatively rather similar to those in ordinary parabolic band 2D carrier systems,[6] thus pointing to the fact that, in spite of its chiral Dirac-like band dispersion, doped graphene in the presence of free carriers is qualitatively similar to doped 2D semiconductor systems.

In summary, we have provided a theoretical calculation for the electron spectral function in 2D doped (i.e., extrinsic) graphene, finding our theory to be in good qualitative agreement with the available experimental data. We show that our calculated electron-electron interaction effects when combined with the corresponding electron-phonon and electron-impurity interaction effects provide a quantitative description for extrinsic graphene ARPES measurements. We believe that our work plus the recent phonon work[9] together now provide a complete theoretical understanding of the quasiparticle properties of extrinsic graphene with no remaining open conceptual questions. We would also like to note that we recently became aware of related work.[15]

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