Unconventional quasiparticle lifetime in undoped graphene

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(Dated: March 6, 2008)

We address the question of how small can the quasiparticle decay rate be at low energies in undoped graphene, where kinematical constraints are known to prevent the decay into particle-hole excitations. For this purpose, we study the renormalization of the phonon dispersion by many-body effects, which turns out to be very strong in the case of the out-of-plane phonons at the $K$ point of the spectrum. We show that these evolve into a branch of very soft modes that provide the relevant channel for quasiparticle decay, at energies below the scale of the optical phonon modes. In this regime, we find that the decay rate is proportional to the cube of the quasiparticle energy. This implies that a crossover should be observed in transport properties from the linear dependence characteristic of the high-energy regime to the much slower decay rate due to the soft phonon modes.

The recent fabrication of single atomic layers of carbon has attracted a lot of attention, as this material (so-called graphene) provides the experimental realization of a system where the low-energy electronic excitations behave as massless Dirac fermions\textsuperscript{[1, 2]}. The main results reported in Refs. [1 and 2] can be understood as a reflection of the linear dependence on momentum of the quasiparticle energy $\varepsilon (\mathbf{k})$. The honeycomb lattice structure of graphene is actually known to lead to a bandstructure with conical shape $\varepsilon (\mathbf{k}) = \pm v_F | \mathbf{k} |$ around the corners of the hexagonal Brillouin zone, with conduction and valence bands meeting at the Dirac points. The relativistic-like invariance arising from the massless Dirac quasiparticles has been shown to be at the origin of a number of remarkable electronic properties, like the finite lower bound of the conductivity at the charge neutrality point\textsuperscript{[3, 4, 5, 6]}, the anomalous integer Hall effect\textsuperscript{[3, 7, 8]}, and the absence of backscattering in the presence of long-range scatterers\textsuperscript{[9]}. More recently, the many-body properties of the graphene layer have been also investigated. One of the relevant issues addressed is whether the quasiparticle properties have to correspond to the expected behavior for a Fermi liquid in two dimensions\textsuperscript{[10, 11]}. In this regard, it has been pointed out that the $e-e$ interactions lead to quite different quasiparticle features in graphene depending on whether the material is doped or not\textsuperscript{[11]}. This can be understood from the particular kinematical constraints of the conical dispersion, that prevent the Dirac quasiparticles from decaying into interband particle-hole excitations\textsuperscript{[12]}. In doped graphene, intraband processes are responsible for the quasiparticle decay, leading to a quadratic dependence on energy of the decay rate\textsuperscript{[11]}. On the other hand, when the Fermi level is at the charge neutrality point, the electron self-energy has a linear dependence on frequency. However, the maximum energy released in the scattering of a quasiparticle with momentum transfer $\mathbf{q}$ is at the boundary of the continuum of particle-hole excitations, which have energy $\geq v_F | \mathbf{q} |$. In situations where the Coulomb interaction remains singular in the limit $\mathbf{q} \to 0$, as it happens in the layers of bulk graphite, a finite spread in the momentum of the quasiparticles is enough to give rise to a finite decay rate\textsuperscript{[12]}. This mechanism must rely however on some effect extrinsic to the 2D system (disorder, for instance), and it would be absent anyhow as soon as the Coulomb interaction is screened beyond a certain distance. In this paper we address the question of whether the quasiparticle decay rate may actually vanish in a graphene layer with the Fermi level tuned at the charge neutrality point. This study is relevant as it faces the possibility of having an electron liquid made of extremely long-lived quasiparticles. Thus, we will look for many-body effects which may give rise to suitable gapless excitations and consequent quasiparticle decay channels in undoped graphene.

It is known that, in the absence of doping, the 2D system does not support plasmon excitations\textsuperscript{[13]}. Yet the polarization of the electron liquid is singular at low energies, and this may be the source of potential instabilities. We will see that there is actually a significant renormalization of the interactions at the large momentum transfer $K$ connecting the two inequivalent Dirac points in graphene. At such large momentum, the singular behavior of the electron polarization tends to amplify the effects of the electron-phonon interaction, which prevails over the Coulomb interaction. Thus, we will show that gapless phonon branches may appear at the $K$ point when graphene is lying on a substrate. The resulting low-energy phonon modes provide then the relevant mechanism for the decay of quasiparticles in undoped graphene, though with a very low decay rate that turns out to be proportional to the cube of the quasiparticle energy.

We begin by considering the Hamiltonian for Dirac quasiparticles in graphene, at energies below the scale of $\sim 1$ eV for which the dispersion can be taken as linear:

$$H_0 = v_F \int d^2k \Psi^{(a)\dagger}(\mathbf{k}) \gamma^{(a)} \cdot \mathbf{k} \Psi^{(a)}(\mathbf{k})$$

(1)

In the above expression, a sum is implicit over the index $a$ accounting for the two different valleys and corresponding Dirac spinors $\Psi^{(a)}$ at opposite corners $K, -K$ in the graphene Brillouin zone. $\gamma^{(a)}$ are different sets of Pauli
matrices for \( a = 1, 2 \), which must be chosen according to the appropriate chirality of the modes at \( K, -K \) as
\[ \gamma^{(1)} \equiv (\sigma_x, \sigma_y), \gamma^{(2)} \equiv (-\sigma_x, \sigma_y) \] 2. As a first step in the development of the many-body theory, we will assume that the quasiparticles interact through a Coulomb potential
\[ V_0(\mathbf{q}) = \frac{e^2}{2\kappa |\mathbf{q}|} \] (2)
with a dielectric constant \( \kappa \) dictated by the coupling to the substrate.

As is well-known, the quasiparticles of the 2D system provide very limited screening of the Coulomb potential in (2). This effect can be assessed by computing the polarization
\[ \Pi_0^{(a,b)}(\mathbf{q}, i\omega) = 4 \text{Tr} \int \frac{d^2 k}{(2\pi)^2} \int \frac{d\omega_k}{2\pi} G^{(a)}(\mathbf{k} + \mathbf{q}, i\omega_k + i\omega) G^{(b)}(\mathbf{k}, i\omega_k) \] with Dirac propagators \( G^{(a)}(\mathbf{k}, i\omega_k) = 1/(\pm i\omega - v_F \gamma^{(a)} \cdot \mathbf{k}) \). At small momentum-transfer, the trace in (3) is taken over excitations in the same valley \( a = b \), with the result that
\[ \text{Tr}(\pm i\omega + v_F \gamma^{(a)} \cdot \mathbf{q})(\pm i\omega + v_F \gamma^{(a)} \cdot \mathbf{k}) = -2\omega \sigma \mp 2v_F^2 \mathbf{q} \cdot \mathbf{k} \]. This leads to an expression for the intravalley polarization \( \Pi_0^{(a,a)}(\mathbf{q}, i\omega) = -\omega^2/8 \sqrt{v_F^2 \mathbf{q}^2 + \omega^2} \). Going back to real frequency \( \omega_q = \sqrt{\omega^2} \), we find a divergence of the polarization at \( \omega_q = v_F |\mathbf{q}| \). This marks actually the threshold for the creation of particle-hole pairs in the electron liquid. The particle-hole continuum is above the maximum energy \( v_F |\mathbf{q}| \) released in the scattering of a quasiparticle with momentum transfer \( \mathbf{q} \). This explains that the quasiparticle decay into particle-hole pairs is forbidden in the case of undoped graphene.

In the case of intervalley scattering of quasiparticles, the polarization is also affected by a similar divergence at \( \omega_q = v_F |\mathbf{q}| \), where \( \mathbf{q} \) stands now for a small deviation around the large momentum \( K \). The computation of the polarization (3) with \( a \neq b \) leads to the trace
\[ \text{Tr}(\pm i\omega + v_F \gamma^{(a)} \cdot \mathbf{q})(\pm i\omega + v_F \gamma^{(b)} \cdot \mathbf{k}) = -2\omega \sigma K - 2v_F^2 q_x k_x + 2v_F^2 q_y k_y \]. This can be assimilated to the above computation for \( a = b \) if the \( y \) component of each momentum is exchanged with the frequency \( \omega \), and an overall sign is introduced. It can be checked by direct calculation that the result for the intervalley polarization corresponds actually to operating that transformation in the above expression for \( \Pi_0^{(a,a)} \), that is,
\[ \Pi_0^{(1,2)}(\mathbf{q}, i\omega) = (p_F^2 + \omega^2/v_F^2)/8 \sqrt{v_F^2 \mathbf{q}^2 + \omega^2} \]. The polarization thus obtained shows a preferred direction in momentum space, which is a reflection of having considered the scattering between two Dirac valleys along the \( x \) direction. The result physically sensible can be obtained by averaging over the processes involving the three equivalent nearest-neighbor valleys of the \( K \) point. These include in particular the valleys rotated by an angle of \( \pm 2\pi/3 \) with respect to the \( x \)-axis. Taking into account the three different contributions, we get the final result for the intervalley polarization
\[ \Pi_0(\mathbf{q}, \omega) = \frac{q^2/2 - \omega^2/v_F^2}{8 \sqrt{v_F^2 \mathbf{q}^2 + \omega^2}} \] (4)

The polarization (4) does not give rise to any singularity when renormalizing the Coulomb interaction, as the Coulomb potential gets dressed at large momentum-transfer \( K \) in the form \( V_0(\mathbf{q}) \approx e^2/(2\kappa K - \Pi_0(\mathbf{q}, \omega)) \). On the contrary, the singular behavior of the intervalley polarization may lead to important effects in the phonon sector. This consideration is relevant for lattice vibrations coupling to the total electron charge, as it happens in the case of the out-of-plane phonons. The electron-phonon interaction can be analyzed in terms of the atomic deformation potential induced by the lattice vibrations 12. When graphene is lying on a substrate, the mirror symmetry of the vibrations perpendicular to the carbon layer is broken, and the on-site deformation potential induces a linear coupling of the out-of-plane phonons to the total electron charge. If we denote by \( \epsilon_q^0, \epsilon_q^+ \) the creation and annihilation operators for the modes around the \( K \) point of a branch of out-of-plane phonons, we can describe the phonon sector by means of kinetic and interaction terms in the hamiltonian:
\[ H_{\text{ph}} = \int \frac{d^2 q}{2\pi} \omega_q(\mathbf{q}) \epsilon_q^\dagger(\mathbf{q}) \epsilon_q \]
\[ H_{\text{e-ph}} = g \int \frac{d^2 k d^2 q}{(2\pi)^2} \Psi^{(a)}(\mathbf{k})(\mathbf{q}) \Psi^{(b)}(\mathbf{k}) (\epsilon_q^0 + \epsilon_q^+) \] (5)

For the characterization of the phonon branch, it will be enough to approximate the energy of the out-of-plane phonons about the \( K \) point by \( \omega_q \approx 70 \text{ meV} \). The electron-phonon coupling \( g \) can be obtained as the atomic deformation potential (of the order of a few eV) times 1/\( \sqrt{m_C a_0} \) (\( m_C \) being the carbon atomic mass) 13.

The intervalley polarization induces a strong renormalization of the out-of-plane phonons in graphene. It has been already pointed out that the interaction with the electronic degrees of freedom may give rise to significant Kohn anomalies in the dispersion of in-plane optical phonons 10,17. The coupling to these branches is given in general by the modulation of the transfer integral between nearest-neighbor atoms in the carbon lattice. In our framework, this gives rise to an electron-phonon vertex proportional to the matrix \( \sigma_x \). When introduced in the computation of the trace in the polarization, such a vertex gives rise to simple scalar products in \( (\mathbf{q}, \omega) \) space, leading to a susceptibility proportional to \( \sqrt{v_F^2 \mathbf{q}^2 + \omega^2} \). However, in the case of phonons coupling to the total electron charge, the particle-hole polarization (4) induces a more profound anomaly in the phonon dispersion. If we approximate the bare phonon propagator about the \( K \) point by \( D_0(\mathbf{q}, \omega) \approx 2\omega_0/(\omega^2 - \omega_0^2 + i\epsilon) \), the renormalized propagator \( D(\mathbf{q}, \omega) \) dressed with the
FIG. 1: Plot of the region corresponding to the particle-hole continuum (shaded area) and the phonon dispersion arising from the solution of Eq. (7).

Particle-hole polarization becomes

$$D(q, \omega) \approx \frac{2\omega_0}{\omega^2 - \omega_0^2 + i\epsilon - 2\omega_0 g^2 \tilde{\Pi}_0(q, \omega)}$$

(6)

The renormalized phonon energies are found by setting to zero the denominator of the propagator (6), which leads to the equation

$$\omega^2 - \omega_0^2 - \frac{g^2}{v_F^2} \omega_0 v_F^2 q^2 / 2 - \omega^2 = 0$$

(7)

It can be checked that the phonon dispersion thus computed becomes gapless at \( q = 0 \), adopting the form of a low-energy branch below the continuum of particle-hole excitations as shown in Fig. 1. At this point, it becomes pertinent however to assess the effects of the Coulomb interaction on the renormalization of the phonon properties.

The Coulomb and the phonon-mediated interaction have to be considered on the same footing when analyzing their role in the renormalization of the phonon propagator. Thus, we can define an intervalley particle-hole susceptibility \( \tilde{\Pi}(q, \omega) \) dressed by the effect of the Coulomb interaction and, therefore, satisfying the equation

$$\tilde{\Pi} = \tilde{\Pi}_0 + \tilde{\Pi}_0 V_0 \tilde{\Pi}$$

(8)

In terms of this susceptibility, the renormalized phonon propagator \( D(q, \omega) \) can be obtained as

$$D^{-1} = D_0^{-1} - g^2 \tilde{\Pi}$$

(9)

Combining the solution of (8) with (9), we arrive at the final expression

$$D(q, \omega) = \frac{2\omega_0 (1 - \frac{\omega^2}{\omega_0^2} \tilde{\Pi}_0(q, \omega))}{\omega^2 - \omega_0^2 + i\epsilon - ((\omega^2 - \omega_0^2) v_F^2 \pi^2 K + 2\omega_0 g^2) \tilde{\Pi}_0(q, \omega)}$$

(10)

The strength of the phonon-mediated interaction is given by the dimensionless coupling \( g^2 / v_F^2 \), which can be estimated as \( \sim 0.1 \). This is smaller than the strength of the Coulomb interaction in typical graphene samples, where \( e^2 / 4\pi \hbar v_F \sim 1 \). However, the latter enters in the denominator of \( \tilde{\Pi}_0 \) with a relative weight \( \omega_0 / 4\pi \hbar v_F K \), which is of the order of \( \sim 0.001 \). Therefore, we see that the Coulomb interaction is not able to balance the effect of the strong renormalization of the phonon propagator at the large momentum transfer \( K \).

We find then that, when graphene is tuned at the charge neutrality point, there are phonon modes with very low energy around the \( K \) point, arising as a consequence of the strong renormalization from the coupling to particle-hole excitations. Strictly speaking, the phonon branches become gapless only at zero temperature in the undoped system. Away from the charge neutrality point and at finite temperature, the divergence of the polarization in (6) will be cut off by either the thermal energy or the effective chemical potential of the system. Above such infrared scales, the phonon dispersion will follow anyhow the trend represented in Fig. 1. The phonon energy \( \omega_{ph}(q) \) obtained from the renormalized propagator is actually given by

$$\omega_{ph}(q) \approx v_F |q| \left( \frac{g^2}{8v_F^2} \right)^2 \frac{\pi^2}{2\omega_0^2} q^2 + \ldots$$

(11)

We stress that this phonon branch lies in any event away from the continuum of particle-hole excitations, opening the possibility to observe well-defined phonon modes of very low energy at the \( K \) point of graphene.

The existence of the soft phonon branch leads to a channel for the decay of quasiparticles in undoped graphene. The maximum energy that can be released by a quasiparticle in a scattering process at any low momentum-transfer is enough to hit the phonon branch (11), so that electron quasiparticles can decay into this type of phonon modes down to arbitrarily low energies (at zero temperature). The quasiparticle decay rate \( \tau^{-1} \) can be computed from the electron self-energy \( \Sigma^{(a)}(k, \omega_k) \) as

$$\tau^{-1} = -\text{Im} \Sigma^{(a)}(k, v_F |k|)$$

$$\approx \text{Im} g^2 \int \frac{d^2 q}{(2\pi)^2} \frac{d\omega_q}{2\pi} G^{(b)}(k - q, v_F |k| - \omega_q) D(q, \omega_q)$$

(12)

which amounts to making the convolution of the imaginary part of the electron propagator with that of \( D(q, \omega_q) \).

In Eq. (12), the imaginary part of \( G^{(b)} \) enforces the constraint \( \omega_q = v_F |k| - v_F |k - q| \). For that frequency, the phonon propagator picks up an imaginary contribution only from the phonon branch (11). We have actually

$$\tau^{-1} \approx \frac{\pi g^2}{2} \int \frac{d^2 q}{(2\pi)^2} \delta(Q(q, \Omega_q))$$

(13)
where $\Omega_q \equiv v_F|k| - v_F|k - q|$ and

$$Q(q, \Omega_q) = \frac{\Omega_q^2 - \omega_0^2}{2\omega_0} - \frac{g^2 v_F^2 q^2 / 2 - \Omega_q^2}{v_F 8 \sqrt{v_F^2 q^2 - \Omega_q^2}}$$

(14)

The integral over $q$ can be done by trading the azimuthal variable of integration $\phi$ by $\Omega_q$. Thus we get

$$\tau^{-1} \approx \frac{1}{2\pi} g^2 \int_0^{2\pi} dq |q| \int_0^{\Omega_q} d\Omega_q$$

$$\left| \frac{\partial \phi}{\partial \Omega_q} \right| \left| \frac{\partial Q}{\partial \Omega_q} \right|^{-1} \delta(\Omega_q - \omega_{ph}(q))$$

(15)

The expression (15) leads to different behaviors depending on whether the quasiparticle energy is well above or below the scale $\omega_0$. In the range where $v_F|k| \gg \omega_0$, it is easy to see that the Jacobian $|\partial \phi/\partial \Omega_q|$ scales as $|k|/|q| \sqrt{4k^2 - q^2}$, while $|\partial Q/\partial \Omega_q|^{-1}$ does not scale with momentum. The quasiparticle decay rate shows then a linear dependence on energy $\tau^{-1} \sim (g^2/v_F^2) v_F|k|$, in agreement with previous analyses of the decay due to optical phonons [15]. On the other hand, when the quasiparticle energy is below $\omega_0$, we find that $|\partial \phi/\partial \Omega_q|$ scales as $\sim \omega_0 \sqrt{|k| - |q|}/\sqrt{|k|v_F^2 q^2}$. Moreover, we also have $|\partial Q/\partial \Omega_q|^{-1} \sim v_F^2 |q|^3/\omega_0^3$. We get then a decay rate

$$\tau^{-1} \approx \frac{1}{16\pi v_F^2} \frac{g^4 |k|^3}{\omega_0^3} \int_0^{1} dx x^2 \sqrt{1 - x}$$

(16)

We arrive at the result that, in the case of undoped graphene, the quasiparticle decay rate cannot vanish at low energies, even below the scale $\omega_0$ of the out-of-plane phonons, where it must be proportional to the cube of the quasiparticle energy.

The behavior (16) differs significantly from the rate obtained for a long-range Coulomb interaction, which is proportional to the quasiparticle energy [11, 13]. We remark that the two behaviors correspond actually to quite different conditions. When the Coulomb interaction remains long-ranged, as in the layers of bulk graphite, the singular character of the potential [2] leads to a jump in the electron self-energy at $\omega_k = v_F|k|$. A spread in momentum of the quasiparticles (induced for instance by disorder) may be invoked to obtain a finite quasiparticle decay rate by taking the limit $\omega_k \to v_F|k| + 0^+$. We have to bear in mind however that, in graphene, the divergence of the Coulomb potential may be cut off by some finite screening length $l$. If we assume a potential of the form $V_0(q) = e^2/\sqrt{q^2 + l^2}$, we can estimate the quasiparticle decay rate to lowest order in the Coulomb interaction as

$$\tau^{-1} \approx \lim_{\epsilon \to 0} e^4 \int_0^{2\pi} dq |q| \int_0^{\Omega_q + \epsilon} d\Omega_q$$

$$\frac{q^2}{\sqrt{|k| - |q|} \sqrt{\epsilon - (\Omega_q - v_F|q|)}} (q^2 + l^2) \sqrt{\Omega_q^2 - v_F^2 q^2}$$

(17)

We see that the decay rate in this approach turns out to be proportional to $(e^2/v_F^2) v_F l^2 |k|^3$. It is only after sending the screening length to infinity that the divergence of the Coulomb potential at $q = 0$ is able to change the scaling of the integrand in (17), leading to a decay rate proportional to the quasiparticle energy.

In conclusion, we have shown that the dispersion of phonons that couple to the total electron density undergoes a strong renormalization when graphene is tuned to the charge neutrality point. This reflects in the behavior of the out-of-plane phonons, which turn out to develop a gapless branch at the $K$ point of the spectrum. In situations where graphene is very lightly doped, the mentioned renormalization will still give rise to a branch of very soft phonons, with a small gap proportional to the effective chemical potential (as measured from the charge neutrality point). We have seen that this branch provides the relevant channel for the decay of quasiparticles below the typical scale of the out-of-plane phonons, $\omega_0 \approx 70$ meV. Thus, even at such low energies, the quasiparticle decay rate does not vanish in undoped graphene, though it becomes very suppressed, with a dependence proportional to the cube of the quasiparticle energy. The present analysis may then be useful to interpret the results of transport experiments in undoped or very lightly doped graphene, where a crossover should be observed from the linear decay rate characteristic of the high-energy regime to the much slower cubic dependence found in the paper.

Acknowledgments

We thank F. Guinea and F. Sol as for very fruitful discussions. The financial support of the Ministerio de Educación y Ciencia (Spain) through grant FIS2005-05478-C02-02 is gratefully acknowledged. E.P. is also financially supported by CNISM and Fondazione Cariplo-Prot.0018524.

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