Unconventional Quasiparticle Lifetime in Graphene

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We address the question of how large can the lifetime of electronic states be at low energies in graphene, below the scale of the optical phonon modes. For this purpose, we study the many-body effects at the K point of the spectrum, which induce a strong coupling between electron-hole pairs and out-of-plane phonons. We show the existence of a soft branch of hybrid states below the electron-hole continuum when graphene is close to the charge neutrality point, leading to an inverse lifetime proportional to the cube of the quasiparticle energy. This implies that a crossover should be observed in transport properties, from such a slow decay rate to the lower bound given at very low energies by the decay into acoustic phonons.

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The recent fabrication of single atomic layers of carbon has attracted a lot of attention, as this material (so-called graphene) provides the realization of a system where the electrons have conical valence and conduction bands, therefore behaving as massless Dirac fermions [1,2]. The consequent relativisticlike invariance 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 (CNP) [3–6], the anomalous integer Hall effect [3,7,8], and the absence of backscattering in the presence of long-range scatterers [9].

More recently, the many-body properties of graphene have been also investigated. The analyses have mainly focused on the behavior of the quasiparticles (QPs) arising from Coulomb scattering [10,11]. The undoped system has isolated Fermi points which coincide with the vertices of the conical dispersion (known as Dirac points), leading to kinematical constraints that prevent the QPs from decaying into electron-hole (e-h) pairs [12,13]. The maximum energy released in the scattering of a QP with momentum transfer $\mathbf{q}$ is at the boundary of the e-h continuum, which has energy $\geq v_F|\mathbf{q}|$, in terms of the Fermi velocity v_F of the system. 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 QP is enough to give rise to a finite decay rate, which becomes linear in the QP energy [12]. This mechanism requires, however, the presence of disorder or some other effect extrinsic to the electron system. Moreover, as soon as the Coulomb interaction is screened beyond some length l, the decay rate becomes proportional to l^2, producing the crossover to a cubic dependence on the QP energy.

In this Letter we address the question of how large can the lifetime of electronic states be when graphene is close to the CNP. The scattering of QPs by acoustic phonons provides an upper bound to the QP lifetime at very low energies. This channel leads to a decay rate proportional to the square of the QP energy, with a prefactor given by the ratio between the speed of sound and the Fermi velocity. There are however experimental results pointing at the existence of other sources of QP scattering at low energies, which do not follow the expected trend from the acoustic phonons [14]. Thus, it becomes pertinent to investigate the low-energy excitations and decay channels arising below the scale of the in-plane optical phonons. We will see that there is actually a strong coupling between e-h pairs and out-of-plane phonons at the momentum transfer connecting the two inequivalent Dirac points in graphene. This effect turns out to give rise to a branch of hybrid states with energy below the e-h continuum. We will show that the resulting soft modes provide then the relevant channel for the scattering of QPs, leading to a low decay rate proportional to the cube of the QP energy below the scale of the out-of-plane phonons ($\approx$ 70 meV).

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

$$H_0 = v_F \int d^2k \Psi^{(a)\dagger}(\mathbf{k}) \boldsymbol{\gamma}^{(a)} \cdot \mathbf{k} \Psi^{(a)}(\mathbf{k}). \quad (1)$$

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. $\boldsymbol{\gamma}^{(a)}$ 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 $\boldsymbol{\gamma}^{(1)} \equiv (\sigma_x, \sigma_y)$, $\boldsymbol{\gamma}^{(2)} \equiv (-\sigma_x, \sigma_y)$ [7].

The many-body effects can be analyzed by computing the polarizations

$$\Pi_0^{(a,b)}(\mathbf{q}, i\bar{\omega}_q) = 4\mathrm{Tr} \int \frac{d^2k}{(2\pi)^2} \int \frac{d\bar{\omega}_k}{2\pi} \times G^{(a)}(\mathbf{k}+\mathbf{q}, i\bar{\omega}_k + i\bar{\omega}_q) G^{(b)}(\mathbf{k}, i\bar{\omega}_k) \quad (2)$$

with propagators $G^{(a)}(\mathbf{k}, i\bar{\omega}_k) = 1/(i\bar{\omega}_k - v_F\boldsymbol{\gamma}^{(a)} \cdot \mathbf{k})$. At small momentum transfer, the trace in (2) is taken over excitations in the same valley $a = b$, with the result that $\mathrm{Tr}(i\bar{\omega}_q + v_F \boldsymbol{\gamma}^{(a)} \cdot \mathbf{q})(i\bar{\omega}_k + v_F \boldsymbol{\gamma}^{(a)} \cdot \mathbf{k}) = -2\bar{\omega}_q \bar{\omega}_k + 2v_F^2 \mathbf{q} \cdot \mathbf{k}$. This leads to an expression for the intravalley polarization $\Pi_0^{(a,a)}(\mathbf{q}, i\bar{\omega}_q) = -\mathbf{q}^2/8\sqrt{v_F^2\mathbf{q}^2 + \bar{\omega}_q^2}$ [15]. Going back to real frequency $\omega_q = i\bar{\omega}_q$, we find a divergence of the polarization at $\omega_q = v_F|\mathbf{q}|$, which marks the threshold for the creation of *e-h* pairs.

In the case of intervalley scattering of QPs, 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 (2) with $a \neq b$ leads to the trace $\mathrm{Tr}(i\bar{\omega}_q + v_F \boldsymbol{\gamma}^{(a)} \cdot \mathbf{q}) \times (i\bar{\omega}_k + v_F \boldsymbol{\gamma}^{(b)} \cdot \mathbf{k}) = -2\bar{\omega}_q\bar{\omega}_k - 2v_F^2 q_x k_x + 2v_F^2 q_y k_y$. This can be assimilated to the above computation for $a = b$ if the y component of each momentum is exchanged with the frequency $\bar{\omega}$, and an overall $-$ sign is introduced. The result that we obtain is $\Pi_0^{(1,2)}(\mathbf{q}, i\bar{\omega}_q) = (q_x^2 + \bar{\omega}_q^2/v_F^2)/8\sqrt{v_F^2\mathbf{q}^2 + \bar{\omega}_q^2}$. This polarization 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 three equivalent nearest-neighbor valleys of the K point, which leads to the intervalley polarization

$$\tilde{\Pi}_0(\mathbf{q}, \omega_q) = \frac{\mathbf{q}^2/2 - \omega_q^2/v_F^2}{8\sqrt{v_F^2\mathbf{q}^2 - \omega_q^2}}. \tag{3}$$

The polarization (3) may lead to important effects when phonons are taken into account. In this respect, the relevant lattice vibrations are those coupling to the total electron charge, which singles out the instance of the out-of-plane phonons. 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 electron charge [16]. If we denote by $b_\mathbf{q}^\dagger (b_\mathbf{q})$ the creation (annihilation) operators for out-of-plane phonon modes around the K point, we can describe the coupling by adding a term to the Hamiltonian

$$H_{e\text{-ph}} = g \int d^2k d^2q \Psi^{(a)\dagger}(\mathbf{k} + \mathbf{q})\Psi^{(b)}(\mathbf{k})(b_\mathbf{q} + b_{-\mathbf{q}}^\dagger). \tag{4}$$

For simplicity, we will approximate the energy of the out-of-plane phonons about the K point by a constant value $\omega_0 \approx 70$ meV. Then, the electron-phonon coupling g can be obtained as the atomic deformation potential (of the order of several eV) times $1/\sqrt{m_C\omega_0}$ (m_C being the carbon atomic mass) [17]. The strength of the phonon-mediated interaction will be given by the dimensionless coupling g^2/v_F^2, which can be estimated as ≈ 0.3 (for a deformation potential $D \approx 10$ eV).

The coupling of out-of-plane phonons and *e-h* pairs in (4) implies that none of them correspond to eigenstates of the total Hamiltonian. To obtain the propagation of the states with well-defined energy, we must diagonalize the quadratic form in the action of the system

$$(\rho_\mathbf{q} \phi_\mathbf{q}) \begin{pmatrix} \frac{\omega_0}{v_F^2}\tilde{\Pi}_0^{-1}(\mathbf{q}, \omega) & \frac{g}{v_F} \\ \frac{g}{v_F} & \frac{1}{\omega_0}D_0^{-1}(\omega) \end{pmatrix} \begin{pmatrix} \rho_\mathbf{q} \\ \phi_\mathbf{q} \end{pmatrix}, \tag{5}$$

where $\rho_\mathbf{q} \equiv (v_F/\sqrt{\omega_0}) \int d^2k \Psi^{(a)\dagger}(\mathbf{k} + \mathbf{q})\Psi^{(b)}(\mathbf{k})$, $\phi_\mathbf{q} \equiv \sqrt{\omega_0}(b_\mathbf{q} + b_{-\mathbf{q}}^\dagger)$, and $D_0(\omega) \approx 2\omega_0/(\omega^2 - \omega_0^2 + i\epsilon)$ represents the bare propagator of the optical phonons. The eigenvalues of the quadratic form are

$$D_\pm^{-1}(\mathbf{q}, \omega) = \frac{1}{2}\left(\omega_0^{-1}D_0^{-1}(\omega) + \frac{\omega_0}{v_F^2}\tilde{\Pi}_0^{-1}(\mathbf{q}, \omega) \pm \sqrt{\left(\omega_0^{-1}D_0^{-1}(\omega) - \frac{\omega_0}{v_F^2}\tilde{\Pi}_0^{-1}(\mathbf{q}, \omega)\right)^2 + 4\frac{g^2}{v_F^2}}\right). \tag{6}$$

D_+ and D_- correspond to the propagation of hybrid states made of a phonon and an *e-h* pair. At large values of $|\mathbf{q}|$, the only pole in ω is found in $D_+(\mathbf{q}, \omega)$, which becomes close to the phonon propagator $D_0(\omega)$ (times ω_0). At small $|\mathbf{q}|$, however, there is a real pole only for $D_-(\mathbf{q}, \omega)$, which can be approximated by $(v_F^2/\omega_0)\tilde{\Pi}_0(\mathbf{q}, \omega)$ up to a slight correction. The dispersion of the states with well-defined energy can be obtained from the position of the pole as a function of $|\mathbf{q}|$, which interpolates between the linear edge of the *e-h* continuum at $|\mathbf{q}| \to 0$ and the almost dispersionless branch at large $|\mathbf{q}|$, as represented in Fig. 1. We observe that, in the hybrid states made of a phonon and an *e-h* pair, the former enters with a relative weight given by $(g\omega_0 D_0(\omega)/v_F)^2$ at the value of the pole (in D_+ or D_-), as shown from the diagonalization of (5). Thus we see that,

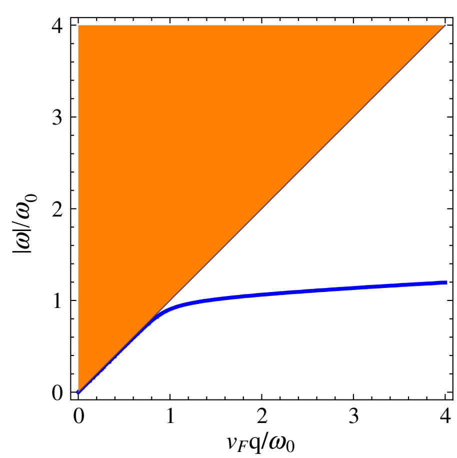

FIG. 1 (color online). Plot of the region corresponding to the *e-h* continuum (shaded area) and the dispersion of the hybrid states made of an optical phonon and an *e-h* pair, obtained from the poles of D_+ and D_- for $g/v_F = 1$.

even in the limit $|\mathbf{q}| \to 0$, the weight of the phonon is comparable to that of the e-h pair in the hybrid state.

It is remarkable that, in the case of the out-of-plane phonons, the interaction with the electronic degrees of freedom gives rise to an effect more pronounced than the Kohn anomalies observed in the dispersion of in-plane optical phonons [18,19]. For $v_F|\mathbf{q}| < \omega_0$, the hybrid state has the character of a bound state made of the e-h pair and the out-of-plane phonon, since its energy is below the threshold for the excitation of the e-h pair. The value of the pole $\omega_{\rm bs}$ in $D_-(\mathbf{q}, \omega)$ is actually given by

$$\omega_{\rm bs}(\mathbf{q}) \approx v_F|\mathbf{q}| - \left(\frac{g^2}{8v_F^2}\right)^2 \frac{v_F^3|\mathbf{q}|^3}{2\omega_0^2} + \ldots. \qquad (7)$$

The formation of bound states of an electron or an exciton with an optical phonon has been discussed long ago in semiconductors [20] and in the two-dimensional electron gas in a magnetic field [21]. In the case of graphene, the branch of hybrid states becomes very soft because of the singular dependence of the polarization (3). It is worth mentioning that this phenomenon is not significantly altered when $\tilde{\Pi}_0$ is dressed by non-RPA interactions. The bare polarization can be corrected for instance by scattering mediated by phonons between the electron and the hole in the one-loop diagram. However, this interaction may only give rise to subdominant terms when compared to the diagrams summed up in the above RPA scheme, which accounts for the largest number of fermion loops at each perturbative level. More relevant may be the effect of the Coulomb scattering at low momentum transfer since, as shown in Ref. [22], that gives rise to ladder diagrams which have an extra logarithmic singularity at the threshold, in comparison to the RPA diagrams at the same perturbative level. The point is that such a Coulomb scattering of the electron and the hole in the polarization has the effect of an attractive interaction, enhancing the effect of the phonon-exchange in the RPA encoded in (6). Thus, the main corrections to our RPA scheme may only lead to a shift in the position of the threshold for e-h excitation [22], without introducing qualitative changes in our discussion of the hybrid states.

The low-energy branch below the e-h continuum leads to a channel for the decay of QPs, as the maximum energy that these can release in a scattering process is enough to hit the branch (7). Taking the phonon-mediated interaction as the relevant source of scattering, we integrate out the e-h pairs in (5) to obtain the phonon propagator $D(\mathbf{q}, \omega)$ dressed with the polarization (3)

$$D(\mathbf{q}, \omega) \approx \frac{2\omega_0}{\omega^2 - \omega_0^2 + i\epsilon - 2\omega_0 g^2 \tilde{\Pi}_0(\mathbf{q}, \omega)}. \qquad (8)$$

The pole in the propagator (8) corresponds again to the dispersion plotted in Fig. 1. The QP decay rate τ^{-1} can be computed from the electron self-energy $\Sigma^{(a)}(\mathbf{k}, \omega_k)$ as

$$\tau^{-1} = -\mathrm{Im}\Sigma^{(a)}(\mathbf{k}, v_F|\mathbf{k}|)$$
$$\approx \mathrm{Im} 2ig^2 \int \frac{d^2q}{(2\pi)^2} \frac{d\omega_q}{2\pi} G^{(b)}(\mathbf{k}-\mathbf{q}, v_F|\mathbf{k}| - \omega_q) D(\mathbf{q}, \omega_q). \qquad (9)$$

In Eq. (9), the imaginary part of $G^{(b)}$ enforces the constraint $\omega_q = v_F|\mathbf{k}| - v_F|\mathbf{k} - \mathbf{q}|$. For that frequency, the phonon propagator picks up an imaginary contribution only from the branch (7). We have actually

$$\tau^{-1} \approx \pi g^2 \int \frac{d^2q}{(2\pi)^2} \delta(Q(\mathbf{q}, \Omega_{\mathbf{q}})), \qquad (10)$$

where $\Omega_{\mathbf{q}} \equiv v_F|\mathbf{k}| - v_F|\mathbf{k} - \mathbf{q}|$ and $Q(\mathbf{q}, \Omega_{\mathbf{q}})$ is the real part of $D^{-1}(\mathbf{q}, \Omega_{\mathbf{q}})$. After trading in (10) the azimuthal variable of integration ϕ by $\Omega_{\mathbf{q}}$, we get

$$\tau^{-1} \approx \frac{1}{2\pi} g^2 \int_0^{|\mathbf{k}|} dq|\mathbf{q}| \int_0^{v_F|\mathbf{q}|} d\Omega_{\mathbf{q}} \left| \frac{\partial \phi}{\partial \Omega_{\mathbf{q}}} \right| \times \left| \frac{\partial Q}{\partial \Omega_{\mathbf{q}}} \right|^{-1} \delta(\Omega_{\mathbf{q}} - \omega_{\rm ph}(\mathbf{q})). \qquad (11)$$

The expression (11) leads to different behaviors depending on whether the QP energy is above or below the scale ω_0. In the range where $v_F|\mathbf{k}| \gg \omega_0$, it is easy to see that the Jacobian $|\partial\phi/\partial\Omega_{\mathbf{q}}|$ scales as $\sim|\mathbf{k}|/|\mathbf{q}|\sqrt{4\mathbf{k}^2 - \mathbf{q}^2}$, while $|\partial Q/\partial\Omega_{\mathbf{q}}|^{-1}$ does not scale with momentum. The QP decay rate shows then a linear dependence on energy $\tau^{-1} \sim (g^2/v_F^2)v_F|\mathbf{k}|$, in agreement with previous analyses of the decay due to optical phonons [23]. On the other hand, when the QP energy is below ω_0, we find that $|\partial\phi/\partial\Omega_{\mathbf{q}}|$ scales as $\sim\omega_0\sqrt{|\mathbf{k}| - |\mathbf{q}|}/\sqrt{|\mathbf{k}|}v_F^2\mathbf{q}^2$. Moreover, we also have $|\partial Q/\partial\Omega_{\mathbf{q}}|^{-1} \sim v_F^3|\mathbf{q}|^3/\omega_0^3$. We get then a decay rate

$$\tau^{-1} \approx \frac{1}{8\pi} \frac{g^4}{v_F} \frac{|\mathbf{k}|^3}{\omega_0^2} \int_0^1 dx x^2\sqrt{1-x}. \qquad (12)$$

We may compare the result (12) with the rate arising from the decay into acoustic phonons. In this case the phonon propagator takes the form $D_0(\mathbf{q}, \omega) \approx v_s^2\mathbf{q}^2/(\omega^2 - v_s^2\mathbf{q}^2 + i\epsilon)$, v_s being the speed of sound in graphene. The decay rate from acoustic phonons $\tau_{\rm ac}^{-1}$ is given by an expression like (9), but with a prefactor which can be written as $\tilde{g}^2/\omega_D$, in terms of the Debye frequency ω_D and the coupling $\tilde{g}^2$ accounting for the effect of both longitudinal and transverse phonons [24]. Following the same steps as before, we get

$$\tau_{\rm ac}^{-1} \approx \frac{1}{8\pi}\tilde{g}^2 \frac{1}{\omega_D} \int_0^{2|\mathbf{k}|} dq|\mathbf{q}| \left| \frac{\partial\phi}{\partial\Omega_{\mathbf{q}}} \right|_{\Omega_{\mathbf{q}} = v_s|\mathbf{q}|} v_s|\mathbf{q}|,$$
$$\approx \frac{1}{4\pi}\tilde{g}^2 \frac{v_s}{v_F} \frac{|\mathbf{k}|^2}{\omega_D} \int_0^2 dx x \frac{1}{\sqrt{4-x^2}}. \qquad (13)$$

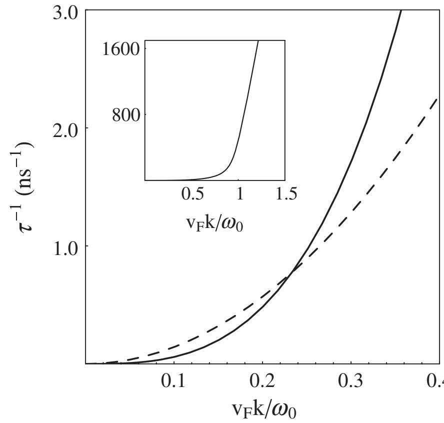

FIG. 2. Plot of the contributions to the decay rate arising from the low-energy branch of hybrid states at the K point (full line) and from acoustic phonons (dashed line). The inset shows the decay rate from the hybrid states over a larger scale, where it is seen the crossover from cubic to linear dependence on QP energy at the scale ω_0 ($\approx$70 meV).

We find that the decay rate $\tau_{\rm ac}^{-1}$ has a quadratic dependence on energy, being further suppressed by the ratio v_s/v_F. There must be therefore a crossover between the range of energies where the relevant contribution to the decay rate is given by (12) and the lower energy regime in which the decay is dominated by the acoustic phonons. This is shown in Fig. 2, where the plots corresponding to the two contributions (11) and (13) have been obtained assuming a value of the deformation potential $D = 10$ eV for the out-of-plane phonons and twice that value for the acoustic phonons.

We arrive at the conclusion that, apart from the acoustic phonons, there are also soft modes arising from the hybridization of out-of-plane phonons with e-h pairs, which contribute to the scattering of QPs at low energies. Such an hybridization is stronger as the Fermi level becomes closer to the CNP of graphene. In general, the contribution of the hybrid modes to the QP scattering will be switched off below an energy scale given by the chemical potential of the system, as this acts as an infrared cutoff in the polarization. This is consistent with the results of Ref. [14] for those measurements closer to the CNP. In such cases, the measures of the resistivity have shown that the linear temperature regime characteristic of the scattering by acoustic phonons ends at a scale which is $\sim$150 K. This agrees well with the minimum width of the range of dominance of the acoustic phonons predicted from the scale of the crossover in Fig. 2.

The results we have obtained may serve to estimate the mean free paths that can be expected in clean graphene samples. From the plot in Fig. 2, we observe that coherent transport can be achieved in graphene up to distances well above the micron scale, at QP energies of the order of several tens of meV. The present analysis may then be useful to interpret the results of transport experiments in graphene around the CNP, where a crossover should be observed from the quadratic decay rate characteristic of the acoustic phonons to the enhanced decay arising from the low-energy hybrid states made of out-of-plane phonons and e-h pairs.

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