Velocity Renormalization and Carrier Lifetime in Graphene from Electron-Phonon Interaction

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We present a first-principles investigation of the phonon-induced electron self-energy in graphene. The energy dependence of the self-energy reflects the peculiar linear bandstructure of graphene and deviates substantially from the usual metallic behavior. The effective band velocity of the Dirac fermions is found to be reduced by 4-8 %, depending on doping, by the interaction with lattice vibrations. Our results are consistent with the observed linear dependence of the electronic linewidth on the binding energy in photoemission spectra.

The recent fabrication of single-layer graphene has attracted considerable interest because low-energy charge carriers in this material have dispersion curves similar to Dirac fermions with zero rest mass and constant group velocity. Because of the peculiar electronic structure of graphene, electrons and holes exhibit exceptionally large mobilities, and the density of states can be tuned over a wide range by applying a gate voltage. These properties make graphene a promising candidate for new-generation electronic and spintronic devices.

Angle-resolved photoemission spectroscopy (ARPES) is used as a powerful tool for investigating quasiparticle behavior with extremely fine energy and momentum resolution. The photoelectron intensity provides information about the energy vs. momentum dispersions of the charge carriers and the associated lifetimes. Recent photoemission experiments performed on graphene showed a peculiar dependence of the hole lifetime on the binding energy, as well as a significant velocity renormalization. The measured carrier lifetime has been discussed within a model including three different decay channels: electron-phonon (e-ph) scattering, electron-plasmon scattering, and electron-hole pair generation. The linear dependence of the linewidth on the binding energy was attributed to the generation of electron-hole pairs. The phonon-induced lifetime was assumed to be energy-independent as found in conventional metallic systems. A subsequent theoretical work analyzed the carrier lifetimes in graphene by adopting a two-dimensional electron-gas model, and concluded that the experimental results could be explained without invoking the e-ph interaction.

In this work we investigate the e-ph interaction in graphene within a first-principles approach. We calculated the electron self-energy arising from the e-ph interaction using a dense sampling of the scattering processes in momentum space, and we extracted the velocity renormalization and the carrier lifetimes from the corresponding real and imaginary parts of the self-energy, respectively. Our analysis shows that the self-energy associated with the e-ph interaction in graphene is qualitatively different from that found in conventional metals. The imaginary part of the self-energy shows a linear energy dependence above the phonon emission threshold, which directly reflects the bandstructure of graphene. The real part of the self-energy leads to a Fermi velocity renormalization of 4-8 % depending on doping. We further propose a simple analytical model of the electron self-energy capturing the main features of our first-principles calculations. Our calculation allows us to assign the low-energy kink in the measured photoemission spectrum and part of the linear energy-dependence of the electronic linewidths to the e-ph interaction.

The e-ph interaction in graphene is treated within the Migdal approximation. The contribution to the electron self-energy $\Sigma_{nk}(E; T)$ arising from the e-ph interaction at the temperature $T$ is:

$$
\Sigma_{nk}(E; T) = \sum_{m,\nu} \int \frac{d\mathbf{q}}{A_{BZ}} \left| g_{mn,\nu}(\mathbf{k}, \mathbf{q}) \right|^2 \times \left[ \frac{n_{\nu} + 1 - f_{mk+q}}{E - \varepsilon_{mk+q} - i\delta} + \frac{n_{\nu} + f_{mk+q}}{E - \varepsilon_{mk+q} + i\delta} \right],
$$

where $\varepsilon_{nk}$ is the energy of an electronic state with band index $n$ and wavevector $\mathbf{k}$, and $h\omega_{\nu}$ the energy of a phonon with wavevector $\mathbf{q}$ and branch index $\nu$. $f_{mk}$ and $n_{\nu}$, are the Fermi-Dirac and Bose-Einstein distribution functions, respectively. The integration extends over the Brillouin zone (BZ) of graphene of area $A_{BZ}$ and the sum runs over both occupied and empty electronic states and all phonon branches. The e-ph matrix element is defined by $g_{mn,\nu}(\mathbf{k}, \mathbf{q}) = \langle mn | q + \Delta V_{\nu\mathbf{q}} | nk \rangle$, $\Delta V_{\nu\mathbf{q}}$ being the change in the self-consistent potential due to a phonon with wavevector $\mathbf{q}$ and branch index $\nu$, while $| nk \rangle$, $| nk + \mathbf{q} \rangle$ indicate Bloch eigenstates. Equation (1) takes into account the anisotropy of the e-ph interaction in $k$-space, as well as retardation effects through the phonon frequency in the denominators.

The electronic structure was described within the local density approximation to density-functional theory. Valence electronic wavefunctions were expanded in a plane-waves basis with a kinetic energy cutoff of 60 Ry. The core-valence interaction was treated by...
means of norm-conserving pseudopotentials. Lattice-
dynamical properties were computed through density-
functional perturbation theory. We modeled an iso-
lated graphene by a honeycomb lattice of carbon atoms
within a periodic supercell. The graphene layers were
separated by 8.0 Å of vacuum, and the relaxed C-
C bond-length was 1.405 Å. Doped graphene was mod-
eled by varying the electronic density and introducing
a neutralizing background charge. We first calculated
electronic and vibrational states and the associated
separated graphene by a honeycomb lattice of carbon atoms
that were calculated using a first-principles interpola-
tion based on electron and phonon Wannier functions.

The fine sampling of the BZ was found to be crucial for convergence
of the self-energy. In the calculation of the self-energy we used a broadening parameter δ of 10 meV, comparable with the resolution of state-of-the-art photoemission ex-
periments. The calculations were performed with the electron and phonon occupations [Eq. (1)] corresponding to T = 20 K to make connection with the ARPES ex-
pertiment. In what follows, we discuss the computed
electron self-energy by focusing on a straight segment
perpendicular to the ΓK direction and centered at the K
density of 1000 × 1000 k and k + q points in the irreducible wedge of the BZ by
point in the BZ [Fig. 1].

We note here that within 2.5 eV from the Dirac point,
the angular dependence of the self-energy is insignificant
(at fixed energy E) [17]. As a consequence, the e-phonon
coupling parameter λn(k) = −∂ ReΣnk(E)/∂E|E=E_D is
isotropic in k-space.

Figure 1 shows the calculated imaginary part of the electron self-energy arising from the e-phonon interaction at T = 20 K (solid lines), for (a) intrinsic, (b) electron-doped, and (c) hole-doped graphene. The self-energy Σk(εk) was evaluated along the reciprocal space line segment shown in the upper-left corner. The Fermi level and the Dirac point are shown schematically in each case. We also show the computed imaginary part of the self-energy for a conventional metal (dashed lines) [Ref. [18]].

Electron energy ε_D (eV)

Im Σk(ε_D) (meV)

Figure 2 shows the real part of the electron self-energy
electron-doped and the hole-doped systems [Fig. 1(b)] and
electron energy ε_D (eV)

Re Σk(ε_D) (meV)

Figure 2(c), respectively] can be rationalized by a similar
energy threshold hω_{ph} (i.e., at −|E_D| − hω in Fig. 1), there are no allowed final states for electronic transitions through
Figure 2(b) shows the real part of the electron self-energy
arising from the e-phonon interaction, for intrinsic and for
electron-doped graphene. The behavior of the hole-
doped system is qualitatively similar to the electron-
doped case. While in conventional metals the real part of the self-energy decays at large hole energies (E < −hω_{ph}) [Fig. 2(b), dashed line], the self-energy in graphene shows a monotonic increase in the same energy range [Fig. 2(b), solid line]. Since the wavevector dependence of the self-energy in graphene within a few eV from the Fermi level is negligible [i.e., Σnk(E) ≈ Σn(E)] [17], we obtained the quasiparticle strength Z_{nk} = (1 − ∂ ReΣnk/∂E)^{-1} by
We repeated our calculations by restricting either the 

tering processes contributing to the electron lifetimes.

When only the in-plane optical phonon

and real (lower panels) part for the intrinsic system (left) as

well as for the electron-doped system (right). Note that the

horizontal energy ranges differ from those shown in Fig. 4.

FIG. 3: Comparison between the electron self-energy obtained

from a first-principles calculation (solid lines) and a single-

parameter model (dashed lines): imaginary (upper panels) and

real (lower panels) part for the intrinsic system (left) as

and real (lower panels) part for the intrinsic system (left) as

well as for the electron-doped system (right). Note that the

horizontal energy ranges differ from those shown in Fig. 4.

evaluating $(1 - d \text{Re}\Sigma_{\mathbf{k}}(\varepsilon_{\mathbf{k}})/d\varepsilon_{\mathbf{k}})^{-1}$. In all cases con-

sidered, the e-ph interaction was found to reduce the non-

interacting quasiparticle strength down to at most

$Z_{\mathbf{k}} = 0.93$ at the Fermi level. This suggests that a

quasiparticle picture is still appropriate at low energy, the

e-ph interaction largely preserving the weakly perturbed

Fermi-liquid behavior. The quasiparticle strength is re-

lated to the velocity renormalization through $1 - Z_{\mathbf{k}}^2 =

(v_{\mathbf{k}} - v_{\mathbf{k}}^0)/v_{\mathbf{k}}^0$ and $v_{\mathbf{k}}^0$ being the non-

interacting and the interacting velocity, respectively. The

velocity renormalization is plotted in Fig. 2(c) and Fig. 2(d)

d for the intrinsic and the electron-doped system, respectively.

The velocity renormalization at the Fermi level was found

to increase with the doping level, and amounts to $-4\%,$

$-8\%,$ and $-6\%$ in the intrinsic, the electron-doped, and

in the hole-doped system considered here. Our results in-
dicate that the velocity of Dirac fermions in graphene is af-
fected by the e-ph interaction. This bears important

implications for the transport properties of graphene-
based electronic devices.

In order to provide a simplified picture of the e-ph in-
teraction in graphene, we analyzed the various e-ph scat-

tering processes contributing to the electron lifetimes.

We repeated our calculations by restricting either the energy

$h\omega_{\mathbf{q}}$, or the momentum transfer $\mathbf{q}$ in Eq. 1 to

limited ranges. When only the in-plane optical phonon

modes between 174 and 204 meV are taken into account

in Eq. 1, the electron linewidth is found to deviate from the full ab initio result by 15\% at most. In contrast, when the momentum integration in Eq. 1 is restricted to small regions around the high-symmetry points $\Gamma$ and $K$, the linewidth is found to deviate significantly from the full calculation, indicating that a proper account of the entire BZ is essential. Based on this analysis, we devised a simplified single-parameter model of the e-ph interaction in graphene. We assumed: (i) linear electronic dis-

persions up to a few eV away from the Dirac point [19], (ii) an Einstein model with the effective phonon energy $h\omega_{\mathbf{q}}$ set to that of the highest degenerate zone-center mode, (iii) an effective e-ph vertex $g$, independent of the electron and phonon momenta [17]. The e-ph matrix element $g$ represents a free parameter in our simplified model, and has been determined by matching the model self-energy with the full ab initio result. Within these assumptions, and with the Fermi level set to zero, the imaginary part of the self-energy reads

$$\text{Im} \Sigma(E) = \frac{\sqrt{3}a^2}{16} \alpha_G^2 g^2 |E - \text{sgn}(E) h\omega_{\mathbf{q}} - E_D|,$$  \quad (2)

whenever $|E|$ exceeds the characteristic phonon energy $h\omega_{\mathbf{q}}$, and vanishes otherwise. In Eq. 2, $a$ is the lattice parameter in Bohr units, $\alpha_G = e^2/4\pi\hbar = 2.53$ is the effective fine structure constant of graphene, and $g$ is the average e-ph matrix element in Rydberg units. The fitting to our calculated ab initio self-energy gave $g = 3.5 \times 10^{-2}$ Ry. The real part of the model self-energy can be straightforwardly obtained from Eq. 2 through Kramers-Krönig relations. Figure 3 shows that this simplified model is in fairly good agreement with the full first-principles calculation. Therefore, despite its simplic-
ity, our single-parameter model captures the qualitative
features of the e-ph interaction in graphene.

In Fig. 4 we compare our first-principles calculations with the width of the momentum distribution curve (MDC) measured by ARPES experiments at 20 K on graphene with a similar doping \[13\]. The width \(\Delta k_{\text{MDC}}\) of the MDC was calculated taking into account renormalization effects through \(\Delta k_{\text{MDC}} = Z_{\text{ph}} 2 \text{Im} \Sigma_{\text{ph}} / \hbar v_{\text{F}}\). Figure 4 shows that, contrary to previous findings \[6\], the e-ph interaction plays a significant role in reducing the carrier lifetime in graphene, as it accounts for about a third of the measured linewidth at large binding energies. The e-ph contribution to the width of the MDC is found to increase linearly at large binding energy, in agreement with experiment.

In conclusion, we have computed from first-principles calculations the Fermi level was set in order to simulate the sample with 2.1 \(\times 10^{13}\) electrons/cm\(^2\) in Fig. 3 of Ref. 6.

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\[16\] Our calculated phonon dispersions are in excellent agreement with previous first-principles results of Ref. 13.

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\[18\] The e-ph interaction of a conventional metal was modeled by assuming constant density of states, an Einstein spectrum, and a constant e-ph matrix element [S. Engelsberg and J. R. Schrieffer, Phys. Rev. 131, 993 (1963)]. These quantities were determined in such a way to match the height of the graphene self-energy near the Fermi level.

\[19\] We used an energy cutoff of 6 eV. The resulting self-energy was found to be largely insensitive to this choice.

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Note added: After submission of this manuscript we became aware of a related work whose results are in agreement with our conclusions \[22\].