Charge-Carrier Screening in Single-Layer Graphene

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The effect of charge-carrier screening on the transport properties of a neutral graphene sheet is studied by directly probing its electronic structure. We find that the Fermi velocity, Dirac point velocity, and overall distortion of the Dirac cone are renormalized due to the screening of the electron-electron interaction in an unusual way. We also observe an increase of the electron mean free path due to the screening of charged impurities. These observations help us to understand the basis for the transport properties of graphene, as well as the fundamental physics of these interesting electron-electron interactions at the Dirac point crossing.

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Unlike normal metals where charge carriers and impurities are highly screened by the Fermi sea, the introduction of charges to a neutral graphene sheet has several competing effects on its transport properties, due to the screening of the electron-electron interaction and the screening of long-range impurities (such as charges or vacancies). While the former is expected to renormalize the Fermi velocity and the Dirac point velocity [1], the latter could lead to a decrease of the quasiparticle scattering rate or an increase in the electron mean free path [2,3]. These effects are important for those applications, such as spintronics, where the electron mean free path is a more relevant quantity than the conductivity [4]. Although the effect of electronic screening has been extensively studied in the past [1,3,5–15], revealing unusual behavior upon the introduction of charge carriers on the graphene sheet [16], most of these works have focused on the renormalization of the Fermi level [17,18] and cannot address the important question of how the various electronic effects can renormalize the Dirac cone or otherwise contribute to conduction.

Angle-resolved photoemission spectroscopy (ARPES) is an ideal tool to probe the electronic properties of graphene [19–25]. In many of these works, however, the starting graphene layer is highly doped, and as a consequence the effect of charge carriers on the electronic screening is more difficult to discern. Similarly, in the case of undoped graphene, the focus has been on how the dielectric screening of the substrate has an impact on the electronic dispersions [23,24]. Therefore, the questions of how charge-carrier screening affects the Dirac cone dispersion and how it differs from dielectric screening remain open questions.

Here, we demonstrate the effects of charge-carrier screening on a graphene sheet: With the progressive deposition of small quantities of potassium, we observe a singularity in the Fermi velocity and Dirac point velocity, an overall renormalization of the valence band, a decrease in the quasiparticle scattering rate, and qualitatively different behaviors from the case of dielectric screening. These results demonstrate the many ways in which charged impurities can have an impact on the transport properties of graphene.

The hexagonal boron nitride (h-BN) samples were prepared by chemical vapor deposition growth on a Cu film, followed by transfer to the h-BN substrate and hydrogen annealing. The graphene was placed on mechanically exfoliated flakes of h-BN, many layers high (opaque to visible light), which were in turn supported on a doped Si wafer with native oxide. The sample preparation was nearly identical to that described in past references (the samples in our experiment were not patterned) [26–28]. Our ARPES investigation was performed at beam line 12.0.1.1 at the Advanced Light Source, at a pressure better than \(3 \times 10^{-11}\) torr, with a sample temperature of 15 K and photon energy 50 eV. The sample was annealed at 700 °C in UHV prior to measurement. The sample was electron doped \textit{in situ} by potassium deposition with a SAES Getters alkali metal dispenser at 15 K, under which conditions the potassium atoms sit above the graphene surface in a disordered arrangement [21,29]. The \textit{in situ} deposition allowed us to study the same position on the sample as potassium was progressively added.

Much attention has recently been focused on the properties of h-BN as a substrate for graphene electronics [26–28,30]. Graphene/h-BN has significantly improved transport properties and fewer charged impurities than previously studied graphene/SiO\(_2\) systems [31]. ARPES spectra of graphene/h-BN are shown in Fig. 1. Following the maximum intensity, one can clearly observe nearly linear energy spectra, characteristic of Dirac electrons [32]. As potassium is added to the sample, the Dirac point appears and moves to higher binding energy [indicated by the black arrows in (d)–(h)], and the charge density of the sample increases.
rates are expected to increase with charge density \[34\]. The
fers from short-range impurity scattering, where scattering
magnitude, the MDC widths decrease. This behavior dif-
tional to the square root of the charge density. We observe
with increasing doping \[8\].

As potassium is added, the spectral widths do not
increase significantly, which is surprising since impurities
often broaden ARPES spectra. In Fig. 2 we examine this
effect, showing the widths of momentum distribution
curves (MDCs, or intensity profiles as a function of mo-
mentum) for different dopings. Panel (a) shows that the
spectral widths vary almost linearly as a function of bind-
ing energy but have an overall offset: The sample with
higher charge density (larger \(k_F\)) has smaller (sharper)
MDC widths.

To quantify this further, in (b), we plot the widths of
MDCs at the Fermi level, as a function of the Fermi
momentum \(k_F\). The width of an MDC at the Fermi level
is proportional to the quasiparticle scattering rate or inverse
mean free path of the photohole \[3,33\], and \(k_F\) is propor-
tional to the square root of the charge density. We observe
that, as the carrier concentration increases in absolute
magnitude, the MDC widths decrease. This behavior dif-
ers from short-range impurity scattering, where scattering
rates are expected to increase with charge density \[34\]. The
decrease in the MDC widths also cannot be due to other
many-body effects, which generally have vanishing con-
thributions to the spectral widths at the Fermi level \[35\].
Therefore, the screening of long-range impurity scattering
is the only remaining explanation for the sharpening of
spectral features: When the increase in charge density
improves the screening of long-range impurity potentials,
the quasiparticle scattering rate is expected to decrease
with increasing doping \[8\].

![Figure 1](image1.png)

**FIG. 1** (color online). ARPES spectra of graphene on \(h\)-BN. (a) ARPES spectrum of single-layer graphene along the \(\Gamma\)-\(K\) direction. This is the direction along which all of our data are
analyzed in subsequent figures. (b) ARPES spectrum along the \(K\)-\(K'\) direction (perpendicular to \(\Gamma\)-\(K\)) shows that the Dirac point
is at the Fermi level. The spectra in panels (a), (b) have been
normalized by the area under the MDCs. (c) The pointlike Fermi
surface of graphene. (d)–(h) Doping dependence along the \(\Gamma\)-\(K\) direction, with Fermi \(k\) vectors corresponding to \(k_F = 0.0035, 0.0037, 0.0126, 0.0206, 0.0282 \text{ Å}^{-1}\), respectively.

![Figure 2](image2.png)

**FIG. 2** (color online). The screening of charged impurities can lower the quasiparticle scattering rate. (a) The imaginary self-
energy \((\text{Im}\Sigma)\), proportional to the width of ARPES spectra and inversely proportional to the quantum lifetime of the photohole,
shows for two different dopings \((n)\) is proportional to \(k_F^2\). In addition to the roughly linear binding energy dependence for
each spectrum is an overall offset between them: The spectra
with added potassium have sharper spectral features than the as-
grown sample, a naively counterintuitive finding. (b) A compa-
nion of the MDC widths at the Fermi level shows that
increasing the charge density sharpens spectral features by
screening the interaction between quasiparticles and impurities.
The dashed line is a \(1/k_F\) fit to the data, which allows us to
extract an impurity density of \(n_{\text{imp}} \sim 1.6 \times 10^{11} \text{ cm}^{-2}\). The inset
shows the raw MDCs at the Fermi level, confirming that the
blue (slightly darker) curve is sharper than the red (slightly
brighter) one.

Figure 2(b) can be compared to theoretical calculations of the scattering rate from long-range impurities \[6,8\].
The dashed line shows a fit to a \(1/k_F\) behavior, where the
constant of proportionality gives the impurity density

\[
\text{Im} \Sigma = \alpha^2 n_{\text{imp}} v_F \pi I(2\alpha)/k_F + \text{const.} \tag{1}
\]

Here, \(\text{Im}\Sigma\) is the imaginary self-energy, \(\alpha\) is the effective
fine-structure constant of graphene, \(n_{\text{imp}}\) is the impurity
density, \(v_F\) is the Fermi velocity, \(I(2\alpha)\) is a dimensionless
constant \[we use \(I(2\alpha) = 0.22\) \[10\], and const is an
overall offset. Using \(\alpha = 0.78\) (discussed below) and \(v_F =
0.85 \times 10^6 \text{ m/s}\) (the bare local density approximation
velocity), we find \(n_{\text{imp}} = (1.94 \pm 0.37) \times 10^{11} \text{ cm}^{-2}\),
which is typical for graphene/\(h\)-BN \[31\], smaller than the
impurity density of graphene on SiO\(_2\) (typically \(\geq 10^{12} \text{ cm}^{-2}\)
\[36\], and 1 order of magnitude smaller than the potassium
density of the highest doping in Fig. 1 \((1.3 \times 10^{12}, \text{ assum-
ing} \sim 1 \text{ electron donated per potassium atom} \[37\]).

To further investigate the effect of long-range screening,
in Fig. 3 we report the doping dependence of the graphene
band structure. The electronic energy-momentum disper-
sion of graphene can be obtained by fitting Lorentzian line
shapes to the MDCs and extracting peak positions as a
function of energy. For as-grown graphene/\(h\)-BN, the
extracted dispersion has been displayed as the solid black
line in Fig. 3(a), illustrating the logarithmic velocity
enhancement in the vicinity of the Dirac point.

As the charge density increases, the Fermi velocity
decreases. The band dispersions near the Fermi level are
plotted for several dopings in Fig. 3(b). The Fermi velocity,
as a function of doping [39]. Interaction is also known to renormalize the Dirac cone [1]. Electron interaction, the Dirac point velocity may be the most fundamental quantity [1]. Overall, for the purposes of understanding the electron-phonon renormalization become significant, causing the Fermi shift of the Fermi energy would lead to a logarithmic Fermi divergence near the Dirac point, even a rigid [38], with a van Hove singularity at higher dopings [21].

In Fig. 3, we observe the differences between two separate types of electronic screening effects: screening by the graphene charge carriers, which has been modified in this study by changing the number of charge carriers through potassium deposition, and screening by the dielectric environment, which can be modified by changing the dielectric substrate [24]. In both cases, the Fermi level and Dirac point velocities are modified, varying linearly with $e$ and logarithmically with $k_F$ or doping. On the other hand, for charge-carrier screening, the inverse screening length $q_s$ varies linearly with $k_F$ and with $1/e$, given by [3]

$$q_s = 4\alpha k_F.$$  

One might therefore expect the differences between these screening effects to be observed at high values of momentum, where $k \gg q_s$.

So, in Fig. 4(a), we compare the doping dependence of the graphene valence band dispersions over a larger range of energy and momentum than Fig. 3(c). At lower values of momentum (near the Dirac point), the band velocities decrease as a function of doping, but, at higher momenta, this trend begins to reverse, with increasing velocity as a function of doping near 0.1 Å⁻¹. In contrast, Fig. 4(b) (and proportionally to the slope of the ARPES dispersions, decreases by a factor of 4, with a maximum of $2.0 \times 10^6$ m/s, as the Fermi $k$ vector increases by 2 orders of magnitude. These results have been plotted as the red crosses in Fig. 3(d) and confirm the results of past experiments [17,18].

However, there are several effects that cooperate to reduce the Fermi velocity as a function of doping. For instance, the bare band of graphene along the $\Gamma-K$ direction is known to decrease in velocity away from the Dirac point [38], with a van Hove singularity at higher dopings [21]. Given that the band dispersion in Fig. 3(a) demonstrates a logarithmic divergence near the Dirac point, even a rigid shift of the Fermi energy would lead to a logarithmic Fermi velocity dependence [23]. Finally, the electron-phonon interaction is also known to renormalize the Dirac cone as a function of doping [39].

Therefore, in order to separate the electron-electron renormalization from other many-body and bare-band effects, in Fig. 3(c), we show the electronic dispersions in the vicinity of the Dirac point as a function of doping, and the velocities have been plotted in Fig. 3(d) [40]. The Fermi velocity and Dirac point velocity roughly coincide for low dopings. However, as the charge density increases, the effects of the bare-band velocity and electron-phonon renormalization become significant, causing the Fermi velocity to be only half as large as the Dirac point velocity. Overall, for the purposes of understanding the electron-electron interaction, the Dirac point velocity may be the most fundamental quantity [1].

The Dirac point velocity can be fitted with a logarithmic dependence [1,18]

$$v_D^* = v_D \frac{r_s^0}{4(1 + \alpha k_F^2)} \ln \left( \frac{k_F}{k_F^0} \right) + \text{const},$$  

where $r_s = e^2/\nu e$ gives the value of the dielectric constant and the constant $\alpha$ introduces the same fit parameter as Ref. [18], allowing $e$ to effectively increase with charge density. From the fit, we obtain $\alpha = 720 \AA^2$ and $v_D r_s^0/4 = (0.168 \pm 0.014) \times 10^6$ m/s. Using the bare local density approximation value of $v_D = 0.85 \times 10^6$ m/s, we obtain $\alpha = 0.78$ or $e_0 = 3.3$. This value of $e$ compares well with the reported logarithmic fit to the binding energy dependence in undoped graphene/h-BN [24], where $e_0 = 4.22$, and with the expected dielectric screening, being $e_0 = (e_{h-BN} + 1)/2 = 4.02$ [41].
For $k \gg k_F$, the extent of the renormalization (or magnitude of the self-energy) is found to be strongly dependent on the dielectric constant $\varepsilon$ but weakly dependent on the screening by charge carriers or $k_F$. This confirms that the electron-electron interaction is indeed a long-range interaction, with a variable length scale due to the concentration of free charges in graphene. The ways in which charge-carrier screening and dielectric screening modify the Dirac cone are illustrated in Figs. 4(c) and 4(d), respectively.

In conclusion, we have demonstrated some of the detailed ways in which the addition of charge carriers to a graphene sheet can have an effect on transport properties and the renormalization of the Dirac cone. The electron-electron and electron-impurity interactions are found to be long-range interactions, and in both cases the addition of charge carriers is shown to decrease the length scale and strength of the interaction. The increase in charge density is also shown to renormalize the Dirac cone in a distinct manner from dielectric screening. These results illustrate the differences between charge-carrier screening and dielectric screening in graphene, illuminating the transport behavior of graphene while demonstrating the interesting differences between the electronic interactions of graphene and those of ordinary metals.

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Ref. [24]) shows that, when the band dispersions of graphene on different dielectric substrates are compared, increasing the dielectric constant leads to uniformly smaller band velocities at any given value of momentum or energy.

To make this comparison more straightforward, Fig. 4(b) shows three graphene dispersions: (i) as-grown graphene/h-BN, (ii) as-grown graphene/SiC(0001), and (iii) doped graphene/h-BN. The doping for (iii) was chosen so that the band velocity near the Dirac point would match that of (ii). The cartoons in (c) and (d) illustrate the renormalization effect on the Dirac cones for charge-carrier screening and dielectric screening, respectively. In (c), the renormalization is primarily restricted to low momenta. In (d), the renormalization extends to all momenta within our range of measurement and becomes larger in magnitude at higher momenta. See Supplemental Material [42] for further discussion of data analysis techniques.

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