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 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.

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, the latter could lead to a decrease of the quasiparticle scattering rate or an increase in the electron mean free path. These effects are important for those applications, such as spintronics, where the electron mean free path is a more relevant quantity than the conductivity. Although the effect of electronic screening has been extensively studied in the past, revealing unusual behavior upon the introduction of charge carriers on the graphene sheet, most of these works have focused on the renormalization of the Fermi level, 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. 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. Therefore, the question of how charge carrier screening affects the Dirac cone dispersion and how it differs from dielectric screening remains an open question.

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 h-BN samples were prepared by CVD 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). Our ARPES investigation was performed at beamline 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 to 700°C in UHV prior to measurement. The sample was electron-doped in situ by potassium deposition with an SAES Getters alkali metal dispenser at 15K, under which conditions the potassium atoms sit above the graphene surface in a disordered arrangement. The 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 hexagonal boron nitride (h-BN) as a substrate for graphene electronics. Graphene/h-BN has significantly improved transport properties and fewer charged impurities than previously studied graphene/SiO$_2$ systems. ARPES spectra of graphene/h-BN are shown in Figure 1. Following the maximum intensity, one can clearly observe nearly linear energy spectra, characteristic of Dirac electrons. As potassium is added to the sample, the Dirac point appears and moves to higher binding energy (indicated by the black arrows in panels d-h), and the charge density of the sample increases.

As potassium is added the spectral widths do not increase significantly, which is surprising since impurities often broaden ARPES spectra. In Figure 2 we examine this effect, showing the widths of momentum distribution curves (MDCs, intensity profiles as a function of momentum) for different dopings. Panel (a) shows that the spectral widths vary almost linearly as a function of binding energy, but have an overall offset: the sam-

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constant of proportionality gives the impurity density: \( \alpha \). Here \( \Im \Sigma \) 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 \( I(2\alpha) \approx 0.22 \)), and \( \text{Const} \) is an overall offset. Using \( \alpha = 0.78 \) (discussed below) and \( v_F = 0.85 \times 10^6 \text{m/s} \) (the bare LDA velocity), we find \( n_{\text{imp}} = (1.94 \pm 0.37) \times 10^{11} \text{cm}^{-2} \), which is typical for graphene/h-BN \(^{30}\), smaller than the impurity density of graphene on SiO\(_2\) (typically \( \geq 10^{12} \text{cm}^{-2} \)) \(^{36}\), and an order of magnitude smaller than the potassium density of the highest doping in figure 1 \( (1.3 \times 10^{12} \text{ cm}^{-2} ) \), assuming \( 1 \) electron donated per potassium atom \(^{37}\).

To further investigate the effect of long-range screening, in Figure 3 we report the doping dependence of the graphene bandstructure. The electronic energy-momentum dispersion of graphene can be obtained by fitting Lorentzian lineshapes 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 figure 3a, 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 panel 3b. The Fermi velocity, proportional to the slope of the ARPES dispersions, decreases by a factor of 4, with a maximum of \( 2 \times 10^6 \text{ m/s} \), as the Fermi k-vector increases by two orders of magnitude. These results have been plotted as the red open circles in panel 3d 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 \( \Gamma-K \) di-
The Dirac point velocity can be fitted with a logarithmic dependence on the Fermi energy, allowing for effective relationships between 

\[ v_D = v_D^0 \frac{r_s^0}{4(1 + ak_F^2)} \ln \left( \frac{k_F}{k_F^0} \right) + \text{Const.} \]

where \( r_s \equiv e^2/\varepsilon \varepsilon \) gives the value of the dielectric constant, and the constant \( a \) introduces the same fit parameter as Ref. \[18\], allowing \( \varepsilon \) to effectively increase with charge density. From the fit we obtain \( a = 720 \text{Å}^2 \), and \( v_D^0 = 0.168 \pm 0.014 \times 10^6 \text{m/s} \). Using the bare LDA value of \( v_D = 0.85 \times 10^6 \text{m/s} \), we obtain \( \alpha = 0.78 \) or \( \varepsilon^0 = 3.3 \). This value of \( \varepsilon \) compares well with the reported logarithmic fit to the binding energy dependence in undoped graphene/h-BN \[24\] where \( \varepsilon^0 = 4.22 \); and with the expected dielectric screening, being \( \varepsilon^0 = (\varepsilon_{\text{BN}} + 1)/2 = 4.02 \text{[41]} \),

In figure 4, 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 \( \varepsilon \), 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 \( 1/\varepsilon \), given by \[23\]:

\[ q_s = 4\alpha k_F. \]

One might therefore expect differences between these screening effects to be observed at high values of momentum, where \( k > q_s \),

So in figure 4a we compare the doping-dependence of the graphene valence band dispersions over a larger range of energy and momentum than figure 3c. 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, figure 4b (and 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.
FIG. 4: Dirac cone renormalization by charge-carrier screening. (a) Electronic dispersions near the Dirac point show higher binding energy behavior than figure 3c. The inset shows the difference between the experimental bands and an arbitrary straight line. Panel (b) illustrates the difference between charge carrier screening and dielectric screening. Three dispersions are shown: (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 panels (c) and (d) illustrate the renormalization effect on the Dirac cones for charge-carrier screening and dielectric screening, respectively. Panel (c), the renormalization is primarily restricted to low momenta. In panel (d), the renormalization extends to all momenta within our range of measurement, and becoming larger in magnitude at higher momenta.

To make this comparison more straightforward, figure 4b shows three graphene dispersions: (i) as-grown graphene/h-BN; (ii) as-grown graphene/SiC(0001); and (iii) doped graphene/h-BN. (i) and (ii) have different substrates and therefore different dielectric environments; (i) and (iii) have the same substrate but different charge carrier concentrations; and (ii) and (iii) have different dielectric screening and charge carrier concentrations. While (ii) and (iii) have similar band velocities in the vicinity of the Dirac point, overlapping for small values of k, these dispersions diverge for larger values of k. For k >> k_F the extent of the renormalization (or magnitude of the self-energy) is found to be strongly dependent on the dielectric constant e_r, 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 figures 4c and 4d, 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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