Multiband Transport in Bilayer Graphene at High Carrier Densities

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We report a multiband transport study of bilayer graphene at high carrier densities. Employing a poly(ethylene)oxide-CsClO₄ solid polymer electrolyte gate we demonstrate the filling of the high energy subbands in bilayer graphene samples at carrier densities |n| ≥ 2.4 × 10¹³ cm⁻². We observe a sudden increase of resistance and the onset of a second family of Shubnikov de Haas (SdH) oscillations as these high energy subbands are populated. From simultaneous Hall and magnetoresistance measurements together with SdH oscillations in the multiband conduction regime, we deduce the carrier densities and mobilities for the higher energy bands separately and find the mobilities to be at least a factor of two higher than those in the low energy bands.

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Multiband transport is common for many complex metals where different types of carriers on different pieces of the Fermi Surface (FS) carry electrical currents. Conduction in this regime is controlled by the properties of the individual subbands, each of which can have distinct mobilities, band masses, and carrier densities. Other changes to the single-band conduction model include inter-band scattering processes and mutual electrostatic screening of carriers in different subbands, which alters the effective strength of the Coulomb potential and hence adjusts the strength of electron-electron and electron-charged impurity interactions.

To understand electronic conduction in this regime, it is desirable to study the properties of the individual bands separately and compare these to the properties in the multiband regime. This was achieved in 2-dimensional electron gases (2DEGs) formed in GaAs quantum wells, where the subbands can be continuously populated and depopulated by inducing parallel magnetic fields. In these 2DEGs, an increased overall scattering rate due to interband scattering was observed upon the single- to multiband transition, along with changes in the effective Coulomb potential which led to the observation of new filling factors in the fractional quantum Hall effect.

Bilayer graphene (BLG), with its multiband structure and strong electrostatic tunability, offers a unique model system to investigate multiple band transport phenomena. BLG’s four-atom unit cell yields a band structure described by a pair of low energy subbands (LESs) touching at the charge neutrality point (CNP) and a pair of high energy subbands (HESs) whose onset is ∼ ±0.4 eV away from the CNP (Fig.1(a)). Specifically, the tight binding model yields the energy dispersion [7]:

\[ \epsilon_{1,2}(k) = \pm \sqrt{\frac{\gamma_1^2}{4} + \frac{\Delta^2}{4} + v_F^2 k^2} = \pm \sqrt{\frac{\gamma_1^2}{4} + v_F^2 k^2(\gamma_1^2 + \Delta^2)}, \]

where the upper and lower index indicates the conduction (+) and valence (-); and LES (1) and HES (2), \( k \)

is the wave vector measured from the Brillouin zone corner, \( v_F \approx 10^6 \) m/s is the Fermi velocity in single layer graphene, \( \gamma_1 \approx 0.4 \) eV is the interlayer binding energy, and \( \Delta \) is the interlayer potential asymmetry. Interestingly, since a perpendicular electric field \( E \) across the sample gives rise to an interlayer potential difference \( \Delta \),
it opens up a gap in the spectrum of the LES [8, 10, 13] and is furthermore predicted to adjust the onset energy of the HES. Whereas the LESs have been widely studied, the HESs, with their expected onset density of \(n \gtrsim 2.4 \times 10^{13} \text{ cm}^{-2}\) [8], have thus far not been accessed in transport experiments. This can mainly be attributed to the carrier density limitations set by the dielectric breakdown of the conventional SiO\(_2\)/Si back gates, which do not permit the tuning of carrier densities above \(n \approx 0.7 \times 10^{13} \text{ cm}^{-2}\) (\(\epsilon_F \approx 0.2 \text{ eV}\)).

In this letter, we report multiband transport in bilayer graphene. Using an electrolytic gate, we were able to populate the HES of bilayer graphene, allowing for both the LES and HES to be occupied simultaneously. The onset of these subbands is marked by an abrupt increase of the sample resistivity, most likely due to the opening of an interband scattering channel, along with the appearance of a new family of Shubnikov-de Haas (SdH) oscillations associated with the HES. A detailed analysis of the magneto- and Hall resistivities in combination with the HES SdH oscillations in this regime enables us to estimate the carrier mobilities in each subband separately, where we observe a two-fold enhanced mobility of the HES carriers as compared to the LES carriers at the same band densities.

Bilayer graphene devices were fabricated by mechanical exfoliation of Kish graphite onto 300 nm thick SiO\(_2\) substrates, which are backed by degenerately doped Si to form a back gate. The samples were etched into a Hall bar shape with a typical channel size of \(\sim 5 \mu\text{m}\) and then contacted with Cr/Au (0.5/30 nm) electrodes through beam lithography (Fig. 1(c) inset). In order to access the HES we utilized a recently developed solid polymer electrolyte gating technique [11, 14], which was recently shown to induce carrier densities beyond values of \(n > 10^{14} \text{ cm}^{-2}\) [14] in single layer graphene. The working principle of the solid polymer electrolyte gate is shown in Fig.1(b). Cs\(^+\) and ClO\(_4^-\) ions are mobile in the solid matrix formed by the polymer poly(ethylene)oxide (PEO). Upon applying a gate voltage \(V_{ce}\) to the electrolyte gate electrode, the ions form a thin Debye layer a distance \(d \sim 1 \text{ nm}\) away from the graphene surface. The proximity of these layers to the graphene surface results in huge capacitances per unit area \(C_{eg}\), enabling extremely high carrier densities in the samples. While CsClO\(_4\) has almost the same properties as the typically used LiClO\(_4\) salt, we find a reduced sample degradation upon application of the electrolyte on top of the sample, resulting in considerably higher sample mobilities.

One major drawback of the electrolyte gate for low temperature studies is that it cannot be tuned below \(T < 250 \text{ K}\), where the ions start to freeze out in the polymer and become immobile (though leaving the Debye layers on the bilayer surface intact) [12, 14]. A detailed study of the density dependent transport properties at low temperatures can therefore be quite challenging. In order to overcome this issue, we employ the electrolyte gate just to coarsely tune the density to high values (\(|n| < 10^{14} \text{ cm}^{-2}\) at \(T = 300 \text{ K}\), followed by an immediate cooldown to \(T = 2 \text{ K}\). We then use the standard SiO\(_2\)/Si back gate to map out the detailed density dependence of the longitudinal sheet resistivity \(\rho_{xx}\) and the Hall resistance \(RH\), from which we extract the total carrier density of the sample \(n_H = B/eRH\), with \(B\) the magnetic field and \(e\) the electron charge. Here we find the back gate capacitance to be \(C_{bg} = 141 \text{ aF/\mu m}^2\), almost unaltered by the presence of the Debye layers on top of the sample.

In this experiment, we have measured \(\rho_{xx}\) and \(RH\) of more than 3 BLG devices as a function of the back gate voltage \(V_{bg}\) at various fixed \(V_{eg}\) corresponding to the wide density range of \(n_H \sim \pm 8 \times 10^{13} \text{ cm}^{-2}\). Fig.1(c) shows \(\rho_{xx}\) and \(RH\) for a representative device for 3 selected cool-downs at \(V_{eg} = -1.7\), -0.4, 1 V from left to
right, corresponding to a pre-doping level of \( n_H = (-2.9, 0, 2.9) \times 10^{13} \text{ cm}^{-2} \). For low doping levels (\( V_{eg} = -0.4 \text{ V} \), Fig.1(c middle) we observe the expected Dirac Peak in \( \rho_{xx} \) and the ambipolar transition of \( R_H \) as \( V_{bg} \) sweeps through the CNP. Away from the CNP, \( \rho_{xx} \) and \( R_H \) decrease as \( |H_{\text{Hall}}| \) increases, as was observed before in BLG samples [9]. For the strongly pre-doped gate sweeps however (Fig.1(c) left and right), we observe another set of right), marked by the “spikes” of increased resistivity which form LL fans converging into the onset point of the HES. All traces show periodic oscillations in \( B^{-1} \) allowing us to obtain the SdH density, \( n_{SDH} = \frac{\pi}{2} (B^{-1}) \), assuming that each LL is spin and valley degenerate. Whereas for all \( |n_H| < n^* \) we find that the obtained \( n_{SDH} \approx n_H \), indicating that the SdH oscillations are solely from a single band (i.e., the LES), for \( |n_H| > n^* \) the obtained \( n_{SDH} \) values are much smaller than the simultaneouly measured \( n_H \) values. This behavior can be well explained by assuming that these SdH oscillations reflect only the small fraction of charge carriers lying in the HES. For \( |n_H| > n^* \) we hence are able to extract the occupation densities of the LES (\( n_{LES} \)) and HES (\( n_{HES} \)) from \( n_{LES} = n_H - n_{SAH} \) and \( n_{HES} = n_{SAH} \). Fig. 3(b) shows the \( n_{LES} \) and \( n_{HES} \) in this regime as a function of the total carrier density \( |n_H| \). For each fixed \( V_{eg} \), the obtained \( n_{LES} \) and \( n_{HES} \) increase as \( |n_H| \) increases (adjusted by \( V_{bg} \), for both electrons and holes. Interestingly, we notice that the \( n_{LES}(n_H) \) are slightly larger for larger \( |V_{eg}| \) while the trend is opposite for the HES, i.e. \( n_{HES}(n_H) \) are smaller for larger \( |V_{eg}| \), even though their \( n_H \) values are in similar ranges. These general trends can be explained by an increase of the interlayer potential difference \( \Delta \) for increased values of \( |V_{eg}| \), which are predicted by the tight-binding model in Eq.1 to result in an increase of the onset density (energy) of the HES.

While a precise quantitative determination of the expected shift in the onset density of the HES as a function of \( V_{eg} \) and \( V_{bg} \) requires a self-consistent calculation of \( \Delta(V_{eg}, V_{bg}) \) and would go beyond the scope of this paper, we can still qualitatively test the above prediction. This is possible since \( \Delta \) is mostly controlled by \( V_{eg} \), which has a much stronger coupling to the BLG sample than the \( V_{bg} \), thus allowing us to approximately treat \( \Delta \) as a constant for fixed \( V_{eg} \). Since the experimental traces displayed in Fig.3(b) correspond to different values of \( V_{eg} \) but the same range of \( V_{bg} \), \( \Delta \) is different for each trace and can be extracted from the theoretical fits from Eq. 1, with \( \Delta \) as the only fitting parameter. Indeed for all 4 traces we find good agreement with the theoretical fits; we clearly observe an enhanced onset density (energy)
for the traces with larger set potential differences across the sample, which is in good qualitative agreement with theoretical predictions.

We now turn our attention to the transport properties of BLG in the limit of \( n_H > n^* \). The filling of these sub-bands creates a parallel transport channel in addition to the one in the LES, thus defining the transport properties in this regime by two types of carriers with distinct mobilities \( \mu_{1,2} \), effective masses \( m^*_{1,2} \) and sub-band densities \( n_{1,2} \) (here the index corresponds to the LES(1) and HES(2)) [13, 14]. In sharp contrast to a single band Drude model, where \( \rho_{xx}(B) \) does not depend on the \( B \) field, in a two-carrier Drude theory it is expected to become strongly modified, resulting in a pronounced \( B \) field dependence [15]:

\[
\rho_{xx}(B) = \frac{n_1\mu_1 + n_2\mu_2 + (n_1\mu_1^2 + n_2\mu_2^2)B^2}{e((n_1\mu_1 + n_2\mu_2)^2 + \mu_1^2\mu_2^2(n_1 + n_2)^2B^2)},
\]

Fig. 4(a) shows magnetoresistance traces for different fixed Hall densities \( n_H \). Close to the CNP, where only the LES are populated (Fig.4(a) black trace), the \( \rho_{xx}(B) \) traces are nearly flat as expected from the one-fluid Drude theory. When the density is increased and the HES starts to fill up, however, we observe a smooth transition to an approximately parabolic \( B \) field dependence, resulting in a strong increase of \( \rho_{xx} \) of up to 25% from 0 T to 8 T. Using the previously extracted carrier densities in the two bands \( n_{1,2} \) we can now fit the \( \rho_{xx}(B) \) traces with the two-carrier Drude model in Eq.2, with the mobilities of the two subbands \( \mu_{1,2} \) as the only fitting parameters. As shown in Fig.4(b) the experimental finding are in excellent agreement with the theory, allowing us to deduce the values of \( \mu_{1,2} \) with good accuracy. Moreover, the ability to extract the mobilities of the HES allows us now to characterize the HES in more detail.

In Fig.4(c) we plot the extracted mobilities of the HES \( \mu_2 \) against the carrier density in the HES \( n_2 \) and compare it to the mobilities \( \mu_1 \) of the LES at a similar range of subband densities in the LES \( n_1 \). We find that the mobilities in the HES are at least a factor of two higher than those in the LES. Considering that the effective carrier masses are similar for the LES and the HES, this feature of the HES may be due to the enhanced screening of charged impurity scatterers at higher carrier densities, effectively reducing the scattering rate of the HES carriers on these scatterers. A more detailed theoretical study is required, however, to undertake a quantitative analysis of this problem.

In conclusion, using a polymer electrolyte gate we have achieved two-band conduction in bilayer graphene. We have found that the filling of these bands above a Hall density of \( |n_H| > 2.4 \times 10^{13} \text{ cm}^{-2} \) is marked by an increase of the sample resistivity by \( \sim 10\% \) along with the onset of SdH oscillations. From simultaneous Hall and magnetoresistivity measurements, as well as the analysis of the SdH oscillations in the two carrier conduction regime, we have characterized the distinct carrier densities and mobilities of the individual subbands, where we have found a strongly enhanced carrier mobility in the HES of bilayer graphene.

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