Limitations to carrier mobility and phase-coherent transport in bilayer graphene

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We present transport measurements on high-mobility bilayer graphene fully encapsulated in hexagonal boron nitride. We show two terminal quantum Hall effect measurements which exhibit full symmetry broken Landau levels at low magnetic fields. From weak localization measurements, we extract gate-tunable phase coherence times $\tau_\phi$ as well as the inter- and intra-valley scattering times $\tau_1$ and $\tau_\tau$. While $\tau_\phi$ is in qualitative agreement with an electron-electron interaction mediated dephasing mechanism, the analysis of $\tau_1$ and $\tau_\tau$ points to local strain fluctuation as the most probable mechanism for limiting the mobility in high-quality bilayer graphene.

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Bilayer graphene (BLG) is an interesting material system to explore phase-coherent mesoscopic transport with unique electronic properties. In contrast to single-layer graphene, in BLG a band gap can be opened by an external electric field $\mathbf{E}$ making local depletion of the two-dimensional electron gas (2DEG) possible similar to III/V semiconductor heterostructures. This is an important prerequisite for implementing state-of-the-art phase-coherent quantum device concepts. In contrast to conventional 2DEGs, the massive Dirac fermion nature of the quasi-particles in BLG results in an unconventional quantum Hall effect and promises unique quantum interference properties.

So far, the observable transport phenomena in BLG devices suffer from the limited device quality which is most likely a consequence of the high sensitivity of BLG on the surrounding environment, in particular to the substrate material. Recent developments in device fabrication have shown that a significant improvement in sample quality can be obtained by replacing conventional SiO$_2$ with hexagonal boron nitride (hBN). This material provides an ultra-flat substrate for graphene and enables the realization of the high-mobility samples that are required to study, e.g., quantum phase transitions in the lowest quantum Hall state or superlattice effects such as the Hofstadter butterfly. However, despite these improvements, it remains difficult to experimentally address the microscopic mechanisms that limit its charge carrier mobility and phase-coherence in high-quality bilayer graphene.

To address these important questions, we present and discuss diffusive transport measurements on bilayer graphene fully encapsulated in hBN. Our fabrication technique allows to obtain high-mobility samples, which show a well-developed quantum Hall effect and a full degeneracy breaking of the zero Landau level around $B=6$ T. To investigate the limits of phase-coherent transport and to gain insights on the limitations to carrier mobility in these devices, we perform weak localization measurements.
measurements. From these measurements, we extract the inter- and intra-valley scattering times, as well as the phase-coherence time. Our results indicate that the main source of dephasing in high-quality BLG is the electron-electron interaction, and that mobility is not limited by inter-valley scattering processes. Moreover, we observe that the mean-free path quantitatively matches the intra-valley scattering length over a wide range of carrier densities. Our findings point at intra-valley scattering as the main limitation to mobility in BLG. We discuss local strain fluctuations as the possible source of these mobility-limiting scattering events.

Bilayer graphene flakes encapsulated in hBN were fabricated using a van der Waals pick up technique similar to the one described by Wang et al. [18]. Top and bottom hBN flakes vary between 10 to 30 nm. After stacking, the hBN-BLG-hBN sandwiches were placed on a highly doped silicon substrate with a 285 nm SiO$_2$ top-layer. A chromium hard-mask is patterned by electron-beam (e-beam) lithography, followed by metal evaporation and lift-off. Subsequently, the samples are exposed to a SF$_6$/O$_2$ plasma and a chemical wet etching step is used to remove the chromium mask. The structured sandwiches are then contacted by an additional e-beam step followed by evaporation of Cr/Au with a thickness of 5 nm/100 nm. Finally, the samples are annealed in a tube oven in an Ar/H$_2$ atmosphere for 3 hours at 275 °C. An illustration of a contacted device is shown in Fig. 1(a). The fabrication technique described above allows to make BLG devices of very high-quality, as proven by transport experiments.

In this Letter, we present measurements performed on a two-terminal device with a length $L \approx 16 \mu$m and a width of $W \approx 7 \mu$m (see Fig. 1(b)), placed in a He$^3$ cryostat with a base temperature of $T = 300$ mK (unless stated otherwise). In Fig. 1(c) we show the conductivity of this device as function of back-gate voltage $V_g$ applied to the highly doped silicon substrate at a temperature of 77 K. The charge neutrality point is observed to be at a back-gate voltage $V_g^0 = -10$ V, indicating electron doping in our sample. By taking a gate lever arm of $\alpha = 6.5 \times 10^{10} \text{ cm}^{-2} \text{V}^{-1}$ (see details below) we extract from the linear increase of the conductivity above $|\Delta V_g| = |V_g - V_g^0| \approx 10$ V a lower limit of the hole mobility of $\mu_h \approx 40,000 \text{ cm}^2/\text{Vs}$ and of the electron mobility of $\mu_e \approx 50,000 \text{ cm}^2/\text{Vs}$ (see dashed lines in Fig. 1(c)). These mobility values have been reproduced on a number of encapsulated BLG devices, and are among the highest reported for BLG on substrates.

Another indication of the high-quality of the sample can be deduced from two-terminal quantum Hall measurements. In Fig. 1(d) we show the transconductivity $d\sigma/dn$ as function of perpendicular magnetic-field $B$ and charge carrier density $n = \alpha \Delta V_g$. Dashed lines are exemplarily marking the filling factors $\nu = -12, -8, -4, 4, 8, 12$ and the arrows indicate the positions of the filling factors $\nu = -4, -2, 2, 4$. The eightfold degenerate zero Landau level unambiguously confirms the bilayer nature of the investigated flake. Additionally, we observe a degeneracy lifting into doubly degenerate Landau levels at $B \approx 2.5$ T, and full degeneracy breaking at a magnetic field as low as $B \approx 6$ T (see squares and dots in Fig. 1(d) respectively), which is a direct signature of the very high-quality of our sample. The full symmetry breaking is consistent with observations made in previous experiments [19, 20] and can be attributed to Coulomb and exchange interactions. In Fig. 1(e) we show the differential conductance $dI/dV_b$ as function of carrier density $n$ at constant magnetic field $B = 2.5$ T (solid line). The dashed line is the result of a numerical calculation for an ideal BLG sample with the same aspect ratio of our device, following the approach of Ref. [17] and using the parameters discussed in Ref. [21]. The experimental data (solid line) closely follows the model of ideal BLG at charge carrier densities above $0.5 \times 10^{12} \text{ cm}^{-2}$. The discrepancy at lower carrier density might be either explained by a small band gap resulting from an asymmetric doping of the top and bottom graphene layer or by the fact that symmetry breaking is not included in the model. From this calculation we also extract a gate lever arm of $\alpha = 6.5 \times 10^{10} \text{ cm}^{-2} \text{V}^{-1}$, which is in good agreement with the slope of the dashed lines in Fig. 1(d) as well as with a conventional plate capacitor model with the SiO$_2$ and the bottom hBN flake as gate dielectrics.

We next focus on weak localization (WL) measurements, from which we extract three fundamental time scales of our device: the phase coherence time $\tau_\phi$, the inter-valley scattering time $\tau_i$ as well as the intra-valley scattering time $\tau_s$. In Fig. 2 we show the experimentally observed WL dip, i.e. the change in conductivity at finite magnetic field with respect to the one at $B = 0$ T: $\Delta \sigma(B) = \langle \sigma(n, B) - \sigma(n, B = 0) \rangle \Delta n$. Fig. 2(a) shows $\Delta \sigma(B)$ at three different temperatures ($T = 0.3$ K, 1.1 K and 2.7 K) for a carrier density $n$ close to the charge neutrality point while Fig. 2(b) shows similar data taken at $n = 3.2 \times 10^{11} \text{ cm}^{-2}$. Each trace is obtained by

![FIG. 2. (color online) Magneto conductivity at low magnetic fields and different temperatures $T = 0.3$ K, 1.1 K and 2.7 K for (a) $V_g = -10$ V (charge carrier density $n$ close the charge neutrality point) and (b) $V_g = -5$ V (corresponding to $n = 0.32 \times 10^{12} \text{ cm}^{-2}$). Fits to the weak localization model for BLG are displayed by the dotted lines.](image-url)
carrier density, we observe a significant increase of graphene \[25\], and \[\tau\] of the extracted phase-coherence time by over one order of magnitude up to \[\tau_f\] (Fig. 3(a)). These values are only a factor 3-4 larger than the values of \[\tau_0\] extracted from our WL measurements which is illustrated by the dashed line in Fig. 3(a) where a scaling factor of 3.5 is included. This finding provides a strong indication that electron-electron interaction - which is the only scattering mechanism included in the AAK theory \[26\] - is the dominant source of dephasing in our encapsulated BLG device.

In Figs. 3(b) and 3(c) we show the temperature dependence of \[\tau_0\] close to the charge neutrality point and at \[n = +3.2 \times 10^{11}\] cm\(^{-2}\) respectively. Similarly to Fig. 3(a), the solid line illustrates the estimates for \[\tau_0\] obtained from the AAK theory \((\tau_0^{-1} \sim T, \text{see equation above})\). Above \(T = 400\) mK (below \(1/T = 2.5\) K\(^{-1}\) \[\tau_0\] is inversely proportional to the temperature, which is qualitative agreement with AAK theory (see dashed line in Fig. 3(c))). However, at lower temperatures \[\tau_0\] shows a saturation behavior that points at the existence of another superimposed dephasing mechanism, not related to electron-electron interaction.

We focus now on the inter- and intra-valley scattering times \[\tau_i\] and \[\tau_0\] (also see schematic inset in Fig. 4(b)), which are related to the scattering processes that limit the mobility of our device (see discussion below). The inter-valley scattering time \[\tau_i\] as function of the carrier density \(n\) is shown in Fig. 4(a). In contrast to the phase-coherence time, \[\tau_i\] shows no clear dependence on the carrier density over a wide range of \(n\). This behavior is roughly consistent with the current understanding of inter-valley scattering processes in BLG which are caused by short-range scattering centers, such as lattice defects or ad-atoms. These can account for the large momentum transfer that is needed to scatter an electron from one valley to the other, and give rise to a density-independent inter-valley scattering time \[27\]. Approximating observed values of \[\tau_i\] with their weighted arithmetic mean \(\tau_i \approx 40\) ps (see dashed line in Fig. 4(a)), we can obtain an estimate for the density of resonant scatterers (impurities) in our sample \(n_i \approx m^*/(8\pi\tau_i) \approx 9 \times 10^7\) cm\(^{-2}\) \[27\] \[28\]. Taking into account the device geometry, this gives a total number of short-range scatterers of \(\approx 100\) which is a considerably low number compared to the total number of carbon atoms (roughly \(10^{10}\) for \(16 \times 7\mu m\)) and considering that the scatterers are most likely located at the edges of the BLG structure.

The intra-valley scattering time \[\tau_s\] is shown in Fig. 4(b). At low carrier density \(|n| < 0.4 \times 10^{12}\) cm\(^{-2}\) \[\tau_s\] is smaller than 0.1 ps, indicating that in this regime the second term on the right hand side of Eq. (1) is negligible and does not contribute to the overall shape of the WL dip, in agreement with previous findings \[15\].

At higher carrier densities, we obtain values of \[\tau_s \approx 5\] - 30 ps, which are anyhow at least one order of magnitude smaller than \[\tau_0\] and \[\tau_i\]. Here, due to the strong trigonal

\[
\Delta \sigma(B) = \frac{e^2}{\pi h} \left[ F \left( \frac{B}{B_0} \right) - F \left( \frac{B}{B_0+2B_i} \right) \right] + \frac{2e^2}{\pi h} F \left( \frac{B}{B_0+2B_i+B_s} \right) \quad (1)
\]

Here \(F(z) = \ln z + \Psi \left( \frac{1}{2} + \frac{1}{z} \right)\), where \(\Psi\) is the digamma function, and \(B_{0,i,s}\) is the gate voltage at which the mobility is maximum. The diffusion coefficient \(D\) is given by the relation \(D = h g_{\phi,i,s} / 4m^*\), where \(m^* = 0.033\) m\(_0\) is the effective carrier mass in bilayer graphene \[25\], and \(g_{\phi,i,s} = \sigma h / e^2\) is the dimensionless conductivity at \(B = 0\) T. The intra-valley scattering time \(\tau_s\) is approximately equal to \(m^*/(8\pi\tau_i) \approx 9 \times 10^7\) cm\(^{-2}\) \[27\] \[28\]. Taking into account the device geometry, this gives a total number of short-range scatterers of \(\approx 100\) which is a considerably low number compared to the total number of carbon atoms (roughly \(10^{10}\) for \(16 \times 7\mu m\)) and considering that the scatterers are most likely located at the edges of the BLG structure.

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warping effect in BLG [24], the intra-valley scattering time $\tau_i$ as extracted from WL measurements is not directly related to a chirality-breaking scattering process but is given by $\tau^{-1}_i = \tau^{-1}_m + \tau^{-1}_w$, where $\tau_w$ is the dominating trigonal-warping time and $\tau_m$ is the single-valley chirality-breaking time [24].

More insight on the role of inter- and intra-valley scattering processes as mobility-limiting factors can be obtained by looking at the corresponding characteristic length scales. In Fig. 4(c) we plot $L_\phi$, $L_i$ and $L_*$, which are related to the respective time scale by $L_{\phi,i,*} = \sqrt{D \tau_{\phi,i,*}}$ together with the mean free-path, $l_m = h\mu \sqrt{\pi n}/e$ (dashed line). The phase coherence length $L_\phi$ can be tuned up to 6 $\mu$m at sufficiently large densities, which, to the best of our knowledge, is significantly larger than all values previously reported in literature [15, 22, 23]. Most importantly, $L_\phi$ can be on the order of the sample width, making the presented BLG-hBN sandwich system interesting for future phase-coherent interference experiments. As for the phase-coherence time, the experimental values of $L_\phi$ are reasonably close to the upper bound for the dephasing length set by the electron-electron interaction according to AAK theory (solid line in Fig. 4(c)).

The inter-valley scattering length, $L_i$, in our sample (triangles in Fig. 4(c)) is about 0.4 $\mu$m at low carrier densities (roughly a factor 2 larger than $L_\phi$), and it increases up to 5 $\mu$m for larger $n$. In this regime $L_i$ exceeds $l_m$ roughly by one order of magnitude, ruling out inter-valley scattering due to short-range disorder as the mechanism limiting mobility in BLG. The intra-valley scattering length $L_*$ on the other hand, appears to be in quantitative agreement with $l_m$. The described observations clearly point at intra-valley scattering as the main limitation to mobility in high-quality BLG samples.

In literature, the main sources of intra-valley scattering in single- and bilayer-graphene have been associated with long-range disorder due to either charged impurities (Coulomb scatterers) [23, 51], or to local mechanical deformations, i.e. strain fluctuations [32]. However, differently from single-layer graphene and in agreement with earlier experiments on BLG samples [22, 23], we can exclude that the limitations to mobility come from Coulomb scatterers. This is a consequence from the simple fact that, if Coulomb scattering was the limiting mechanism, the conductivity would show a different dependence on the carrier density ($\sigma \sim n^\alpha$ and $1 < \alpha < 2$, where $\alpha$ is a density-dependent exponent [22]), than the linear behavior $\sigma \sim n$ reported in Fig. 1(c) and many other measurements [34, 35]. Vice versa, it can be shown that local strain fluctuations in BLG leads to the correct dependence $\sigma \sim n^2$ [36]. We can therefore conclude that the electron mobility in our sample is limited by intra-valley scattering events that are most likely caused by local strain fluctuations. This conclusion agrees well with evidence we have from confocal Raman experiments that high-mobility samples exhibit reduced local strain fluctuations [37], as well as with recent studies on single-layer graphene, which also identified mechanical deformations as the main source of mobility-limiting processes [23]. This in turn strongly suggests that the transport properties of both single- and bilayer graphene are hindered by the same physical mechanism.

In conclusion we performed low temperature transport measurements of high-mobility bilayer graphene encapsulated in hexagonal boron nitride. The high-quality of our device is proven by the full degeneracy-breaking of the zero Landau level at a magnetic field as low as $B = 6$ $T$. From weak localization measurements, we extract information on the dephasing time as well as the inter- and intra-valley scattering times and the corresponding characteristic length-scales. We observe phase-coherence times which are close to the limits imposed by electron-electron interaction, as well as phase-coherence lengthsls comparable with the sample size. Moreover, we can unambiguously conclude that intra-valley scattering rather than inter-valley scattering is the limiting mechanism for electron transport in BLG, and we discuss strain fluctuations as the most probable source of mobility-limiting scattering processes.

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