

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 τ_ϕ as well as the inter- and intravalley scattering times τ_i and τ_* , respectively. While τ_ϕ is in qualitative agreement with an electron-electron interaction-mediated dephasing mechanism, electron spin-flip scattering processes are limiting τ_ϕ at low temperatures. The analysis of τ_i and τ_* 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 [1]. In contrast to single-layer graphene, in BLG a band gap can be opened by an external electric field [2,3], making local depletion of the two-dimensional electron gas (2DEG) possible similar to III/V heterostructures. This is an important prerequisite for implementing state-of-the-art phase-coherent quantum device concepts [4,5]. In contrast to conventional 2DEGs, the massive Dirac fermion nature of the quasiparticles in BLG results in an unconventional quantum Hall effect [6,7] and promises unique quantum interference properties [8]. 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. Recent developments in device fabrication have shown that a significant improvement in sample quality can be obtained by replacing conventional SiO₂ with hexagonal boron nitride (hBN) [9]. This material provides an ultraflat substrate for graphene [9,10] 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 [11] or superlattice effects such as the Hofstadter butterfly [12–14]. However, despite these improvements, it remains difficult to experimentally address the microscopic mechanisms that limit carrier mobility and phase coherence in high-quality BLG.

To address these important questions, we present diffusive transport measurements on BLG fully encapsulated in hBN. Our fabrication technique allows us 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 [15]. From these measurements, we extract the inter- and intravalley scattering times, as well as the phase-coherence time. Our results indicate (i) that the main sources of dephasing in high-quality BLG are the electron-electron interaction as well as electron spin-flip scattering and (ii) that mobility is not limited by intervalley scattering processes. Moreover, we observe that the mean-free path quantitatively matches the intravalley scattering length over a wide range of carrier densities. Our findings point at intravalley scattering as the main limitation to mobility in BLG. We discuss local strain fluctuations as the possible source of these mobility-limiting scattering events.

Here, we present two-terminal measurements on bilayer graphene encapsulated in hBN; see the schematic in Fig. 1(a). The investigated device has length $L \approx 16 \mu\text{m}$ and width $W \approx 7 \mu\text{m}$; see Fig. 1(b). Detailed information of the sample fabrication, which is similar to Ref. [17], is given in Supplemental Material [18]. All measurements are performed in a He³ cryostat with a base temperature of $T = 300$ mK (unless stated otherwise). In Fig. 1(c), we show the conductivity of this device as a function of back-gate voltage V_g at $T = 77$ K. The charge neutrality point is 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.

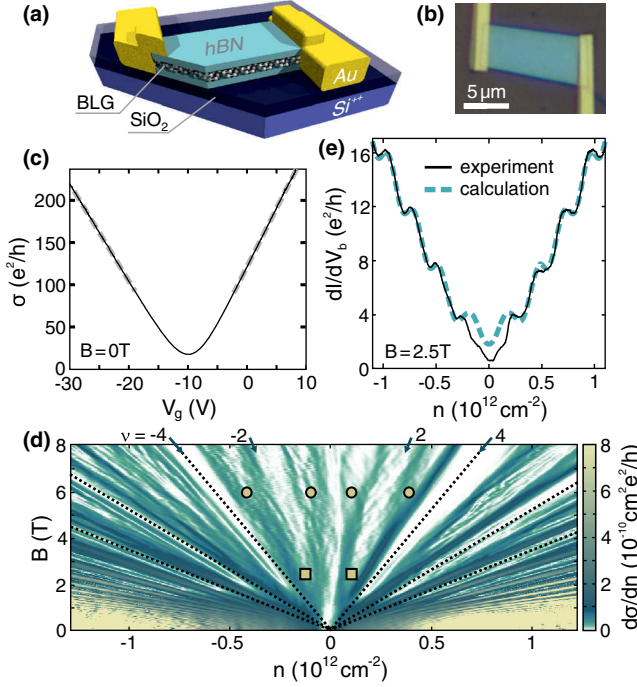


FIG. 1 (color online). (a) Illustration and (b) optical image of a contacted hBN-BLG-hBN heterostructure. (c) Conductivity σ vs V_g of the device shown in (b) at $T = 77$ K. Here we subtracted a contact resistance $R_C = 600 \, \Omega$ for electrons and $650 \, \Omega$ for holes by following Ref. [16]. For more details on R_C , please see Supplemental Material [18]. (d) Transconductivity $d\sigma/dn$ as a function of n and perpendicular B field. (e) The solid line shows dI/dV_b in dependence of n at $B = 2.5$ T. The result of a theoretical calculation is shown by the dashed line.

In Fig. 1(d), we show the transconductivity $d\sigma/dn$ as a function of the perpendicular B field and carrier density $n = \alpha \Delta V_g$. Dashed lines mark the filling factors $\nu = \pm 12, \pm 8, \pm 4$. The eightfold degenerate zero Landau level unambiguously confirms the bilayer nature of the investigated flake [7]. Additionally, we observe a degeneracy lifting into doubly degenerate Landau levels at $B \approx 2.5$ T and full degeneracy breaking at $B \approx 6$ T [see squares and dots in Fig. 1(d)], which is a direct signature of the high quality of our sample and consistent with Refs. [19,20]. In Fig. 1(e), we show the differential conductance dI/dV_b as a 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. [21] and using parameters discussed in Ref. [22]. The experimental data closely follow 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 explained either 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 agreement with the slope of the dashed lines in Fig. 1(d) as

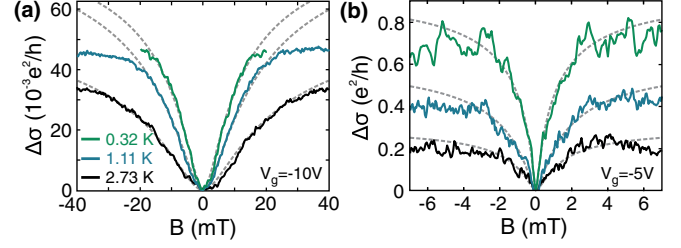


FIG. 2 (color online). Magnetoconductivity at low magnetic fields and different temperatures $T = 0.3, 1.1$, and 2.7 K for (a) $V_g = -10$ V (charge carrier density n close to the charge neutrality point) and (b) $V_g = -5$ V (corresponding to $n = 0.32 \times 10^{12} \, \text{cm}^{-2}$). Fits to the weak localization model for BLG are displayed by the dotted lines.

well as with a plate capacitor model with the SiO₂ and the bottom hBN flake as the gate dielectric.

We next focus on weak localization (WL) measurements, from which we extract three fundamental time scales of our device: the phase-coherence time τ_ϕ and the intervalley scattering time τ_i , as well as the intravalley scattering time τ_* . In Fig. 2, we show the experimentally observed WL dip, i.e., the change in conductivity at a finite magnetic field with respect to the one at $B = 0$ T: $\Delta\sigma(B) = \langle \sigma(n, B) - \sigma(n, B = 0) \rangle_{\Delta n}$. Figure 2(a) shows $\Delta\sigma(B)$ at three different temperatures 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 averaging 20 traces over a small density interval $\Delta n = 2.6 \times 10^9 \, \text{cm}^{-2}$. The resulting magnetoconductivity traces exhibit an increase with positive and negative B field followed by a saturation, which is consistent with previous WL experiments on BLG [15,23,24].

To obtain the time scales mentioned above, we fit to each trace the theoretical model for WL in BLG (see dashed lines in Fig. 2) derived by Kechedzhi *et al.* [25]:

$$\Delta\sigma(B) = \frac{e^2}{\pi h} \left[F\left(\frac{B}{B_\phi}\right) - F\left(\frac{B}{B_\phi + 2B_i}\right) \right] + \frac{2e^2}{\pi h} F\left(\frac{B}{B_\phi + B_i + B_*}\right). \quad (1)$$

Here $F(z) = \ln z + \Psi(\frac{1}{2} + \frac{1}{z})$, where Ψ is the digamma function, and $B_{\phi,i,*} = \hbar/(4eD\tau_{\phi,i,*})$. The diffusion coefficient D is given by the relation $D = \hbar g_\square/4m^*$, where $m^* = 0.033m_0$ is the effective carrier mass in BLG [26] and $g_\square = \sigma\hbar/e^2$ is the dimensionless conductivity at $B = 0$ T. We focus first on the behavior of the extracted phase-coherence time τ_ϕ . This time scale reaches a minimum of approximately 20 ps around the charge neutrality point [see Fig. 3(a)]. For larger carrier density, we observe a significant increase of τ_ϕ by over one order of magnitude up to $\tau_\phi \approx 240$ ps for $|n| = 0.8 \times 10^{12} \, \text{cm}^{-2}$. Such a gate-tunable phase-coherence time—also measured on a second sample

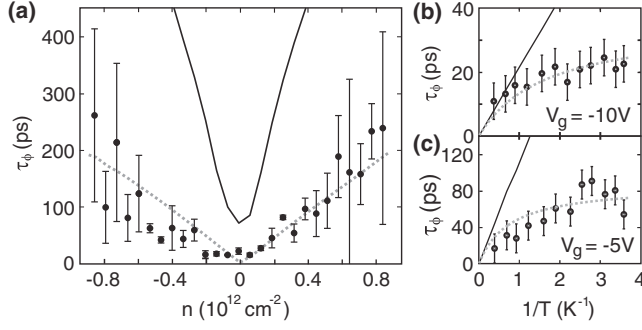


FIG. 3. (a) shows the n dependence of the phase-coherence time τ_ϕ at $T = 300$ mK. Here, the black line displays the theoretical result by AAK [27]. The dashed line highlights the result modified due to a finite electron spin-flip scattering time $\tau_{sf} = \beta|n|$ with $\beta = 3 \times 10^{-22}$ cm² s. (b) and (c) display the temperature dependence of τ_ϕ for carrier densities n (b) very close to the charge neutrality point ($V_g = -10$ V) and (c) at $n = 3.2 \times 10^{11}$ cm⁻² ($V_g = -5$ V). The solid and dashed lines resemble the same theoretical models as in (a). From (c), we extract $\tau_{sf} = 88$ ps in good agreement with the fit in (a) from which we obtain $\tau_{sf} = 96$ ps for $n = 3.2 \times 10^{11}$ cm⁻². All error bars are extracted from multiple measurements of each data point.

(see Supplemental Material [18])—has not yet been observed for single- and bilayer graphene. However, it is in qualitative agreement with a scattering mechanism based on electron-electron interactions as predicted by Altshuler-Aronov-Khmelnitsky (AAK) for a two-dimensional system: $\tau_\phi^{-1} = k_B T \ln g_\square / (\hbar g_\square)$ [27], where k_B is the Boltzmann constant and $g_\square$ is the dimensionless conductivity (introduced above), which can be directly extracted from the measured conductivity. Hence, without any adjustable parameter, we obtain the solid line in Fig. 3(a). These values are a factor of 3–4 larger than the values of τ_ϕ extracted from our WL measurements, meaning that there must be an additional source for dephasing. This becomes even more apparent when investigating the temperature dependence of the extracted τ_ϕ at different carrier densities, as shown in Figs. 3(b) and 3(c). Similarly to Fig. 3(a), the solid line illustrates the estimates for τ_ϕ obtained from the AAK theory ($\tau_\phi^{-1} \sim T$). Above $T = 400$ mK (below $1/T = 2.5$ K⁻¹), the experimentally extracted τ_ϕ is inversely proportional to the temperature. However, at lower temperatures, τ_ϕ shows a saturation behavior. To account for these discrepancies, we follow Ref. [28] and include an additional inelastic electron spin-flip scattering time τ_{sf} , leading to an overall scattering rate which is the sum of the spin flip and the AAK decoherence rate, $\tau_\phi^{-1} = \tau_{sf}^{-1} + k_B T \ln g_\square / (\hbar g_\square)$. We use this expression to estimate τ_{sf} and show that by assuming a linear carrier density dependency ($\tau_{sf} \sim |n|$) we obtain good agreement with all our experimental data [see dashed lines in Figs. 3(a)–3(c)]. The extracted τ_{sf} values are on the order of 100 ps (see Fig. S4 of Supplemental Material [18]), which is in agreement with earlier transport studies on graphene, where magnetic impurities were identified as a dominant

phase scattering source [29]. Although the apparent n dependence of τ_{sf} is in contradiction to recent studies of spin scattering in single-layer graphene [30], it may well be in agreement with resonant spin-flip scattering in BLG [31].

We focus now on the inter- and intravalley scattering times τ_i and τ_* [see the inset in Fig. 4(b)], which are related to the scattering processes that limit the mobility of our device (see the discussion below). The intervalley scattering time τ_i as a function of n is shown in Fig. 4(a). In contrast to the phase-coherence time, τ_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 intervalley scattering processes in BLG which are caused by short-range scattering centers, such as lattice defects or adatoms. 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 intervalley scattering time [32]. Approximating observed values of τ_i with their weighted arithmetic mean $\tau_i \approx 40$ ps [see the dashed line in Fig. 4(a)], we can obtain an estimate for the density of resonant scatterers (impurities) in our sample of $n_i = m^*/(8\hbar\tau_i) \approx 9 \times 10^7$ cm⁻² [32,33]. Taking into account the device geometry, this gives a total number of short-range scatterers of ≈ 100 , which is a considerably low number compared to the total number of carbon atoms (roughly 10^{10} for 16×7 μ m) and considering that the scatterers are most likely located at the edges of the BLG structure.

The intravalley scattering time τ_* is shown in Fig. 4(b). At low carrier density ($|n| < 0.4 \times 10^{12}$ cm⁻²), τ_* 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 n , we obtain values of $\tau_* = 5$ –30 ps, which are at least one order of magnitude smaller than τ_ϕ and τ_i . Here, due to the strong trigonal warping effect in BLG, the intravalley scattering time τ_* as extracted from WL measurements is not directly related to a chirality-breaking scattering process but is given by $\tau_*^{-1} = \tau_w^{-1} + \tau_z^{-1}$, where τ_w is the dominating

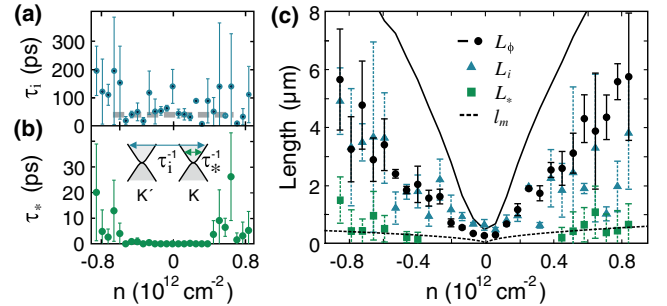


FIG. 4 (color online). (a) and (b) show the n dependence of the intervalley scattering time τ_i and intravalley scattering time τ_* , respectively. (c) illustrates the corresponding length scales L_ϕ , L_i , and L_* and the mean-free path l_m (dashed line). The result of AAK is highlighted by the black solid line.

trigonal-warping time and τ_z is the single-valley chirality-breaking time [25].

More insight on the role of inter- and intravalley scattering processes as mobility-limiting factors can be obtained by looking at the corresponding characteristic length scales. In Fig. 4(c), we plot L_ϕ , 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, $l_m = \hbar\mu\sqrt{\pi n}/e$ (dashed line). The phase-coherence length L_ϕ can be tuned up to 6 μm at sufficiently large densities, which, to the best of our knowledge, is significantly larger than all values previously reported in the literature [15,23,24]. Most importantly, L_ϕ 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_ϕ 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)], but limited by the spin-flip scattering length $L_{\text{sf}} = \sqrt{D\tau_{\text{sf}}}$, which is on the same order.

The intervalley scattering length L_i in our sample [triangles in Fig. 4(c)] is about 0.4 μm at low carrier densities (roughly a factor of 2 larger than L_ϕ), and it increases up to 5 μm for larger n . In this regime, L_i exceeds l_m by roughly one order of magnitude, ruling out intervalley scattering as the mechanism limiting mobility in BLG. This observation clearly points at intravalley scattering (being the only alternative) as the mobility-limiting process in high-quality BLG samples. Additionally, we find that the intravalley scattering length L_* (which is determined by both the trigonal warping effect and the single-valley chirality breaking time) is roughly one order of magnitude lower than L_i , but similar to l_m .

In the literature, the main sources of intravalley scattering have been associated with long-range disorder due either to charged impurities (Coulomb scatterers) [34–36] or to local strain fluctuations [37]. However, differently from single-layer graphene and in agreement with earlier experiments on BLG samples [38], 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 α is a density-dependent exponent [39,40]) than the linear behavior $\sigma \sim n$ reported in Fig. 1(c) and other measurements [38,41]. Vice versa, it can be shown that local strain fluctuations in BLG lead to the correct dependence $\sigma \sim n$ [42]. We therefore conclude that the electron mobility in our sample is limited by intravalley scattering events that are most likely caused by local strain fluctuations. This conclusion agrees with evidence we have from Raman experiments that high-mobility samples exhibit reduced strain fluctuations [43], as well as with recent studies on single-layer graphene, which also

identified mechanical deformations as the main source of limiting mobility [44]. This in turn strongly suggests that the transport properties of both single- and bilayer graphene are hindered by the same physical mechanism. To our knowledge, random strain fluctuations are most likely introduced by sample fabrication, where mechanical exfoliation, predeposition on rough SiO_2 , graphene-substrate interactions, and different thermal expansion coefficients may give rise to local mechanical deformations. However, further work is needed to understand the detailed mechanisms.

In conclusion, we performed transport measurements on high-mobility bilayer graphene encapsulated in hBN. From WL measurements, we extract information on the dephasing time as well as the inter- and intravalley scattering times and the corresponding characteristic length scales. We observe phase-coherence lengths comparable with the sample size as well as phase-coherence times close to the values imposed by electron-electron interaction but limited by spin-flip scattering at low temperatures. Surprisingly, this spin-flip scattering time is more than an order of magnitude lower than observed in nonlocal spin-value measurements [45]. Moreover, we can unambiguously conclude that intravalley scattering rather than intervalley 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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