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Linear magnetoresistance in compensated graphene bilayer

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We report a nonsaturating linear magnetoresistance in charge-compensated bilayer graphene in a temperature range from 1.5 to 150 K. The observed linear magnetoresistance disappears away from charge neutrality ruling out the traditional explanation of the effect in terms of the classical random resistor network model. We show that experimental results qualitatively agree with a phenomenological two-fluid model taking into account electron-hole recombination and finite-size sample geometry.

Classical magnetoresistance is a perfect tool for experimental studies of multicomponent electronic systems where conventional theory of electronic transport predicts a quadratic dependence of the resistance on the weak applied magnetic field followed by a saturation in classically strong fields. While most materials do exhibit the quadratic behavior there is a fast growing number of experiments reporting observations of linear magnetoresistance (LMR) in a wide variety of novel materials including multilayer graphene15, topological insulators24,25, Dirac16–19 and Weyl20–22 semimetals, transition-metal dichalcogenides23 as well as in narrow-gap semiconductors22 and three-dimensional (3D) silver chalcogenides23–25.

Semiclassical linear magnetoresistance has been predicted for 3D metallic slabs with complex Fermi surfaces and smooth boundaries22,24, for strongly inhomogeneous, granular materials25, and for compensated two-component systems with quasiparticle recombination29. Purely quantum effects (and screening of charged impurities) lead to LMR in zero-gap band systems with linear dispersion in the case where all carriers belong to the first Landau level20,21. In weak fields, quantum interference in two-dimensional electron systems yields an interaction correction33 to resistivity that is linear in the Zeeman magnetic field.

The extreme quantum limit of Refs. 30–33 has been realized in graphene26 and in Bi2Se3 nanosheets27. The quantum theory was also reported28 to be applicable to the novel topological material LuPdBi. The classical theory of Ref. 28 was recently used to interpret the behavior of hydrogen-intercalated epitaxial bilayer graphene1. It was argued that large samples of epitaxial bilayer graphene contain a “built-in mosaic tiling” due to the dense dislocation networks31, making it an ideal material to realize the random network model of Ref. 29. At the same time, neither theory can explain LMR in homogeneous topological insulators35 and neutral two-component systems21,23.

In this paper we report results of a systematic experimental analysis of magnetotransport in exfoliated bilayer graphene. Precisely at charge neutrality, we have observed nonsaturating LMR in a wide range of magnetic fields in Hall bars of widths 0.5, 0.95, and 2.0 µm in a temperature range from 1.5 to 150 K. Deviations from charge neutrality lead to eventual saturation of the magnetoresistance. Our key experimental findings are not accounted for within the random resistor network model. Indeed, this model is insensitive to the relative concentration of different types of charge carriers and thus cannot explain the observed saturation of the magnetoresistance away from charge neutrality. This model also does not explain the transition between the quadratic dependence at very weak magnetic fields and LMR observed at higher fields35. The extreme quantum limit is unlikely to be reached in our system at 150 K for both electrons and holes31. Moreover, the excitation spectrum in bilayer graphene is quadratic, which rules out the quantum theory of Refs. 30–31.

We are able to explain our results in terms of a semiclassical description of finite-size, charge-compensated two-component systems in moderately strong, classical magnetic fields36. The key element of the physical picture of Ref. 29 is the electron-hole recombination29. When external magnetic field is applied, recombination processes allow for a neutral quasiparticle flow in the lateral direction relative to the electric current37. Although such neutral current cannot be directly detected in our measurements, its presence leads to redistribution of charge carriers over the sample area influencing the nonuniform profile of the electric current in the sample. As a result, the sample is essentially split into the bulk and edge regions, which contribute to the total sheet resistance of the sample as parallel resistors. The bulk and edge resistances exhibit qualitatively different dependence on the magnetic field yielding LMR. Away from charge neutrality a nonzero Hall voltage is formed leading to the observed saturation of the magnetoresistance.
I. EXPERIMENTAL DETAILS AND SAMPLE CHARACTERIZATION

We have prepared the sample by placing the exfoliated bilayer graphene sheet on the substrate consisting of a highly doped Si wafer covered by a 330nm-thick SiO₂ film. Subsequently, the sample was patterned into a triple Hall bar device, see Fig. 1(b) for an atomic force microscope (AFM) image. The sample consists of three sections 2, 0.95, and 0.5 μm wide. The length of each Hall bar is 1.8 μm. The sample was purified using an AFM tip (instead of annealing) which allowed us to decrease the concentration of the charged impurities on top of graphene considerably. The carrier concentration n in the sample can be varied up to 5 × 10¹² cm⁻² by applying a gate voltage V₉ to the conducting substrate, which acts as a back gate.

Magnetotransport was studied by four-probe method with simultaneous measurements of longitudinal Rₓₓ and transverse Rₓᵧ resistances in perpendicular magnetic fields from 0 to 7 T and in a temperature range from 1.5 to 150 K passing an ac current with an amplitude of 10 μA through the sample.

To characterize the sample and to define the charge neutrality point (CNP), the field effect (FE) was measured for each section of the device. Figure 1(a) shows the FE dependences measured at B = 0 T and T = 25 K for the three sections of the device. All three sections exhibit a graphene typical FE with a sharp maximum corresponding to the CNP. The precise value of V₉ cor-
responding to CNP depends on the Hall bar width and is shifted from 0.8 V in the widest section of the sample toward 3.6 V in the medium and 10.4 V in the narrowest Hall bar. The maximum resistivity in the widest and middle sections is 5.6 kΩ while exceeding 6.2 kΩ for the narrowest section.

The electron and hole mobilities were estimated from the conductivity at 25 K using the one-band model (see Fig. 1(c)). The electron and hole densities necessary for this estimate were obtained from the measured Shubnikov-de Haas oscillations at low temperatures. The resulting mobilities increase with the width of the sample; we have obtained the following values for the mobilities of the narrowest, medium, and widest sections of the sample far away from charge neutrality: 2200, 3000, and 3800 cm²/Vs for holes and 2600, 3000, and 3800 cm²/Vs for electrons.

The dependence of the mobility on the width of the sample is attributed to scattering of carriers on the sample edges and is described in Ref. 38. Although the above mobilities are not very high, the samples are of a good quality having a clear manifestation of CNP and exhibiting the quantum Hall effect (see Fig. 1(d)). Measurements of the Hall resistance in the wide section of the sample at 1.5 K in relatively high magnetic field 9.5 T (Fig. 1(d)) demonstrate the features inherent to bilayer graphene following from filling factors in the Hall plateaus equal to ν = ±4, ±8, ±12. In strong magnetic field the neutrality point is shifted towards higher gate voltages, see Figs. 1(e) and 2. For the wide section of the sample at 12 T, CNP corresponds to V₉ = 7 V. This effect has also been observed in other sections of the sample.
II. LINEAR MAGNETORESISTANCE

We have measured the longitudinal resistance for all three sections of the sample and the Hall resistance between widest and medium sections in the interval of gate voltages from −20 to 32.2 V with the step δVg = 2.4 V that includes CNP for all three sections. The data for the wide section of the sample at T = 1.5K are shown in Fig. 2(a). To reduce the conductance fluctuations, further measurements were performed at higher temperatures: 25, 50, 100, and 150 K. At such high temperatures quantum effects, e.g. Landau quantization, are not detectable.

The magnetoresistance data for the thin section of the sample at the four temperatures are shown in Fig. 3. The data show linear behavior close to the neutrality point (the green curve corresponding to the gate voltage Vg = 10.6 V). Away from neutrality, the data show linear behavior for an intermediate range of magnetic fields followed by a saturation at stronger fields. Similar results were obtained for the other two sections of the sample. At the same time, the Hall resistance grows in amplitude near charge neutrality. Finally, in contrast to the theoretical predictions of Ref. 29, see Eqs. (1), with the parameters given in Table I, the experimental data; the dashed (blue) line represents the the-}

| TABLE I: Microscopic parameters obtained from analyzing the experimental data with the theory (1) for the three sections of the sample, see Figs. 1 and 2. |
|---------------------------------|----------------|----------------|----------------|
| W                              | µ               | ℓ<sub>0</sub>  |
| narrow                         | 0.52μm          | 0.43μm         |
| medium                         | 0.95μm          | 0.35m<sup>2</sup>/Vs |
| wide                           | 2μm             | 0.79μm         |
|                                | 0.42m<sup>2</sup>/Vs | 1.2μm         |
challenge for an analytic theory. Nevertheless, we may attempt to analyze the measured data with the help of the existing theory of Ref. 29. The simplest version of this theory (applicable to a particle-hole symmetric system with parabolic dispersion and energy-independent impurity scattering rate) yields the following expressions for the longitudinal and Hall resistivities of a two-component system near charge neutrality:

$$R_{xx} = R_0 \frac{1 + \mu^2 B^2}{1 + \mu^2 B^2 \left[ \frac{\tanh(W/\ell_R)}{W/\ell_R} \right] \left( 1 - \frac{n^2}{\rho^2} \right) + \frac{n^2}{\rho^2}}. \quad (1a)$$

$$R_{xy} = \frac{R_0}{\rho} \frac{(1 + \mu^2 B^2) \mu B}{1 + \mu^2 B^2 \left[ \frac{\tanh(W/\ell_R)}{W/\ell_R} \right] \left( 1 - \frac{n^2}{\rho^2} \right) + \frac{n^2}{\rho^2}}. \quad (1b)$$

Here $n$ and $\rho$ are the charge and quasiparticle densities, $\mu$ is the mobility (which is assumed to be the same for both electrons and holes), $W$ is the sample width, $R_0$ is the zero-field resistivity, and $\ell_R$ is the field-dependent recombination length. Assuming that the dominant recombination process is the impurity-assisted electron-phonon coupling that can occur anywhere in the sample with equal probability, the recombination length found in Ref. 29 is given by

$$\ell_R = \frac{\ell_0}{\sqrt{1 + \mu^2 B^2}}; \quad \ell_0 = 2 D \tau_R,$$

where $D$ is the diffusion coefficient and $\tau_R$ is the recombination time in zero magnetic field. As a result, in classically strong fields ($\mu B \gg 1$) and for $W \gg \ell_R$ the magnetoresistance (1a) close to the charge neutrality point is linear, $R_{xx} \approx R_0 W \mu B/\ell_0$.

Using the measured parameters of our sample in the above expressions, we find that the theory predicts a magnetoresistance that is stronger than what is actually observed in our experiment. However, our results can be quantitatively described by Eqs. (1a) and (1b) if we introduce an empiric expression for the recombination length

$$\ell_R = \frac{\ell_0}{\sqrt{1 + \mu^2 B^2}} \quad (1c)$$

with $\mu < \mu$. This modification turns out to effectively account for the following issues: (i) electron-hole asymmetry, (ii) energy dependence of the electron-hole recombination length and mobility, and (iii) spatial inhomogeneity of the sample. In Figs. 4 and 5 we used $\mu \approx 0.5 \mu$.

Electron-hole asymmetry manifests itself in the nonzero Hall resistivity at charge neutrality. Moreover, for any value of the carrier density the Hall resistivity is a nonmonotonous function of the magnetic field. As mentioned above, this effect also leads to the apparent drift of the charge neutrality point (in terms of the applied gate voltage) with the external magnetic field.

At temperatures lower than the Debye energy the dominant recombination process involves electrons and holes near the bottom of the band. Indeed, far away from the neutrality point, kinematic constraints preclude the “direct” process where an electron from the upper band is scattered into an empty state in the lower band by means of single acoustic phonon emission. Instead, such “hot” electrons require an additional scatterer (e.g. an impurity or a second phonon) for recombination to take place. In contrast, electrons close to the neutrality point in bilayer graphene are slow enough so that the direct, single-phonon recombination is allowed. Hence, within the kinetic equation approach the effective length scale describing the recombination processes depends on energy. Similarly, the impurity scattering time or carrier mobility is strictly speaking energy-dependent as well. Now, the macroscopic description of Ref. 29 involves quantities that are averaged over the quasiparticle spectrum. Taking into account the existence of the several distinct recombination processes, we arrive at the conclusion that after thermal averaging, the typical recombination length $\ell_R$ may be described by slightly different effective parameters as compared to, e.g., Drude conductivity.
The width dependence of the carrier mobility indicates that the edge region of the sample is characterized by stronger scattering. As a result, all parameters describing electronic transport acquire an effective coordinate dependence across the sample. Since in strong magnetic fields the current is mostly flowing near the sample edges\cite{29}, we expect that the effective recombination length $\ell_R$ is determined by the lower mobility typical of the near-edge region.

Theoretical results shown in Figs. 4 and 5 were obtained by using expressions (1) with the parameters listed in Table I. The theory \( \mu = 1 \) assumes that electrons and holes have the same mobility. While plotting Figs. 4 and 5 we have treated the mobility as a free parameter instead of using the values quoted in Sec. 1 (see Fig. 1(a)) since experimentally one can reliably determine mobility only far away from the neutrality point, where the classical Hall resistivity exhibits the standard behavior $R_{xy} = B/(ne)$. Close to charge neutrality, the mobility may deviate from such experimental values due to electron-hole interaction processes similar to the drag effect. Far away from charge neutrality this interaction is ineffective since with exponential accuracy only one band is partially filled and contributes to low-energy physics. In contrast, close to the neutrality point, both electrons and holes participate in transport and hence one has to take into account their mutual scattering. The resulting change of the mobility does not exceed 50\% in accordance to theoretical expectations.

The values of the recombination length $\ell_0$ shown in Table I show significant dependence on the sample width (roughly, $\ell_0 \sim W$). We interpret this observation as an indication of a much larger recombination length that would characterize a very large (in theory – infinite) sample (if it were possible to fabricate without strong structural disorder\cite{4,34}). Assuming that the electron-hole recombination is dominated by electron-phonon interaction (either impurity- or edge-assisted), we argue that in narrow samples the phonon spectrum is modified (compared to an idealized infinite system), leading to a much shorter recombination length of the order of the sample width.

The Hall resistance \( R_{xy} \) is expected to vanish at the neutrality point. However, as we have already mentioned, in our sample the neutrality point shifts toward higher gate voltages when a strong magnetic field is applied. In order to account for this effect, we have used the ratio $R_{xy}/R_{xx} = \mu B n / \rho$ to extract the field-dependent quantity $\mu B n / \rho$ from the experimental data. Using thus obtained dependence in Eq. (1b), we find good quantitative agreement between the calculated and measured values, see Fig. 4. At the same time, the longitudinal resistivity \( \rho \) is much less sensitive to small deviations of density. Using the extracted values of $\mu B n / \rho$ in Eq. (1b) does not lead to visible changes in the calculated curve shown in Fig. 4.

The shift of CNP with magnetic field was observed directly, see Figs. 1 and 2. Assuming that the maximum of the longitudinal resistivity corresponds to CNP, we can extract the field dependence of the chemical potential (and hence, carrier densities) from the data. Using thus obtained dependence, we recalculated the Hall resistance, see the brown curve in Fig. 1(b). The result shows reasonable agreement with the data, with the visible deviations may stem from the mismatch of temperatures in the two data sets in Figs. 2 and 3 (\( T = 1.5 K \) and $T = 150 K$, respectively).

Finally, away from the neutrality point the data shows a tendency towards saturation in high magnetic fields, see Fig. 3(b). The theoretical fits where performed with a set of parameters depending on the gate voltage and taking into account the shift of CNP with magnetic field. In particular, the mobility appeared to show a slight increase from 0.35$m^2$/Vs (close to CNP, see Table I) to 0.44$m^2$/Vs at \( V_g = -3.8 V \). At the same time, in that range of gate voltages the recombination length $\ell_0$ appears to be almost unchanged from the value shown in Table I.

III. CONCLUSIONS

In this paper we reported the experimental observation of linear magnetoresistance in narrow bilayer graphene samples. The observed behavior is in good qualitative agreement with the two-fluid model of Ref. 29. The observed effect is specific to the charge neutrality point. Away from neutrality the magnetoresistance shows an approximate linear behavior only in a limited intermediate range of magnetic fields followed by a tendency to saturation. Our observations are incompatible with the quantum theory of Refs. 30,31 and with the random resistor network model of Ref. 28, but are accounted for in the semiclassical theory of two-component compensated systems of Ref. 29.

Using an empirical modification of the simplest theoretical model \( \mu = 1 \), we were able to describe our data in a quantitative fashion. A microscopic theory accounting for the physics that is beyond the simplest version of the two-fluid model of Ref. 29 should be based on the quantum kinetic equation\cite{41,43}. Further aspects of the phenomenon of linear magnetoresistance are the subject of future experimental work, especially in numerous novel materials.

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In wide samples of graphene monolayers a square-root magnetoresistance has been observed in Ref. 44 near the Dirac point, resulting from the combination of linear dispersion of carriers and short-range character of scatterers\cite{44}.

In Ref. 44, the authors observed a square-root magnetoresistance near the Dirac point in graphene monolayers. This behavior is a result of the linear dispersion of carriers and the short-range character of scatterers in wide samples of graphene.

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