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Taming the magnetoresistance anomaly in graphite

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ABSTRACT

At low temperatures, graphite presents a magnetoresistance anomaly which manifests as a transition to a high-resistance state (HRS) above a certain critical magnetic field B_c. Such HRS is currently attributed to a c-axis charge-density-wave taking place only when the lowest Landau level is populated. By controlling the charge carrier concentration of a gated sample through its charge neutrality level (CNL), we were able to experimentally modulate the HRS in graphite for the first time. We demonstrate that the HRS is triggered both when electrons and holes are the majority carriers but is attenuated near the CNL. Taking screening into account, our results indicate that the HRS possess a strong in-plane component and can occur below the quantum limit, being at odds with the current understanding of the phenomenon. We also report the effect of sample thickness on the HRS.

1. Introduction

Graphite is a quasi-compensated semimetal in which charge carriers possess high electronic mobility and low effective masses [1]. These allow the material to reach the quantum limit at modest values of $B \approx 7$ T (the smallest magnetic field for which only the lowest energy Landau level (LL) remains populated) [2]. At higher magnetic fields however, graphite hints at some exotic properties, such as the occurrence of the fractional quantum Hall effect, the possibility of magnetic-field-induced superconductivity and the existence of a magnetic-field-induced high resistance state (HRS) [3–5]. The latter has been thoroughly investigated since its first experimental observation in the 1980's, and still sparks off debate to date [5–10]. It manifests as a single or multiple sharp bump(s) of the sample resistance as a function of magnetic field, usually triggered at $B > 25$ T [6–10].

Much experimental work has been devoted to verify the origin of this state (see Ref. [11] for a review). Despite earlier reports suggesting that the HRS is an in-plane phenomenon, current consensus is that it is triggered along the c-axis direction at the lowest Landau level in graphite [6,12]. Early theoretical attempts by Yoshioka and Fukuyama invoked the surging of a charge-density-wave (CDW) transition along the sample c-axis, caused by a 3D to 1D dimensionality reduction due to the quantum limit [13]. In this context, the occurrence and subsequent suppression of the CDW state have been attributed to the crossing between the Fermi level and the lowest Landau spin subbands at increasing magnetic fields.

Albeit it is widely accepted that the effect is caused by electron-electron interactions, experimentalists still struggle to verify the physical mechanisms responsible for the HRS. Currently, reports support different hypothesis, which include the formation of a CDW, a spin-density wave or the opening of an excitonic gap. All expected to occur along graphite's c-axis at the lowest Landau level [9,10,14–16].

Different approaches to understand the nature of the HRS have been attempted in the past decades, highlighting various aspects of the phenomenon. For example, early angle-resolved measurements by Timp et al. showcased the c-axis component of magnetic field as the sole responsible for the HRS, supporting the presence of a CDW in graphite. Similar experiments recently performed by Zhu et al. however, showed a suppression of the HRS with tilting angle, weighting in favour of an excitonic gap taking place [6,17]. As another example, although earlier models ascribed the HRS to a CDW triggered by a single spin subband in the lowest Landau level of graphite [13], recent measurements by Fauque et al. at $B = 80$ T demonstrated a re-entrant behavior, suggesting its triggering by more than one spin subband [10].

Although such works provide tantalizing hints of the HRS origin, current experimental data does not allow for an indisputable implication of the bands and carriers responsible for the phenomenon. One solution for this problem would be to monitor the

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HRS for doped graphite samples. In doing so, one could actively depopulate different spin subbands of electrons and holes by changing the sample's chemical potential. To our best knowledge, attempts in this topic are currently carried out by performing ionic implantation in bulk graphite or testing different sample qualities. These approaches do not allow one to reliably separate the effects caused by induced disorder from the ones caused by the intended doping [11,14]. This becomes critical when comparing different samples, as differences on disorder and native charge carrier concentration can affect the phenomenon under consideration, causing seemingly similar experiments to produce potentially diverging conclusions.

In order to address this issue, in the present work, we verify the behavior of the HRS in mesoscopic graphite by controlling the material's chemical potential (i.e. the position of the Fermi level within the band structure) by gating our device through its Charge Neutrality Level (CNL). In doing so, we were able to electrostatically dope the sample without modifying other critical parameters (such as cristalinity, geometry and quality of contacts). Our results provide the first experimental observation that the properties of the HRS change non-monotonically with graphite's averaged charge carrier density. We observe that the HRS survives outside the quantum limit and is triggered by both electrons and holes, approximately symmetric to each other with respect to the CNL. By accounting for charge screening we also infer a strong two-dimensional character of the HRS, although an off-plane degree of freedom seems necessary for the phenomenon. Our results shed new light on the subject, suggesting that the HRS in graphite has a large in-plane component and might take place at Landau levels with *n*>0.

2. Results and discussion

The experiments shown here were carried out in mesoscopic Highly Oriented Pyrolytic Graphite (HOPG) exfoliated from a bulk crystal with mosaicity of 0.30^o [18] - the Full width at Half Maxima (FWHM) extracted from x-ray rocking-curve measurements. The device had approximate in-plane dimensions of 5 μm × 5 μm (see Fig. 1) and a thickness of 35 nm. The sample was deposited atop a 30 nm-thick BN crystal previously placed on a N-dopped Si substrate coated with 300 nm of SiO_2. The sample was contacted with electron-beam lithography for longitudinal and Hall measurements. A backgate voltage in the range $-30\,V \le V_g \le +30V$ was used to modulate its charge carrier concentration.

Magnetoresistance measurements were carried out at $T = 4.2$ K with pulsed magnetic fields up to $B = 55$ T. To avoid thermal stress, the sample was kept at constant temperature between measurements. The longitudinal magneto-resistance (MR) was positive at low magnetic fields and reached a broad maximum at B_{max}, which was followed by a region of negative slope. The main features of the measurements were reproducible during the increase and decrease of the magnetic field pulse. Hall resistance curves (presented in the suppl. Material [19]) showed a pronounced non-linear behavior for all values of V_g, tending towards zero at large B. These results are qualitatively similar to observations in macroscopic graphite [9,14]. Unfortunately, such behavior cannot allow for a reliable determination of the sample charge carrier concentration, as the simplest fitting of the Hall curves would rely on the two fluid model for which at least four independent parameters (carrier density and mobility for electrons and holes) are necessary [20]. The fitting procedure further gains in complexity in the realistic cases of magnetic-field-dependent mobility, occurrence of partial charge screening or when additional sub-band contributions are considered.

In our sample, the HRS shows a critical field $B_c > 38$ T. This corresponds to the magnetic field for which $R_{xx}(B)$ deviates from its smooth background, as indicated in Fig. 2a. Such state is strongly influenced by the backgate voltage, showing a variation of its relative intensity and a non-monotonic shift of B_c with V_g. In order to correlate such changes with the alteration of the charge carrier concentration in the material, we analyzed the Shubnikov de Haas (SdH) oscillations present in all measurements. To determine their frequencies, we employed the method used in Refs. [21,22]. Unfortunately, this method only yields the value of the dominant SdH frequency (f_{SDH}), specially if other components have a much lower intensity. The obtained f_{SDH} reached up to 75 T when varying V_g between -30 V and $+15$ V, as shown in Fig. 3a (the quantum oscillations are shown in the suppl. material [19]). Such f_{SDH} is tenfold higher than values typically found in the literature for pristine graphite (which ranges between 4.5 T and 7 T) [23]. We did not observe sharp variations of the oscillations's Berry phase with V_g (see the suppl. material [19]).

Our results show a linear dependency between f_{SDH} and V_g for $V_g < 15$ V, characterizing the sample as a quasi-2D system [24]. For $V_g > +15$ V, f_{SDH} became almost constant with V_g, presumably due to enhanced Coulomb screening caused by an excess of charge carriers. A linear extrapolation of the data for $V_g < 15$ V found $f_{SDH} = 3.3|V_g + 7.6|$ (f_{SDH} given in Tesla and V_g in volts), suggesting that compensation in our device (equivalent electron and hole population) occurs around $V_{CNL} = -7.6$ V. This is shown in Fig. 3a. At this voltage, B_c shows a local maximum, presenting minima at the nearly symmetrical values with respect to V_{CNL} $V_g \approx -15$ V and $V_g \approx +5$ V. At these values we also observed an increase of the relative intensity of the HRS ($\Delta R_{HRS}/R(B_{max})$, see Fig. 3b), followed by its reduction at larger doping. This observation is highlighted in the colormap of Fig. 2b, which shows two local maxima above 40 T caused by the HRS (marked by dashed circles), quasi-symmetric with respect to the CNL.

We can account for the occurrence of charge screening in our device by associating the modulation of f_{SDH} to a change of the tree-dimensional charge carrier density (electrons or holes) η_{3D} in the sample. For this, we use a simplified model for a quasi-2D electron gas

$$\eta_{3D} = 2e\frac{S}{(2\pi)^2}\frac{1}{c_0}\alpha. \quad (1)$$

In it, *e* is the electronic charge, *S* is the 2-dimensional in-plane Fermi surface cross-section, α is a constant related to the dimensionality of the material and $c_0 = 0.335$ nm the interlayer distance in graphite. Under the assumption of decoupled graphene layers, $\alpha = 1$, whereas if one considers the existence of a closed Fermi

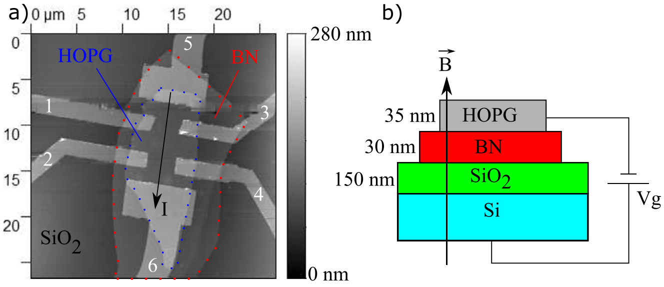

Fig. 1. a) AFM image of the HOPG sample studied here. The blue (red) dots mark the boundaries of the HOPG (BN) in the device. The numbers on the figure are used to identify the contacts. The electrical current is applied between contacts 5 and 6. R_{xx} is measured between contacts 1 and 2 and R_{xy} between contacts 2 and 4. b) Cross-section schematic of the device, showing how the backgate voltage was applied and the direction of the magnetic field. (A colour version of this figure can be viewed online.)