QUANTUM HALL EFFECT IN HIGHLY ORIENTED PYROLYTIC GRAPHITE

Heiko Kempa[1], Pablo Esquinazi[1] and Yakov Kopelevich[2]
[1] *Superconductivity and Magnetism Division, Universität Leipzig, Linnéstrasse 5 D-04103 Leipzig, Germany*
[2] *Instituto de Física, Universidade Estadual de Campinas, Unicamp 13083-970, Campinas, Sao Paulo, Brasil*

Corresponding author e-mail address:esquin@physik.uni-leipzig.de

Introduction

The occurrence of the quantum-Hall-effect (QHE) is not restricted to perfectly two-dimensional (2D) systems but can be observed also in highly anisotropic layered systems [1] as well as in 200-layer quantum-well structures [2]. Surprisingly, for the paradigm of strong anisotropic systems, graphite, no clear evidence for the QHE has been published. An indication for the possible existence of a QHE at the quantum limit (magnetic fields B > 2T) in graphite has been recently reported [3]. New experimental results [4] indicate a much larger anisotropy, a much smaller coupling between the graphene layers and a larger influence of the two-dimensionality of the sample on the transport properties of highly oriented graphite than previous studies reported. Among these findings is the magnetic-field induced metal-insulator-transition (MIT), which shows significant similarities to the one discovered in Si-MOSFETs and other dilute 2D electron systems [5]. As in graphite [4], in some of these 2D systems the MITs have been shown to be connected to quantum-Hall-insulator-transitions at high magnetic fields [6]. After these experimental observations and the theoretical prediction for an integer QHE in graphite [7], it is reasonable to expect the occurrence of quantum-Hall-states in graphite. Using two different experimental arrangements we show in this contribution that an integer QHE-like response is obtained for highly anisotropic and relatively small samples and electrodes distance.

Experimental

The main obstacle to obtain clear evidence for the intrinsic Hall effect of ideal graphite is the sample quality, i.e. the internal short circuits between graphene planes caused by defects and impurities. As a characterization of the sample quality and orientation of our samples we used the full width at half maximum (FWHM) of the rocking curves. The smaller the FWHM and the distance between voltage electrodes, the clearer are the plateaus in the Hall effect, as can be seen in Fig. 1 were we show data from Ref. [3] for three samples with different FWHM. The samples studied in this work are highly oriented graphite (HOPG) obtained from Union Carbide (sample HOPG-1), from Advanced Ceramics (sample HOPG-2) and from the research institute ``Graphite" in Moscow (sample HOPG-3) with FWHM equal to 0.24°, 0.40° and 0.64°, respectively, and a Kish graphite sample with FWHM ~2°. The measured anisotropy between the c-axis and in-plane resistivities is ~ 3 x 10^4 for the HOPG samples and ~10^2 for Kish graphite. The longitudinal resistance as a function of magnetic field of all three HOPG samples has been measured with the conventional ac-method, where four electrodes made of silver paint are placed on top of a surface parallel to the graphene layers. Using a van der Pauw configuration consisting of four point-like electrodes at the corners of a surface parallel to the graphene layers, as shown in Fig. 1, we obtain the diagonal ρ_{xx} and off-diagonal ρ_{xy} elements of the resistivity matrix [8]. The current is applied between two diagonally opposite contacts and the voltage is measured between the other two contacts. Inverting the two dimensional matrix we obtain the two conductivities σ_{xx} and σ_{xy}. To obtain the absolute values we have measured the effective penetration depth of the current λ ~ 10 μm.

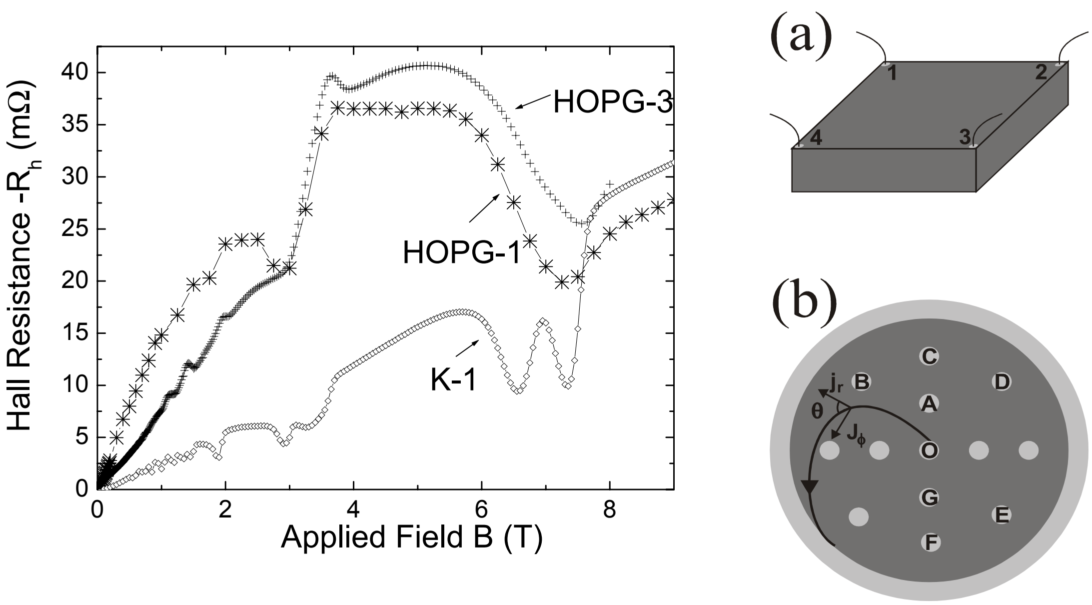

Figure 1. *Left figure*: Hall resistance for three samples (K-1: Kish graphite (R_h/10) at T = 1.5 K, HOPG-1 at 4.2 K and HOPG-3 at 2 K), taken from Ref. [3], measured with the conventional method. The FWHM decreases from K-1 to HOPG-1. *Right figure*: (a) Van der Pauw configuration. Current is applied between the diagonally opposite electrodes and voltage is measured between the other electrodes. (b) View from top onto the Corbino disk geometry with the used electrodes to measure the voltage. The current path is schematically shown with the radial j_r and azimuthal component j_ϕ and the Hall angle θ. The outer ring has a diameter ~ 1mm and the inner electrodes ~ 0.1mm.

A Corbino-disk configuration has been prepared from a 200nm gold layer by e-beam lithography on top of a surface parallel to the graphene layers of the sample HOPG-2 as shown in Fig. 1. In this configuration, the current is applied between the inner point-like electrode and four contacts along the outer ring-like electrode to assure homogenous current distribution. The voltage is measured between any of the other contacts in between them. The magnetic field is applied perpendicular to the graphene layers as in the other experiments. For all geometries the applied current is of the order of 1mA, which is known to be in the ohmic regime and for which no heating effects are observed. The voltage is measured with a LR-700 resistance bridge from Linear Research Inc., which works at low ac-frequency.

Results and Discussion

I. Results with the Van der Pauw configuration

The essential signature of the QHE are the plateaus in a Hall measurement at constant temperature. In Fig. 2 we show that regular plateaus are observable in the off-diagonal element of the conductivity tensor σ_{xy}. The transverse conductivity value at the position of the apparent first plateau-like feature is $\sigma_{xy} \sim 25 \times 10^3\ \Omega^{-1}m^{-1}$, see Fig.2. Within experimental error this value agrees with the measured separation of the plateaus $\Delta\sigma_{xy} \approx 22 \times 10^3\ \Omega^{-1}m^{-1}$. Assuming an exponential decay of the current with the distance from the surface and taking into account that $\lambda >> c/2$, the distance between the graphene layers c/2=0.337 nm, we obtain that the transverse conductivity for a single layer at the first plateau $\sigma^{\square}_{xy}$ as well as the separation between them is (0.2 ± 0.02) e^2/h. Considering the simplicity of the model and the geometrical errors involved in the determination of the absolute values (~ 30%), the quantitative agreement with the expected value for the integer QHE is reasonable. Future experiments should clarify whether the quantitative deviation is related to geometrical errors, the effective distance between graphene layers or to a more sophisticated origin related to the nature of the massless Dirac-Fermions.

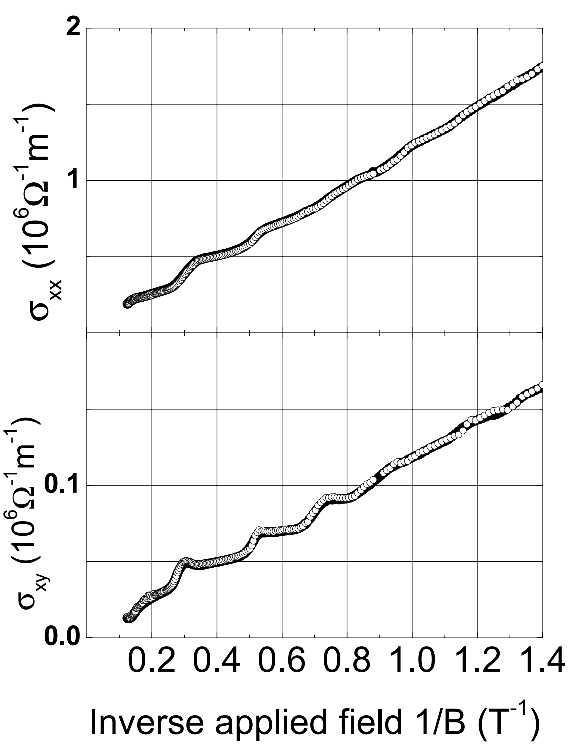

Figure 2. The components of the conductivity tensor vs. inverse field for the sample HOPG-3 measured in van der Pauw-configuration at 0.1 K (o) and 2 K (●). In the off-diagonal element regular plateaus are obtained, indicative of the quantum-Hall-effect.

II. Results with the Corbino-disk configuration

In a homogenous, isotropic (along the planes) 2D sample, the application of a constant current *I* would result in a two-dimensional radial current density $j_r = I/2\pi r$ at a point at distance *r* from the center, which is deflected by the magnetic field by the Hall-angle θ with $\tan\theta = \sigma_{xy}/\sigma_{xx}$. The result is a current path along logarithmic spirals [9], as shown in Fig.1. Following Ref. [10], the relative resistance increase with field between center and outer electrode has a new term, which depends quadratically on the Hall angle. Upon the field dependence of the mobility of the carriers one may expect to see the Hall effect influence in this geometry. If we neglect this influence the voltage between any two contacts at the distances r_1 and r_2 from the center can be calculated from

$$V = \frac{1}{\sigma_{xx}} \int_{r_1}^{r_2} j_r dr = \frac{I}{2\pi\sigma_{xx}} \ln\frac{r_2}{r_1}\,. \quad (1)$$

As expected, the voltage vanishes between contacts at the same distance from the center. The circulating current component is caused by the Lorentz force perpendicular to the electric field and therefore it is a non-dissipative current, which does not contribute to the voltage. We show below that when for a given pair of electrodes inside the Corbino disk the ``longitudinal"-like signal is minimized (i.e. $r_1 \approx r_2$, see Eq.(1)), still a voltage is measured that resembles the Hall signal obtained with the Van der Pauw method.

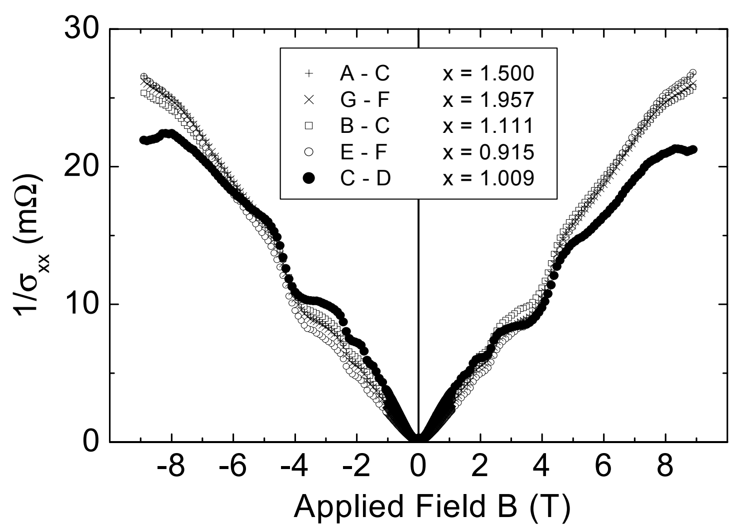

Figure 3. Inverse diagonal component of the conductivity tensor vs. field of the sample HOPG-2 obtained in Corbino-configuration at 2 K. The designations of the voltage contacts are given in the legend and correspond to the ones in Fig. (1). σ_{xx} is calculated from (1) assuming a relation of the distances of the voltage contacts $x = r_2/r_1$ which leads to the best scaling onto the curve for the contact pair A-C.

We show in Fig. 3 the field dependence of the inverse measured voltage obtained from measurements on various contact pairs. Two of them (A-C and G-F) provide the longitudinal magnetoconductivity, which perfectly agrees with that measured with the conventional method. Three pairs at nominally the same distance from center (B-C, E-F and C-D) should show no voltage. In contradiction to the expectations, the voltage between pairs located at nominally the same distance from center is not negligible. The first explanation for this observation would be that this is due to a misplacement of the contacts giving rise to a radial distance between them. To check this, the data in Fig. 3 is presented as follows. For the contact pair A-C, σ_{xx} is calculated from (1) assuming the nominal value for x = 0.45 mm/0.30mm=1.5. For the other pairs, x is chosen in such a way, that the curves scale onto the one for the pair A-C.

As can be seen in Fig.3 the curve for the pair G-F scales very well assuming x=1.957, corresponding to a shift of the inner contact from the nominal one by 70µm. The same holds for the pairs B-C and E-F, and therefore we would conclude that those signals are mainly determined by the ``nominal" longitudinal magnetoresistance. The pair C-D, however, has the smallest nominal misplacement (x=1.009), but shows the most significant deviation from the purely longitudinal magnetoresistance curve A-C. Interestingly, C-D is the contact pair where we observe the clearest plateau-like structures similar to the ones measured in the van der Pauw configuration, as shown in the upper part of Fig. 4. The periodicity of the plateaus (see inset in Fig.4) agrees very well with that obtained from the van der Pauw configuration. The inverse-field period of the plateaus is $\Delta(1/B) \approx 0.17\ T^{-1}$. Using the relation between this period and the 2D electron system we obtain a 2D electron density $\approx 2.85 \times 10^{15}\ m^{-2}$. From the clear plateaus and the estimate done above we may conclude therefore, that the measured voltage between the electrodes C-D at large enough fields is not related to the longitudinal magnetoresistance.

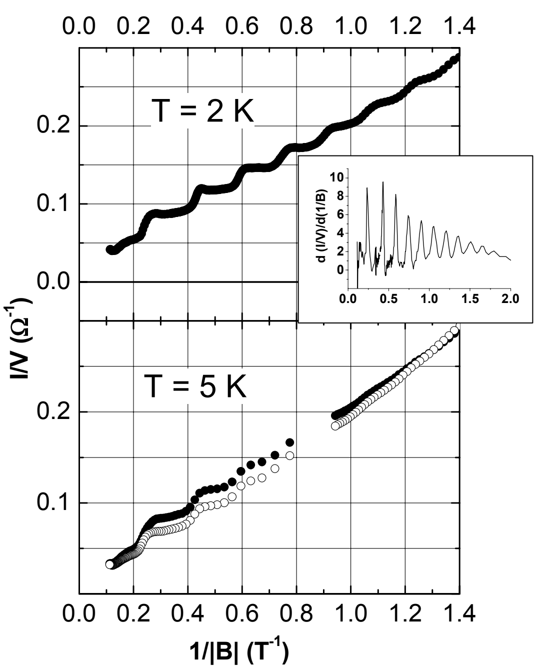

Figure 4. Current divided by the measured voltage between electrodes C and D in the Corbino geometry. Top: data at 2 K. Bottom: Curve taken at 5 K at positive (●) and negative (o) fields. Inset shows the derivative of the data as a function of the inverse field.

III. Discussion

A vanishing longitudinal resistance in the vicinity of the fields where the Landau levels are filled is a concomitant of the QHE, which appears in 2D systems at very low temperatures [11]. However, the longitudinal resistance of graphite does not show any minima or a decreasing behavior with field, see Figs. 2 and 3. The positive magnetoresistance of graphite is very large and, upon sample, is nearly linear in field from fields as low as ~100 Oe and without any sign of saturation up to 9 T (see Fig.2). Note that the magnetoconductance at high fields, where the Shubnikov - de Haas (SdH) oscillations due to the Landau level quantization occur, shows a striking similarity with the transverse conductivity, see Fig.2. The origin of a linear magnetoresistance in graphite as well as in other semimetals and polycrystalline metals is a matter of current discussion, see e.g. Refs. [12,13] and references therein. The simple two-band model [14] does not explain the quasi-linear field dependence observed in all the three samples at low and high fields, unless one assumes ad-hoc field dependences for the electron and hole mobilities and densities.

Abrikosov proposed that the linear magnetoresistance of graphite is due to the small density of the massless carriers in a regime where the energy difference between lowest Landau levels is larger than the temperature and bandwidth along c-axis (small hopping between layers)[15]. The fact that a quasi-linear magnetoresistance is observed at low fields casts doubts whether the transport properties of real graphite can be appropriately described by this ``quantum magnetoresistance" model. Experimental evidence based on a large number of transport measurements with different contact distributions on the same sample indicates that the internal disorder of a graphite sample influences the field and temperature dependence of the transport properties in such an extent that these samples should be considered as a disordered semimetal.

One possible explanation for: (a) the unexpected Hall-like signal at the electrodes C-D, (b) the absence of minima in the nominal longitudinal magnetoresistance, (c) the striking similarity between this and the Hall signal, as well as (d) the non-saturation of the nominal longitudinal magnetoresistance at high fields, may be related to the mobility disorder, extrinsic or intrinsic, of the oriented graphite samples. Numerical calculations based on a disordered-semiconductor model in which the transverse conductivity is included by selecting a special arrangement of simple “gyrators” [12] indicate that the magnetoresistance can be governed by the Hall resistance at high enough fields. Similar ideas were discussed in the literature in the last 50 years [10,16]. We note that this Hall-like signal should be independent of the field direction (switching the field direction will also switch the transverse direction of the current within the disordered network). That means that the measured magnetoresistance at C-D should be field polarity independent in case the Hall current paths are perfectly symmetric. To check this we performed measurements at the two field directions, i.e. +z and -z. The results shown in Figs. 3 and 4 indicate that the observed voltage does not change sign and does depend only slightly on the field direction, probably due to non perfect symmetrical current paths as expected for a real disordered system.

Conclusions

In summary, transport measurements obtained in different configurations in HOPG samples show evidence for an integer QHE in agreement with theoretical expectations. From different experimental facts, e.g. similar field and angle dependence for longitudinal and c-axis resistivities as well as the electrode position dependence of the transport properties, we conclude that even the most ordered HOPG sample contains disorder, which influences the transport properties. Within the disordered semiconductor model it may be possible to understand several of the observations done in this work, in particular the absence of the minima in the nominal longitudinal magnetoresistance, that usually accompany the plateaus in the transverse resistivity. Our results cast doubts on the significance of 3D Fermi surface models with electrons and holes pockets for ideal graphite as well as on the validity of the quantum magnetoresistance model as the explanation for experimental results obtained in real graphite samples. Future work should clarify the field dependence of the resistance at very low fields, where according to the disordered model the Hall component contribution should be negligible and a crossover to the classical B^2 dependence should appear. More time and technical skill will be needed to clarify the transport properties of ideal graphite for which samples of much smaller size are necessary.

Acknowledgements

We thank M. Ziese for discussions. P.E. gratefully acknowledges correspondence with M. M. Parish and Nai-Yi Cui. The stay of Y.K. is supported by the DFG under Le 758/20-1. This work was supported by DFG Grant ES 86/6-3.

References

[1] Störmer HL, Eisenstein JP, Gossard AC, Wiegmann W, Baldwin K. Quantization of the Hall effect in an anisotropic three-dimensional electronic system. Phys. Rev. Lett. 1986; 56: 85-88.

[2] Zhang B, Brooks J, Wang Z, Simmons J, et al. In-plane magnetic-field effect on transport properties of the chiral edge state in a quasi-three-dimensional quantum well structure. Phys. Rev. B 1999; 60: 8743-8747.

[3] Kopelevich Y, Torres JHS, Ricardo da Silva R, Mrowka F, et al. Reentrant metallic behavior of graphite in the quantum limit. Phys. Rev. Lett. 2003; 90: 156402-1 – 156402-4.

[4] Kopelevich Y, Esquinazi P, Torres JHS, Ricardo da Silva R, Kempa H. Graphite as a highly correlated electron liquid. Advances in Solid State Physics 43: 207 (Springer, Berlin, 2003).

[5] Abrahams E, Kravchenko SV, Sarachik MP. Metallic behavior and related phenomena in two dimensions. Rev. Mod. Phys. 2001; 73:251-267.

[6] Hanein Y, Nenadovic N, Shahar D, Shtrikman H, et al. Linking insulator-to-metal transitions at zero and finite magnetic fields. Nature 1999; 400: 735-737.

[7] Zheng Y, Ando T. Hall conductivity of a two-dimensional graphite system. Phys. Rev. B 2002; 65: 245420-1 – 245420-12.

[8] Montgomery HC. Method for Measuring Electrical Resistivity of Anisotropic Materials J. App. Phys. 1971; 42: 2971-2975.
[9] Beer AC, Galvanomagnetic Effects in Semiconductors, Academic Press, New York, 1963.
[10] Weiss H, Welker H. Zur transversalen magnetischen Widerstandsänderung von InSb. Z. für Physik 1954; 138: 322-329.
[11] Jiang HW, Johnson CE, Wang KL, Hannahs ST. Observation of magnetic-field-induced delocalization: Transition from Anderson insulator to quantum Hall conductor. Phys. Rev. Lett 1993; 71:1439-1443. Hilke M, Shahar D, Song SH, Tsui DC, et al. Symmetry in the insulator–quantum-Hall–insulator transitions observed in a Ge/SiGe quantum well. Phys. Rev. B 1997; 56: R15545-R15548.
[12] Parish MM, Littlewood PB. Non-saturating magnetoresistance in heavily disordered semiconductors. Nature 2003; 426: 162-165.
[13] Abrikosov AA. Quantum linear magnetoresistance Europhys. Lett. 2000; 49: 789-793.
[14] Kelly BT. Physics of Graphite, Applied Science Publishers LTD, London and New Jersey (1981).
[15] Abrikosov AA. Phys. Quantum magnetoresistance of layered semimetals. Phys. Rev. B 1999; 60: 4231-4234.
[16] Wick RF. Solution of the Field Problem of the Germanium Gyrator. J. Applied Phys. 1954; 25: 741-756.