Absence of Metal-Insulator-Transition and Coherent Interlayer Transport in oriented graphite in parallel magnetic fields

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Abstract

Measurements of the magnetoresistivity of graphite with a high degree of control of the angle between the sample and magnetic field indicate that the metal-insulator transition (MIT), shown to be induced by a magnetic field applied perpendicular to the layers, does not appear in parallel field orientation. Furthermore, we show that interlayer transport is coherent in less ordered samples and high magnetic fields, whereas appears to be incoherent in less disordered samples. Our results demonstrate the two-dimensionality of the electron system in ideal graphite samples.

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Recent experimental and theoretical studies of graphite have renewed the interest in this system [1,2,3,4,5,6,7,8,9,10]. Experimental results show that, contrary to the common belief, the transport and magnetic properties of graphite cannot be accounted for by semiclassical models. Experiment and theory raise a number of questions concerning the coupling between the graphite layers. The understanding of the transport properties is of primary interest and can provide a fundamental contribution to the physics of two-dimensional (2D) systems in general. In this letter we deal with two important open questions:

1. A MIT appears both in the in-plane [1,3,4] and out-of-plane [5] resistivity induced by a magnetic field $B$ applied perpendicular to the graphite layers, i.e. $B||c$-axis. Based on magnetization data [2] and the found 2D scaling similar to that in the MIT of Si-MOSFETs (metal-oxide-semiconductor field-effect-transistor) [11], Mo-Ge [12] and Bi-films [13], the MIT in graphite has been
interpreted in terms of superconducting fluctuations [1,3]. It has been shown that the density of states at the Fermi level is enhanced through topological disorder, thus leading to the possible occurrence of localized ferromagnetism and superconductivity [7].

Other interpretation of the MIT in graphite uses the idea that a single graphite layer is the physical realization of the relativistic theory of (2+1)-dimensional Dirac fermions due to the linear dispersion in the spectrum of quasiparticle excitations in the vicinity of the corners of the Brillouin zone [14]. Taking this into account and the large Coulomb coupling constant for graphite [5,8], a magnetic catalysis (MC) has been proposed [9]. This original explanation assumes that a magnetic field perpendicular to the graphite layers breaks the chiral symmetry and opens a gap in the spectrum of the quasiparticles at the corners of the Brillouin zone. This effect is interpreted as the enhancement of the fermion dynamical mass through electron-hole pairing, i.e. a transition to an excitonic insulating state.

Which role does the field direction play? The experimental evidence indicates that the MIT in Si-MOSFETs occurs independently of the orientation of the magnetic field and, therefore, has been considered to be driven solely by spin-dependent effects [15]. On the other hand, the MC in graphite would be possible only for the case $B \parallel c$, i.e. the transition should be absent in the parallel case [8]. To our knowledge there is no experimental proof published in the literature that the MIT is absent in oriented graphite samples for fields applied parallel to the graphite planes. A clear experimental evidence for the absence or not of the MIT would give an important hint to search for its origin. This is one of the tasks of our experimental work.

2. In a recent work we have shown that the metalliclike behavior of the out-of-plane $c$–axis resistivity $\rho_c$ is directly correlated to that of the in-plane resistivity [5] and that the intrinsic $\rho_c(T)$ of an ideal graphite sample would not be metalliclike. These results cast doubts about the interlayer transfer integral value of $\sim 0.3$ eV used all over the literature [20,6] in which neither the electron-electron interaction nor charge fluctuations were taken into account [21]. In principle, for such a large interlayer transfer one would expect coherent transport for the interlayer magnetoresistance at low temperatures. In this work we provide an answer to the question whether the $c$–axis transport in graphite is or not coherent and whether this (in)coherent transport is influenced by sample inhomogeneities.

We have measured the following samples: a highly oriented pyrolytic graphite (HOPG) manufactured at the Research Institute "Graphite" in Moscow and denoted as HOPG-3 [3]. The X-ray rocking curve for this sample, see Fig. 1(a), exhibits a full width at half maximum (FWHM) of $(0.64 \pm 0.05)^\circ$. We take this value as a measure for the misalignment of the graphite layers within
Fig. 1. (a) X-ray rocking curve of the sample HOPG-3. (b) Angle dependence of the in-plane resistivity (●) for the same sample in a magnetic field of 9 T at 2 K. The field parallel to the sample at 90° is defined at the resistivity minimum. A second run (+) is stopped at 90°.

the sample with respect to each other. Further two HOPG samples, one from Union Carbide Corp. with a FWHM of 0.24° (HOPG-1) and the other from Advanced Ceramics Corp. with a FWHM of 0.40° (HOPG-2), have been measured. The fourth sample was a Kish graphite sample with a FWHM of 1.6° and a in-plane resistivity at 2K and zero field ∼ 100 times smaller than for
the HOPG samples. AC resistivity measurements were performed by a conventional four-probe method. The samples were fixed in a rotating sample holder inside the bore of a 9T superconducting solenoid. Temperature stability was better than 2 mK in the whole temperature range.

The crucial point of the magnetoresistance measurements in parallel field is the misalignment of the sample with respect to the field. The ultimate limit for the alignment of the sample would be that of the graphite layers within the sample. To adjust the sample to the magnetic field we measured the angle dependence of the in-plane resistivity with the current always perpendicular to the field. The result is shown in Fig. 1(b) for the HOPG-3 sample. Here 90°, i.e. field parallel to the sample, is defined at the minimum of the resistivity. The high angle resolution, the strong angle dependence of the resistivity and the high sensitivity of its measurement, as well as the excellent reproducibility of the absolute angle (of the order of the angle resolution, see Fig. 1(b)) allow us to align the sample parallel to the field with an accuracy of ±0.02°, well below the FWHM of the rocking curve.

Figure 2(a) shows the results of the in-plane resistivity with \( B \parallel c \). We note that the metalliclike phase, i.e. \( d\rho/dT > 0 \), is observed between a maximum \( T_{\text{max}} \) and a minimum temperature \( T_{\text{min}} \) and for fields below a critical field \( B_c \). The difference between \( T_{\text{max}} \) and \( T_{\text{min}} \) decreases as the field approaches \( B_c \), i.e. \( T_{\text{max}}(B_c) = T_{\text{min}}(B_c) \), and the metalliclike phase disappears. Figure 2(b) shows these data together with fits of \( T_{\text{max}}(B) \) to the experimentally found [1] relation

\[
T_{\text{max}}(B) \propto \left( 1 - \frac{B^2}{B_1^2} \right),
\]

and of \( T_{\text{min}}(B) \) to the relation [4]

\[
T_c(B) \propto \sqrt{B - B_0},
\]

where \( B_0 \) and \( B_1 \) are free parameters, the first is referred as the offset field [10]. The extrapolation of those relations serves to determine the critical field at their crossing which for the present case is \( B_c^\perp = (0.12 \pm 0.01)\text{T} \).

The results obtained for \( B \perp c \) (aligned as shown in Fig. 1(b)) are shown in Fig. 3(a) and the corresponding \( T_{\text{min}} \) and \( T_{\text{max}} \) in Fig. 3(b). Whereas \( T_{\text{min}}(B) \) follows a similar relation as in the former case, \( T_{\text{max}}(B) \) is reproduced by Eq. (1) only in the low field range. At high fields the empirical relation \( T_{\text{max}} \propto e^{-\frac{B}{B_c}} \) is found. We note that the intersection of \( T_{\text{max}}(B) \) and \( T_{\text{min}}(B) \) is at \( B_c^\parallel = (11.3 \pm 0.1)\text{T} \). The dominant contribution to the misalignment is the misalignment of the graphite layers with respect to each other and is taken
Fig. 2. (a) In-plane resistivity of the sample HOPG-3 as a function of temperature for magnetic fields $B = 0, 0.02, 0.04, 0.06, 0.08, 0.09, 0.10, 0.11, 0.12 \, \text{T}$ (bottom to top) applied $\parallel c$. Upward (downward ) arrows mark the minima (maxima) of the resistivity. (b) Temperatures of the maxima and minima of the resistivity as a function of field together with fits for $T_{\text{max}}$: Eq. (1) ($-\cdot$) with $B_1 = 0.13 \, \text{T}$; $T_{\text{min}}$: dotted line corresponds to Eq. (2) with $B_0 = 65.9 \, \text{mT}$. The intersection of the curves gives the critical field $B_c$. 
Fig. 3. (a) In-plane resistivity of the sample HOPG-3 as a function of temperature for magnetic fields $B = 0, 1\ldots 9$ T (bottom to top) applied $\perp c$, aligned as in Fig. 1(b). (b) Temperatures of the maxima and minima of the in-plane resistivity as a function of field together with fits for $T_{\text{max}}$: Eq. (1) ($-$) with $B_1 = 5.83$ T and dashed line $\propto \exp(-B/B_2)$ with $B_2 = 5.11$ T; $T_{\text{min}}$: dotted line corresponds to Eq. (2) ($B_0 = 1.59$ T).

to be $(0.64 \pm 0.05)\degree$ (see Fig. 1). The perpendicular component of $B_c^\parallel$ is then $(0.13 \pm 0.01)\text{T}$. This value is within the error equal to $B_c^\perp$. From this we conclude that the MIT is, if not solely driven by the magnetic field perpendicular to the graphite layers, by far dominated by it.
We would like to note two details of the field-driven transition shown in Fig. 2(a) for sample HOPG-3, as well as in previous publications [3,4,5]. First, in the “insulating” side of the transition at $B \geq B_c$ we would expect that the resistivity $\rho \rightarrow \infty$ for $T \rightarrow 0$. Instead we observe always a saturation or, upon applied field, a weak logarithmic increase of the resistivity decreasing temperature [16]. The saturation for $T \rightarrow 0$ in the insulating side of the transition has been reported for various 2D systems as, for example, in Mo-Ge films [12], GaAs/AlGaAs heterostructures [17] and Josephson junction arrays [18]. There is no consent on the origin of this saturation. Whatever the reason for it is, the main result of this part of the work, i.e. that only the normal component of the applied field to the planes drives the transition, remains untouched.

Second, one would tend to underestimate the real magnitude of the effect driven by the applied field because of the relative small change of the resistivity shown in Fig. 2(a). However, we stress that the relative change in the resistivity with field depends on the sample characteristics. Experimental data from different graphite samples show a change at low temperatures between $\sim 20\%$ up to more than one order of magnitude for a field of the order of 1 kOe [3,4,5,19]. Qualitatively speaking, the transition is similar for all samples studied.

We discuss now shortly the origin of the MIT in graphite. First we note that Eq. (72) from Ref. [10] ($T_c(B) \propto (1 - (B^2_0/B^2)^\sqrt{B})$ fits the data for $T_{\text{min}}(B)$ as well as Eq. (2) with similar $B_0$. In Ref. [10] the authors argue that the offset field $B_0 \simeq \pi n c/e$ ($n$ is the charge density) is model independent and is related to the minimum field required to fill the lowest Landau level, necessary condition to deblock the electrons for pairing and to produce the excitonic gap. For the majority carriers $n \sim 10^{11}\text{cm}^{-2}$ and $B_0 \sim 2\text{T}$ in clear disagreement with the experimental result $\sim 0.06\text{T}$. But, if we take the minority carrier density $n \sim 10^9\text{cm}^{-2}$ [20] we get roughly the measured $B_0$. Nevertheless and since MC is a 2D phenomenon, it is unclear whether this assumption is valid. We note that recent results [5] including those from this work indicate that for HOPG samples, the coupling between planes is much weaker than previously assumed, casting doubts about the correctness of a 3D Fermi surface with two types of carriers with different densities and effective masses for ideal graphite [20].

Returning to the superconducting scenario which is supported by the magnetization data [2], we may relate $T_{\text{max}}(B)$ to the critical temperature of a system of superconducting islands in a semiconducting matrix [1]. In this case mesoscopic effects play a role. Indeed, the observed behavior in the parallel case shows a change of curvature of $T_{\text{max}}(B)$ similar to that found theoretically for disordered 2D superconductors [22]. We note also that an exponential decay with field for $T_c(B)$ has been predicted for 2D superconductors with weakly Josephson-coupled local superconducting islands [23]. Because in the parallel
Fig. 4. Angle dependence around 90° of the out-of-plane resistivity of the Kish graphite (a), HOPG-2 (b) and HOPG-1 (c) samples at $B = 9$ T and at 2 K.

case the MIT occurs at large fields, we are able to observe the anomalous decay of $T_{\text{max}}$ in contrast to the transverse case, see Fig. 2(b). On the other hand we may argue that the reason for the difference in the behavior of $T_{\text{max}}(B)$ may be due to its sensitivity to the intrinsic misalignment of the graphene planes of the sample that shows a gauss-like distribution.

We discuss now the interlayer transport. One possible way to test coherent transport across the graphite layers is given by the measurement of a maximum in the angle dependence of $\rho_c(\theta)$ at magnetic fields parallel to the layers [24,25,26]. This peak should be absent for incoherent interlayer transport but observed if the inequality $\omega_c \tau > 1$ holds, where $\omega_c$ is the cyclotron frequency and $\tau$ the relaxation time of the carriers. Coherent transport means therefore that band states extend over many layers and a 3D Fermi surface can be defined. In the other case, incoherent transport is diffusive and neither a 3D Fermi surface nor the Bloch-Boltzmann transport theory is applicable [24]. In order to check this and the role played by disorder we have performed measurements of the out-of-plane electrical resistivities with high angle resolution.

Figure 4 shows the results for three samples at 9 T and 2 K. We observe that a weak coherent peak in $\rho_c$ around the parallel orientation (90°) occurs and this is larger the larger the FWHM of the corresponding rocking curve. The asymmetry seen in the angle dependence is in part due to the small experimental
misalignment of the surface of the sample and to the lack of crystal perfection. The coherent peak decreases as expected with field. Our results indicate that lattice defects not only affect the transport as scattering centers but they contribute to enhance the coupling between the layers giving rise to a 3D-like electronic spectrum and coherent transport. The absence of coherent peak in ideal samples may be related either to incoherent transport or that $\omega_c\tau < 1$ holds. Although the validity of semiclassical criteria for incoherent transport is under discussion [25] we use the Ioffe-Regel-Mott maximum metallic resistivity $\rho_{\text{max}}$ criterion to evaluate coherent transport [27]. We obtain that only for the HOPG-1 and -2 samples $\rho_c > \rho_{\text{max}}$ in the whole $T$– and $B$–range, in agreement with the absence of coherent peak.

From our results we draw the following conclusions: The MIT in HOPG is triggered only by a magnetic field perpendicular to the graphite layers. Therefore, it is unlikely that spin effects play a significant role in the MIT. The absence of the MIT in the parallel field orientation supports the theoretical approach of Refs. [8,9,10], but there is apparently no quantitative agreement [10]. On the other hand, the influence of possible superconducting fluctuations on the MIT cannot be ignored. The transport perpendicular to the graphite layers in highly oriented and less disordered samples appears to be incoherent, demonstrating the quasi-2D character of the electron system of graphite. Samples defects lead to a better coupling between the layers, a 3D-like behavior and coherent interlayer transport, added to the possible local enhancement of the density of states which may generate local superconductivity and ferromagnetism [7].

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References

[1] Y. Kopelevich, V. V. Lemanov, S. Moehlecke, J. H. S. Torres, Phys. Solid State 41, 1959 (1999).

[2] Y. Kopelevich, P. Esquinazi, J. H. S. Torres, S. Moehlecke, J. Low Temp. Phys. 119, 691 (2000).
[3] H. Kempa et al., Solid State Commun. 115, 539 (2000).
[4] M. S. Sercheli et al., Solid State Commun. 121, 579 (2002).
[5] H. Kempa, P. Esquinazi, Y. Kopelevich, Phys. Rev. B 65, 241101(R) (2002).
[6] J. Gonzáles, F. Guinea, M. A. H. Vozmediano, Phys. Rev. Lett. 77, 3589 (1996).
[7] J. Gonzáles, F. Guinea, M. A. H. Vozmediano, Phys. Rev. B 63, 134421 (2001).
[8] D. V. Khveshchenko, Phys. Rev. Lett. 87, 206401 (2001).
[9] D. V. Khveshchenko, Phys. Rev. Lett. 87, 246802 (2001).
[10] E. V. Gorbar et al., cond-mat/0202422.
[11] P. Phillips et al., Nature 395, 253 (1998).
[12] N. Mason and A. Kapitulnik, Phys. Rev. Lett. 82, 5341 (1999).
[13] N. Markovic, C. Christiansen and A. M. Goldman, Phys. Rev. Lett. 81, 5217 (1998).
[14] G. W. Semenoff, Phys. Rev. Lett. 53, 2449 (1984).
[15] S. V. Kravchenko et al., Phys. Rev. B 58, 3553 (1998).
[16] Y. Kopelevich et al., cond-mat/0209442.
[17] J. Yoon, C. C. Li, D. Shahar, D. C. Tsui, and M. Shayegan1, Phys. Rev. Lett. 84, 4421 (2000).
[18] H. S. J. van der Zant, F. C. Fritschy, W. J. Elion, L. J. Geerligs, and J. E. Mooij Phys. Rev. Lett. 69, 2971 (1992).
[19] C. Ayache, Ph.D. Thesis, Grenoble 1978 (unpublished).
[20] B. T. Kelly, in Physics of Graphite, Applied Science, London/New Jersey, 1981.
[21] M. A. H. Vozmediano, M. P. López-Sancho, F. Guinea, Phys. Rev. Lett. 89, 166401 (2002).
[22] B. Spivak and Fei Zhou, Phys. Rev. Lett. 74, 2800 (1995).
[23] V. M. Gallitski and A. I. Larkin, Phys. Rev. Lett. 87, 087001 (2001).
[24] P. Moses, R. H. McKenzie, Phys. Rev. B 60, 7998 (1999).
[25] J. Singleton et al., Phys. Rev. Lett. 87, 117001 (2001).
[26] J. Wosnitza et al., Phys. Rev. B 65, 180506(R) (2002).
[27] J. J. McGuire et al., Phys. Rev. B 64, 94503 (2001).