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Field-induced metal-insulator transition in the *c*-axis resistivity of graphite

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We show that the resistivities perpendicular ρ_c and parallel ρ_a to the basal planes of different graphite samples show similar magnetic-field-driven metal-insulator transitions at a field $B_c \sim 0.1$ T applied parallel to the *c* axis. Our results demonstrate the universality of the recently found scaling in ρ_a of graphite and indicate that the metalliclike temperature dependence of ρ_c is directly correlated to that of ρ_a. The similar magnetoresistance found for both resistivities, the violation of Kohler's rule, and the field-induced transition indicate that the semiclassical transport theory is inadequate to understand the transport properties of graphite.

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Although a considerable amount of studies has been performed on graphite and related compounds, their transport properties are still not well understood. The scientific interest on graphite has been recently renewed by recent magnetization[1] and transport[2] results on highly oriented pyrolytic graphite (HOPG). These show irreversible magnetization[1] that resembles that of a superconducting (SC) loop even at room temperature suggesting the existence of localized SC domains in topological disordered regions.[3] This result is indirectly supported by the recently found magnetic-field-driven superconductor-insulator-type transition (SIT) in the in-plane resistivity of a HOPG sample.[2] Remarkably, this field-driven transition showed a similar scaling as found for two-dimensional (2D) disordered superconductors as well as for Si metal-oxide-semiconductor field-effect transistors (MOSFET's).[4]

It has been speculated that in graphite impurity-assisted hopping mechanism may affect the electronic transport along the *c* axis. In this case the conduction across the graphite planes might be influenced by the in-plane transport in a given temperature (T) range. The aim of our research is to check whether the *c*-axis resistivity $\rho_c(T,B)$ shows a similar SIT with a similar scaling as the in-plane resistivity $\rho_a(T,B)$. Evidence for this speculation has important consequences to solve an old and basic problem of the transport in graphite, namely, to what extent the metalliclike behavior of ρ_c is an intrinsic property of the transport along the *c* axis and whether the semiclassical theories are appropriate to account for the observed effects. Furthermore, it is important to prove experimentally the universality of the 2D scaling found in graphite.

We have studied the resistivities $\rho_a(T,B)$ and $\rho_c(T,B)$ of four samples: two HOPG samples, sample 1 from Advanced Ceramics [rocking curve (full width at half maximum) FWHM $\simeq 0.4°$], sample 2 (from the Research Institute "Graphite" (Moscow), FWHM $\simeq 0.4°$) and samples 3 and 4, Kish graphite crystals. The typical length and width of the samples was ~ 2 mm and thickness between 0.1 mm and 0.5 mm. The in-plane resistivity ρ_a was measured attaching four contacts on the sample surface with silver paint. The absolute values were obtained with other configuration that assures a uniform current distribution. The *c*-axis resistance was measured attaching two contacts on each of the surfaces (one in the middle and other surrounding it and contacted on the rest surface). A resistance bridge (LR700) was used with an ac current $\leqslant 1$ mA. The field was applied normal to the graphene layers. The temperature stability was better than 2 mK in the whole range.

The left panels of Figs. 1–3 show both resistivities as a function of T at constant fields for samples 1–3, respectively. At zero applied field the basal-plane resistivity shows the typical behavior reported in the literature: whereas for sample 1 ρ_a shows a metallic behavior up to $T \sim 100$ K, (~ 220 K for sample 2), sample 3—which shows the lowest resistivity—up to the highest measured $T \sim 270$ K. Similar absolute values as well as the observed maximum of ρ_c for samples 1 and 3 were reported in the literature.[5,6] Note also that sample 2 with the highest $\rho_c(0)$ shows no maximum but a saturation below ~ 10 K. The basal-plane rest resistance ratio $R_{RR} = R(300K)/R(4.2)$ has been tentatively taken in the literature as a measure of sample perfection.[5] A speculative interpretation of the observed nonmonotonic behavior of ρ_c vs R_{RR} has been given in terms of the influence of laminar defects and microcraks. Samples with $R_{RR} < 10$, for example, were considered of poor perfection. We note, however, that this sample classification might be misleading because topological defects may play a role other than scattering centers, but can lead to superconducting as well as ferromagnetic domains.[3] In this case large R_{RR} values do not necessarily mean high quality. We believe that a broad spectrum of transport data of graphite may need to be reinterpreted.

A clear magnetic-field-driven transition from a metallic to a semiconductinglike behavior is observed in both ρ_a and ρ_c, see Figs. 1–3. Remarkably, this transition is observed at fields $B \sim 0.1$ T in both resistivities. The observed behavior in $\rho_a(T,B)$, in particular the leveling at low T, is similar to that reported in 2D films.[7] The minimum in $\rho_a(T)$, which develops at intermediate fields in sample 3, is similar to that measured in a Si MOSFET (Ref. 8) but located at 100 times higher T and observed at 10 times smaller fields.[9] In spite of

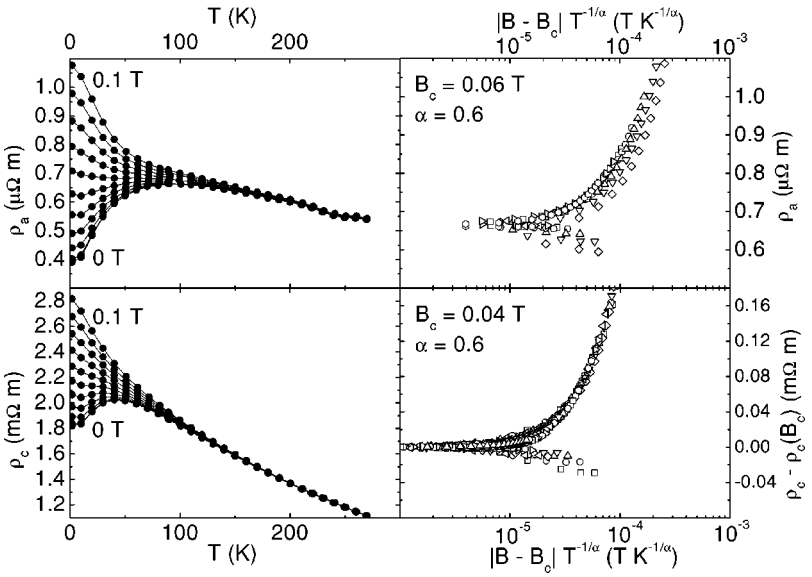

FIG. 1. Left panels: In-plane ρ_a and c-axis ρ_c resistivities as a function of temperature at constant fields for sample 1. The field step between two curves is 0.01 T. Right panels: Scaling behavior of the resistivities. The experimental line ρ_c ($B_c = 0.04$ T) has been subtracted from the ρ_c data. The symbols are the data points obtained between 30 K and 120 K. The symbols in the lower figure correspond to the data taken between 40 K and 270 K.

these similarities, the transition in graphite may have other origin as in Si MOSFET's. We note that the metal-insulator transition (MIT) in 2D electron systems is measured in the in-plane resistivity as a function of the magnetic field applied parallel as well as normal to the film plane.[4] Recent high-resolution angle-dependent experiments[10] reveal, however, that the MIT in graphite is driven mainly by the perpendicular component of the field providing a clear evidence of the 2D character of graphite in agreement with recent theoretical work.[11]

In the ideal case the separatrix line obtained at a critical field $B = B_c$ is a line where the resistivity remains temperature independent and separates the metallic branch from the semiconducting branch. It has been shown that for 2D disordered superconductors as well as for Si MOSFET's, for fields near the MIT the in-plane resistivity follows a scaling when it is plotted as function of the scaling variable $|B - B_c| T^{-1/\alpha}$, α being a critical exponent. In a similar way we determined B_c as well as α from $\rho_a(T,B)$, see the right upper panels in Figs. 1–3. Taking into account that the separatrix line is not an ideal horizontal line in our samples, we obtain a reasonable scaling in a restricted temperature range. Both parameters are similar for the three samples and agree with those reported in Ref. 2. The exponent $\alpha = 0.6 \pm 0.05$ coincides with that for Si MOSFET's (Ref. 12) and ultrathin a-Bi films.[13] A similar scaling with the same exponent α but with a smaller B_c is obtained for ρ_c at 30 K $\leqslant T \leqslant$ 60 K for samples 1 and 3. Sample 2 c-axis resistivity shows no metallic behavior at zero field and therefore no apparent scaling. Based on this scaling a SIT in MOSFET's has been suggested.[14] The evidence for Cooper pairing of the carriers, however, is still a matter of discussion in the literature and no consent has been achieved yet.[4]

As shown in superconducting amorphous In-O films[15] the scaling can be considerably improved if one assumes that in addition to the contribution to the conductivity that shows the SIT, an additional T-dependent contribution affects the resistivity. It has been speculated that this contribution can be

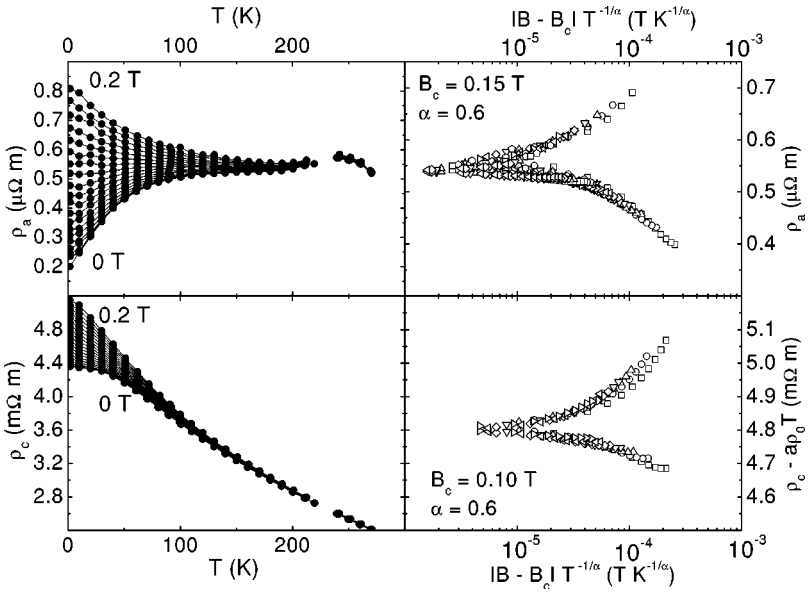

FIG. 2. The same as Fig. 1 but for sample 2. Right panel: The symbols are the data points obtained between 30 K and 200 K. The symbols of the lower figure are obtained from the data between 30 K and 110 K. The parameter $a = -0.0013$ K^{-1} and $\rho_0 = 4.7$ mΩm.

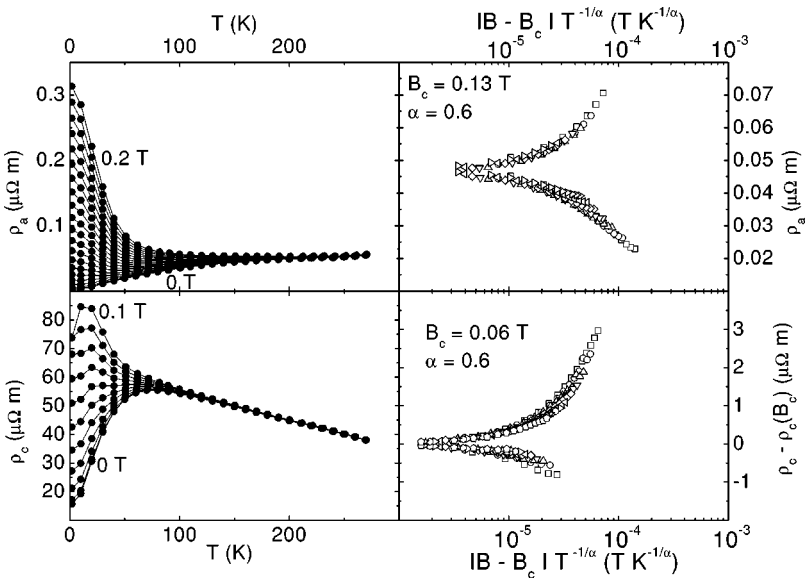

FIG. 3. The same as Fig. 1 for sample 3, $\rho_a(2\ \mathrm{K},0)=0.0015\ \mu\Omega$ m. Right panel: The symbols are the data points obtained between 50 K and 130 K. The symbols of the lower figure correspond to the data taken between 90 K and 200 K.

due to the conductivity of normal electrons.[15] Within the same idea and assuming that the intrinsic *c*-axis resistivity of graphite has a nonmetallic T dependence, which adds as background to the field-dependent part, we show in the lower right panels of Figs. 1–3 the scaling obtained if we subtract from the measured $\rho_c(T,B)$ the line $\rho_c(T,B_c)$ or a linear T dependence $a\rho_0 T$ [sample 2, a is a free parameter and $\rho_0=\rho_c(T\rightarrow 0,B\sim 0.1\ \mathrm{T})$]. In this case the T range for scaling is considerably enhanced. Obviously, a similar inprovement of the T range for scaling is obtained also for the in-plane resistivity after subtracting a T-dependent background. The value of the exponent α remains the same. These results indicate the universality of the MIT found in graphite for both resistivities and of its scaling.

Graphite can be considered as a dilute electron system with a Coulomb coupling constant $r_s=1/(\pi n_{2D})^{1/2}a_B^*$ where the 2D carrier density is given by $n_{2D}=n_{3D}d$ with $d=0.335$ nm the interplane distance, $a_B^*=\epsilon\hbar^2/e^2m^*$ the effective Bohr radius, $\epsilon=2.8$ the dielectric constant, and m^* the effective mass of the carriers. Taking literature values for the majority (minority) carriers,[16] one obtains $r_s\sim 5(10)$ suggesting that the 2D electron system in graphite might be considered as a strongly correlated liquid and the effects due to the electron-electron interaction should be important. We note that $n_{2D}\sim 10^{11}\ \mathrm{cm}^{-2}$ practically coincides with that reported for a Si MOSFET.[4]

One important characteristic observed in several of the measured 2D systems with a MIT is that the critical sheet resistance R_s at the separatrix is of the order of the quantum unit h/e^2. Within the scaling theory[17] of a SIT, $R_s=h/4e^2\simeq 6.4\ \mathrm{k}\Omega$ at B_c. Assuming that the input current decreases exponentially in the *c* direction with a penetration depth λ and taking into account the distance d between graphene sheets, we estimate $R_s\simeq(b/l)R/[1-\exp(-d/\lambda)]$ for a single graphene layer, where R is the measured longitudinal resistance at the sample surface, b is the sample width, and l is the distance between voltage contacts.[18] With measured parameters we obtain at the separatrix $R_s\simeq a(h/4e^2)$ with $a=1/3.2(1/3.7)$ for sample 1(2), whereas for sample 3 $a=1/27$, using $\lambda\simeq 10\ \mu$m obtained from measurements with different configurations. The difference in sheet resistance between samples 1, 2, and 3 may be related to a difference in the coupling between layers due to lattice defects, such as stacking faults or in-plane defects, or may indicate the lack of a unique critical resistance as observed in disordered thin films.[19]

We note that lattice defects may not only affect the transport as scattering centers but they may also contribute to enhance the electronic coupling between layers giving rise to a quasi-3D electronic spectrum with coherent transport along the *c* axis. Field-angle-dependent experiments around the direction parallel to the planes show a weak coherent peak in *c*-axis resistivity for Kish graphite (but not for the HOPG samples) as expected from theoretical considerations for coherent transport.[20]

The T dependence of ρ_c in HOPG has been interpreted[6] in terms of a semiclassical model with parameters such as the carrier density $n(T)$, the stacking-fault spacing l, and the potential barrier formed on the plane of stacking disorder ΔE. The increase of ρ_c with T at $T<100$ K was attributed to an interlayer transfer of carriers, which provides the mixing of the *c* axis and in-plane transport. It was further speculated that the interaction of the carriers with phonons on the planes determines the T dependence.[6] We note first that according to the model[6] and from the comparison of the absolute values of ρ_c of our samples, sample 2 should have an extremely large stacking-fault density. This is at odds with x-ray diffraction results that indicate that this kind of lattice defect is absent in high-quality HOPG.[21] Second, from our results it is obvious that the increase of ρ_c with T at $T<100$ K cannot be attributed simply to electron-phonon scattering. Nevertheless, the concept of conduction-path mixing mechanism is appealing. This can be realized if the conduction path of the electrons along the *c* axis is in part short circuited by lattice defects or impurities as was usually speculated in literature. The conduction-path mixing may be also related to the situation found in layered high-temperature superconductors, where in-plane charge fluctuations influence the *c*-axis

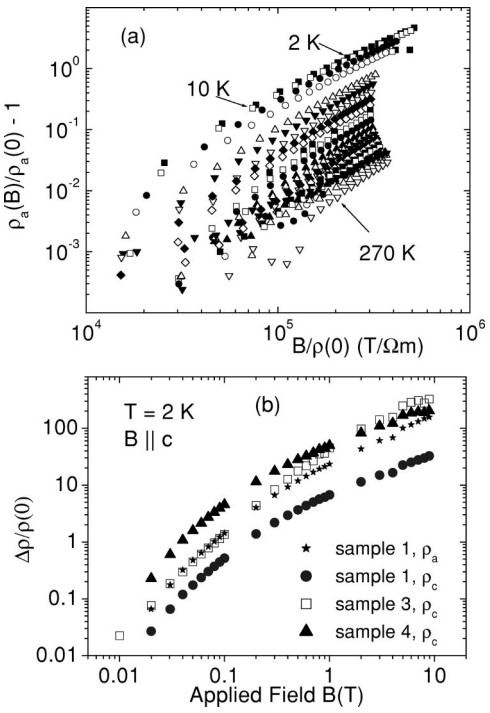

FIG. 4. (a) Kohler's plot of the in-plane magnetoresistance data for sample 1. Between 10 K and 270 K each curve has been taken every 10 K. (b) Magnetoresistance $\Delta\rho=\rho_{a,c}(B)-\rho_{a,c}(0)/\rho_{a,c}(0)$ as a function of the applied field at $T=2$ K for samples 1, 3, and 4.

transport.[22] Our results cast also doubts about the estimate of ~ 0.3 eV for the interlayer transfer integral used in literature.[16] This is not surprising since neither the electron-electron interaction nor charge fluctuations[23] were taken into account.

Regarding the field dependence of ρ_c one may argue that the field can decrease the carrier concentration at low T inducing an increase in ρ_a and ρ_c. However, this appears to be at odds with the increase of $n(T)$ by a factor of 2 with a field of 0.2 T obtained from magnetoresistance measurements and within a semiclasical model.[24] As noted in the literature,[24] the mean mobility in graphite cannot be estimated rigurously with the usual two-band model since the resistivity does not follow the predicted B^2 dependence. Arbitrary definitions to circumvent this problem did not provide a satisfactory description of the measurements. Our results indicate clearly that Kohler's rule is not fulfilled in graphite, neither in the *a* nor in the *c* direction—as can be seen in Fig. 4(a) for ρ_a—implying that the semiclassical picture does not apply. Furthermore, we demonstrate in Fig. 4(b) that the low-temperature magnetoresistance in the *c* direction, shown for two Kish graphite samples and sample 1, is basically the same as the in-plane one. This result also indicates the correlation between both conductivities and contradicts the semiclassical model since for $B||c||J$ no magnetoresistance in the *c* direction is expected.

Concluding, we proved that both in-plane and out-of-plane resistivities in graphite show a MIT with a scaling similar to 2D electron systems and indicate that the metallic-like behavior of $\rho_c(T,B)$ is related to $\rho_a(T,B)$. Our overall results show that semiclassical models cannot account for the magnetotransport properties of graphite.

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