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Hall effect and magnetoresistance in low dimensional magnetic systems

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We have measured the Hall effect and magnetoresistance of stages 5 and 9 $FeCl_3$-graphite intercalated compounds in a magnetic field up to 20 T and at several temperatures. Our preliminary results indicate that the Hall coefficient (R_H) of stage 5 is positive at all field values and has insignificant variation between room and liquid helium temperature. In stage 9, however, R_H has a complicated field and temperature dependence. The magnetoresistance of the two stages at all temperature and field values is positive and stage 5 exhibits strong Shubinkov–de Haas oscillations at helium temperature. Fourier transforms of these oscillations are performed and the results indicate that stage 5 has a dominant fundamental frequency which is attributed to the hole carriers, whereas stage 9 exhibits a modulation of several frequencies which are related to different carrier pockets.

I. INTRODUCTION

The extremely high anisotropy in most of the physical properties of highly oriented pyrolytic graphite (HOPG) makes this system an interesting candidate for low dimensional studies. Moreover this anistropy can be increased by the intercalation of acceptor or donor compounds into the graphite.[1] Because, electronically, only the graphite layers next to the intercalant change significantly upon intercalation,[1,2] the intercalation of magnetic species into the graphite can thus provide a layered magnetic compound with variable spacing between the magnetic layers.

In this work, we report the measurements of the Hall effect and the magnetoresistance and its quantum oscillations of stages 5 and 9 $FeCl_3$-graphite intercalated compounds (GIC's). Measurements of the low-field magnetic properties of this system were performed by several authors,[3–5] the results of the magnetic susceptibility measurements indicate the existence of a low-temperature phase transition at 1.7 K. In high stages, where there are enough inner layers to screen out the interactions between the magnetic intercalant layers, one can expect the behavior to approximate a two-dimensional magnetic system. Quantum oscillations (QO), which provide information about the Fermi surfaces and density of carriers, were observed in some GIC's.[6,7]

II. EXPERIMENT

The $FeCl_3$-GIC samples were prepared using a standard two-zone furnace technique where stage index was controlled by the temperature difference between the graphite host (HOPG) and the $FeCl_3$ powder. The samples were in the form of thin rectangular plates of dimensions $1.5\times0.5\times0.1$ cm^3. The samples measured were characterized by means of x-ray diffraction and Mössbauer effect. Galvanomagnetic measurements were performed in high magnetic field at the Francis Bitter National Magnet Laboratory (MIT). Layers of about $1.5\times0.5\times0.001$ cm^3 were peeled from the samples and then mounted on flat substrates. The five-probe dc technique was used to measure the Hall voltage and the transverse magnetoresistance.

III. RESULTS AND ANALYSIS

A. The Hall effect

The Hall resistivity versus the applied magnetic field at 4.2 K (1,4), 77 K (2,5), and 300 K (3,6) is shown in Fig. 1 for stages 5 (1,2,3) and 9 (4,5,6). Throughout this work the field H was always parallel to the c axis. As shown in the figure, in stage 5 the Hall voltage is positive and nearly linear with the field at all temperature and field values indicating the dominance of hole carriers in this stage. Stage 9, however, exhibits a different character at room temperature since the Hall voltage fluctuates between positive and negative values. The Hall coefficient, thus, has a complicated field dependence at room temperature which might indicate the near equality of the electron and hole densities. This could be also described as a low- to high-field transition, where the condition $\omega_c\tau > 1$ holds for holes and does not for electrons. The Hall coefficient of HOPG (Refs. 8 and 9) at low temperatures exhibits similar behavior to that of stage 9 at room temperature. As shown in Fig. 1, at 4.2 and 77 K

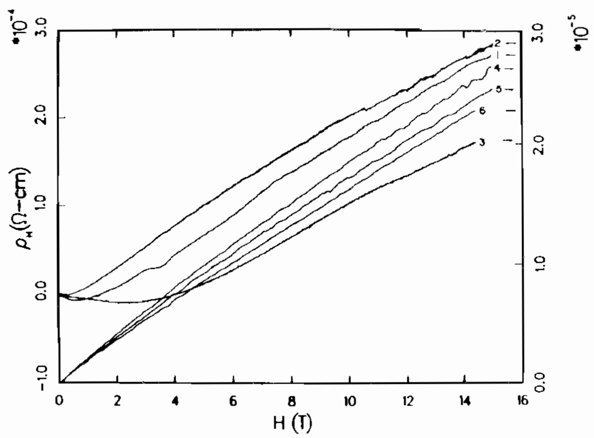

FIG. 1. The Hall resistivity vs the applied field of stages 5 and 9 at helium (1,4), nitrogen (2,5), and room temperatures (3,6), respectively.

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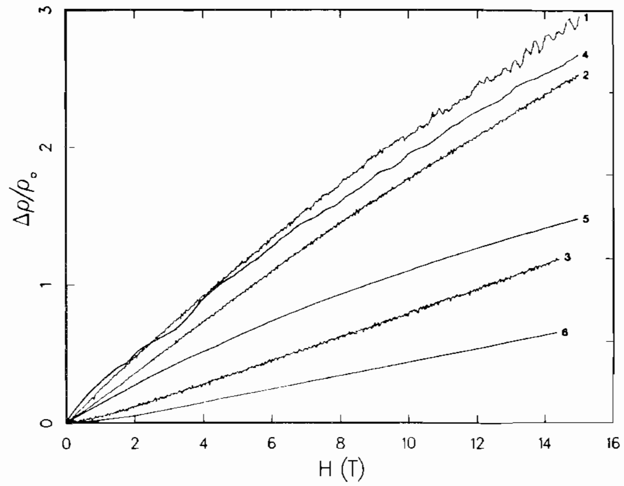

FIG. 2. The magnetoresistance versus the applied field of stages 5 and 9 at helium (1,4), nitrogen (2,5), and room temperatures (3,6), respectively.

temperatures, the Hall voltage is positive at all the field values for stage 9, however, it is not as linear in the field as that of stage 5. Therefore, the Hall coefficient of stage 5 has the least complicated field dependence relative to those of stages 2, 9, and HOPG (see Ref. 9 for data on HOPG and stage 2).

At this stage of our work, we have not performed a detailed investigation of the temperature dependence of the Hall coefficient or the magnetoresistance. However, qualitative information can be extracted from our preliminary results. The Hall resistivity data shown in Figs. 1 and 2 for stages 5 and 9 and those of HOPG and stage 2 (Ref. 9) indicate that the Hall coefficient of stage 5 is less sensitive to the variation with temperature relative to the other stages and HOPG as well. The Hall coefficient R_H for one- and two-carrier models, is related to the carrier mobilities μ_h, and μ_e, and to the carrier densities n_h and n_e by the following equations:

$$R_H = \frac{-1}{n_e ec} \quad \text{or} \quad \frac{1}{n_h ec}, \tag{1}$$

$$R_H = \frac{1}{n_e e}\left(\frac{n_h/n_e - (\mu_e/\mu_h)^2}{(n_h/n_e + \mu_e/\mu_h)^2}\right). \tag{2}$$

According to Eq. (1), the Hall coefficient R_H would be constant between 77 and 4.2 K only if the carrier concentration is constant in this temperature range. However Eq. (2), the two-carrier model, indicates that the constancy of R_H is controlled by the temperature dependence of the four quantities, n_h, n_e, μ_h, and μ_e, or at least the carrier concentration and the mobility ratio. It would be unrealistic to assume the constancy of these four quantities. Therefore, based on the field and temperature dependence of the Hall coefficient, the conduction in stage 5 is dominated by one kind of charge carrier. Our measurements of the c-axis conductivity in this system[10] indicate that stage 5 has the smallest c-axis conductivity of all the stages of $FeCl_3$-graphite compounds.

The behavior of the Hall data of stage 9 is different from that of stage 5; it has a complicated field and temperature dependence similar to that of HOPG. At room temperature the Hall voltage changes sign from negative to positive values at a field of about 4 T. Thus, the single-carrier model of Eq. (1) is not applicable at this temperature and one has to use the two-carrier model of Eq. (2) to qualitatively analyze these data. Because of the difference in the effective masses of the electrons and holes, one can expect the high field to drift holes more than electrons. Thus, at room temperature holes contribute dominantly to the Hall voltage in the high-field region. In terms of Eq. (2), this result can be described as a change of the inequality

$$\frac{n_h}{n_e} < \left(\frac{\mu_e}{\mu_h}\right)^2 < 1$$

in low field to

$$\frac{n_h}{n_e} > \left(\frac{\mu_e}{\mu_h}\right)^2$$

in high field. Thus, it is possible that one could have a positive Hall coefficient even when $n_h/n_e < 1$. At low temperatures the Hall voltage is dominantly contributed by holes and then exhibits very weak QO at helium temperature.

B. Magnetoresistance

The tranverse magnetoresistivity $\Delta\rho/\rho_0$ as a function of the applied field at 4.2 K (1,4), 77 K (2,5), and 300 K (3,6) is presented for stages 5 (1,2,3) and 9 (4,5,6) of $FeCl_3$-graphite compounds in Fig. 2. The value of $\Delta\rho$ increases with the field at all temperatures; there is no sign of saturation up to a field of 20 T in stage 5, but in stage 9 there is a weak indication of saturation at very high fields. At room and nitrogen temperatures the two stages exhibits similar field and temperature dependence, however, at helium temperature, although the over all behavior is still the same, the size of the change in $\Delta\rho$ is different. The magnetoresistance of both stages oscillates at helium temperature, but the magnitudes of the oscillations in stage 5 are relatively larger and faster than those of stage 9. The magnetoresistance of HOPG, and stages 2, 5, and 9 exhibit similar field and temperature dependance.

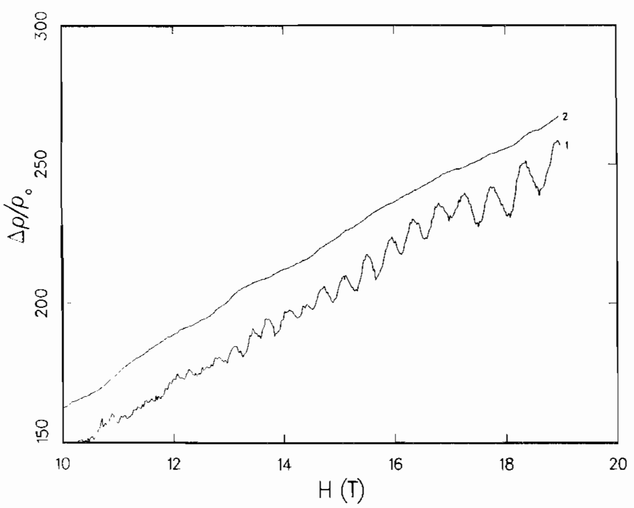

FIG. 3. The oscillations in the magnetoresistance versus the field at helium temperature of stage 5 (1) and stage 9 (2) (stage 5 data are multiplied by 70).

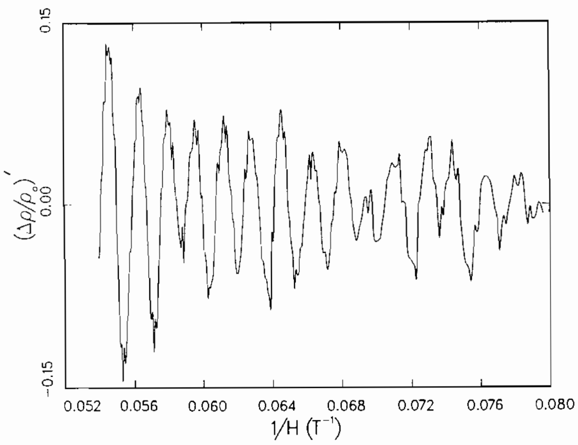

FIG. 4. The oscillations in the magnetoresistance versus the inverse field of stage 5 (nonoscillatory part is subtracted).

The similarities between the magnetoresistance data of HOPG and the three stages of $FeCl_3$ suggest that the transport mechanism in this system can be described in terms of a two-dimensional model. The scattering of the charge carriers is, thus, an in-plane mechanism controlled by the graphite lattice and the density of free carriers. The increase of the number of free carriers due to the charge transfer between the graphite and the intercalant is accounted for by the sharp decrease in the magnitude of $\Delta\rho$ of the three stages relative to that of HOPG. Magnetoresistance data are, generally, analyzed on the basis of a simple two-carrier model in which an average Hall mobility could be obtained from $\mu^* = (\Delta\rho/\rho_0 H^2)^{1/2}$. For conductors that obey a one-carrier model, the carrier density (n) determined by $ne\mu^*\rho_0 = 1$ should be equal to that determined by Eq. (1). However, these simple one- and two-carrier models fail to give adequate results for many GIC's. Details of these calculations will be given in a later paper.

The QO of the magnetoresistance of stages 5 and 9 are plotted as a function of the applied field in Fig. 3 (note that the data of stage 5 are multiplied by a factor of 70). As shown in the figure, the size of the oscillations relative to the base line in stage 5 is significantly larger than that of stage 9. The oscillations in the electric resistivity arise from the oscillations in the relaxation time for the scattering carriers. Because these oscillations are a manifestation of the periodic variation in the density of states at the Fermi energy, the large amplitude of the oscillations in stage 5 can be attributed to a longer in-plane relaxation time. In Fig. 4 the oscillations in stage 5 are plotted as a function of the inverse field. As shown in the figure, the fundamental frequency is modulated by other very weak harmonics. As it was pointed out in the previous section, this result is consistent with the Hall data which indicate that in stage 5 the transport mechanisms are dominantly controlled by one kind of charge carrier.

Fourier transforms of these oscillations are displayed in Fig. 5 for stages 5 and 9. The dominant frequency of stage 5 which is attributed to the hole oscillations occurs at about 588 T. In stage 9, as shown in the figure, oscillations of different frequencies are observed at 0.55, 4.4, and 8.8 T. Some high frequencies of insignificant amplitudes are also observed in stage 9. It is clear that some of these frequencies in stage 9 are due to the HOPG layers in the system. The large frequency of stage 5 indicates a similar large size in the extremal Fermi surface cross-sectional area which is a measure of the carrier density at the Fermi energy.

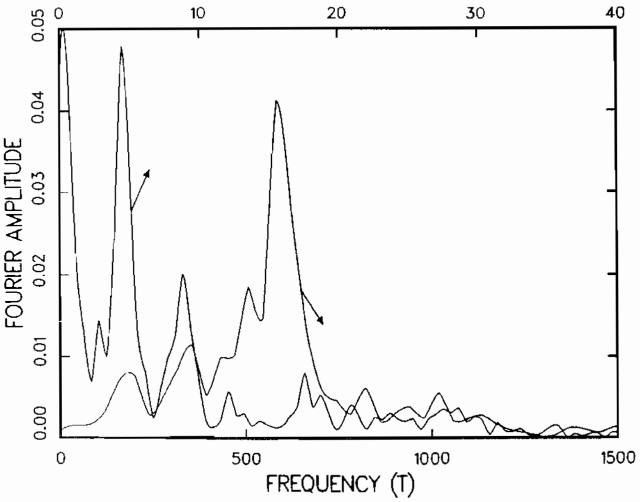

FIG. 5. Fourier transform of data obtained from stages 5 and 9 at helium temperature. The top x-axis refers to stage-9 data, where the bottom one refers to stage-5 data.

ACKNOWLEDGMENTS

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