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Investigation of interlayer coherency and angular-dependent magnetoresistance oscillations in magnetic graphite intercalation compounds

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Abstract

We investigated the interlayer coherency and angular-dependent magnetoresistance oscillations (AMROs) in magnetic graphite intercalation compounds (GICs), stage-1 $FeCl_3$ GIC (S = 5/2) and stage-5 $MoCl_5$ GIC (S = 1/2), where sufficiently strong π–d coupling is expected. Shubnikov–de Haas (SdH) oscillations were observable in a limited field-angle region. Standard AMRO was observed for both GICs, but the background of the angular-dependent magnetoresistance cannot be interpreted by the semi-classical model, based on the Boltzmann coherent transport theory. The resistance peak in magnetic fields parallel to the conduction layers is observed only for stage-5 $MoCl_5$ GIC, demonstrating coherent transport. The results are discussed in terms of magnetic scattering in the characteristic stacking structure of GICs.

Keywords: Transport measurements, Magnetotransport, Graphite and related compounds

1. Introduction

The angular effects of interlayer magnetoresistance have been extensively studied in layered quasi-one-dimensional (Q1D) or quasi-two-dimensional (Q2D) conductors [1]. In these conductors, the fundamental concept of electronic transport is based on the coherent motion of electrons in energy bands or Bloch states. However, in some cases, the simple semi-classical Boltzmann transport theory fails. Recent theoretical studies argue that the standard angular-dependent magnetoresistance oscillation (AMRO) could appear even in a system where interlayer transport is weakly incoherent [2,3]. Incoherent interlayer transport is expected when the intra-layer scattering rate (τ^{-1}) is much larger than the interlayer hopping integral (t_c). In this situation, the resistance peak in a magnetic field parallel to the layers is not observable because the interlayer conductivity is proportional to the tunneling rate between two adjacent layers, and the Fermi surface is only defined within the layers [2,3]. To investigate interlayer coherency, magnetoresistance measurements were performed in magnetic graphite intercalation compounds (GICs), since their interlayer coupling can be systematically controlled by the interaction between the conducting π-electrons in the graphene sheets and the localized magnetic moments [4,5]. Among various compounds, a $FeCl_3$ GIC is reported to show antiferromagnetic transitions of the large Fe *3d*-magnetic moments (S = 5/2) at T~4 K [4]. Such strong π–d coupling is expected to provide new insights into the mechanism of electronic transport. At various $MoCl_5$ GICs stages (S = 1/2) [6], interlayer coherency is also expected to be controlled by a change in the stage number (n). In this paper, we report the angular dependence of the magnetoresistance of two GICs, stage-1 $FeCl_3$ and stage-5 $MoCl_5$.

2. Experimental

Samples were synthesized from highly oriented pyrolytic graphite (HOPG, Union Carbide) for $FeCl_3$ and $MoCl_5$ GICs [4,6]. Resistance was measured by a conventional four-probe ac technique with electric current along the *c*-

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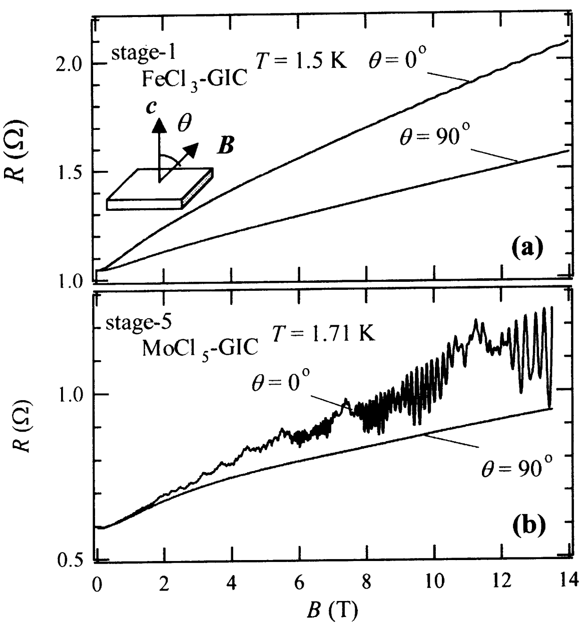

Fig. 1. Shubnikov–de Haas (SdH) oscillations of stage-1 $FeCl_3$ GIC at 1.5 K (a) and stage-5 $MoCl_5$ GIC at 1.71 K (b). Angle θ is defined in the inset.

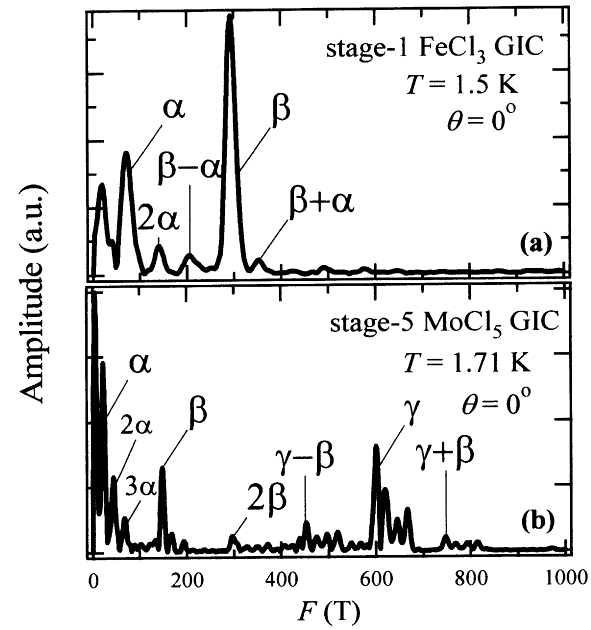

Fig. 2. Fourier transform spectra of the SdH oscillations for stage-1 $FeCl_3$ GIC (a) and stage-5 $MoCl_5$ GIC (b).

axis, which is perpendicular to the conduction (*ab*) plane. Four gold wires (Ø 10 μm) were attached to the samples by silver paste. The experiments were conducted using a ^{4}He cryostat with 15 T superconducting magnets with a sample rotator at NIMS.

3. Results and discussion

Fig. 1 shows the field dependence of the resistance of stage-1 $FeCl_3$ GIC at 1.5 K (a) and the stage-5 $MoCl_5$ GIC at 1.71 K (b). Both GICs show a ρ_c (θ = 0°) > ρ_c (θ = 90°) over the whole field region. At high magnetic fields, Shubnikov–de Haas (SdH) oscillations were successfully observed in both compounds. Fourier transform spectra of both SdH oscillations are shown in Fig. 2. For stage-1 $FeCl_3$ GIC, the spectrum shows the presence of two fundamental frequencies: α=75 T and β=300 T, which correspond to Fermi cylinder radii of k_α=0.0475 and k_β=0.0954 Å^{-1}, respectively, and their combined frequencies (Fig. 2a). For stage-5 $MoCl_5$ GIC, the spectrum shows three strong peaks at α=21.6 T, β=145 T and γ=588 T, which correspond to Fermi cylinder radii of k_α=0.026, k_β=0.066 and k_γ=0.134 Å^{-1}, respectively. There are also other peaks, however, they can be interpreted as harmonics and combined frequencies (Fig. 2b). Such low frequencies can be ascribed to the small closed pockets formed by the reconstruction of the original π-bands of the graphene layers due to the formation of a superlattice of the intercalants. The SdH oscillations are observed only for θ <20° for stage-1 $FeCl_3$ GIC, where θ is the angle between the magnetic field and the *c*-axis, described in the inset to Fig. 1. Similarly to stage-1 $FeCl_3$ GIC, the amplitude of the SdH oscillations for stage-5 $MoCl_5$ GIC rapidly decreases with increasing θ. The angle dependence of the SdH oscillations is well explained by the 1/cos θ dependence, suggesting that all frequencies arise from the Q2D cylindrical Fermi surfaces. These experimental results show that the AMRO would be observed in these systems.

Fig. 3 shows the angular-dependent magnetoresistance of stage-5 $MoCl_5$ GIC for different magnetic fields at T =1.71 K. The oscillatory behavior in the low-angle region is caused by SdH oscillations which remain until T ~ 30 K. At higher angles, a series of distinct peaks are identified. Since these peaks show no magnetic field dependence, they are ascribed to an AMRO effect, which was firstly discovered in a Q2D organic conductor [1]. The standard AMRO model predicts that the peaks appear at the angles defined by [2,3]:

$$\gamma \tan\theta_n = (n - 0.25)\pi \qquad (1)$$

where γ (= $l_c k_F$ for Q2D systems) and n are the geometric factor and arbitrary integer number, respectively. Although this formula has been first derived for the Q2D coherent systems, it is even valid for the Q2D weakly incoherent systems [2,3]. When we insert l_c = 22.83 Å for stage-5 $MoCl_5$ GIC [6], we can evaluate the radius of the Fermi cylinder as k_F ~ 0.13 Å^{-1}. The Fermi cylinder radius of 0.13 Å^{-1} coincides with k_γ, determined by SdH measurements (*B*//*c*-axis). Another remarkable feature in Fig. 3 is that an anomalous magnetoresistance background and peaks at θ=90° are both observed for stage-5 $MoCl_5$ GIC. Fig. 4 shows angular dependent magnetoresistance for B=13.5 T at various temperatures. With increasing temperature, the anomalous background is robust, while the peak at θ = 90° is suppressed. Stage-1 $FeCl_3$ GIC also shows an AMRO effect and an anomalous magnetoresistance background;

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