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ANGULAR DEPENDENT MAGNETORESISTANCE OSCILLATION IN STAGE-2 IBr GRAPHITE INTERCALATION COMPOUND*

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Abstract – The angular dependent magnetoresistance oscillation (ADMRO) of the stage-2 IBr graphite intercalation compound (GIC) was studied between 1.8 K and 110 K in magnetic fields between 1 and 9 T. There was a series of peaks in the c-axis resistance as the field direction was changed from the c axis to the (001) plane. The field independence of the angular positions of the peaks, showing that the oscillation is not from the Shubnikov-de Haas effect, and the presence of the oscillation at high temperatures indicate that the effect is semiclassic. The location and the relative amplitudes of the peaks show that the ADMRO in this compound does not follow the predictions for the symmetrical corrugation model of the cylindrical Fermi surface. The reason is attributed to the presence of two cylindrical Fermi surfaces and zone folding of the Fermi surface by the periodicity of the IBr lattice in this compound.

Keywords – Graphite intercalation compound, ADMRO effect

1. INTRODUCTION

There is an angular dependent magnetoresistance oscillation (ADMRO) in two dimensional conducting materials. It is observed in the c-axis resistivity as the magnetic field direction is changed between the c axis and the (001) plane and consists of a set of peaks in this rotation. Two-dimensional conducting planes have a straight cylindrical Fermi surface directed perpendicular to the conducting planes. A coupling between the planes causes a small undulation or corrugation along the cylindrical Fermi surface. The peaks in the magnetoresistance occur at magnetic field directions at which the cross section of the Fermi surface in planes perpendicular to the magnetic field has the same area along the whole Fermi surface. As such, it gives information about the Fermi surface and the type of conduction, as well as being a useful tool for understanding conduction in two-dimensional materials.

The ADMRO was observed first in the organic conductor θ-$(BEDT\text{-}TTF)_2I_3$ [1], followed by a report for β-$(BEDT\text{-}TTF)_2IBr_2$ [2] which has a quasi-two dimensional electronic structure. The oscillations were periodic in tanθ, where θ is the direction between the magnetic field and the direction normal to the two-dimensional plane. The angular positions of the oscillation were independent of the magnetic field intensity, while the amplitude of the oscillation depended weakly on the temperature of the sample. The ADMRO has also been observed in $(TMET\text{-}STF)_2BF_4$, $(TMTSF)_2ClO_4$, and β”-$(ET)_2SF_5\text{-}CH_2CF_2SO_3$, in which information about the Fermi surface was derived [3-5].

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The two-dimensional nature of the GaAs/Al_xGa_{1-x} as an artificial semiconductor superlattice also supports the ADMRO [6]. However it was relatively weak because the heavy doping necessary to make the metallic carrier density degraded the carrier mobility. The observation did show that ADMRO can occur in a general family of quasi-two dimensional metals. Thus, it can occur in graphite intercalation compounds (GIC's) that are quasi- two dimensional and have a large resistance anisotropy. It was observed in stage-2 $SbCl_5$ GIC [7], however there were some deviations from the classical behavior of the ADMRO effect. It was also observed in the $CuCl_2$ and $CoCl_2$ GIC's [8], but the behavior differed from the standard model. Thus it is of interest to investigate the ADMRO effect in other GIC's to test the standard model of ADMRO and to obtain information about their Fermi surfaces.

It has been shown that the stage-2 IBr GIC has a resistance ratio of 10^5 between the c axis and the (001) plane and is pseudo two dimensional [9]. An investigation of the c-axis magnetoresistance as a function of magnetic field direction was undertaken to obtain ADMRO for the purpose of testing the theory of ADMRO and the Fermi surface of the GIC. It was shown from the de Haas van Alphen effect that zone folding by the periodicity of the IBr lattice results in many Fermi surface pieces [10]. One question is: how does this zone folding affect ADMRO? The purpose was also to determine whether c-axis conduction was by a hopping mechanism or through the conduction band. If the conduction in a GIC is mainly governed by the non-intrinsic hopping process, the electronic transport properties of the compound simply reflect two dimensional electronic states of each graphite sheet, and therefore the c-axis magnetoresistance is dependent on the magnetic field component along this axis. But, if the carriers are conducted through the conduction band, the large, but finite c-axis resistivity of the compound indicates that the Fermi surface should not be too far from a cylindrical shape with the symmetry axis parallel to the c axis. In this case, the angular dependence of the magnetoresistance is influenced by a small corrugation of the cylindrical Fermi surface.

The standard model for the ADMRO uses a cylindrical Fermi surface with an undulation along the k_z direction perpendicular to the conducting plane. As Yamaji [11] pointed out, there is a periodic variation in the cross-sectional area of the Fermi surface as the magnetic field is rotated from the c axis which results in a $\tan\theta$ dependence of the magnetoresistance. Yagi et al [12] showed that this is a semiclassical effect rather than the quantum effect that results in the de Haas Shubnikov effect.

Yagi et al [13] calculated the ADMRO for four symmetries of the undulation of the Fermi surface and showed that the behavior depends markedly on the corrugation symmetry of the cylindrical Fermi surface. In particular, some types of corrugation symmetries yield an inverted peak and valley structure which may have some relevance to the inverted peak structure of the ADMRO experimentally observed in some organic conductors.

2. EXPERIMENTAL METHOD

The stage-2 IBr GIC was produced by placing highly oriented pyrolytic graphite (HOPG) and IBr in a sealed reaction tube and heating it to a temperature of 50-55 °C with the graphite and intercalant at the same temperature for two weeks. The prepared sample had a thickness of approximately 0.3 mm and a surface of 2mmx3mm. It was characterized by (00l) x-ray diffraction. The spectrum revealed a pure staged sample with no diffraction peaks from mixed stages with a periodic repeat distance Ic of 10.5 A. The electrical contacts for c-axis resistivity measurements were formed with silver paste and the measurements made with the two current contacts with the two potential contacts on opposite sides of the cleaved surface and one pair at each end of the sample. The magnetoresistance measurements were taken with an Oxford Instruments Mag Lab EXA system with a fixed current of 10 mA and magnetic fields up to 9 T.

The sample was rotated between -90° to +90° in steps of 1 degree with respect to the field direction. The temperature was controlled with an uncertainty of 0.01 K at temperature between 1.8 K and 110 K.

3. EXPERIMENTAL RESULTS

The data of the c-axis resistivity were taken by rotating the sample between -90° and +90° with respect to the field direction in a plane perpendicular to (001). At low fields there is a cos θ dependence with a maximum at θ=0, a minimum near θ=±90°, and symmetry with respect to the c axis as shown in Fig. 1 for a field of 1 T. At high fields there is considerably more structure as shown in Fig. 2 for a field at 9 T. There is still a maximum at 0°, but as the magnetic field approaches 4.4 T, another peak appears at ±57°.These peaks become more predominant as the field is increased and two new peaks begin to appear at ± 33° and ± 76°, as the field is increased to 9 T (as shown in Fig. 3). However the directions of the resistance maxima are independent of the magnetic field strength.

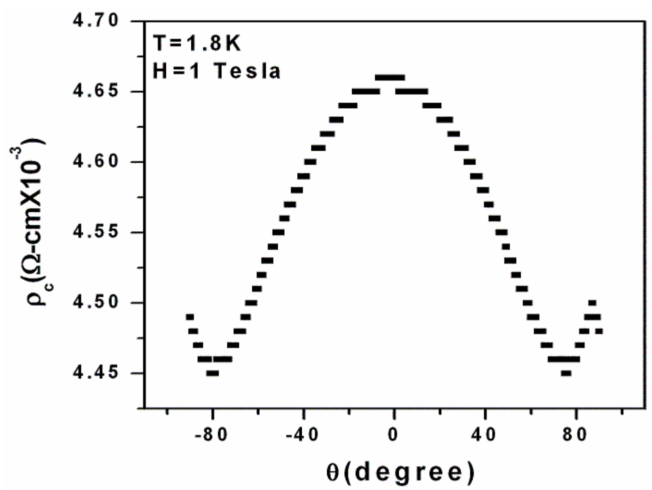

Fig. 1. Angular dependence of the c-axis resistivity of stage-2 IBr GIC at 1.8 K and 1 T

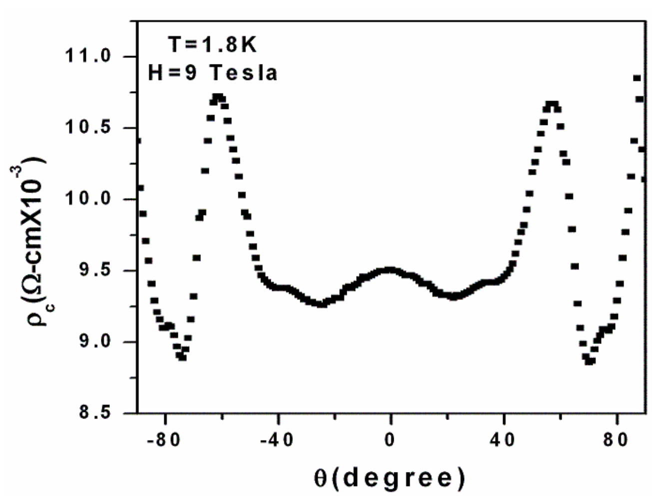

Fig. 2. Angular dependence of the c-axis resistivity of stage-2 IBr GIC at 1.8 K and 9 T

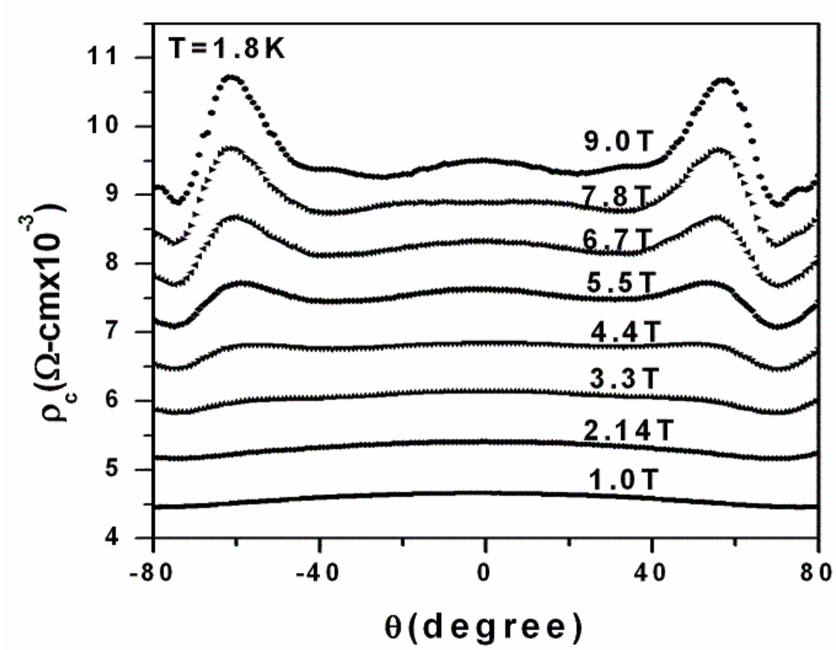

Fig. 3. Angular dependence of the c-axis resistivity of stage-2 IBr GIC at 1.8 K for fields between 1 T and 9 T

The structure exists over a wide range of temperature. The small peaks become less distinct and the maxima a little smaller as the temperature is increased. Data for 5 K and 110K are compared in Fig. 4. The resistivity increases linearly with magnetic field strength as shown in Fig. 5 for the largest peak.

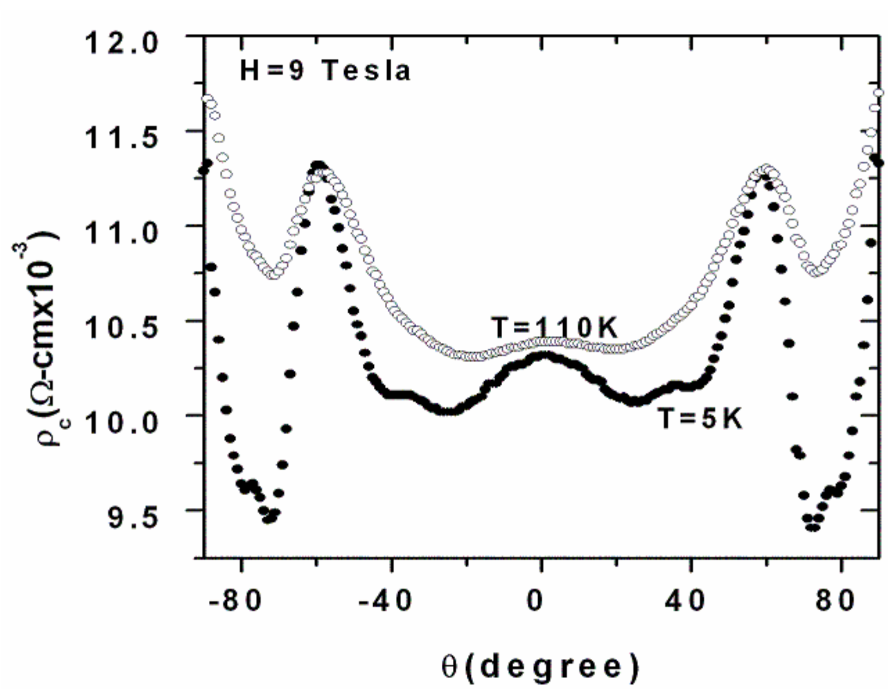

Fig. 4. Angular dependence of the c-axis resistivity of stage-2 IBr GIC at 9 T and at temperatures of 5 K and 110 K

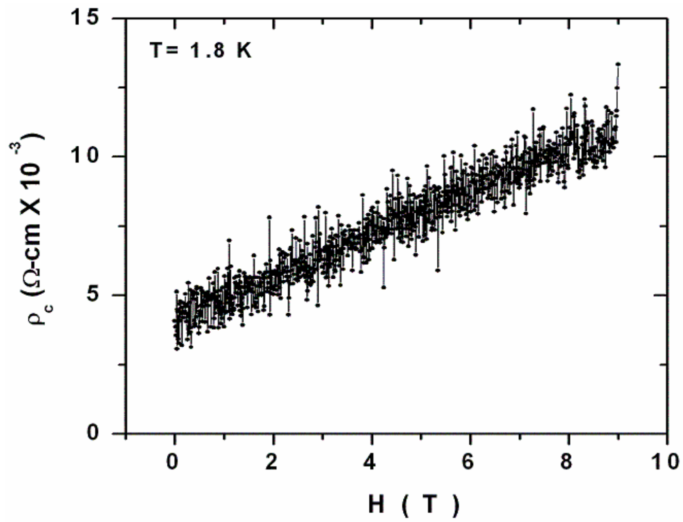

Fig. 5. Magnetic field dependence of the c-axis resistivity at 1.8 K and $\theta=57^{o}$

4. DISCUSSION

The ADMRO effect is observed in two-dimensional compounds at high magnetic fields [12]. At low fields the effect of a corrugated Fermi surface in the magnetoresistance is negligible, and therefore the component of the magnetic field along the c axis has a major effect on the magnitude of the resistivity in two-dimensional systems. In this case, the angular dependence of the magnetoresistance has a cos θ dependence. The magnetoresistance of the stage-2 IBr GIC shows a maximum at 0^{o} and a minimum close to $\theta = \pm 90^{o}$ at 1T as shown in Fig. 1. As is seen from the Fig., there is a little evidence of the ADMRO effect on the experimental results at this magnetic field. Therefore, little ADMRO behavior is expected below 1 T in this compound. In comparison with the other GIC's it is interesting to mention that the ADMRO effect appears above 2 T in stage-2 $CoCl_2$ GIC [8], and is not detected in stage-2 $FeBr_2$ GIC, even at 9 T [14].

The presence of the structure in the angular dependence of the magnetoresistance in stage-2 IBr GIC at a high magnetic field (shown in Fig. 2) is attributed to a semiclassical effect for the following reasons. The angular values of the maxima are independent of the magnetic field. This rules out the Shubnikov-deHaas (SdH) effect as a cause, because the period of the SdH oscillation depends on the magnetic field. Secondly, the fact that the structure is observed over a wide range of temperatures from 1.8 K to 110 K means that it is not a quantum effect from Landau levels which would be washed out by kT broadening at high temperatures. Thirdly, there is a linear magnetic field dependence in the magnetoresistance without any indication of SdH oscillations up to 9 T, as presented in Fig. 5.

The ADMRO effect in stage-2 IBr GIC also establishes that the conduction of the carriers along the c axis is mainly governed by the conduction band and the hopping process, which has a major role in the conductivity of stage-2 $FeBr_2$ GIC [14], but has very little contribution in this compound.

For a Fermi surface described by

$$E_k=(k_x^2+k_y^2)/2m-2t\cos(I_c\, k_z) \qquad (1)$$

Yamaji [11] determined the angular dependence of the ADMRO effect and showed that the positions of the peaks of the magnetoresistance satisfy

$$I_c k_f \tan\theta = \pi(n-1/4) \quad (2)$$

where n is an integer, k_f is the radius of the Fermi cylinder, and t in equation 1 is the interlayer transfer integral.

Carriers in stage-2 IBr GIC , like the other stage-2 GIC's, are in two different energy bands according to the theory of Blinowski et al [15]. Two de Haas van Alphen (dHvA) frequencies of 455T and 1106 T were identified with these bands [10]. The Fermi surface consists of two cylinders directed along the c axis from theory and experiment. The radius of the cross sections are $k_1=0.118 A^{-1}$ and $k_2=0.183\ A^{-1}$ from the dHvA measurements. Since both cylinders can contribute to the ADMRO effect, its interpretation is not so simple. A second complication arises from the fact that the IBr has an ordered structure with a cell ($\sqrt{183}R26.33^o X 2\sqrt{3}R0^o$) [16, 17]. Translation of the Fermi surface cylinders by this periodicity results in zone folding. The folded Fermi surface was observed with up to 30 dHvA frequencies [10]. It has to be determined whether folding affects the ADMRO effect.

The directions of the magnetic field at the maxima at 9 T and at 1.8 K are measured to be 33^o, 57^o, and 76^o. The theoretical prediction of equation 2 is 62^o, 77^o, and 82^o for cylinder 1 with $k_1=0.118\ A^{-1}$ and 51^o, 71^o, and 77^o for cylinder 2 with $k_f=0.183\ A^{-1}$ for n=1-3, respectively. Since these are not in agreement with the experimental values, equation 2 is generalized to

$$I_c k_f \tan\theta = \pi(n-\varphi) \quad (3)$$

where φ is an experimentally determined phase factor. This is reasonable because φ depends on the type of corrugation of the Fermi surface according to Yamaji [11] and Iye [7] and was found to have a value of 0.39 in the ADMRO investigation of stage-2 $SbCl_5$ GIC [7]. The best fit of equation 3 to θ_{exp} is for φ= 0.6 for $k_2=0.183\ A^{-1}$ giving values of 33^o, 66^o, and 76^o. The prediction for the Fermi surface cylinder with $k_1= 0.118\ A^{-1}$ is not in agreement, and this surface does not appear to contribute to the ADMRO.

The amplitude of the ADMRO peaks is also interesting to investigate. It is small for 33^o and 76^o peaks and is very large for the 57^o peak. The relative amplitudes depend on the corrugation symmetry of the cylindrical Fermi surface according to Yamaji [11] and Iye [7].

The s-type corrugation as given by equation 1 results in the largest peak for n=1 and decreasing size as n increases. This is not observed here. For a p-type corrugation there is an inverted peak and valley structure which is also not observed here. The d_{xy}-type corrugation has a 4-fold symmetry, while the d_{xx} type corrugation exhibits a 2-fold symmetry around the Fermi surface. Thus, the relative amplitudes of the peaks depend on the Fermi surface corrugation, but are different from what is observed here. However, since the ADMRO depends so much on the corrugation symmetry, it is plausible that the effect can be explained by some sort of corrugation or undulation of the Fermi surface. This could be fundamental to the basic cylinder or caused by the zone- folding process. Thus the theoretical investigation of this compound is more complicated and a simple model of the geometrical configuration of the Fermi surface is not sufficient to explain the ADMRO effect.

5. CONCLUSIONS

The ADMRO effect was observed in stage-2 IBr GIC above 1 T where a series of peaks in the c-axis resistance appeared as the field direction was rotated from the c axis to the (001) plane. The field independence of the angular values of the maxima showing that the oscillation is not from the SdH effect and the presence of the ADMRO at high temperatures (up to 110 K) indicate that the effect is entirely a semiclassical effect. The location of maxima at 33^o, 57^o and 76^o and the relative amplitude of the peaks at low temperature and high magnetic field show that the ADMRO in stage-2 IBr GIC does not follow the predictions for the symmetrical corrugation model of the cylindrical Fermi surface as presented by Yamaji [11] and Iye [7]. This model may not be applicable for the stage-2 IBr GIC because there are two cylindrical Fermi surfaces as predicted by Blinowski et al [15] and there is zone folding of the Fermi surface by the periodicity of the IBr lattice. However, by generalizing the equations for the corrugation model to equation 3, a best fit is obtained for the phase angle of 0.6 for the carriers in the larger cylindrical Fermi surface, indicating that the carriers in the other Fermi cylinder do not appear to contribute to the ADMRO in the stage-2 IBr GIC.

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