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# Two-Dimensional Weak Localization Effect in Stage-4 MoCl<sub>5</sub> Graphite Intercalation Compound

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The c-axis resistivity  $\rho_c(H, T)$  of stage-4 MoCl<sub>5</sub> GIC between 1.9 and 50 K has been measured with and without an external magnetic field along the c axis (0 $\leq$ H $\leq$ 44 kOe). The interior G layers form a bottleneck to the c-axis conduction. The T and H dependence of  $\rho_c$  is mainly determined from that of the bottleneck resistivity proportional to the in-plane resistivity of interior graphite layers. A logarithmic behavior of  $\rho_c$  in the form of In(T) and In(H) indicates that the two-dimensional weak localization occurs in the interior graphite layers. The T and H dependence of the in-plane conductivity derived from  $\rho_c$  is discussed in terms of scaling relation predicted from the theory of two-dimensional weak localization.

Keywords: weak localization; negative magnetoresistance; graphite intercalation compounds

#### INTRODUCTION

In a previous paper [1], we have reported the temperature (T) dependence of the c-axis resistivity ( $\rho_c$ ) for stage-2 to 6 MoCl<sub>5</sub> GICs in the presence of an

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external magnetic field H along the c axis. We have found (i) a metallic behavior in stage 2, (ii) a logarithmic behavior at low T in stage 3 and 4, and a negative magnetoresistance (n-MR) at low T and weak H in stage 3 to 5, and (iii) a semi-conductor-like behavior in high stages (5, 6). We have shown that these results can be qualitatively explained within the framework of a two-dimensional (2D) band model with hopping conduction mechanism. The logarithmic behavior and negative magnetoresistance are due to a 2D weak localization effect (WLE) occurring in the interior graphite (G) layers [2]. In this paper we have undertaken an extensive study on the T dependence and H dependence of  $\rho_c$  for stage-4 MoCl<sub>5</sub> GIC, where the magnetic field is applied along the c axis. These results are discussed in the light of the theories on 2D WLE.

#### EXPERIMENTAL PROCEDURE

The c-axis resistivity  $\rho_c$  was measured using a system of SQUID magnetometer (Quantum Design, MPMS XL-5) with an external device control and an ultra low field capability option. Before setting up a sample at 298 K, a remanent magnetic field in a superconducting magnet was reduced to one less than 3 mOe using an ultra low field capability option. For convenience, hereafter this remanent field is noted as the state of H=0. The sample was cooled down to 1.9 K for typically 8 hours for a annealed case and 0.5 hours for a quenched case. Then the c-axis resistivity was measured with increasing T from 1.9 to 50 K with and without H along the c axis.

#### RESULT

The value of  $\rho_c$  for the quenched system is relatively larger than that for the annealed system for the same T and H, reflecting the nature of disordered state in graphite layers. Figures 1(a) and (b) show the T dependence of  $\rho_c$  for the annealed system in the presence of H along the c axis. Similar behavior is observed for the quenched system. Figure 2 shows the H dependence of  $T_{min}$  for both the annealed and quenched systems. The temperature  $T_{min}$  decreases with increasing H at low H, exhibiting a local minimum around H = 2 kOe, and increases with further increasing H:  $T_{min}$  = 46 K at H = 20 kOe.

In order to analyze the T dependence of  $\rho_c$  in the presence of H, we assume that the resistivity is described by

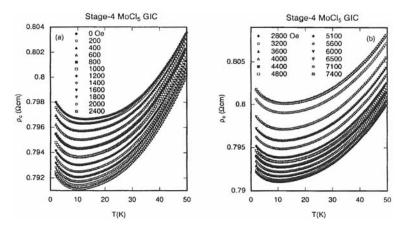


FIGURE 1 (a) and (b) T dependence of  $\rho_c$  with various H for stage-4 MoCl<sub>5</sub> GIC in the annealed system. H // c.

$$\rho_{c} = \rho_{0} - \rho_{1} \ln(T) + \rho_{2} T + \rho_{3} T^{2}$$
 (1)

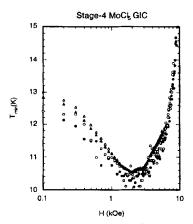


FIGURE 2 H dependence of  $T_{min}$ . Experimental values of  $T_{min}$  for the annealed system ( $\bullet$ ) and the quenched system ( $\circ$ ). Calculated values of  $T_{min}$  for the annealed system ( $\triangle$ ) and for the quenched system ( $\triangle$ ).

where  $\rho_0$ ,  $\rho_1$ ,  $\rho_2$ , and  $\rho_3$  are positive constants dependent on H, the second logarithmic term is due to the 2D WLE, and the third and fourth terms are due to the scattering of carriers by phonons. The value of  $T_{min}$  can be calculated from  $d\rho_c/dT=0$ .

We find that the data of  $\rho_c$  vs T with  $1.9 \le T \le 25$  K for both the annealed and quenched systems can be well described by Eq.(1) for each H below 5.6 kOe. The parameters  $\rho_1$  to  $\rho_4$  for each H are determined by the least squares fit of the data to Eq.(1): for example,  $\rho_0 = 0.79869 \pm 0.00001 \ \Omega cm, \rho_1$ 

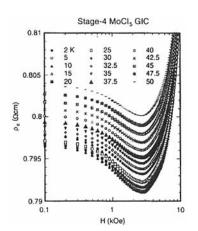


FIGURE 3 H dependence of  $\rho_c$  for stage-4 MoCl<sub>5</sub> GIC in the annealed system at various T. H // c.

=  $(1.038 \pm 0.024) \times 10^{-3} \Omega \text{cm}$ ,  $\rho_2$ =  $(1.863 \pm 0.534) \times 10^{-5} \Omega \text{cm K}^{-1}$ , and  $\rho_3$  =  $(2.269 \pm 0.120) \times 10^{-6} \Omega \text{cm K}^{-2}$  at H = 0 for the annealed system. In Fig.2 we also show the calculated values of  $T_{\text{min}}$  for both the annealed and quenched systems. The experimental data of  $T_{\text{min}}$  vs H are in good agreement with the calculated ones.

The ratio  $\rho_1/\rho_0$  tends to increase slightly with increasing H:  $\rho_1/\rho_0 = 0.0013$  at H = 0 to 0.00138 at H = 5.6 kOe for the annealed system. In the previous paper [1] we have shown that the following

relation is valid in stage-4 MoCl<sub>5</sub> GIC:  $\rho_1/\rho_0 = (e^2/2\pi^2\hbar \sigma_{2D}^0)(\alpha p + \gamma)$ . Note that  $\sigma_{2D}^0$  (= 1.0776 x 10<sup>-2</sup>  $\Omega^{-1}$ ) corresponds to the in-plane conductivity defined by  $I_0/\rho_0^1$ , where  $\rho_0^0$  (= 18  $\mu\Omega$ cm at 4.2 K) [3] is the in-plane resistivity and  $I_c$  (= 19.396  $\pm$  0.014 Å) is the c-axis repeat distance for stage-4 MoCl<sub>5</sub> GIC. Using the ratio  $\rho_1/\rho_0 = 1.30$  x  $10^{-3}$  the constant  $(\alpha p + \gamma)$  can be estimated as 1.14, which is comparable with 0.90 in the previous paper [1].

Figure 3 shows the H dependence of  $\rho_c$  with  $1.9 \le T \le 50$  K for the annealed system. For each T  $\rho_c$  decreases with increasing H at low H, exhibits a local minimum around H = 2.5 kOe, and  $\rho_c$  increases with further increasing H. The sign of the difference  $\Delta \rho_c$  [= $\rho_c$ (T,H) -  $\rho_c$ (T,H=0)] is negative for  $0 \le H \le 6 - 7$  kOe due to the 2D WLE. Because of the Boltzmann term  $\sigma_B^{2D}$  which may drastically decrease with increasing H above 6 - 7 kOe the sign of  $\Delta \rho_c$  becomes positive. For  $0.6 \le H \le 2.0$  kOe the H dependence of  $\rho_c$  is described by a logarithmic term [ $\rho_c = b_1 - b_2 \ln(H)$ ], due to the 2D WLE. The ratio  $b_1/b_0$  is dependent on T. It decreases with increasing T:  $b_1/b_0 = (3.93 \pm 0.05) \times 10^{-3}$  at 1.9 K and (2.14  $\pm 0.05) \times 10^{-3}$  at 50 K.

#### DISCUSSION

We assume that the c-axis resistivity of stage-4 MoCl<sub>5</sub> GIC is proportional to the in-plane resistivity tensor of the interior G layers  $(\rho_{xx})$ , where  $\rho_{xx} = A\rho_c$  and A is constant. Then the correction term of the conductivity tensor (X) in the interior G layers can be expressed by

$$X = \frac{\Delta\sigma(T, H)}{\sigma(T, H = 0)} = \frac{\rho_c(T, H = 0)}{\rho_c(T, H)} \frac{1}{1 + \frac{\lambda^2 H^2}{[\rho_c(T, H)]^2}} - 1,$$
 (2)

where  $\lambda = R_H/A$  and  $R_H$  is the Hall coefficient defined by  $R_H = 1/nec = 6.25 \times 10^{10}/n$ . The carrier density n for stage-4 MoCl<sub>5</sub> GIC is related to  $N_{2D}$  by  $n = N_{2D}/I_c (= 5.16 \times 10^{20}/cm^3)$ , where  $N_{2D}$  is typically  $10^{14}/cm^2$  for the 2D band model [4]. Using the value of A (=  $\rho_a/\rho_c = 18 \mu\Omega cm/0.8 \Omega cm = 2.25 \times 10^{-5})$  [3], we have  $\lambda = 6.25 \times 10^{10}/An = 5.39 \times 10^{-6}$ .

The value of  $\lambda$  for stage-4 MoCl<sub>5</sub> GIC can be determined as follows. First we calculate the H dependence of X for 1.9 $\leq$ T $\leq$ 50 K when the parameter  $\lambda$  is changed between  $\lambda = 2 \times 10^{-6}$  and  $2 \times 10^{-5}$  around the expected value  $\lambda = 5.39 \times 10^{-6}$ . For each  $\lambda$  we find that the H dependence of X at T = 2.0 K in the limited

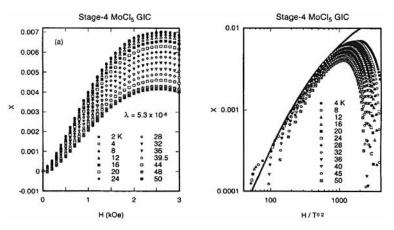


FIGURE 4 (a) H dependence of the conductivity X with  $\lambda = 5.3 \times 10^{-6}$  for various T. (b) Scaling plot of X with  $\lambda = 5.3 \times 10^{-6}$  as a function of H/TP (p = 0.2) for various T. The solid line is described in the text.

field range between 0.5 kOe and 2 kOe can be well fitted to a logarithmic form defined by  $c_1 + c_2 \ln(H)$ . Then we make a plot of  $c_2$  as a function of  $\lambda$ . The value of  $c_2$  linearly decreases with  $\lambda$  for  $3 \times 10^{-6} \le \lambda \le 7 \times 10^{-6}$ :  $c_2 = 0.00413 - 37.65\lambda$ . An appropriate value of  $\lambda$  (= 5.30 x 10<sup>-6</sup>) is chosen under the condition that  $c_2$  coincides with the value of  $b_1/b_0$  (= 3.93 ± 0.05) x 10<sup>-3</sup> at 1.9 K. This value of  $\lambda$  is very close to the expected value of  $\lambda$  (= 5.39 x 10<sup>-6</sup>).

Figure 4(a) shows the H dependence of X with  $\lambda = 5.30 \text{ x } 10^{-6}$  at various T. The value of X is positive at least below 7 - 8 kOe and has a peak around H = 2.6 - 2.7 kOe, which are independent of T for 1.9 $\leq$ T $\leq$ 50 K. For any fixed H the value of X decreases with increasing T. Figure 4(b) shows the log-log plot of X with  $\lambda = 5.30 \text{ x } 10^{-6}$  as a function of H/TP with p = 0.2, where p is the exponent of relaxation time for inelastic scattering  $\tau_E$ :  $\tau_E \approx \text{T}^{-p}$ .

Here we discuss the H and T dependence of X in terms of the theory of 2D WLE. The conductivity is predicted to be expressed by a scaling function  $f(H/H_{\rm E})$ , where  $f(x) = \ln(1/x) - \Psi(1/2 + 1/x)$ ,  $\Psi(x)$  is a digamma function, and  $H_{\rm E}$  is proportional to  $1/\tau_{\rm E}$ . As shown in Fig.4(b) it seems that almost all the data of X with  $\lambda = 5.3 \times 10^{-6}$  for T<34 K fall on the solid line described by a scaling function  $X = -B f(H/H_{\rm E})$  for  $H/T^{0.2}$ <800 where B = 0.0074 and  $H_{\rm E} = \beta TP$  with  $P = 0.20 \pm 0.02$  and P = 90. For  $P = 0.20 \pm 0.02$  and P = 90. For  $P = 0.20 \pm 0.02$  and P = 90. For  $P = 0.20 \pm 0.02$  and P = 90. For  $P = 0.20 \pm 0.02$  and P = 90. For  $P = 0.20 \pm 0.02$  and P = 90. For  $P = 0.20 \pm 0.02$  and P = 90. For  $P = 0.20 \pm 0.02$  and P = 90. For  $P = 0.20 \pm 0.02$  and P = 90. For  $P = 0.20 \pm 0.02$  and P = 90. For  $P = 0.20 \pm 0.02$  and P = 90. For  $P = 0.20 \pm 0.02$  and P = 90. For  $P = 0.20 \pm 0.02$  and P = 90. For  $P = 0.20 \pm 0.02$  and P = 90. For  $P = 0.20 \pm 0.02$  and P = 90. For  $P = 0.20 \pm 0.02$  and P = 90.

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