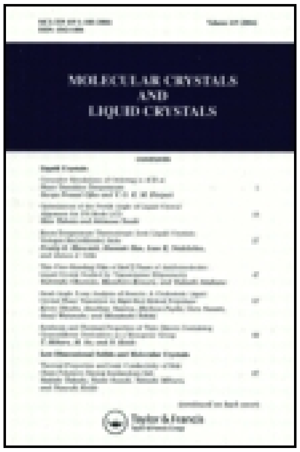

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Theory of the c-Axis Conduction and Longitudinal Negative Magnetoresistance in $MoCl_5$ GICs

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A general theory on the c-axis resistivity ρ_c and longitudinal magneto-resistance $\Delta\rho_{cL}/\rho_0$ in graphite intercalation compounds (GICs) is presented and theory provides a reasonable explanation for the observed results of $MoCl_5$ GICs.

Keywords: graphite intercalation compounds; c-axis conduction

1. INTRODUCTION

In the past two decades, extensive studies were performed for various kinds of graphite intercalation compounds(GICs), however, the mechanism on the c-axis conduction is still in a controversy[1]. We focus on the GICs with large A-value, where $A = \rho_c/\rho_a$ lies over a range of 10^3~10^6 in acceptor GICs.

Our view on the c-axis conduction mechanism is as follows[2,3]. Carriers in the π-band spend most of the time in the diffusive motion along the basal plane and occasionally make transitions to the adjacent graphite(G) layers with or without the intervening intercalate(I) layers. In the low stage GICs (stage n=1,2) the transition rate is given by $1/\tau \approx (H_{tr})^2/\hbar\Gamma$, where H_{tr} is the transfer Hamiltonian across the intercalant layers and $\Gamma/\hbar$ denotes the scattering rate in the basal plane. In the case of H_{tr} being temperature independent the above relation explains the metallic T-dependence of ρ_c in the low stage since $\rho_c \propto \Gamma$.

To come to a definite conclusion on the c-axis conduction mechanism, theoretical investigations are performed in this article. In Sec.2 the formula of ρ_c for stage-1 and 2 is derived and Sec.3 is devoted to the calculations on ρ_c and the longitudinal magnetoresistance for intermediate stage compounds.

2. c-Axis Resistivity in Low Stage GICs

The c-axis conductivity $\sigma_c = \rho_c^{-1}$ is related to the diffusion constant D_c as

$\sigma_c = e^2 D_c N(E_F)/V$; V: sample volume, N: density of states, (2.1)

where E_F is the Fermi energy and $D_c = \ell_c^2/\tau_c$. ℓ_c denotes the diffusion length along the c-axis and $\ell_c = I_c$ (repeat distance) for stage-1. As discussed in Sec.1 $1/\tau_c$ is given by

$$1/\tau_c \approx (H_{tr})^2/\hbar\Gamma \approx (N_I/N)V_0^2/\hbar\Gamma, \tag{2.2}$$

where the transfer Hamiltonian is assumed in the form:

$$H_{tr} = N^{-1/2}\sum_{\alpha} V(z,r)\Delta(r - R_\alpha). \tag{2.3}$$

N denotes the number of unit cells, $\Delta(r - R_\alpha)$ the two-dimensional(2D) Kronecker delta function, and $V(z,r)$ is the scattering potential inducing the transitions across the intercalant layers. Equation(2.3) is a conduction path Hamiltonian introduced by Shimamura [3]. N_I in Eq.(2.2) is the number of the conduction path and V_0 is the interlayer matrix element of V(x.r). We take a notice that the conduction path Hamiltonian in Eq.(2.3) should not be taken in the literal sence of the word. It implies a transfer Hamiltonian across the intercalant layers. A detailed calculation yields the formula for σ_c [4].

$$\sigma_c = -\frac{8\pi e^2 d_I^2}{\hbar V}\left(\frac{N_I}{N}\right)\sum_c\sum_k V_0^2(i)\frac{\partial f(E_i(k))}{\partial E_i(k)}\frac{1}{\Gamma(E_i(k))}, \tag{2.4}$$

where d_I denotes the thickness of GIG-sandwich layer, i the band index and f is the Fermi function. Γ includes a contribution from the electron-in-plane phonon interaction and so ρ_c shows a metallic T-dependence, which explains the behavior in low stage GICs (see: Fig.1).

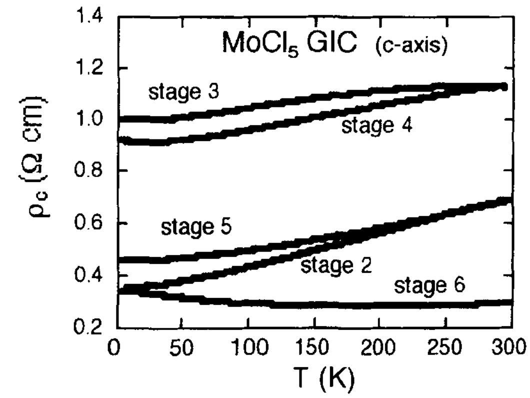

FIGURE 1 Temperature dependence of ρ_c for stage-2 to 6 $MoCl_5$ GICs.

3. c-Axis Conduction and Longitudinal Magnetoresistance for Intermediate GICs

The calculation is performed in terms of a series-resistance model:

$$\begin{cases} \rho_c \cong \{d_I \rho_c(GIG) + d_G(n-1)\rho_c(GG)\} \\ (n-1)\rho_c(GG) = \sum_{i=1}^{n-1} \rho_{GG}(i, i+1) \\ I_c = d_I + (n-1)d_G \qquad (n > 3) \end{cases} \tag{3.1}$$

where d_G represents the distance between nearest graphite layers without an intercalant layer. $\rho_c(GIG)$ is given by σ_c^{-1} calculated in Sec.2. $\rho_{GG}(i, i+1) = \sigma_{GG}(i, i+1)^{-1}$ denotes the resistivity corresponding to the transition $i \leftrightarrow i+1$ without across an intercalant layer.

The T-dependence of ρ_c changes from metallic-like to semiconductor-like with stage number n. To account for this feature, the T-dependent transfer Hamiltonian inducing $i \leftrightarrow i+1$ transition is introduced:

$$H_{GG} = H_{GG}^{(I)} + H_{GG}^{(e-p)} \tag{3.2}$$

where I denotes the elastic scattering term and e-p the electron-out-of phonon scattering. The expression for $\sigma_{GG}(i, i+1)$ becomes [4]

$$\sigma_{GG}(i, i+1) = (8e^2 d_G{}^2 / I_c \hbar^3)(m_i \Gamma_i^{-1} + m_{i+1}\Gamma_{i+1}^{-1}) A_{GG}(i, i+1) \tag{3.3}$$

$$A_{GG}(i, i+1) = |\langle i | H_{GG}^{(I)} | i+1 \rangle|^2 + |\langle i | H_{GG}^{(e-p)} | i+1 \rangle|^2 . \tag{3.4}$$

Here, we employed an intuitive model that the i-th layer is described by a band with effective mass m_i. From Eqs. (3.3) and (3.4) we have

$$\rho_c = \left(\hbar^4/16e^2\right) \times \left\{\left(d_I m_b \tau_b B_{GG}\right)^{-1} + \sum_i \left[\left(d_G/2\right)\left(m_i\tau_i + m_{i+1}\tau_{i+1}\right)A_{GG}(i, i+1)\right]^{-1}\right\}, \quad (3.5)$$

where $B_{GG} = N_I V_0^2/N$, m_b is the effective mass for the bounding layer, and τ_b is the corresponding basal plane relaxation time. $\left(m_i\tau_i + m_{i+1}\tau_{i+1}\right)$ decreases with T, while A_{GG} increases with T. $\rho_{GG}(i, i+1)$ term for the inter-interior transition plays a major role in ρ_c and it makes a bottleneck in the c-axis conduction because of the small charge density and small density of states in these layers. This explains the stage dependence in ρ_c vs T curve illustrated in Fig.1. This situation in ρ_c is essentially different from the one in ρ_a, since each layer contribution to ρ_a makes a parallel-resistance and the bounding layer with large carrier density plays a dominant role in the a-axis conduction.

Now we discuss the longitudinal magnetoresistance in terms of Eq. (3.1) which yields the equation:

$$\Delta\rho_{cL}/\rho_0 = -\left\{d_I r_b\left(\Delta\sigma_{GIG}/\sigma_{GIG}\right) + d_G\sum_i r(i, i+1)\left(\Delta\sigma_{GG}(i, i+1)/\sigma_{GG}(i, i+1)\right)\right\}I_c^{-1} \quad (3.6)$$

$$\rho_c(GIG) = \rho_{GIG} = \sigma_{GIG}^{-1}, \; r_b = \rho_{GIG}/\rho_0, \; r(i, i+1) = \rho_{GG}(i, i+1)/\rho_0, \quad (3.7)$$

σ_{GIG} and $\sigma_{GG}(i, i+1)$, which are given by Eqs.(2.4) and (3.3) respectively, include τ_b, τ_i and τ_{i+1}. Magnetic field along the c-axis affect the basal plane conductivity $\sigma_\alpha = e^2 n_\alpha \tau_\alpha / m_\alpha$ $(\alpha = b, i, i+1)$. This explains the reason for finite longitudinal magnetoresistance and we can apply the weak localization theory to $-\Delta\sigma_\alpha/\sigma_\alpha$ in Eq. (3.6). $\Delta\sigma_{GG}(i, i+1)/\sigma_{GG}(i, i+1)$ corresponding to the inter-interior transition plays an important role in $\Delta\rho_{cL}/\rho_0$ since $r(i, i+1) > r_b$. Consequently the negative magnetoresistance is expected from this term since the condition:

$$L_0 << L_H << L_{in} \quad , \quad L_H = \sqrt{\hbar c/4eH} \quad (3.8)$$

is satisfied at low temperature and in weak magnetic field, where L_0 and L_{in} represent the diffusion length for the elastic scattering and inelastic scattering, respectively[5]. Figure 2 illustrates $\Delta\rho_{cL}/\rho_0$ for stage-4 $MoCl_5$ GIC and the similar behavior was obtained in stage-5[6].

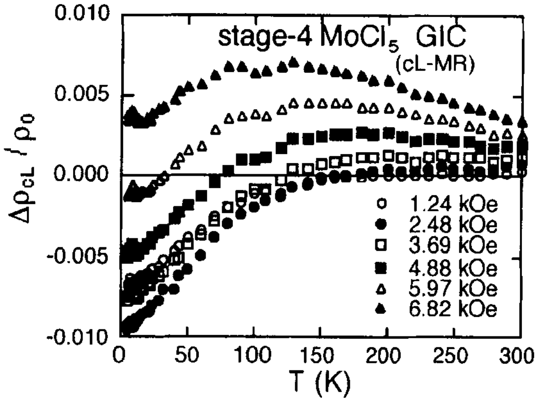

FIGURE 2 Temperature dependence of c-axis longitudinal magneto-resistance for stage-4 $MoCl_5$ GIC.

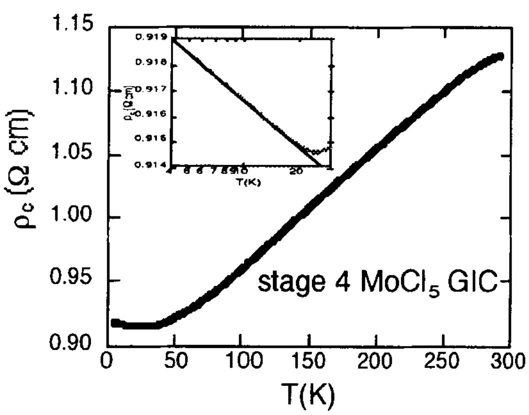

FIGURE 3 ρ_c vs T curve for stage-4 $MoCl_5$ GIC, where ρ_c vs $\log_{10} T$ is shown in the inset.

On the other hand stage-2 $MoCl_5$ GIC does not exhibit the negative magnetoresistance since the bounding layer with large density of states does not satisfy Eq.(3.8). logT-dependence of ρ_c vs T curve in the intermediate stage GICs provides an evidence for the weak localization effect [6] (see: Fig.3). In contrast with the c-axis conduction no negative transverse magnetoresistance $\Delta\rho_{aT}/\rho_0$ and no logT-dependence in ρ_a were observed irrespective of stage number. This is due to the situation that ρ_a is expressed by a parallel-resistance model and the bounding layer contribution is predominant in the a-axis conduction.

Finally we present the following comment. The present paper does not claim that the bounding layers do not give rise to a negative magnetoresistance and logT-dependence in GICs. Actually, these results were observed in the low-stage acceptor fiber GICs.[7] However, they are related to the basal-plane conduction process and not related to ρ_c mentioned in the present paper. Most important point in our article is that ρ_c and $\Delta\rho_{cL}/\rho_0$ are related to the basal plane conduction process.

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