

c-axis conductivity and thermoelectric power in graphite intercalation compounds

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The measurements of the *c*-axis conductivity σ_c at room temperature for acceptor graphite intercalation compounds typically show very small values of σ_c , ranging from $\sim 10^{-1}$ to ~ 10 ($\Omega \text{ cm}$) $^{-1}$. The formula $\sigma_c = Ne^2 l_c / m_c v_c$ leads to a mean-free path $l_c \ll 10^{-8}$ cm. This implies that the *c*-axis conduction is not a band conduction but a hopping process. The model for the phonon-assisted and impurity-assisted hopping conduction is presented and the diffusion thermoelectric power along the *c* direction is also calculated. Reasonable orders of magnitude are obtained for these transport coefficients. The phonon-assisted hopping conductivity increases with temperature, while the impurity-assisted hopping conduction is temperature independent at low temperatures and decreases with temperature in the region where the phonon scattering process is predominant. The diffusion thermoelectric power S_c is proportional to temperature and takes the form which is essentially equivalent to the one in metallic conductors.

I. INTRODUCTION

The electrical conductivity in graphite intercalation compounds (GIC's) is highly anisotropic. Typical values for the *a*- and *c*-axis conductivities for a number of donor and acceptor compounds are listed in Table I.^{1,2} As is shown in Table I, the behavior of the *c*-axis conductivity σ_c is very different in donor compounds and acceptor compounds. Namely, donor intercalation into graphite increases σ_c , while acceptor intercalation tends to decrease σ_c .

The typical value of σ_c in acceptor compounds at room-temperature ranges from $\sim 10^{-1}$ ($\Omega \text{ cm}$) $^{-1}$ to ~ 10 ($\Omega \text{ cm}$) $^{-1}$. If we apply the simple Drude formula $\sigma_c = Ne^2 l_c / m_c v_c$, it yields a mean free path $l_c \ll 10^{-8}$ cm. This implies that the *c*-axis conduction in acceptor compounds is not a band conduction but a hopping conduction. On the other hand, l_c of the low-stage donor compounds are much larger than those of the acceptor compounds and then the conduction is a band conduction.

Hopping conduction in GIC's is different from that in the usual small polaron,³ since the carriers in GIC's are completely delocalized in the basal plane. In this respect the situation is analogous to the transverse conduction in strong magnetic field.⁴ In this case carriers can freely move along the magnetic field direction, while in the plane perpendicular to the magnetic field, the carriers are confined in the Landau orbit.

We can formulate a theory of the *c*-axis conduction in a similar way to the Miller-Abrahams theory in impurity conduction.⁵ In this approach, the current density is given by a net flow of carriers per unit time across an intercalant layer, and this process can be calculated by introducing an interplay of the transfer Hamiltonian from one graphite layer to an adjacent layer and the electron-phonon interaction or the impurity potential. Diffusion thermoelectric power S_c is calculated in a similar way. In Sec. II the hopping conductivity due to the phonon-assisted process is obtained and S_c is also calculated. In

this case σ_c increases with temperature and S_c takes the form which is essentially equivalent to the one in degenerate conductors. In Sec. III a similar calculation is carried out in the case of impurity scattering. σ_c is temperature independent at low temperatures and decreases with increasing temperature in the region where the phonon scattering is important. Finally, in Sec. IV the conclusions are summarized.

II. PHONON-ASSISTED HOPPING CONDUCTION

The Hamiltonian employed in the following argument is

$$H = H_0 + H_{tr} + H_{e-ph}, \quad (2.1)$$

where

$$H_0 = \sum_n \sum_{s,k} E_s(k) a_{s,k}^\dagger(n) a_{s,k}(n), \quad (2.2)$$

TABLE I. Conductivity values of some typical donor and acceptor graphite intercalation compounds at room temperature (Refs. 1 and 2).

Intercalant	Stage	σ_a [$(\Omega \text{ cm})^{-1}$]	σ_c [$(\Omega \text{ cm})^{-1}$]
HOPG	∞	2.5×10^4	8.3
K	1	1.1×10^5	1.94×10^3
K	2	1.7×10^5	1.97×10^2
K	5		7.0
Li	1	2.5×10^5	1.8×10^4
HNO ₃	1	1.7×10^5	2.0
AsF ₅	1	5.0×10^5	0.23
AsF ₅	2	6.3×10^5	0.24
AsF ₅	3	5.8×10^5	0.26
FeCl ₃	1	1.1×10^5	10.0
FeCl ₃	2	2.5×10^5	1.6
SbCl ₅	6		1.2
SbCl ₅	10		3.0

$$H_{\text{tr}} = N^{-1/2} \sum_n \sum_s \sum_{k,k'} J_s(\vec{k} - \vec{k}') \times [a_{s,k}^\dagger(n+1)a_{s,k}(n) + a_{s,k}^\dagger(n)a_{s,k}(n+1)], \quad (2.3)$$

$$H_{e\text{-ph}} = \sum_n \sum_s \sum_{k,q} iD(\hbar/2d\Omega\omega_q)^{1/2} |q_z| \times [a_{s,k+q}^\dagger(n)a_{s,k}(n)b_q - a_{s,k-q}^\dagger(n)a_{s,k}(n)b_q^\dagger], \quad (2.4)$$

where H_0 is the two-dimensional unperturbed Hamiltonian, n is an index denoting a graphite layer, and s is an index specifying bands. H_{tr} is the transfer Hamiltonian across intercalated layers and $H_{e\text{-ph}}$ represents the electron-phonon interaction associated with the out-of-plane vibration.^{6,7} The interaction with the out-of-plane mode is important to explain the deviation from a T -linear dependence of the resistivity in graphite.^{6,7} For simplicity, interband transitions are neglected in H_{tr} and $H_{e\text{-ph}}$. N is the carrier density, D the electron-phonon coupling constant, and Ω represents the volume of the crystal. In the long-wavelength approximation, the dispersion relation of the out-of-plane vibration takes the form^{6,7}

$$\omega_q \cong v_s |q_z|, \quad z \parallel c \text{ axis}. \quad (2.5)$$

Equation (2.5) is expected to be a good approximation in GIC's, since the contributions from q_1^2 and q_2^2 terms are very small in graphite.⁷ In graphite we have $v_s \cong 3.96 \times 10^5$ cm/sec,^{6,7} while in GIC's it depends on both the intercalant species and stage numbers,⁸⁻¹⁰ and its magnitude ranges from $\sim 2 \times 10^5$ cm/sec to $\sim 4 \times 10^5$ cm/sec. Along the z direction, the current density and heat current density for $\nabla T = 0$ are given by

$$j_z = (e/\Omega) \sum_s \sum_k v_d(s,k), \quad (2.6)$$

$$w_z = (1/\Omega) \sum_s \sum_k [E_s(k) - E_F] v_d(s,k),$$

where the drift velocity $v_d(s,k)$ takes the form

$$v_d(s,k) = z_0 \sum_p [W_s(n,k \rightarrow n+1,p) - W_s(n+1,p \rightarrow n,k)]. \quad (2.7)$$

Here, W_s denotes a hopping rate across the intercalate layer and z_0 is an average hopping distance along the c axis. The diffusion thermoelectric power is given by

$$S_c = w_z / T j_z. \quad (2.8)$$

The transition rate $W_s(n,k \rightarrow n+1,p)$ is expressed by

$$W_s(n,k \rightarrow n+1,p) = \frac{2\pi}{\hbar} \sum_i \left| \frac{\langle n;s,\vec{k} | H' | \vec{i} \rangle \langle \vec{i} | H' | n+1;s,\vec{p} \rangle}{[E(\vec{k}) - E(\vec{i})]} \right|^2 \delta(E(\vec{k}) - E(\vec{p}) + eEz_0), \quad (2.9)$$

$$H' = H_{\text{tr}} + H'_{e\text{-ph}},$$

where \vec{k} , \vec{i} , and \vec{p} include electronic and phonon states, and E is an electric field applied parallel to the c axis. Without the electric field $W_s(n,k \rightarrow n+1,p)$, $W_s(n+1,p \rightarrow n,k)$ becomes zero. Using an approximation $J_s(k-k') = J = \text{const}$, we obtain the following expression for $W_s(n,k \rightarrow n+1,p) - W_s(n+1,p \rightarrow n,k)$:

$$W_s(n,k \rightarrow n+1,p) - W_s(n+1,p \rightarrow n,k)$$

$$= \frac{2\pi}{\hbar} \frac{\hbar(DJ)^2}{2d\Omega v_s} \sum_q |q_z| \left\{ \sum_{p'} [E_s(k) - E_s(p) \mp \hbar\omega_q]^{-2} [1 - f(p')] \times \left[f(k)[1 - f(p)] \begin{pmatrix} N_q + 1 \\ N_q \end{pmatrix} - f(p)[1 - f(k)] \begin{pmatrix} N_q \\ N_s + 1 \end{pmatrix} \right] \delta_{\vec{k}, \vec{p}' \pm \vec{q}} \times \delta(E_s(k) - E_s(p) \mp \hbar\omega_q + eEz_0) + \sum_{k'} [E_s(k) - E_s(k')]^{-2} [1 - f(k')] \times \left[f(k)[1 - f(p)] \begin{pmatrix} N_q + 1 \\ N_q \end{pmatrix} - f(p)[1 - f(k)] \begin{pmatrix} N_q \\ N_q + 1 \end{pmatrix} \right] \delta_{\vec{k}, \vec{p} + \vec{q}} \times \delta(E_s(k) - E_s(p) \mp \hbar\omega_q + eEz_0) \right\}, \quad (2.10)$$

where N_q denotes the phonon occupation number and f is the Fermi distribution function and $(N_q \mp q)$ indicates phonon-absorption and -emission processes.

Retaining the linear terms in electric field and assuming elastic scattering, we get the expression for σ_c from Eqs. (2.7) and (2.10)

$$\sigma_c \cong \frac{2\pi}{\hbar} \frac{e^2 z_0^2}{\Omega N k_0 T} \frac{\hbar(DJ)^2}{2d\Omega v_s} 4 \times \sum 4 \left[-k_0 T \frac{\partial f}{\partial E_s(k)} \right] \frac{[1-f(p')]\delta_{k,q+p'}}{|E_s(k)-E_s(p)|^2} \times \delta(E_s(k)-E_s(p)) N_q |q_z|, \quad (2.11)$$

where the factor of 4 in front of \sum comes from the spin degeneracy and from the two independent edges along HKH in the Brillouin zone and another factor of 4 stems from the summation over p' and k' and over phonon-absorption and -emission processes. \sum in Eq. (2.11) can be represented by

$$\sum \cdots = \sum_s \sum_{k_s, p_s, p'_s} \sum_{q_z} \cdots = \left[\frac{\Omega}{(2\pi)^3} \right]^3 \left[\frac{2\pi}{I_c} \right]^3 \frac{I_c}{2\pi} \times \sum_s \int d\vec{k}_s d\vec{p}'_s d\vec{p}_s d\vec{q}_z \cdots, \quad (2.12)$$

where \vec{k}_s , \vec{p}'_s , and \vec{p}_s are two-dimensional wave vectors perpendicular to the HKH axis, and I_c denotes the repeat distance of the crystal along the c axis. If the band extremum is far from the Fermi level, we can employ the approximation^{11,12}

$$E_s(k) \cong A k_s + \Delta_s, \quad A = \sqrt{3/2} \gamma_0 a, \quad (2.13)$$

and then Eq. (2.11) becomes

$$\sigma_c \cong \frac{2e^2}{\pi^3} \left[\frac{z_0}{I_c} \right]^2 \left[\frac{\Omega}{N} \right] \frac{(DJ)^2}{dv_s} \frac{q(T)^2 F(\Theta/T)}{A^4} \times \sum_s \int dE_s(k) k_s dE_s(p') dE_s(p) p \times \frac{E_s(p') - \Delta_s}{|E_s(k) - E_s(p')|^2} [1 - f_s(p')] \delta(E_s(k) - E_F) \times \delta(E_s(k) - E_s(p)), \quad (2.14)$$

where

$$q(T) = \frac{k_0 T}{\hbar v_s}, \quad F(\Theta/T) = \int_0^{\Theta/T} dx x (e^x - 1)^{-1}, \quad (2.15)$$

$$k_0 \Theta = \hbar v_s q_{\max}.$$

We note that Eq. (2.14) diverges at $E_s(k) = E_s(p')$. To remove this divergence, let us introduce a lifetime of the ini-

tial state \hbar/Γ and replace $|E_s(k) - E_s(p')|^{-2}$ by $\{[E_s(k) - E_s(p')]^2 + \Gamma^2\}^{-1}$. Moreover, we introduce an approximation

$$1 - f(x) \cong \begin{cases} 0, & x \leq -1 \\ \frac{1}{2}(x+1), & -1 \leq x \leq 1 \\ 1, & x \geq 1 \end{cases} \quad (2.16)$$

where $x = [E_s(p') - E_F]/k_0 T$. Finally, we obtain

$$\sigma_c \cong \frac{2e^2}{\pi^3} \left[\frac{z_0}{I_c} \right]^2 \left[\frac{\Omega}{N} \right] \frac{(DJ)^2}{dv_s} \frac{q(T)^2 F(\Theta/T)}{A^4} \sum_s k_{Fs}^2 G_s, \quad (2.17)$$

where k_{Fs} is the Fermi momentum of the s band and G_s takes the form

$$G_s = -1 + \frac{\Gamma}{\epsilon} \tan^{-1} \left[\frac{\epsilon}{\Gamma} \right] + \frac{1}{2} \ln \left[\frac{E_B^2}{\epsilon^2 + \Gamma^2} \right] + \frac{\pi(E_F - \Delta_s)}{2\Gamma}, \quad (2.18)$$

Here $\epsilon \cong k_0 T$, and E_B is a quantity with the order of the bandwidth. The most important term in G_s is the last term. At low temperatures this term is temperature independent, while at high temperatures it decreases as T^{-1} . Since $\Theta \cong 100$ K,^{6,7} $F(\Theta/T)$ is a temperature-dependent function except at low temperatures. $F(\Theta/T)$ is given by

$$F(\Theta/T) = \sum_{n=1}^{\infty} \frac{1}{n^2} \left[1 - e^{-n\Theta/T} \left(1 + \frac{n\Theta}{T} \right) \right], \quad (2.19)$$

and then

$$F(x) \cong \begin{cases} \sum \frac{1}{n^2} \cong \zeta(2) \cong 1.6, & x \gg 1 \\ x, & x \ll 1. \end{cases} \quad (2.20)$$

In consideration of the temperature dependence of $q(T)^2$, $F(\Theta/T)$, and G_s , the temperature dependence of the phonon-assisted c -axis conductivity is given by

$$\sigma_c \propto \begin{cases} T^2 & \text{at low temperatures} \\ T & \text{independent at high temperatures} \end{cases}, \quad (2.21)$$

since at low temperatures Γ and $F(\Theta/T)$ are temperature independent, while at high temperatures $F(\Theta/T) \cong \Theta/T$ and Γ^{-1} is also proportional to T^{-1} . The temperature dependences of the c -axis resistivity of higher stage SbCl_5 -GIC's (Ref. 13) and stage-5 K-GIC (Ref. 14) are shown in Figs. 1 and 2. The anomaly in certain SbCl_5 -GIC near 210 K shown in Fig. 1 has its origin in the commensurate-incommensurate transition.¹⁵

The observed behaviors of stages-4, -6, and -10 SbCl_5 -GIC's below 200 K are qualitatively explained by using Eq. (2.21), and the observed temperature dependence in Fig. 2 above ~ 30 K is not inconsistent with Eq. (2.21). At low temperatures, impurity scattering becomes

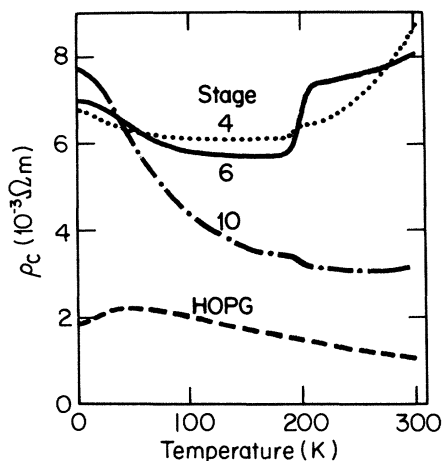


FIG. 1. Temperature dependence of the c -axis resistivity ρ_c of higher stage of SbCl_5 graphites (Ref. 13).

predominant and σ_c approaches a constant value. To evaluate the magnitude of σ_c , let us consider a stage-2 compound with the following set of parameters (see Refs. 7, 8–10, and 1 for D , v_s , and Δ_2 , respectively):

$$\begin{aligned} N/\Omega &= 10^{21} \text{ cm}^{-3}, \quad D = 4 \text{ eV}, \quad J = 0.005 \text{ eV}, \\ v_s &= 2.5 \times 10^5 \text{ cm/sec},^{8-10} \quad d = 2.5 \text{ g cm}^{-3}, \quad E_F = 0.8 \text{ eV}, \\ \Delta_1 &= 0, \quad \Delta_2 = \gamma_1 = 0.39 \text{ eV}, \quad \Theta = 100 \text{ K}, \\ \Gamma &= 2 \times 10^{-14} \text{ erg}, \quad A = 6.732 \times 10^{-8} \text{ eV cm}, \\ z_0 &= I_0, \quad T = 300 \text{ K}, \end{aligned} \quad (2.22)$$

and we obtain

$$\sigma_c \cong 0.1 (\Omega \text{ cm})^{-1}. \quad (2.23)$$

This is of the same magnitude as those of $A_s\text{F}_5$ compounds in Table I. In Eq. (2.22) the transfer integral J is an unknown parameter and is chosen so as to fit the conductivity data. Γ is estimated by using the relaxation time τ associated with the room-temperature resistivity values.¹ The magnitude of Eq. (2.23) is an order of 1

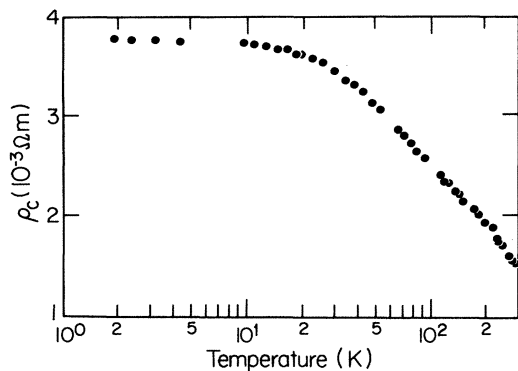


FIG. 2. Temperature dependence of the c -axis resistivity of stage-5 graphite potassium intercalation compounds (Ref. 14).

smaller than those of the observed values in Figs. 1 and 2. Although there is some ambiguity in the parameters of Eq. (2.22); such a discrepancy may not be serious.

From Eqs. (2.6), (2.8), and (2.14) the diffusion thermoelectric power S_c can be easily obtained as follows:

$$S_c = \frac{\pi^2 k_0^2 T}{3 E_F} \frac{2 \sum_s (E_F - \Delta_s)^2}{\sum_s (1 - \Delta_s/E_F)(E_F - \Delta_s)^2}. \quad (2.24)$$

Equation (2.24) is essentially equivalent to the expression in usual metallic conductors. By inserting the parameters employed in Eq. (2.22) into Eq. (2.24), we get

$$S_c = 18.4 \mu\text{V/K} \text{ at } 300 \text{ K}, \quad (2.25)$$

which is in qualitative agreement with the observed results shown in Fig. 3.¹⁴ Except for the low-temperature anomaly of S_c for stage-5 graphite potassium compounds, the linear dependence on temperature in Fig. 3 is consistent with the theoretical expectation given by Eq. (2.24). If we insert $D = 16 \text{ eV}$ and $v_s = 2.10 \times 10^6 \text{ cm/sec}$ for the in-plane vibration, which plays an important role in the basis plane conduction,^{1,6,7} into Eq. (2.17), we obtain a very small value for $\sigma_c < 0.01 (\Omega \text{ cm})^{-1}$. This indicates that out-of-plane vibration is much more important in c -axis conduction.

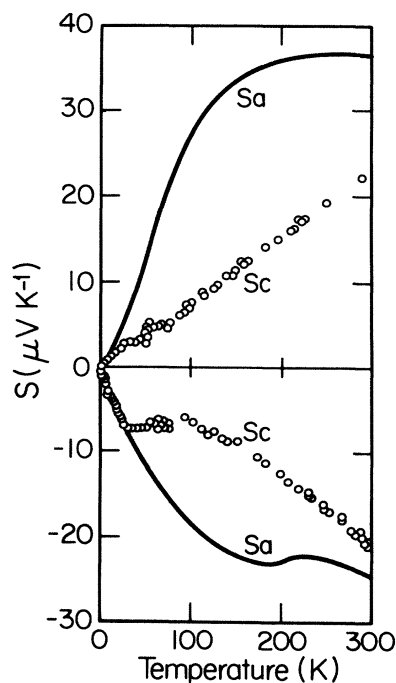


FIG. 3. Temperature variations of the c -axis thermoelectric power S_c of a stage-2 graphite- FeCl_3 compound (open circle, positive) and stage-5 graphite-potassium compound (open circle, negative). For comparison, results for the in-plane thermoelectric power S_a are shown (Ref. 14).

III. IMPURITY-ASSISTED HOPPING CONDUCTION

A similar calculation to the one in Sec. II can be carried out in the case of localized charged-center scattering. The presence of the charged centers is expected in the form of defects with localized charge (traps) in the intercalant layers. We assume that the scattering potential is given by

$$H_I = \frac{4\pi Z e^2}{\Omega \kappa} \sum_I \sum_q \frac{e^{i\vec{q}\cdot(\vec{r}-\vec{I})}}{q^2 + q_D^2}, \quad (3.1)$$

where κ denotes the dielectric constant, and q_D is the screening constant due to mobile carriers. σ_c takes the form

$$\sigma_c \cong \frac{32}{\hbar I_c} \frac{e^2}{\kappa^2 q_D^4} \left[\frac{z_0}{I_c} \right]^2 \frac{N_I}{N} \left[\frac{JZe^2}{A^2} \right]^2 \sum_s k_{Fs}^2 G_s, \quad (3.2)$$

where N_I denotes the scattering center concentration. In the Thomas-Fermi approximation, q_D is given by

$$\kappa q_D^2 = 4\pi e^2 N(E_F), \quad (3.3)$$

where $N(E_F)$ denotes the density of states at the Fermi level. $N(E_F)$ is obtained by

$$\begin{aligned} N(E_F) &= \frac{2 \times 2}{(2\pi)^3} \frac{(2\pi)^2}{I_c} \sum_s \int \frac{dk}{dE_s} dE_s k \delta(E_s - E_F) \\ &\cong \frac{2}{\pi} \frac{1}{AI_c} \sum_s k_{Fs}. \end{aligned} \quad (3.4)$$

At low temperatures, G_s given by Eq. (2.18) is temperature independent, since Γ is limited by point-center scattering. Assuming $N_I/N = 10^{-2}$, $Z=2$, and $\Gamma = 2 \times 10^{-15}$ erg which is an order of 1 smaller than the value in Eq. (2.22), we obtain

$$\sigma_c \cong 1.4 (\Omega \text{ cm})^{-1}, \quad (3.5)$$

which is in qualitative agreement with the observed values of the low-stage compounds in Figs. 4 and 5. σ_c given by

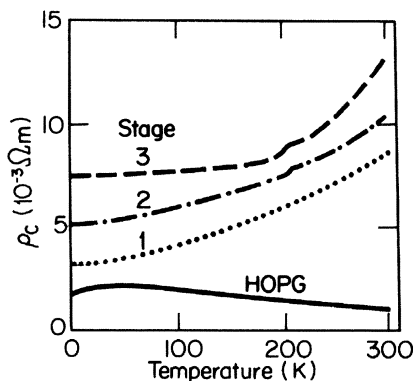


FIG. 4. Temperature dependence of the c -axis resistivity of lower stage SbCl_3 graphite (Ref. 13).

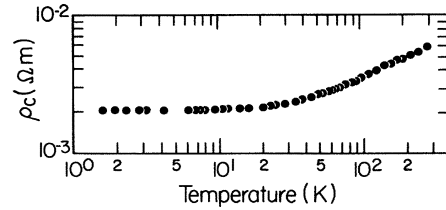


FIG. 5. Temperature variation of the c -axis resistivity of a stage-2 graphite- FeCl_3 intercalation compound (Ref. 14).

Eq. (3.2) decreases with T at high temperature, and this trend is consistent with Figs. 4 and 5. The thermoelectric power S_c is also given by Eq. (2.24).

The total conductivity is given by the sum of Eqs. (2.17) and (3.2) as the following:

$$\sigma_c = (\sigma_c)_{\text{phonon}} + (\sigma_c)_{\text{impurity}}, \quad (3.6)$$

where $(\sigma_c)_{\text{phonon}}$ and $(\sigma_c)_{\text{impurity}}$ have opposite temperature dependences. The different behaviors of the c -axis resistivity between low-stage compounds and high-stage compounds (see Figs. 2–5) can be explained as follows. In low-stage compounds many impurities or defects are introduced in the intercalation process. Then the localized charged-center scattering is more effective than the phonon scattering even at high temperatures. On the other hand, in high-stage compounds the phonon scattering becomes predominant at high temperatures.

IV. CONCLUSIONS

A model for the c -axis conduction and thermoelectric power in graphite intercalation compounds is presented. The c -axis conduction in GIC's is a hopping conduction without activation energy and is induced through the interplay between the transfer Hamiltonian H_{tr} and the electron-phonon interaction or the Coulomb potential due to the localized center. H_{tr} makes a carrier transfer from the n th layer to the $n+1$ or $n-1$ layers across the intercalant layers. The c -axis conductivity is given by the sum of the phonon-assisted hopping term and impurity-assisted contribution:

$$\sigma_c = (\sigma_c)_{\text{phonon}} + (\sigma_c)_{\text{impurity}}.$$

We conclude the following:

(1) $(\sigma_c)_{\text{phonon}}$ is proportional to T^2 at low temperatures and constant at high temperatures. This term is dominant for high-stage compounds, which exhibit a temperature dependence similar to that for $(\sigma_c)_{\text{phonon}}$.

(2) $(\sigma_c)_{\text{impurity}}$ is constant at low temperatures and decreases with increasing temperature at high temperatures. This behavior is in qualitative agreement with the observed results in low-stage compounds, where $(\sigma_c)_{\text{impurity}}$ is expected to be predominant. The intercalation process introduces many defects and/or impurities in the crystal, so that impurity-assisted hopping process is expected to be dominant even at high temperatures for the low-stage compounds.

(3) The temperature dependence of the thermoelectric power S_c is essentially equivalent to the one in usual metallic conductors, and the calculated value is in good

agreement with the observed ones.

(4) In $(\sigma_c)_{\text{phonon}}$ the interaction with the out-of-plane vibration plays an important role.

(5) Assuming the magnitude of the transfer integral $J=0.005$ eV, we can get a reasonable order of magnitude for $(\sigma_c)_{\text{phonon}}$ and $(\sigma_c)_{\text{impurity}}$. S_c does not include J .

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