Electrical Resistivity and Magnetoresistance of Graphite Fibers
II. Theoretical

Ko Sugihara, I. Rahim, M.Z. Tahar and M.S. Dresselhaus
Massachusetts Institute of Technology
Cambridge, MA 02139

Abstract. Theories of the electrical resistivity and magnetoresistance in benzene derived graphite fibers (BDF) are presented. An ellipsoidal band model is employed and three scattering processes due to crystallite boundaries, ionized impurity potentials and phonons, are taken into account. The observed linear magnetoresistance is ascribed to the microstructure of BDF.

Introduction

Electrical resistivity and magnetoresistance measurements on pregraphitic carbons have been theoretically interpreted by making use of a simple two-band (STB) model. However, in providing a more quantitative interpretation, a more detailed model is needed. In this article the electrical resistivity and magnetoresistance of benzene-derived graphite fibers (BDF) are treated by introducing an ellipsoidal model. The electrical resistivity is calculated by considering three different scattering processes -- phonon scattering, and scattering processes due to crystallite boundaries and ionized impurity potentials. The ionized impurity scattering provides a contribution of the same order to the relaxation rate as that due to boundary scattering, and is responsible for the oscillatory magnetoresistance in mesophase pitch derived fibers.[1] The Boltzmann equation is solved in the presence of a magnetic field and its solution is applied to BDF.

Electrical Resistivity of Graphite Fibers

The resistivity vs. temperature curve in single crystal graphite exhibits a sublinear temperature dependence.[2] The increase in the carrier concentration with temperature compensates the increasing scattering rate due to the electron-phonon interaction.[3] More pronounced effects are observed in BDF which are shown in Fig. 1.[4] In these samples, the three-dimensional structure is not so well established and the energy overlap and the Fermi energy are smaller than their corresponding values for single crystal graphite.

An ellipsoidal band model which is symmetric in electrons and holes is introduced:

$$E_i = \pm\Delta \pm \frac{\hbar^2}{2m_a} k_a^2 + \frac{\hbar^2}{2m_z} k_z^2 \quad , \; i = n \text{ or } p, \tag{2.1}$$

where 2Δ is the energy overlap. By introducing an anisotropic factor defined by

$$\S = \left(\frac{m_z}{m_a}\right)^{1/2}, \text{ and } K = (K_a, K_z) = (k_x, k_y, \S k_z), \tag{2.2}$$

Eq. (2.1) becomes

$$E_i = \pm\Delta \pm \frac{\hbar^2}{2m_a} K^2 \quad , \; (i = n \text{ or } p) \tag{2.3}$$

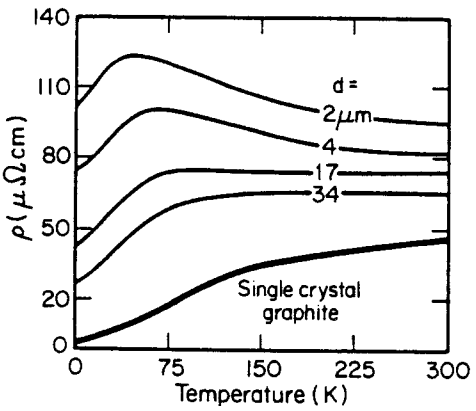

Figure 1. Resistivity vs. temperature four benzene-derived fibers ($T_{HT} \approx 3500°C$) with diameter 2, 4, 17 and 34 μm and for HOPG.[4]

If the electron and hole concentrations are equal, the Fermi energy E_F is located at $E_F = 0$. For simplicity, this assumption is employed. The relaxation rate is given by:

$$1/\tau = 1/\tau_b + 1/\tau_I + 1/\tau_{ph} = v/L + 1/\tau_I + CT^n, \tag{2.4}$$

where L denotes the average crystallite dimension, $1/\tau_I$ is the ionized impurity scattering rate and the last term represents the phonon scattering. The power n is ~ 1.2.[3] We employ the approximation:

$$\tau = \left[\frac{L}{v}\left(1 + \frac{\nu_I + CT^n}{\langle v\rangle/L}\right)\right]^{-1} , \qquad (2.5)$$

where $\nu_I = 1/\tau_I$ and $\langle v\rangle$ is an average velocity. The electrical resistivity is obtained as follows:

$$\rho(T) = \frac{3\pi^2\hbar^3}{2e^2L}\left(\frac{1}{m_a m_z}\right)^{\frac{1}{2}} \frac{1+\frac{\nu_I + CT^n}{\langle v\rangle/L}}{k_oT\,\ln(1+e^{\beta\Delta})} . \qquad (2.6)$$

At intermediate temperatures, the $\rho(T)$ vs. T curve has a maximum value, consistent with Fig. 1. $1/\tau_I$ take the form:

$$1/\tau_I = \frac{2\pi}{\hbar} N_I \overline{|v(q)|^2} \frac{D(E_F)}{4} , \qquad (2.7)$$

where N_I is the impurity concentration, $v(q)$ the Fourier component of the scattering potential and $D(E_F)$ denotes the total density of states. In the Thomas-Fermi scheme, $v(q)$ is

$$v(q) = \frac{4\pi Ze^2}{\varepsilon}\frac{1}{q^2+q_s^2} , \quad q_s^2 = \frac{4\pi e^2 D(E_F)}{\varepsilon} , \qquad (2.8)$$

where ε is the dielectric constant and Z is the effective valence of an ionized center. Inserting $m_z = 0.04\ m_o$, $\Delta = 0.01$ eV and $\varepsilon = 4$, we obtain

$$D(E_F) = 2.31 \times 10^{32}/\text{erg cm}^3, \quad q_s = 1.29 \times 10^7 \text{cm}^{-1}.$$

In the present model $q_{max} \approx 2(k_z)_{max} \approx 4 \times 10^7 \text{cm}^{-1}$, and Eq.(2.7) takes the value $\tau_I = 5 \times 10^{-12}$ sec, which is comparable to τ_b where $N_I = 3 \times 10^{18}/\text{cm}^3$ has been used.

Magnetoresistance of Graphite Fibers

As shown schematically in Fig. 2, each graphite layer in a BDF sees a different magnetic field and a different electric field. Solution of the Boltzmann Equation is assumed to be $f = f_o - (v\cdot\phi)\,\delta f_o/\delta E$. Inserting this into the Boltzmann Equation, we find

$$\phi = \left\{-e\tau E - e(e/c)^2\tau^3 \begin{bmatrix} 0 \\ H_y/m_a m_z \\ H_z/m_a \end{bmatrix} E\cdot H + (e^2/c)\tau^2 \right.$$

$$\left. \times \begin{bmatrix} E_y H_z/m_a - E_z H_y/m_z \\ -E_z H/m_a \\ E_x H_y/m_a \end{bmatrix} \right\} \times [1 + (\omega_c(\theta)\tau^2\}^{-1} , \qquad (3.1)$$

where

$$\omega_c(\theta) = \frac{eH}{m(\theta)c} , \quad \frac{1}{m(\theta)^2} = \frac{\sin^2\theta}{m_a m_z} + \frac{\cos^2\theta}{m_z^2} . \qquad (3.2)$$

The current density j_x is composed of two terms.

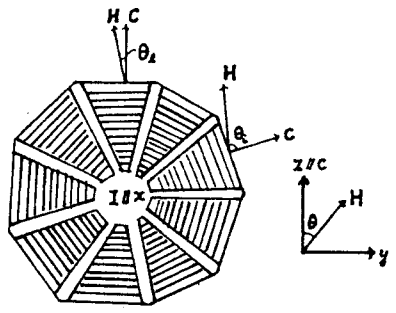

Figure 2. Cross section of a benzene-derived graphite fiber where H is perpendicular to I and I is along fiber axis.

$$j_x = \sum_{i=1}^{n} \sigma_{xx}(i) E_x + \sum_{i=1}^{n} \sigma_{xy}(i)\, E_y(i) \qquad (3.3)$$

By averaging over θ, σ_{xy} vanishes. Then, we have

$$j_x = \Delta\rho/\rho_o = \sigma_{xx}(0)/\sigma_{xx}(H) - 1 . \qquad (3.4)$$

After averaging over θ, σ_{xx} becomes

$$\sigma_{xx}(H) = \sum_{\lambda=n\cdot p} \sigma_{xx}^{(\lambda)}\{(1 + \omega_z^{(\lambda)}\omega_a^{(\lambda)}\tau_\lambda^2)$$

$$\times(1 + \omega_a^2\tau_\lambda^2)\}^{-\frac{1}{2}} \qquad (3.5)$$

Since $m_z \gg m_a$, $\omega_z\omega_a\tau^2$ is much smaller than unity if the field intensity is not so strong. Then, in the region of $(\omega_a\tau)^2 \gg 1$ we have $\Delta\rho/\rho_o \propto H^{1.7}$ which is consistent with our observations.

References

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