Negative Magnetoresistance of Graphite Fibers

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Abstract. A mechanism is proposed to explain the oscillatory negative magnetoresistance in mesophase pitch-derived graphite fibers. To explain the observed second minimum in $\Delta\rho/\rho_0$, a field-dependent scattering process due to ionized impurity potentials is taken into account.

Introduction

Some kind of disordered carbons with no long range three-dimensional ordering exhibit a negative magnetoresistance at low temperatures.[1] This problem has attracted a good deal of attention by many investigators, and several attempts have been made to explain this interesting phenomenon.[1-4] Among them, only the Bright theory[5] can account for the observed behavior of the negative magnetoresistance. In view of its importance, an outline of the Bright theory is summarized in the following.

1. A two-dimensional band model is employed as an appropriate starting point, since pregraphitic carbons lack three-dimensional layer correlations. The density of states associated with the two-dimensional Landau levels[6,7] is introduced. The effect of disorder on the density of states is considered by taking into account the Gaussian level broadening caused by the collision process and thermal broadening.

2. A small three-dimensional overlap effect is included by adding an excess density of states N_0 to the n = 0 Landau level.

3. An acceptor concentration of $N_a \sim 10^{18}/cm^3$ located at $E_a \sim 10^{-2}$eV is assumed. The carrier concentration n and p are functions of N_0, N_a, E_a, T and H. A simple two-carrier model is used to express the resistivity. Four adjustable parameters N_0, N_a, E_a and the mobility μ are chosen to fit the observed results.

The increase of the density of states and carrier concentration with magnetic field makes the magnetoresistance negative. With further increase in the magnetic field, the magnetoresistance becomes positive due to the presence of the factor $(\omega_c\tau)^2$, where ω_c denotes the cyclotron frequency.[5]

Recently, Woolf reported an unusual magnetoresistance behavior in mesophase pitch derived fiber (PDF)[8] which is shown in Fig. 1. The behavior in regions (I) and (II) can be explained by the Bright theory.[5] However, the Bright mechanism cannot provide any interpretation for the behavior in regions (III) and (IV) (see Fig. 1). It should be noted that the second minimum in the magnetoresistance is located at H ≈ 6.6 Tesla independent of temperature. Woolf[8] suggested that this second minimum might be related to the quantum limit in three-dimensional graphite which appears at H ≈ 7 Tesla. However, Woolf could not provide any mechanism for regions (III) and (IV).

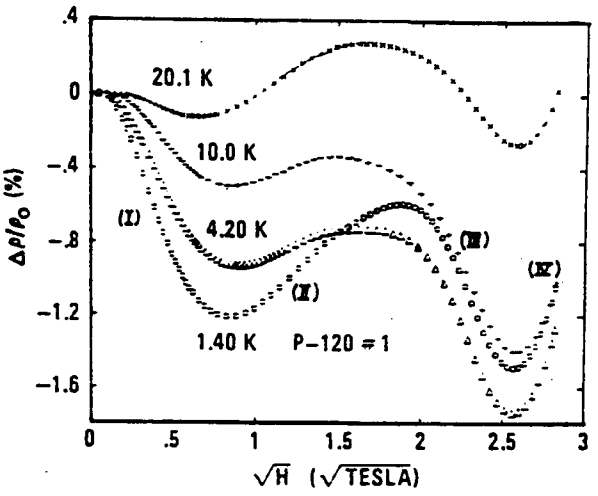

Figure 1. Transverse magnetoresistance $\Delta\rho/\rho_0$ vs. $\sqrt{H}$ for a p-120 fiber at various temperatures (Ref. 8).

In this article an alternate theory is proposed to explain the observed oscillatory magnetoresistance, with particular reference to the behavior (III) and (IV). In the Bright theory[5], the mobility $\mu = e\tau/m^*$ and the level broadening are assumed to be independent of the magnetic field. However, a field dependent relaxation process is considered and is necessary to explain the observed features in Fig. 1.

II. Oscillatory Magnetoresistance in Mesophase Pitch Derived Graphite Fibers

As is shown in Fig. 2, the mesophase PDF has a radial microstructure. Electric current flows along the fiber axis and the magnetic field is applied perpendicular to the fiber axis.[8] We

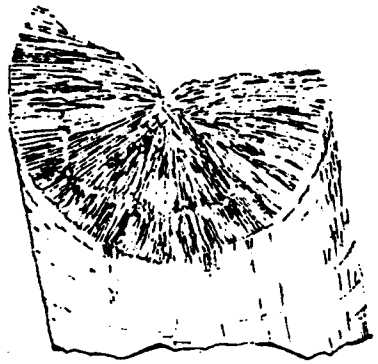

Figure 2. Microstructure of mesophase pitch derived graphite fibers.

assume that each graphitic layer contributes independently to the total conductivity σ, and σ is given by

$$\sigma(H) = \sum_i \sigma_i = \sum_i e\, n_i^+ \mu_i [1 + (n_i^- \beta_i / n_i^+)^2]/(1+\beta_i^2), \quad (2.1)$$

where $n_i^\pm = n_i \pm p_i$, $\beta_i = \omega_i \tau_i$, $\omega_i = eH_i/m^*c$, $H_i = H\cos\theta_i$ and θ_i denotes the angle between H and the c-axis of the i-th layer. The dominant relaxation processes at low temperatures consist of two contributions:

$$1/\tau = 1/\tau_b + 1/\tau_I, \quad (2.2)$$

where $1/\tau_b$ denotes the crystallite boundary scattering and $1/\tau_I$ is the ionized impurity scattering. $1/\tau_b$ is field-independent, while $1/\tau_I$ depends on the field intensity. The ionized impurity potential is given by

$$V(r) = 1/\Omega \sum_n \sum_q v(q) e^{iq\cdot(r-R_n)},$$

$$v(q) = [(4\pi Z e^2)/\varepsilon]\, [1/(q^2+q_s^2)], \quad (2.3)$$

where Ω is the crystal volume, ε the dielectric constant, Z the effective valence of an ionized scattering center and q_s the screening constant which in the Thomas-Fermi scheme becomes:

$$q_s^2 = 4\pi e^2\, D(E_F)/\varepsilon. \quad (2.4)$$

$D(E_F)$ denotes the density of states at the Fermi level and is given by[5]

$$D(E_F) = aH + \text{constant}. \quad (2.5)$$

The constant term comes from the additional density of states N_0. For H ≈ 5 Tesla, q_s is on the order of $\sim 10^7 cm^{-1}$, and we have

$$v(q) \approx v(0) \propto D(E_F) \quad (2.6)$$

The relaxation time τ_I is easily obtained as follows:

$$\frac{1}{\tau_I} = \frac{2\pi}{h} N_I\, |v(0)|^2\, \frac{D(E_F)}{4} \propto D(E_F)^{-1}, \quad (2.7)$$

where N_I indicates the ionized impurity concentration. Because of Eq.(2.5), $1/\tau_I$ decreases with increasing H. This result was already obtained by McClure and Spry in their solution of the quantum transport problem in graphite.[9] In the weak field region, the effect of the magnetic field on the mobility μ_i is small since $1/\tau_b > 1/\tau_I$ and in regions (I) and (II) in Fig. 1, the Bright theory[5] is applicable. With further increase of the field, the decrease of $1/\tau_I$ becomes appreciable in the layers with large crystallites where $1/\tau_I \lesssim 1/\tau_b$. These layers make a large contribution to the total conductivity σ, especially at low temperature. Accordingly, $\Delta\rho/\rho_o$ decreases again in region (III) and finally it increases with H due to the factor $(\omega_i\tau_i)^2$ and the ineffectiveness of $1/\tau_I$ relative to $1/\tau_b$ at high fields. The effect of temperature on the present theory is negligible at low temperatures where $E_F \gg k_oT$ is realized. Therefore, the constant location of the second minimum can be explained. In the quantum limit region in graphite, the observed magnetoresistance does not exhibit any negative magnetoresistance nor a minimum.[10] Therefore, Woolf's suggestion does not provide a mechanism for explaining the second minimum.

To check the present theory, further detailed experimental studies are desirable. The Bright mechanism and the present model cannot be applicable to the negative magnetoresistance problem in intercalated graphite fibers[11] because of their large Fermi energy. The effect of spin disordering scattering is too small to account for the observed magnitude of the negative magnetoresistance in intercalated graphite fibers.

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