Electrical Properties of Graphitic Pitch-Based Fibers at High Pressure

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Introduction

The unusual electronic properties of carbon fibers are currently of interest from several points of view. For example, adjustment of fiber formation conditions and heat-treatment temperature allows one to vary the degree of three-dimensional ordering along the c axis and study its effect on the electronic structure. In addition, the properties of well-graphitized production-type fibers are of practical importance since these fibers can be intercalated to yield high-strength, high-conductivity materials for aerospace applications.

One unusual feature of PAN- and pitch-based fibers is their pronounced negative magnetoresistance at low temperatures[1] in contrast to the positive magnetoresistance typically observed in single-crystal graphite and in most metals. Bright[1] has developed a theoretical model for the negative magnetoresistance which is in reasonable agreement with data on a wide variety of partially-ordered carbon fibers and glassy carbons. In this model the negative magnetoresistance is attributed to a magnetic-field-dependent variation in carrier concentration arising from field-induced changes in the Fermi energy and the energies of the (collision-broadened) Landau levels. Bright's model is important because it suggests that the conduction process, even in rather disordered samples, involves extended (Bloch) states rather than localized states and hopping processes.

Since Bright's model involves carriers (of density N_0) arising from band overlap produced by partial c-axis ordering, it can be tested by magnetoresistance measurements under hydrostatic pressure, which in single-crystal graphite produces large changes in N_0.[2] In this report we present data on a well-graphitized commercially-available pitch-based fiber (Union Carbide P-100) taken at liquid-helium temperatures in fields up to 1.2 T at hydrostatic pressures up to ∿16 kbar.

Experimental Methods

Samples (5-7 mm in length) were mounted perpendicular to the magnetic field on copper wires with conductive epoxy. Resistivity and magnetoresistance were measured by four-terminal methods using alternating current of 5 μA rms at 19 Hz. Hydrostatic pressures were generated by the clamp method using a 1:1 mixture of isoamyl alcohol and n-pentane as a pressure-transmitting medium[3] and were measured by a superconducting tin manometer.

Results

Typical curves of magnetoresistance vs magnetic field strength (B) at 4.2 K and 1.5 K are shown in Fig. 1, where the magnetoresistance is defined in the usual way, i.e., $\Delta\rho/\rho_0=[\rho(B)-\rho(0)]/\rho(0)$. A surprising result is the substantial temperature dependence of $\Delta\rho/\rho_0$ between 1.5 K and 4.2 K despite the fact that ρ_0 itself varied by <0.5% over this interval.

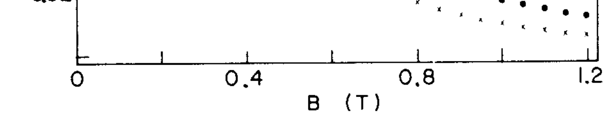

Figure 1. Magnetoresistance vs magnetic field strength (B).

The pressure dependence of $\Delta\rho/\rho_0$ for fields of 0.1, 0.5, and 1.2 T at 4.2 K is shown in Fig. 2. Numbers above the data points indicate the order in which the data were taken - no irreversible sample damage is evident. At the lower field strengths, $|\Delta\rho/\rho_0|$ decreases approximately linearly at the rate $d\ln(|\Delta\rho/\rho_0|)/dP=-0.006\ \mathrm{kbar}^{-1}$.

Discussion

Bright[1] has shown that for sufficiently low magnetic field and temperature $\Delta\rho/\rho_0$ varies quadratically with B. In Fig. 3 we plot $\Delta\rho/\rho_0$ vs B^2 for low fields and in fact find approximately quadratic behavior at fields below ∿0.1 T. The data over our entire field range (Fig. 1) are quantitatively very similar to Bright's results[1]

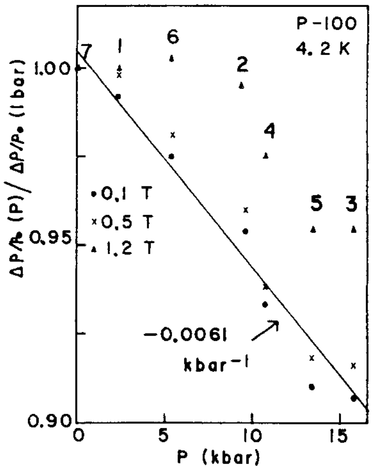

Figure 2. Magnetoresistance vs pressure.

for a fiber for which his model showed $N_o=5.8\times10^{24}$ m^{-3}, $\mu=0.37$ $m^2(V\ s)^{-1}$, where μ is the carrier mobility. Inserting these numbers into Bright's approximate expression for the low-field magneto-resistance, we find that the approximation can be further simplified to yield

$$\Delta\rho/\rho_o \sim -(\ell n2)\,\frac{e^2\gamma_o{}^2 a_o{}^2\tau^2}{\pi c_o \hbar^4 N_o}\,B^2, \qquad (1)$$

where γ_o is the in-plane interaction energy, a_o and c_o the a-axis and c-axis lattice parameters respectively, and τ the relaxation time. Deriving τ from Bright's value for μ and his estimate of the carrier effective mass (0.025 m_o) and using published values of γ_o, a_o, and c_o,[2] we find that equation (1) is in reasonable quantitative agreement with the data of Fig. 3.

Since equation (1) appears to be valid at low fields, we may use it to estimate the pressure dependence of N_o. Combining our pressure data at low fields (Fig. 2) with reported data on the pressure dependence of c_o (-0.0028 $kbar^{-1}$)[2] and τ (0.003 $kbar^{-1}$)[4] in single crystals and assuming γ_o and a_o vary only weakly with pressure,[2] we find $d\ell nN_o/dP \sim 0.015$ $kbar^{-1}$. This value is smaller than that observed in single crystals (0.038 $kbar^{-1}$)[2] and in very-highly-ordered gas-derived fibers (0.019 $kbar^{-1}$).[5] This trend may arise from the diminished c-axis order in fibers. However, the agreement with Bright's model is quite reasonable.

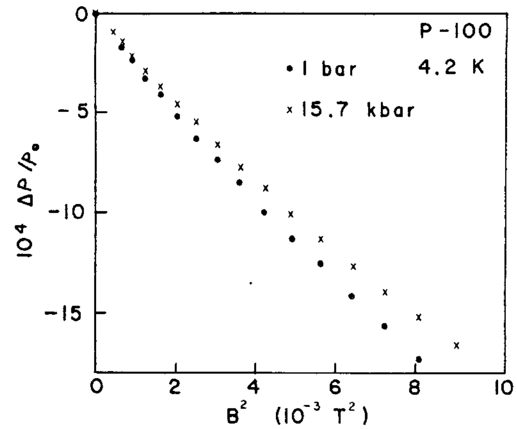

Figure 3. Magnetoresistance vs B^2 at low fields.

Conclusions

Interpretation of our data in terms of Bright's model yields a value for the pressure dependence of the carrier concentration which is in fair agreement with observations on single-crystal graphite and very-highly-ordered fibers. This lends support to the model and its assumption that conduction in fibers is by extended states rather than by hopping. However, we are unable at this time to understand the marked temperature dependence of the magnetoresistance at temperatures below 4.2 K. Further studies on less-well-graphitized samples are in progress.

Acknowledgments

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