Non-Ohmic Transport in the Magnetic-Field-Induced Charge-Density-Wave Phase of Graphite

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A striking nonlinear transport behavior has been observed in the recently discovered high-magnetic-field phase of graphite. The phenomenon is reminiscent of the collective charge-density-wave transport observed in some classes of linear-chain compounds such as $NbSe_3$. This supports our identification of the new phase with a charge-density-wave state. The principal direction of the charged-density wave vector is perpendicular to the magnetic field in contrast to the prediction of the Yoshioka-Fukuyama theory.

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The electron-hole gas in graphite undergoes a phase transition under the conditions of high magnetic field and low temperature.[1-3] The transition temperature T_c as a function of magnetic field B is empirically given by[2,3]

$$T_c = T^* \exp(-B^*/B), \quad (1)$$

where T^* and B^* are fitting parameters whose values have been determined as 69 K and 1047 kG, respectively. This functional form is basically understood as a BCS formula $T_c \sim 1.14\epsilon_F \exp[-1/N(\epsilon_F)V]$ for a mean-field–type phase transition. Here, ϵ_F is the Fermi energy, $N(\epsilon_F)$ is the density of states at ϵ_F, and V is the "pairing" potential. The B dependence of the exponent in Eq. (1) arises from the linear dependence of $N(\epsilon_F)$ on B because of the Landau degeneracy factor.

Yoshioka and Fukuyama (YF)[4] have proposed a model which is based on the general theory by Fukuyama[5] of the charge-density-wave (CDW) instability of a magnetic-field-induced one-dimensional electron gas. According to the YF model, the CDW wave vector is $\mathbf{Q} = (0,0,2k_F)$, where k_F is the Fermi wave number, so that the charge-density modulation is along the magnetic field direction.

In this Letter, we report a peculiar non-Ohmic transport behavior in this new phase of graphite, which bears some resemblance to those observed in the CDW phases of linear-chain compounds including $NbSe_3$, TaS_3, $(TaSe_4)_2I$, and $K_{0.3}MoO_3$.[6-9] Single-crystal flakes of Kish graphite[10] were used for the transport measurements. Typical dimensions were $3\times 1\times \text{mm}^3$; the smallest dimension corresponds to the c axis or the direction perpendicular to the layer plane. The resistivity measurements were done by the four-probe method. The electrical contacts were made by silver paint. A pulse technique was used for the high-current measurements to avoid Joule heating. Typical pulses width and repetition rate were 10 μsec and 100 Hz, respectively. Magnetic fields up to 230 kG supplied by a Bitter magnet were applied parallel to the c axis of the samples, and the transverse magnetoresistivity ρ_{xx} was measured. The samples were cooled down to 450 mK in a ^{3}He evaporation cryostat.

The inset of Fig. 1 shows a trace of ρ_{xx} for the whole magnetic field range. The sharp increase of ρ_{xx} was seen at $B = 210$ kG for this temperature is the manifestation of the phase transition. The main frame of Fig. 1 shows traces of ρ_{xx} in the high-magnetic-field range for different current values. It is seen that as the current is increased, the resistivity behavior above the transition point is drastically changed. A self-heating effect as a possible cause of the effect has been ruled out by varying the width and repetition rate of the current pulses. A most convincing evidence against the self-heating effect is provided by the current independence (despite its temperature sensitivity) of the transition point as seen in Fig. 1.

Figure 2 shows the conductivity σ_{xx} as a function of electric field E for different magnetic fields ranging from 210 to 230.9 kG. At $B = 210$ kG, where the system is in the normal state at this temperature $T = 496$ mK, the conductivity is independent of E (i.e., Ohmic) up to the highest E value. For higher B, the system is in the ordered state. There, a peculiar non-Ohmic behavior is observed. The non-Ohmic behavior is characterized as a switching of the conductivity from a low-E-limit value σ_0 to a high-E-limit value $\sigma_0 + \sigma_1$, in a finite E range centered around E_0 (~ 100 mV/cm typically). The fact that the characteristic field E_0 is as low as ~ 100 mV/cm rules out the single-particle Zener tunneling as the possible mechanism for the non-Ohmic behavior. The condition for the Zener

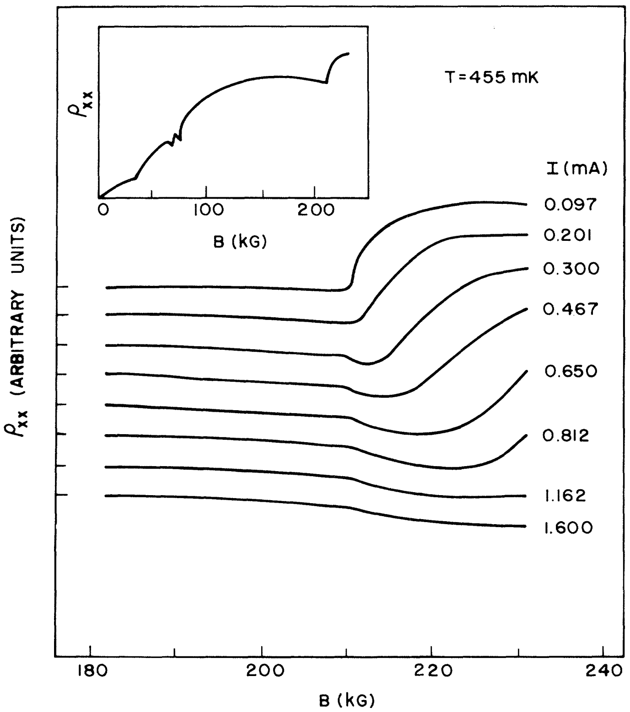

FIG. 1. The traces of ρ_{xx} in the high-magnetic-field region for different current values. Each trace is shifted as indicated in the figure for clarity. The inset shows the trace for the whole magnetic field range in the low current limit.

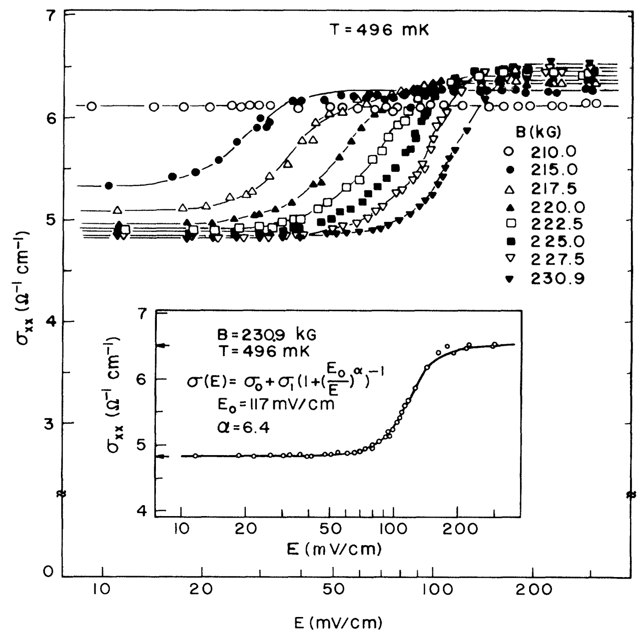

FIG. 2. The magnetoconductivity as a function of the electric field at different magnetic fields. The inset shows an example of the best fit with Eq. (2).

tunneling to occur is $eE\langle r\rangle \sim \Delta$, where Δ is the gap and $\langle r\rangle$ denotes the spatial extent of the electronic wave function. Since $\langle r\rangle$ should be of the order of the magnetic length l (~ 60 Å at $B=200$ kG), $E_0 \sim 100$ mV/cm gives $eE_0\langle r\rangle \sim 10^{-7}$ eV. Such a value is orders of magnitude smaller than the gap $\Delta \sim k_B T_c \sim 10^{-4}$ eV. Another possibility worth considering is the drift motion of carriers in the crossed electric and magnetic fields. However, the drift velocity is estimated as $c(E/B) \sim 10^2$ cm/sec (where c is the light velocity), and is far too small to cause any effect.

This non-Ohmic transport phenomenon is reminiscent of those observed in linear-chain compounds such as $NbSe_3$,[6-9] which are ascribed to the electric field depinning of CDW condensates. It is reported by Nakamura *et al.*[11] that ρ_{xx} measured at radio frequencies behaves differently from the dc counterpart with respect to the anomaly associated with the phase transition. This may be related with the present observation, and may be explained in terms of the collective response of a CDW condensate. It would be interesting to study the frequency dependence of the conductivity in more detail.

The present observation supports our identification of the new phase with a magnetic-field-induced CDW state.[12] The alternative possibility of an excitonic-insulator state is inconsistent with the present result, since the electrically neutral excitonic condensate cannot contribute to the transport. Secondly, the CDW wave vector **Q** is not in the direction of magnetic field. A CDW along the magnetic field direction, as modeled by YF,[4] cannot explain the observed motion of the CDW condensate in the direction perpendicular to the magnetic field. Preliminary data on the longitudinal magnetoresistivity indicate that ρ_{zz} does not exhibit such a large change at the transition as ρ_{xx} does. Therefore, the principal charge-density modulation appears to be in the plane perpendicular to the magnetic field direction. Theoretical models such as one developed by Fukuyama, Platzman, and Anderson[13] for two-dimensional systems may be more relevant to the present problem of the magnetic-field-induced CDW state of graphite than the one-dimensional model of the YF theory.[4] The preferred direction of the CDW wave vector with respect to the magnetic field direction is discussed by Fukuyama[5] for the case of isotropic electron mass.

Although the non-Ohmic behavior is similar to that observed in $NbSe_3$, there are differences. The onset of the non-Ohmic behavior in the present case is rather gradual, in contrast with a sharp threshold behavior in $NbSe_3$. The electric field dependence of the conductivity can be fitted with an empirical formula,

$$\sigma_{xx}(E) = \sigma_0 + \sigma_1[1 + (E_0/E)^{\alpha}]^{-1}, \qquad (2)$$

rather than an exponential form found successful in

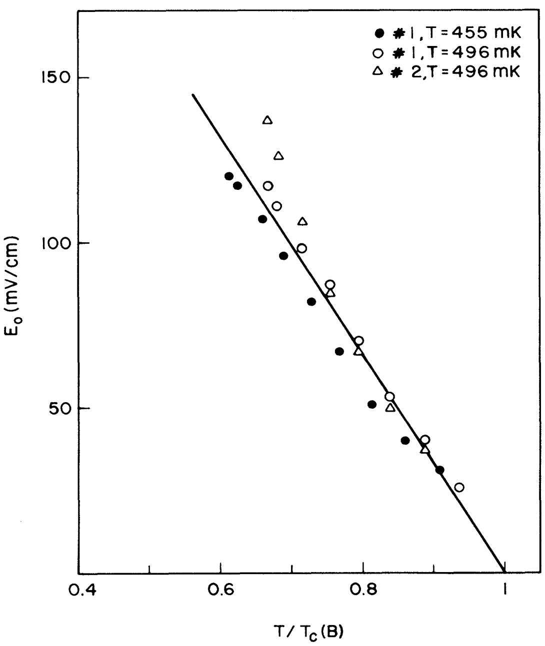

FIG. 3. The dependence of the characteristic electric field E_0 on $T/T_c(B)$. The data were taken by varying B [and thereby changing $T_c(B)$ according to Eq. (1)] while keeping T constant.

the case of $NbSe_3$.[6,7] Here, E_0 gives the characteristic field for the switching from the low-E-limit conductivity σ_0 to the high-E-limit conductivity $\sigma_0+\sigma_1$. The parameter α scales the sharpness of the switching. An example of the best fit of Eq. (2) is shown in the inset of Fig. 2, where $E_0=117$ mV/cm and $\alpha=6.4$, when $B=230.9$ kG ($T_c=741$ mK) and $T=496$ mK. The dependence of E_0 on $T/T_c(B)$ is shown in Fig. 3. Three sets of data are shown in this figure, two sets for one sample at two different temperatures, and the third set for another sample. The characteristic field E_0 decreases approximately linearly with $T/T_c(B)$ as $T/T_c(B)$ approaches unity from below. Though the same functional form [Eq. (2)] was valid for both samples and the values of E_0 were roughly in agreement between them, the value of α was found to be about 6 for one sample and about 3 for the other. One of the possible interpretations for the sample dependence of α is that the functional form of Eq. (2) reflects the distribution of the pinning strength at different pinning sites, while the individual depinning transitions are sharp.

In conclusion, the observation of non-Ohmic transport supports our identification of the high-magnetic-field phase of graphite with a CDW state. However, the present results reopen the question of the mechanism for the phase transition, because the CDW state realized here is clearly more complicated than the one modeled in the YF theory.

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