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Anomalous electrical conduction in graphite-vaporized films

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ABSTRACT

A transition to a higher electrical-conduction state is observed at temperatures between 30 K and 115 K in graphite-vaporized films, depending on preparation conditions. This transition may be related to carbon clusters [C_n with n = 30–100] being contained in graphitic amorphous carbon.

Recent reports on soccer-ball-shaped C_{60} molecules, one of the 'fullerenes', have attracted great interest (Kroto, Heath, O'Brien, Curl and Smalley 1985, Kratschmer, Lamb, Fostiropoulos and Huffman 1990), since they are a new type of superconducting material. Superconductivity has been reported in alkali-metal-doped compounds of C_{60}: K_3C_{60} with a critical temperature T_c = 18 K (Hebard, Rosseinsky, Haddon, Murphy, Glarum, Palstra, Ramirez and Kortan 1991) and Rb_2C_{60} with T_c = 30 K (Holczer, Klein, Gruner, Huang, Kaner, Fu, Whetten and Diederich 1991).

Solid carbon containing C_{60} molecules is synthesized from soot produced by arc vaporization of graphite in a 100-Torr atmosphere of helium (Kratschmer et al. 1990). Graphitic amorphous carbon films are known to be prepared by arc vaporization of graphite in a vacuum of 10^{-6} Torr. An unconventional temperature dependence of the d.c. conductivity, \sigma_d \propto T^p with p = 2–15, has been reported in such films (Shimakawa and Miyake 1988, 1989, Shimakawa and Kameyama 1989). A certain amount of C_n (n = 30–100) should also be present in arc vaporized films prepared at a relatively high atmospheric pressure. In this Letter, we report the transition from low to high d.c. conductivity states at temperatures between 30 and 115 K in these films, which has never been observed in normal films prepared at 10^{-6} Torr.

Thin films (0.2–1 \mu m) of carbon were prepared on Corning 7059 glass substrate by arc vaporization of graphite under pressures of between 1 \times 10^{-5} and 3 \times 10^{-4} Torr atmosphere of air (not in He or Ar). The deposition rate was approximately 100 Å s^{-1}. Planar gap-cell electrodes using Al or Au contacts were fabricated (gap spacing 0.1 mm, gap width 5 mm) to measure the d.c. conductivity. Films were peeled off from substrates to measure the X-ray diffraction.

Figure 1 shows the X-ray diffraction pattern of powder in the arc-vaporized graphite prepared at 3 \times 10^{-4} Torr. In this figure, a small but sharp peak around 2\theta = 26^\circ corresponding to the main peak of crystalline graphite is observed, indicating the presence of microcrystallites of graphite. A broad peak at 2\theta = 24^\circ and a small and broad peak around 2\theta = 44^\circ are attributed to amorphous graphite. The sharp peak
Fig. 1

X-ray diffraction pattern for arc-vaporized graphite prepared at $3 \times 10^{-4}$ Torr (sample no. 3).

$(26^\circ)$ disappears in the samples evaporated in a higher vacuum ($10^{-6}$ Torr). This indicates that the amorphous structure accounts for most of the sample.

Figures 2 and 3 show examples of the temperature-dependent resistance $R_{dc}$ for three samples nos. 1, 2, and 3, respectively which were all arc-evaporated in a relatively poor vacuum (in the range $10^{-4}-10^{-5}$ Torr during evaporation), of which sample 3 was deposited under the lowest vacuum level ($3 \times 10^{-4}$ Torr). We use the resistance $R_{dc}$ here because the exact film thickness has not been measured. Roughly estimated resistivities at room temperature are 1000, 2 and 1 $\Omega$ cm for sample nos. 1, 2, and 3, respectively. Transitions from low to high conducting states are evident (step-like transition) at 47 K for no. 1, 30 K for no. 2, and 115 K for no. 3. Although the magnitude of the change of $R_{dc}$ and the transition temperature depend on preparation conditions (pressure during evaporation) which are not controllable at present, nevertheless for a given sample, the discontinuities were observed on both heating and cooling in a temperature-cycling experiment. It should be noted, however, that only low-vacuum-deposition carbons exhibit these transitions.

It is considered that semiconducting behaviour may continue to the lowest measured temperature in the host graphitic amorphous carbon and that the transition to a very high conducting state occurs within small volume fractions (a binary distribution of high resistivity and low resistivity after the transition). This is just the percolation problem (Kirkpatrick 1973). The effective resistivity in the system is given by two different resistivities for a binary distribution of resistivity (effective-medium percolation theory; EMPT) (Kirkpatrick 1973). Using EMPT, a decrease of about 20% in the resistance at 115 K, for example in no. 3 as shown in fig. 3, can be explained by the presence of 5% volume-fraction of the high-conducting state ($10^{-6}$ $\Omega$ cm for the high conducting state is used for calculation). The large change of $R_{dc}$ found in no. 1 (fig. 2) can be attributed to large volume fraction (around 30%) of the high-conducting state which is very close to the percolation threshold (33 vol.% (Kirkpatrick 1973). It is suggested that the high-conducting state may be related to $C_6$ molecules.

The resistance $R_{dc}$ before the transition (higher temperature) is proportional to $T^{-\rho}$ with $\rho = 3.5$, 2.2, and 3.0 for nos. 1, 2 and 3, respectively, which may be interpreted in
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Fig. 2

Temperature dependence of resistance of sample no. 1.

Fig. 3

Temperature dependence of resistance in samples nos. 2 and 3.
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terms of the weak-coupling multiphonon hopping of carriers (Shimakawa and Miyake 1988, 1989, Shimakawa and Kameyama 1989). This unfamiliar property of $R_{de} (T \approx 2)$ has also been reported in the c-axis conductivity in crystalline copper-oxide superconductors (Ito, Takagi, Ishibashi, Ido and Uchida 1991).

In summary, we have found transitions from low to high electrical conducting states at temperatures in the range 30–115 K in graphite vaporized films which may contain C$_n$ ($n = 30–100$) molecules. A mass spectroscopy study is now being undertaken and a detailed argument will be elaborated in a future publication.

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REFERENCES


