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Revealing properties of single-walled carbon nanotubes under high pressure

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Abstract

It was found by the x-ray diffraction experiment under hydrostatic pressure that the carbon nanotubes are compressed easily with a high volume compressibility of 0.024 GPa^{-1} . The single-walled carbon nanotubes are polygonized when they form bundles of hexagonal close-packed structure and the inter-tubular gap is smaller than the equilibrium spacing of graphite. Under high pressure, further polygonization occurs to accommodate the extra amount of volume reduction. The ratio of the short and the long diagonals in the hexagonalized cross section is found to have changed from 0.991 at zero pressure to 0.982 at 1.5 GPa pressure, when the Bragg reflection from the nanotube lattice diminished. Accompanying polygonization, a discontinuous change in electrical resistivity was observed at 1.5 GPa pressure, suggesting a phase transition had occurred.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Carbon nanotubes have been extensively studied after they were first discovered about ten years ago [1], due to their extraordinary properties and their promising potential for nanotechnology applications [2]. Because of the finite lateral dimension and perfection in atomic structure, carbon nanotubes are expected to exhibit physical behaviours fundamentally different from those of graphite [3–6]. Electronically, a carbon nanotube can be either a semiconductor or a metal, depending on its diameter and helicity, and the electronic band structure is sensitive to the cross-sectional geometry of the concerned nanotube.

We have applied hydrostatic pressure to single-walled carbon nanotubes produced by laser evaporation and here we report the results on the structural and transport property changes derived from *in situ* synchrotron x-ray diffraction analysis and four-probe measurements of the electrical resistivity conducted at various temperatures.

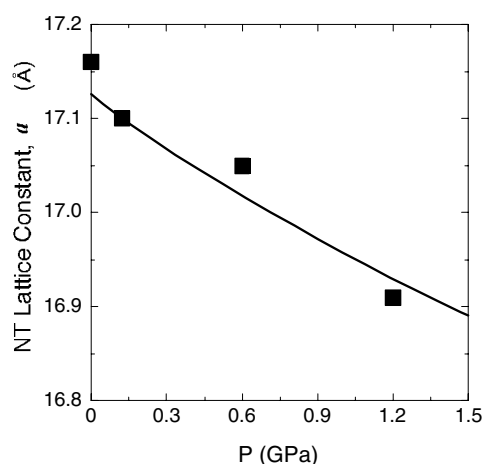


Figure 1. Linear dependence of the trigonal nanotube lattice constant on applied hydrostatic pressure. The volume compressibility of 0.024 GPa^{-1} was obtained for nanotubes of 14 \AA diameter. The solid curve is for calculated lattice constant.

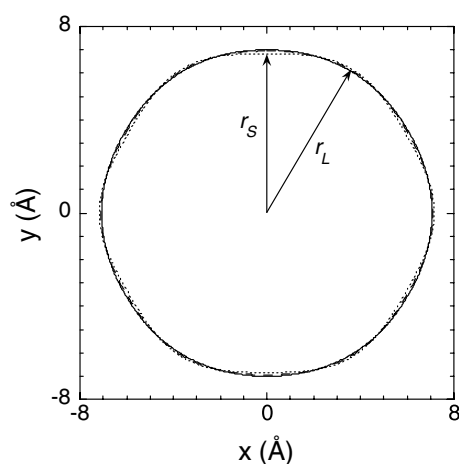


Figure 2. Morphological evolution of nanotube under increasing pressure.

2. Experimental details

Carbon nanotubes were produced by laser evaporation of graphite target at 1200°C using Ni/Co as catalyst. The thus-produced carbon nanotubes often form raft-like bundles which are stacked together in hexagonal close-packing [7]. The trigonal lattice diffracts x-rays with detectable intensity and therefore allows us to monitor the structural changes under various pressures.

High pressure was generated with a diamond anvil cell (DAC) using a liquid medium in order to obtain hydrostatic pressure. *In situ* synchrotron x-ray diffraction intensity data were collected using an imaging plate to assure high resolution. Electrical resistivity measurements were carried out using the four-probe method under various pressures generated by a piston apparatus. The resistivity measurement was performed at temperatures from 2 K up to room temperature (300 K).

3. Results and discussion

Single-walled carbon nanotubes were found to follow a smooth change of volume under pressure up to 2 GPa. Figure 1 shows the dependence of the lattice constant of the nanotube lattice on the applied hydrostatic pressure up to 2 GPa, beyond which the characteristic nanotube lattice reflection becomes too weak to be identifiable. As can be seen from the figure, the deformation of the nanotube lattice followed a smooth behaviour. From figure 1, the volume compressibility, defined by $\nu = -(1/V)(dV/dP)$, where V and P are the sample volume and the applied pressure, respectively, was obtained to be 0.024 GPa^{-1} .

The polygonization of the nanotubes can be specified by the ratio r_S/r_L , where r_S and r_L are the short and the long radial dimensions of the polygonized cross-section, respectively, as shown in figure 2. The inter-tubular separation between neighbouring nanotubes in the raft-like bundle is obtained from energetic calculations [8] using the Tersoff potential [9] which successfully explain the pressure dependence of the lattice constant as shown in figure 1.

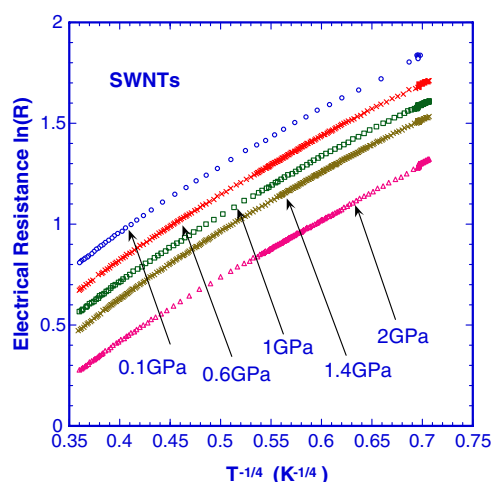


Figure 3. Electrical resistivity versus temperature of single-walled carbon nanotubes under various hydrostatic pressures. Semiconducting behaviour was apparent as shown in the curves.

It was found that when nanotubes are brought together, slight polygonization takes place even at zero pressure ($\eta = 0.991$) due to the van der Waals forces between neighbouring nanotubes. At 1.5 GPa pressure, the polygonization factor was obtained to be 0.982.

It has been shown theoretically that the electronic structure of carbon nanotubes is very sensitive to the topological change when their cross-sections are polygonized from circular to polygonal [10]. In particular, it was suggested that when a semiconducting nanotube is polygonized, its electronic band gap will be changed due to the faceting that induces $\sigma^*-\pi^*$ hybridization and lowers the symmetry of the otherwise cylindrical tubules.

Figure 3 shows a set of experimental data of the electrical resistance of the single-walled carbon nanotube material versus temperature at various pressures up to 2 GPa. From the relationship between electrical resistivity and temperature, the nanotube material under all pressures employed in the present experiment showed a semiconducting behaviour [11]. The band gaps deduced from the measured data are given in figure 4. When the pressure is below 1.5 GPa, the band gap exhibited a monotonic increase with increasing pressure. This phenomenon is attributed to the fact that, as the pressure increased, the band gap of the semiconducting nanotubes increases as a result of polygonization. However, at 1.5 GPa pressure, a structural phase transition is suggested to have occurred in which the polygonized nanotubes become elliptical in cross-section which led to the sudden drop in band gap as shown in figure 4. The morphological evolution of nanotubes versus pressure is schematically illustrated by the insets in figure 4. On the other hand, as has been discussed theoretically [12], the band gap of semiconducting nanotubes increases slightly as the ellipticity increases.

4. Conclusions

The volume compressibility of single-walled carbon nanotubes of 14 Å diameter has been measured to be 0.024 GPa^{-1} . Combined with the results obtained from numerical computations, it was found that the nanotubes must have been polygonized under the hydrostatic pressure employed in the present experiment. Measurement of the transport properties showed that the nanotubes exhibited a semiconducting behaviour with a narrow

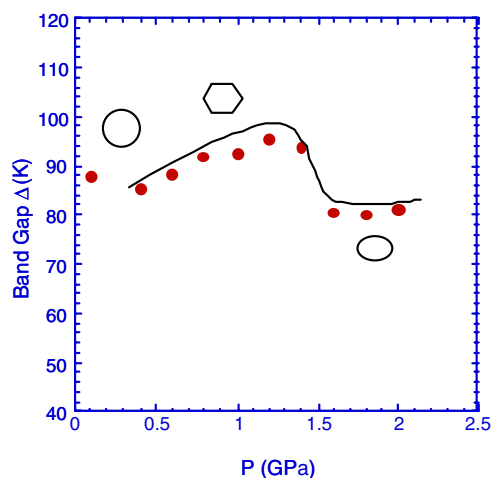


Figure 4. Pressure dependence of the band gap obtained from the electrical resistance on temperature dependence above 200 K. The band gap increases with pressure initially followed by a sudden drop at 1.5 GPa. The insets illustrate the morphological evolution of the cross-section of nanotubes at various pressures.

band gap. The polygonization due to the applied external pressure enlarged the band gap and a phase transition from trigonal to a new structure of lower symmetry is proposed to account for the abrupt drop of band gap observed experimentally.

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