EFFECT OF HYDROGEN IMPURITY ON THE THERMAL EXPANSION OF SINGLE-WALLED CARBON NANOTUBE BUNDLES

Popov S.N., Vinnikov N.A., Gavrilko V.G., Dolbin A.V.*, Esel'son V.B., Manzhelii V.G., Sundqvist B. (1)

B.Verkin Institute for Low Temperature Physics and Engineering of the NAS of Ukraine
47 Lenin Ave., Kharkov 61103 , UKRAINE
(1)Umea University, Department of Experimental Physics, 90187 Umea, Sweden
*Fax: 38 (044) 340 3370 E-mail: dolbin@ilt.kharkov.ua

Introduction

Carbon nanotubes (CNT) attracted attention of a large number of scientists around the world because of it unusual physical properties. And the articles related to it are increasing every day. But the effect of different gas impurities on the thermal expansion of single-walled carbon nanotube (SWNT) bundles is still unexplored. We should point that low-temperature investigations of thermal expansion can give us important data about dynamics of such systems.

Results and discussion

We have investigated the sample of pressure-oriented nanotubes. According to the results of [1], at this pressure nanotubes become aligned in plane perpendicular to pressure vector if the plate height not exceeds 0.4 mm. The average deviation of nanotubes from alignment plane is ~4° in this case. A cylindrical sample was compressed of pressure (11000 bar)-oriented SWNT plates, each being up to 0.4 mm thick. The sample was 7.2 mm high and 10 mm in diameter.

Radial thermal expansion of pure and H2-saturated SWNT samples was investigated using a low-temperature capacitance dilatometer in temperature range of 2.2—28 K.

Linear thermal expansion coefficient (LTEC) for pure nanotubes is negative below 5.5 K. This is because the Grüneisen coefficient of transverse acoustic vibrations is negative in two-dimensional (graphene) or quasi-two-dimensional (graphite, nanotubes) carbon systems. We found that at T ~ 5.5 K the LTEC of SWNT bundles become positive. The formation of SWNT bundles generates three-dimensional features in the system. As a result, the negative Grüneisen coefficients of such system decrease in magnitude and/or become positive. The hysteresis of LTEC of pure SWNT-bundles was found. Hysteresis is due to nonequilibrium state of the pressure-oriented nanotubes, and it becomes prominent on heating. Thermocycling within narrow temperature range (heating and cooling on 1—2 K) helps in achieving equilibrium state and makes hysteresis disappear.

Addition of hydrogen impurity changes LTEC significantly. Very high increase of LTEC values in case of hydrogen impurity is due to contribution of hydrogen molecules to vibrational spectra of "SWNT-bundles — H_2 impurity" system. Measurement results of LTEC of the "SWNT-bundles — H_2 impurity" system is shown of Fig. 1.

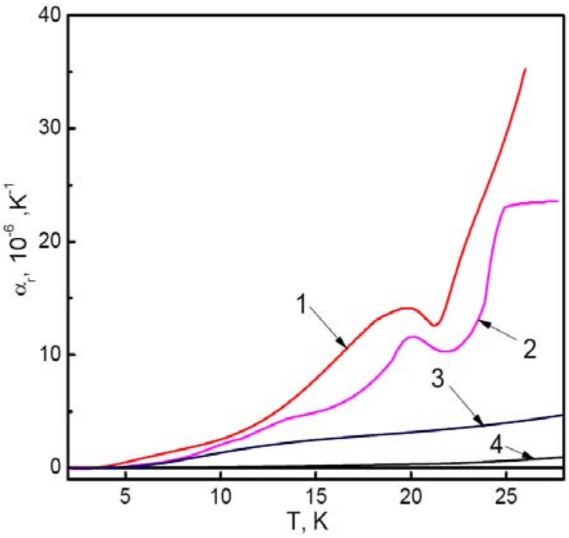

Fig. 1. Radial thermal expansion of SWNT:
1) with H_2 impurity;
2) after partial removal of H_2 impurity;
3) with Xe impurity;
4) pure SWNT.

It is necessary to point that doping with hydrogen caused the shift of the temperature interval where negative thermal expansion was observed to lower temperatures (2.2—3 K). Narrowing of temperature interval of negative thermal expansion in case of H_2 and Xe impurity is caused by impact of impurity molecules on the transverse vibrations of nanotubes. The filling of interior sites of nanotubes with H_2 molecules results in lowering of the amplitude and in raising the frequency of the transverse vibrations of nanotube surface atoms. This leads to lowering of contribution of negative part and shifts its temperature interval to lower temperatures.

It is seen that LTEC values of SWNT — H_2 system is much higher than that for SWNT — Xe.

It shows that H_2 molecules which can penetrate to interstitial and interior sites of nanotube makes significant contribution to LTEC values of H_2—SWNT system, because these sites are unaccessible for impurity molecules in case of Xe impurity[3].

It is also necessary to point that there is no hysteresis of LTEC in H_2-doped SWNT bundles.

Conclusions

Dilatometric investigations of low-temperature thermal expansion of single-walled carbon nanotube bundles showed:

1. Radial thermal expansion coefficient of SWNT-bundles rises significantly when doping with H_2.
2. Temperature interval of negative thermal expansion of H_2-doped SWNT is shifted to low temperatures in comparison with pure SWNT.

References

1. Benidab N., Almairac R., Sauvajol J.-L., Rols S., *J. of Appl. Phys.* **93**, 1769 (2002).

2. Dolbin A.V., Esel'son V.B., Gavrilko V.G., Manzhelii V.G., Vinnikov N.A., Popov S.N. and Sundqvist B., *Fiz. Nizk. Temp.* **35**, to be published (2009)

3. Kuznetsova A., Mawhinney D.B., Naumenko V., Yates J.T., Jr., J. Liu, Smalley R.E., *Chem. Phys. Lett*, 2000;321: 392-296,