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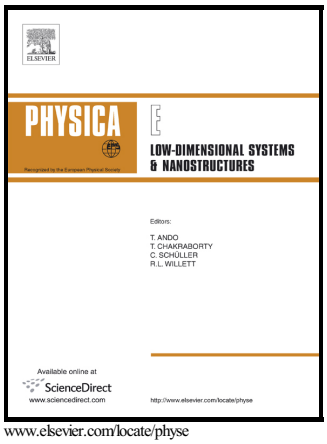

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Vibrational heat capacity of carbon nanotubes at low and ultra-low temperatures

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To measure a heat capacity of a single carbon nanotube is impossible nowadays and experiments are performed with the nanotubes bundles. However, the existing theories of carbon nanotubes heat capacity do not take into account an interaction between an individual nanotube and its environment. Here we show that due to this fact the known theories overestimate the heat capacity in the ultra-low temperature limit. In this region the main contribution to heat capacity of a single free nanotube is caused by the bending phonon mode with a quadratic dispersion. At T=1 K different known models of the phonon dynamics lead to the theoretical specific heat which is at least 10 times *greater* than the experimentally measured value. To overcome this contradiction we take into account the interaction between an individual nanotube and its environment. As a result, the nanotube loses its Goldstone degrees of freedom and essential reconstruction of the lowest-energy part of its phonon spectrum occurs. The theory proposed explains experimental data on thermal capacity in the ultra-low temperature range and simultaneously demonstrates better agreement with experiments in the region 2.5K<T<40K than the preceding models.

Keywords: carbon nanotubes, low temperatures, heat capacity, phonon spectrum, pinning, 2D membranes.

I. Introduction

Since their discovery [1], carbon nanotubes (CNTs) are investigated due to their unique physical properties [2-4]. Recent finding of extremely high thermal conductivity of graphene [5] has stimulated a new wave of interest to the theory of thermal properties of low-dimensional graphene-related systems [6, 7] including CNTs. Their heat capacity is one of those important thermal characteristics, which is poorly understood to date. Unfortunately, there are only a few experimental articles presenting results of single-walled carbon nanotubes (SWCNTs) specific heat measurements in the low temperature (LT) region. For the first time specific heat temperature dependences were measured in the ranges 0.6<T<210 K [8] and 2<T<300 K [9]. Later, some experiments were carried out at ultra-low temperatures (ULT) [10, 11]. As it is known, the main contribution to heat capacity of CNTs and their bundles comes from phonon modes even at ULT.

Currently, the two groups of approaches to theoretical studies of CNTs vibrational heat capacity exist. The first one include the methods based on computational modeling of SWCNTs vibrational dynamics, in particular, force-constant models [12-15] and methods of molecular dynamics [16, 17]. Large number of atoms in CNTs leads to huge amount of computation. Besides, complex multiparticle interaction potentials are to be used in order to yield satisfactory results. The authors of theoretical papers [12, 14, 15-17] state that their results are in quite good quantitative agreement with experimental data [9, 10]. However, in [12] it is reported that specific heat temperature dependence T^x has two regimes: linear at LT, then quadratic. On the contrary, in [17] it is stated that T^x function is quadratic at LT and then linear. Moreover, model [14] predicts the existence of three regimes instead of two in the temperature range 0<T<100 K: x=1/2 at ULT (approximately at 0.5-0.8 K), then x=1 at T<5 K and x>1 at T>5 K. Slight adjustments to the results of [14] were made in [15] by specifying that 0.6<x<1 in the second regime. Nevertheless, the specific heat temperature dependence obtained in [15] is in good agreement with experimental data [9] only in the range 2<T<30 K.

Another well-known group of methods for SWCNTs thermal characteristics calculation is based on different continuous models, which usually consider SWCNT as a thin cylindrical shell [18-21].