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Magnon specific heat and free energy of Heisenberg ferromagnetic single-walled nanotubes: Green's function approach

Bin-Zhou Mi [a,b,*], Liang-Jun Zhai [c], Ling-Ling Hua [a]

[a] Department of Basic Curriculum, North China Institute of Science and Technology, Beijing 101601, China
[b] Department of Physics, School of Mathematics and Physics, University of Science and Technology Beijing, Beijing 100083, China
[c] The School of Mathematics and Physics, Jiangsu University of Technology, Changzhou 213001, China

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ABSTRACT

The effect of magnetic spin correlation on the thermodynamic properties of Heisenberg ferromagnetic single-walled nanotubes are comprehensively investigated by use of the double-time Green's function method. The influence of temperature, spin quantum number, diameter of the tube, anisotropy strength and external magnetic field to internal energy, free energy, and magnon specific heat are carefully calculated. Compared to the mean field approximation, the consideration of the magnetic correlation effect significantly improves the internal energy values at finite temperature, while it does not so near zero temperature, and this effect is related to the diameter of the tube, anisotropy strength, and spin quantum number. The magnetic correlation effect lowers the internal energy at finite temperature. As a natural consequence of the reduction of the internal energy, the specific heat is reduced, and the free energy is elevated.

1. Introduction

In recent years, magnetic nanotubes (MNTs) have attracted great interest in experimental [1–20] and theoretical [18,21–40] studies due to their fascinating properties originating from low dimensionality and quantum fluctuations. Of all the physical properties of MNTs, the thermodynamic and magnetic behavior plays a crucial role in potential applications, particularly in ultra-high-density magnetic storage devices, biotechnology, nanomedicine, and nanoelectronic devices, etc. Seen from the perspective of the theory, the magnetic properties of MNTs can be very easily described by the well-known Ising model and Heisenberg model using various theoretical techniques, such as Monte Carlo simulations [31–34], the effective-field theory [35–39], and many-body Green's function method (MBGFM) of the quantum statistical theory [40].

The effects of magnetic correlations (MCs) on thermodynamic properties in ferromagnetic single-walled nanotubes (FM-SWNTs) with Heisenberg model are to be further investigated within the MBGFM. The aim of the present paper is to have a satisfactory investigation of the thermodynamic quantities of FM-SWNTs, including internal energy, free energy, and magnon specific heat. To do so, we follow the routine proposed in Ref. [41], and derive an expression for computing the internal energy, which contains transverse magnetic correlation (TMC) and longitudinal magnetic correlation (LMC), and valid for spin quantum number $S \geq 1$. With this formula, we are able to evaluate the other two thermodynamic quantities and thoroughly analyze the effect of the MCs. The cases where an external magnetic field is introduced are also investigated. We find that the MCs play an important role since they change the internal energy remarkably. The MCs depress the thermal motion of the system, so that they lower the internal energy, magnon specific heat, and elevate the free energy.

In Section 2 we present our model and put down the Heisenberg exchange Hamiltonian of FM-SWNTs. Then we briefly outline the formulas derived by the MBGFM. In Section 3, numerical computation is carried out and the role of temperature, spin quantum number, diameter of the tube, anisotropy strength and external magnetic field on thermodynamic quantities are carefully analyzed. Furthermore, the effects of MCs on internal energy and other thermodynamic quantities are clearly demonstrated. At last, Section 4 presents our concluding remarks.

2. Model and formulas

We consider a Heisenberg ferromagnetic single-walled nanotubes (HFM-SWNTs) with armchair type [31,40]. In this model, the spins, localized on the sites of a square lattice wall, were assumed

* Corresponding author at: Department of Basic Curriculum, North China Institute of Science and Technology, Beijing 101601, China.
E-mail address: mbzfjerry2008@126.com (B.-Z. Mi).

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to interact via a Heisenberg exchange coupling limited to nearest neighbor (nn) sites. Moreover, a uniaxial anisotropy was assumed to favor the nanotube axis, and a magnetic field was applied along such an easy direction. The Hamiltonian is given as follows:

$$H = -\frac{1}{2}J\sum_{[i,j]} S_i \cdot S_j - D\sum_i (S_i^z)^2 - B_z\sum_i S_i^z \tag{1}$$

The first term represents the Heisenberg exchange energy with exchange parameter J. The subscripts i and j denote the lattice sites, and $[i, j]$ mean that the nn exchanges are involved. The second term describes the uniaxial anisotropy. It was indeed possible for the uniaxial anisotropy to appear in real nanotube materials [1–3]. Note that z-axis labels the directions of tube axis. The anisotropy strength of D is usually believed to be less than J by two orders of magnitude. The last term stands for Zeeman energy when an external magnetic field B_z is applied along the tube axis. In this paper, we set Boltzman constant $k_B=1$. In calculation, we fix $J=100$, and all parameters are taken as dimensionless quantities. There are N and m sites along the axis and circumference of the tube, respectively.

The MBGFM is a powerful means [42–48] to calculate magnetization and other thermodynamic quantities [49–58] of the Heisenberg model since this method takes into account the spin fluctuation, and is valid in the whole temperature range. In order to study magnetic and thermodynamic properties of HFM-SWNTs, we introduce the retarded Green's function

$$G_{ij}^R(t-t') = \langle\langle S_i^+(t);\, B(t')\rangle\rangle^R, \tag{2}$$

where $B = e^{uS_j^z}S_j^-$, and u is a parameter. After time Fourier transformation, the retarded Green's function is denoted as $\langle\langle S_i^+;\, B\rangle\rangle^R(\omega) = G_{ij}^R(\omega)$, and then we obtain the equation of motion:

$$(\omega + i0^+)\langle\langle S_i^+;\, B\rangle\rangle^R(\omega) = \langle [S_i^+,\, B]\rangle + \langle\langle [S_i^+,\, H];\, B\rangle\rangle^R(\omega) \tag{3}$$

The higher order Green's function appearing in the equation of motion is decoupled by random phase approximation (RPA). As for the term concerning single-ion anisotropy term in Eq. (1), we adopt the Anderson and Callen's decoupling [44,45].

Now the Green's function is further Fourier transformed along the tube axis with periodic boundary condition. The wave vector component, denoted as p, is within the first Brillouin zone. As for circumferential direction, the discrete Fourier transformed is taken with periodicity condition. The site number m along perimeter given, the argument after transformation, denoted as q, should meet following condition:

$$q = \frac{2\pi n}{ma}\ (n = 0,\, 1,\, 2,\, \ldots,\, m-1). \tag{4}$$

Then the Fourier transformation of the Green's function is written as

$$G_{ij}^R(\omega) = \frac{1}{N}\sum_p \frac{1}{m}\sum_{n=0}^{m-1} G^R(p,\, q,\, \omega)e^{ipa(i_z-j_z)}e^{i2\pi n(i_y-j_y)/m}. \tag{5}$$

The subscripts z and y label the directions of tube axis and circumference, respectively. The perimeter of the tube is ma, and the diameter d of the tube is determined by $\pi d = ma$. Hereafter we also simply refer m as diameter.

The retarded Green's function $G^R(p, q, \omega)$ can be expressed as follows:

$$G^R(p,\, q,\, \omega) = \frac{[S_i^+,\, B]}{\omega + i0^+ - E(p,\, q)}, \tag{6}$$

where the expression of energy spectrum is

$$E(p,\, q) = \langle S^z\rangle[J(0) - J(k)] + 2DC\langle S^z\rangle + B_z. \tag{7}$$

In Eq. (7),

$$J(0) = 4J, \tag{8}$$

$$J(k) = 2J[\cos(pa) + \cos(qa)], \tag{9}$$

and

$$C = 1 - \frac{1}{2S^2}[S_b - \langle (S^z)^2\rangle], \tag{10}$$

where $S_b = S(S+1)$. Note that we use $\langle S^z\rangle$ to denote magnetization and S to denote spin quantum number. Following the Callen's method proposed in Ref. [46], the magnetization of arbitrary S is expressed [47,48] as follows,

$$\langle S^z\rangle = \frac{(\Phi+1+S)\Phi^{2S+1} - (\Phi - S)(\Phi+1)^{2S+1}}{(\Phi+1)^{2S+1} - \Phi^{2S+1}}, \tag{11}$$

and

$$\langle (S^z)^2\rangle = S(S+1) - (1+2\Phi)\langle S^z\rangle, \tag{12}$$

where

$$\Phi = \frac{1}{N}\sum_p \frac{1}{m}\sum_q \frac{1}{e^{\beta E(p,q)} - 1}. \tag{13}$$

Where $\beta = 1/T$, the inverse of temperature.

The internal energy is a primary quantity of the system, which is simply the thermodynamic average of the Hamiltonian. Let us discuss the formula of the internal energy. It contains three terms: the Heisenberg exchange energy (hereafter called as magnetic correlation energy), the anisotropy energy, and the Zeeman energy. Under the mean field approximation (MFA), the Heisenberg exchange energy in Eq. (1) per lattice site was decoupled [49] as follows:

$$\left\langle -\frac{1}{2}J\sum_{[i,j]} S_i \cdot S_j \right\rangle \Big/ Nm = -\frac{1}{2}J(0)\langle S^z\rangle^2. \tag{14}$$

Its essence is to neglect the transverse and longitudinal correlation between site i and its neighbor j. In other words, the MFA neglects the spin wave excitation. While for the internal energy or the specific heat, that involve TMC and LMC between spins, one has to go beyond MFA. In fact, in a pure Heisenberg magnetic system, the magnetic spin correlation contains two parts: the TMC and the LMC, and both of them are nonzero at or above the magnetic phase transition temperature [50,51]. In this sense, the magnetic spin correlations effect has a considerable contribution to the thermodynamics properties of HFM-SWNTs.

Magnon specific heat in title means considering the magnon contribution to specific heat. This paper, being the continuation of our previous research of the magnetic correlation in ferromagnetic single-walled nanotubes [52], will be devoted to the evaluation of the magnetic spin correlations contribution to the specific heat and free energy.

Several energies are defined as follows: longitudinal correlation energy LCE $E_{LC} = -\frac{1}{2Nm}\sum_{i,j} J_{ij}\langle S_i^z S_j^z\rangle$, transverse correlation energy TCE $E_{TC} = -\frac{1}{2Nm}\sum_{i,j} J_{ij}\langle S_i^+ S_j^-\rangle$, magnetic correlation energy MCE $E_{MC} = E_{TC} + E_{LC}$, mean field energy MFE $E_{MF} = -\frac{1}{2}J(0)\langle S^z\rangle^2$ and internal energy $E = \frac{\langle H\rangle}{Nm}$. Please note that they are all the energies per lattice site. For a precise theoretical treatment we follow the routine of Ref. [41] where the longitudinal and transverse correlation functions were calculated carefully. The expressions of these energies as follows:

$$E_{\mathrm{TC}} = -\langle S^z\rangle\Phi_a, \tag{15}$$