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Magnetic correlations in ferromagnetic single-walled nanotubes

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ABSTRACT

The magnetic correlations, including transverse magnetic correlation (TMC) and longitudinal magnetic correlation (LMC), of 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 magnetic correlations are carefully calculated. An interesting result is that for the two smallest spin quantum numbers $S=1$, and 3/2, the LMC around the Curie point is negative, demonstrating that the neighboring spins in ferromagnetic single-walled nanotubes are antiparallel to each other along the tube axis direction in spite of the ferromagnetic exchanges between them, while it is not so along the transverse direction. This is due to the fact that the quantum spin fluctuation is believed anisotropic. The effect of the LMC is always in contrary to that of the TMC effect: if one is stronger, the other is weaker.

1. Introduction

Since the discovery of carbon nanotubes (CNTs) [1], tubular structures have been widely investigated because of their fascinating structure and unique properties. Following the discovery, different chemical compositions of the nanotubes have been successfully synthesized by various methods [2–9]. More recently, the enthusiasm of studying magnetic nanotubes (MNTs) has been rising from both experimental [10–26] and theoretical [27–46] standpoints due to their promising applications, such as ultrahigh-density magnetic storage devices, biomagnetic sensors, nanomedicine, molecular devices, catalysts, and nanoelectronic devices, etc. Research on MNTs is expanding into newer and broader fields, including both physical properties and possible unique applications in various areas.

There have been several theoretical methods for the magnetic and thermodynamic properties of MNTs, such as micromagnetic simulation [27–32], continuum theory of ferromagnetism [33–36], Monte Carlo simulations [37–40], *ab initio* density functional theory calculations [41], effective-field theory [42–45], and many-body Green's function method (MBGFM) of quantum statistical theory [46]. As far as we know, the magnetic correlation (MC) effect of ferromagnetic single-walled nanotubes (FM-SWNTs) has not yet been satisfactorily investigated. This paper is devoted to this point.

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 MC are carefully analyzed. At last, Section 4 presents our concluding remarks.

2. Model and formulas

We have a two-dimensional (2D) square lattice with the nearest neighbor distance being a and a spin situated in each lattice site. In Fig. 1 one unit cell is depicted. Then the plane is rolled up along one of the coordinate axes, say w_1 direction in Fig. 1, to form a nanotube with a diameter d. This kind of rolling is just what was named as armchair type tube [27,46]. Zigzag type tube is similarly formed by rolling along the diagonal direction of a cell, i. e., w_2 direction in Fig. 1. Theoretical study showed that the magnetic properties of FM-SWNTs under fixed diameter are independent of rolling helicity [46]. Therefore, we only study the case of armchair type tubes.

The Hamiltonian is given as

$$H = -\frac{1}{2}J\sum_{[i,j]} \mathbf{S}_i\cdot\mathbf{S}_j - D\sum_i (S_i^z)^2 - B_z\sum_i S_i^z. \quad (1)$$

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 nearest neighbour (nn) exchanges are involved. The second term describes the uniaxial anisotropy. Note that z-axis labels the directions of tube axis. The anisotropy

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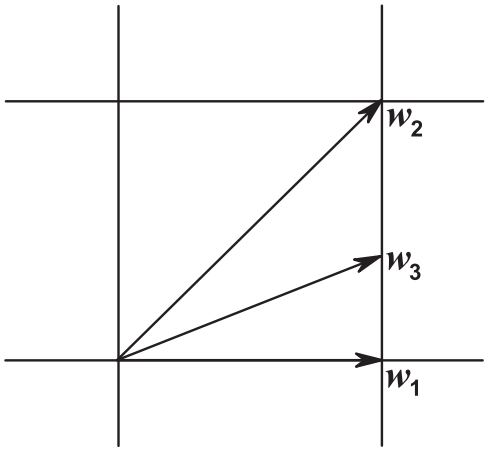

Fig. 1. One unit cell of a 2D monolayer with square lattice and three possible rolling directions.

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.

In a pure Heisenberg magnetic system, the MC contains two parts: transverse magnetic correlation (TMC) and longitudinal magnetic correlation (LMC). They are defined as $C_T = \sum_{[i,j]} \langle S_i^+ S_j^- \rangle$ and $C_L = \sum_{[i,j]} \langle S_i^z S_j^z \rangle$ [48,49], respectively. The TMC is easily calculated by means of the well-known spectral theorem [50–52]. The LMC, however, is not easy to evaluate precisely. Some further approximations are resorted. Under the mean field approximation (MFA), the LMC was decoupled by [53] $C_L = \sum_{[i,j]} \langle S_i^z \rangle \langle S_j^z \rangle$ which became zero when temperature is at or above the magnetic phase transition temperature (T_M). Recently, a more satisfactory expression of the LMC beyond the MFA decoupling was obtained [47], and it was found that LMC in some three dimensional magnetic systems (bcc and fcc lattice) was nonzero at T_M. This formulism was successfully applied in Ref. [54]. Following the routine proposed in Ref. [47], we can realize the better physical picture of the MC in the FM-SWNTs.

As long as the internal energy of a system is known, it is easy to calculate other thermodynamic quantities such as free energy, specific heat, entropy and so on. Three energies are defined as follows: longitudinal correlation energy LCE $E_{LC} = -\frac{1}{2Nm} J \sum_{[i,j]} \langle S_i^z S_j^z \rangle$, transverse correlation energy TCE $E_{TC} = -\frac{1}{2Nm} J \sum_{[i,j]} \langle S_i^+ S_j^- \rangle$ and internal energy $E = \frac{\langle H \rangle}{Nm}$. Please note that they are all the energies per lattice site.

The MBGFM is employed following the standard routine [46–52,55–57]. Here we briefly outline the formulas without presenting the detailed derivation. The well-known spectral theorem and its derivative with respect to time t help us to calculate various thermodynamic quantities. The TMC and LMC are expressed as

$$C_T = \frac{2\langle S^z \rangle \Phi_a}{J}, \tag{2}$$

$$\begin{aligned} C_L\,(S=1) &= \frac{1}{2J}\{[2-\langle S^z\rangle - 3\langle (S^z)^2\rangle]\Phi_a + [-2-3\langle S^z\rangle + 3\langle (S^z)^2\rangle]\Phi_b\} \\ &\quad + 4\langle S^z\rangle - \frac{D}{J}[2\langle (S^z)^2\rangle - \langle S^z\rangle - 1] - \frac{B_z}{J}(\langle S^z\rangle - 1), \end{aligned} \tag{3}$$

$$\begin{aligned} C_L\,(S>1) &= \frac{1}{J(S_b+1)}\{[S_b - (2S_b+1)\langle S^z\rangle - 3\langle (S^z)^2\rangle + 4\langle (S^z)^3\rangle]\Phi_a \\ &\quad + [-S_b - 3\langle S^z\rangle + 3\langle (S^z)^2\rangle]\Phi_b + J(0)(\langle (S^z)^3\rangle + S_b)\langle S^z\rangle \\ &\quad + D[2\langle (S^z)^4\rangle + \langle (S^z)^3\rangle - 2(S_b+1)\langle (S^z)^2\rangle + (S_b-1)\langle S^z\rangle + S_b] \\ &\quad + B_z[S_b - (S_b+1)\langle S^z\rangle + \langle (S^z)^3\rangle]\}, \end{aligned} \tag{4}$$

and $S_b = S(S+1)$. Here, we use $\langle S^z \rangle$ to denote magnetization and S to denote spin quantum number. Φ_a, and Φ_b are expressed as

$$\Phi_a = \frac{1}{N}\sum_p \frac{1}{m}\sum_q \frac{J(p,q)}{e^{\beta E(p,q)}-1}, \tag{5}$$

and

$$\Phi_b = \frac{1}{N}\sum_p \frac{1}{m}\sum_q \frac{E(p,q)}{e^{\beta E(p,q)}-1}, \tag{6}$$

where $\beta = 1/T$, the inverse of temperature. In Eqs. (5) and (6),

$$E(p,q) = [4J - J(p,q) + 2DC]\langle S^z\rangle + B_z, \tag{7}$$

and

$$J(p,q) = 2J[\cos(pa) + \cos(qa)], \tag{8}$$

where

$$\langle S^z\rangle = \frac{(\Phi+1+S)\Phi^{2S+1} - (\Phi-S)(\Phi+1)^{2S+1}}{(\Phi+1)^{2S+1} - \Phi^{2S+1}}, \tag{9}$$

and

$$\Phi = \frac{1}{N}\sum_p \frac{1}{m}\sum_q \frac{1}{e^{\beta E(p,q)}-1}. \tag{10}$$

The correlation functions $\langle (S^z)^2\rangle$, $\langle (S^z)^3\rangle$ and $\langle (S^z)^4\rangle$ and the coefficient C were expressed in Ref. [47]. The perimeter of the tube is ma, and the diameter d of the tube is determined by $\pi d = ma$. Hereafter we simply refer m as the diameter. The quantity p is the wave vector along the z direction and takes the values within the first Brillouin zone. The value of q is determined by $q = \frac{2\pi n}{ma}$, $(n = 0, 1, 2, \ldots, m-1)$. Eqs. (2)–(10) are the transcendental equations of TMC and LMC. From these equations, they can be calculated as a function of spin quantum number S, temperature T, diameter of the tube m, anisotropy strength D and external magnetic field B_z.

Under the MFA, the LMC was decoupled for the present lattice as follows:

$$C_L = 4\langle S^z\rangle^2. \tag{11}$$

As the diameter of the tube m goes to infinite, a single-walled nanotube approaches a 2D monolayer. For the sake of comparison, we also calculate the MC of the 2D monolayer. For $S=1/2$, the anisotropy term in Eq. (1) does not play a role. Therefore, in the present paper, we study the cases of $S=1$, 3/2, 2, 5/2, 3.

3. Results and discussions

Fig. 2(a) plots the temperature dependence of the TMC and LMC for several diameters of tubes and five S values, where the external magnetic field B_z is absent. At zero temperature, $T=0$ K, the TMC is zero, which reveals that along the z direction, the neighboring spins are strictly parallel to each other. With temperature increasing, the TMC increases while the LMC decreases until the Curie point (T_C). This means that the longitudinal correlation effect becomes weaker and the transverse correlation effect stronger. The nearest neighbor spins gradually deviate from the parallel configuration along the z direction as temperature rises. A