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Theoretical investigations of magnetic properties of ferromagnetic single-walled nanotubes

Bin-Zhou Mi [a], Huai-Yu Wang [b,*], Yun-Song Zhou [a]

[a] Department of Physics, Capital Normal University, Beijing 100048, China
[b] Department of Physics, Tsinghua University, Beijing 100084, China

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ABSTRACT

The magnetic behaviors of ferromagnetic single-walled nanotubes are systematically investigated by use of the many-body Green's function method of quantum statistical theory. The spontaneous magnetization, absolute value of ferromagnetic energy, area of hysteresis loop and coercivity increase with diameter of the tubes and spin quantum number, and decrease with temperature. Curie temperature increases with diameter and spin quantum number. As the diameter of the tube tends to infinity, all the numerical results approach to those of a two-dimensional monolayer. The dependences of initial susceptibility on temperature and diameter below and above Curie point are contrary. The calculated results are compared with experimental results where possible, and are qualitatively in agreement with the latter. The Curie temperature is determined by the tube diameter and independent of rolling helicities.

1. Introduction

In recent years, the fundamental research on low-dimensional materials has been unfolding following the vigorous development of nanotechnology. In particular, considerable efforts have been made in the area of quasi-one-dimensional (1D) nanostructured materials, such as nanotubes, nanowires, and nanorods. More recently, ferromagnetic (FM) nanotubes have been successfully fabricated by various methods [1–5], and their magnetic properties have been investigated experimentally [3–9]. Magnetic nanotubes have been attracting much attention due to their potential applications, such as ultrahigh-density magnetic storage devices, biotechnology, nanomedicine, and nanoelectronic devices, etc.

There have been several theoretical methods for the investigation of magnetic nanotubes, such as micromagnetic simulation [9–11], continuum theory of ferromagnetism [12–15], and Monte Carlo simulations [16]. We believe that the discrete lattice effects and the quantum mechanical effects are of importance for the magnetic materials with nanometer-scale. Therefore, a description of the magnetic properties of nanotubes with discrete sites based on quantum mechanism is inevitable. Indeed, Konstantinova [16] simulated the magnetic structures of single-walled nanotubes with Heisenberg interaction and dipole interaction. Because of the limitation of computation time, the tubes they studied were rather thin, i.e., the lattice sites in circumference are less and equal to 12. We think the investigations of thicker nanotubes and other magnetic properties such as Curie temperature, hysteresis loop and coercivity, etc. are also desirable. In this paper, we present a systematic investigation of ferromagnetic single-walled nanotubes. The magnetic properties are explored as far as possible. This paper is organized as follows. In Section 2 we put down the Heisenberg exchange Hamiltonian of tubes based on the observation of experiment [3]. Then we briefly outline the formulas derived by use of the many-body Green's function method (MBGFM) of quantum statistical theory. In Section 3, numerical computation is carried out and the results are compared with experiments where possible. We demonstrate that no matter what rolling helicity is, the magnetic physical quantities are determined as long as the tube diameter is known. At last, Section 4 presents our concluding remarks.

2. Model and method

The magnetic single-walled nanotubes investigated in this paper are constructed as follows. First, we have a monolayer of two-dimensional (2D) square lattice with spins situated at the lattice sites. The distance between the nearest neighboring site is a. In Fig. 1 one unit cell is depicted. Then the monolayer is rolled up along one of the coordinate axes, say $\boldsymbol{w}_1$ direction in Fig. 1, to form a nanotube with a diameter d. The site number of circumferential direction is denoted as m. This kind of rolling is just what was named as armchair type in Ref. [16]. Here we do not

* Corresponding author. Tel.: +86 1062788193; fax: +86 1062781604.
E-mail address: wanghuaiyu@mail.tsinghua.edu.cn (H.-Y. Wang).

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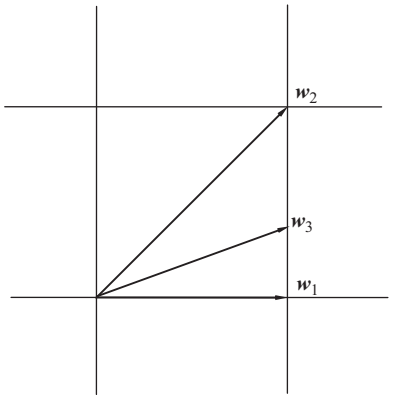

Fig. 1. One unit cell of a 2D monolayer with square lattice and the possible rolling directions.

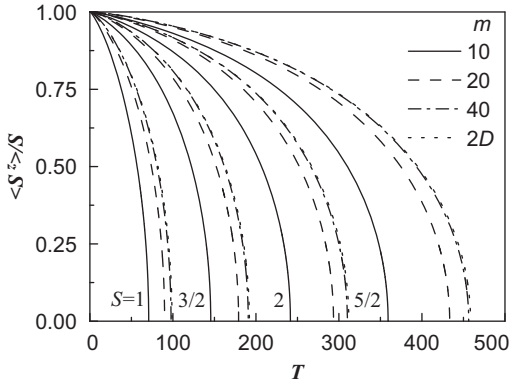

Fig. 2. The spontaneous magnetization as a function of temperature for several diameters of the tube and four S values. The results of a 2D monolayer are also plotted for comparison.

depict the geometrical structure of the nanotubes after rolling. Readers can refer to Fig. 2 in Ref. [16].

According to experimental observation, the easy axis of Fe nanotubes could be parallel to the tube axis [3]. Based on this fact we put down the Heisenberg exchange Hamiltonian as follows:

$$H = -\frac{1}{2}J\sum_{(i,j)}\mathbf{S}_i\mathbf{S}_j - K_2\sum_i (S_i^z)^2 - B_z\sum_i S_i^z. \tag{1}$$

The first term represents the Heisenberg exchange energy. The subscripts i and j denote the lattice sites, and J is the FM exchange parameter. Here only the nearest neighbor exchange is considered. The second term describes the uniaxial anisotropy showing the z-axis to be the easy axis, which can be either single-ion anisotropy or shape anisotropy or both. The strength of K_2 is usually believed to be less than J by orders of magnitude. The last term stands for Zeeman energy when an external magnetic field is applied along the z-direction. In calculation, we set $J=100$, $K_2=1$, and all parameters are taken as dimensionless quantities. In Ref. [16] the dipole interaction was also included. However, it was pointed out that when the dipolar and the Heisenberg interactions were introduced simultaneously, the spin configurations were very similar to the ones in the case of nearest-neighboring interactions. In fact, if both a uniaxial anisotropy ($K_2>0$) and a very small dipolar interaction ω exist, $\omega/K_2 \ll 1$, $\omega/J \ll 1$, then the dipole interaction can be neglected. In Ref. [16] $\omega/J=0.001$, which corresponded to $\omega/K_2=0.1$. Therefore, we do not consider the dipole interaction in this paper. Since only single-walled nanotubes are studied, we do not concern the possibility of the formation of domain walls. Therefore, the calculated magnetic hysteresis results in this paper are based on the coherent mode mechanism.

In studying magnetic properties of materials, the many-body Green's function method (MBGFM) has been a powerful mean [17–19] since this method takes into account the spin fluctuation, and is valid in the whole temperature range. The retarded Green's function is constructed by raising and lowering spin operators

$$G_{ij}^R(t-t') = \langle\langle S_i^+(t); S_j^-(t')\rangle\rangle^R = -i\theta(t-t')\langle[S_i^+(t), S_j^-(t')]\rangle. \tag{2}$$

After time Fourier transformation, the retarded Green's function is denoted as $\langle\langle S_i^+; S_j^-\rangle\rangle^R(\omega) = G_{ij}^R(\omega)$, and then we obtain the Eq. of motion

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

The higher order Green's function appearing in the equation of motion is decoupled by random phase approximation (RPA). As for the term concerning anisotropy term in Eq. (1), we adopt the Anderson and Callen's decoupling [20,21].

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:

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

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

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

where N is the site number along the tube axis. 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:

$$G^R(p,q,\omega) = \frac{2\langle S^z\rangle}{\omega+i0^+-E(p,q)}, \tag{6}$$

where the expression of energy spectrum is

$$E(p,q) = \langle S^z\rangle[4J-J(p,q)] + 2K_2C\langle S^z\rangle + B_z. \tag{7}$$

In Eq. (7),

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

and

$$C = 1-\frac{1}{2S^2}\left[S(S+1)-\langle(S^z)^2\rangle\right]. \tag{9}$$

Note that we use $\langle S^z\rangle$ to denote magnetization and S to denote spin quantum number. From Eqs. (7)–(9), we see that the energy spectrum has just the same form as that of a 2D plane. In this sense we can regard p and q in Eqs. (5)–(8) as two components of a vector $\boldsymbol{k}$: $\boldsymbol{k}=(p,q)$. One only needs to keep in mind that one of the vector components, q, takes the values determined by Eq. (4). As m tends infinitely large, the nanotube tends to be a 2D monolayer. This guarantees that as the diameter of the tube rises, all the physical quantities will tend to those of a 2D monolayer. The magnetization of arbitrary S is expressed [17]