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Blocking temperature in nanocrystalline systems with "alloy-like" ferromagnetic–antiferromagnetic heterogeneous morphology

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ABSTRACT

A modified Monte Carlo Metropolis method is performed to simulate the blocking temperature (T_B) in an "alloy-like" ferromagnetic $(FM)_x$−antiferromagnetic $(AFM)_{1-x}$ heterogeneous system with geometrical frustration. It is found that the blocking temperature, at which the field-cooled (FC) and zero-field-cooled (ZFC) magnetization curves are splitting, changes little for $x \leq 0.5$ initially, then decreases obviously with the increase in x. Some discrete error bars emerge for large x owing to the superparamagnetic or agglomerate behavior of the small antiferromagnet. Using a thermal fluctuation model, an analytic expression for T_B as a function of x is obtained. By calculating the curves of temperature derivative of the difference between FC and ZFC magnetizations and analyzing the distribution of energy barriers, we interpret the dependence of T_B on different proportions of ferromagnetic phase in detail.

1. Introduction

Over the last few years interest in the research on the synthesis of magnetic nanoparticles (NPs) and on the characterization of their properties has grown enormously. A common characteristic of nanostructured magnetic materials is the coexistence of two or more phases, magnetically and/or structurally different, which are modulated on a length scale of the order of a nanometer [1–4]. Most of the NPs systems are heterogeneous and prepared artificially by physical and chemical techniques, such as milling, chemical reaction, gas condensation, electrodeposition, or microemulsion, and so on [5]. Moveover, some of them can come from the nature, e.g., ferrite particles [6] and perovskite cobaltite [7,8], and so forth. The alloys [9] of Co, Fe, or Ni and various Heusler alloys [10,11] obtained by sol–gel or conventional arc melting method as well as nanogranular Co/CoO, Ni/NiO systems produced by ball milling and redox [12,13] have revealed tremendous potential from application point of view.

When this heterogeneous system consisting of a ferromagnetic (FM) and an antiferromagnetic (AFM) phases is cooled in a magnetic field, it may exhibit an additional unidirectional anisotropy due to the magnetic interaction at the interface. This phenomenon called exchange bias (EB) was first discovered in the passivated Co NPs five decades ago [14]. In addition to the FM/AFM systems, the EB may be observed in the samples involving the ferrimagnetic (FI/FI, FM/FI) [15] or spin-glass (SG) [16] systems. Although extensive research has been conducted on this subject both experimentally and theoretically by virtue of its potential for applications in fields such as ultrahigh-density magnetic recording [17] and spin-valve/tunnel junction sensor [18], the EB phenomena in the FM/AFM systems consisting of powders or other types of bulk materials have received relatively little attention mainly because of the lack of a well-defined surface between the different phases [19]. Therefore from either scientific research or application point of view bulk materials exhibiting the EB properties are of intense interest. For further background on the EB effects, we refer to the recent review by Nogués et al. [20].

Since the ambient temperature of the magnetic recording devices increases as their heads are reading or writing, the thermal stability of EB is of concern for head design and governs the choice of recording media [21]. In the EB systems, there is an important blocking temperature (T_B), at which the EB vanishes, viz., the superparamagnetic behavior indeed occurs. Single domain particles are subject to the thermal activation leading to a magnetic transition over an energy barrier ΔE [22]. The characteristic time to overcome the energy barrier is usually estimated in the framework of the Arrhenius–Néel statistical switching model:

$$\frac{1}{\tau} = \upsilon_0 \exp(-\Delta E/k_B T), \tag{1}$$

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where T denotes the absolute temperature, and k_B denotes the Boltzmann constant. The energy barrier ΔE results usually from the magnetic lattice anisotropy and the magnetic dipole coupling. $\upsilon_0 = 10^9 - 10^{12}\,s^{-1}$ is the attempt frequency to overcome the energy barrier. In 1972, Fulcomer and Charap employed a "superparamagnetic" model to determine the effect of the thermal fluctuations on the EB in the polycrystalline FM/AFM bilayers, in which the AFM layer is approximated by an assembly of grains without the inter-grain exchange coupling through the thickness t_{AF} [23]. Typically, T_B is smaller than the Néel temperature (T_N) of the antiferromagnet, and it is dependent on the thickness of t_{AF} of the AFM layer as [24–27]

$$\frac{T_N(\infty)-T_B(t_{AF})}{T_N(\infty)} = \left(\frac{\xi_0}{t_{AF}}\right)^{\delta}, \tag{2}$$

where ξ_0 denotes the zero-temperature magnetic correlation length, and δ denotes the so-called "shift exponent". Up to now, a plentiful literatures have discussed the dependence of T_B on the AFM phase of systems. Unfortunately, most of the works focus on the FM/AFM layered systems, the research on T_B in the heterogeneous systems without clear surfaces is lacking.

In experiments, the research on the effect of ratio varies of FM to AFM phase of systems on T_B has been engaged in the various metal alloys or oxides with high geometrical frustration [9,13,19,28], and many new phenomena have been observed. However, a definite relation between the blocking temperature and the phase content in such systems has not been obtained directly. In this paper, we employ a frustrated "alloy-like" FM/AFM model to investigate the effect of ratio varies of ferromagnet to antiferromagnet on the blocking temperature T_B, and analyze the reason of this relation based on the thermal stability of the antiferromagnet. In Section 2, we adopt a classical Heisenberg model to describe the "alloy-like" $(FM)_x(AFM)_{1-x}$ heterogeneous system and introduce our simulation method, then present the simulation results and discussion in Section 3. The summary is given in Section 4.

2. Model and Monte Carlo method

A classical Heisenberg model with a hexagonal closed-packed (hcp) lattice is considered. The three-dimensional hcp lattice itself presents frustration due to the geometrical topology, furthermore, it is one of the commonest and most important crystalline structures in the metals and compounds [29]. For the sake of simulating the different ratios of FM to AFM phase, a fraction x of randomly selected sites was FM, while the residual sites, $1-x$, are set as AFM phase, forming an $(FM)_x(AFM)_{1-x}$ structure. The Hamiltonian in an external field is given by

$$\begin{aligned}\mathcal{H} = &-J_{FM}\sum_{\langle i,j\in FM\rangle}\mathbf{S}_i\cdot\mathbf{S}_j - \sum_{i\in FM}K_{FM}(\mathbf{S}_i\cdot\hat{\mathbf{e}}_i)^2\\ &-J_{AF}\sum_{\langle i,j\in AFM\rangle}\mathbf{S}_i\cdot\mathbf{S}_j - \sum_{i\in AFM}K_{AF}(\mathbf{S}_i\cdot\hat{\mathbf{e}}_i)^2\\ &-J_{IF}\sum_{\langle i\in FM, j\in AFM\rangle}\mathbf{S}_i\cdot\mathbf{S}_j - \mathbf{H}\cdot\sum_i\mathbf{S}_i,\end{aligned} \tag{3}$$

where $\mathbf{S}_i$ denotes the classical spin and $\hat{\mathbf{e}}_i$ denotes the unit vector in the direction of the easy axis of spin at site i. The angular brackets in the sums denote a summation over the nearest neighbors only. The first term gives the exchange interaction between the spins in the FM phase; J_{FM} denotes the FM exchange coupling constant. The second term gives the anisotropy energy of the FM phase; K_{FM} denotes the uniaxial anisotropy constant of spins in the FM phase. The third term gives the exchange interaction between the spins in the AFM phase; J_{AF} denotes the AFM exchange coupling constant. The fourth term gives the anisotropy energy of the AFM phase; K_{AF} denotes the uniaxial anisotropy constant of spins in the AFM phase. The fifth term gives the exchange interaction between the spins at the FM/AFM interface; J_{IF} denotes the FM/AFM interfacial exchange coupling constant. The last term gives the Zeeman energy, and $\mathbf{H}$ is supposed to be along the z-axis. We take the anisotropy axes of the interior of the FM and AFM phases along the z-axis. However, in experiments it is usually observed that the state of nonuniform distribution of spin direction at the FM/AFM interface exists [30]. Additionally, Malozemoff has presented that only using the random-field model at the rough FM/AFM interface can fit the experimental results [31]. Therefore, randomly oriented anisotropy axes for the spins at the FM/AFM interface are selected in the present paper.

We set $J_{FM}=1.0$, $K_{FM}=0.1$, $J_{AF}=-J_{FM}/2=-0.5$, $K_{AF}=10.0$, and $J_{IF}=J_{FM}/2=0.5$. H is given in units of $J_{FM}/g\mu_B$, T in units of J_{FM}/k_B, and K_{FM}, K_{AF} in units of J_{FM}.

Because the bulk material will be studied in the paper, while simulations are only performed on the finite systems. Therefore, in order to study the magnetic properties of the materials, we use the periodic boundary condition to eliminate the boundary effects [32]. In order to compute the energy barriers, we introduce a modified MC Metropolis method. It is well-known that the reversal of a single moment can be described by using a Stoner–Wohlfarth model [33]. When the moment rotates from one state to another, it must jump over the energy barriers between the two states. Hence in our simulation, we not only consider the thermal fluctuation of spins but also calculate the energy of each spin with respect to its polar and azimuthal angles, and judge whether there are energy barriers between the new and initial states, thus, determine the flipping probability [34,35].

For the sake of obtaining the information of T_B and distribution of energy barriers, the zero-field-cooled (ZFC) and field-cooled (FC) magnetizations will be studied, and the protocol to simulate the magnetizing process is as follows. The ZFC magnetization curve is obtained by cooling in the zero field from a high temperature, at which the AFM spins show the superparamagnetic behavior, to a low temperature and measuring the magnetization at stepwise increasing temperatures in an external field. The FC magnetization curve is obtained by measuring at stepwise decreasing temperatures in the same magnetic field. In the present paper, the ZFC and FC magnetization curves are both measured along the z-axis, and H is set by 1.0. We find that the Curie temperature (T_C) as $x=1$ and the Néel temperature (T_N) as $x=0$ of the present model are approximately 3.25 and 0.72, respectively. During the simulation, 20 000 MC steps per spin are used; the thermal average of the magnetization is performed with 10 000 MC steps after the initial 10 000 MC steps, which were discarded for the thermalization.

3. Results and discussion

For the purpose of elucidating the relation between x and T_B in the $(FM)_x(AFM)_{1-x}$ heterogeneous systems in detail, we calculate the magnetizations of systems with the x ranging from 0.02 to 0.98 in steps of $\Delta x=0.02$. The FC and ZFC magnetization curves and their differences of some typical x values are given in Fig. 1. It is shown that both values of FC and ZFC magnetizations increase with the increase in x at high temperature region, but at low temperature, this monotonous behavior is no longer valid for the ZFC magnetization behavior. In experiments, the superparamagnetic blocking temperature (T_B^{SP}) is determined by observing the temperature point, at which FC and ZFC magnetization curves are splitting [36]. As shown in Fig. 1(a), the splitting points are