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Dependence of exchange bias on core/shell relative dimension in ferromagnetic/antiferromagnetic nanoparticles

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ABSTRACT

We employ a modified Metropolis Monte Carlo simulation to study the effect of bimagnetic core/shell relative dimension on exchange bias in ferromagnetic/antiferromagnetic nanoparticles. The exchange bias field is inversely proportional to the ferromagnetic shell thickness in the antiferromagnetic (core)/ferromagnetic (shell) nanoparticles, while in the nanoparticles with an opposite core/shell structure the exchange bias behavior is complex and distinguished in different ranges of the ferromagnetic core radius. The work elucidates unambiguously how the core and shell dimensions optimize the exchange bias in nanoparticles.

1. Introduction

Recently, new advanced preparation techniques offered an opportunity to fabricate several-nanometer magnetic particles with narrow size distributions and different compositions through a variety of procedures, either embedded in a solid matrix or diluted in organic solvents [1]. Nanoparticles could display interesting new phenomena that can be associated either with intrinsic characteristics of the individual particle such as finite-size and surface effects or with collective properties of the nanoparticle ensemble such as dipolar interactions and exchange mediated couplings [2–5]. On the other hand, a possibility that core/shell magnetic nanoparticles are used as storage media for magnetic recording is appealing due to their substantially improved thermal stabilities. This fact was stimulated from a report by Skumryev et al. [6] showing a principle that a superparamagnetism limit could be beaten by means of an exchange bias (EB) effect, which raised prospective hopes to further miniaturize storage devices. An indication of EB is a shift of hysteresis loop along the field axis after field cooling or depositing in the materials composed of ferromagnetic (FM)/antiferromagnetic (AF) interfaces [7–10], termed as EB field (H_E). As mentioned earlier, to meet the requirement of ultra-high density recording, the size of a spintronic device is continuously reduced, and thus the role of the size in the EB properties becomes significant and needs to be elucidated further.

A large number of experimental and theoretical studies have been conducted on the core/shell size dependences of EB in FM/AF nanoparticles, including commonly FM (core)/AF (shell) [11–22] and inverted structures [23–25]. As compared to the planar systems where interfaces are fully compensated or non-compensated due to the nature of interfacial coupling or the roughness caused by defects, the interfaces in the core/shell nanoparticles incorporate naturally the roughness (i.e., the noncompensation) arising from the surface effects associated with the finite sizes of the particular geometry. For instance, in naturally oxidized Fe or Co nanoparticles, a critical diameter, below which the surface spin-glass state and EB both disappeared abruptly, existed due to an increasing surface-to-volume ratio with reduced sizes [11,12]. Also in the oxidized Co nanoparticles, a nonmonotonic behavior of the EB versus CoO shell thickness was obtained through carefully controlling the oxidation process, and the maximum value of H_E just corresponded to the core and shell that have similar dimensions [15]. Similarly, as a powerful tool, computer simulations also calculated the EB properties in nanoparticles and found that H_E might either decrease monotonously or oscillatory with increasing particle sizes [19,21]. Distinct phenomena observed indicate that it is still a challenge to establish a universal dependence

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of EB on the core/shell dimensions, since many extrinsic factors in the nanogranular systems (e.g., domain formation, interfacial roughness, impurities, grain boundaries, and collective effects) could influence the EB properties to differing extents in nontrivial and somewhat uncontrollable ways [15,18]. Moreover, many real core/shell nanoparticles are prepared through growing the shell at the expense of the core, that is, the core and shell cannot be varied independently [8,14,15,23,24].

In this paper, two opposite core/shell structures of the nanoparticles with a constant radius are proposed to study the effect of an increasing core accompanying a decreasing shell, called core/shell relative dimension, on the EB properties in FM/AF nanoparticles by performing a modified Metropolis Monte Carlo simulation. Different trends are obtained in two structures, and the behaviors of the magnetizations at the core/shell interfaces and of the coercive fields along the decreasing and increasing branches of hysteresis loops as well as the images of microscopic spin configurations are given to interpret the numerical findings. In the next section, the models of nanoparticles and the simulation method are described, respectively. In Section 3, we present the simulation results meanwhile discuss them. A conclusion is drawn in the final section.

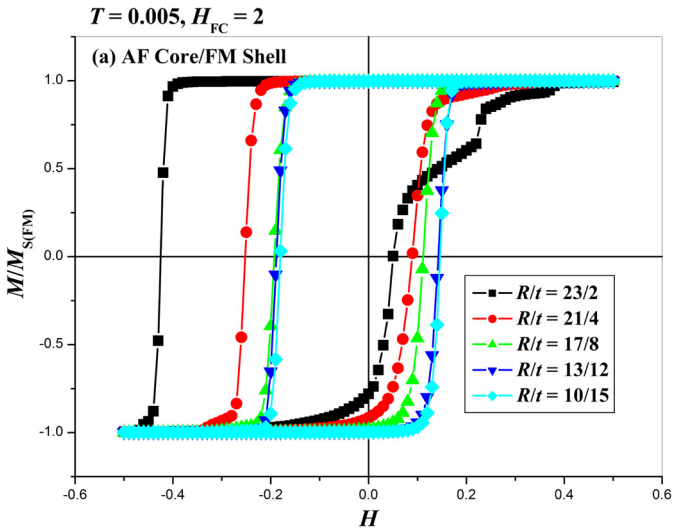

Fig. 1. Ferromagnetic hysteresis loops in the nanoparticles with opposite core/shell structures and representative core/shell relative dimensions at low temperature after field cooling, where $M_{S(FM)}$ is a saturated value of ferromagnetic magnetization.

2. Model and Monte Carlo simulation

At an atomic level, the core/shell magnetic nanoparticles could be modeled using a simple cubic lattice of the classical Heisenberg spins with unit vectors. Taking a lattice constant as 1, we select the nanoparticle model with a constant radius of $R_0 = 25$, composed of a core radius (R) and a shell thickness (t). Therefore, in the simulation, R increases while t decreases, and vice versa.

Open boundary conditions are used to the outermost layer of the shell, resulting in 65 267 spins in the model. Each of the spins experiences a uniaxial anisotropy. Meanwhile, it couples to its nearest neighbors with Heisenberg exchange interactions. Under an external magnetic field (H), the Hamiltonian is expressed simply as

$$\mathcal{H} = -K\sum_i \left(S_i^z\right)^2 - J\sum_{\langle ij\rangle} S_i \cdot S_j - H\sum_i S_i, \tag{1}$$

where $S_{i(j)}$ represents an atom spin at site $i(j)$. Core/shell single-site anisotropy energies with an easy axis along the axis z are first given in Eq. (1). To obtain a well hysteresis loop, $K = K_{FM} = 0.01$ is suggested, if the spins belong to the FM component. Elsewhere, $K = K_{AF} = 2$ is set for stabilizations of the AF component to induce a pronounced EB effect in the FM component via core/shell interfaces. Next, exchange energies between the spins in the core, shell, and interfaces, commonly related to the Curie points of materials, are exhibited, where $J = J_{FM} = 1$ in the FM component (meanwhile, serving as a unit energy) while $J = J_{AF} = -1/2$ in the AF component are due to the fact that Curie temperatures in the ferromagnets are higher than Néel temperatures of their AF oxides in general. For maximizing the interfacial effects, highly crystalline and oriented core/shell interfaces are considered with $J = J_{IF} = 1$ [15,26]. Finally, a Zeeman energy is added, and H is applied along the axis z.

To update spins, a modified Metropolis Monte Carlo simulation, which exactly calculates the spin energies with respect to polar and azimuthal angles to look for real energy barriers, is employed. Detailed description has been published in the previous works [27–31] to which the reader could be referred. 5×10^3 Monte Carlo steps per spin are used to average magnetic quantities after the other 5×10^3 Monte Carlo steps per spin performed for thermalizations. The simulation starts at a high temperature ($T = 1.0 > T_N$), and the nanoparticle model has a random configuration. With decreasing temperature in a step of $\Delta T = 0.005$, the configuration is being optimized under an external magnetic field ($H_{FC} = 2$). At the desired temperature ($T = 0.005$), hysteresis loops are recorded through cycling H between 0.5 and -0.5 to study the EB properties.

3. Results and discussion

Firstly, Fig. 1 gives the FM hysteresis loops in the nanoparticles with opposite core/shell structures and representative core/shell relative dimensions. For the AF (core)/FM (shell) nanoparticles, the FM coercive field at the decreasing branch decreases rapidly for an initial decrease of R/t, while a gradual increase of the coercive field is obtained on the other side with decreasing R/t. Finally, they both level out for small values of R/t. Moreover, a shoulder magnetization behavior could be observed after the magnetization reversal at the increasing branch in the nanoparticles with the largest value of R/t ($= 23/2$). In the FM (core)/AF (shell) nanoparticles, the FM hysteresis loops cannot be closed for small values of R/t due to the large J_{IF}, which causes a failure to extract H_E. Hence, the results are excluded. When R/t is above 10/5, shift directions of the FM hysteresis loops are not estimated until a R/t value of 16/9. The loops gradually move towards the positive field direction with increasing R/t from 16/9 to 22/3, and the coercive field at the decreasing branch is more sensitive to R/t than that at the increasing branch. A further increase in R/t could result in an unexpected shift of the loop again. To exhibit systematically the EB properties in the core/shell nanoparticles, H_E is calculated through $H_E = (H_{LEFT} + H_{RIGHT})/2$, where H_{LEFT} and H_{RIGHT} are the FM coercive fields at the decreasing and increasing branches, and presented in Fig. 2. As shown in the insert of Fig. 2(a), H_E is inversely proportional to the FM shell thickness (t_{FM}) in the AF (core)/FM (shell) nanoparticles, in consistence with experimental observations and theoretical predictions in the granular [32] and planar systems [33–38]. The EB properties become complex in the FM (core)/AF (shell) nanoparticles. Only in the intermediate range of the FM core radius ($R_{FM} \sim 16$–22), H_E roughly linearly decreases with increasing R_{FM}; otherwise, H_E exhibits an oscillatory behavior.

Fig. 3 depicts the AF magnetization behaviors at the interfaces and the FM coercive field behaviors to interpret the EB phenomena observed in the AF (core)/FM (shell) nanoparticles. After field cooling, the AF magnetization at the interfaces is saturated due to