Effects of dipolar interactions on magnetic properties of Co nanowire arrays*

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Magnetic properties and magnetization processes of Co nanowire arrays with various packing densities are investigated by means of object-oriented micromagnetic framework (OOMMF) software package with finite difference micromagnetic simulations. The packing density of nanowires is changed with the diameter, number of nanowires and center-to-center spacing between the wires. The magnetization reversal mechanism and squareness of the hysteresis loops of the nanowire arrays are very sensitive to the packing density of nanowires. Clear steps and plateaux on the demagnetization are visible, which turns out that dipolar interactions among the wires have a significant influence on switching field.

Keywords: micromagnetic simulations, cobalt nanowires, magnetization reversal mechanism, magnetic properties

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1. Introduction

The magnetic properties of the 3d metal and alloy magnetic nanowire array have been extensively investigated because of their potential applications in ultrahigh-density magnetic recording media and future permanent magnets.[1–8] Kittel first obtained the high coercivity in cobalt particle resulting from shape anisotropy through calculations.[9] Nowadays, advances in fabrication techniques, such as polyol method, have allowed the preparation of nanostructured systems with very interesting physical properties. Pousthomis *et al.* prepared Co nanorods with mean diameters ranging from 7–25 nm and lengths ranging from 30–300 nm by the polyol method, and a high coercivity of 7.2 kOe was obtained for Co nanorods with a mean diameter of 9 nm.[10] Anagnostopoulou *et al.* prepared Co rods and assembled them from wet suspensions under a magnetic field, and the magnets prepared with the rods assemblies exhibited energy product, $(BH)_{max}$ exceeding 18.85 MGOe.[11] However, the theoretically predicted upper limit of the coercivity and the maximum energy product of single Co nanowire are 16.6 kOe $(1/2\mu_0 M_s + H_a)$ and 81 MGOe, respectively.[12] There is still a potential to obtain larger coercivity and energy product for Co nanowires. Further theoretical studies on magnetic properties of Co nanowires have been carried out. Westphalen *et al.* have studied the magnetization reversal in nanowires with a spiral shape and found that the magnetization reversal is rather inhomogeneous.[13] Vila *et al.* have studied the magnetic vortex in nanowire with transverse easy axis.[14] Ott *et al.*[15] and Zighem and Mercone[16] have independently studied the effect of the shape of elongated magnetic particles on the coercivity.[15,16] Note that the effective dipolar interactions among the nanowires have a strong influence on the macroscopic magnetic behavior of the system.[17] Nevertheless, the details of the magnetization processes occurring in arrays of closely packed magnetic nanowires are still unknown.

In this paper, the effects of dipolar interaction on the coercivity and energy product of the Co nanowire assemblies with the micromagnetic simulations and the mechanism of the magnetization reversal process of nanowires array are investigated.

2. Methods

Systematically, the effects of the dipolar interaction on magnetic properties and magnetization processes in array of Co nanowires are investigated using OOMMF micromagnetic simulations.[18–20] These simulations are performed with an algorithm-based finite difference method. Equilibrium structures of the magnetization are characterized by local energy minima. The total energy E_{tot} of a given arrangement of the magnetization in a ferromagnetic sample reads

$$E_{tot} = E_{an} + E_{exc} + E_{dem} + E_{Zee}, \tag{1}$$

where the terms on the right-hand side denote the anisotropy energy, the exchange energy, the stray field energy, and the Zeeman energy, respectively.[21] These contributions are usually sufficient for micromagnetic simulations. For given material parameters and the external field $\boldsymbol{H}$, the total energy of

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the sample can be written as

$$E = A(r)\left(\frac{\nabla \boldsymbol{M}}{M_{\rm s}}\right)^2 - K(r)\frac{(\boldsymbol{M}\cdot\boldsymbol{n})}{M_{\rm s}^2} - \mu_0 \boldsymbol{M}\cdot\boldsymbol{H} - \frac{1}{2}\mu_0 \boldsymbol{H}_{\rm d}(r)\cdot\boldsymbol{M}, \quad (2)$$

where A and K are the exchange and crystalline anisotropy constants, and $\boldsymbol{H}$ and $\boldsymbol{H}_{\rm d}(r)$ are the applied field and magnetostatic self-interaction fields, respectively. The standard demagnetization energy term is built on the assumption that the magnetization is constant in each cell. Thus, the demagnetization field (stray field) can be defined as

$$\boldsymbol{H}_{\rm d} = -N\boldsymbol{M}, \quad (3)$$

where N is the demagnetizing factor in the direction of the magnetization.[22] In the experiment, each wire is a single crystal with the c-axis (002) along the long axis of the wire.[12] Thus, the external field is applied along the long axis of the wires and the closed hysteresis loops are simulated between the external fields $+20$ kOe to -20 kOe in steps of 0.1 kOe. The sample is discretized into cube elements of regular size with respect to the wire diameter and length.[21,23–25] According to the previous report on hard/soft multilayers and skyrmions,[26–31] the cell size of sample should be smaller than the crystalline exchange length ($L_{\rm mc} = \sqrt{A/K}$) and the magnetostatic exchange length ($L_{\rm me} = \sqrt{2A/(\mu_0 M_{\rm s}^2)}$) as well. In cobalt, the $L_{\rm mc}$ and $L_{\rm me}$ are 6.08 nm and 4.73 nm, respectively. To obtain the accurate results and appropriate calculated time, the unit cell sizes of single wire and arrays are $1\times1\times2$ nm^3 and $2\times2\times4$ nm^3, respectively. In this simulation process, the cylindrical Co nanowires are set to be 200 nm long with the maximal number of 16 and the distance spacing is varied up to 60 nm. Moreover, the saturation magnetization and exchange stiffness are very close to those of corresponding bulk materials in nanowire array when the diameter increases up to 10 nm. The material parameters are as follows:[14,19] saturation magnetization $M_{\rm s} = 17.86$ kGs, crystalline anisotropy constant $K_1 = 0.54\times10^7$ erg·cm^{-3}, and exchange constant $A = 20\times10^{-7}$ erg·cm^{-1}. In this paper, higher order terms of the magnetocrystalline anisotropy are ignored for simplicity, which makes the squareness of each hysteresis loop rectangular. However, the shape of hysteresis loop and the coercivity may change if the fourth order term is taken into account in this calculation.[32] When the ratio of the fourth order term to the second one in the magnetocrystalline anisotropy coefficient, λ, is smaller than -2, the shape of the hysteresis loop would change while the coercivity will keep constant. When $-2 \le \lambda \le 0.25$, both the shape and coercivity of the hysteresis loop keep constant. When λ is larger than 0.25, the shape of the hysteresis loop will change and the coercivity will increase slightly. The ratio λ is about $1/3$ for cobalt, which will result in a 1.4% increase of the coercivity in this calculation. More details and discussion for the effect of higher order terms of the magnetocrystalline anisotropy can be found in Ref. [32].

3. Results and discussion

Figure 1 shows the experimental and simulated hysteresis loops with the field applied parallel to the long axis for Co nanowires. The experimental aligned hysteresis loop of Co nanowires assembly is obtained via a solvothermal chemical process, providing an ultrahigh coercivity of 10.3 kOe at room temperature with a mean diameter of 15 nm and a mean length of 200 nm, which is the highest value ever reported so far for Co nanowires.[12] Before measurement, the Co nanowires had been prepared by sonicating the toluene dispersion then mixed with epoxy. This composite is then incorporated into a mold aligned under an external magnetic field of 20 kOe.[12] In order to compare the computed results with the experimental results and to understand the effects of the dipolar interaction on magnetic properties and magnetization processes in array of Co nanowires, the magnetic properties and magnetization processes of single Co nanowire and a 16 nanowires array are investigated by means of OOMMF micromagnetic simulations. The center-to-center spacing of the simulated Co nanowire array is 60 nm with nanowire diameter $D = 15$ nm, wire length $L = 200$ nm. Figure 1 shows that the magnetic properties of 16-nanowire array matched better with the experimental value than that of single wire. The simulated value of the coercivity of the array is 10.4 kOe, which is in good agreement with the obtained experimental value,[12] whereas the coercivity of a single nanowire is 11.08 kOe. Significantly, the corresponding coercivity is increased by 6.5% compared with that of the array. In addition, the hysteresis loop of the single nanowire exhibits a typical square loop whereas the hysteresis loop of the array exhibits a little worse squareness. The difference in coercivity and squareness of the hysteresis loops between

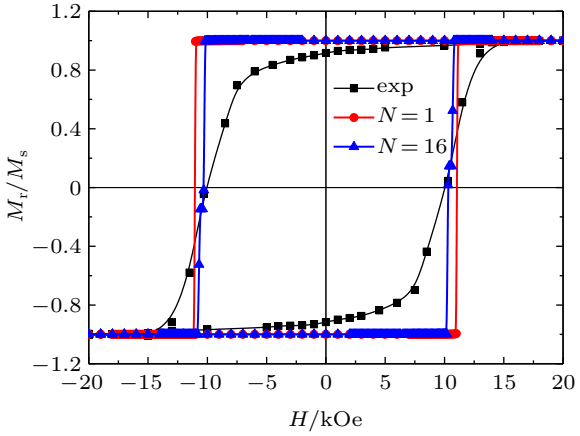


Fig. 1. (color online) Experimental[12] and simulated hysteresis loops with the field applied parallel to the long axis for Co nanowires assemblies. The red line with circles represents the hysteresis loop of one Co nanowire for comparison.

single Co nanowire and the array demonstrates that the dipolar interactions between the nanowires cannot be ignored. However, the remanence is not well matched, owing to the less orientation and uniformity morphology of the magnetic measurement sample.[10]

To further understand the dipolar interactions in Co nanowires array, a systematic simulation for different numbers of nanowires and various center-to-center interspaces between the wires is analyzed. To compare with the experimental result, nanowire diameter is set to be a constant value $D = 15$ nm. The magnetic measurement sample consists of a large array of magnetic nanowires, however, the maximal size of the simulated array is not sufficient to expect an exact reproduction of the experimental value. Modeling an array with macroscopic dimensions is impracticable compared with experimental value due to the long computational time of calculation. In order to account for the interaction between the neighboring wires in the simulation, the hysteresis loops with up to 16 interacting nanowires are simulated, which is the largest set used in the calculation.[21] Figure 2(a) shows the coercivity decreases with increasing the number of interacting wires. The simulated coercivity approaches well to the experimental value, as the number of interacting wires increases up to 16 and center-to-center spacing between the wires is 60 nm. The simulations clearly show that the dipolar interaction is

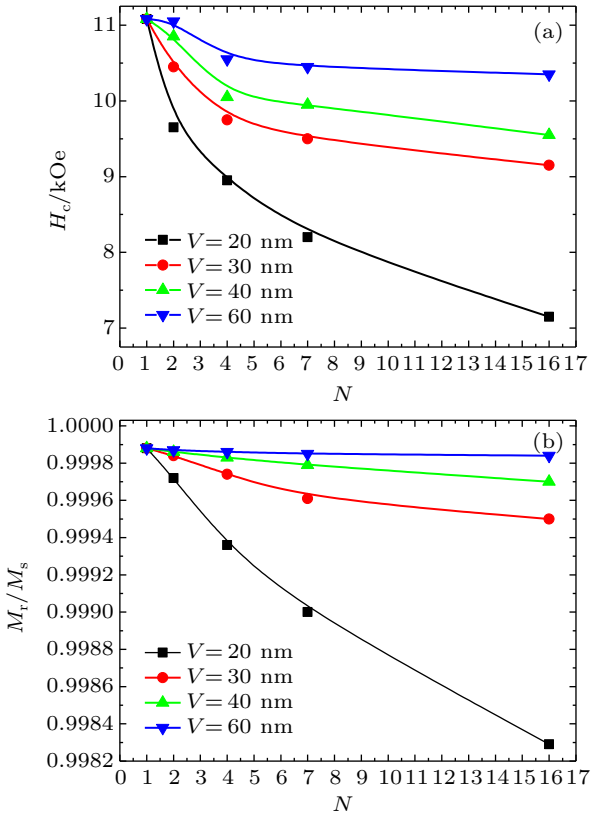


Fig. 2. (color online) Plots of coercivity (a) and the remanence ratio (b) of arrays versus the number of nanowires (N) for different center-to-center spacing between the wires (V), with the diameter of nanowires (D) being 15 nm.

crucial for the coercivity of an array of magnetic nanowires. For the same number of nanowires, coercivity decreases with the decrease of the center-to-center spacing between the wires. Especially for the nanowire spacing equal to 20 nm, the close neighbor nanowires are almost connected with each other, making the coercivity decline obviously. The remanence ratio of arrays of various numbers of nanowires to center-to-center spacing between the wires is shown in Fig. 2(b). However, the effect is not significant as indicated by the magnetization loops with remanence ratio close to 1, obtained for all nanowire arrays.

It is already well known that the dipolar interactions affect considerably the macroscopic magnetic behavior of array of nanowires. High packing fraction for nanowires assemblies is an important factor for obtaining a high energy product. However, high packing fraction for nanowire assembly results in lower coercivity. Thus, the optimal packing density is very important for obtaining high magnetic energy product and coercivity. For this purpose, Co nanowire arrays with various packing densities are simulated and the numbers of nanowires ($N = 16$) in all nanowire arrays are also fixed. The coercivity increases with the decrease of diameter and the increase of aspect ratio, which is in good agreement with the enhanced shape anisotropy.[15] For fixed diameter of nanowires, coercivity decreases with the increase of the packing density. The ideal $(BH)_{max}$ of 46.43 MGOe is obtained for an array by combining a high packing density ($P = 0.628$) and a small diameter ($D = 10$ nm) as shown in Fig. 3. The performance of Co nanowires makes them good candidates in the new rare-earth free permanent magnets. The large discrepancy in coercivity between the theoretical calculations and the experimental measurements might result from the dipole interaction among the nanowires, incomplete alignment of the nanowires, and structural imperfections and surface defects of the nanowires.[10,33]

Obviously, the higher the packing density, the stronger the magnetostatic interaction among the Co nanowires will be. Increasing the distance of center-to-center spacing between the wires from 20 to 60 nm while keeping the diameter of the wires at 15 nm and number of nanowires at 16, the remanence ratio increases up to nearly 1 and the coercivity rises from 7.2 kOe up to 10.4 kOe as shown in Fig. 4. The packing density decreases with the decreasing of the center-to-center spacing between the wires, resulting in a reduction in macroscopic interactions among the nanomagnets and an increase of the switching field of the individual nanowire. These trends are in good agreement with the result reported by Hertel.[21] Moreover, an interesting phenomenon is found that the clear steps and plateaux on the demagnetization curve become more visible as the distance of center-to-center spacing between the wires decreases. The complexity in studying the phenomenon of steps and plateaux is that the magnetic field resulting from

the dipolar interaction depends on the magnetization state of each nanowire, which, in turn, is influenced by the effective field of neighboring nanowires.[34] This might be due to the increase of interactions among the nanowires in the array and the enhancement of the switching field of the individual nanowire.[17]

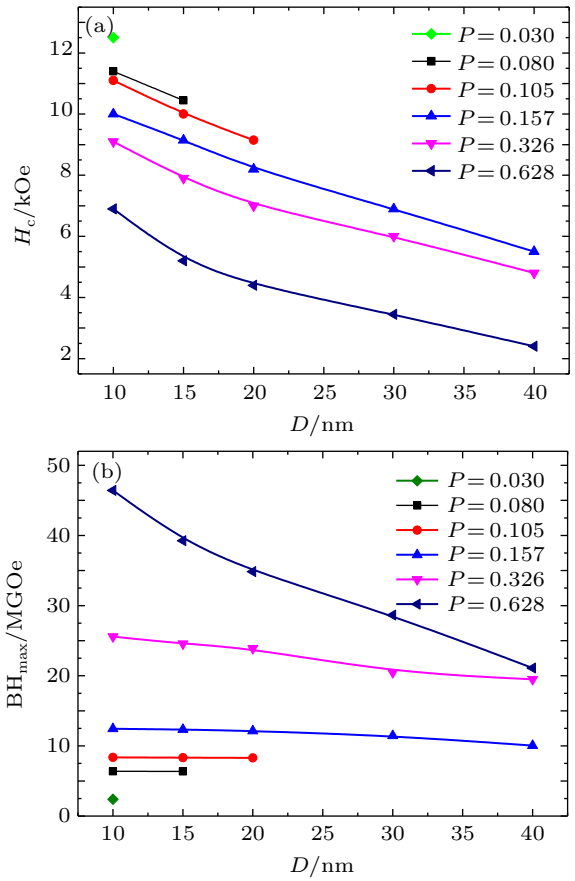


Fig. 3. (color online) (a) Coercivity and (b) magnetic maximum energy product of nanowires array with different values of diameter (D) and packing density (P), the numbers of nanowires (N) is 16.

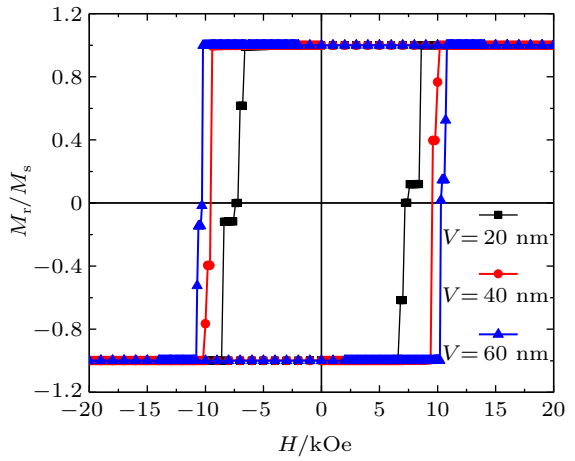


Fig. 4. (color online) Hysteresis loops of arrays with different center-to-center spacing between the wires. The numbers of nanowires (N) is 16 and the diameter of nanowires (D) is 15 nm.

We investigate magnetization configuration of one nanowire array ($D = 15$ nm, $V = 40$ nm) as shown in Fig. 5. The magnetization configuration of the Co nanowires array near the coercivity can give an insight into the magnetization reversal. Since the nucleation starts at the end of the wire and the rest of the wire remains mostly homogeneously magnetized, we only plot the magnetization state in the region of the top and bottom of Co nanowires array (Fig. 5(a)) and magnified view on the top of one nanowire (Fig. 5(b)). An applied field of -9.5 kOe results in most of the magnetic moments for nanowires being switched in the direction of the applied field. However, this field is not strong enough to switch a single wire (-11.08 kOe). The reversal of some wires occurs because the stray field of neighboring wires is added to the external field and thus leads to a higher field to switch the magnetic moment.[34] From the magnified view on the top of one nanowire, the magnetic moments start to rotate out from the easy axis at the ends of the nanowires, implying that the nucleation mode for the nanowires with $D = 15$ nm deviates slightly from homogeneous rotation, and can be observed. In this connection, the dipolar interaction from neighboring sites can trigger early nucleation events, leading to significant changes of magnetic properties. Hence, the reversal mode could play a major role in the study of dynamical properties and the reversal mode of the demagnetization process might be much more complex in experiment.

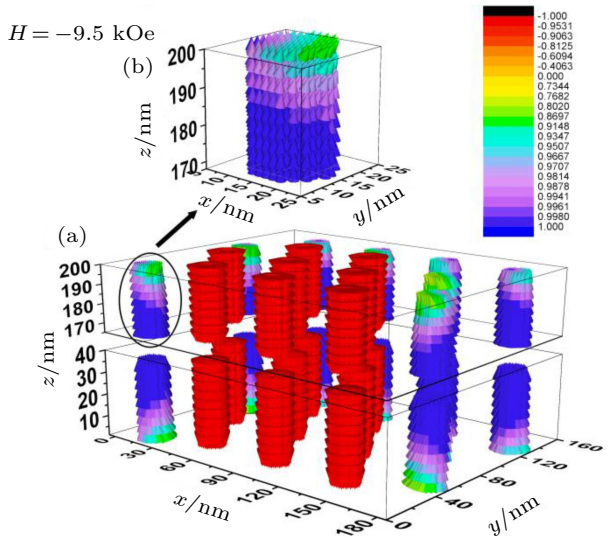


Fig. 5. (color online) (a) Magnetization states at the top and bottom of array for Co nanowires ($D = 15$ nm, $V = 40$ nm) in a magnetic field of $H = -9.5$ kOe. (b) Magnified view on the top of one nanowire. The colors represent the z component of the magnetization.

4. Conclusions

The magnetic properties and magnetization processes of the Co nanowires array with various packing densities are investigated by means of micromagnetic simulation. The magnetization reversal mechanism and coercivity of the nanowires are very sensitive to the packing density of nanowires. The effects of dipolar interaction on the coercivity and the energy

product of nanowires array are enhanced with the increasing of packing density. The magnetization reversal mechanism plays a major role in squareness of the hysteresis loops in nanowires array. The results can be explained considering the dipolar field created by the neighboring nanowires. It is clearly shown that this field is responsible for the change of the reversal field of the wires, leading to the occurrence of plateaux in the demagnetization process.

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