Magnetization reversal process and magnetic relaxation of self-assembled Fe$_3$Pt nanowire arrays with different diameters: Experiment and micromagnetic simulations

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Magnetization reversal process and magnetic relaxation of Fe$_3$Pt nanowire arrays fabricated by electrodeposition have been investigated by means of micromagnetic simulation and magnetization measurements. Time dependence of magnetization reveals that the nanowire arrays exhibit a strong magnetic viscosity effect. The magnetic viscosity coefficients $S$, which are dependent on temperature and magnetic field, achieve maximum values near the coercivity. The activation volume $V^*$, defined as the effective volume involved in the magnetization reversal process, increases with temperature as $T^{1/2}$ and linearly with the diameter, which is consistent with the theoretical prediction of the thermally activated magnetization reversal process.

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I. INTRODUCTION

Magnetic properties of magnetic nanowires arrays have been extensively investigated because of their potential applications in ultrahigh-density magnetic recording media as well as their significance in fundamental research. Owing to the strong magnetic shape anisotropy, magnetic nanowire arrays exhibit both high coercive fields and squareness ratio with the easy magnetization direction along the wire axes. The fundamental issues include the shape anisotropy, the dipolar interaction between the nanowires, and size effect on magnetic properties. Among them, the understanding of the magnetization reversal process for magnetic nanowires is of primary importance, and consequently, the emphasis has been put on the magnetization reversal mechanism and related magnetic behavior. Several experimental techniques, such as micro-superconducting quantum interference device (SQUID) magnetometry, ferromagnetic resonance, and magnetic force microscopy have been employed to investigate the magnetization reversal process of nanoparticles and nanowires. The results showed that the magnetization reversal of particles could be described by the classical Néel-Brown model for rotation in unison. However, as far as the magnetic wires are concerned, the magnetization reversal appears to proceed with inhomogeneous reversal mode, such as curling or nucleation followed by the propagation of the domain wall.

It is well known that the thermal stability is very critical to the high-density magnetic recording media with areal density of Tbits/in$^2$. Therefore, in addition to the static magnetic properties, the potential application of the nanowire arrays also depends on the hysteresis properties and dynamical behavior concerning the thermally activated magnetization reversal process and magnetic relaxation. Although such experiments have been performed on bulk materials and magnetic thin films, the detailed magnetic viscosity behavior for nanowire arrays has rarely been reported.

In our previous work, shape anisotropy and temperature dependence on the magnetic properties with thermally activated reversal mechanism of the Fe$_3$Pt nanowire arrays have been investigated. In this work, we simulated the magnetization reversal process of a hexagonal cell of 4×4 array for Fe$_3$Pt nanowires with $d$=10 nm. The simulated coercivity is in good agreement with the experimental one. Furthermore, time dependence of magnetization indicates that the nanowire arrays exhibit a strong magnetic viscosity effect, which was discussed in terms of the Stoner-Wohlfarth model.

II. EXPERIMENT

The Fe$_3$Pt nanowire arrays were fabricated by ac deposition into the anodic aluminum oxide (AAO) templates with different diameters. The AAO templates were prepared by a two-step anodization process controlled by the anodization voltage and electrolyte such as sulfuric acid and oxalic acid. Transmission electron microscopy (TEM) indicates that the nanowire diameters vary from about 8 to 60 nm. Fe$_3$Pt nanowires were electrodeposited into self-assembled nanoporous templates by ac deposition in the electrolytes consisted of 45 g/l HBO$_3$, 0.03M FeSO$_4$, and 0.01M Pt(NO$_3$)$_2$(NH$_3$)$_2$ with the pH value of 3.0–3.3. The ac deposition process was conducted under 15 V$_{ac}$ with the frequency of 200 Hz for 300 s. The structural characterization of the Fe$_3$Pt nanowire arrays was performed using x-ray diffraction pattern and TEM, which indicate that the Fe$_3$Pt nanowires have fcc structure with the [110] preferred orientation along the nanowire axes.

The magnetic properties of the Fe$_3$Pt nanowire arrays have been investigated by means of SQUID magnetometry with the applied field parallel and perpendicular to the nanowire axes, respectively. It was revealed that the shape magnetic anisotropy plays a dominant role in the overall magnetic anisotropy of the nanowire arrays, driving the easy magnetization axis along the wire axes. The temperature dependence of coercivity has been interpreted by the thermally activated magnetization reversal process with the Stoner-Wohlfarth model. In order to understand the magnetization reversal process at ground state, micromagnetic simulation with the three-dimensional version of the OOMMF code was performed to simulate the hysteresis loop of the Fe$_3$Pt nanowire arrays. In addition, we have investigated the magnetic relaxation of the Fe$_3$Pt nanowire arrays.
The unit cell size is 1.25 nm, the center-to-center spacing between the wires is 40 nm, and interacting wires increase up to 16.23 kOe. In order to take into account the interaction with neighboring wires and, at the same time, use reasonable computational time, the following model and parameters were assumed on the basis of our experimental results.

(1) A hexagonal cell of $4 \times 4$ array of Fe$_3$Pt nanowires was chosen to simulate the magnetization reversal process. The nanowire diameter $d=10$ nm, wire length $L=400$ nm, the center-to-center spacing between the wires is 40 nm, and the unit cell size is $1.25 \times 1.25 \times 10$ nm$^3$.

(2) Although it is well known that the magnetic properties change significantly in nanostructure, the saturation magnetization and exchange stiffness are very close to those of corresponding bulk materials in nanowire arrays when the diameter is up to 10 nm. Therefore, it is reasonable to use approximately the saturation magnetization of $M_s = 1250$ emu/cm$^3$ and the exchange stiffness of $A=2.57 \times 10^{-12}$ A/m for the bulk Fe$_3$Pt.\(^{25}\)

(3) Fe$_3$Pt is known as soft magnetic with easy magnetization axes along the $[100]$ direction, and the nanowire arrays fabricated by electrodeposition are disordered fcc structures with $[110]$ preferred direction along the nanowire axis. The Fe$_3$Pt nanowire arrays exhibit strong uniaxial magnetic anisotropy due to the shape anisotropy, which is much higher than magnetocrystalline anisotropy. From the magnetic hysteresis loops it could be seen that the switching field is much higher than the magnetocrystalline anisotropy field, which is usually several hundreds of oersteds. Although its magnetocrystalline anisotropy constant is not known, the fcc structure with high symmetry for Fe$_3$Pt suggests its magnetocrystalline anisotropy constant is the same order of magnitude as that for Fe. Previous investigation for Fe nanowire arrays indicated that the shape anisotropy constant is 1 order of magnitude larger than that of the magnetocrystalline anisotropy constant $K_1$.\(^{21,25}\) Hence, the magnetocrystalline anisotropy can be neglected reasonably.

The simulated hysteresis loop for the Fe$_3$Pt nanowire arrays is plotted in Fig. 1. For comparison, the hysteresis loop for one single Fe$_3$Pt nanowire was also simulated and plotted in Fig. 1. The coercivity decreases from 3.7 kOe for one single nanowire to 2.8 kOe for $4 \times 4$ array of Fe$_3$Pt nanowires. The simulated value of coercivity is in good agreement with the measured coercivity $H_{C0}$. However, the remanence squareness $S_r$ exists with a relatively large deviation, which is related to the surface magnetic moment of the magnetic moments deviating from the wire axes of the nanowires.

The magnetization configuration of the Fe$_3$Pt nanowire arrays near the coercivity can give insight into the magnetization reversal. Since the nucleation starts at the end of the wire and the rest of the sample remains mostly homogeneously magnetized, we only plot the magnetization state in the region of 40 nm from the bottom of Fe$_3$Pt nanowire arrays [Fig. 2(a)] and a full view of magnetization configuration for one central nanowire [Fig. 2(b)]. An applied field of $-3.0$ kOe results in most of the magnetic moments for nanowires being switched in the direction of the applied field ($z$ direction). Note that this field is not strong enough to switch a single decoupled wire (3.7 kOe). The reversal of some wires occurs because the stray field of neighboring wires adds to the external field and leads to a higher field to switch the magnetic moment. From the magnified view on the top of one nanowire [Fig. 2(c)], it can be seen that 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 $d=10$ nm deviates slightly from homogeneous rotation.

The magnetization reversal and coercivity of the nanowires are very sensitive to the nanowire diameter. Shtrikman and co-workers have predicted that for perfect infinite cylinders with different diameters, the magnetization reversal occurs by coherent rotation, magnetization curling, and buckling mode.\(^{26,27}\) Usually coherent or uniform rotation occurs when the nanowire diameters are smaller than the critical diameter $d_{coh} = \sqrt{24A/\mu_0 M_s^2}$.\(^{28}\) When the diameters are larger than the critical diameter $d_{coh}$, the magnetization reversal process occurs by magnetization curling mode or buckling mode.

For the magnetization curling mechanism, coercive fields decrease with increasing diameter and could be written as...
The magnetic field of... have switched for... have been fit... theoretical prediction has shown that... of one central nanowire. The colors represent the... are the fitted results using the hexagonal cell of... the nanowire diameter at 5 and 300 K, respectively. Fig. 3 plots the coercivity parallel to the... where $A$ is the exchange stiffness, $k$ is the geometry dependent parameter, and $d$ is the diameter of the nanowires. Figure 3 plots the coercivity parallel to the nanowire axes as a function of nanowire diameter at 5 and 300 K, respectively.

\[
H_{ci} = \frac{2 \pi k A}{\mu_0 M_s} d^2 + H_a, \tag{1}
\]

where $A$ is the exchange stiffness, $k$ is the geometry dependent parameter, and $d$ is the diameter of the nanowires. The solid lines in Fig. 3 are the fitted curves for the buckling reversal mode, the dependence of coercive fields on nanowire diameter can be written as

\[
H_{ci} = \frac{2 \pi k A}{\mu_0 M_s} \frac{1}{d^{2/3}} + H_a. \tag{2}
\]

The dotted line is the fitted result for the curling reversal mode. The fitted exchange parameters are $A = 1.25 \times 10^{-12}$ A/m and $A = 8.02 \times 10^{-13}$ A/m at 5 and 300 K, respectively, which are in agreement with the values of the magnetic materials.

Since the homogeneous rotations and the curling mode represent extreme nucleation modes with vanishing exchange energy and vanishing stray fields, respectively, theoretical prediction has shown that there may exist another reversal mode, i.e., buckling reversal mode. The buckling mode can be described approximately as a homogeneous rotation for each cross section, whereas with a sinusoidal variation along the cylinder axis. For the magnetization buckling reversal mode, the dependence of coercive fields on nanowire diameter can be written as

\[
\Delta E = KV \left(1 - \frac{H}{H_0}\right)^m. \tag{3}
\]

The temperature dependence of the coercivity has been interpreted by the above thermally activated reversal mechanism with $m=2.0$, suggesting the nanowires exhibit similar behavior as that of the Stoner-Wohlfarth model, corresponding to the nanowires with the easy magnetization direction along the wire axes. The exponential time dependence of the magnetization $M(t)$ would show a strong magnetic viscosity effect. For the nanowire arrays, by applying an external magnetic field, the probability of the magnetization jumped over the energy barrier is $P(t) = e^{-t/\tau}$, where $\tau = \tau_0 e^{-\Delta E/kT}$. The exponential time dependence of magnetization has been attributed to the single energy barrier height. However, exponential behavior has seldom been observed due to different energy barriers coming from the distribution of the anisotropy and size in real systems. The grains with smaller volumes or anisotropy will decay faster and the ob-
observed dynamics of the magnetization decay is more like the superposition of the exponential decays with different energy barriers. As a result, the observed time dependence magnetization is approximately linearly dependent on the logarithm of time as follows:

\[ M(t) = M_0 [1 - S(T,H) \ln t], \]  

where \( S(T,H) \) is the magnetic viscosity coefficient caused by the thermally activated magnetization reversal. The magnetic relaxation measurement procedure is as follows: the temperature is fixed at the desired value and saturated with a positive applied field, then the desired negative reversed field is set and held constant, and the magnetization decay measurements \( M(t) \) have been measured for thousands of seconds.

Figure 4 illustrates the time dependence of magnetization \( M(t) \) for the FePt nanowire arrays with the wire diameter \( d=10 \text{ nm} \) at 300 K in different fields. It appears that the magnetization has logarithmic time dependence, exhibiting a typical relaxation behavior. Since the magnetic relaxation behavior is significant around \( H_c \), we chose the applied fields of 0.85\( H_c \), 0.88\( H_c \), and 0.89\( H_c \) as an example to plot in Fig. 4.

According to Eq. (4), the magnetic viscosity coefficients \( S(T,H) \) can be obtained by fitting the time dependence of magnetization. The small deviation in the \( M/M_0 \) observed at 1100 and 1250 s for applied fields of 0.88\( H_c \), and 0.89\( H_c \) is due to the measurement error. Figure 5 shows the field dependence of magnetic viscosity coefficients \( S(T,H) \) at different temperatures for FePt nanowire arrays, from which it can be seen that the magnetic viscosity coefficients increase with increasing temperature. The viscosity coefficients \( S \) present the peak values near the coercive fields \( H_c \) with different temperatures. The maximum values of the magnetic viscosity coefficients \( S_{\text{max}} \) are observed to increase with increasing temperature, as plotted in the inset of Fig. 5.

Bruno et al. have reported that single activation energy barrier fails to describe the observed relaxation phenomena with perpendicular easy magnetization. If it is assumed that there exists a certain distribution of the energy barrier, it should be

\[ V^* = \frac{k_B T}{M_s \left( \frac{H_2 - H_1}{\ln t_2 - \ln t_1} \right) M_{\text{irr}}} \]  

Usually the activation volume is dependent on temperature and wire diameter. As discussed above, magnetization reversal is initiated by thermally activated reversal process with the energy barrier \( \Delta E = K \nu_0 (1-H/H_0)^2 \). According to the definition of the activation volume \( V^* = -(1/M_s)(\partial \Delta E/\partial H) \), activation volume can be given by

\[ V^* = \frac{1}{2} V_0^{1/2} \left[ \frac{k_B T \ln(f_0 \tau)}{K} \right]^{1/2}, \]  

where \( K \) is the anisotropy constant, \( f_0 \) is the attempt frequency, and \( \tau \) is the relaxation time. Due to the nonlinearity of the energy barrier, thermal activation volume \( V^* \) is usually different from the physical volume \( V_0 \).

Figure 6(a) shows the activation volume \( V^* \) as a function of temperature for the wire diameter \( d=10 \text{ nm} \). It can be seen that by increasing the temperature, the activation volumes \( V^* \) increase with the values from 30 to 240 nm\(^2\). By fitting the experimental points with Eq. (7), it can be seen...
that the single wire volume is \( V_0 = \frac{1}{3} \pi d^2 L \), the activation volume \( V^* \) should be proportional to the wire diameter \( d \). Figure 6(b) shows that the activation volume \( V^* \) increases with diameter from 200 nm\(^3\) for \( d = 8 \) nm up to 2000 nm\(^3\) for \( d = 60 \) nm. The activation volume \( V^* \) is 1 order of magnitude smaller than that of the wire volume for all the wire diameters, which confirms the localized mechanism with thermally activated reversal process.

IV. CONCLUSION

In summary, magnetization reversal mechanism and magnetic relaxation of the Fe\(_3\)Pt nanowire arrays with different diameters have been investigated by means of micromagnetic simulation and magnetization measurements. The nanowire arrays show strong magnetic viscosity effect, which is much evident at high temperatures and strongest near the coercive fields of the nanowire arrays. The activation volume dependence on temperature and diameter is consistent with the prediction of the thermally activated magnetization reversal mechanism.

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FIG. 6. (a) Activation volume \( V^* \) dependence on temperature for the wire diameter \( d = 10 \) nm. (b) Activation volume \( V^* \) as a function of wire diameter at 300 K. The solid points are the experimental data and the solid lines are the fitting results with Eq. (7). Obviously that the activation volume \( V^* \) is roughly consistent with \( \Theta^{1/2} \) for the temperature range of 5–300 K. Assuming
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