Thermal activation of magnetization in $Pr_2Fe_{14}B$ ribbons*

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The aftereffect field of thermal activation, which corresponds to the fluctuation field of a domain wall, is investigated via specific measurements of the magnetization behavior in $Pr_2Fe_{14}B$ nanocrystalline magnets. The thermal activation is a magnetization reversal arising from thermal fluctuation over an energy barrier to an equilibrate state. According to the magnetic viscosity and the field sweep rate dependence of the coercivity, the calculated values of the fluctuation field are lower than the aftereffect field and in a range between those of domain walls and individual grains. Based on these results, we propose that the magnetization reversal occurs in multiple ways involving grain activation and domain wall activation in thermal activation, and the thermal activation decreases the coercivity by ∼0.2 kOe in the $Pr_2Fe_{14}B$ ribbons.

Keywords: thermal activation, magnetization reversal, fluctuation field, aftereffect field

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1. Introduction

Nanostructured magnets arouse interest since they can be used to test the basic concepts of ferromagnetism, and possibly applied in permanent magnets with a giant energy product and in high density magnetic storage.[1–5] The process of magnetization reversal, sensitive to the phase composition, microstructure, etc, has long been a hot subject.[6–18] Basically, two models of coercivity mechanism, i.e., domain nucleation and domain wall pinning, are generally accepted to describe the mechanism of magnetization reversal. Due to different sample preparation methods and different analysis methods, the process of magnetization reversal varies widely.[19–22]

Actually, in nanocrystalline magnets with high interface-to-volume ratios, the interfaces between grains, like defects, could be domain nucleation centers or pinning sites of domain walls in magnetization reversal. These contribute to a very complicated process of magnetization reversal. Magnetic activation, which arises from thermal fluctuation over an energy barrier to an equilibrate state[23–25] or from the exchange coupling overcoming the anisotropy energy barrier,[26] is a magnetization reversal and attracts much attention.[27–30] The fluctuation field, determined by the activation volume, is generally used to check the magnetization reversal process. In this paper, we investigate the aftereffect of thermal fluctuation via specific measurements of magnetization behavior, and check the fluctuation field and activation volume in $Pr_2Fe_{14}B$ ribbons. It is expected that these are helpful for understanding the thermal activation process and magnetization reversal in nanocrystalline magnets.

2. Experiment

Isotropic nanostructure Pr–Fe–B ribbons with the nominal composition of $Pr_{12}Fe_{82}B_6$ were obtained directly by using the melt spinning method, in which a molten alloy was quenched on the surface of a copper wheel rotating with the optimal surface velocity of 24 m/s in argon atmosphere of high purity. The only phase detected by X-ray diffraction is the $Pr_2Fe_{14}B$ one, and according to the Scherrer method, it is estimated that the average grain size is 22.8 nm. The curves of time-dependent magnetization and recoil loops were measured with a superconducting quantum interference device (SQUID) at the temperature of 300 K. The measurements were performed along the longitudinal direction of ribbons and no demagnetization factor was corrected.

3. Results and discussion

Figure 1 shows the hysteresis loop for $Pr_2Fe_{14}B$ ribbons at the temperature of 300 K. The squareness of the hysteresis loop is good, which indicates that the sample is relatively homogeneous in magnetic properties. Due to the homogeneous magnetic properties, the magnetization reversal is more uniform and it is helpful for further investigating the thermal activation. The thermal activation is a magnetization reversal over the energy barrier, and it decreases the coercivity due to the thermal fluctuation.[23–25] So the thermal activation has an aftereffect in the behavior of magnetization reversal. For the first time, we investigate the aftereffect of thermal activation via many families of recoil loops with the following method. As shown in Fig. 2, after magnetic saturation in a positive direction, the negative field is applied and kept for 10 min

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(waiting time), and then the field is cycled to zero and back at a number of negative field values $H^{\max}$. Here $H^{\max}$ is the maximum applied field for a certain minor loop with successive increases of 0.1 kOe. Figures 2(a)–2(c) show the recoil loops after the field is fixed at 10 kOe, 13 kOe, and 14 kOe, respectively, for 10 min. There is a large drop of magnetization during the waiting time while keeping the field fixed, and the magnetization does not decrease drastically until the field $H^{\max}$ is increased by a critical value of 0.2 kOe. During the waiting time while keeping the applied field fixed, a drop of magnetization results from the thermal activation. The thermal activation is strongly dependent on time. The probability that magnetization reversal occurs after time t is given by $P(t) = 1 - \exp(-t/\tau)$.[26,31] Here τ follows the Arrhenius law, $\tau = \tau_0 \exp(\Delta E/(k_B T))$, where τ_0 is a pre-exponential factor on the order of 10^{-9}s, which corresponds to the Larmor frequency, ΔE is the energy barrier, a difference between the coercivity and the applied field, and $k_B T$ is the activation energy,[23] which is mainly dependent on the temperature. The longer the waiting time is, the more probable the magnetization reversal occurs via overcoming the energy barrier, and the more equilibratory state it reaches in the sample. The thermal activation decreases the coercivity, and even if the applied field is increased by less than the critical field, the magnetization is stable and hardly changes. So the critical field is strongly dependent on the thermal activation and fluctuation field, thus called an aftereffect field $\Delta H_{\rm af}$. In order to check the results, we study the magnetization behavior by another method. As shown in Fig. 3(a) and 3(b), the field is kept fixed for a waiting time of 10 min, and then increased at a low sweep rate of 5 Oe/s. As the field starts to increase, the magnetization state keeps nearly stable, and as the field is increased by a certain amount, the magnetization changes drastically. The tangents of the curves of magnetization keeping stable and magnetization changing drastically have a crosspoint at which the field has a difference of critical field with the field kept fixed. It is verified that all the measured critical fields, i.e., aftereffect fields, are 0.2 kOe (shown in the inset of Fig. 3(a)).

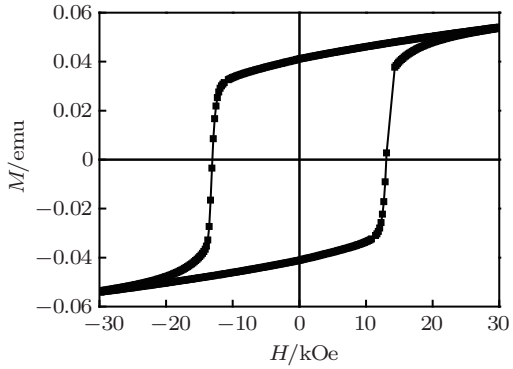

Fig. 1. Hysteresis loop for $Pr_2Fe_{14}B$ ribbons at the temperature of 300 K.

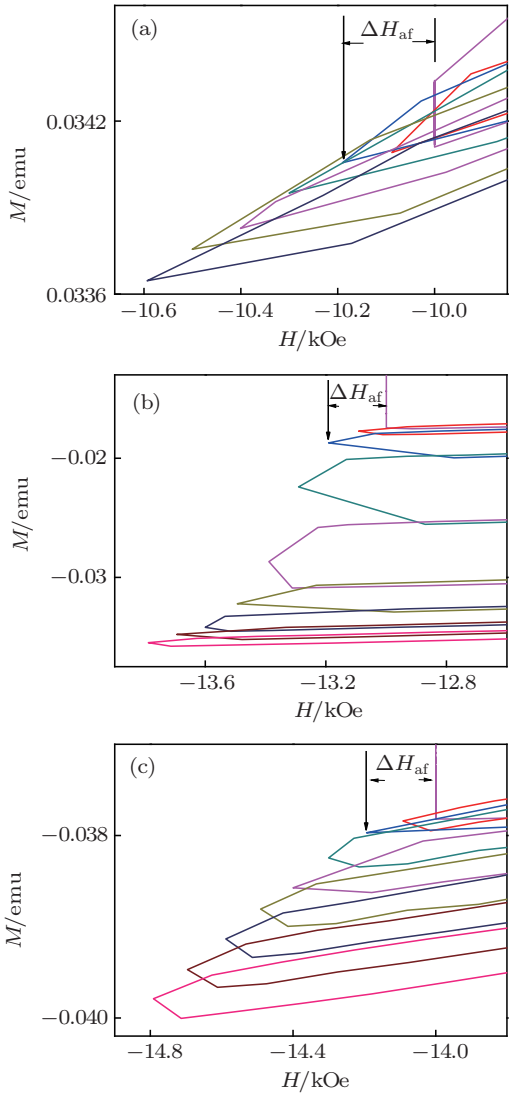

Fig. 2. (color online) Recoil loops while cycling the field to zero and back at a number of negative fields after keeping the field fixed at (a) 10 kOe, (b) 13 kOe, and (c) 14 kOe for 10 min.

The aftereffect field results from the thermal activation and is supposed to depend on the magnetization reversal process. Since the aftereffect fields are the same at different applied fields in these measurements, possibly they reflect the intrinsic properties in magnetization reversal. Actually, in nanocrystalline magnets, the magnetization reversal process is rather complicated. It is believed that domain walls are at the interface where the domain wall energy is probably a minimum. The interface between grains, like defects, could be domain nucleation centers or pinning sites of domain walls. If the nucleation of a reversed domain is dominant for magnetization reversal in thermal activation, the activation volume corresponds to that of the domain wall. According to the formula $\delta = \pi\sqrt{A/K}$ ($A = 7.8 \times 10^{-12}$ J/m is the exchange coefficient, and $K = 5.6$ MJ/m^3 is the magnetoanisotropy at the temperature of 300 K),[32] the domain wall thickness δ is about 3.7 nm for $Pr_2Fe_{14}B$ magnets. The domain wall length and width are compatible with the grain dimension. We suppose that the effective dimension (mean size) of the domain wall is 5.5 nm,

which should be the intrinsic properties in the magnets. According to the formula $H_f = kT/vM_s$ ($k = 1.38 \times 10^{-23}$ J/K, $T = 300$ K, $v = (5.5 \times 10^{-9}$ m$)^3$, $M = 1.55$ T),[23] the fluctuation field of domain walls is 0.2 kOe, which corresponds to the aftereffect field obtained in the measurements. It seems that the magnetization reversal in thermal activation occurs in the way of domain wall activation in these measurements.

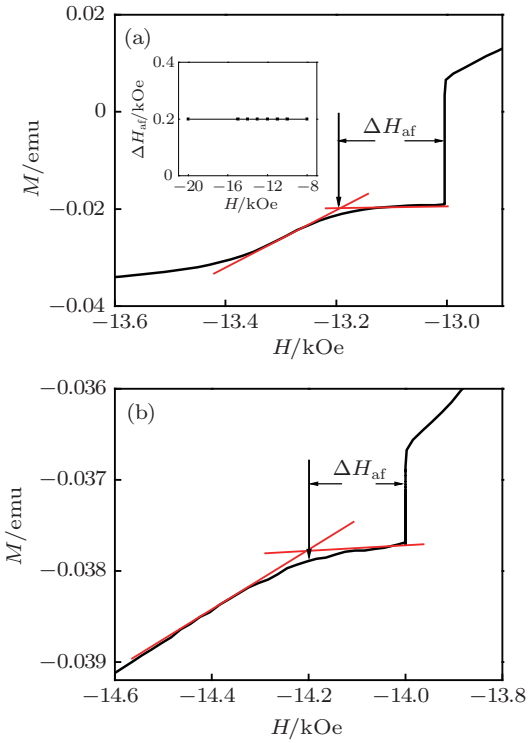

Fig. 3. (color online) Demagnetization curves while the field is increased at a rate of 5 Oe/s after keeping the field fixed for 10 min. The inset shows the measured aftereffect field.

If the domain wall pinning effect is strong and the magnetization reversal occurs in the way of depinning, i.e., the domain wall jumping from the grain boundary to the next pinning center,[23,24] the activation volume is the grain volume.[7,28,29] The fluctuation field H_f could be obtained by the two equivalent definitions of activation energy, $E = kT$ and $E = vM_sH_f$.[23] According to the formula $H_f = kT/vM_s$ ($k = 1.38 \times 10^{-23}$ J/K, $T = 300$ K, $v = (22.8 \times 10^{-9}$ m$)^3$, $M =$ 1.55 T), the fluctuation field is very small, about 0.003 kOe for the $Pr_2Fe_{14}B$ ribbons. Given the inhomogeneous microstructure, we assume that the grain size is distributed in a wide range, 10–50 nm, thus the fluctuation field is in the range of 0.000027–0.034 kOe, much less than the aftereffect field ΔH_{af} obtained in the measurements. According to the formula $H_f = S/x_{irr}$,[23] the sample has less viscosity and less activated magnetization. It seems that the thermal activation does not occur by the activation of individual grains.

In order to further probe the thermal activation process, we investigate the fluctuation field and activation volume by measuring the magnetic viscosity of the sample.[25,30] The applied field is kept fixed for time t and the magnetic viscosity S is obtained from the time dependent magnetization, $M_{(t)} = M_0 - S\ln(t_0 + t)$, where M_0 and t_0 are constants.[23,24] Figure 4(a) shows the irreversible susceptibility x_{irr} and the viscosity coefficient S. The fluctuation field can be given as $H_f = S/x_{irr}$,[23] and the activation size is obtained by the formula $d_{active} = \sqrt[3]{v}$, where $v = kT/M_sH_f$. Figure 4(b) shows the measured fluctuation field and activation size. The fluctuation fields are in the range between those of the domain walls and individual grains. The fluctuation field could be obtained by using another method, the field sweep rate dependence of the coercivity given by the formula $H_c(R) = H_f\ln(R) + \text{const}$, where R is the sweep rate of the field.[30] The fluctuation field is 0.056 kOe (the inset in Fig. 4(a)), also more than that of the individual grains and less than that of the domain walls.

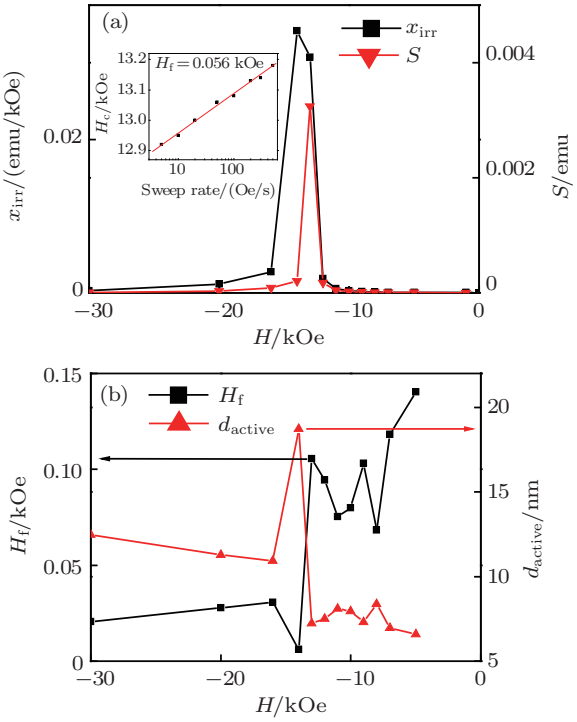

Fig. 4. (color online) (a) Irreversible susceptibility, viscosity coefficient, (b) fluctuation field, and activation size as functions of the applied field. The inset shows the field sweep rate dependence of coercivity for $Pr_2Fe_{14}B$ ribbons.

With these in mind, the process of thermal activation in the $Pr_2Fe_{14}B$ ribbons could be speculated. During the waiting time, in some grains in which the coercive field is 0–0.003 kOe larger than the applied field, the magnetization reversal may occur by the activation of individual grains. This is called pinning/depinning of the domain wall from one interface to the next interface. In some grains in which the coercivity is 0.003–0.2 kOe larger than the applied field, the magnetization reversal occurs by domain wall motion from the interface gradually to the main phase with the driving of thermal fluctuation of the domain walls, simultaneously the domain wall thickness decreases and its energy increases. This is called

the nucleation mechanism, i.e., the domain nucleation at the grain surface in the main phase, and in some papers it is called passage and expansion.[33,34] So the magnetization reversal in thermal activation is multi-process, involving grain activation and domain wall activation. The domain nucleation by thermal activation may decrease the coercivity field by about 0.2 kOe. So the aftereffect field corresponds to 0.2 kOe, which is larger than the measured fluctuation field. The measured fluctuation field and activation size are averages of those of domain walls and individual grains. It can be seen in Fig. 4(b) that at the coercivity field (13 kOe), the measured fluctuation field is close to that of domain walls, so the activation of the domain wall is dominant during the process of thermal activation under the field of 13 kOe. At the field of 14 kOe, there is less magnetic activation, and the activation size is close to the grain size, so the pinning effect on the domain wall motion is dominant during the process of thermal activation.

4. Conclusion

In summary, in $Pr_2Fe_{14}B$ nanocrystalline magnets, the magnetization reversal occurs probably in multiple ways involving grain activation and domain wall activation in thermal activation. The measured fluctuation field and activation size are averages of those of domain walls and individual grains, and the thermal activation decreases the coercivity of grains by ~ 0.2 kOe. So the aftereffect field of thermal activation is 0.2 kOe, which corresponds to the fluctuation field of the domain walls.

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