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Nucleation of reversed domain and pinning effect on domain wall motion in nanocomposite magnets

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The magnetization behaviors show a strong pinning effect on domain wall motion in optimally melt-spun $Pr_8Fe_{87}B_5$ ribbons at room temperature. According to analysis, the coercivity is determined by the nucleation field of reversed domain, and the pinning effect, which results from the weak exchange coupling at interface, makes domain nucleation processes independent and leads to non-uniform magnetization reversals. At a temperature of 60 K, owing to the weak exchange coupling between soft-hard grains, magnetization reversal undergoes processes of spring domain nucleation in soft grains and irreversible domain nucleation in hard grains, and the pinning effect remains strong among hard grains. © *2013 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4817968]

Nanocomposite magnets with a large number of interfaces, at which there exists the effect of exchange coupling between soft magnetic grains with high saturation magnetization and hard magnetic grains bearing high coercivity, have attracted much attention since giant energy product could possibly be achieved.[1] The process of magnetization reversal, sensitive to phase composition, microstructure, etc.,[2–4] has been investigated extensively to improve the magnetic properties.[5–16] Using different analysis methods or different types of samples, the process of magnetization reversal will change from conventional domain nucleation,[12,13] domain wall pinning,[5–7] to self-pinning at soft/hard interface.[14] It seems difficult to find intrinsic properties in the process of magnetization reversal. Due to grain size in the nanometer scale, hybrid structure, a large number of interfaces, and the exchange coupling effect between grains, the mechanism of magnetization reversal becomes rather complicated compared with that in conventional sintered magnets.

Actually, in nanocomposite magnets, the hard grains are in a single-domain state, and a spin disorder is penetrated into soft phase whose penetration length is dependent on applied field.[17] Since domain wall energy is probably minimal at interface, the domain wall stays generally at grain boundary and the interface has a pinning effect on domain wall motion. For example, in nanocrystalline Sm_2Co_{17} based alloys, the phase boundary acts as strong repulsive or trapping barriers for the displacement of domain walls.[18,19] Due to exchange coupling at interface, the magnetization behavior becomes collective among grains and the pinning effect is weak. These contribute to a competitive behavior in domain wall motion. In this paper, we investigate the magnetization reversal via magnetization behavior at room temperature and a temperature of 60 K by using a comparative method in analysis, which is expected to be helpful for better understanding the nucleation of reversed domain, pinning effect and exchange coupling effect in magnetization reversal.

The precursor ingots of $Pr_8Fe_{87}B_5$ and $Pr_{12}Fe_{82}B_6$ were prepared by arc melting method under an argon atmosphere. The ribbons were obtained by using the method of induction melting the ingot in a quartz tube and then blowing the melt onto the surface of a rotating copper wheel with pressurized argon atmosphere. The wheel surface velocity was variable for optimizing the magnetic properties. X-ray diffraction (XRD) was used to confirm a mixture nanostructure of α-Fe and isotropic $Pr_2Fe_{14}B$ phases in $Pr_8Fe_{87}B_5$ ribbons, and a single phase $Pr_2Fe_{14}B$ nanostructure in $Pr_{12}Fe_{82}B_6$ ribbons. Magnetic measurements were performed using Lakeshore VSM and superconducting quantum interference device (SQUID) VSM.

Fig. 1(a) shows the virgin magnetization curve and the field H^{max} dependence of coercivity $H_c(H)$ for optimal melt-spun $Pr_8Fe_{87}B_5$ ribbons at room temperature. Here, $H_c(H)$ and H^{max} are coercivity and maximum applied field, respectively, for a certain minor hysteresis curve. The inset in Fig. 1(a) shows the minor hysteresis loops. The values of magnetization is low and increases slowly at low field less than coercivity field, which indicates that the domain wall pinning is dominant in the process of magnetization reversal.[2–6] The field dependence of coercivity confirms this coercivity mechanism.[2–6] Based on these results, the pinning effect on domain wall motion at interface is understandable. In the virgin state, magnetization of half grains is in the same hemisphere as the applied field. It is believed that domain walls are at the interface where domain wall energy is probably minimal. If the pinning effect is weak at interface, the domain wall motion occurs easily and the sample is magnetized to saturation at low field. So the low susceptibility at low field implies that a strong pinning occurs at interface and the domain wall is less free in initial magnetization process. Basically, magnetization reversal results from hard grains,[20] so the pinning effect more probably arises from the interface at hard grain boundary. However, the exchange coupling effect between grains promotes magnetic moments to arrange in the same direction, and the domain wall energy would increase at interface, thereby resulting in an exchange coupling domain involving multi-grains.[21,22] More probably, the microstructure is inhomogeneous in the ribbons, and the effect of exchange coupling is weaker than that of effective anisotropy at some interfaces where the domain wall exists.

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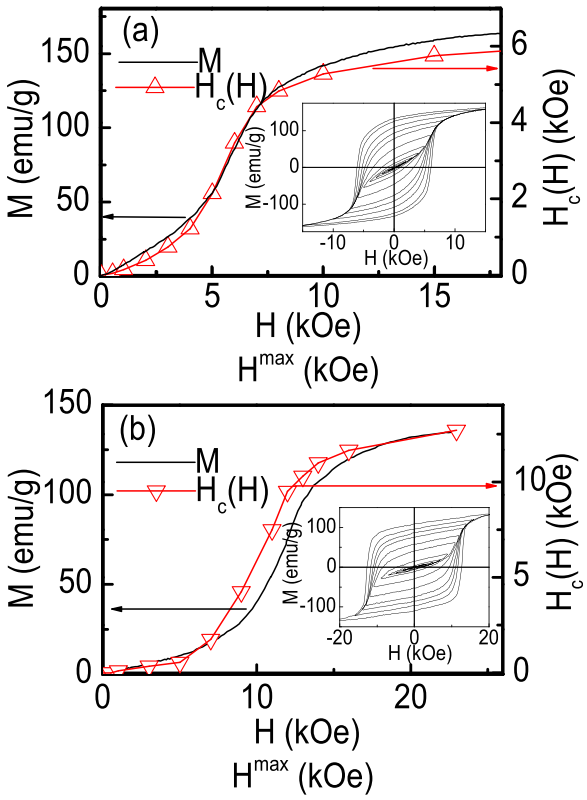

FIG. 1. Initial magnetization curve and field dependence of coercivity for $Pr_8Fe_{87}B_5$ (a) and for $Pr_{12}Fe_{82}B_6$ ribbons (b) at room temperature, and the insets show minor hysteresis loops for $Pr_8Fe_{87}B_5$ (a) and for $Pr_{12}Fe_{82}B_6$ ribbons (b).

The effective anisotropy in soft grains is very weak, so the domain wall is at hard grain boundary and a spin disorder penetrates into the soft phase, whose penetration length is dependent on applied field.[17]

To understand fully the pinning effect on domain wall motion in nanocomposite magnets, for a comparison of analysis, we investigate magnetization behavior in nanocrystalline magnets of $Pr_{12}Fe_{82}B_6$ ribbons. Fig. 1(b) shows initial magnetization curve and field dependence of coercivity at room temperature, which indicate that the magnetization reversal is also governed by domain wall pinning.[2–6] The inset in Fig. 1(b) shows minor hysteresis loops. Even though the pinning effect is strong in both samples, the squareness of hysteresis loop is better for $Pr_{12}Fe_{82}B_6$ ribbons than for $Pr_8Fe_{87}B_5$ ribbons. The squareness is strongly dependent on the process of magnetization reversal, and in order to further investigate the magnetization reversal, we check the nucleation field dependence of coercivity. Fig. 2 shows the dependence of $\mu_0 H_c/J_s$ on $\mu_0 H_N^{\min}/J_s$ in the two samples. Here, H_c is coercivity at a specific temperature, and the minimum nucleation field $H_N^{\min}$ denotes the average value for the nucleation field in hard grains with easy axis aligned randomly. $H_N^{\min} = K_1^{eff}/J_s$, and K_{eff} and J_s are taken in Ref. 23. The relationship between coercivity and the minimum nucleation field is liner and agrees with the modified Brown equation, $\frac{\mu_0 H_c(T)}{J_s(T)} = a_K a_{ex} \frac{\mu_0 H_N^{\min}(T)}{J_s(T)} - N_{eff}$. This result indicates that the nucleation of reversal domain also controls the process of magnetization reversal.[12,13] In both samples, the domain nucleation is necessary and the pinning effect is strong in magnetization reversal, so the difference in the

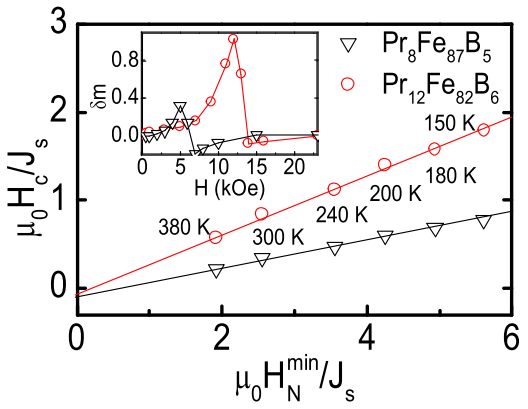

FIG. 2. The dependences of $\mu_0 H_c/J_s$ on $\mu_0 H_N^{\min}/J_s$ for $Pr_8Fe_{87}B_5$ and $Pr_{12}Fe_{82}B_6$ ribbons.

squareness of hysteresis loop possibly results from other factors in the two samples.

The exchange coupling has a great effect on magnetization reversal. The value of δm (Henkel plots) could be used to check the exchange coupling effect between grains, especially between hard grains,[20] which is defined as $\delta_m = [M_d(H) + 2M_r(H)]/M_r - 1$.[24,25] Here $M_r(H)$ is a remanence acquired after the application and subsequent removal of a field H in virgin magnetization, $M_d(H)$ is a remanence obtained after saturation in one direction and then the subsequent application and removal of a field H in the opposite direction. Positive δ_m value is an indication of stronger exchange coupling effect between grains. The inset in Fig. 2 shows δm curves at room temperature, which demonstrate that the effect of exchange coupling in nanocomposites $Pr_8Fe_{87}B_5$ is weaker than in nanocrystallines $Pr_{12}Fe_{82}B_6$. The weak exchange coupling between hard grains possibly results from intergranular soft region.[20]

Bearing these in mind, in nanocomposite magnets, the process of domain nucleation, pinning effect on domain wall motion and exchange coupling between grains can be speculated in magnetization reversal. The nucleation of reversed domain is necessary in magnetization reversal, and magnetization reversal occurs more probably in some grains with low effective anisotropy. However, it is the pinning effect that is responsible for suppressing the domain wall motion into neighbor grains with high effective anisotropy. So in nanocomposite magnets, the nucleation of reversed domain and domain propagation is more independent among grains not only among different magnetic domains, but in same magnetic domain, and the magnetization reversals are nonuniform and demagnetization curve has less perfect squareness. In nanocrystalline magnets, though the pinning effect is strong among magnetic domains, due to strong exchange coupling effect between grains, the nucleation of reversed domains and domain propagations are more dependent among grains, which contributes to a more uniform magnetization reversal and a good squarenenss in hysteresis loop.

It seems that the process of magnetization reversal is much clear in nanocomposite magnets, which is dominated by magnetization behavior in hard grains. The pinning effect originates from the interface where exchange coupling is weak at hard grains boundary, and makes domain nucleation independent among grains. However, in nanocomposites, how does the magnetization behave between soft-hard grains?

Basically, if soft grains are well exchange coupled with hard grains, magnetization reversal keeps coherent in soft and hard grains, and the magnetization process takes place simultaneously in the soft as well as in the hard grain. Actually, owing to inhomogeneous microstructure, some soft grains possibly are imperfectly exchange-coupled with hard grains. In order to understand fully the process of magnetization reversal, we investigate the magnetization behavior at a temperature of 60 K since the exchange coupling between soft-hard grains becomes weak at such a low temperature.[23]

Fig. 3 and the inset show the demagnetization curve and remanence curve for $Pr_8Fe_{87}B_5$ ribbons at temperature of 60 K and at room temperature, respectively. The squareness of demagnetization curve deteriorates at the temperature of 60 K, which evidences that the exchange coupling is weak between soft-hard grains, and magnetization reversal occurs first in soft grains. Yet even so, the squareness of remanence curve is nearly same with that at room temperature, which indicates that the magnetization reversal is reversible in soft grains. So at the temperature of 60 K, owing to the weak exchange coupling between soft-hard grains, magnetic domain nucleation occurs in soft grains at low applied field. From soft grain to hard grain boundary, domain wall energy increases monotonically without a minimum in the domain propagation, and there are no pinning sites in the domain wall motion. As the field is cycled to zero, the magnetization recovers to initial state in soft grains. Thus the domain nucleation is a magnetic spring behavior, which mainly occurs in soft grains. Only as the applied field is increased to nucleation field in hard grains does the irreversible nucleation of reversed domain occur at the hard grain boundary. This magnetization behavior was also observed in Ref. 13 and called as two steps behavior in high-density recording systems and a three-step process in a simple triple-layer system.[26,27]

The reversible behavior of domain nucleation could be also observed in the initial magnetization curve. Fig. 4(a) shows the initial magnetization curve and field dependence of coercivity for $Pr_8Fe_{87}B_5$ ribbons at the temperature of 60 K, and the inset in Fig. 4(a) shows minor hysteresis loops. Since at low field the magnetization increases drastically, it implies that the reversible domain nucleation occurs in soft grains.[2–6] Fig. 4(b) shows the initial magnetization curve and field dependence of coercivity for $Pr_{12}Fe_{82}B_6$ ribbons at the temperature of 60 K, and the inset in Fig. 4(b) shows minor hysteresis loops. The initial magnetization curve is different with that for $Pr_8Fe_{87}B_5$ ribbons, which should be attributed to the reversible nucleation of soft grains in $Pr_8Fe_{87}B_5$ ribbons. Yet even so, via the field dependence of coercivity in the two samples, it is suggested that the domain wall pinning effect remains strong in magnetization reversal in $Pr_8Fe_{87}B_5$ ribbons as well as in $Pr_{12}Fe_{82}B_6$ ribbons. So even though the nucleation field of reversed domain is low in soft grains, the resistance against magnetization reversal is strong in hard grains, and the magnetization behaviors remain independent among hard grains in $Pr_8Fe_{87}B_5$ ribbons at the temperature of 60 K.

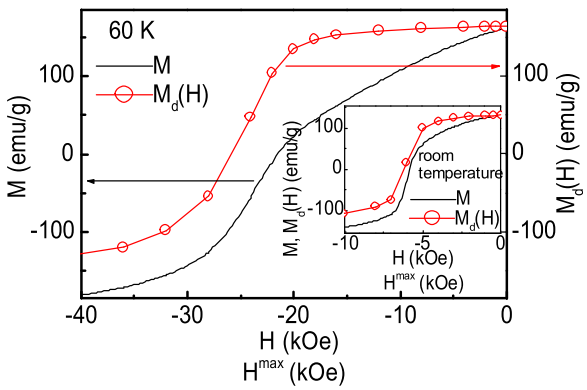

FIG. 3. Demagnetization curve and remanence for $Pr_8Fe_{87}B_5$ ribbons at a temperature of 60 K. The inset shows demagnetization curve and remanence for $Pr_8Fe_{87}B_5$ ribbons at room temperature.

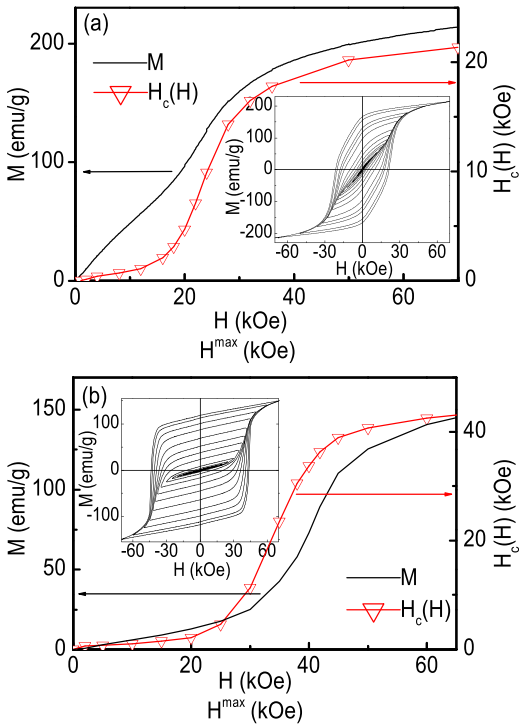

FIG. 4. Initial magnetization curve and field dependence of coercivity for $Pr_8Fe_{87}B_5$ (a) and for $Pr_{12}Fe_{82}B_6$ ribbons (b) at a temperature of 60 K, and the insets show minor hysteresis loops for $Pr_8Fe_{87}B_5$ (a) and for $Pr_{12}Fe_{82}B_6$ ribbons (b).

In summary, in optimal melt-spun ribbons, the magnetization process takes place simultaneously in soft as well as in hard grain due to the optimum exchange coupling between soft-hard grains. According to the nucleation field dependence of coercivity, the nucleation of reversed domain is necessary in magnetization reversal, and the pinning effect on domain wall motion makes the nucleation of reversed domain independent among grains and leads to a more non-uniform magnetization reversal. As exchange coupling becomes weak between soft-hard grains at a low temperature of 60 K, the nucleation of reversed domain occurs first in soft grains, yet even so the pinning effect remains strong at hard grain boundary.

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