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Magnetization and demagnetization behaviours of melt-spun $Pr_{12}Fe_{82}B_6$ and $Pr_8Fe_{87}B_5$ ribbons*

Du Xiao-Bo(杜晓波)[a)b)†], Zhang Hong-Wei(张宏伟)[a)], Rong Chuan-Bing(荣传兵)[a)], Zhang Jian(张 键)[a)], Zhang Shao-Ying(张绍英)[a)], Shen Bao-Gen(沈保根)[a)], Yan Yu(闫 羽)[b)], and Jin Han-Min(金汉民)[b)]

[a)] *State Key Laboratory of Magnetism, Institute of Physics and Centre for Condensed Matter Physics, Chinese Academy of Sciences, Beijing* 100080, *China*

[b)] *Department of Physics, Jilin University, Changchun* 130023, *China*

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Nanocrystalline $Pr_{12}Fe_{82}B_6$ and nanocomposite $Pr_8Fe_{87}B_5$ ribbons have been prepared using a melt spinning technique. Recoil loops have been measured at 20, 200 and 300K. Demagnetization curves are analysed by dividing it into reversible and irreversible portions. High recoil loop susceptibility at low applied field and large reversible change in the demagnetization curve have been found in $Pr_8Fe_{87}B_5$ ribbons, showing that the reversible behaviours in nanocomposite permanent magnets originate primarily from the magnetically soft phase. The hysteresis in recoil loops found in $Pr_8Fe_{87}B_5$ ribbons originates from the soft phase α-Fe that suffers a stress.

Keywords: nanocomposite magnets, recoil loops, permeability, reversible and irreversible behaviour
PACC: 7550B,7550K,7550V, 7560E

1. Introduction

Nanocomposite permanent magnets (NPMs) have attracted much attention since the discovery of remanence enhancement in nanocomposite material comprising $Nd_2Fe_{14}B$ and Fe_3B.[1−3] NPM consists of magnetically hard and soft phases. The hard phase contributes the high anisotropy and the soft phase provides large spontaneous polarization. Because of intergrain exchange coupling (IGEC), the magnetization vector of the soft phase is exchange coupled by that of the hard phase, which can lead to a single magnetic hard phase behaviour shown in demagnetization curve of NPM. For an assembly of small isotropic non-interacting particles with uniaxial anisotropy, the reduced remanence $m_r=J_r/J_s=1/2$ (J_r is the remanent polarization, J_s the saturation polarization) is predicted by the Stoner–Wohlfarth model.[4] Meanwhile, m_r is larger than 1/2 for isotropic NPM.[1−3] The enhancement of remanence is of great benefit to high energy product $(BH)_{max}$. The theoretical value of $(BH)_{max}$ is about 1 MJ/m^3 in anisotropic NPM.[5] So a considerable interest has been aroused in the past decade by these new rare-earth based magnets in which the content of expensive rare-earth component is reduced.[6−12]

In comparison with single-phase nanocrystalline magnet, the presence of magnetically soft phase in nanocomposite magnet brings significant changes in magnetization and demagnetization behaviours. The reversible behaviour becomes pronounced. The hard magnetic properties are changed as well. The saturation polarization and remanence increase, while the coercivity falls off evidently. Investigating the similarity and difference in magnetic behaviours between single-phase and two-phase magnets is helpful to understanding the physics of magnetization and demagnetization processes.

In this work, we measured the recoil loops of $Pr_{12}Fe_{82}B_6$ (single-phase) and $Pr_8Fe_{87}B_5$ (two-phase) melt-spun ribbons at different temperatures. Reversible and irreversible behaviours in the magnetization and demagnetization processes were investigated in detail.

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†E-mail: dxb@g203.iphy.ac.cn

2. Experimental detail

Ribbons with composition of $Pr_{12}Fe_{82}B_6$ and $Pr_8Fe_{87}B_5$ were prepared by melt-spinning at optimum wheel speeds. Obtained ribbons are about 2mm wide and 30μm thick. Structure of the ribbons is determined by x-ray diffraction with Cu $K\alpha$ radiation in a Rigaku Rint. Magnetic measurements were performed using a superconducting quantum interference device magnetometer (MPMS-7). The field was applied along the longitudinal direction of the ribbon so the demagnetization factor could be neglected.

3. Results and discussion

According to XRD patterns, the $Pr_{12}Fe_{82}B_6$ ribbons mainly consist of $Pr_2Fe_{14}B$ phase. Obtained remanence J_r, coercivity $\mu_{0i}H_c$ and maximum energy product $(BH)_{max}$ at 300K are 0.91T, 1.13T and 143kJ/m^3, respectively. $Pr_8Fe_{87}B_5$ ribbons mainly consist of $Pr_2Fe_{14}B$ and α-Fe. The J_r, $\mu_{0i}H_c$ and $(BH)_{max}$ are 1.19T, 0.52T and 140kJ/m^3, respectively.

Figure 1 shows the recoil loops along both magnetization and demagnetization branches for $Pr_{12}Fe_{82}B_6$ and $Pr_8Fe_{87}B_5$ ribbons, measured at 20K, 200K and 300K, respectively. The recoil loops are obtained as follows: starting from a point in magnetization or demagnetization curve, the field is decreased to zero and then increased to the starting point.

It can be seen in Fig.1 that the recoil loops of $Pr_{12}Fe_{82}B_6$ ribbons are linear for both magnetization and demagnetization branches. According to Ref.[13], such a linear change of magnetization with applied field originates from the moment rotation away from easy axis in misaligned grains. This conclusion is further confirmed by our calculation based on the moment rotation model. For an isotropic $Pr_2Fe_{14}B$ magnet comprising single-domain particles without interaction, the calculated susceptibilities at field about 2–5T are 0.03, 0.07 and 0.13μ_0, respectively, corresponding to the temperatures of 20, 200 and 300K. These results are roughly in agreement with the experimental values of 0.05, 0.06 and 0.14 μ_0. The increase of susceptibility with increasing temperature, as manifested by both experiment and calculation, is due to the fast decline of anisotropy field with increasing temperature, since the departure of magnetization vector away from the easy axis becomes easy as the anisotropy field is weakened.

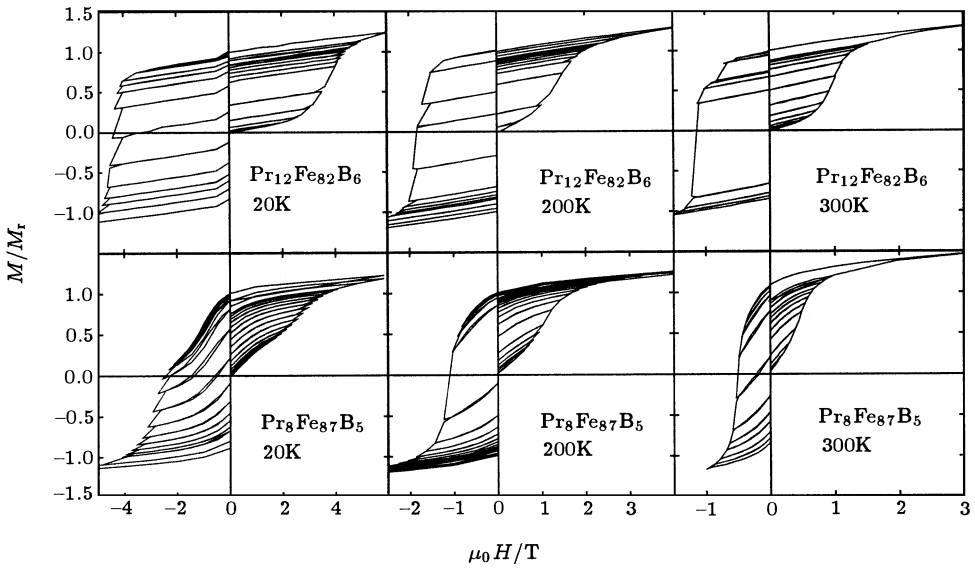


Fig.1. Recoil loops of $Pr_{12}Fe_{82}B_6$ and $Pr_8Fe_{87}B_5$ ribbons at 20, 200 and 300K.

The situation is quite different in $Pr_8Fe_{87}B_5$ ribbons. The recoil loops are no longer linear. In the magnetization branch, the susceptibility of recoil loops is large at first and declines with increasing applied field. The large susceptibility at low field is due to reversible rotation of α-Fe soft phase. At high field, the rotation of soft phase has completed. Hence the susceptibility is primarily contributed by the reversible rotation of hard phase and approaches gradually to the corresponding value of $Pr_{12}Fe_{82}B_6$ ribbons. In the demagnetization branch, the susceptibility of recoil loops is even larger due to a large rotation of soft

phase moments from the directions approximately opposite to the external field to the direction parallel to the external field. In the magnetization branch the rotation of soft phase moments begins at angles near the direction of applied field. Hence the change of polarization is small. In addition, the easier reversal of soft phase leads to a circular demagnetization curve in the second quadrant while the demagnetization curve is quadrate for $Pr_{12}Fe_{82}B_6$ ribbons.

Another feature, namely the hysteresis in recoil loops, especially in the demagnetization branch, is found in $Pr_8Fe_{87}B_5$ ribbons. Similar behaviour is also observed in $Sm_2Fe_{15}Si_2C/\alpha$-Fe nanocomposite magnet.[14] Considering that no hysteresis is found in the recoil loops of $Pr_{12}Fe_{82}B_6$ ribbons, it is safe to say that this phenomena is associated with the presence of soft phase α-Fe. But just ascribing the hysteresis to that of α-Fe seems unreasonable, because the coercivity of α-Fe is only about several hundreds A/m, while maximum coercivity measured in the recoil loops is about 70kA/m. A possible reason is the strain that the α-Fe grains suffer.[15] In $Nd_2Fe_{14}B/\alpha$-Fe nanocomposite ribbons,[16] $Nd_2Fe_{14}B$ hard phase grains suffer no obvious stress. In contrast, α-Fe soft phase grains suffer a stress (tensile) of approximate 1GPa. As estimated in Ref.[17], the coercivity of α-Fe grains in nanocomposite magnet from the strain consideration is about 50kA/m, being consistent with the experimental value of 70kA/m in the order of magnitude.

Analysis of the demagnetization curve in terms of reversible and irreversible portions is helpful for understanding the physical essential about the demagnetization behaviour. The irreversible portion is defined as $\Delta m_{\rm irrev}=[J_{\rm r}\text{-}J_{\rm d}(H)]/2J_{\rm r}$,[2] where $J_{\rm r}$ is saturation remanent polarization, $J_{\rm d}(H)$ is dc field demagnetization remanent polarization acquired after saturation in one direction and subsequent application and removal of a dc field H in the opposite direction. Reversible portion is defined as $\Delta m_{\rm rev}=[J_{\rm d}(H)-J(H)]/2J_{\rm r}$,[2] $J(H)$ is the polarization in demagnetization curve when a reverse field H is applied.

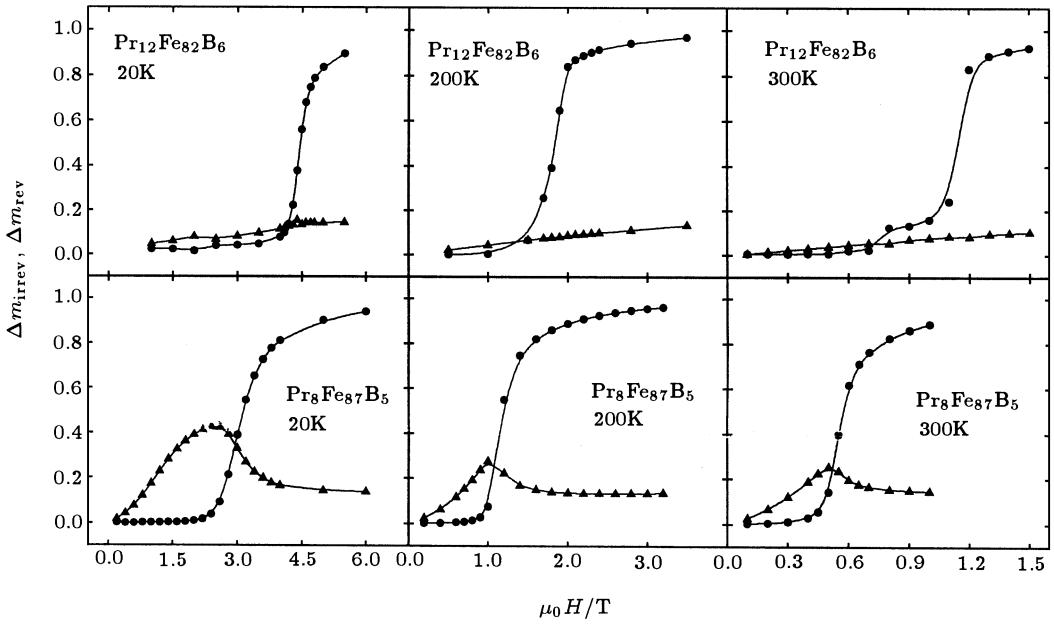


Fig.2. Reversible $\Delta m_{\rm rev}$ (▲) and irreversible portions $\Delta m_{\rm irrev}$ (●) of demagnetization curves versus reverse field for $Pr_{12}Fe_{82}B_6$ and $Pr_8Fe_{87}B_5$ ribbons, measured at 20,200 and 300K.

The change of $\Delta m_{\rm irrev}$ and $\Delta m_{\rm rev}$ with reverse field H are shown in Fig.2. One can notice that $\Delta m_{\rm irrev}$ varies with H in the same way for both $Pr_{12}Fe_{82}B_6$ and $Pr_8Fe_{87}B_5$ ribbons. They remain constant for small reverse field H and increase rapidly once H exceeds a critical field which, according to Ref.[2], is the nucleation field $H_{\rm n}$ of the hard phase. The steep rise in $\Delta m_{\rm irrev}$ arises from the irreversible reversal of hard phase. As temperature increases, $H_{\rm n}$ decreases due to the decline of anisotropy field of $Pr_2Fe_{14}B$ hard phase. In addition, $H_{\rm n}$ of $Pr_8Fe_{87}B_5$ ribbon is smaller than that of $Pr_{12}Fe_{82}B_6$ ribbons at the same temperature. This is because the $Pr_2Fe_{14}B$ hard phase in $Pr_8Fe_{87}B_5$ ribbons reverses easily when

they are pulled by the neighbouring soft phase which has already turned their polarization into the direction of H.

The field dependence of $\Delta m_{\rm rev}$ is different for $Pr_{12}Fe_{82}B_6$ and $Pr_8Fe_{87}B_5$ ribbons. For $Pr_8Fe_{87}B_5$ ribbons, $\Delta m_{\rm rev}$ rises from zero with increasing H and reaches a maximum value at $H_{\rm n}$. This is caused by the competition between external H and IGEC. The external field attempts to pull the moments of soft phase into its own direction, but the neighbouring hard phases attempt to maintain the soft phase in its previous direction through IGEC. This competition makes the moments in the core of soft grains reversed but leaves the moments near the surface unchanged. The stronger the reverse field is, the more the moments of soft phase reverse. So $\Delta m_{\rm rev}$ increases gradually with increasing external field. Once the external field exceeds $H_{\rm n}$, partial hard phase reverses irreversibly, causing the neighbouring soft phase to reverse irreversibly as well. Then $\Delta m_{\rm rev}$ falls off gradually. The peak value of $\Delta m_{\rm rev}$ declines with increasing temperature due to the decline of $H_{\rm n}$. As for $Pr_{12}Fe_{82}B_6$ ribbons, $\Delta m_{\rm rev}$ increases monotonically with the increasing external field. No doubt, it is the sequence of moment rotation away from the easy axis to the direction of applied field in misaligned grains.

4. Conclusion

Melt-spun $Pr_{12}Fe_{82}B_6$ and $Pr_8Fe_{87}B_5$ ribbons were prepared by melt spinning. Recoil loops were measured at 20, 200 and 300K. The linear recoil loops of $Pr_{12}Fe_{82}B_6$ ribbons originate from the moment rotation away from the easy axis in misaligned grains, while for $Pr_8Fe_{87}B_5$ ribbons, the recoil loops are not linear. Large susceptibility at low field originates from the reversible reversal of soft phase. The hysteresis in the recoil loops results from the strain the α-Fe grains suffer.

The irreversible portion of demagnetization curve shows similar behaviour for both $Pr_{12}Fe_{82}B_6$ and $Pr_8Fe_{87}B_5$ ribbons. $\Delta m_{\rm irrev}$ rises suddenly when the reverse field reaches the nucleation field of hard phase associating with the irreversible reversal of hard phase. The reversible portion of demagnetization curve is different for $Pr_{12}Fe_{82}B_6$ and $Pr_8Fe_{87}B_5$ ribbons. For $Pr_8Fe_{87}B_5$ ribbons, $\Delta m_{\rm rev}$ is primarily derived from the reversal of soft phase. It rises with increasing reverse field and shows a peak when the applied field equals to the nucleation field of hard phase, due to the competition between the external field and IGEC. For $Pr_{12}Fe_{82}B_6$ ribbons, $\Delta m_{\rm rev}$ increases monotonically with the increase of external field.

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