AIP | Applied Physics Letters

Non-uniform magnetization reversal in nanocomposite magnets

Z. B. Li, M. Zhang, B. G. Shen, and J. R. Sun

Citation: Appl. Phys. Lett. **102**, 102405 (2013); doi: 10.1063/1.4795445
View online: http://dx.doi.org/10.1063/1.4795445
View Table of Contents: http://apl.aip.org/resource/1/APPLAB/v102/i10
Published by the American Institute of Physics.

Non-uniform magnetization reversal in nanocomposite magnets

Z. B. Li, M. Zhang, B. G. Shen,[a] and J. R. Sun
State Key Laboratory for Magnetism, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, People's Republic of China

(Received 23 December 2012; accepted 1 March 2013; published online 11 March 2013)

Magnetization reversal and exchange coupling are investigated in Pr-Fe-B melt-spun ribbons. In nanocomposite magnets, not only does the coercivity decrease but also magnetization reversal becomes more non-uniform in hard grains. The non-uniform magnetization reversal, resulting in a deterioration of squareness in hysteresis loop and a drop of the maximum Henkel plot value, mainly is caused by random arrangement of easy axes and intergranular soft regions among hard grains even with well exchange coupling between soft-hard grains in these ribbons. It is expected that the uniformity in magnetization reversal could be improved with the perfection of easy axes alignment in anisotropy nanocomposites. © *2013 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4795445]

Nanocomposite magnets consisting of a fine mixture with magnetically hard and soft phase have attracted many attentions because of their potential application in permanent magnets with energy product achieving to 1 MJ/m^3.[1] However, the energy product obtained experimentally in laboratory is far from that predicted theoretically.[2–5] The soft-phase provides high saturation magnetization, but effective anisotropy and coercivity decrease simultaneously.[1,6] The mechanism of exchange coupling between grains was extensively investigated theoretically and experimentally. The process of magnetization reversal was studied in many ways, from nucleation of reversed domains to self-pinning at the soft/hard grains interface.[7,8] The theoretical model and the attainable maximum energy product were questioned and reexamined.[9–13] It is widely accepted that the nanostructure, i.e., grain size and interfacial structure, and the arrangement of easy axes in hard grains are key factors in the improvement of magnetic properties.[14]

In nanocomposite magnets with an ideal microstructure, the volume fraction of soft phase reaches 90% and thus giant energy product is obtained theoretically.[1] However, in laboratory as the volume fraction of soft-phase increases to nearly 40%, coercivity reduces dramatically, and magnetic properties decrease simultaneously.[7] Even in the films with microstructure more closed to theoretical model, the content of soft-phase is limited.[15–17] Most of the previous works focused on soft-grain size and exchange coupling between soft-hard grains.[18,19] Actually, in nanocomposite magnets, the intergrain interaction involves exchange coupling not only between soft and hard grains but also between hard grains. In our work, with the increase of soft-phase content, the squareness of hysteresis loop deteriorates unfortunately, which has rarely attracted enough attention to be investigated deeply. For a thorough understanding of exchange-spring magnets, in this letter, we investigate the origin of the deterioration of squareness from the angles of irreversible magnetization reversal and exchange coupling between grains.

The precursor ingots Pr-Fe-B were prepared by arc melting method under an argon atmosphere. The ribbons were obtained directly by induction melting the ingot in a quartz tube and then ejecting the melt onto the surface of a rotating copper wheel. The surface velocity of copper wheel was varied in the range of 15–30 m/s in order to optimize the magnetic properties. The phase composition of the ribbons was examined by x-ray diffraction (XRD) using Cu Kα radiation. Magnetic measurements were performed using superconducting quantum interference device vibrating sample magnetometer (SQUID VSM) at temperature of 300 K.

Fig. 1 shows the X-ray diffraction patterns of optimally melt-spun ribbons with compositions varied from $Pr_{12}Fe_{82}B_6$ to $Pr_7Fe_{88.5}B_{4.5}$. XRD confirms that all samples contain isotropic $Pr_2Fe_{14}B$ structure phase. With a decrease of Pr atomic percent, the intensity of α-Fe structure diffraction peaks becomes stronger, which indicates that the content of α-Fe increases. The phase contents are calculated from the elemental composition assuming a two phase mixture of α-Fe and $Pr_2Fe_{14}B$ in these samples. In addition, via the room temperature saturation magnetizations the α-Fe contents can also be obtained. The saturation magnetization of sample is acquired by the formula $M = M_s(1 - \frac{A}{H} - \frac{B}{H^2})$, and the room temperature saturation magnetizations of α-Fe and $Pr_2Fe_{14}B$ are taken as 218 emu/g and 167 emu/g, receptively. The results for all samples with different compositions are summarized in Table I. The data obtained with the two

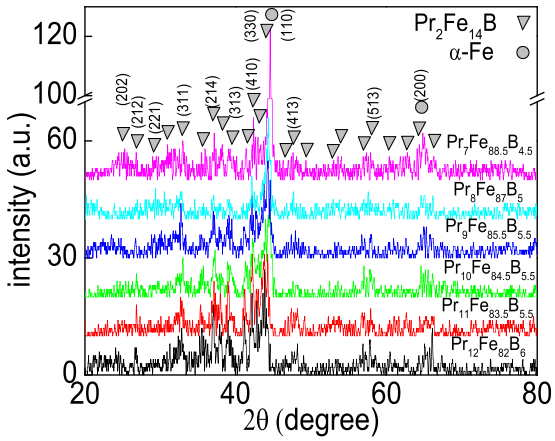

FIG. 1. The XRD patterns of all samples.

[a]Author to whom correspondence should be addressed. Electronic mail: shenbg@iphy.ac.cn.

TABLE I. Comparison of the weight fraction of α-Fe as determined by initial nominal stoichiometry and by calculation method of saturation magnetizations.

Composition	M_s (emu/g)	Theoretical content of α-Fe (wt. %)	Calculated content of α-Fe (wt. %)
$Pr_{12}Fe_{82}B_6 \approx Pr_2Fe_{14+\delta}B$ ($\delta = 0$)	165.2	0	0
$Pr_{11}Fe_{83.5}B_{5.5} \approx Pr_2Fe_{14+\delta}B$ ($\delta = 1.18$)	168.5	5.8	2.9
$Pr_{10}Fe_{84.5}B_{5.5} \approx Pr_2Fe_{14+\delta}B$ ($\delta = 2.9$)	172.4	13	10.6
$Pr_9Fe_{85.5}B_{5.5} \approx Pr_2Fe_{14+\delta}B$ ($\delta = 5$)	176.7	20.5	19
$Pr_8Fe_{87}B_5 \approx Pr_2Fe_{14+\delta}B$ ($\delta = 7.75$)	181.8	28.5	29
$Pr_7Fe_{88.5}B_{4.5} \approx Pr_2Fe_{14+\delta}B$ ($\delta = 11.3$)	187.5	36.7	40.2

methods are a little different, and this may be attributed to a little amount of residual amorphous phase at grain boundary, which is beneficial for the improvement of magnetic properties in nanocomposites.[20]

Fig. 2 shows the hysteresis loops of the optimally melt-spun ribbons with different Pr atomic percent. With an increase of α-Fe content (a decrease of Pr atomic percent), the coercivity decreases. The squareness of hysteresis loop is determined by the ratio of the integral $\int_0^{-H_c} M/M_s\, dH$ to $M_r/M_s * H_c$ (the product of remanence and coercivity) in the second quadrant. The dependence of squareness on the Pr atomic percent is shown in the inset of Fig. 2. With a decrease of Pr atomic percent the squareness deteriorates. Also explained that in all samples except $Pr_7Fe_{88.5}B_{4.5}$, soft grains are well exchange-coupled with hard grains because the squareness does not greatly decrease, and the coercivity keeps high among same kind of samples with same composition.[21]

The decrease of coercivity has been investigated theoretically and is basically attributed to the decrease in effective anisotropy in nanocomposites.[1,6] In nanocomposites with well exchange coupling between soft-hard grains, irreversible magnetization reversal originates from hard grains, and in soft grains the magnetization is more reversible. In order to further probe the potential origin of the squareness deterioration, we investigate irreversible components of magnetization. Fig. 3 shows irreversible susceptibility x_{irr}/M_s versus the reduced field H/H_c, which is obtained from the recoil loops measured at temperature of 300 K. With an increase of α-Fe content (a decrease in Pr atomic percent),

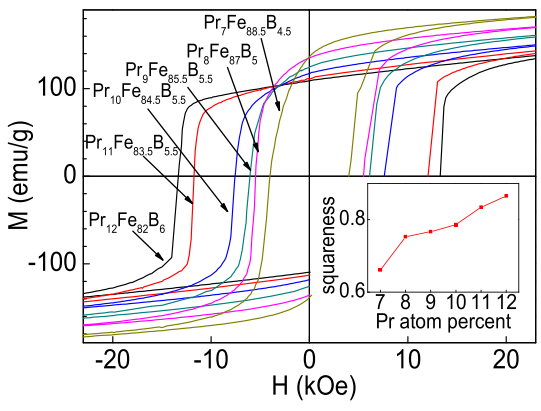

FIG. 2. The hysteresis loops of all samples. The inset shows the dependence of squareness on Pr atomic percent.

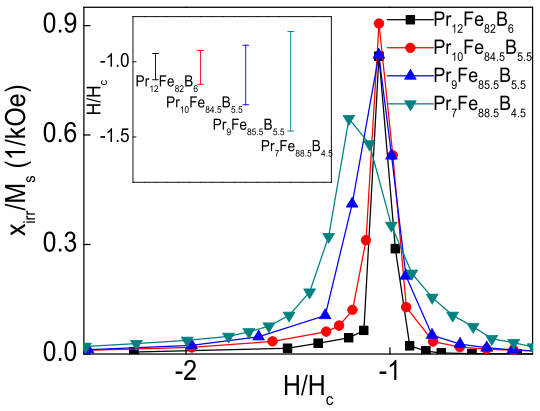

FIG. 3. The dependence of irreversible susceptibility on reduced field. The inset shows the main distribution of irreversible susceptibilities with different Pr atomic percent.

the main distribution of irreversible susceptibility (more than 0.2 times of the maximum irreversible susceptibility) is more decentralized on H/H_c (shown in the inset of Fig. 3), which indicates that magnetization reversal becomes more non-uniform. Without doubt, the non-uniform magnetization reversal results in the squareness deterioration of hysteresis loop, and the question arises, what causes the non-uniform magnetization reversal in hard grains.

In isotropic magnets each hard grain is different in effective anisotropy with easy axes oriented randomly. In nanocrystallines, it is that owning to exchange coupling between hard grains the process of magnetization reversal is cooperative and more uniform. So in nanocomposites, the non-uniform magnetization reversal may result from weak exchange coupling between grains.[1,22] In order to check the exchange coupling effect, Henkel plot is used, which is defined as $\delta m = [M_d(H) + 2M_r(H)]/M_r - 1$.[23,24] Here $M_d(H)$ is a remanence obtained after the application and subsequent removal of a field H in initial magnetization process in a fresh sample, $M_r(H)$ after saturation in one direction and then the subsequent application and removal of a field H in the reverse direction. Positive value of δm indicates that exchange coupling is dominant over the dipolar interaction. Fig. 4 shows the dependence of δm on H/H_c at temperature of 300 K for all optimally melt-spun samples. The plot of δm versus H/H_c shows a peak at the field around coercivity H_c. With a decrease in Pr atomic percent (an increase of

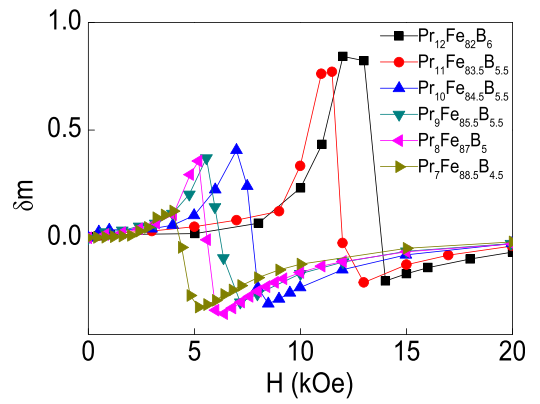

FIG. 4. δm curves (Henkel plots) of all samples.

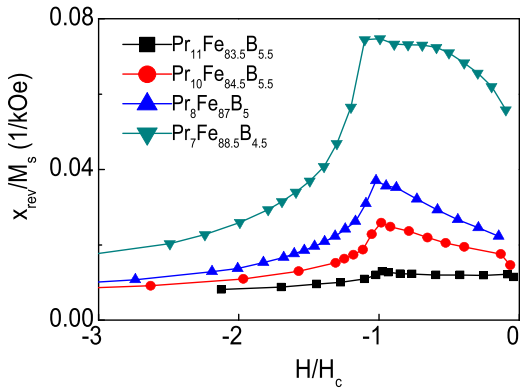

FIG. 5. The reversible susceptibility versus reduced field at temperature of 300 K.

soft-phase content), the maximum value of δm decreases. It demonstrates that the decrease of squareness and non-uniform magnetic reversal arise from the weakening of exchange coupling between grains in nanocomposite magnets.

In nanocomposites, exchange coupling exists not only between soft-hard grains but also between hard grains. It seems unreasonable to attribute the non-uniform magnetic reversal entirely to exchange coupling between soft-hard grains in these samples. In the formula of $\delta m = [M_d(H) + 2M_r(H)]/M_r - 1$, $M_r(H)$, and $M_d(H)$ are remanences that are irreversible components of magnetization. The irreversible magnetization arises from magnetization reversal in hard grains for well exchange coupling nanocomposites. More possibly, Henkel plots could check the effect of exchange coupling directly between hard grains in these samples. With these in mind, the non-uniform magnetization reversal and the deterioration of squareness seem to result from the weakening of exchange coupling between hard grains.

To get the knowledge about the weakening effect of exchange coupling between hard grains, we take soft regions as intergranular layers among hard grains. Zhang *et al.* presented that there is no change in the maximum value of δm in nanocrystallines with 3 nm width intergranular layers using simulation method.[25] This result nearly consists with that for $Pr_{11}Fe_{83.5}B_{5.5}$ ribbons, in which α-Fe content is a little and δm value has only a little drop compared with $Pr_{12}Fe_{82}B_6$ ribbons. However, the simulation for magnets with larger width layers has not been further developed. As soft-phase content increases, the width of some intergranular layers could be thought to increase simultaneously. As the maximum value of δm decreases greatly in nanocomposites with large soft-phase content, magnetization reversal is assured to be more independent among hard grains. With these in mind, the non-uniform magnetization reversal could be understood more easily. In isotropic nanocomposites, in hard grains with easy axis much deviation from the field direction the effective anisotropy decreases much, thus magnetization is more probably reversed prior to that with less deviation from the field direction. So magnetization reversal is more independent among hard grains as if exchange coupling between hard grains becomes weak, which results in a deterioration of squareness in hysteresis loop.

The magnetization reversal, involving irreversible and reversible components, depends in part on that of soft regions and exchange coupling between soft-hard grains.[22] Fig. 5 shows the dependence of reversible susceptibility x_{rev}/M_s on H/H_c at temperature of 300 K. With the increase of soft-phase content the reversible susceptibility increases whereas magnetization reversal becomes more non-uniform. Actually, in isotropic nanocomposite magnets with a variation of anisotropy, reversible magnetization involves not only a portion of soft phase but also of hard phase.[26,27] Most probably, owing to exchange coupling between soft-hard grains while soft grains have strong resistance against magnetization reversal, the coercivity in hard grains decreases and magnetization reversal of hard grains becomes more reversible rather than more dependent and uniform, especially in isotropic nanocomposites. Ideally, it can be argued that if each hard grain is identical in effective anisotropy with the easy axes aligned strictly along same direction, magnetization reversal would be uniform even with independent magnetic behavior among hard grains, so the squareness of hysteresis loop could be improved and giant energy product could be achieved.

In summary, in nanocomposite magnets, with an increase of soft-phase content, due to random arrangement of easy axes and intergranular soft layers among hard grains, magnetization reversal becomes much non-uniform, irreversible components is distributed in a more scatter range, and the maximum value of δm drops, which result from the less dependent magnetization behavior among hard grains. It strongly suggests that the improvement in easy axes alignment and the thickness reduction in soft regions are necessary for further improvement of magnetic properties in nanocomposite magnets.

The present work was supported by the Knowledge Innovation Project of the Chinese Academy of Sciences.

[1]R. Skomski and J. M. D. Coey, Phys. Rev. B **48**, 15812 (1993).
[2]A. M. Gabay, Y. Zhang, and G. C. Hadjipanayis, Appl. Phys. Lett. **85**, 446 (2004).
[3]H. Zeng, J. Li, J. P. Liu, Z. L. Wang, and S. Sun, Nature (London) **420**, 395 (2002).
[4]V. V. Nguyen, C. Rong, Y. Ding, and J. P. Liu, J. Appl. Phys. **111**, 07A731 (2012).
[5]R. Chen, S. Guo, J. Wang, D. Lee, and A. Yan, J. Appl. Phys. **109**, 07A756 (2011).
[6]R. W. Gao, W. C. Feng, H. Q. Liu, B. Wang, W. Chen, G. B. Han, P. Zhang, H. Li, W. Li, Y. Q. Guo, W. Pan, X. M. Li, M. G. Zhu, and X. Li, J. Appl. Phys. **94**, 664 (2003).
[7]J. Bauer, M. Seeger, A. Zern, and H. Kronmüller, J. Appl. Phys. **80**, 1667 (1996).
[8]G. P. Zhao, X. L. Wang, C. Yang, L. H. Xie, and G. Zhou, J. Appl. Phys. **101**, 09K102 (2007).
[9]H. Fukunaga and H. Mukaino, J. Rare Earth **22**, 146 (2004).
[10]M. K. Griffiths, J. E. L. Bishop, J. W. Tucker, and H. A. Davies, J. Magn. Magn. Mater. **234**, 331 (2001).
[11]H. Fukunaga and H. Nakamura, Scr. Mater. **44**, 1341 (2001).
[12]C. B. Rong, H. W. Zhang, R. J. Chen, S. L. He, and B. G. Shen, J. Magn. Magn. Mater. **302**, 126 (2006).
[13]T. Schrefl, J. Fidler, and H. Kronmüller, Phys. Rev. B **49**, 6100 (1994).
[14]N. Jones, Nature (London) **472**, 22 (2011).
[15]W. Liu, Z. D. Zhang, J. P. Liu, L. J. Chen, L. L. He, Y. Liu, X. K. Sun, and D. J. Sellmyer, Adv. Mater. **14**, 1832 (2002).
[16]J. Zhang, Y. K. Takahashi, R. Gopalan, and K. Hono, Appl. Phys. Lett. **86**, 122509 (2005).

[17]H. Kato, T. Nomura, M. Ishizone, H. Kubota, T. Miyazaki, and M. Motokawa, J. Appl. Phys. **87**, 6125 (2000).
[18]Z. Q. Jin, H. Okumura, Y. Zhang, H. L. Wang, J. S. Muñoz, and G. C. Hadjipanayis, J. Magn. Magn. Mater. **248**, 216 (2002).
[19]J. E. Shield, Y. Liu, R. Marr, Z. Chen, and B. M. Ma, IEEE Trans. Magn. **40**, 2901 (2004).
[20]X. Y. Zhang, Y. Guan, and J. W. Zhang, Appl. Phys. Lett. **80**, 1966 (2002).
[21]D. Goll, M. Seeger, and H. Kronmüller, J. Magn. Magn. Mater. **185**, 49 (1998).
[22]E. F. Kneller and R. Hawig, IEEE Trans. Magn. **27**, 3588 (1991).
[23]O. Henkel, Phys. Status Solidi **7**, 919 (1964).
[24]P. E. Kelly, K. O'Grady, P. I. Mayo, and R. W. Chantrell, IEEE Trans. Magn. **25**, 3881 (1989).
[25]H. W. Zhang, C. B. Rong, X. B. Du, J. Zhang, S. Y. Zhang, and B. G. Shen, Appl. Phys. Lett. **82**, 4098 (2003).
[26]Y. Choi, J. S. Jiang, J. E. Pearson, S. D. Bader, and J. P. Liu, Appl. Phys. Lett. **91**, 022502 (2007).
[27]Z. B. Li, B. G. Shen, and J. R. Sun, J. Appl. Phys. **113**, 013902 (2013).