AIP Journal of Applied Physics

Origin of recoil hysteresis in nanocomposite Pr8Fe87B5 magnets

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

Citation: J. Appl. Phys. **113**, 013902 (2013); doi: 10.1063/1.4772606
View online: http://dx.doi.org/10.1063/1.4772606
View Table of Contents: http://jap.aip.org/resource/1/JAPIAU/v113/i1
Published by the American Institute of Physics.

JOURNAL OF APPLIED PHYSICS **113**, 013902 (2013)

Origin of recoil hysteresis in nanocomposite $Pr_8Fe_{87}B_5$ magnets

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

(Received 27 July 2012; accepted 29 November 2012; published online 2 January 2013)

The physical mechanism of recoil hysteresis loops has been subject to dispute in recent years. This paper deals with the origin of recoil hysteresis in nanocomposite $Pr_8Fe_{87}B_5$ magnets. It is shown by investigating the recoil loops and magnetic activation of melt-spun ribbon of $Pr_8Fe_{87}B_5$ at different temperatures that the openness of recoil hysteresis loops is strongly dependent on the magnetic activation as the negative field (opposite to magnetization) is cycled to zero in a remanent state. The hysteresis of recoil loops originates from the reversible magnetization reversal of hard phase, which is probably caused by the energy of isotropic exchange coupling overcoming the energy barrier of magnetocrystalline anisotropy. So, the hysteresis is codetermined by the exchange coupling and variation of anisotropy, thus the openness has a linear relationship with the product of magnetic moments of activation and reversible susceptibility. © *2013 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4772606]

I. INTRODUCTION

Nanocomposite magnets with exchange coupling between hard and soft magnetic phases may be a potential candidate for high energy product permanent magnets and ultrahigh-density recording media.[1–4] Measuring the recoil loops by cycling the field to zero and back at a number of negative field values could characterize the strength of exchange coupling and the distribution of effective anisotropy. Descending and ascending hysteresis branches along the recoil loops usually do not exactly overlap, which can be quantified by the loop openness. The openness can be small or even absent in a single phase nanocrystalline magnet, and big for a hard/soft nanocomposite magnet. It was thought that the hysteresis of recoil loops is indicative of breakdown of exchange coupling between soft phase and hard phase, and the openness is associated with the decoupled volume in the soft phase.[5–8] Since the openness was found to be present both in hard phase and in soft phase by element-specific recoil loop measurements recently,[9] the recoil hysteresis loops and its physical origin have re-attracted much attention and subsequently given rise to dispute. The experiments and simulations demonstrated that the recoil hysteresis loops results from or strongly depends on the anisotropy variation in hard phase,[9] unstable magnetic moments affected by thermal fluctuation,[10] strong intergrain exchange coupling,[11] and unstable magnetization behavior in grain boundary and soft-phase regions.[12] Different experiments and simulations lead to different views on the origin of recoil hysteresis loops. It is worth further ascertaining, which one of the views is the ultimate cause and the physical origin of recoil hysteresis loops.

In the present work, we measured the recoil loops and magnetic moments of activation of $Pr_8Fe_{87}B_5$ melt-spun ribbon at the temperatures of 300 K, 180 K, 60 K, and 10 K. With the variation of temperature, the strength of exchange coupling, the irreversible susceptibility, and openness of recoil loops are changed. The openness is strongly dependent on the activated magnetization reversal as the negative field is cycled to zero in a remanent state. These give a way to investigate the physical origin of recoil hysteresis in nanocomposite magnets.

II. EXPERIMENT

Nanocomposite structure $Pr_2Fe_{14}B/\alpha$-Fe ribbons with the nominal composition of $Pr_8Fe_{87}B_5$ were obtained by direct melt spinning method with optimal quenching rate, 22 m/s of copper wheel speed. X-ray diffraction confirms that the samples contain $Pr_2Fe_{14}B$ and α-Fe structure phases. According to the Scherrer method, the mean grain sizes of $Pr_2Fe_{14}B$ and α-Fe phases are 20.9 nm and 16.5 nm, respectively. The magnetic recoil loops were measured with superconducting quantum interference device (SQUID) VSM at the temperatures of 300 K, 180 K, 60 K, and 10 K. The magnetic moments of activation were measured under the applied field increased from the state of remanence and under the field cycled to zero, respectively.

III. RESULTS AND DISCUSSION

Figures 1(a)–1(c) show the recoil loops of sample measured at a field ramp rate of 50 Oe/s at the temperatures of 300 K, 180 K, and 60 K, respectively. The measurement process at points a, b, c, and d in recoil loops is shown in Fig. 1(b). As illustrated in Fig. 1(b), ΔM_{open} refers to the maximum of openness and H_R denotes the field of start of each recoil loop. As temperature decreases, the coercivity of sample increases, however, the exchange coupling does not keep up with the increase of coercivity and the squareness of demagnetization curve decreases.[13] Figure 2 shows the hysteresis loops measured at different ramp rates of field at the temperatures of 300 K and 60 K. The opennesses measured at field ramp rates of 400 Oe/s and 200 Oe/s are greater than those measured at field ramp rates of 50 Oe/s and 5 Oe/s, which indicates that the openness is time dependent. However,

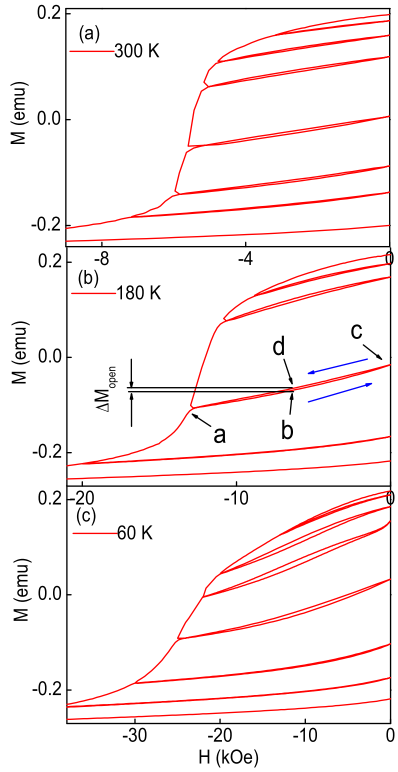

FIG. 1. Recoil loops measured at the temperatures of 300 K (a), 180 K (b), and 60 K (c).

the opennesses measured at 50 Oe/s and 5 Oe/s are not much different, and the hysteresis of recoil loop is stable. These indicate that even though the openness is time dependent, the field ramp rate and time are not the fundamental cause of the hysteresis of recoil loop.

Figure 3 shows the dependences of openness ΔM_{open}, irreversible susceptibility x_{irr}, and reversible susceptibility x_{rev} on recoil field H_R of each pair of loops. The irreversible susceptibility reflects the distribution of coercivity and effective anisotropy of hard phase, while the reversible susceptibility reflects the strength of exchange coupling between grains. In Fig. 3(a), the values of field H_R corresponding to maxima of irreversible susceptibility and reversible susceptibility are the same, which indicates the effect of exchange coupling is strong and compatible with the effective anisotropy of hard phase at the temperature of 300 K. The corresponding field H_R of maximum openness is the same as those of irreversible susceptibility and reversible susceptibility. However, with the decrease of temperature, the corresponding field H_R of

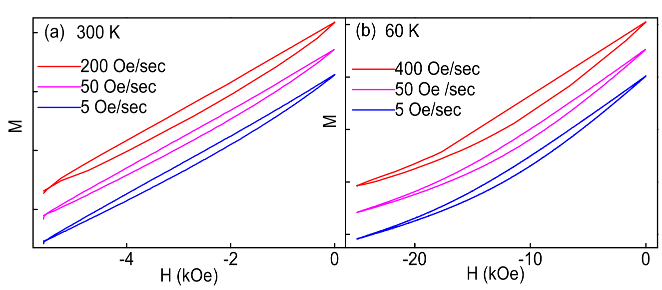

FIG. 2. Hysteresis loops measured at different ramp rates of field at the temperatures of 300 K (a) and 60 K (b).

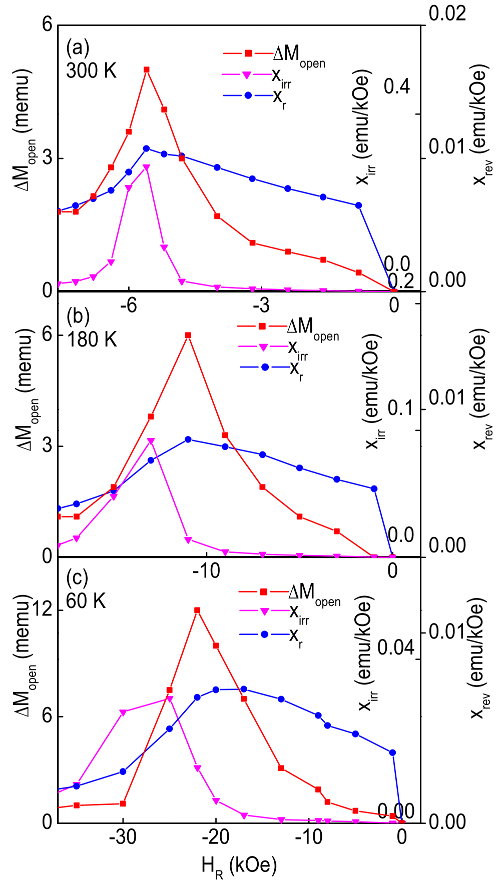

FIG. 3. Dependences of openness, irreversible susceptibility, and reversible susceptibility on field H_R at the temperatures of 300 K (a), 180 K (b), and 60 K (c).

maximum openness is different from that of irreversible susceptibility or reversible susceptibility. It seems that neither the anisotropy nor the effect of exchange coupling alone determines the hysteresis of recoil loops. As seen in Fig. 3, at the beginning of recoil loop family measurement (H_R close to 0), reversible susceptibility x_{rev} dominates, most of which is believed to be ascribed to the soft phase, however, ΔM_{open} is nearly zero. So it is not possible that the reversible magnetization of soft phase results in the hysteresis of recoil loops.

In order to probe clearly the physical origin of openness, we measured the magnetic moments of activation ΔM_{act-a} under the field kept fixed at H_R for 300 s, after a sweep from zero at remanence to H_R, and we also measure ΔM_{act-c} under field kept fixed at zero for 300 s, after cycling it back from H_R to zero. Figure 4 shows the magnetic activation with the field swept from 0 to -5.6 kOe and with the field cycled from -5.6 to 0 kOe at the temperature of 300 K. Beside, there is a thermally activated magnetization reversal as the field swept from 0 to -5.6 kOe due to the irreversible magnetization,[14,15] the reversible magnetization reversal of thermal activation is found as the field is cycled to 0 kOe, as shown in Fig. 4(b).

Figure 5 shows the dependences of magnetic moments of activation and openness on field H_R. With the increase (decrease) of ΔM_{act-c}, the openness increases (decreases) simultaneously. Obviously, the openness is strongly dependent

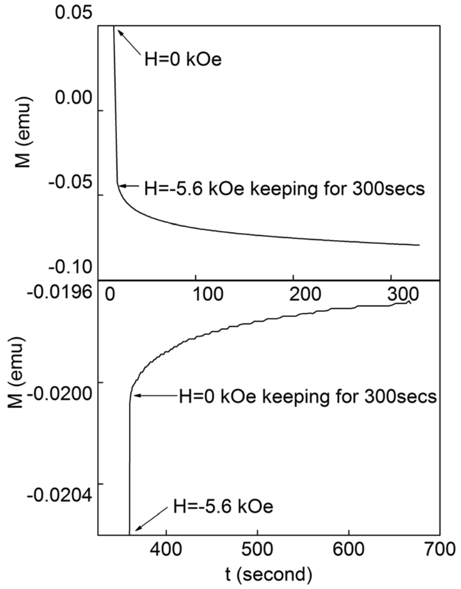

FIG. 4. Magnetic activation (a) with the field swept from 0 to −5.6 kOe and (b) with the field cycled from −5.6 to 0 kOe at the temperature of 300 K.

on ΔM_{act-c} rather than ΔM_{act-a}. The magnetic viscosity is obtained from the time dependent magnetization under field kept fixed for a time t and obeys the equation, $M_{(t)} = M_0 - S\ln(t_0 + t)$, where M_0 and t_0 are the constants, and $\Delta M_{act} = M_{(t)} - M_{(0)}$.[14,15] The magnetic moments of activation and magnetic viscosity arise from the thermal activation of irreversible magnetization for overcoming energy barriers

$$S = x_{irr}H_f,$$

where H_f is the fluctuation field.[14] The irreversible magnetization originates from the magnetization reversal of hard phase overcoming the energy barrier of anisotropy in well exchange-coupled nanocomposite magnets.[1,3] So the openness results from the reversal of magnetic moments of some hard grains switching from the negative magnetoanisotropy direction to positive direction as the field is cycled between the negative field H_R and zero. These consist with the finding that reversible magnetization involves a portion of the hard phase.[9,16] It would be reasonable and more accurate to say that the magnetic viscosity arises from the thermal activation overcoming the anisotropic energy barrier rather than irreversible magnetization.

What results in the activated magnetization reversal of hard phase as the field is cycled between the negative field H_R and zero? Figure 6 shows the dependences of openness on $\Delta M_{act-c} * x_{rev}$. At all the temperatures of 300 K, 180 K, and 60 K, there are linear relationships between the openness and the product of magnetic moments of activation and reversible susceptibility. The reversible susceptibility reflects the strength of exchange-spring between grains. It is probable that the effect of exchange coupling plays an important role in the reversible magnetization reversal of hard phase. As the field is cycled from H_R to zero at remanence, because of the driving of exchange coupling with isotropic nature between grains, the un-reversed magnetic moments of grains pull those revered and make some of them overcome the stumbling block of the energy barrier of anisotropy and turn toward the same direction with (or without) the help of thermal fluctuation field. The probability that the magnetic moments of hard grains switch from the state at point b to the state at point d after a time is given by $P(t) = 1 - \exp(-t/\tau)$,[17] where τ obeys the Arrhenius law. $\tau = \tau_0\exp(\Delta E/(k_BT))$, where τ_0 is a pre-exponential factor on the order of 10^{-9} (s), which is corresponding to the Larmor frequency. ΔE is an energy barrier, the sum of anisotropy, exchange coupling, external field, and dipolar energy. So the openness of recoil loops is time (field ramp rate) and temperature dependent (thermal fluctuation).[10,12] The magnetization reversal of some soft grains follows that of hard grains, so the openness of recoil loops is amplified by the soft phase.[11]

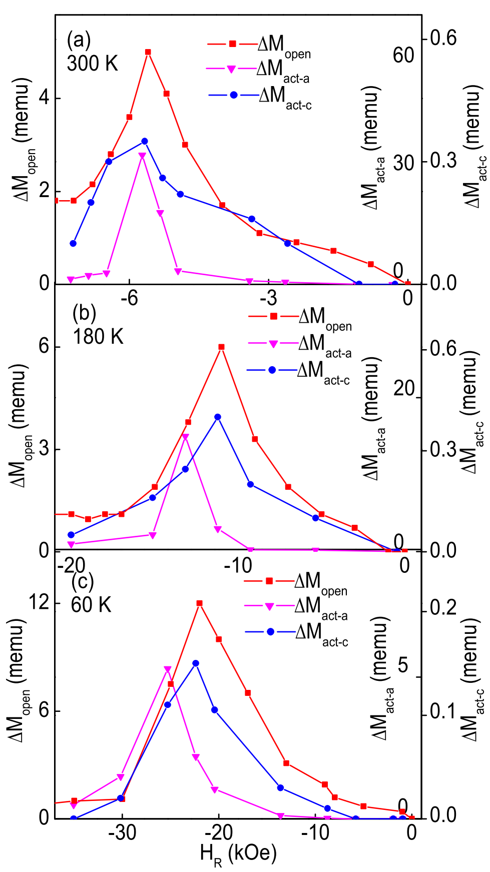

FIG. 5. Dependences of magnetic moments of activation and openness on field H_R at the temperatures of 300 K (a), 180 K (b), and 60 K (c).

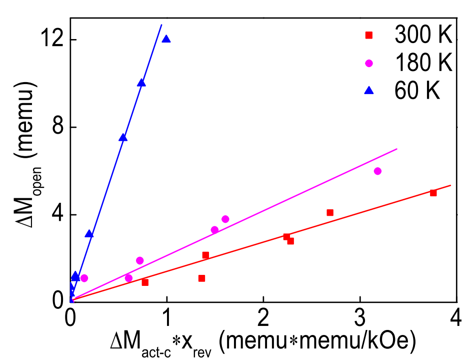

FIG. 6. Dependences of openness on the product of magnetic moments of activation and reversible susceptibility $\Delta M_{act-c} * x_{rev}$ at the temperatures of 300 K (a), 180 K (b), and 60 K (c).

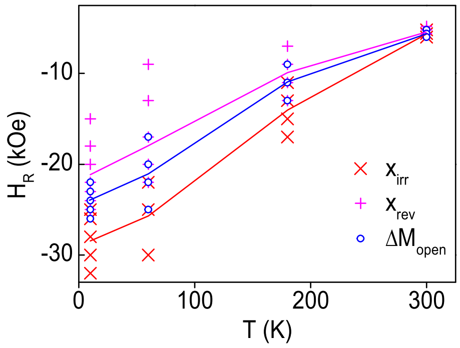

FIG. 7. Distributions of corresponding field H_R of ΔM_{open}, x_{irr}, and x_{rev} with higher values at different temperatures. The solid lines are fitted lines for those of openness, irreversible susceptibility, and reversible susceptibility with the variation of temperature.

The hysteresis of recoil loops originates from the magnetization reversal of hard phase as the field is cycled to zero, which is probably caused by the isotropic exchange coupling overcoming the energy of magnetoanisotropy. Field ramp rate and temperature have influence on the openness. However, they are not the root cause of hysteresis. ΔE is an energy barrier, the sum of magnetoanisotropy, exchange coupling, and external field with neglecting the dipolar energy. So, it is possible that the magnetization reversal reflects the competition between exchange coupling and anisotropy energy under the external field. Figure 7 shows the distributions of corresponding field H_R of ΔM_{open}, x_{irr}, and x_{rev} with higher values at different temperatures. At the temperature of 300 K, the exchange coupling between grains is strong, so the distributions of corresponding field H_R of ΔM_{open}, x_{irr}, and x_{rev} with higher values are nearly the same. With the decrease of temperature, the coercivity becomes gradually greater than the strength of exchange coupling, and the distribution of corresponding field H_R of openness is between those of irreversible susceptibility and reversible susceptibility. The openness seems to be a compromise between the strength of exchange coupling and the anisotropy energy, so it is possibly the result of competition among these types of energies.

IV. CONCLUSIONS

In this paper, the recoil loops and magnetic moments of activation in nanocomposite $Pr_8Fe_{87}B_5$ magnets have been measured at different temperatures. The thermally activated magnetization reversal is found as the negative field is cycled to zero at remanence, on which the openness of recoil hysteresis loops is strongly dependent. The hysteresis of recoil loops originates from the reversible magnetization reversal of hard phase, which is probably caused by the exchange coupling overcoming the energy barrier of magnetoanisotropy. So, the hysteresis is codetermined by the exchange coupling and the variation of anisotropy, and the openness has a linear relationship with the product of magnetic moments of activation and reversible susceptibility. The magnetization reversal of some soft grains follows that of hard grains, so the openness of recoil loops is amplified by the soft phase.

ACKNOWLEDGEMENTS

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]S. D. Bader, Rev. Mod. Phys. **78**, 1–15 (2006).
[3]H. Zeng, J. Li, J. P. Liu, Z. L. Wang, and S. Sun, Nature **420**, 395–398 (2002).
[4]E. E. Fullerton, J. S. Jiang, C. H. Sowers, J. E. Pearson, and S. D. Bader, Appl. Phys. Lett. **72**, 380 (1998).
[5]K. Kang, L. H. Lewis, J. S. Jiang, and S. D. Bader, J. Appl. Phys. **98**, 113906 (2005).
[6]C. L. Harland, L. H. Lewis, Z. Chen, and B. M. Ma, J. Magn. Magn. Mater. **271**, 53 (2004).
[7]D. Goll, M. Seeger, and H. Kronmüller, J. Magn. Magn. Mater. **185**, 49 (1998).
[8]D. C. Crew, J. Kim, L. H. Lewis, and K. Barmak, J. Magn. Magn. Mater. **233**, 257 (2001).
[9]Y. Choi, J. S. Jiang, J. E. Pearson, S. D. Bader, and J. P. Liu, Appl. Phys. Lett. **91**, 022502 (2007).
[10]C. Rong, Y. Liu, and J. P. Liu, Appl. Phys. Lett. **93**, 042508 (2008).
[11]B. Zheng, H. W. Zhang, S. Zhao, J. Chen, and G. Wu, Appl. Phys. Lett. **93**, 182503 (2008).
[12]C. Rong and J. P. Liu, Appl. Phys. Lett. **94**, 172510 (2009).
[13]H. W. Zhang, C. B. Rong, X. B. Du, S. Y. Zhang, and B. G. Shen, J. Magn. Magn. Mater. **278**, 127 (2004).
[14]E. P. Wohlfarth, J. Phys. F: Met. Phys. **14**, L155–L159 (1984).
[15]N. H. Hai, N. Chau, D. Ngo, and D. T. H. Gam, J. Magn. Magn. Mater. **323**, 3156 (2011).
[16]K. V. O'Donovan, J. A. Borchers, C. F. Majkrzak, O. Hellwig, and E. E. Fullerton, Phys Rev. Lett. **88**, 067201 (2002).
[17]H. Kronmüller and M. Fahnle, *Micromagnetism and the Microstructure of Ferromagnetic Solids* (Cambridge University Press, 2003), p. 420.