APPLIED PHYSICS LETTERS **88**, 042504 (2006)

Mechanism of the anomalous temperature dependence of coercivity in Sm(Co,Fe,Cu,Zr)$_z$ high-temperature magnets

Chuan-bing Rong[a)]
State Key Laboratory of Magnetism, Institute of Physics and Center for Condensed Matter Physics, Chinese Academy of Sciences, Beijing 100080, China and Department of Physics, University of Texas at Arlington, Arlington, Texas 76019

Hong-wei Zhang and Bao-gen Shen
State Key Laboratory of Magnetism, Institute of Physics and Center for Condensed Matter Physics, Chinese Academy of Sciences, Beijing 100080, China

J. Ping Liu
Department of Physics, University of Texas at Arlington, Arlington, Texas 76019

(Received 19 September 2005; accepted 18 December 2005; published online 24 January 2006)

Coercivity mechanism of the precipitation-hardened Sm(Co,Fe,Cu,Zr)$_z$ high-temperature magnets was found to vary with temperature by measurements of reversible magnetization. The magnetization reversal is mainly controlled by domain-wall pinning when temperature is lower than 800 K, while is mainly dominated by nucleation at higher temperature. It is interesting to find that the anomalous temperature dependence of coercivity $H_c(T)$ near Curie temperature of the 1:5 phase is caused by the fast drop of intergranular exchange coupling. This mechanism can also explain the coercivity behavior that the anomalous $H_c(T)$ tends to disappear with increasing z value. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2167795]

Recently, the SmCo-based permanent magnets have attracted considerable attention due to the need for developing magnets for high-temperature applications. Moreover, the anomalous temperature dependence of the coercivity $H_c(T)$, i.e., a positive temperature coefficient in a certain temperature range, is found in the precipitation-hardened Sm(Co,Cu,Fe,Zr)$_z$ magnets.[1–7] The high coercivity in these magnets originates from a complex microstructure consisting of a superposition of cellular and a lamellar structure.[8–10] Although much effort has been paid to understand the anomalous coercivity, up until now the mechanism of the anomalous $H_c(T)$ in Sm(Co,Cu,Fe,Zr)$_z$ magnets is still the subject of debate. The major proposal is domain-wall pinning mechanism which suggests that H_c should be proportional to the difference of the domain-wall energy density between the 2:17 and 1:5 phases.[2–4,11] Another point is that the anomalous $H_c(T)$ is caused by a temperature-induced transition from a repulsive to an attractive pinning of domain wall and then to the nucleation of reversed domain at the temperature higher than Curie temperature of 1:5 phase ($T_C^{1:5}$).[1,12] Due to the difficulty in explaining the dependence of coercivity on the Cu content, Gabay *et al.*[5,6] suggested that $H_c(T)$ is exclusively controlled by the nucleation of reversed domain. However, there still are some unclear coercivity behavior in these precipitation-hardened Sm(Co,Cu,Fe,Zr)$_z$ magnets using the above models. For example, the tendency for the anomalous $H_c(T)$ decreases with increasing z value.

In this work, measurements of reversible and irreversible magnetization have been used to examine the coercivity mechanism of the Sm(Co,Cu,Fe,Zr)$_z$ magnets magnets at different temperature. It is interesting to find that the coercivity mechanism varies with temperature. Being different from point of views of domain-wall pinning and nucleation mechanisms, we found that the anomalous $H_c(T)$ is caused by the fast drop of exchange coupling interaction near the temperature of $T_c^{1:5}$.

Ingots with nominal composition Sm(Co$_{\mathrm{bal}}$Cu$_{0.1}$Fe$_{0.1}$Zr$_{0.03}$)$_z$ with z=6.5, 7.0, and 7.5 were prepared by arc-melting technique. The ribbons were obtained using the melt-spinning technique with the surface speeds of the Cu wheel about 15 m/s. Subsequently, the samples were isothermally annealed at 1123 K for 3 h, followed by a slow cooling (rate of 1 K/min) to 673 K and then quenched to room temperature (RT). Crystalline structure was determined by x-ray diffraction (XRD) with Cu $K\alpha$ radiation. XRD analysis shows that all the ribbons are isotropic. A single TbCu$_7$-type phase is obtained in the as-spun ribbons and is further segregated into 1:5 and 2:17 phases after precipitation hardening. The magnetic measurements were performed in a superconducting quantum interference device magnetometer and a Lakeshore vibrating sample magnetometer with maximum applied field of 5 and 2.4 T, respectively. The Curie temperatures of 1:5 and 2:17 phases ($T_c^{1:5}$ and $T_c^{2:17}$) are obtained from the measured thermomagnetic curves. The determined $T_c^{1:5}$ of the samples with z=6.5–7.5 are 940, 900, and 870 K, respectively. $T_c^{2:17}$ is fixed at 1120 K for different z value.

Figure 1 shows the $H_c(T)$ curves of the Sm(Co,Cu,Fe,Zr)$_z$ magnets. It shows an maximum at temperature $T_{\max}$=800 K for the sample with z=6.5. This anomaly of coercivity disappears in the samples with z=7.0 and 7.5. It is interesting to note that although the shape of $H_c(T)$ curve varies with the increase of z value, the coercivity almost converges at the same values above 800 K. According to either pinning or nucleation mechanism,[5,11] $T_{\max}$ should be equal to $T_c^{1:5}$. To study the relation between them, $T_c^{1:5}$ of the sample with z=6.5 is indicated by arrows in Fig. 1. As can be seen clearly that $T_{\max}$ is far lower than $T_c^{1:5}$.

[a)] Electronic mail: crong@uta.edu

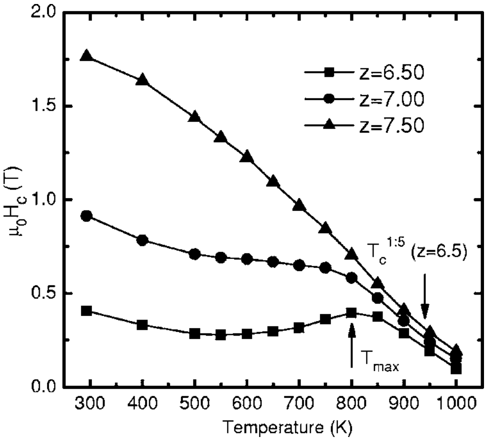

FIG. 1. $H_c(T)$ curves of the precipitation-hardened Sm(Co,Fe,Cu,Zr)$_z$ magnets.

Earlier studies showed that the Cu content in the center of cell boundary is far higher than that of the 1:5/2:17 interface in the precipitation-hardened magnets.[3] This should lead to the wide distribution of $T_c^{1:5}$ because it depends sensitively on the Cu content.[13] Thus, $T_{\max}$ may be strongly related to $T_c^{1:5}$ of the center of the cell boundary, which is smaller than the $T_c^{1:5}$ determined by the thermomagnetic analysis. This cause the large difference between $T_{\max}$ and $T_c^{1:5}$.

It was proposed by Crew *et al.*[14,15] that the measurements of reversible magnetization J_{rev} and its dependence on irreversible magnetization J_{irr} could be used to distinguish the magnetization reversal of domain-wall pinning and nucleation of reversed domain. Figure 2 shows the reduced reversible magnetization m_{rev} $(=J_{\text{rev}}/J_r)$ versus reduced irreversible magnetization m_{irr} $(=J_{\text{irr}}/J_r)$ of the Sm(Co,Fe,Cu,Zr)$_{6.5}$ magnet at different temperatures. The curves display a behavior characteristic of domain wall pinning at temperature below 800 K. At low field, m_{rev}, which is proportional to the amount of domain-wall bowing, increases due to the fast increases of the domain walls nucleate and the domain-wall area. After a certain stage in reversal, the total area of domain walls reaches a maximum and m_{rev} decreases as reversal proceeds further. Thus, the m_{rev} versus m_{irr} curves exhibit a minimum and are very similar to the results of Crew *et al.*[14] The pinning coercivity mechanism at RT is also proven by the angular dependence of coercivity of other recent works.[16,17] It is interesting to note that the value of the minimum m_{rev} decreases with the increase in temperature. This means that the pinning field in the Sm(Co,Cu,Fe,Zr)$_z$ magnets weakens with elevating temperature. By further increasing the temperature above 800 K, m_{rev} decreases linearly with increasing m_{irr}. This suggests that the reversal mechanism at high temperature is primarily associated with nucleation, and domain walls play a small part in the reversible magnetization component. The transition of coercivity mechanism from domain-wall pinning to reversed-domain nucleation supports the the angular coercivity analysis reported in Ref. 16.

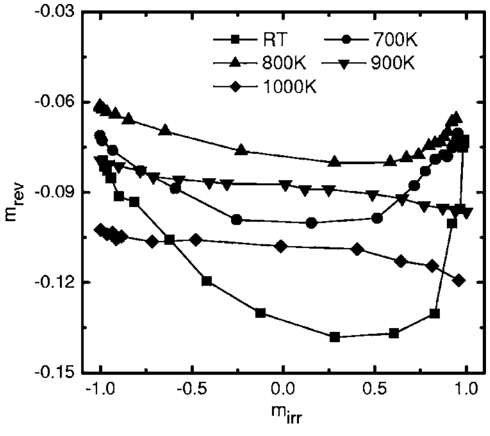

FIG. 2. m_{rev} vs m_{irr} of the Sm(Co,Fe,Cu,Zr)$_{6.5}$ magnets at different temperatures.

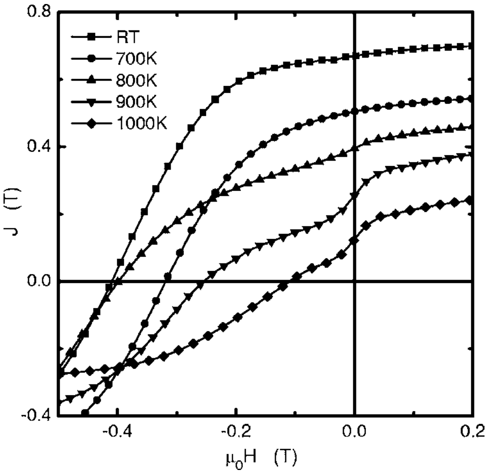

FIG. 3. The demagnetization curves of the Sm(Co,Fe,Cu,Zr)$_{6.5}$ magnet at different temperatures.

Although the anomalous $H_c(T)$ can be explained by the gradual change of the magnetization reversal mechanism from pinning to nucleation, the reason why the anomalous $H_c(T)$ disappears by increasing the z value is still unclear.

Figure 3 shows the demagnetization curves of the Sm(Co,Fe,Cu,Zr)$_{6.5}$ sample at different temperatures. The demagnetization curves show a good single-phase magnetic behavior at $T < 800$ K. However, an two-step demagnetization behavior appears on the demagnetization curves at $T \geqslant 800$ K. Since the exchange constant of 1:5 phase drops quickly as the temperature approaches $T_c^{1:5}$, the intergranular exchange coupling (IGEC) between the 1:5 and 2:17 phases decreases quickly, as well. This is proven by the $\delta m(H)$ plots of the Sm(Co,Fe,Cu,Zr)$_{6.5}$ sample at different temperatures as given in Fig. 4. It can be seen that the maximum positive δm values decrease with increasing temperatures, which means the fast drop of IGEC in the magnets.[18] When temperature is above 900 K, IGEC is very low. As a result, the 1:5 phase can not be exchange hardened completely by the 2:17 cellular phase at high temperature. This is the reason for the appearance of turning point on the demagnetization curves.

Since Sm(Co,Fe,Cu,Zr)$_z$ magnets have a special cellular microstructure, i.e., the 2:17 cells are surrounded by the 1:5 cell boundary, the coercivity of the Sm(Co,Fe,Cu,Zr)$_z$ magnets can be described by the theory of single-phase permanent magnets if the 1:5 cell boundary phase is considered as the defects of the 2:17 matrix phase. That is to say, the coercivity of Sm(Co,Fe,Cu,Zr)$_z$ magnets can also be described by a phenomenological expression[19,20]

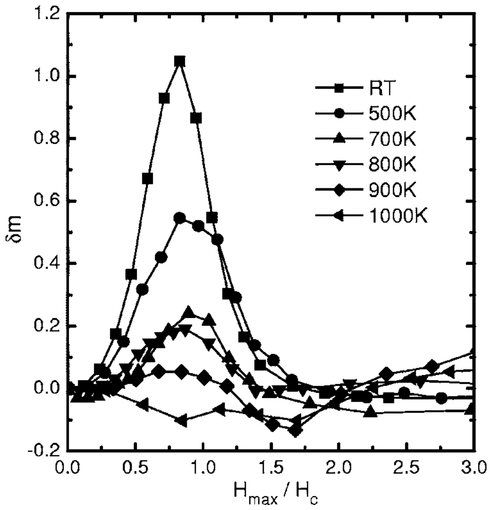

FIG. 4. $\delta m(H)$ of the $\mathrm{Sm(Co,Fe,Cu,Zr)}_{6.5}$ magnet at different temperatures.

$$\mu_0 H_c = \alpha_{\mathrm{ex}} \alpha_k \mu_0 H_a - N_{\mathrm{eff}} J_s, \qquad (1)$$

where α_k is the real-structure-dependent Kronmuller parameter and N_{eff} is a magnetostatic interaction parameter. α_{ex} describes the deleterious effect of exchange coupling between neighboring grains on the coercivity. In general, α_{ex} is equal to 1.0 for the exchange decoupled magnets, while it decreases with the increase in IGEC. For $\mathrm{Sm(Co,Fe,Cu,Zr)}_z$ magnets, α_k depends only weakly on temperature since the cellular microstructure of the precipitation-hardened magnets does not change with temperature. On the contrary, α_{ex} is very low at RT due to the strong IGEC as proven in Fig. 4 and is approximately equal to 1.0 above $T_c^{1:5}$ since the 2:17 cells are completely isolated by the 1:5 boundaries. Combined with the monotonous decrease of anisotropy field, the temperature dependence of coercivity is the competitive result of the temperature dependences of microstructure parameter $\alpha_{\mathrm{ex}}(T)$ and anisotropy field $H_a(T)$. To investigate the influence of temperature on $\alpha_{\mathrm{ex}}(T)$ quantitatively, Fig. 5 gives the temperature dependence of remanence ratio m_r($=J_r/J_s$) of the $\mathrm{Sm(Co,Fe,Cu,Zr)}_z$ magnets. Generally, the larger m_r means the stronger IGEC in a material. As shown in Fig. 5, m_r is far larger than the Stoner–Wohlfarth theoretical value of one-half at low temperature,[21] while decreases dramatically near $T_c^{1:5}$ in all samples. This proves that IGEC decreases quickly at temperatures near $T_c^{1:5}$. Thus, α_{ex} increases fast due to the sharp drop of IGEC. If the magnitude of the increase in $\alpha_{\mathrm{ex}}(T)$ is faster than of the decrease in $H_a(T)$, the anomalous $H_c(T)$ may be obtained. Furthermore, according to the $m_r \sim T$ relations presented in Fig. 5, α_{ex} increases slower for the sample with larger z value. This is the reason that the anomalous $H_c(T)$ tends to disappear with increasing z value.

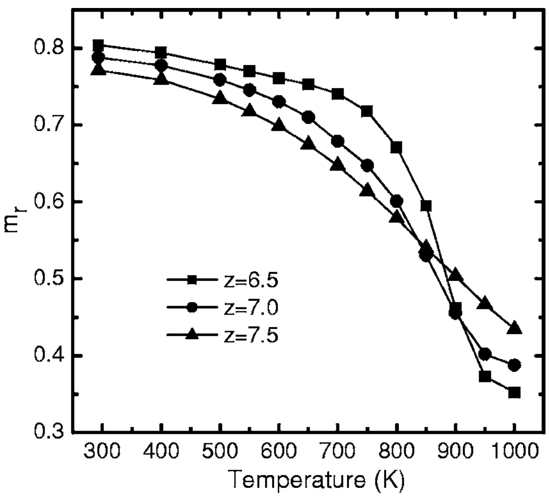

FIG. 5. The temperature dependence of m_r of the $\mathrm{Sm(Co,Fe,Cu,Zr)}_z$ magnets.

In summary, the coercivity mechanism of the precipitation-hardened $\mathrm{Sm(Co,Fe,Cu,Zr)}_z$ magnets has been found to vary with temperature. The magnetization reversal is mainly controlled by domain-wall pinning when $T \leqslant 800$ K, while the reversal is mainly dominated by nucleation at $T > 800$ K. It is interesting to find that $H_c(T)$ of the precipitation-hardened magnets is the competitive result of the temperature dependences of IGEC and anisotropy field. The fast drop of IGEC at the temperature near $T_c^{1:5}$ leads to the fast increase of α_{ex} and the anomalous $H_c(T)$. Since α_{ex} increases slower for the sample with larger z value, the anomalous $H_c(T)$ tends to disappear with increasing z value.

The work was supported by the National Science Foundation of China and the U.S. DoD/MURI under the Grant No. N00014-05-1-0497.

[1] A. G. Popov, A. V. Korolev, and N. N. Shchegoleva, Phys. Met. Metallogr. **69**, 100 (1990).
[2] J. F. Liu, T. Chui, D. Dimitrov, and G. C. Hadjipanayis, Appl. Phys. Lett. **73**, 3007 (1998).
[3] D. Goll, I. Kleinschroth, W. Sigle, and H. Kronmuller, Appl. Phys. Lett. **76**, 1054 (2000).
[4] J. Zhou, R. Skomski, C. Chen, G. C. Hadjipanayis, and D. J. Sellmyer, Appl. Phys. Lett. **77**, 1514 (2000).
[5] A. M. Gabay, W. Tang, Y. Zhang, and G. C. Hadjipanayis, Appl. Phys. Lett. **78**, 1595 (2001).
[6] J. Zhang, H. Liu, C. B. Rong, H. W. Zhang, S. Y. Zhang, B. G. Shen, Y. Q. Bai, and B. H. Li, Appl. Phys. Lett. **83**, 1172 (2003).
[7] C. B. Rong, J. Zhang, H. W. Zhang, X. B. Du, S. Y. Zhang, and B. G. Shen, J. Magn. Magn. Mater. **279**, 143 (2004).
[8] G. C. Hadjipanayis, E. J. Yadlowsky, and S. H. Wollins, J. Appl. Phys. **53**, 2386 (1982).
[9] J. Fidler and P. Skalicky, J. Magn. Magn. Mater. **27**, 127 (1982).
[10] C. B. Rong, H. W. Zhang, J. Zhang, X. B. Du, S. Y. Zhang, and B. G. Shen, J. Appl. Phys. **97**, 33907 (2005).
[11] J. D. Livingston and D. L. Martin, J. Appl. Phys. **48**, 1350 (1977).
[12] D. Goll, H. Kronmuller, and H. H. Stadelmaier, J. Appl. Phys. **96**, 6534 (2004).
[13] E. Lectard, C. H. Allibert, and R. Ballou, J. Appl. Phys. **75**, 6277 (1994).
[14] D. C. Crew, R. C. Woodward, and R. Street, J. Appl. Phys. **85**, 5675 (1999).
[15] D. C. Crew, P. G. McCormick, and R. Street, J. Appl. Phys. **86**, 3278 (1999).
[16] I. Panagiotopoulos, M. Gjoka, and D. Niarchos, J. Magn. Magn. Mater. **279**, 389 (2004).
[17] C. B. Rong, H. W. Zhang, S. L. He, R. J. Chen, and B. G. Shen, Appl. Phys. Lett. **86**, 122506 (2005).
[18] P. E. Kelly, K. O. Grady, P. I. Mayo, and R. W. Chantrell, IEEE Trans. Magn. **25**, 3881 (1989).
[19] D. Goll, M. Seeger, and H. Kronmuller, J. Magn. Magn. Mater. **185**, 49 (1998).
[20] R. Skomski, J. Phys.: Condens. Matter **15**, R841 (2003).
[21] E. C. Stoner and E. P. Wohlfarth, Philos. Trans. R. Soc. London **240**, 599 (1948).