ELSEVIER

Journal of Magnetism and Magnetic Materials 206 (1999) 127–134

Journal of magnetism and magnetic materials

www.elsevier.com/locate/jmmm

# Anomalous magnetic viscosity in bulk-amorphous materials

L. Wang[a], J. Ding[b,*], Y. Li[b], Y.P. Feng[a], X.Z. Wang[b]

[a]*Department of Physics, National University of Singapore, Lower Kent Ridge Road, Singapore 119260, Singapore*
[b]*Department of Materials Science, National University of Singapore, Lower Kent Ridge Road, Singapore 119260, Singapore*

Received 16 February 1999; received in revised form 28 July 1999

**Abstract**

The demagnetization processes and the magnetic viscosity have been studied on a bulk-amorphous $Nd_{60}Fe_{30}Al_{10}$ rod at room temperature. Many unique magnetic properties have been found in this novel hard magnetic material. A clear hysteresis was present on the minor loops, though the total and irreversible susceptibilities exhibited single-phase magnet behavior. A significant magnetic viscosity was evident at positive fields. A large magnetic viscosity was found at negative fields close to the coercivity. The time-dependent magnetization curves were not logarithm-linear and could be well fitted with a logarithm power series with $N = 6$. The fluctuation field was strongly dependent on the magnetic field. The activation volume was calculated to be $15–60 \times 10^{-18}$ cm$^3$. The magnetic viscosity on the minor loops was measured. A non-monotonic behavior was found. 

*PACS:* 75.60.Lr; 75.60.Ej; 75.50.B6

*Keywords:* Bulk-amorphous ferromagnet; Magnetic viscosity; Non-monotonic behavior

## 1. Introduction

Recently, magnetic bulk-amorphous materials have been successfully obtained by the chill-casting process [1,2]. The amorphous (Nd,Pr)–Fe–(Al,Si) alloys have shown high values of coercivity [1,3]. Therefore, the novel magnetic materials are of interest for hard magnetic applications [1,3].

The coercivity mechanism of amorphous materials has not been well understood. The study of magnetic viscosity is well known as a powerful method for the investigation of magnetization and demagnetization processes. The viscosity study can provide information about energy barriers and activation volumes involved in nucleation or pinning mechanisms. Therefore, the results of magnetic viscosity study can be used for optimization of magnetic properties, e.g., coercivity and remanence.

In this paper, we report the viscosity behavior of the hard magnetic bulk-amorphous $Nd_{60}Fe_{30}Al_{10}$ alloy. The main loop viscosity behavior has been described using the theory developed by O'Grady et al. [4]. The fluctuation field and activation volume were determined. A non-montonic magnetic viscosity was found on the minor loops.

Magnetic viscosity, i.e., time dependence of magnetization, is a well-known phenomenon and arises due to a finite time for thermal activation of the magnetization over the energy barrier [5–7]. The time-dependent magnetization is given in general

* Corresponding author. Fax: + 65-776-3604.
*E-mail address:* massdingj@nus.edu.sg (J. Ding)

0304-8853/99/$ - see front matter 
PII: S0304-8853(99)00548-X

as a sum of the reversible and the irreversible magnetizations ($M(t) = M_{irr}(t) + M_{rev}(t)$), where $M_{irr}(t)$ can be obtained as the remanent component after the instantaneous removal of the field $H$ at time $t$. Variations of $M_{rev}$ with time are generally small and arise for example from a change of magnetization-dependent demagnetization field. Here, we consider only the component $M_{irr}(t)$, since magnetic changes by thermal action are irreversible. According to this phenomenology, the time dependence of the remanent magnetization is given by

$$M_{irr} = M_{irr}(0)e^{-t/\tau}. \quad (1)$$

Here $\tau$ is the relaxation time and in zero field $\tau^{-1}$ is given as

$$\tau^{-1} = f_0 e^{-\Delta E/kt}, \quad (2)$$

where $\Delta E$ is the energy barrier for reversal.

For the case where the distribution function of the barrier $\Delta E$ remains approximately constant during the time of observation, the formalism of Eqs. (1) and (2) gives [5]:

$$M_{irr}(t) = M_0 + C \ln(t). \quad (3)$$

Most magnetic materials can be well-described with the linear logarithm function (Eq. (3)). Eq. (3) has been widely used for the study of the magnetic viscosity for different types of magnetic materials, such as Alnico, $BaFe_{12}O_{19}$, ferritin, $SmCo_5$ and NdFeB [5,8–10].

Magnetic viscosity behavior in spin glass and other disordered systems does not obey Eq. (3). O'Grady et al. [4] develop a theory to explain these phenomena. This theory shows that the curvature of the $M(t)$ versus $\ln(t)$ curve is strongly dependent on the form of the distribution of energy barriers. They give a logarithm power series to describe the $M(t)$ versus $\ln(t)$ curve:

$$M_{irr} = M_0 + \sum_{i=1}^{n} C_i \ln(t)^i. \quad (4)$$

An irreversible change of the magnetization may be described using the following differential equation [11]:

$$dM_{irr} = S d\ln(t) + \chi_{irr} dH, \quad (5)$$

where $\chi_{irr}$ is the irreversible susceptibility.

In Eq. (5), $\chi_{irr}\, dH$ is the field dependent part of $dM_{irr}$. Néel [6], therefore, suggested that the effect of the thermal fluctuations ($S d\ln(t)$) may be represented as a fictitious field, which he called the fluctuation field. The definition of the fluctuation field is [7]

$$S = H_f \cdot \chi_{irr} \quad (6)$$

If the $M(t)$ versus $\ln(t)$ is not linear, Eq. (6) should be generalized [11]:

$$H_f(H, t) = \frac{S(H, t)}{\chi_{irr}(H, t)}, \quad (7)$$

where $S$ can be determined [11] as

$$S(H, t) = \left.\frac{\partial M_{irr}}{\partial \ln(t)}\right|_H \quad (8)$$

An alternative but equivalent definition of the fluctuation field is obtained directly from the constitutive relation [11]

$$H_f = -\left.\frac{\partial H}{\partial \ln(t)}\right|_{M_{irr}}. \quad (9)$$

Measurement of $H_f$ can be used to evaluate an activation volume [7].

$$V_a = \frac{kT}{M_s H_f} \quad (10)$$

$M_s$ is the saturation magnetization.

The activation volume represents the average volume of thermal activation (nucleation or domain wall motion). The activation volume $V_a$ is associated with the domain wall thickness for many hard magnetic materials [9,10,12].

## 2. Experimental procedure

The bulk-amorphous sample studied in this work is a cylindrical $Nd_{60}Fe_{30}Al_{10}$ rod with a diameter of 1 mm and a length of 3 mm prepared by chill casting [1,3]. The density of the rod was estimated to be 7.7 g/cm$^3$. The X-ray diffraction and Mössbauer spectroscopy studies confirmed that the rod consisted of a single amorphous phase.

The magnetic study was performed using a vibrating sample magnetometer (Oxford VSM) at room

temperature with a maximum applied magnetic field of 50 kOe. The internal magnetic field $H_i$ was estimated using a demagnetization field factor of 0.13 for cylindrical sample [13].

The hysteresis loop (Fig. 1) was measured using the field ramping rate of 20 kOe/min. The minor loops in Fig. 1 were taken after waiting for 2 min at negative field. The sample was then brought $+0.5$ kOe with a ramping rate of 5 kOe/min. $M_{irr}$ was obtained as the remanent component after the removal of the field $H$. The irreversible susceptibility $\chi_{irr}$ was calculated by $dM_{irr}/dH_i$, while the total susceptibility $\chi_{tot}$ was determined by the differential of $dM/dH_i$ on the mean hysteresis loop.

For the viscosity measurements (Fig. 2), the rod was first saturated with a positive field of 50 kOe. The applied magnetic field $H_a$ was then brought to the desired field with a ramping rate of 20 kOe/min. The sample was held at the constant applied field for 1800 s (30 min), during which the change of magnetization (the viscosity) was measured. The magnetic viscosity study was also performed on the minor loop. The sample was brought to $-4$ kOe with a ramping rate of 20 kOe/min after saturation with $+50$ kOe and then brought back to the desired fields on the minor loop (Fig. 5). The sample was held at a constant field for 900 s (15 min).

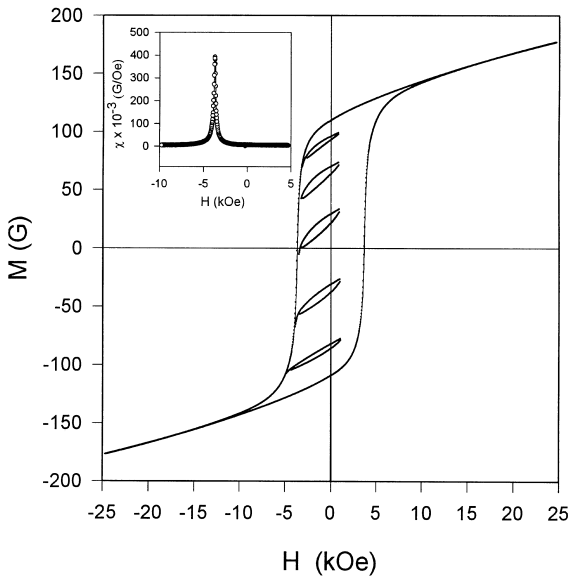


Fig. 1. The hysteresis loop and the minor loops of the amorphous $Nd_{60}Fe_{30}Al_{10}$ rod. The ramping rate is 20 kOe/min. The total susceptibility as a function of the magnetic field is shown in the attached plot.

## 3. Result and discussion

Fig. 1 is the hysteresis loop and the minor loops of the amorphous rod. The $Nd_{60}Fe_{30}Al_{10}$ rod is a hard magnet with a coercivity of 3.7 kOe (the ramping rate was 20 kOe/min). All the minor loops (especially those taken after the demagnetization with a field close the coercivity) are open, indicating the presence of magnetic hysteresis (Fig. 1). This behavior is different from that of conventional permanent magnets (based on a hard magnetic crystalline phase $BaFe_{12}O_{19}$, $SmCo_5$ or $Nd_2Fe_{14}B$), by which the minor loops are straight lines and no hysteresis is present [8]. Hysteresis existing on minor loops has been reported in nanocomposites consisting of a mixture of hard and soft phases [14] due to exchange coupling of the hard and soft phases. The hysteresis on the minor loops in Fig. 1 indicates irreversible magnetization processes. The mechanism of the demagnetization processes is to be investigated.

The total susceptibility is shown in the inset to Fig. 1. A sharp peak appears at the magnetic field, which is nearly identical to the coercivity. The irreversible susceptibility had a field-dependence similar to that of the total susceptibility in Fig. 1. This behavior is typical of single-phase magnets [9,14], by which irreversible magnetization processes mainly occur at magnetic fields nearby the coercive field.

For the amorphous $Nd_{60}Fe_{30}Al_{10}$ rod, the magnetic viscosity has been observed in a wide field range. Fig. 2 shows the time-dependent magnetization curves at different constant applied fields $H_a$ from positive 5 to negative $-10$ kOe after the magnetization with the positive field of 50 kOe. It is surprising that a significant magnetic viscosity was present at positive fields, as shown in Fig. 2a. The magnetic viscosity is detectable even at positive 10 kOe. As is well known, magnetic viscosity occurs because of the thermal instability, corresponding to the flip of magnetic domains into a more stable

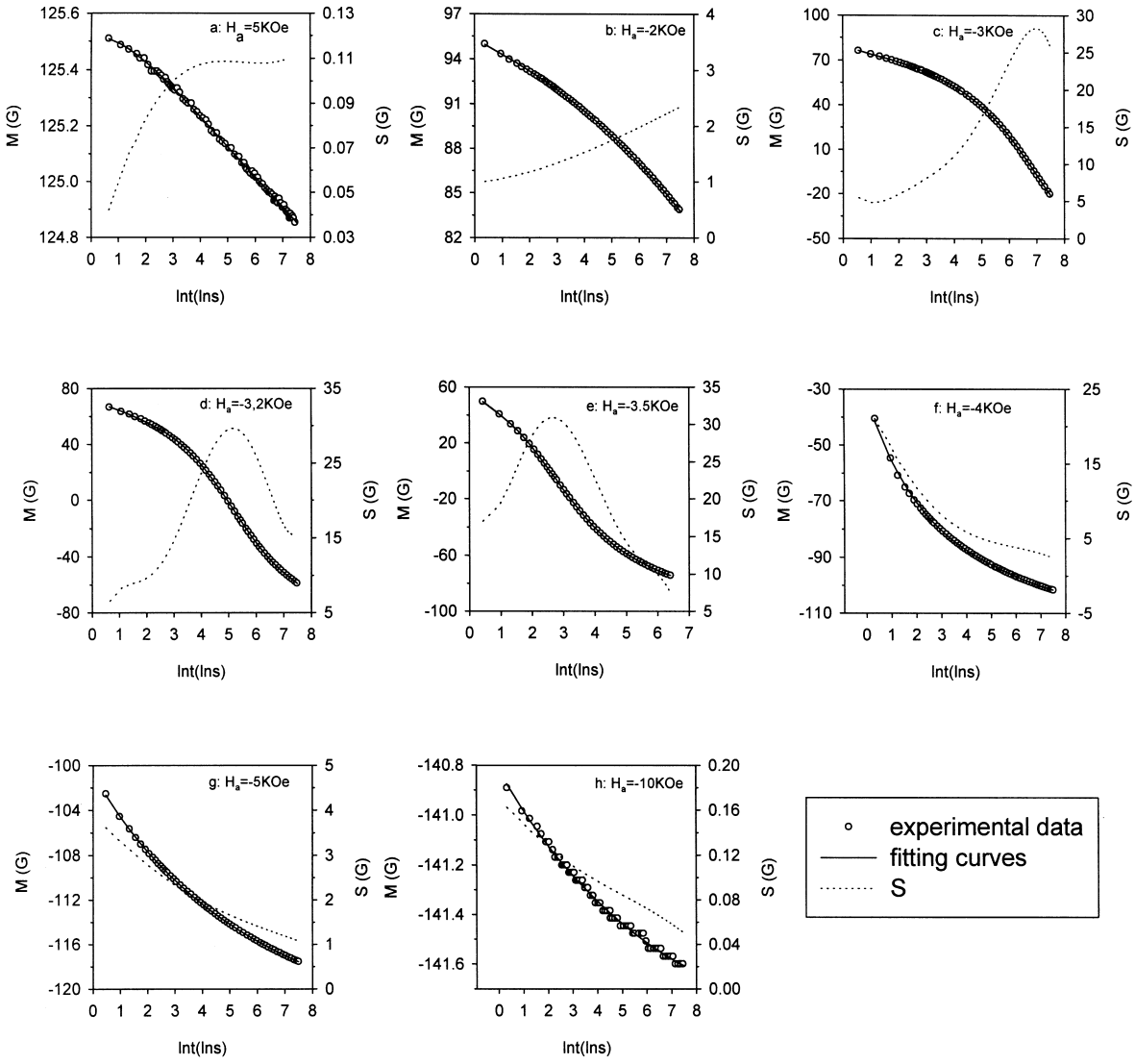


Fig. 2. The time-dependent magnetization curves taken at different constant applied magnetic fields. The solid lines are the fitting curves using the logarithm power series with $n = 6$. The dashed lines are the viscosity coefficient $S$ as a function of the time. The time scale is ln(s).

direction, e.g., from positive to negative under the application of a negative field after the saturation in the positive direction. The origin of the viscosity at positive fields has to be investigated. The viscosity at positive fields is probably related to the hysteresis on the minor loop in Fig. 1.

At negative magnetic fields, the magnetic viscosity increased with increasing negative field. The highest viscosity (i.e., the largest change in magnetization) was found in the field range close to the coercivity. When the negative field further increases beyond the coercivity, the magnetic viscosity decreases and becomes nearly unmeasurable at a field of 15 kOe or above. This change of the viscosity is often found in other magnetic materials, such as mechanical alloying $SmCo_5$ and melt-spun NdFeB [9,10].

As shown in Fig. 2, the time-dependent magnetization curves of the amorphous $Nd_{60}Fe_{30}Al_{10}$ rod cannot be described using the linear logarithm function (Eq. (3)). A large deviation from the linear logarithm is found in the magnetization curves taken at magnetic fields near the coercive field (3–5 kOe).

The non-linearity of the magnetic viscosity has been reported previously [12,15–17]. Street et al. [12] have proposed the modified equation of $M(t) = M_0 + C\ln(t + t_0)$ (with $M_0$, $C$ and $t_0$ as constants) for fitting non-linear magnetization curves. For magnetization curves in Fig. 2, the magnetization curves taken at positive fields can be well fitted with the modified linear-logarithm function $M(t) = M_0 + C\ln(t + t_0)$. However, the modified linear-logarithm function cannot be used for fitting the magnetization curves taken at negative fields, especially magnetization curves taken at magnetic fields close to the coercivity. The curvature of the time-dependent magnetization changed from concave downwards to upwards, when the magnetization changed its sign from positive to negative, as shown in Figs. 2d and e. This has shown that the magnetic viscosity coefficient $S$ is not a time-independent constant and is strongly dependent on the irreversible magnetization [11].

O'Grady et al. [4] have derived theoretically the time-dependent magnetization curve for spin glass and other disorder systems, for which a log-normal distribution of particle volumes (corresponding to the energy barriers) is assumed. A non-linearity of the viscosity was obtained from the theoretical analysis [4]. They proposed a logarithm power series for fitting their time-dependent magnetization curves. In this work, we fit the magnetization curves in Fig. 2 using a logarithm power series as shown in Eq. (4).

If $n = 1$, Eq. (4) reduces to Eq. (3). For magnetization curves taken at magnetic fields near the coercivity, a power series with $n$ up to 6 is required. All the magnetization curves including those taken at positive fields can be well-fitted with the logarithm power series with $n = 6$, as shown by the solid lines in Fig. 2. This result implies that the bulk-amorphous $Nd_{60}Fe_{30}Al_{10}$ material may have a log-normal distribution of energy barriers.

The non-linearity in Fig. 2 shows that the viscosity coefficient $S$ is time dependent. As reported by Lyberators and Chantrell [11], the viscosity coefficient $S$ can be calculated as Eq. (8) ($S(H, t) = \partial M_{irr}/\partial \ln(t)$ at constant field).

The viscosity coefficient $S$ is plotted in Fig. 2 as a function of the time $t$ at the different constant fields. It can be seen that $S$ is nearly a constant at positive fields, if the time is long enough (Fig. 2a). For the magnetization curves taken at low negative fields, $S$ increases with the increasing time. For the magnetization curves taken at negative 3–3.5 kOe (i.e., just below the coercivity), $S$ increases initially with the time, reaches a maximum when the magnetization is around zero, and then decreases with the time. For the magnetization curves taken at higher negative fields above the coercivity, $S$ decreases continuously with the time. As reported previously [17], the time-dependent magnetization curves of the Tb–Fe–Co thin films are similar to those of the amorphous $Nd_{60}Fe_{30}Al_{10}$ rod at the magnetic fields nearby the coercivity (Figs. 2d and e). Non-linearity of the viscosity has been reported in $Fe_3O_4$ small particle systems [15] and Cu–Mn spin glasses [16].

The fluctuation field can be evaluated from the viscosity coefficient $S$ and the irreversible susceptibility. An alternative expression of the fluctuation field is shown as Eq. (9) ($H_f = -\partial H/\partial \ln(t)$ at constant $M_{irr}$) [11].

Fig. 3 shows the internal magnetic field $H_i$ as a function of $\ln(t)$ at different constant irreversible magnetizations. These data points were taken from a set of time-dependent magnetization curves, which were measured at different positive applied magnetic fields (3.0–3.1 kOe). In this relatively narrow field range (3.0–3.1 kOe), the magnetic field has a linear relationship with $\ln(t)$ at a constant irreversible magnetization. All the linear lines in Fig. 3 are parallel to each other, indicating that the fluctuation field $H_f$ is a constant in the narrow field interval. Similar results have been reported for Tb–Fe–Co thin films [17].

Fig. 4 is the plot of the fluctuation field as a function of the magnetic field. $H_f$ in Fig. 4 was estimated from different sets of viscosity curves in narrow field ranges as described in Fig. 3. $H_f$ was found to be constant, if the field range is small enough [17].

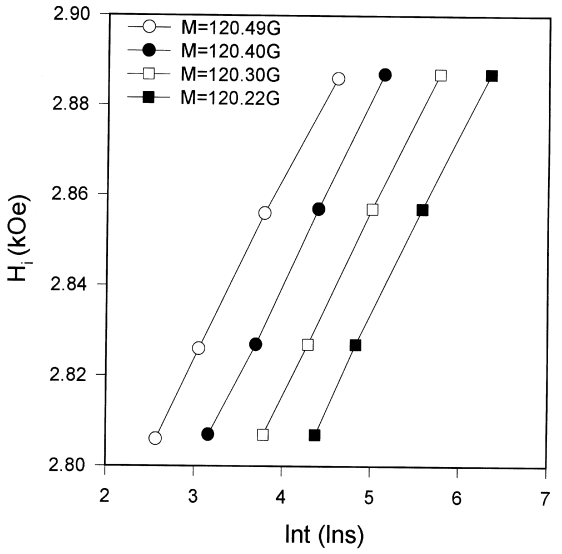


Fig. 3. The internal magnetic field $H_i$ versus ln($t$) at different constant irreversible magnetizations. The data points were obtained from the time-dependent magnetization curves taken at different constant applied fields. The time scale is ln(s).

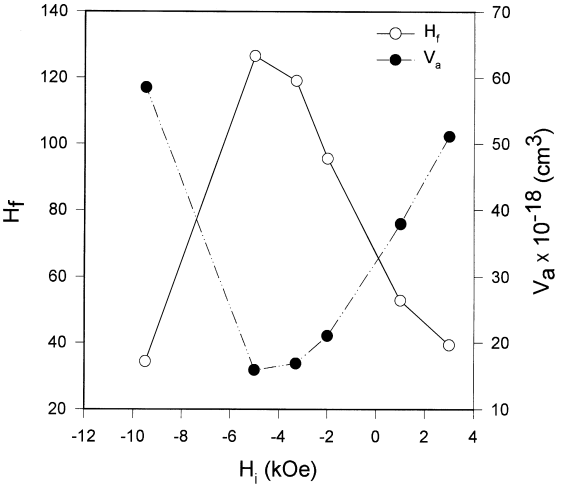


Fig. 4. The fluctuation field $H_f$ and the activation volume $V_a$ as functions of the internal magnetic field $H_i$.

For positive fields, the fluctuation field $H_f$ is relatively low in the range of 30–40 Oe (Fig. 4). The fluctuation field increases with increasing negative field, and the maximum of 120–130 Oe is found in the field range close to the coercivity (4–5 kOe). The fluctuation field then decreases with increasing negative field, when the negative field is beyond the coercivity. $H_f = 35$ Oe was measured at the magnetic field of $-9.5$ kOe.

From the fluctuation field $H_f$, the activation volume $V_a$ can be calculated, which is the volume of magnetic domains involved in the thermal activation [11].

The activation volume is also plotted in Fig. 4 as a function of the internal magnetic field. The activation volume at positive fields is approximately $50 \times 10^{-18}$ cm$^3$. $V_a$ decreases with increasing negative fields and is equal to $15\text{–}20 \times 10^{-18}$ cm$^3$ at magnetic fields nearby the coercivity. These values of the activation volume are comparable to the reported values of other magnetic materials, such as $SmCo_5$ and NdFeB magnets [9,10,12].

Fig. 5 is the non-monotonic behavior of the magnetic viscosity on the minor loop after demagnetization with $-4$ kOe. The viscosity is still in the negative direction at $-3$ kOe. At lower magnetic fields $-2.25$ and $-1.95$ kOe, clear non-monotonic magnetic viscosity is found (see Figs. 5b and c). If the magnetic field is brought back far enough ($-1$ kOe), the magnetic viscosity will be monotonic again (see Fig. 5d). However, the direction is positive this time. Nanocomposites, such as Ni/NiO and $Nd_2Fe_{14}B/Fe_3B$ [18,19], have been reported possessing such character. The mechanism of this behavior in nanocomposites $Nd_2Fe_{14}B/Fe_3B$ is due to the interaction between the hard magnetic phase $Nd_2Fe_{14}B$ and soft magnetic phase $Fe_3B$. The mechanism of the non-monotonic behavior in amorphous magnetic material is still not well established. More investigation is needed.

## 4. Conclusions

In conclusion, the bulk-amorphous $Nd_{60}Fe_{30}Al_{10}$ rod has shown many unique magnetic properties in this study. A clear hysteresis was present on the minor loop, though the total and irreversible susceptibilities exhibited the single-phase magnet behavior. A significant viscosity was observed at positive fields. The viscosity is detectable even at

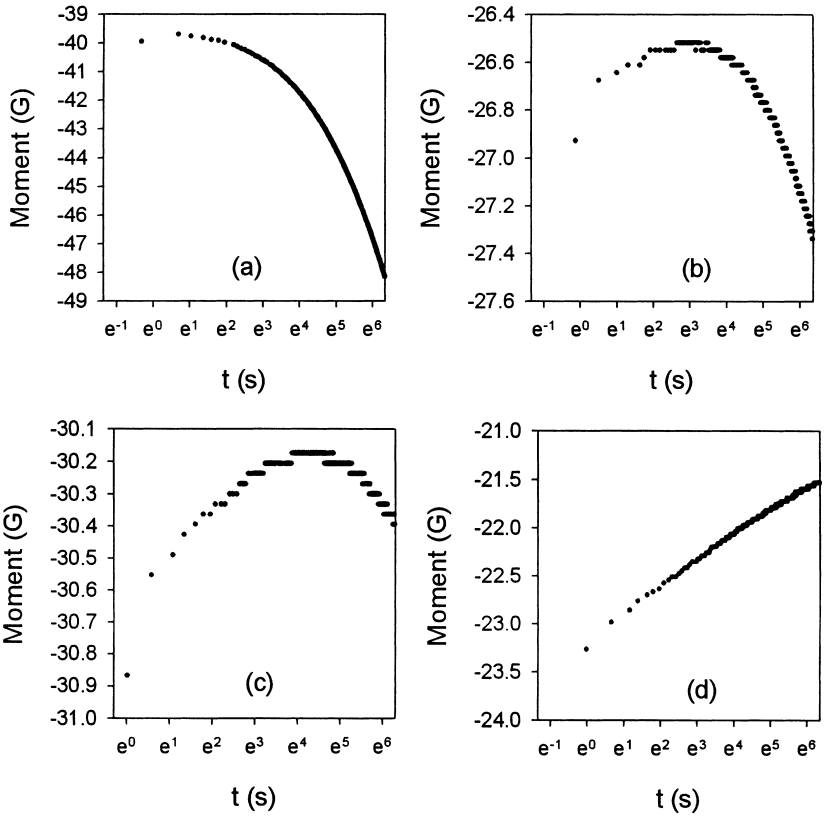


Fig. 5. The time-dependent magnetization curves taken at different applied magnetic fields on the minor loops. The rod was first saturated at a positive field of 50 kOe. Then, the field was brought to $-4$ kOe, finally to the desired fields on the minor loop: (a) $-3$ kOe, (b) $-2.25$ kOe, (c) $-1.95$ kOe, (d) $-1$ kOe.

positive 10 kOe after the magnetization at positive 50 kOe. The viscosity at positive fields is an unusual magnetic phenomenon. The time-dependent magnetization curves could not be described with a linear logarithm function, especially for the magnetization curves taken at magnetic fields nearby the coercivity. All the magnetization curves were well-fitted with the logarithm power series with $n = 6$, indicating a log-normal distribution of the energy barriers. The fluctuation field $H_f$ was strongly dependent on the magnetic field. $H_f$ was 100–130 Oe when the magnetic field was close to the coercivity, resulting in an activation volume of $15–20 \times 10^{-18}$ cm$^3$. Clear non-monotonic magnetic viscosity is found on minor loops.

## References

[1] A. Inoue, T. Zhang, A. Takeuchi, Sci. Rep. RITU A 44 (1997) 261.
[2] A. Inoue, H. Koshida, T. Itoi, A. Makino, Appl. Phys. Lett. 73 (1998) 744.
[3] J. Ding, Y. Li, X.Z. Wang, J. Phys. D 32 (1999) 713.
[4] K. O'Grady, M. El-Hilo, R.W. Chantrell, J. Appl. Phys. 76 (1994) 6368.
[5] R. Street, J.C. Woolley, Proc. Phys. Soc. B 63 (1950) 509.
[6] L. Néel, Phys. Radium 11 (1950) 49.
[7] E.P. Wohlforth, J. Phys. F 14 (1984) 2155.
[8] J.R. Friedman, U. Voskoboynik, M.P. Savachik, Phys. Rev. B 56 (1997) 10793.
[9] J. Ding, P.A.I. Smith, P.G. McCormick, R. Street, J. Magn. Magn. Mater. 161 (1996) 303.
[10] R. Street, D. Bingham, R.K. Day, J.B. Dunlop, J. Phys. C 8 (1988) 629.

[11] A. Lyberatos, R.W. Chantrell, J. Phys.: Condens. Matter 9 (1997) 2623.
[12] R. Street, L. Folks, S. Hirosawa, Sixth Symposium on Magn. Anisotropy and Coercivity in Rare Earth-Transition Metal Alloys, Pittsburgh, 25 Oct. 1990.
[13] D.X. Chen, J.A. Brug, R.B. Goldfarb, IEEE Trans. Magn. 27 (1991) 3601.
[14] P.G. McCormick, J. Ding, E.H. Feutrill, R. Street, J. Magn. Magn. Mater. 157–158 (1996) 7.
[15] A. Labarta, O. Iglesias, L. Balcolls, F. Badia, Phys. Rev. B 48 (1993) 10240.
[16] R.V. Chamberlin, G. Mozurkewich, R. Orbach, Phys. Rev. Lett. 52 (1984) 868.
[17] R. Street, S.D. Brown, J. Appl. Phys. 76 (1994) 6386.
[18] D.S. Geoghegan, P.G. McCormick, R. Street, J. Magn. Magn. Mater. 177–181 (1998) 937.
[19] L. Folks, E.H. Feutrill, R. Street, Eigth International Symposium on Magnetic Anistropy and Coercivity in RE-TM Alloys, Birmingham, UK, 15 September 1994, p. 415.