Quasilogarithmic magnetic viscosity in perpendicularly anisotropic Nd–Fe–B films

Q. Yao [a], R. Grössinger [b], W. Liu [a,*], W.B. Cui [a], F. Yang [a], X.G. Zhao [a], Z.D. Zhang [a]

[a] Shenyang National Laboratory for Materials Science, Institute of Metal Research and International Center for Materials Physics, Chinese Academy of Sciences, Shenyang 110016, People's Republic of China
[b] Institute for Solid State Physics, Vienna University of Technology, Wiedner Haupstrasse 8-10, A-1040 Vienna, Austria

ARTICLE INFO

Abstract: The quasilogarithmic magnetic viscosity of the perpendicularly anisotropic Nd–Fe–B films is determined by measuring the time dependent magnetization in coercive fields at 300 K–4.2 K. The distribution of energy barrier heights in the films is believed to be the origin of this stretched exponential magnetic relaxation. Another feature exhibited in the anisotropic Nd–Fe–B films is the nonmonotonic temperature dependence of the magnetic viscosity coefficients, which are extracted by the decay slopes of the linear quasilogarithmic magnetic aftereffect curves.

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1. Introduction

Time dependence of magnetization in ferromagnetic materials, known as magnetic viscosity or magnetic aftereffect, is of great practical and theoretical significance. Following the first observation of the magnetic viscosity by Preisach [1], a physical mechanism of thermally-activated reversal processes of magnetic domains was developed by Street and Woolley [2] in order to interpret the magnetic viscosity. Accordingly, the magnetic hysteresis was explained as a system of domains and domain walls caused by a complicated free energy landscape, with many relative minima corresponding to metastable states. The energy barriers between the metastable energy minima can be overcome by thermal agitation, which causes an irreversible reversal process with magnetization relaxing to the thermodynamic equilibrium state and a consequent decay of magnetization with time. The transition frequency of the magnetic moment in this relaxation process is usually estimated by the Arrhenius–Néel statistical switching model [3]:

$$f = f_0 \exp(-E_B/k_B T) \tag{1}$$

where f_0 is a frequency constant of the order of 1010–1012 Hz, k_B the Boltzmann constant, T the absolute temperature and E_B the energy barrier depending on the external field and temperature.

The relaxation time τ is given by $1/f$ as:

$$\tau = f_0^{-1} \exp(E_B/k_B T) \tag{2}$$

which characterizes the mean time needed to cross a local energy barrier to another energy state. Obviously, the smaller the relaxation time τ and the higher the transition frequency f, the higher the rate of the magnetic relaxation.

In most permanent magnetic materials, the magnetization relaxes very slowly towards its ground state in presence of large anisotropy energy barriers. Therefore, a constant reverse field (called holding field H_0) is usually applied to a magnetically saturated sample to accelerate the magnetic viscosity testing in the hard magnets, via lowering the effective anisotropy energy barriers. Over a limited range of time and assuming a flat energy barrier distribution, the experimental data of the magnetization $M(t)$ can be described by a logarithmic time dependence [4,5]

$$M(t) = M_0 - S\ln(t) \tag{3}$$

where S is the magnetic viscosity coefficient determining the rate of the magnetic relaxation and M_0 is the magnetization of the sample at the start of the viscosity measurement ($t = 0$). To prevent the $\ln(t)$ from going to infinity, a reference time t_0 is usually introduced to establish a $\ln(t + t_0)$ dependence of M:

$$M(t) = M_0 - S\ln(t + t_0) \tag{4}$$

The magnetization decay with time due to the magnetic viscosity becomes pronounced in nanostructured magnetic systems [6–7]. Whether in particulate or continuous films, the thermal loss effect of magnetization is manifested by the

* Corresponding author.
E-mail address: wliu@imr.ac.cn (W. Liu).

reduction in magnetic switching unit size. Also the magnetization should become strongly time-dependent as the grain size and the activation volume largely decrease in nanocrystalline permanent magnetic thin films [8–11]. However, there are relatively few studies for temperature dependence of magnetic aftereffect in perpendicularly anisotropic magnetic films, particularly at very low temperature.

In the present work, the time dependent magnetization is measured over a range of temperatures (300 K–4.2 K) for the nanocrystalline Nd–Fe–B films with perpendicular anisotropy. The quasilogarithmic magnetic relaxation is observed and studied for each of the samples at all temperatures. This is followed by a discussion of the nonmonotonic temperature dependence of magnetic viscosity coefficients for these perpendicularly aniso-tropic Nd–Fe–B films.

2. Experimental details

Mo(50 nm)/Nd–Fe–B (x)/Mo(50 nm) films (x=300 nm, 500 nm, 800 nm, and 6.5 μm) were deposited onto heated (100) oriented Si substrates in a high vacuum chamber using dc magnetron sput-tering [12]. The Mo overlayer was deposited for corrosion protec-tion and the Mo underlayer was added to resist the oxidation by the oxygen atoms diffusing from the SiO_2 surface of the Si substrate. The chamber was evacuated to a background pressure better than 1.5×10^{-5} Pa and all depositions were made at a constant total Ar gas pressure of 0.7 Pa. A sintered $Nd_{16}Fe_{71}B_{13}$ alloy target and a commercial Mo target of higher than 99.9% purity were used. The deposition rate of 9.5 Å/s was adopted during sputtering, calibrated by weighing the films. Pre-sputtering of the NdFeB target was carried out for 30 min to remove any oxidized layer on the target surface. During deposition, the temperature of the substrate was fixed at 923 K.

After deposition, the crystallographic structures and the perpendicular alignment of the films were analyzed by means of X-ray diffraction (XRD) using Cu Kα radiation [12]. After saturation with applying fields H up to 90 kOe, the time-dependent magne-tization $M(t)$ of the Nd–Fe–B films were recorded over 1800 s at 300 K–4.2 K in constant reverse fields H_0 equal to the coercivities H_c, using a Quantum Design PPMS-9H physical properties mea-surement system (PPMS). The H_0 was applied to maximize the relaxation rates and the viscosity coefficients. Before the $M(t)$ measurements at all temperatures, H_c of the films were deter-mined by the same PPMS as the fields reducing M to zero on the demagnetization curves with H up to 90 kOe. All magnetic measurements were performed by applying H perpendicular to the film plane and with the diamagnetic signals from the Si substrates being subtracted.

3. Results and discussion

Fig. 1 shows that the coercivities of the anisotropic Nd–Fe–B films increase monotonically as the temperature decreases from 300 K to 4.2 K. This is due to the increased anisotropy field of $Nd_2Fe_{14}B$ phase, despite its spin-reorientation at 135 K [13]. At 300 K, the coercivities of the anisotropic Nd–Fe–B films increase from 4.2 kOe with 300 nm thickness to 11.4 kOe with 6.5 μm thickness. Microstructure and magnetic characterizations justify the films' strong out-of-plane magnetic anisotropy with the easy axes of the columnar grains normal to the film plane [12]. With increasing the film thickness from 300 nm to 6.5 μm, the Nd–Fe–B films' perpendicular alignment is weakened gradually.

With H_c determined, magnetic aftereffects were measured for all the films over 1800 s at various temperatures under the
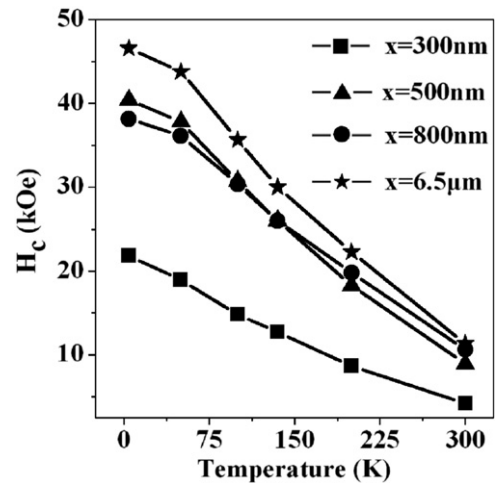

Fig. 1. Temperature dependence of coercivities for the Nd–Fe–B films with film thickness x=300 nm, 500 nm, 800 nm and 6.5 μm.

constant reverse fields H_0 ($=H_c$), by following the procedure described in the experimental section. A typical set of these measurements is given in Fig. 2(a), where the magnetization is plotted against time for the 6.5 μm anisotropic Nd–Fe–B film at 300 K–4.2 K. It is seen from Fig. 2(a) that the film magnetization decreases as relaxation progresses at all temperatures, with the relaxation rates being distinctive at different temperatures. How-ever, the Nd–Fe–B film magnetization decay is not a complete linear dependence on the logarithm of time (ln(t)) as expressed by Eq. (3). For example, Fig. 2(b) exhibits the magnetization as a function of log-time for the 6.5 μm Nd–Fe–B film at 300 K–4.2 K. It is demonstrated that the film magnetization decays linearly on the log-time aftereffect curves only in a limited time interval (around 20–1800 s) at all temperatures. From 0 s to 20 s, the magnetization relaxes nonlinearly with ln(t), indicating the nonexponential characteristic of the magnetic viscosity for the Nd–Fe–B film. Furthermore, upon introducing a reference time constant (t_0 = 10 s) in log-time, the time behavior of the Nd–Fe–B film magnetization can be perfectly fitted to Eq. (4) which establishes a linear dependence of magnetization on ln($t+t_0$). As shown in Fig. 2(c), the 6.5 μm Nd–Fe–B film magnetization decreases linearly with ln($t+10[s]$) at 300 K–4.2 K. It may be that the PPMS records the magnetic decay a short time after the relaxation starts with H_0 being applied, causing the small nega-tive M_0 (-29 to -16 Gs). The M_0 of 12 Gs for the 135 K relaxation needs to be further investigated. Under the holding reverse field of H_0 ($=H_c$), the magnetic relaxation of the perpendicularly anisotropic film starts from a energy minimum state of nearly zero net magnetization, with one spin up for every spin down which is aligned perpendicular to the film plane. Thereafter, thermal activation and H_0 jointly drive the film magnetization to gradually decay with time over local energy barriers. Due to the fluctuations of the local magnetocrystalline anisotropy or the pinning of the domain-wall motion, the disorder of energy barrier heights creates a dispersion of relaxation times for activation grains in the anisotropic Nd–Fe–B film. This relaxation process causes the M to deviate from a linear dependence on ln(t), giving rise to the quasilogarithmic or the stretched exponential magnetic viscous effects. Accordingly, the magnetic viscosity coefficients S in the coercive fields are extracted from the slopes of these linear quasilogarithmic aftereffect curves to characterize the magnetic relaxation rates at various temperatures.

Another feature appearing in these anisotropic Nd–Fe–B films is the nonmonotonic temperature dependence of the magnetic viscosity coefficients S, which has also been detected in CoCr films