ELSEVIER

Contents lists available at ScienceDirect

# Journal of Alloys and Compounds

journal homepage: www.elsevier.com/locate/jallcom



# Mechanical damping in nanostructured Nd<sub>60</sub>Fe<sub>30</sub>Al<sub>10</sub> magnetic alloys

H.R. Salva<sup>a,c</sup>, L.M. Fabietti<sup>b,c</sup>, A.A. Ghilarducci<sup>a,c</sup>, S.E. Urreta<sup>b,\*</sup>

- a Centro Atómico Bariloche, Comisión Nacional de Energía Atómica, Instituto Balseiro, Universidad Nacional de Cuyo, 8400 Bariloche, RN, Argentina
- <sup>b</sup> Facultad de Matemática, Astronomía y Física, Universidad Nacional de Córdoba, Ciudad Universitaria, 5000 Córdoba, Argentina
- <sup>c</sup> CONICET, Argentina

### ARTICLE INFO

Article history: Received 10 July 2008 Received in revised form 14 May 2009 Accepted 4 October 2009 Available online 13 October 2009

Keywords: Amorphous materials Rapid-solidification Mechanical properties Internal damping Dynamic elastic modulus

### ABSTRACT

Nanostructured  $Nd_{60}Fe_{30}Al_{10}$  magnetic alloys were obtained by melt spinning at a tangential wheel speed of 5 m/s. Magnetic and stress relaxation processes were investigated in the as cast condition. Low frequency mechanical spectroscopy studies (dynamic elastic torsion modulus and internal damping measurements) were conducted at fixed frequencies between 0.1 and 1 Hz, in the range [150–480 K]. The low temperature internal damping and dynamic elastic modulus exhibited a large heating–cooling hysteresis and two damping peaks: one due to a stress induced relaxation mechanism operating near  $200 \, \text{K} \, [1 \, \text{Hz}]$  and another one, at about  $260-270 \, \text{K}$ , which could not be correlated with any step in the modulus, indicating that it is not anelastic in origin. The high temperature damping was described by an exponential background and a broad peak contribution centered at about  $400 \, \text{K} \, (1 \, \text{Hz})$ . The resulting maximum is frequency dependent, but it is little sensitive to applied fields up to  $30 \, \text{mT}$ . On contrary, the elastic modulus behavior associated to this maximum resulted largely affected by frequency, the external fields and the magnetic state of the sample.

© 2009 Elsevier B.V. All rights reserved.

## 1. Introduction

It has been demonstrated that rapidly solidified  $Nd_{60}Fe_{30}Al_{10}$  bulk metallic glass (BMG) alloys are not entirely amorphous materials. For this composition the microstructure consists [1–3] of nanoscopic Nd-rich phases (hcp Nd, Nd(Fe,Al)<sub>2</sub>,  $\delta$ -NdFeAl phase) embedded in a major ferromagnetic Fe-rich amorphous phase, also containing small clusters of the  $\mu$ -NdFeAl phase [4]. The relatively high coercivity of these largely amorphous alloys has been attributed to strong pinning of domain walls by well dispersed, small Nd-rich nanophases [2] or to magnetic exchange coupling between the ferromagnetic clusters and these antiferromagnetic (Nd(Fe,Al)<sub>2</sub>,  $\delta$ -NdFeAl) nanoscopic phases [3].

In addition to the hard magnetic properties, these BMG alloys are found to show extensive stress and magnetization relaxation near room temperature, even at small or even zero internal fields. It is found that thermal activation of magnetization mechanisms is quite efficient as indicated by the large values of the mean fluctuations field measured (of about 14 mT at the coercive field [5]). The stress relaxation mechanisms, on the other hand, are responsible for the relatively large internal damping (ID) measured

and the interesting dynamic elastic modulus behavior between 150 and 500 K [6], for a quite large frequency range [0.01 Hz to 1 kHz]. These previous mechanical spectroscopy results showed an ID peak near 200 K (1 Hz) originated in a relaxation mechanism, and a broad frequency independent maximum at about 270 K, tentatively associated to  $\delta$  phase small particles, antiferromagnetically ordering below 250 K [3]. Above room temperature the ID spectrum was found to increase monotonously with temperature and show frequency dependence, but it was little sensitive to an applied field up to 30 mT. The dynamic elastic shear modulus G was found to undergo the classical step associated to anelastic relaxations at 200 K (1 Hz), but it exhibited large anomalies between the temperatures of magnetic ordering of the  $\delta$  phase (250 K) and that of the Fe-rich  $\mu$  phase (460 K).

In this article we report new details concerning the ID and modulus G as functions of temperature in a  $Nd_{60}Fe_{30}Al_{10}$  alloy processed by melt spinning at a wheel speed of 5 m/s.

# 2. Experimental procedures

Ingots of  $Nd_{60}Fe_{30}Al_{10}$  alloy were processed from the elements Nd (99.95%), Fe (99.99%) and Al (99.99%) in an arc furnace under Ar atmosphere. They were then melt spun at tangential wheel speeds of  $5\,m/s$  (V5) to obtain ribbons 2–3 mm wide and 150  $\mu$ m thick. Magnetic measurements were conducted in a VSM magnetometer for fields up to 1.5 T, at 300 and 480 K. The internal damping ID and the dynamic shear modulus G were measured as functions of temperature in an automated sub-resonant forced torsion pendulum at fixed frequencies between 0.1 and 1 Hz [150–500 K],

<sup>\*</sup> Corresponding author. Tel.: +54 351 433 4051; fax: +54 351 433 4054. E-mail address: urreta@famaf.unc.edu.ar (S.E. Urreta).

The ID spectra are analyzed considering peak contributions ( $ID_P$ ) superimposed to a background ( $ID_B$ ) given by [7]:

$$ID_B = a_0 + \frac{a_1}{T} \exp\left[-\frac{\Delta G_B}{k_B T}\right] \tag{1}$$

$$ID_{P} = ID_{MAX} \frac{1}{\cosh\left(\frac{\Delta G_{P}}{k_{B}}\left(\frac{1}{T_{P}} - \frac{1}{T}\right)\right)}$$
 (2)

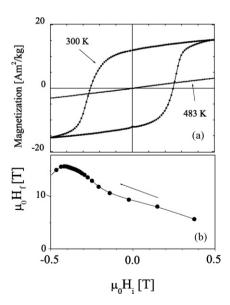
where  $a_0$  and  $a_1$  are constants,  $ID_{MAX}$  the peak height,  $\Delta G_B$  and  $\Delta G_P$  are the activation energies of the mechanisms contributing to the background and the peak, respectively, and  $k_B$  and T are the Boltzmann constant and the absolute temperature.

The thermally activated magnetic relaxation was characterized by estimating the mean fluctuations field  $\mu_0 H_f = -(\partial H_i / \partial \ln R)|_j$  from major hysteresis loops  $J(H_i)$ , traced at different field rates R as described in [5]. This fictitious field measures the effect of thermal fluctuations on the stability of the sample magnetic polarization.

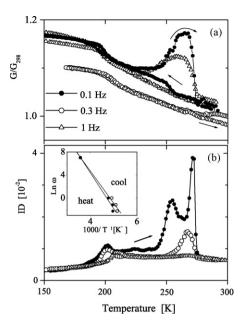
## 3. Results and discussion

The room temperature values of the mean fluctuations field is shown in Fig. 1, as a function of the applied field, together with the hysteresis loops measured at two temperatures: one above (483 K) and another one below (300 K) the Curie temperature of the Fe-rich phase ( $T_C$ =460 K). As expected, the sample is paramagnetic at high temperature and hard ferromagnetic below  $T_C$ . The actual value of the fluctuations field is 14.5 mT at the coercive field, which leads to an activation length of 7 nm if the spontaneous polarization of the  $\mu$  phase is considered ( $J_S$ =0.85 T). It may be observed that the fluctuations field is quite large at zero applied field, suggesting that thermally activated changes in the magnetic microstructure are relatively easy near and above room temperature.

The low temperature ID spectra and the corresponding dynamic shear modulus *G*, measured at three different frequencies, are shown in Fig. 2. The spectra measured during heating are plotted after subtraction of a background increasing with temperature given in Eq. (1). The peak observed at 200 K (1 Hz) shifts to lower temperature when the measuring frequency decreases and it is accompanied by a sharp step in the elastic modulus confirming that it originates in an anelastic relaxation mechanism. Assuming that it is the same peak previously observed at 280 K (1090 Hz) [6] the activation energy was estimated from the Arrhenius plot



**Fig. 1.** (a) Hysteresis loops showing a hard ferromagnetic behavior at 300 K and a paramagnetic one at 483 K, above the Curie temperature (460 K). (b) The mean fluctuations field as a function of the applied field, measured along the major demagnetization loop.

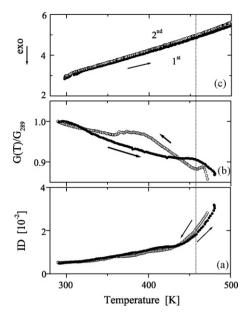


**Fig. 2.** (a) Dynamic shear modulus G as a function of temperature during thermal cycles at different frequencies. (b) ID spectra measured during heating. Inset: Arrhenius plot corresponding to the ID peak at 210 K (1 Hz).

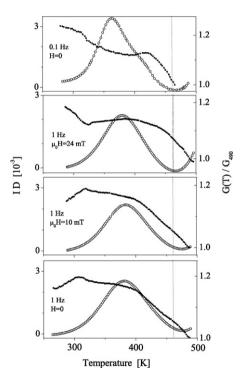
shown in the inset of Fig. 2b. The peak temperatures are different for heating and cooling measurements, so the peak parameters resulted  $\Delta G_H = 51.7 \, \text{kJ/m} \, (0.54 \, \text{eV})$  and  $\tau_{0H} = 10^{-12\pm1} \, \text{s}$  during heating and  $\Delta G_C = 46.3 \, \text{kJ/m}$  and  $\tau_{0C} = 10^{-11\pm1} \, \text{s}$  during cooling. These parameters are consistent with a stress induced relaxation mechanism, controlled by interstitial atomic diffusion, more likely of H atoms [8].

As previously reported [6], local ID maxima are also detected in the temperature range between 240 and 270 K. After background subtraction, it is clear that peak heights increase as the measurement frequency decreases. Such contributions, found in the same temperature range for frequencies between 0.01 Hz and 1 kHz, are likely to originate in a structure or magnetic transformation. The large increase in the dynamic shear modulus detected during heating is reproducible during subsequent measurements, and it becomes more evident as frequency decreases to 0.01 Hz. This modulus behavior is characteristic of transformations and may be connected with the paramagnetic  $\delta$  phase small particles, which order antiferromagnetically below 250 K [3], as previously suggested.

The ID and the relative elastic shear modulus measured during heating and cooling the alloy above room temperature up to 500 K are shown in Fig. 3. These curves are practically reproduced during subsequent measurements. No large structural changes were detected during these thermal cycles, as shown by the DSC curves measured during the first and the second heating of the as cast alloy, indicating that damping is not due to irreversible changes in the atomic microstructure. The ID and the elastic modulus G both showed heating/cooling hysteresis, being particularly large in the modulus below the Curie of the  $\mu$  phase, indicated in the plot by the vertical line. In this high temperature range the ID spectra were not well fitted by only an exponential background as that given in Eq. (1), being the low temperature flank of a high temperature peak [6]. Instead, it was necessary to consider the superposition of a background and a broad maximum near 400 K (1 Hz). Fig. 4 illustrates the maximum resulting after background subtraction (Fig. 4a) and the elastic modulus (Fig. 4b), measured during cooling from 500 K. It may be observed that small magnetic fields slightly affect the maximum position and height, and that it



**Fig. 3.** Internal damping ID (a), the relative shear modulus  $G/G_{298}$  measured during heating and cooling the alloy between 300 K and 500 K (b), and the DSC heat flux (c), measured during the first and the second heating of an as cast sample.



**Fig. 4.** Internal damping ID after background subtraction, measured during cooling, for different frequencies and applied magnetic fields and the corresponding relative elastic shear modulus  $G/G_{480}$ .

shifts to lower temperature when frequency reduces. The dynamic modulus dependence on frequency, field and temperature is quite complex, and cannot be systematized by a simple anelastic relaxation mechanism. It is then confirmed that the major anomalies in the modulus coincide with the local maximum in the internal dissipation detected between the two Curie temperatures of the  $\delta$  phase (250 K) and that of the Fe-rich  $\mu$  phase (460 K). In this range small paramagnetic Nd-rich phases (2 nm [3]) are finely dispersed in a Fe-rich matrix containing ferromagnetic clusters of the  $\mu$ -NdFeAl phase.

### 4. Conclusions

The low frequency internal damping spectra of  $Nd_{60}Fe_{30}Al_{10}$  magnetically hard alloys could be described, in the temperature range between 150 and 500 K, as the superposition of four contributions: an exponential background, continuously increasing with temperature, a relaxation peak at 200 K, involving interstitial diffusion of H atoms, and two broad damping maxima, with large anomalies in the dynamic modulus G associated to them, lying at about 270 and 400 K.

The maximum at 270 K is tentatively associated to the magnetic ordering of the  $\delta$  phase, while the origin of the other one, covering practically the temperature range between room temperature and the magnetic ordering temperature of the  $\mu\text{-like}$  phase, remains unclear.

## Acknowledgement

This work has been partially supported by SECyT-UNCOR, SECyT-UNC and CONICET, Argentina.

## References

- [1] S. Schneider, A. Bracchi, K. Samwer, M. Seibt, P. Thiyagarajan, Appl. Phys. Lett. 80 (no 10–11) (2002) 1749–1751.
- [2] R. Sato Turtelli, D. Triyono, R. Grössinger, H. Michor, J.H. Espina, J.P. Sinnecker, H. Sassik, J. Eckert, G. Kumar, Z.G. Sun, G.J. Fan, Phys. Rev. B 66 (2002) 054441.
- [3] R.W. McCallum, L.H. Lewis, M.J. Kramer, K.W. Dennis, J. Magn. Magn. Mater. 299 (2) (2006) 265–280.
- [4] C.E. Rodríguez Torres, A.F. Cabrera, F.H. Sánchez, O.V. Billoni, S.E. Urreta, L.M. Fabietti, J. Magn. Magn. Mater. 267/1 (2003) 92–96.
- [5] O.V. Billoni, S.E. Urreta, L.M. Fabietti, J. Magn, Magn. Mater. 265 (2) (2003) 222–233.
- [6] G.C. Tarnowski, H.R. Salva, A.A. Ghilarducci, O.V. Billoni, S.E. Urreta, L.M. Fabietti, Phys. B: Phys. Condens. Matter 354 (1-4) (2004) 220-223.
- [7] A.S. Nowick, B.S. Berry, Anelastic Relaxation in Crystalline Solids, Academic Press, New York/London, 1972.
- [8] S. Takeuchi, T. Yagi, T. Imai, R. Tamura, Mater. Sci. Eng. A 375–377 (2004) 455–459.