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$Nd_{60}Fe_{30}Al_{10}$ Glass Forming Magnetic Alloys: a Mechanical Spectroscopy Study at the 300- 560 K Temperature Range

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Abstract. The ferromagnetic amorphous phase in rapidly solidified $Nd_{60}Fe_{30}Al_{10}$ glass forming alloys is investigated in melt spun ribbons (100 μm thick) and in chill cast cylinders (2 mm diameter). The amorphous resulting for these two different quenching rates were characterized by their room temperature hysteresis loops, magnetization and differential calorimetric measurements in the temperature range 300K-900K. The mechanical damping was explored in the 300-560 K temperature range, by measuring the internal friction and the shear modulus in a forced inverted pendulum operating in the frequency range 0.1-10Hz. Simultaneously, the electrical resistance of the samples was measured. The internal friction spectra of both, ribbons and a cylinder, exhibit a local maximum at about 500K, arising in a relaxation mechanism. After some thermal cycles the peak parameters stabilize reaching an apparent activation enthalpy of 1.5 eV and a limit relaxation time $\tau_0 \approx 0.4\text{-}2.5 \cdot 10^{-17}$s. In both samples, the electrical resistance largely decreases during the first heating run to 560K and remains unchanged during subsequent thermal cycles. No changes in the elastic modulus or in the damping properties are detected at the Curie temperature of the alloys.

Introduction

$Nd_{60}Fe_{30}Al_{10}$ alloys exhibit, for a wide range of quenching rates from the melt, a quite large volume fraction of a ferromagnetic amorphous phase, which is responsible for the hard magnetic properties observed [1,2]. The alloy also contains minor crystalline phases identified as dhcp-Nd and the δ-NdFeAl stable phases, which are paramagnetic above room temperature. The ferromagnetic phase is a nanocomposite [3], with small Nd rich crystallites (2-10 nm) embedded in a Fe rich amorphous (or crystalline nanocluster containing [4]) phase. The relatively high coercivity of these alloys is attributed to a mechanism of strong pinning of domain walls by the small Nd rich paramagnetic crystallites in the amorphous phase.

The Curie temperature of the alloy is about 450K – 510K and after crystallization (at about 780K, depending on the quenching rate) it becomes paramagnetic. In this work we explore, using mechanical spectroscopy, the evolution of two microstructures, obtained by quenching the $Nd_{60}Fe_{30}Al_{10}$ alloy at different rates, when heated to the temperature range where pre-crystallization atomic rearrangements in the amorphous matrix are expected.

Experimental

The alloy (nominal composition $Nd_{60}Fe_{30}Al_{10}$) was prepared by arc melting of 99.99 % pure Al and 99.9 % pure Nd and Fe; it was further processed by melt spinning onto a single copper wheel at tangential speed of 5m/s to obtain ribbons (100μm thick) and also by suction casting into a water chilled mould 2mm diameter and 5 cm long.

Characterization of samples were performed by calorimetric and magnetic measurements. The microstructure evolution was monitored by DSC calorimetric measurements sweeping the temperature range between 300 K and 900 K at 5 K/min. The magnetic properties of the alloys, hysteresis loops and magnetic moment vs. temperature curves, were measured in a Vibrating

Sample Magnetometer (VSM). Anelastic measurements were done using a forced inverted torsion pendulum to measure the internal friction (IF) and the anelastic torsion modulus (G) in the frequency range 0.1-10 Hz. The electrical resistance was simultaneously monitored during the successive thermal cycles imposed to the samples during IF and G measurements. The temperature range used was 300-560 K with 1 K/min for increasing or decreasing temperature. Ribbons were measured in a very light pendulum (3 mm diameter) while one of 5 mm diameter was used for cylinders; in both cases they were 50 cm long. Three ribbons of the same batch and one cylinder were measured. All measurements were done in 200 mmHg helium gas atmosphere.

Results

The DSC first and second scans corresponding to the ribbon sample are shown in Fig. 1. No evident exothermic effects are detected in the range 300K- 600K; at about 760K an exothermic peak develops, associated to general crystallization. The same behavior is observed in the cylinder. This indicates that no new phases appear during heating to 760 K, however, limited atomic rearrangements in the amorphous phase cannot be excluded.

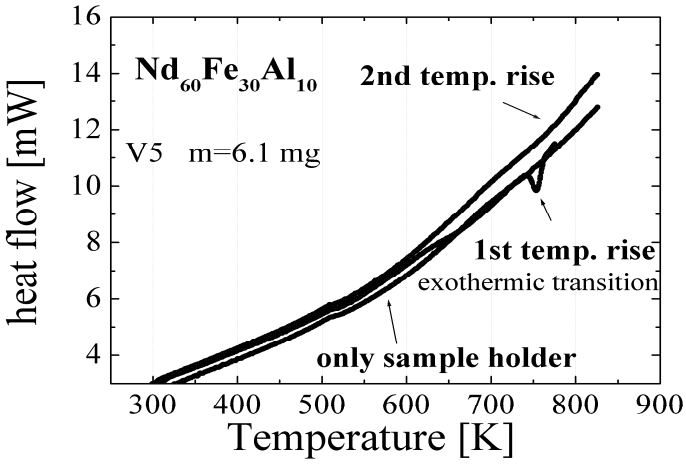

Fig. 1. DSC scans of an as cast ribbon. No evident exothermic effects are found in the range 300K-600K. The exothermic peak observed at 760K is attributed to crystallization.

The paramagnetic/ferromagnetic transition takes place near 460 K in ribbons and at about 470K in cylinders (Fig. 2). These distinct Curie temperatures can be explained by considering that the amorphous phases in ribbons and cylinders differ in composition or in the extent of the Fe-rich clustering. These different ferromagnetic phases should be responsible for the slightly higher saturation polarization in the cylinder (Fig. 3), leaving practically unchanged the coercive field, mainly determined by the size and distribution of the Nd rich, small paramagnetic crystallites, embedded in the amorphous matrix.

The IF spectra show a peak at around 500 K in both, ribbons and cylindrical samples, which attenuates and shifts to low temperature during the subsequent heating-cooling cycles.

After about two cycles the IF peak reaches a stationary height (Figs. 4-5). The electrical resistance (Fig. 6) also decreases during heating; it drops nearly 20 % during first heating up to a temperature very close to the magnetic transition (~ 450 K), where it shows a small dip, and then it further decreases in 7 % as temperature increases up to 560 K; then, its value remains low during the subsequent thermal cycles. The internal damping and the electrical resistance, quite sensitive to the internal stress state and to the atomic arrangement in the sample; change only during the few initial cycles, indicating that the metallic matrix evolves to a more relaxed condition.

The peak temperature depends on frequency and G shows the characteristic step near the peak temperature, indicating that the effect arises in a thermally activated relaxation mechanism. In ribbons, the activation enthalpy and the relaxation time τ_0 (Fig. 7) changes as the number of cycles increases.

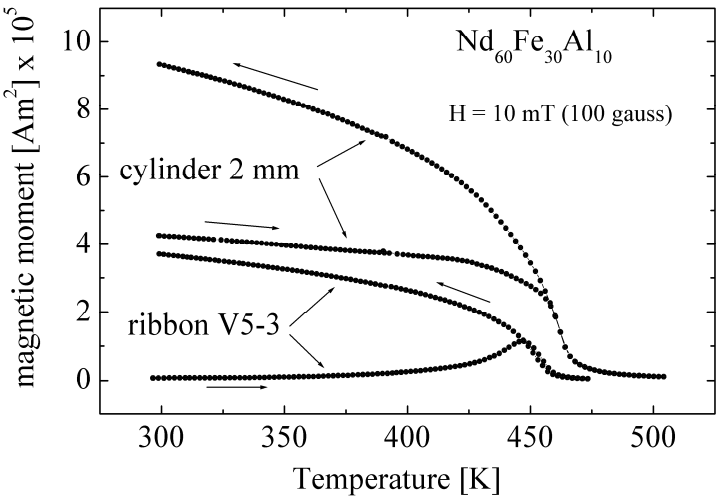

Fig. 2. Magnetic moment as a function of temperature, showing Curie temperatures of 460K and 470 K for ribbons and cylindrical samples, respectively.

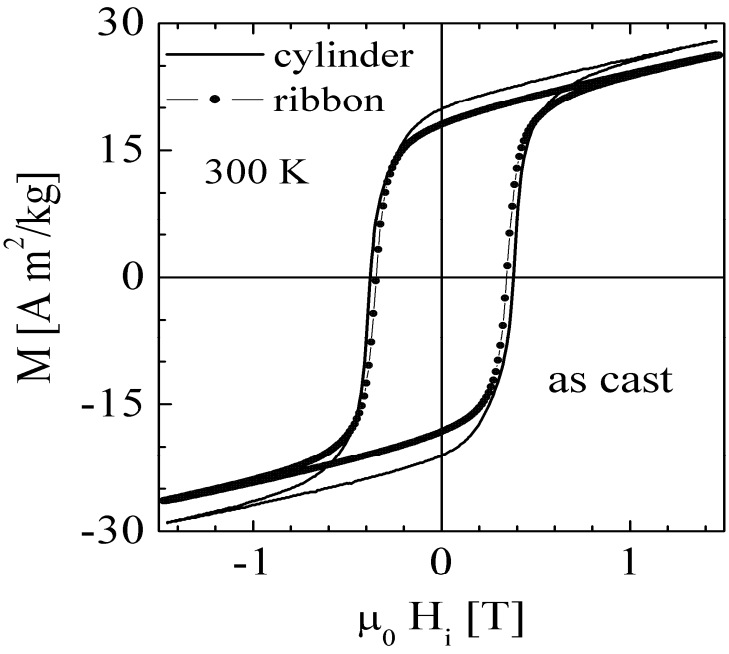

Fig. 3. Room temperature hysteresis loops for the microstructures investigated, exhibiting a quite similar hard magnetic behavior.

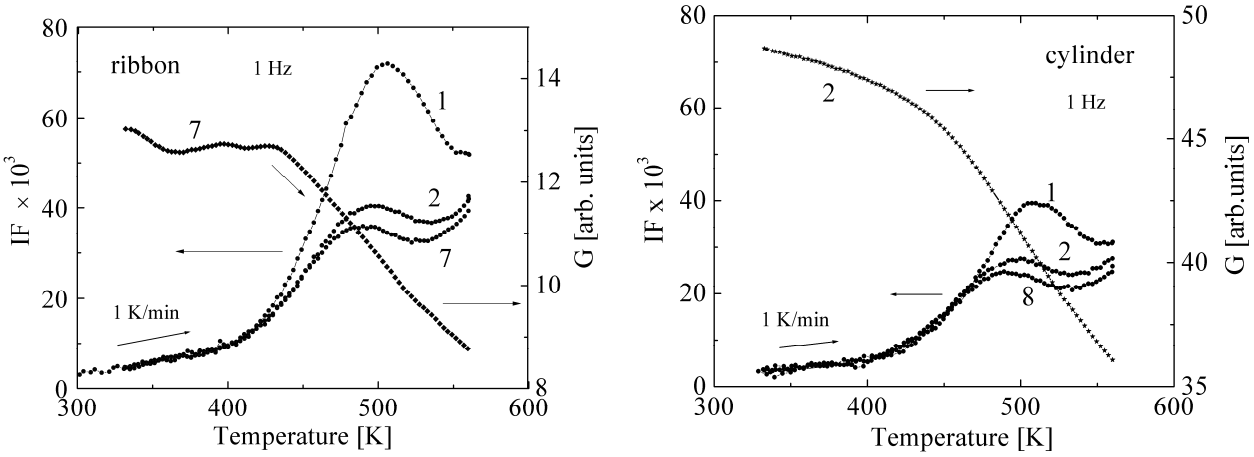

Fig. 4. Internal friction IF as a function of temperature in the ribbon and cylinder samples. The numbers at the side of the data indicates the nth thermal cycle. The dynamic elastic modulus for cycle 7 (ribbon) and 2 (cylinder) are also shown. Measurements are taken during heating at 1 K/min.

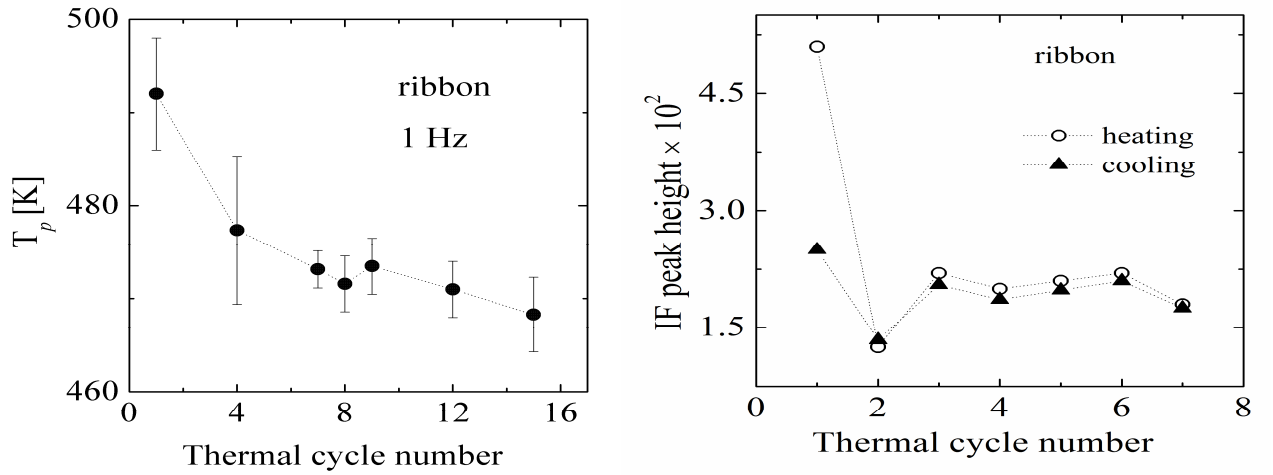

Fig. 5. Evolution of the anelastic peak parameters in ribbons as the number of measurement thermal cycles increases. After a few cycles the relaxation peak becomes stationary.

In ribbons, the activation enthalpy decreases from 1.9 eV to 1.5 eV while τ_0 increases from $4x10^{-21}$ s to $2.5x10^{-17}$ s. In the cylinder (Fig. 8), cooled at a lower rate, the activation parameters have only a small change after 22 thermal cycles; the final values are 1.55 eV and $0.4x10^{-17}$ s, respectively. The effect of the deformation amplitude on the internal damping was investigated: IF vs deformation amplitude measurements performed in the cylinder showed no change in peak temperature or height (< 0.5 %) for amplitudes changing by a factor four. It is worth to note that no changes in the IF or the elastic modulus G take place when crossing the Curie temperature in both samples; this indicates that the peak has no magnetomechanical contribution.

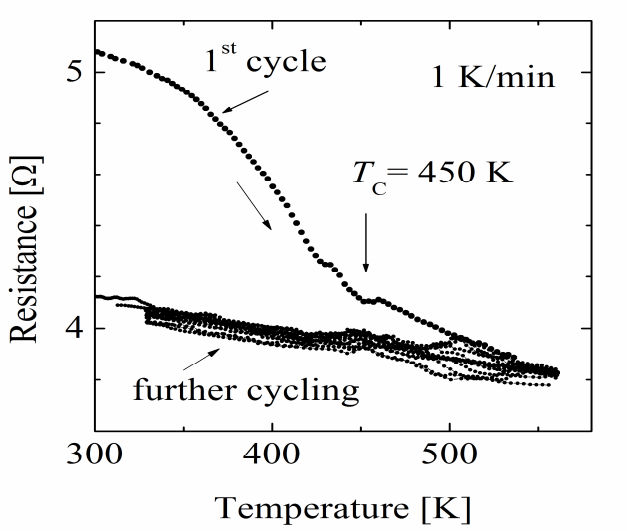

Fig. 6. Electrical resistance as a function of temperature simultaneously monitored during the internal friction measurements of Fig. 4. As observed in the internal friction spectra, the largest irreversible changes take place during the first heating to 560K.

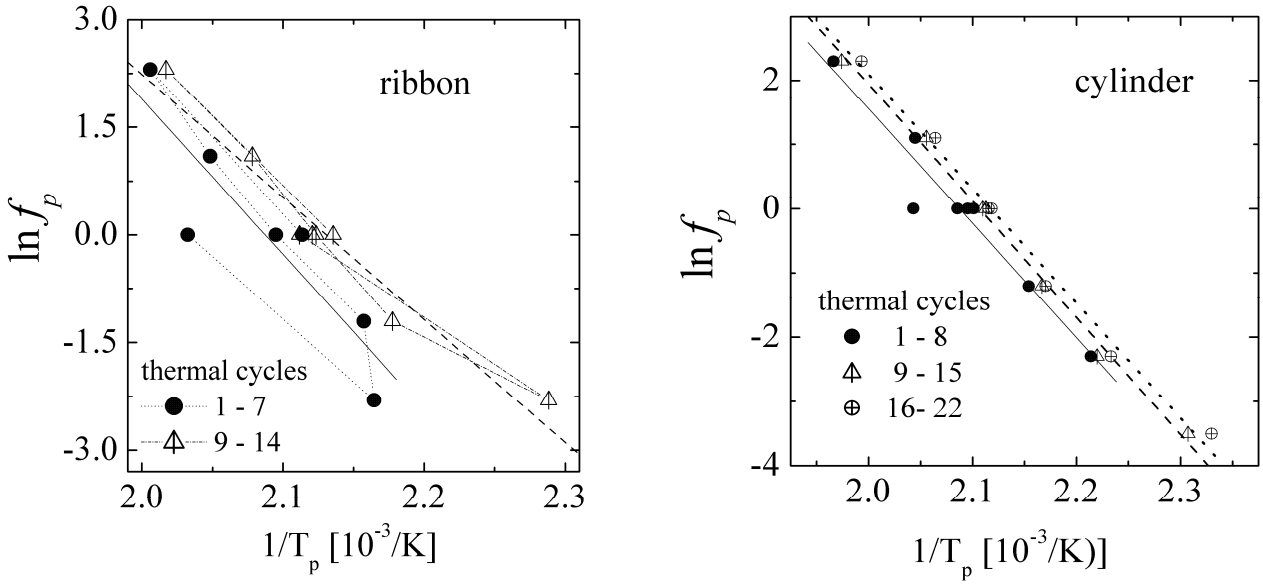

Fig. 7. Arrhenius plots corresponding to ribbons and cylinder illustrating the effect of successive heating-cooling cycles on the apparent activation enthalpy and limit relaxation time of the damping mechanism.

Summary

The internal friction damping spectra show a relaxation peak in both, ribbons and cylindrical samples, with similar activation parameters: 1.5 eV and $1x10^{-17}$ s for the apparent activation enthalpy and the limit relaxation time, respectively.

The internal friction peak of the cylinder is nearly the half of that of the ribbons. This may indicate that the origin of the observed relaxation is in the amorphous phase of the alloy, because the cylinder (lower temperature cooling rate) has a lower proportion of amorphous phase than ribbons.

The IF peak parameters are amplitude independent as no change in the peak temperature or height (< 0.5 %) were detectable for deformations amplitudes changing by a factor of four.

No detectable effect of the 450 K magnetic transformation on the IF peak or modulus is observed, indicating that this relaxation is not magnetomechanical in origin.

Internal friction damping and electrical resistance measurements indicate that structural rearrangements actually take place during the few initial thermal cycles up to 560K, leading to a quite stationary microstructure, while characterization calorimetric techniques did not reveal any structural change in this range, where pre-crystallization atomic ordering is expected. The variations of peak height and temperature during the initial cycles are attributed to irreversible atomic rearrangements in the amorphous phase when approaching the crystallization temperature (750 K).

Electronic stable state is achieved faster than the mechanical structural state.

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