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FREQUENCY, DC-FIELD AND TEMPERATURE DEPENDENCE OF THE AC-SUSCEPTIBILITY OF $Nd_{60}Fe_{30}Al_{10}$ ALLOY

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The temperature dependence of the initial AC-susceptibility of melt-spun $Nd_{60}Fe_{30}Al_{10}$ ribbons shows an anomaly, which depends strongly on the heat treatment and the superposing DC-field. The anomaly is suppressed by DC-field. Additionally, the position and the amplitude of the anomaly are frequency dependent. However, the time dependence of the AC-susceptibility is observable for temperatures above 100 K. The Analysis of the susceptibility (measured with and without superposing DC-field) as function of the time at different temperatures using the Arrhenius law was performed.

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1. Introduction

Quasi amorphous Nd-Fe-(Al, Si) exhibits hard magnetic properties [1-5]. Quasi-amorphous because in these materials very small particles are always present in the amorphous matrix, but due to small size the X-ray diffraction patterns are typical for an amorphous material. In the as-cast state, microstructure, coercive field (H_c) and Curie temperature (T_C) are strongly dependent on the quenching rate (QR) and/or the production method. Generally at room temperature, the coercive field of the melt-spun ribbons increases with the thickness becoming comparable to that of bulk material (prepared by a copper mould casting). The coercive field at room temperature (RT) is almost constant with annealing temperature and shows an exponential behaviour as a function of T_C [4]. However the material becomes paramagnetic at RT after complete crystallisation [1,3]. We have explained the high H_c by a strong pinning of domain walls by inhomogeneities [4]. In our previous works, we showed the existence of an anomaly in the temperature dependence of the AC-susceptibility of the ribbons, which anomaly depends on the annealing and polishing of the surface of the wheel side [6,7]. Additionally, the time dependence of the magnetization, M, and H_c at different temperatures were investigated [6].

In the present work we investigate the temperature dependence of the magnetic properties of $Nd_{60}Fe_{30}Al_{10}$ ribbons as well as bulk material in the as-cast state and after annealing by means of initial AC- susceptibility and hysteresis loops. The influence of the annealing, the effect of frequency and the superposed DC-field on the existing anomaly in the temperature dependence of the AC-susceptibility is shown. Additionally, the time dependence analysis of the initial susceptibility at different temperatures is performed.

2. Experimental procedure

Rapidly quenched $Nd_{60}Fe_{30}Al_{10}$ ribbons were prepared by the melt-spinning technique with a wheel surface speed of 30 m/s. The thickness of the ribbons is of 30 μm. A bulk $Nd_{60}Fe_{30}Al_{10}$ specimen was prepared by pouring the melt directly into a water-cooled cooper mould, obtaining a

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ingot. For the magnetic measurements, a sample of dimensions 2 x 2 x 3 mm was prepared cutting the ingot. X-ray diffraction, XRD, measurements were performed using CoK_{α} radiation. The structural characterizations by means of transmission electron microscopy are reported in [3-8]. The temperature *T*, frequency *f* and Dc-field dependencies of the initial AC-susceptibility (in phase $\chi'(T)$ and out of phase $\chi''(T)$) was measured using a Quantum Design PPMS model 6000 applying a field of 80 A/m. Hysteresis loops were measured between 4.2 and 350 K using a pulsed-field magnetometer with a maximum applied field $\mu_0 H_{max}$ = 22 T. The measurements were performed on both as-cast and annealed materials.

3. Results and discussion

Fig. 1 shows the XRD patterns obtained for the bulk material in the as-cast state and also for the as-cast and heat treated ribbons. In ribbons, even the XRD patterns are typical for an amorphous material, we found a precipitation of small nano-particles of Nd-rich phase through the electron microscopy observation [3,8]. With a heat treatment, the particle size increases. For the sample annealed at 603 K, the new crystalline phases appear, such as Nd and $Nd(Fe_{1-x}Al_x)_2$ crystallites, with a average grains size of 20 nm, embedded in the amorphous matrix. When the ribbon is heat treated at 773 K for 10 min., the specimen is completely crystallised, with crystalline grains > 200 nm. Four crystalline phases are found: cubic Nd, hexagonal Nd, $Nd(Fe_{1-x}Al_x)_2$ and Nd_2Fe_{17} [3]. In the bulk specimen, the crystalline peaks corresponding to Nd and $NdAl_2$ phases already appear in the as-cast state. The temperature dependence of the coercive field and AC-susceptibility reflects very well the presence of these multi-phases (see details in [3,4]).

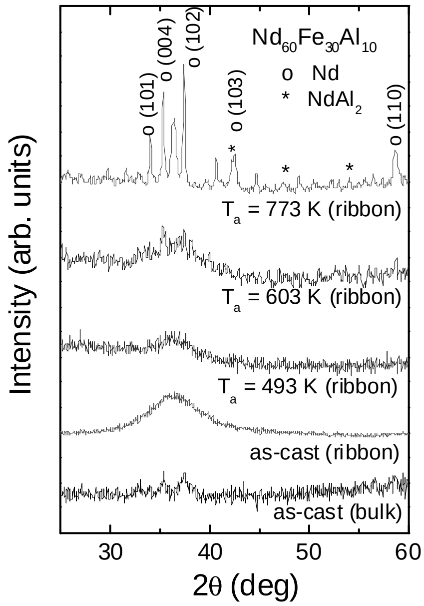

Fig. 1. XRD of the bulk material (as-cast) and ribbons (as-cast and annealed).

Generally at room temperature, the coercive field of the as-cast melt-spun ribbons increases with the thickness becoming comparable to that of the bulk material [2]. The coercive field values of ribbon and bulk are 0.038 and 0.29 T, respectively. The hysteresis loops of the as-cast ribbon and bulk materials measured at 5 K and 300 K are shown in Fig. 2. In Fig. 3 is plotted the coercive field against the temperature, $H_c(T)$, measured on as-cast and annealed at 603 K for 10 min. ribbons and also on the as-cast bulk material. As can be seen, at room temperature, the coercive field of the ribbons is nearly independent on the annealing and on the existence of Nd-rich or $Nd(Fe_{1-x}Al_x)_2$

crystalline phases. For the bulk alloy, the coercive field is relatively high even with the presence of crystals of Nd and $NdAl_2$. For all samples, the curves of the $H_c(T)$ show a maximum close to 70 K with coercive field reaching values between 2.5 and 5 T depending on the preparation method and annealing procedure. Generally, the maximum value of the coercive field reached at around 70 K, in the ribbons is larger than that in the bulk. Below this temperature, the soft magnetic Nd-rich crystalline phase becomes magnetically ordered decreasing then the coercive field. Consequently, the hysteresis loops at low temperatures are typical for a material of multi-phases, as shows Fig. 3, for example the loop measured at 5 K, with maximum field of 22 T.

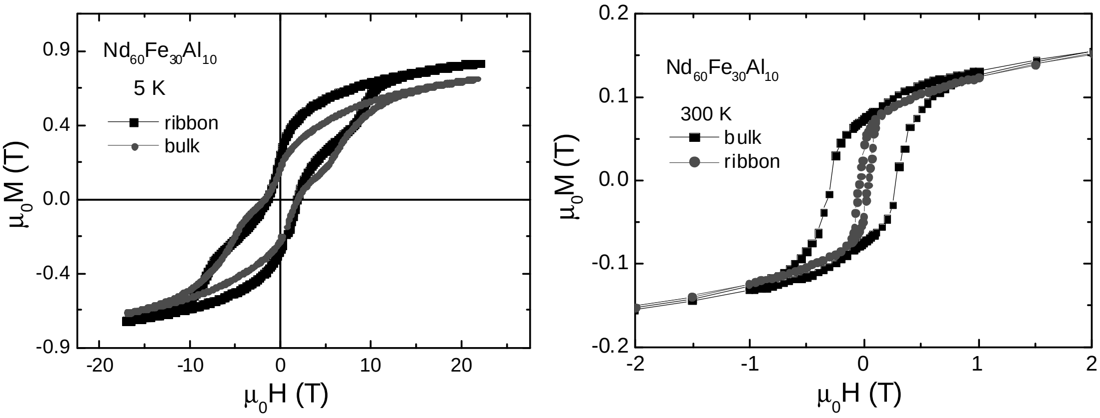

Fig. 2. Hysteresis loops measured at 5 K (left side) and 300 K (right side) on the ribbon and bulk materials, both in the as-cast state.

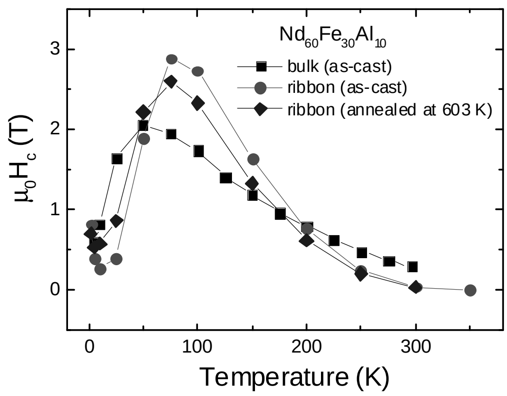

Fig. 3. Coercive field as a function of the temperature of the as-cast and annealed at 603 K ribbons and of the as-cast bulk material.

In our previous work [4], we have described the coercivity mechanism based on the existence of domain walls in the amorphous matrix, which walls are strongly pinned by magnetic in-homogeneities, such as precipitated Nd-rich particles, or by structural defects.

The temperature dependencies of the susceptibility, $\chi(T)$, measured on ribbons with different heat treatments and on the as-cast bulk material are shown in Fig. 4. As can be seen, the curves of $\chi(T)$ of the melt-spun ribbons present an anomaly above 100 K, which the anomaly changes for higher temperatures with increasing of the annealing temperature, and disappears when the sample is completely crystallized. The $\chi(T)$ curve of the ribbon in the crystallized state shows at low temperatures magnetic transitions corresponding to the cubic and hexagonal Nd phases, at 7.5 and 19.9 K, respectively, and to $NdAl_2$ phase, at 76 K. The same magnetic ordering temperatures are found for the as-cast bulk material, indicating then the presence of Nd and $NdAl_2$ phases embedded in amorphous matrix, which the precipitation may have occurred during the preparation of the sample.

It is known that the Curie temperature of Nd-Fe alloys is strongly dependent of the quenching rate QR [9]. The alloys produced with higher QR exhibits lower T_C. Croat has reported that due to different QR the T_C of the Nd-Fe alloys can vary more than 200 K [9]. Therefore, for the existence of the anomaly in the $\chi(T)$ curve, we have explained as a consequence of the non-homogeneity of the quenching rate along the thickness of the ribbon due to the inevitable temperature gradient of melt-spun material that leads to the formation of multimetastable phases. The ribbon surface, which is in contact with the copper wheel, has a significantly higher QR (giving origin to anomaly) than that of the free surface. Upon an annealing of the ribbons, the degree of the amorphous state changes (as lowering the QR) changing then the peak of the anomaly for higher temperatures. In fact, this anomalous disappears when a surface thickness f 9 μm is removed from the wheel side [7].

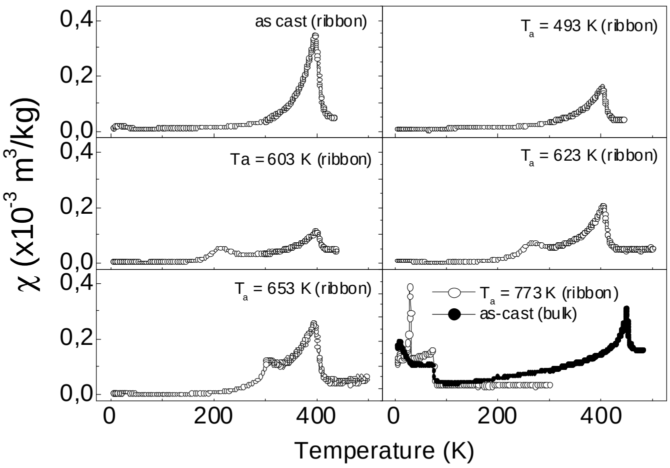

Fig. 4. Temperature dependence of the real part of the susceptibility of the as-cast bulk material and as-cast and annealed at temperatures between 493 and 773 K for 10 min.
ribbons measured with a field of intensity 200 A/m and frequency of 80 Hz.

The temperature dependence of χ' and χ'' measured on the ribbon annealed at 603 K for 10 min at different superposing DC-field to an AC-field of f = 1 kHz is shown in Fig. 5.

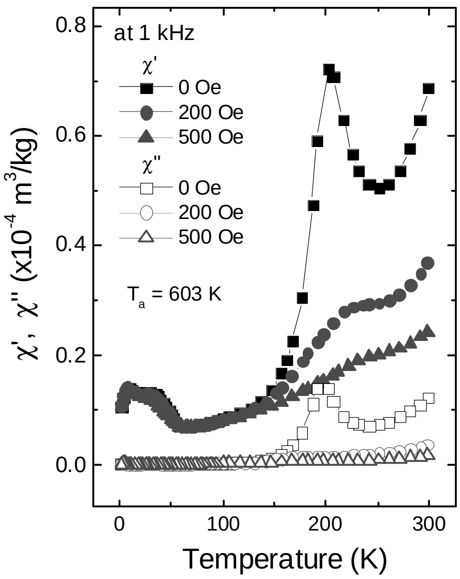

Fig. 5. χ’ and χ’’ as a function of the temperature measured with superposing DC-field of 0, 200 and 500 Oe on the ribbon annealed at 603 K for 10 min.

As in general magnetic transition, the anomalous peak decreases in the intensity when an additional DC-field is applied and almost disappears upon a DC-field of 500 Oe. As was mentioned before, our model for the coercivity mechanism is based on the existence of the domain walls. Then, the main contribution of the application of a DC-field is given by the orientation of domains in the direction of the applied field, resulting in the 180 ° domain walls. When the intensity of the applied field increases, the anomaly must reduce its height due to reduction of the number or thickness of domain walls.

The $\chi'(T)$ and $\chi''(T)$ curves obtained at different f without and with DC-field of $\mu_0 H_{DC}$ = 500 Oe are shown in Figs. 6 and 7, respectively. In both figures, a significant decreasing of both χ' and χ'' with increasing f occurs for temperatures above 100 K, even for low frequencies. It is worth to cite that the pulsed-field rate (or time) dependence of the coercive field of the same sample was also found for temperatures higher than 100 K [6]. These sweep rate effects have observed for all other samples investigated in this work.

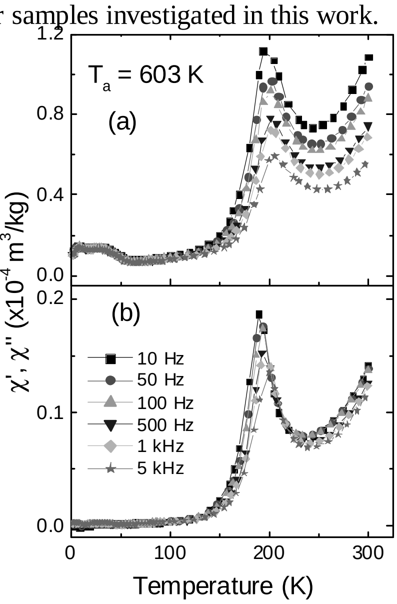

Fig. 6. χ' (a) and χ'' (b) as a function of the temperature measured at frequencies between 10 and 5000 Hz on the ribbon heat treated at 603 K for 10 min.

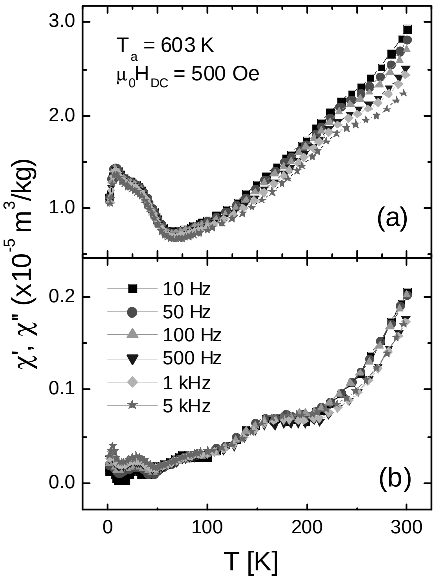

Fig. 7. χ'(a) and χ''(b) as a function of the temperature measured with superposing DC-field to Ac-field at frequencies between 10 and 5000 Hz on the annealed ribbon.

Because both χ' and χ''decrease with f and also for the region of low frequencies, the effect of the damping dominated by eddy currents can be neglected. Considering this hypothesis, the decreasing of the susceptibility can be attributed mainly to the reversible time dependence of the initial susceptibility.

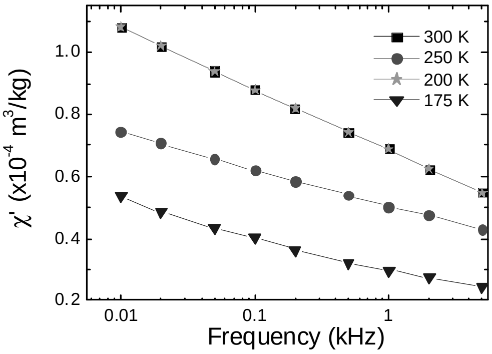

Fig. 8. χ' as a function of the f determined form Fig. 6 at temperatures 175, 200, 250 and 300 K.

Fig. 8 shows the susceptibility at different fixed T plotted against the frequency, determined from Fig. 6. The decreasing of χ' with logarithmic frequency is observed for all T and also for that by applying a DC-field. In the present work, the origin of the reversible time dependence of the initial susceptibility process is still unclear. However, it is clear that due to the logarithmic behavior, a distribution of relaxation time τ exists, which in condition $1/f < \tau$ causes a loss in the susceptibility. A detailed theoretical investigation is in progress, which result can contribute to understanding of the time dependence of the coercive field.

4. Conclusions

The AC-susceptibility measurements showed the influence of the frequency and superposing DC-field on the existent anomaly in $\chi(T)$ curves. The DC-field suppresses the anomaly and diminishes the intensity. The frequency dependence of the susceptitibility at a fixed temperature obeys the logarithmic law indicating then that the relaxation process exhibits a distribution of the relaxation time. At room temperature, the coercive field of the ribbons is lower than that of the bulk, but for lower temperatures, the coercive field of the ribbons becomes larger.

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