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Anisotropy energy distribution determined by mössbauer spectroscopy in a metallic glass

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Abstract

The distribution of frozen-in magnetic anisotropy in as-quenched $Fe_{73.5}Si_{13.5}Cu_1Nb_3B_9$ amorphous melt-spun ribbons was studied by Mössbauer effect spectroscopy, using the temperature-dependent magnetoelastic effect produced on the metallic glass by 1 μm Al coatings. Al coatings were deposited by RF sputtering at $T \approx 350\,K$ on both sides of the amorphous ribbons. Estimated magnetic anisotropy values were below $1\,kJ/m^3$, with preeminence of anisotropy energy densities lower than $300\,J/m^3$.

Keywords: Amorphous ribbons; Magnetoelastic effects; Mössbauer spectroscopy; Sensor devices

1. Introduction

Intrinsic stresses determine the domain structure in amorphous metal melt-spun ribbons. The application of external stresses also induces magnetoelastic anisotropy and modifies the domain structure. For example, tensile stresses favor a longitudinal magnetization easy axis [1] (for positive magnetostriction). Magnetoelastic effects have been currently used in several magnetoelastic sensor devices. Recently, sensing devices have been proposed where melt-spun amorphous ribbons are glued on one or both sides with materials of different nature. The components exhibiting different thermal expansion coefficients make these composite multilayers good candidates for temperature sensors [2–4]. In these devices, sensing is performed through permeability, which is modified by the stress. Consequently, in-depth knowledge about mechanical stresses in those materials is also of interest for practical reasons. In most of these prototypes, 20 μm-thick melt-spun amorphous ribbons and different metallic coatings with thickness between a few micrometers and several tens of micrometers, stuck to the ribbons by means of a special glue, were used.

In this paper, it will be shown that Mössbauer effect (ME) spectroscopy offers the possibility of obtaining information on the magnetoelastic effects created by melt-spinning processing and other sources of stresses (e.g., those generated by glued in the above-mentioned sensor devices). ME spectroscopy presents the advantage of sampling the distribution of magnetization direction within the sample bulk as well as in the surface neighborhoods. The mapping is performed easily by evaluating the relative intensities of absorption lines [5]. This methodology averages the information over the whole material, therefore providing only the mean value of the magnetization direction. However, as essentially just domains that follow either an easy axis parallel to the ribbons' surface or perpendicular to it have been observed in these melt-spun materials [6], the amount of in-plane and out-of-plane magnetic moment can be readily estimated.

The objective of this work is to present original results about the distribution of bulk anisotropies induced by the stresses frozen-in during the ribbons fabrication, as well as

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after sputtering films of Al on both sides of an amorphous ribbon.

2. Experimental

Twenty micron thick amorphous $Fe_{73.5}Si_{13.5}Cu_1Nb_3B_9$ alloy ribbons were prepared by the melt-spinning technique. Pieces of ribbons were coated on both sides with 1 μm-thick Al layers by RF sputtering at a temperature of approximately 350 K; we will label these samples as *AaA* (Al/amorphous/Al) trilayers.

Mössbauer experiments were carried out with a standard constant acceleration spectrometer, under transmission geometry, using a ^{57}Co*Rh* source of about 20 mCi.

3. Experimental results and discussion

Fig. 1 shows some of the Mössbauer spectra obtained from the ***AaA*** sample at different temperatures. The patterns of six broad absorption lines are the typical ones observed for magnetic metallic glasses. The mean orientation of Fe magnetic moments can be calculated from the Mössbauer spectrum using the well-known expression for the absorption line relative intensities $3{:}z{:}1{:}1{:}z{:}3$, where $z = 4\sin^2(\theta)/(1+\cos^2(\theta))$, θ being the angle between the incident γ-ray and the direction of the magnetization [5]. The incident photon direction was along the normal to the ribbon surface. As shown in Fig. 1, the relative intensities of the absorption lines change with temperature.

The temperature dependence of the intensity ratio of lines 2–3, $I_{23} = z$ can be better observed in Fig. 2 for coated and uncoated amorphous ribbons. While I_{23} strongly depends on temperature for the ***AaA*** sample, within experimental error it remains constant for pure amorphous ribbons.

For the trilayers used in this work, important tensile or compressive stress appears on the individual layers when

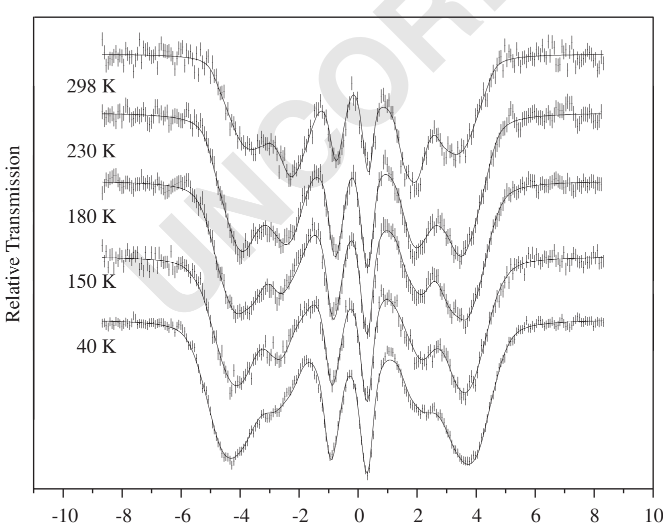

Fig. 1. ME spectra of *AaA* sample obtained at different temperatures.

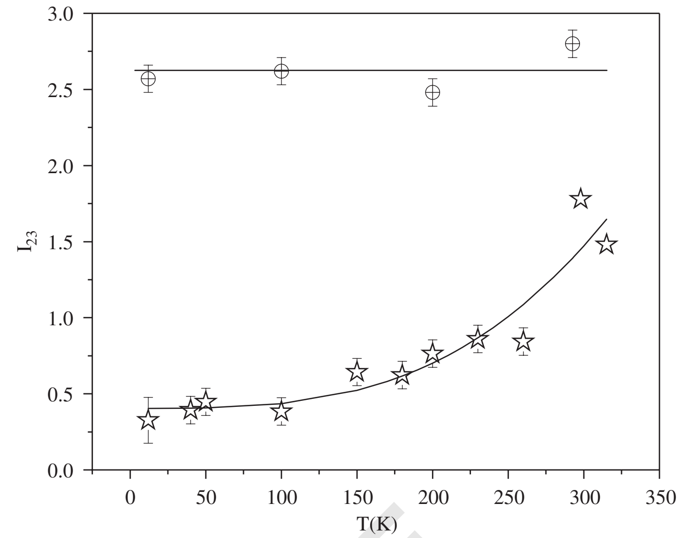

Fig. 2. Temperature dependence of I_{23} for pure amorphous (circles) and *AaA* samples (stars). The full lines are only guide to the eyes.

temperature T is changed. The effective linear thermal expansion coefficient α_{eff} of the ***AaA*** system can be estimated from a simple physical model [2].

The strain undergone by the metallic glass for a temperature variation ΔT, relative to that which would occur in the absence of the coating is given by $\Delta\varepsilon = \varepsilon - \varepsilon_{\mathrm{Am}} = (\alpha_{\mathrm{eff}} - \alpha_{\mathrm{Am}})\,\Delta T$ and the stress it experiences by

$$\sigma = E_{\mathrm{Am}}\Delta\varepsilon = E_{\mathrm{Am}}(\alpha_{\mathrm{eff}} - \alpha_{\mathrm{Am}})\Delta T, \qquad (1)$$

where α_{Am} ($\approx 12\times10^{-6}\,\mathrm{K}^{-1}$) and E_{Am} (≈ 150 Gpa) are the linear thermal expansion coefficient and Young's Modulus of the amorphous. In our case, the relative thermal expansion coefficient $\alpha_{\mathrm{eff}} - \alpha_{\mathrm{Am}}$ is about $5.4\times10^{-7}\,\mathrm{K}^{-1}$.

The main energy term affecting magnetization distribution is the local bulk anisotropy present in the amorphous material induced by the ultra-rapid freezing process. Essentially two types of domain patterns have been observed in melt-spun metallic glasses: wide domains that follow a local in-plane easy direction and narrow fingerprint-like ones, which follow an easy direction perpendicular to the surface [6]. A sketch of this situation is given in Fig. 3 which represents one small piece of the ribbon submitted to no external stress where regions of preferential anisotropy parallel $K_{\parallel}$ and perpendicular $K_{\perp}$ to the surface are indicated. The figure also suggests how new out-of-plane magnetization regions would develop when temperature is reduced, due to the increase of a compressive stress generated by the Al layers. In the volume $V_{\parallel}$ where $K \equiv K_{\parallel}$, volumes V_1 and V_2, where the local anisotropy $K \geqslant 3\lambda_S\sigma/2$ and $K \leqslant 3\lambda_S\sigma/2$, can be distinguished. Within V_1 and V_2 the combined effect of stress and anisotropy energy contributions leads to magnetization preferential directions given by $\theta \approx 0$ and $\theta \approx \pi/2$, respectively.

Since in V_1, $I_{23} \approx 4$ and in V_2, $I_{23} \approx 0$, the fraction $f_j = V_j/V$ (V, total volume) can be estimated by means of the expressions

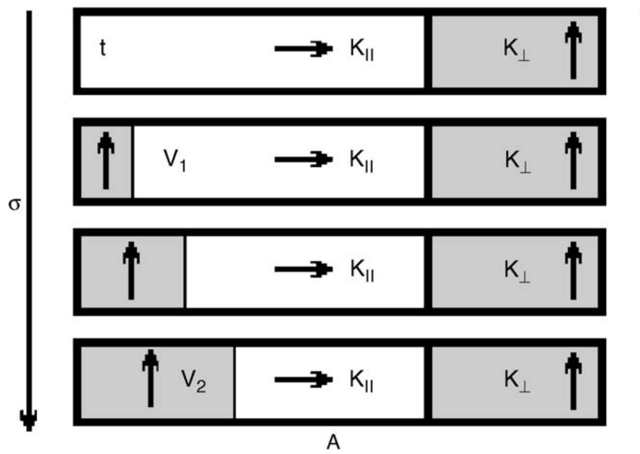

Fig. 3. Regions of 'quenched' in-plane and out-of-plane anisotropy, $K_{\|}$ and $K_{\perp}$. Small arrows indicate preferential magnetization directions. Large arrows indicate sense of increasing temperature and stress. Gray and white zones do not intend to represent the actual space magnetization distribution.

$$V = V + V_{\perp} = V_1 + V_2 + V_{\perp},$$

$$I_{23} = 4f_1,$$

$$f_2 = 1 - f_1 - f_{\perp} = 1 - f_{\perp} - I_{23}/4.$$

Therefore, f_2 can be estimated experimentally, taken into account that $f_{\perp}$ can be estimated from the value of I_{23} obtained from uncoated amorphous ribbon, where $f_2 = 0$,

$$f_{\perp} = 1 - I_{23}(\sigma = 0)/4 \approx 0.36.$$

It is possible now to find a relationship between f_2 and the critical value of $K_{\|}$, $K_C = -3\lambda_S\sigma/2$, by noting that

$$f_2 = \frac{1}{V}\int_{K=0}^{K_C=-3\lambda\sigma/2} \mathrm{d}V(K_{\mathrm{II}}).$$

Fig. 4 is a plot of f_2 vs. K_C built assuming $\sigma = 0$ at 350 K; it gives the distribution of $K_{\|}$ within the material. It indicates that frozen in-plane anisotropy is lower than 1 kJ/m^3. Furthermore, volume fraction f_2 increases more rapidly for small K_C values indicating that regions with in-plane anisotropy $K_C \leqslant 3 \times 10^2\,J/\mathrm{m}^3$ are more abundant than those with higher K_C values.

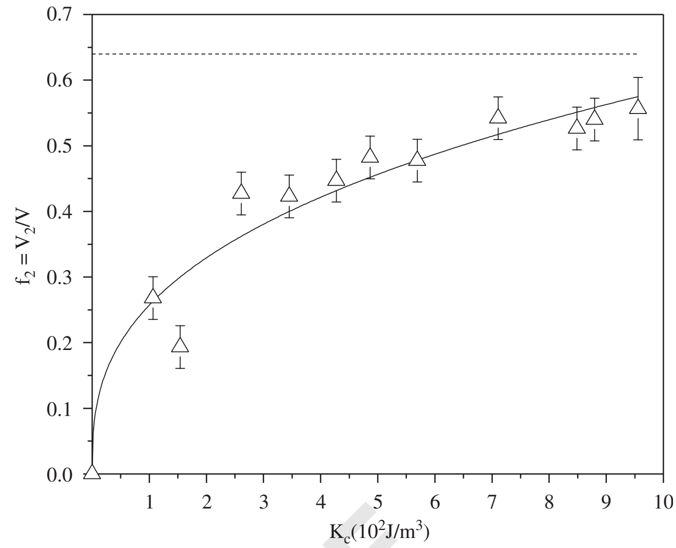

Fig. 4. Empirical estimation of the relationship between the volume fraction where the magnetization is varied from in-plane to out-of-plane by means of the Al-induced compressive stress, and the critical value of in-plane anisotropy for a given stress, $K_C = -\lambda_S\sigma$ (triangles). The full line is a potential function fit to the data. The dashed line represents the expected upper limit of $f_2 = 0.64$.

4. Summary and conclusions

It was found that 1 μm Al coatings on both sides of an amorphous melt-spun ribbon produce noticeable changes of the intensity ratio I_{23} of the ME spectra absorption lines due to the evolution of magnetoelastic effects with temperature. Within the studied temperature range $\mathrm{d}I_{23}/\mathrm{d}T$ takes its largest values near RT, being about 0.001 K^{-1} at 100 K and 0.01 K^{-1} at 293 K. Using a simple model for the distribution of internal stress in pure as-quenched ribbons, the magnitude of the internal stress induced anisotropy $K_{\|}$ could be estimated, as well as its intensity distribution. As the effect of Al coating increases monotonously with its thickness, it is expected that $\mathrm{d}I_{23}/\mathrm{d}T$ values much larger than those presented here can be obtained, thus reducing the temperature interval where main changes take place.

In an on-going work, the study of the effects of different coatings is being extended and deepened, including the investigation of the systems' response at higher temperatures.

Acknowledgments

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