Spin reorientation in Al/Metglas 2605S2/Al trilayers induced by magnetoelastic effect

P. S. Moscon,¹ E. C. Passamani,¹,4) C. Larica,¹ A. Y. Takeuchi,¹ F. H. Sánchez,² P. Mendoza Zélis,² and Elisa Baggio-Saitovitch³

¹Departamento de Física-CCE, Universidade Federal do Espírito Santo, Campus de Goiabeiras, Vitória, Espírito Santo 29075-910, Brazil
²Departamento de Física-FCE, Universidade Nacional de La Plata, C.C.67, 1900 La Plata, Argentina
³Centro Brasileiro de Pesquisas Físicas, R. Dr. Xavier Sigaud 150, Urca, Rio de Janeiro 22290-180, Brazil

(Received 17 March 2008; accepted 1 July 2008; published online 9 September 2008)

Mössbauer spectroscopy, in a broad temperature interval of 12–425 K, has been applied to investigate the spin reorientation dynamics caused by the temperature induced magnetoelastic effect on Al(x μm)/Metglas 2605S2 (20 μm)/Al(x μm) trilayers (x=0; 2.5; 5 and 20). It was found that the angle between the average sample magnetization and gamma ray direction (perpendicular to the sample plane) depends on the Al layer thickness. For temperatures smaller than 260 K, saturation of spin reorientation, which can be controlled by adjusting the Al thickness, was reached for Al thicknesses larger than and equal to 5 μm. For a 20 μm Al thickness, changes in the ⁵⁷Fe atom spin and charge densities have also been observed. A simple spin model has been proposed to describe qualitatively the spin reorientation effect as well as the influence of the Al thickness on the spin reorientation sensitivity. © 2008 American Institute of Physics. [DOI: 10.1063/1.2974801]

I. INTRODUCTION

Soft magnetic melt-spun amorphous ribbons still attract special attention from the scientific community due to the fact that these materials pose fundamental questions not yet clarified¹ and also because of their potential for technological application in electronic devices—such as transformer cores and small motors—and, especially, as sensing elements.¹–⁶

Bimetal sensing devices are usually produced by gluing surfaces of different metal layers. It is considered, in general, that the adhesive material does not play any important role in the induction of mechanical stress when the device temperature is changed or when stress is applied on the device. This relative simple setup has some advantages (cheaper to be produced), but it leads to technical inconveniences because the adhesive is a third material and certainly influences the device properties. In general, the mechanical coupling of the bimetal glued surfaces is weak compared with that obtained if the metal surfaces are homogeneously coupled (as occurs when deposition techniques are used), which may reduce the magnitude of the desired magnetoelastic effect.

The symmetrical trilayer system is a type of sensing device where the magnetostrictive material (soft magnetic amorphous alloy, am) is covered on both sides with a non-magnetic material, which has a thermal expansion coefficient different from the former one. When temperature is modified, this type of setup produces mechanical stress on the magnetic ribbon surfaces inducing a magnetoelastic effect (magnetization reorientation). Therefore, it is generally proposed for the fabrication of temperature sensors.³,⁵ In fact, it has been shown that the magnetoelastic effect leads to inductance changes (in a coil wound around the trilayer) of about 3% K⁻¹.³,⁵ Up to now, however, the basic phenomena involving the spin dynamics of the mentioned trilayer is not known in detail. Considering that the stress on the ribbon surfaces may cause modification in the (i) spin structure, (ii) spin polarization, and (iii) s-electron density around Fe probe atoms, Mössbauer spectroscopy (MS) is a suitable technique to investigate the influence of induced stress on Fe-based soft amorphous ribbons. The mentioned quantities can be directly measured from the Mössbauer spectra without having to apply an external magnetic field. In fact, MS presents the advantage of sampling the average magnetization direction within the samples by performing a calculation of the relative intensities of the second (fifth) and third (fourth) absorption lines (I₂₃ or I₅₄) of a magnetic spectrum. Consequently, the MS method may lead us to a better understanding of the magnetoelastic effects, particularly of those created by the Al layer induced stress.

In this work, we present Mössbauer results obtained at various temperatures from Al/amorphous/Al trilayer samples prepared from melt-spun Metglas 2605S2-like precursors with Al layers symmetrically deposited by sputtering. From these samples, through MS measurements, we obtain information on the influence of the stress caused by the Al layers on these ribbons when a magnetoelastic effect is induced by temperature change.

II. EXPERIMENTAL PROCEDURE

Amorphous Metglas 2605S2 alloy ribbons (am), 20 × 10 mm² and 20 μm thick, were prepared by melt spinning. The ribbons were ultrasound cleaned using three successive baths (neutral Extran detergent, distilled water, and acetone). The cleaned samples were then coated on both sides with aluminum (Al) by dc sputtering. During the Al depositions, the ribbons (substrates) were kept at tempera-
tures close to 410 K. The trilayers were designated as Al(x \, \mu m)/am/Al(x \, \mu m). The Al layer thicknesses (x) were varied from 0 \, \mu m (uncoated sample) to 20 \, \mu m. It is important to stress that the chosen am alloy is known to have a magnetostriction constant $\lambda_{am}$ of about $+32 \times 10^{-6}$, a thermal expansion coefficient ($\alpha_{am}$) of $-6 \times 10^{-6}$ K$^{-1}$, and Young’s modulus ($E_{am}$) of about 110 GPa,5,7 while bulk Al has the following parameters: $\lambda_{Al}=0$, $\alpha_{Al}=23 \times 10^{-6}$ K$^{-1}$, and $E_{Al}$ of about 70 GPa.8,9 For the Al layer thickness ($\sim \mu m$) employed in this work, bulk properties were assumed.

MS experiments were carried out with a standard constant acceleration spectrometer, under transmission geometry, using a $^{57}$CoRh source of about 50 mCi. The trilayer samples were mounted in a sample holder especially designed to avoid external stress on the samples and with the gamma ray direction perpendicular to the ribbon plane. They were put into an APD cryogenics Mössbauer setup and kept in place under the same geometry during each set of measurements at different temperatures from 12 to 300 K, while the Mössbauer radioactive source was always kept at 300 K. High temperature MS measurements were performed in a high vacuum Mössbauer furnace, with the source also kept at 300 K. For $T > 525$ K the changing shape of the Mössbauer spectra indicates a modified magnetic structure, suggesting a structural relaxation process and/or an Al atomic diffusion process. The kinetics of the crystallization process of the Al(x \, \mu m)/am/Al(x \, \mu m) trilayers will be published elsewhere.

The center of gravity shift (CS=chemical shift + second order Doppler shift (SODS)+pressure/stress induced shift) values of the Mössbauer spectra are given relative to the $^{57}$Co:Rh source. The spectra were analyzed using a single component magnetic hyperfine field distribution, assuming a linear relation between CS and $B_{hf}$ to account for small spectra asymmetries. For the fitting procedure, the quadrupole shift (QUA) contribution arising from the perturbation treatment of the combined magnetic-electric interaction and the relative intensity of the second to the third absorption lines (I$_{23}$) were used as fitting parameters, while the (I$_{13}$) ratio was kept fixed at the standard value of 3:1. The QUA values, obtained for the spectra recorded at different temperatures from uncoated and coated samples, are small ($\sim 0.02$ mm/s) and do not allow us to measure the influence of the Al cover layer on the charge symmetry within the studied temperature range.

III. RESULTS

A. Main results

For the trilayers used in this work, important tensile or compressive stress appears on individual layers when temperature ($T$) is changed due to the layers’ different thermal expansion coefficients ($\alpha$). The stress value also depends on their Young’s moduli ($E$) and the cover layer thickness ($x$). This stress induces changes in the magnetic moment distribution, as experimentally indicated in Fig. 1. Based on these considerations, one would expect no thermoelastic effects when the trilayer system is set around the temperature at which the coating layers were deposited. Thus, by taking into account the thermal strains in each layer and applying Newton’s laws, for an Al/am/Al trilayer system, we have

$$\sigma_{am} = \varepsilon_{eff}E_{am} = \frac{\varepsilon_{eff}}{d_{am}}E_{am} \left( \frac{2\varepsilon_{Al}}{d_{am}E_{am}} - \varepsilon_{am} \right) \Delta T$$

where $\varepsilon_{am}$ and $x$ are the amorphous and Al layer thicknesses, respectively, $K_{eff}$ is the effective thermal expansion coefficient, and $\Delta T$ is the temperature variation with respect to the deposition temperature ($\Delta T=T-T_{deposition}$). Using the $\alpha$, $E$, $d_{am}$, and $E$ parameters (Sec. II) of the Al and am layers in Eq. (1), we have plotted in Fig. 2 the effective deformation $\varepsilon_{eff}$ and the stress ($\sigma_{am}$) for two $x$ values. This figure indicates that the magnitude of the stress on the am depends on the cover layer thickness; i.e., increasing $x$ results in an enhancement of the $\sigma_{am}$ stress. Specifically, for $x=20 \, \mu m$ and $\Delta T=420$ K, the estimated stress on the am ribbon reaches about 0.45 GPa. Conversely, it should be mentioned that a hydrostatic pressure of about this size, when applied to magnetic Fe-based materials, can produce considerable changes in spin orientation and charge densities around the $^{57}$Fe nucleus.10

Experimentally, the general features of the temperature dependence of the Al(x)/am/Al(x) trilayers’ Mössbauer

![FIG. 1. Temperature dependence of the Al(x)/am/Al(x) Mössbauer spectra for $x=20 \, \mu m$. The magnetic hyperfine field distribution curves, obtained from the fittings of the Mössbauer spectra, are shown on the right side of this figure.](image)

![FIG. 2. Temperature dependence of the effective deformation ($\varepsilon$) and the stress ($\sigma$), given by Eq. (1), for the Al(x)/am/Al(x) trilayers, with $x=2.5$ and 20. The deposition temperature is assumed to be 0 K.](image)
spectra can be observed in Fig. 1, on the example of the trilayer with $x=20 \, \mu m$. The spectra have six broad, almost symmetric, absorption resonance lines associated with the magnetic hyperfine splitting of the nuclear spin levels in a local field ($B_{hf}$). The line broadening is related to the distribution of the Fe environments found in the amorphous state of the Metglas ribbons.\cite{11,12} Thus, the Mössbauer spectra were analyzed using a distribution of $B_{hf}$, where the fitting results are shown on the right side of Fig. 1.

The temperature dependence of the average magnetic hyperfine field ($\langle B_{hf} \rangle$), the average center of gravity shift ($\langle CS \rangle$), and the 2 to 3 line intensity ratio ($I_{23}$), obtained from the fittings of the spectra, are shown in Figs. 3-5, respectively, for the Al($x$)/am/Al($x$) trilayers with the Al thickness ($x$) indicated in the figures. The $\langle B_{hf} \rangle$ values for the $x=5 \, \mu m$ trilayer (not shown) are similar (within the error bars) to those for the uncoated sample, while the $\langle CS \rangle$ are slightly different for both samples. Furthermore, the $\langle B_{hf}(T) \rangle$ behaviors (Fig. 3) for our trilayers are similar to those found in conventional magnetic materials. However, the results for larger $x$ indicate a decrease in the $\langle B_{hf} \rangle$ values in the low temperature region ($T<260 \, K$), while for $T>280 \, K$ the $\langle B_{hf} \rangle$ values are slightly larger for the coated ribbon sample. At about 425 K (near the deposition temperature of this sample), the $B_{hf}$ for the uncoated and coated samples reach similar values.

For the studied trilayers, the largest effect of the coating on the (CS) values has been obtained from the Al($x=20 \, \mu m$)/am/Al($x=20 \, \mu m$) trilayer. Actually, the temperature dependence of (CS) is nonlinear in the whole temperature interval for the coated ribbon samples ($x \geq 5 \, \mu m$), while for uncoated ribbons it decreases monotonically with temperature (Fig. 4), as expected from the SODS effect contribution at high temperatures.\cite{13}

Figure 5 displays the $I_{23}$ as a function of the temperature for all studied trilayers. The corresponding $\theta$ angles between the average sample magnetization and the gamma ray direction [defined by Eq. (3)] are indicated in the right hand vertical axis. For the uncoated sample, $I_{23}$ is roughly temperature independent, with the average magnetization forming an angle of about 57$^\circ$ relative to the gamma ray direction (perpendicular to the ribbon plane). On the other hand, it can be observed in this figure that for increasing $x$ the spins tend toward the out-of-ribbon plane direction when the temperature is lowered. At 12 K, the $\theta$ values depend strongly on $x$, being 45$^\circ$, 27$^\circ$, and 23$^\circ$, for 2.5; 5; and 20 $\mu m$, respectively. The $\theta$ values of the trilayers with $x \geq 5 \, \mu m$ saturate at temperatures below 260 K; i.e., the Al layer effect is more pronounced in the trilayers with large $x$ values. The fraction of spins that do not completely become perpendicular to the ribbon plane at 12 K can be due to local anisotropy distributions associated with the melt-spinning process,\cite{14} as illustrated in Fig. 6 by polygon pictures inside the amorphous ribbon.
B. Discussion

1. Influence of the Al covering layer on the s-electron spin and charge densities

The average center shift ((CS)) of the Mössbauer spectrum has two contributions associated with the difference of s-electron densities (ρ(0)) of the source and the absorber and with the SODS effect. The first contribution is practically temperature independent, while the second one is strongly temperature dependent. The pressure effect on the sample will change the nucleus-electron interaction magnitude, which is correlated to the resonant photon energy (f) by

\[
\left( \frac{\partial f}{\partial P} \right)_T = K \left( \frac{\partial V}{\partial P} \right)_T + \left( \frac{\partial \rho}{\partial P} \right)_T + \left( \frac{\partial \rho_{ff}}{\partial P} \right)_T.
\]

The first term of Eq. (2) corresponds to the pressure effect on the s-electron density, while the second term is the pressure effect on the lattice vibration state, which is associated, according to the Debye model, with the solid Debye temperature. The second term is, in general, much smaller than the first one and can be usually discarded. Therefore, stress on the absorber may create distortion in the 4s and 3d bands, resulting in a modification in s-electron populations at the \(^{57}\)Fe nucleus, consequently modifying the (CS) values. The pressure (stress) effect in our samples is observed by comparing the (CS) values of uncoated (pure ribbon) and Al coated samples. The uncoated sample, which remains at constant pressure, has (CS) values decreasing monotonically with temperature, which is basically attributed to the SODS effect. On the other hand, the x = 20 \(\mu\)m trilayer has a parabolic-like (CS) thermal behavior, indicating a strong modification in s-electron density caused by the Al cover layers. The (CS) behavior, observed in the x = 20 \(\mu\)m trilayer, indicates that a positive contribution is added to the SODS temperature dependence for \(T < 260\) K, while a negative contribution is added for temperatures > 260 K. Particularly, the (CS) values for the uncoated and for the x = 20 \(\mu\)m trilayer at about 80 and 420 K are equal. At about 420 K, which is close to the deposition temperature range, this behavior would be expected (only SODS effect). However, the result obtained at about 80 K, suggesting that \((CS)_{\text{ribbon}}\) (chemical shift + SODS) \((CS)_{\text{trilayer}}\) (chemical shift + SODS + stress), is not yet clearly understood.

To describe the nonmonotonical \((CS)\) thermal behavior clearly observed for the x = 20 \(\mu\)m trilayer, the following mechanism is proposed. First of all, one has to use the fact that the (CS) values of \(^{57}\)Fe in transition metals decrease practically linearly under hydrostatic pressure (up to 30 GPa). Additionally, we have to take into account the \(I_{23}\) thermal behavior and the fact that the Poisson coefficient for the amorphous ribbon is around 1/3; consequently the longitudinal compression on reducing temperature produces an additional volume reduction. Therefore, the abrupt changes of the \(I_{23}(T)\) behavior for the x = 5 \(\mu\)m trilayers, at the interval 260 K < \(T < 425\) K, can be related to a large nonlinear volume modification in this temperature range. Thus, the above mentioned behaviors may strongly indicate that the biaxial stress field induced by temperature variation on the am layer leads to a nonuniform volume variation. Consequently, for \(T > 260\) K (more negative (CS) values, see Fig. 5), one has to assume that the biaxial stress modifies the 3d orbital to the plane of the ribbon (stress plane) concomitantly with drastic changes in s-electron densities (enhancement of (CS) values). Oppositely for \(T < 260\) K the am volume variation maybe under linear compression stress, and this stress would mainly act on the 3d orbital electronic population in such way as to reduce the 4s-electron densities at the \(^{57}\)Fe nucleus, as suggested by the (CS) behavior within this temperature range. In other words, the \(^{57}\)Fe “cell volume” change is nonuniform under the biaxial stress induced by temperature variation in the Al/am/Al trilayers, resulting in the nonlinear (CS) thermal behavior. This unexpected behavior, caused by the Al biaxial stress, is also reflected in the \(B_{\text{h//}}\) behavior, as shown in Fig. 3.

In summary, the stress induced by temperature variation of the Al/am/Al trilayers causes modification in the s-electron spin and charge densities at the \(^{57}\)Fe nucleus of the Metglas 2605S2 amorphous alloy, as suggested by the results in Figs. 3 and 4, respectively. However, calculations of the electronic structure have to be performed to better understand the observed (CS) and \(B_{\text{h//}}\) behaviors of the Al/am/Al trilayers.

2. Influence of Al thickness on the spin reorientation of Metglas 2605S2 alloy

This section aims to describe qualitatively the spin reorientation phenomenon of the Al(x \(\mu\)m)/am/Al(x \(\mu\)m) trilayers. From Fig. 5, it can be said that the studied Al/am/Al trilayers, with nonvanishing x, display similar behaviors for the \(I_{23}(T)\) quantity; i.e., its value decreases as the temperature is reduced, but this effect is enhanced for thicker Al layers, as discussed above. Therefore, using Eq. (3), one may conclude that the average sample magnetization prefers to be oriented at the out-of-ribbon plane when the temperature is reduced, which is associated with the coupling of the Metglas 2605S2 positive magnetostriction with the compressive stress caused by the Al coating layers.

\[
I_{23}(\theta) = \frac{4 \sin^2(\theta)}{1 + \cos^2(\theta)}
\]
As mentioned above, the largest \( x \) provokes the highest sensitivity of spin reorientation relative to temperature variations within a quite narrow interval around 400 K. The variation in the average magnetization vector (\( dM \)) can be related to the magnetic layer deformation and the material magnetostriction (magneto-mechanical coupling). Thermal deformation of the well mechanically coupled Al and am layers causes stress in the magnetic layer because of the different thermal expansion coefficients. The effect in a specific direction (\( x \)-axis) can be written as

\[
\frac{d\mathbf{M}}{dT} = \frac{\partial \mathbf{M}}{\partial \mathbf{e}} \mathbf{e}_x \frac{dT}{d\tau},
\]

\[
\frac{d\mathbf{M}}{dT} = \frac{\partial \mathbf{M}_x}{\partial \mathbf{e}_x} \frac{dT}{d\tau} + \frac{\partial \mathbf{M}_y}{\partial \mathbf{e}_x} \frac{dT}{d\tau} + \frac{\partial \mathbf{M}_z}{\partial \mathbf{e}_x} \frac{dT}{d\tau},
\]

where \( (\partial \mathbf{e}_x/\partial T)dT = d\mathbf{e}_x \), making

\[
\Gamma = \frac{\partial \mathbf{M}_x}{\partial \mathbf{e}_x} \frac{dT}{d\tau} = \Gamma_x \mathbf{e}_x.
\]

One can write

\[
\frac{d\mathbf{M}}{dT} = \Gamma_x \mathbf{e}_x \frac{dT}{d\tau} = \Gamma_x d\mathbf{e}_x.
\]

\( \Gamma_x \) can be defined as the magneto-mechanonical coefficient vector, where for positive magnetostriction, one has \( \Gamma_{xx} > 0 \), \( \Gamma_{xy} < 0 \), and \( \Gamma_{xz} < 0 \); i.e., traction stress in the \( x \)-direction will promote an increase in the \( x \)-magnetization, while the \( y \) and \( z \) contributions will be reduced, as illustrated in Fig. 6. Therefore, \( \Gamma_x \) is a tensor element associating the three components of the magnetization vector with deformation in the \( x \)-direction (\( de_x \)). Considering tridimensional deformations, the relation between the magnetization direction and deformation can be rewritten as

\[
\Gamma = \begin{bmatrix} \Gamma_{xx} & \Gamma_{xy} & \Gamma_{xz} \\ \Gamma_{yx} & \Gamma_{yy} & \Gamma_{yz} \\ \Gamma_{zx} & \Gamma_{zy} & \Gamma_{zz} \end{bmatrix}.
\]

For isotropic and positive magnetostriction materials like amorphous Metglas, the \( \Gamma \) tensor is symmetric, \( \Gamma_{ij} = \Gamma_{ji} = \Gamma(= -|\Gamma|) \) for \( i = j \) and \( \Gamma_{xx} > 0 \). Thus, the tensor can be rewritten as

\[
\Gamma = \begin{bmatrix} \Gamma_{xx} & -|\Gamma| & -|\Gamma| \\ -|\Gamma| & \Gamma_{xx} & -|\Gamma| \\ -|\Gamma| & -|\Gamma| & \Gamma_{xx} \end{bmatrix}.
\]

For an interfacial compression stress induced by a temperature variation (\( dT \)), one has \( de_{xx} < 0 \), \( de_{yy} < 0 \), and \( de_{zz} > 0 \). In the case of \( de_{xx} = de_{zy} = (de < 0) \) and \( de_{xz} = -\nu d\epsilon \), where \( \nu = 1/3 \) (Poisson coefficient) for the Metglas ribbon, the magnetic reorientation due to the \( \Gamma_x \) tensor under the deformations in the three spatial components can be rewritten as follows:

\[
\begin{bmatrix} dM_x \\ dM_y \\ dM_z \end{bmatrix} = \begin{bmatrix} \Gamma_{xx} - |\Gamma| & -|\Gamma| \\ -|\Gamma| & \Gamma_{xx} \\ -|\Gamma| & -|\Gamma| \end{bmatrix} \begin{bmatrix} d\epsilon \\ d\epsilon \\ d\epsilon \end{bmatrix}.
\]

It should be observed that the \( z \)-magnetization component is favored by the interfacial compressive stress applied in isotropic materials with positive magnetostriction. In other words, it promotes the magnetization reorientation perpendicular to the direction of the deformation. Thus, the amorphous-aluminum interfacial mechanical coupling results in the compressive stress in the planar directions, producing magnetization orientation nearly perpendicular to the ribbon plane, when volume deformation is produced, as experimentally shown in Fig. 5. Additionally, the thermal rate of magnetization reorientation in the deformation direction for a positive magnetostrictive material is proportional to the deformation rate of the magnetic layer; in other words, \( \partial \mathbf{e}_x/\partial T = K_{eff} \). Therefore, the thermal rate of magnetic reorientation increases with \( x \), as experimentally shown in Fig. 5 for different \( x \) values. Unfortunately, our Mössbauer data do not allow us to have numeric values for the \( \Gamma_{xx} \) and \( \Gamma_y \) parameters, but this formalism helps quite well to describe qualitatively the observed spin reorientation phenomenon of our samples.

IV. CONCLUSION

Al(\( x \) \( \mu \)m)/Metglas 2605S2/Al (\( x \) \( \mu \)m) trilayers (\( x = 0 \); 2.5; 5 and 20), prepared by dc magnetron sputtering, have been systematically studied by MS at various temperatures. The influence of Al capping layers on the hyperfine properties of the Metglas 2605S2 is clearly demonstrated. Spin polarization and charge densities are strongly affected by thick Al layers (\( x = 20 \) \( \mu \)m). The ribbon magnetization average direction of a trilayer can be partially adjusted alternatively by varying its temperature or its Al capping layer thickness. Additionally, the angle between the average sample magnetization and the gamma ray direction saturates at lower values for temperatures below 260 K for the trilayers with \( x \geq 5 \) \( \mu \)m, suggesting a saturation of the spin reorientation process. A phenomenological model has been proposed to describe qualitatively the spin reorientation effect and its sensitivity in the trilayer system.

ACKNOWLEDGMENTS

The authors would like to thank FINEP, FAPES, CNPq/MCT, and UFES for the financial support.

7See www.metglas.com/sitemap for Metglas physical properties.