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Conversion-electron Mössbauer spectroscopy studies in amorphous Tb-Fe films

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We have carried out conversion-electron Mössbauer (CEM) spectroscopy studies in rf-sputtered amorphous Tb_xFe_{100-x} films with $x=20$, 24, and 25. The samples present a well-defined uniaxial anisotropy K_u on the order of 1×10^6 erg cm^{-3}, and the magnetization is well aligned along the film normal as shown by rectangular hysteresis loops (both magnetization and polar Kerr). However, the analysis of the CEM spectra shows that the Fe moments deviate strongly from the film normal. The semiangle of the cone is calculated to be 35° for $x=20$ and 25° for $x=24$ and 25. Exchange coupling between Fe and Tb moments and the well-known tendency in some amorphous alloys to form antiferromagnetic coupling between Fe moments are offered as possible explanations for this result. Finally, the average hyperfine field for Fe is found to be 165 kOe with the high-field peak centered at around 190 kOe.

INTRODUCTION

Amorphous films of rare-earth–transition-metal alloys such as Tb-Fe, Gd-Tb-Fe, etc., are being used more and more as magnetooptical storage media and have been the object of intense study for over a decade.[1] Rare-earth metals with strong spin-orbit coupling ($L=0$) give rise to large local anisotropy. Harris, Plischke, and Zuckermann[2] have explained the anisotropy in such amorphous alloys by the random-anisotropy model, namely the local variation in the orientation of the symmetry axis at the rare-earth site. Neutron-diffraction studies have revealed the presence of anisotropy in Tb-Fe alloys.[3] However, thin films of such alloys in the thickness range 50–300 nm present an interesting characteristic, namely, a well-defined uniaxial anisotropy (K_u) making the normal to the film plane the easy axis of magnetization, which incidentally is a prerequisite for magnetooptical recording applications.[4,5] We have recently reported on our low-temperature measurements of K_u in rf-sputtered amorphous (a-) Tb-Fe films.[5] Such films with strong K_u on the order of 10^6 erg cm^{-3} at 290 K present a rectangular M-H and polar-Kerr (θ_K)-H loops. The magnetic saturation is obtained for fields on the order of 5 kOe and also the remanence is almost the same as that at saturation. Nevertheless, it is known that Tb moments are not collinear but show a spread in their direction whereby the net Tb moment measured is smaller than that predicted by theory.[3] It is, therefore, interesting to learn about the spin structure of Fe. Chappert, Coey, Liénard, and Rebouillat[6] have shown by Mössbauer studies that in a-Y-Fe, the Fe spins are noncollinear. But such studies have been carried out on fairly thick specimens (several microns thick) where one no longer has either a well-defined uniaxial anisotropy or a rectangular hysteresis loop. So it is necessary that Fe-spin structure be studied in Tb-Fe thin films with characteristics as defined earlier. Conversion-electron Mössbauer spectroscopy (CEMS) is ideally suited for such studies. By comparing the intensity of the various lines in the spectrum it is possible to calculate the average angle θ the Fe spins make with the film normal and hence the direction of the propagation of γ rays (Fig. 1). Let us recall that once magnetized, the Tb-Fe films retain their magnetized state and so the CEMS experiment is carried out in the absence of any domains, and is therefore equivalent to applying a saturating field. We studied both magnetized and nonmagnetized samples and describe our results here. To our knowledge this is the first report of its kind.

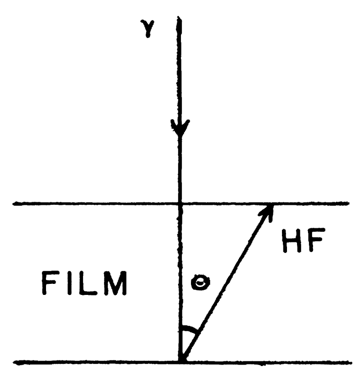

FIG. 1. Schematic diagram of the configuration of different parameters.

EXPERIMENTAL DETAILS

Amorphous Tb_xFe_{100-x} with $x=20$, 24, and 25 (expressed in at.%) were deposited on water-cooled glass substrates by rf sputtering. The details have been published earlier.[5] A rf power of 100 W and an argon pressure of 8 mTorr were used. The magnetic film thickness was around 150 nm. The samples were given a protective coating of 10-nm-thick Al before breaking the vacuum, which is found to be adequate to protect against oxidation (no deterioration of the properties was observed over a period of about one year on samples stored in a descicator) but at the same time permeable for electron capture for CEMS studies. Sample composition was determined by inductively coupled plasma analysis. Both *M-H* and θ_K-*H* loops were also studied.

For CEMS studies, the 7.7-keV conversion and 5.6-keV Auger electron spectra were taken using a source of 50 mCi ^{57}Co in rhodium at room temperature. The proportional counter consisted of a cell through which a mixture of He and 5% CH_4 was allowed to flow.[7] The spectrometer was a conventional one. The Fourier analysis of the spectra was also performed using a method derived from that first proposed by Window.[8]

RESULTS AND DISCUSSIONS

The *M-H* and θ_K-*H* loops are rectangular with similar magnetic characteristics. A typical θ_K-*H* loop for the sample with $x=25$, taken from the substrate side, is shown (laser wavelength $\lambda=632$ nm) in Fig. 2. It is recalled that for $x=20$, the Fe moments dominate whereas for $x=24$ and 25 those of Tb which dominate. This leads to an inversion in the Kerr loop as the magnetooptical effect arises mainly from Fe atoms. The uniaxial anisotropy of the samples is around 1.5×10^6 erg cm^{-3}.

The CEM spectra of the three compositions studied have nearly the same shape. Also both magnetized and unmagnetized samples give similar results. Figure 3(a) shows the spectrum for $x=20$. The main feature is that the signal intensity for $m=0$ transitions is not null. This indicates that the hyperfine field is not rigorously perpendicular to the film plane but forms a cone with a semi-angle θ. An estimation of the average angle between the HF and the film normal (Fig. 1) will be given in what follows.

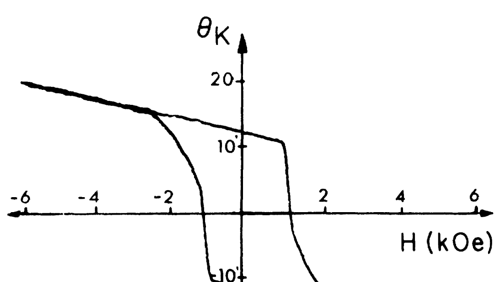

FIG. 2. θ_K-*H* loop for the sample with $x=25$.

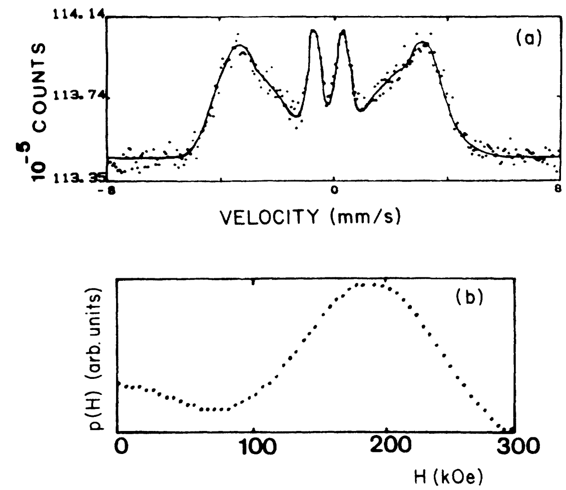

FIG. 3. (a) CEM spectrum for the sample with $x=20$. (b) The HF distribution in sample with $x=20$.

All the spectra were fitted to a field distribution by summing 60 elementary sextuplets with 0.16-mm s^{-1}half linewidth for fields ranging from 0 to 300 kOe. Isomer shift and quadrupole effect were taken as -0.15 and 0-mm s^{-1}, respectively. The HF distribution obtained for $x=20$ is shown in Fig. 3(b). A small fraction of Fe atoms appear to be in weak field. The high-field peak is centered around 190 kOe. The average field over the whole distribution $\bar{H}$ is 165 kOe with a standard deviation (σ_H) of 66 kOe. The intensities of the lines are in the ratio 3:0.8:1:1:0.8:3. The relative intensity [Fig. 3(b)] of $\Delta m=0$ is 0.8 and the value of the angle θ can be calculated as 35°. For $x=24$, using the same fitting procedure, a small decrease of about 5% was found for $\bar{H}$. However, for both $x=24$ and 25, θ was found to be only 25°.

So it is seen that the HF is making an angle in the range 25°–35° with the film normal whereas the net magnetization is well aligned in this direction. Let us recall the following points. The CEM spectra result not only from the spins in the film surface but also from those in the whole film volume. It is of course true that the electron-capture probability is not the same for all the electrons and would depend on the depth from which they are coming. So the contribution to the spectrum from Fe spins at different depths do not have the same weight. It cannot, therefore, be argued that the present result arises from the surface spins which may not be aligned properly. Furthermore, the polar-Kerr effect, which, on the contrary, concerns only a thickness of 10 to 20 nm near the surface, shows that the direction of magnetization does not deviate from the film normal (Fig. 2). The magnetization measurements that give an average effect show that the *M-H* loop is rectangular which again indicates that there is a good alignment of the bulk magnetization along the film normal. So the present results, namely the large value of 25°–35° for θ, could be understood as follows: *M-H* and θ_K-*H* loops give the situation of the overall magnetization. However, there are local fluctuations in the direction of Fe moments which could arise from the following factors: (1) the exchange coupling of Fe to Tb moments which are known to show a large spread. It would appear that Fe-Fe exchange which should tend to align Fe spins parallel to each other is relatively weaker than Fe-Tb exchange. (2) Furthermore, in certain amorphous alloys Fe is known to give rise to competing ferromagnetic and antiferromagnetic interactions. In fact, in Fe-rich amorphous alloys such as Fe-Zr, the local order around Fe atoms is found to have fcc structure, giving rise to antiferromagnetic interactions.[9] We have shown that in Fe-Zr alloys, when Ni is substituted for Fe, an increase in the Curie temperature is observed due to the fact that the local order is modified from fcc structure and more of ferromagnetic coupling ($J > 0$) is favored.[10] So in Tb-Fe also there could be such effects which introduce the spanning of Fe moments. It is interesting to note that for $x = 25$, namely Tb richer film, the deviation of Fe moments is much smaller ($\theta = 25°$) which lends support to the above hypothesis. Of course to get a full alignment of Fe spins, much higher fields would be necessary. Hence, the apparent magnetic saturation obtained for a field of 5 kOe is not very much modified even in field on the order of 12 kOe. Also, in amorphous Y-Fe alloys, the Fe moments do show a very large spread as explained in Ref. 6. However, this material cannot be compared directly to Tb-Fe films discussed here because in Y-Fe there is no uniaxial anisotropy present and it does not show any magnetic saturation in moderate fields or a high remanence.

In conclusion, CEMS study of amorphous Tb-Fe films has shown that the Fe moments are not collinear and deviate strongly from the film normal forming a cone about it. This result is rather unexpected just looking at the rectangular hysteresis loops and the uniaxial anisotropy evidenced from torque curves. It is also noteworthy that for Tb-rich samples the average value of the semiangle of the cone is decreased. It would be interesting to study Fe spin structures in a system where the rare-earth metal has no spin-orbit coupling ($L = 0$) such as Gd, in order to reduce the effect of random anisotropy on the alignment of Fe spins.

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