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CEMS STUDY OF $\text{Fe}_{60}\text{Al}_{40}$ THIN FILMS

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Abstract. – The coevaporation technique offers the possibility to prepare $\text{Fe}_{60}\text{Al}_{40}$ thin films which behave ferromagnetically at 293 K. They show nearly same magnetic properties as cold worked or ion-implanted bulk $\text{Fe}_{60}\text{Al}_{40}$. On the contrary, their behaviour strongly differs at higher temperatures so that the precipitation of iron rich phases is inferred from CEMS analysis.

In a recent paper [1], it has been shown that magnetic $\text{Fe}_{60}\text{Al}_{40}$ films could be prepared by coevaporation. This result is important because bulk $\text{Fe}_{60}\text{Al}_{40}$ having the L_{20} superstructure is paramagnetic at 293 K and previously it could be rendered ferromagnetic at this temperature only under certain conditions, namely after cold working [2] or ion-implantation [3]. These treatments were assumed to create atomic disorder so that a given Fe atom had a good probability to be surrounded by at least three or four Fe atoms which is necessary for him to bear magnetic moments. We describe here further results on the magnetic properties of coevaporated $\text{Fe}_{60}\text{Al}_{40}$ thin films (200 nm) deposited on a Si substrate. Special emphasis will be placed on the thermal behaviour of the films which showed striking features. The magnetic properties were chiefly evaluated with the Mössbauer effect working in scattering geometry (CEMS); additional informations were obtained with a polar Kerr effect measurement systems.

The films were deposited on a Si substrate by coevaporation. Two E-beams installed in a high vacuum chamber were used; the starting pressure was 1×10^{-8} Torr and during deposition it was maintained at about $2 - 3 \times 10^{-8}$ Torr; the deposition rate of each element, typically 0.2-0.3 nm/s, and the thickness of the layer were monitored by two quartz oscillators. Concerning the CEMS technique, the 7.3 keV conversion and 5.6 keV Auger electron spectra were taken using a source of 50 mCi ^{57}Co in rhodium, a proportional counter with a He-5 % CH_4 gas flow and a conventional Mössbauer spectrometer; the counter has been recently described in details in reference [4]. The analysis of the spectra was performed by summing a central Lorentzian line and a distribution of hyperfine fields (Window method [5]). The magnetization was determined using a polar Kerr effect measurement device, the laser of which had a wavelength of 632.8 nm.

The 293 K electron spectrum of an as-coevaporated thin film is presented in figure 1a. The average hyperfine field $\bar{H}$ is found to be 218 kOe while the percentage of magnetic iron is 74 % (see Tab. I). These

Fig. 1. – Room temperature CEM spectra: (a) as-coevaporated, (b) annealing temperature = 573 K, (c) 773 K, (d) 873 K.

results are in good agreement with those determined on both implanted [3] and crushed [2] bulk samples. Figure 2 shows the variation in Kerr rotation angle θ_K as a function of applied magnetic field. Saturation is reached for $\theta_K = 13^\circ$ and $4\pi M = 6.4$ kOe; assuming a density of 6.02 g.cm^{-3} , the magnetic moment is found to be $\sigma = 84.5 \text{ emu/g}$, a result which is also in agreement with the value determined on implanted samples by ferromagnetic resonance [6]. From the above data it is quite satisfactory to note that the three kinds of sample preparation namely cold working, ion-implantation and coevaporation, lead to nearly same magnetic properties.

Fig. 2. – Kerr rotation angle versus magnetic field at 293 K.

We report now the thermal behaviour of the films as determined by CEMS. The sample under study was annealed for one hour under a 10^{-6} Torr vacuum at the required temperature in the range 293-873 K and then the CEM spectrum was taken at 293 K. The spectra are presented in figures 1b, c, d while the corresponding Mössbauer parameters are listed in table I.

Table I. - Mössbauer parameters. Left and right sides correspond to field distribution and paramagnetic central line respectively: $\bar{H}$ = average hyperfine field, σ_H = standard deviation, b = relative intensity of lines 2 and 5, f_f = ferromagnetic fraction, IS = isomer shift with respect to α -iron, Γ = half line width. The large values of Γ determined for spectra 1a, b indicate the paramagnetic fraction is also distributed.

T (K)	$\bar{H}$ (kOe)	σ_H (kOe)	b	f_f (%)	IS (mm/s)	Γ (mm/s)
293	218	51	1.1	74	0.15	0.5
after 1 h at 573	290	18	2	46	0.10	0.5
after 1 h at 773	301	15	0.45	100	—	—
after 1 h at 873	—	—	—	—	0.15	0.26

First, we have to point out that the spectra unambiguously show a magnetic fraction (f_f) up to at least 773 K; at this temperature $f_f = 100\%$ so that no paramagnetic iron is remaining. A paramagnetic spectrum is only obtained after annealing at 873 K; the most probable reason for this change is the diffusion of Si from the substrate into the film. A strong difference then appears with respect to both cold worked and ion-implanted bulk $Fe_{60}Al_{40}$ for which the magnetic interactions disappeared at temperatures slightly higher than 293 K [7]; in this case, $L2_0$ atomic reordering was assumed to take place, thus leading to paramagnetic spectra. On the contrary, the hyperfine field of the coevaporated films increases with increasing temperature up to 301 kOe, a value slightly higher than that of D sites in ordered Fe_3Al , i.e. 290 kOe. This variation in field indicates the presence of iron-rich domains, the relative abundance of which increases up to 100 % at 773 K. Atomic ordering of $L2_0$ type, if any exists,

may contribute to the paramagnetic fraction but from the above result it is clear that the basic phenomenon is rather new phase precipitation than $L2_0$ ordering. This behaviour is first surprising; however, it is to be recalled that the coevaporation technique leads to metastable random alloys exhibiting local fluctuations of composition because the evaporation rate from each source cannot be maintained rigorously constant. In our case, the Mössbauer spectra suggest that at 773 K, significant atomic motion becomes possible enabling the transformation of metastable $Fe_{60}Al_{40}$ into one or several phase(s) of higher stability close to Fe_3Al and α -Fe. Finally it is noteworthy that Godbole *et al.* [8] have already observed same trend in ion-beam-mixed $Fe_{55}Al_{45}$ samples.

As a conclusion we have prepared thin films of $Fe_{60}Al_{40}$ by coevaporation, the magnetic properties of which at room temperature appear to be nearly the same as those of bulk material chemically disordered either by cold working or by ion-implantation. However, at temperatures higher than 293 K, i.e. in the 573-773 K range, the films show a tendency towards new iron rich phase precipitation instead of $L2_0$ ordering as expected from the bulk behaviour.

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