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MÖSSBAUER SPECTRA OF ULTRATHIN Fe FILMS COATED BY MgO

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Abstract.- Mössbauer spectra of thin Fe films (24, 16 and 8 Å) sandwiched with MgO layers showed that the hyperfine field at 4.2 K increased with the decrease of the film thickness. From the computer fitting results with the least square method the hyperfine field at the surface layers of the MgO-coated Fe film is nearly 380 kOe. The spectrum at 4.2 K of the 8 Å film sandwiched with MgO shows the intensity ratio of 3:1:1:1:1:3, which means that the magnetization of the 8 Å Fe film was preferentially perpendicular to the surface due to the surface anisotropy.

This article reports the Mössbauer results on ultrathin Fe films coated by MgO. In an oil-free UHV system, MgO and Fe (50 % enriched in $^{57}$Fe) were heated by an electron beam gun and alternately evaporated onto a mylar substrate at room temperature. The base vacuum was $5 \times 10^{-9}$ torr and the vacuum during deposition was better than $1 \times 10^{-7}$ torr. The depositions of Fe and MgO were repeated for several times to gain enough thickness of $^{57}$Fe for the Mössbauer measurements. The thickness of MgO layers in between the Fe layers was about 400 Å and the first and the final MgO layers were as thick as 500 Å to protect the sample. The average thicknesses of Fe layers in the prepared samples were 24, 16 and 8 Å.

Mössbauer spectra at 300 K and 4.2 K are shown in figure 1 and figure 2, respectively. The magnetic hyperfine fields of the 24 Å film at both temperatures are nearly the same as the bulk values and the intensity ratios are approximately 3:4:1:1:4:3, which means that the magnetization lies in the film plane due to the shape anisotropy. At 300 K, the 16 Å film is partially, and the 8 Å film is entirely superparamagnetic. The spectra at 4.2 K clearly show that the hyperfine field increases with the decrease of the film thickness. This result suggests the magnetic moment of Fe has increased at the interface with MgO. The same tendency was already observed in some cases; Mg$_2$-covered Fe films /1/, Ag-coated Fe films /2/ and Fe fine particles in SiO$_2$ matrix /3/. Electrodeposited surfaces /4/, Cu-coated surfaces /5/ and Sb-coated Fe films /6/ however, showed the opposite tendency; namely the surface hyperfine field is smaller than the inside. The surface magnetic anomaly depends on the characteristic properties of the coating material.

Fig. 1: Mössbauer absorption spectra at 300 K of MgO-coated Fe films.

The spectrum at 4.2 K of the 8 Å film looks unusual since the intensity ratio has differed greatly and also the sample has partially been oxidized. A computer fitting was made with assuming three 6-line sets of Lorentzian lines; one oxide and two metallic components. The result indicates the oxide was 30 % of the total Fe and the hyperfine field increases with the decrease of the film thickness.
field is about 480 kOe. The hyperfine fields of the metallic part were obtained as 355 kOe and 380 kOe. The former nearly corresponds to the bulk value and the latter possibly the surface fraction which is 12% larger than the bulk value.

The intensity ratio of the metallic part, the sum of the two metallic 6-lines, was 3:1:1:1:1:3, which is much closer to the ratio, 3:0:1:1:0:3 than the powder pattern, 3:2:1:1:2:3.

Usually thin ferromagnetic films have in-plane magnetization due to the shape anisotropy. In extremely thin films, however, the "surface anisotropy" plays an important role. An atom in the surface layer always has fewer neighbours and a lower symmetry than in the bulk. An additional anisotropy can exist at surface atoms since the anisotropic interactions which are canceled by symmetry in the interior need not vanish at the surface. The bulk anisotropies (shape anisotropy and crystalline anisotropy) per unit area of the sample are proportional to the thickness and therefore decrease with the decrease of the thickness but the surface anisotropy remains constant. When the film is not very thin, the effect of surface anisotropy is negligible, while in ultrathin films the surface anisotropy can be dominant. If the easy axis due to the surface anisotropy is along the surface normal, the whole magnetization may be oriented perpendicular to the surface. The spontaneous magnetization being surface normal is a clear evidence of such a surface anisotropy. The critical thickness in this case is between 8 and 16Å, which probably depends on the coating material, crystal plane and other parameters such as the flatness of the film. The first report on the perpendicular magnetization was the torque measurements by Gradmann and coworkers [7]. The magnetization of single crystal Fe-Ni film on Cu substrate was along surface normal when the thickness was much thinner than 10Å.

The perpendicular magnetization can be realized only when the film is flat and continuous, therefore the deposited Fe films on MgO substrate seem to be very uniform. An Auger spectroscopic study by Kanaji et al. showed that Fe films grow in an monolayer by monolayer fashion on a substrate of single crystal MgO [8]. An electron diffraction pattern for the present sample was powder's one and the relationship between the crystal planes and the surface anisotropy is not clear. For a quantitative discussion on the surface anisotropy, measurements with well defined crystal planes are required.

References

/6/ Hine, S., Shigematsu, T., Shinjo, T., and Takada, T., this conference.