I. 10. Mössbauer Spectroscopy of Fe/Ag Layered Structure

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Introduction

Recently considerable effort has been devoted to synthesize or to analyze artificially structured materials (ASM) in the field of materials science or solid state physics. Among them, artificial synthesized lattice has been one of the most intensively studied subjects. Here, in the present work layered structure of Ag and Fe are studied. Namely, Ag and Fe are stacked alternatively for several 10 layers and Mössbauer spectroscopy (MS) and Rutherford back scattering spectroscopy (RBS) have been performed. The purpose of MS is to examine the phase of Fe layers and also the magnetization direction as a function of the layer thickness and annealing temperature. The RBS is employed to determine the exact thickness of the layers and also to examine the structure change of layers upon annealing.

Experimental

Ag and Fe layered structure was synthesized by a conventional vacuum evaporation method. The layer thickness during the evaporation was monitored and controlled by a film thickness meter utilizing the resonant frequency change of a crystal oscillator. The vacuum was maintained at $3 \times 10^{-6}$ during the evaporation.

MS spectra were measured in a transmission mode using $^{57}$Co(Rh) source with stacking 8-16 specimens so as that the total Fe layer thickness becomes 1 $\mu$m. Among used substrates, Al foil (for cooking use) and natural mica have been found to contain a trace amount of Fe giving rise to a weak paramagnetic line near 0 mm/s in Mössbauer spectrum. Artificial mica (Mylar) or polyimid film (Kapton) was found not to contain Fe. RBS spectrum was measured by 2 or 4 MeV - He ions with the scattering angle of 170 deg.

Result and Discussion

Fig. 1 shows the Mössbauer spectra for Ag/Fe layer structured specimens. Here x, y and n in $(x\text{ Ag, } y\text{ Fe})_n$ indicated in Fig. 1 signify layer thickness of Ag, Fe in (nm) and the total number of the layers, respectively.
Experimental findings and corresponding physical significance may be summarized as follows:

1. All the spectra have sextet absorption lines showing that Fe is in an $\alpha$-Fe state (ferromagnetic bcc phase) down to 2 nm (20A).

2. The separation between line 1(-5 mm/s) and 6 (5 mm/s) is almost independent from the Fe layer thickness ($t_{Fe}$). This shows that the hyperfine field (Hf) is independent of the thickness down to 2 nm for Ag/Fe layered structure. This is in contrast to the result for Fe-Cu case where part of Fe was found to be $\gamma$-phase (paramagnetic fcc phase) when the $t_{Fe}$ is reduced below 10 nm (100A)\(^1\).

3. The relative intensity of line 1 and 2(-3 mm/s), namely, $I_2/I_1$ is about 1.3 when $t_{Fe}$ is below 50 nm. Since the relative intensity is given as,

$$I_1/I_2 = 4 \sin^2 (\theta) / 3(1 + \cos^2 (\theta)),$$

where $\theta$ is the angle between the magnetization direction and $\gamma$-ray direction, this shows the magnetization direction is in the plane of the layers. This in-plane magnetization is quite usual for thin film Fe specimen and has been known caused by macroscopic shape effect.

4. A weak paramagnetic line is always present for specimens $t_{Fe} > 25$ nm and it becomes larger for $t_{Fe} < 5$ nm. This line, however, is found to be caused by a trace amount of Fe in the substrate Al foil and has nothing to do with Fe in the layered structure. This is confirmed by the spectra of the specimens prepared on polycrystalline substrate where the paramagnetic line is completely missing.

Figure 2 shows a result of annealing in an UHV for one of specimens [Ag(3 nm)/Fe(2 nm)]\(_{160}\) in Fig. 1. The duration of annealing was 15 hrs. After the annealing above 700K, a paramagnetic line grows near 0 mm/s and only one paramagnetic line is present after 863K annealing without any ferromagnetic lines. The same annealing effect was found for all the specimens in Fig. 1. To study the cause of the paramagnetic line after the 863K annealing, RBS spectrum was measured for one of the specimens in Fig. 1[Ag(5 nm)/Fe(5 nm)]\(_{21}\) before and after the annealing.

The result is shown in Fig. 3 where the lower one is for before annealing and the upper one for after the annealing. The spectrum before the annealing, namely in the as prepared state, shows a discrete spectrum which reveals that a layer structure is indeed formed in the present method of specimen preparation. The spectrum after the annealing, however, reveals that this layered structure is completely washed out by the annealing; Ag layers have evaporated out of the specimens during the annealing at 863K and also the substrate Al diffuses to the specimen surface. Moreover, some part of Fe diffuses into Al substrate forming an Al-Fe alloy. Therefore it is quite likely that the paramagnetic line in Fig. 2 after the 863K annealing is caused by the Al-Fe alloy and has nothing to do with the
anomaly of Fe magnetic properties expected for layered structure. Indeed a similar paramagnetic line has been reported by Nasu et al for Al-Fe alloys 2).

Different from the Cu-Fe case, no anomaly has been found for the Fe layer in Fe-Ag layered structure for the thickness investigated in the present, namely $t_{Fe} > 2$ nm. This is probably due to the large difference in lattice parameter between fcc Ag and bcc Fe, which prevents a coherent growth of Fe film on Ag film. Further studies are now in progress for other systems as Fe-Pb, Fe-Cd or et cet.

References

Fig. 1. Mössbauer spectra of Fe-Ag layered structure in as-evaporated state. In \((xAg, yFe)n\), \(x\), \(y\) and \(n\) signify layer thicknesses in nano-meter and \(n\) signify the total (Fe + Ag) layer number.
Fig. 2. Mössbauer spectra after an isochronal (150K interval and for 15 hrs) annealing for the (3Ag, 2Fe)$_{160}$ specimen.
Fig. 3. RBS spectrum for (5Ag, 5Fe)\(_2\) specimen before (lower) and after (upper) annealing at 863K. A similar spectrum is obtained for the specimen in Fig. 2.