Asymmetric interfaces in Fe/Ag and Ag/Fe bilayers prepared by molecular beam evaporation

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Abstract

Single layers of Fe and Ag, as well as Fe/Ag (iron deposited first) and Ag/Fe bilayers were prepared by molecular beam evaporation onto Si. The samples were investigated with backscattering spectrometry (BS) and atomic force microscopy (AFM). BS spectra of Fe/Ag and Ag/Fe indicate a significant difference at the interface. In the case of Fe/Ag the Ag peak has a long tail at the interface, while for Ag/Fe the interface is abrupt. The tail in the Fe/Ag spectrum is too large to be caused by double or plural scattering. According to AFM, the effect of surface roughness is also negligible. In spite of the fact that Fe and Ag are completely immiscible in equilibrium, this tail, however, suggests that some Ag is located in the Fe layer. After annealing, both samples show mixing between the two layers; this is much larger again for Fe/Ag.
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1. Introduction

Fe/Ag multilayers have been intensively studied due to their giant magnetoresistive behaviour [1–3]. Their physical properties strongly depend on the interface quality [4,5].

Although, Fe and Ag are completely immiscible in equilibrium [6], asymmetric interface structures between Fe and Ag layers deposited by pulsed laser deposition have been reported by Gupta et al. [7]. It was assigned to ion implantation: the relatively high-energy (10–200 eV) Fe and Ag ions produced during the deposition process have different ranges. An alternative explanation was suggested by Etienne and Massies [8]. When depositing Ag on Fe or Fe on Ag by MBE (molecular beam evaporation), the different fundamental growth-modes (layer-by-layer growth or 3D island formation) lead to different interface roughness.

These assumptions have been confirmed by Mössbauer spectroscopy [9]. The initial growth of Fe on Ag has been found to be three-dimensional; only after depositing 4.7 monolayers the Fe film grows by a quasi-layer-by-layer mode. In contrast with this, the interface of Fe/Ag (Fe deposited first) was found to be very sharp.

In this paper, backscattering spectrometry (BS) was used to investigate the interface properties of the above system. To decrease the effect of "ion implantation", our samples were prepared by MBE, where the energy of the impinging atoms is about 0.1 eV and the corresponding range is about 0.1 nm. The surface roughness of the samples was determined by atomic force microscopy (AFM) and was taken into account in the BS simulations. According to our results, the difference of the Fe/Ag and Ag/Fe interfaces is too large to be assigned to the surface roughness of the sample alone.

The asymmetry at the interface could be enhanced by annealing the samples.

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2. Experimental

Fe and Ag single layers, as well as Fe/Ag (Fe deposited first) and Ag/Fe bilayers were prepared by MBE onto Si(1 1 1) substrates under ultrahigh vacuum (UHV) conditions. To remove the native oxide, each substrate was first etched chemically by HF then annealed in UHV at 600 °C for 30 min. The base pressure was 1.4×10^{-10} mbar and during the growth process it increased to 2×10^{-9} mbar. Both Fe and Ag were evaporated by e-guns. During the growth process both e-guns ran simultaneously; the beams were controlled by pneumatic shutters. The energy of deposited atoms was about 0.1–0.15 eV corresponding to their thermal energy. The evaporation rate was 0.04 nm/s and 0.05 nm/s for Fe and Ag, respectively. To improve the lateral homogeneity of the layers, the samples were rotated during evaporation.

The surface roughness of the samples was determined by AFM. 1 μm × 1 μm AFM images were recorded by a Nanoscope E instrument operating under ambient conditions with a scan rate of 1 Hz. The height distribution of each sample was determined by the surface-roughness analysis software of the instrument.

For better detection of light impurities (e.g. C and N) in the layers, the BS measurements were performed with 3560-keV He$^+$ ions obtained by a 5 MV Van de Graaff accelerator. The samples were measured at tilts of 7° and 60°. The detector was mounted at a scattering angle of 165°. For each sample the two BS spectra were evaluated by the RBX code [10] using the same layer structure. Assuming bulk densities of pure elements, the measured thicknesses were 55 and 73 nm for the Ag and Fe single layers and 78/44 and 61/105 nm for the Fe/Ag and Ag/Fe bilayers, respectively. The experimental error of the layer thickness is about 5%.

The annealing was carried out in Ar atmosphere (500 °C, 1 h).

3. Results

The BS spectra taken on the bilayer-samples are shown in Fig. 1. In the case of Fe/Ag the Ag peak shows a long
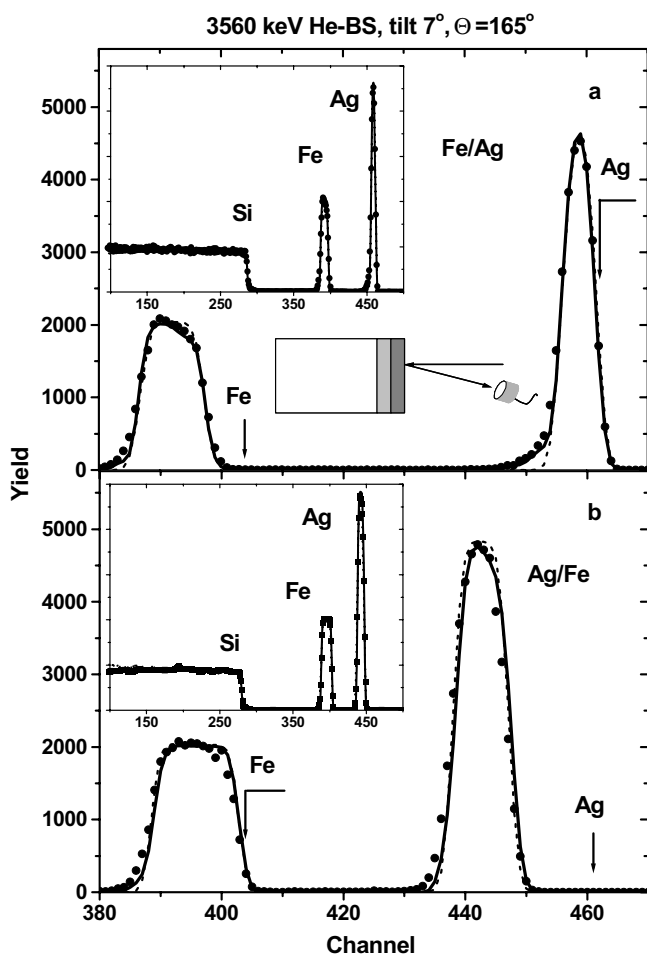

Fig. 1. BS spectra taken on the bilayer-samples together with two simulations: (a) for Fe/Ag and (b) for Ag/Fe. Dots: measured spectra, dashed line: simulation taking into account the roughness distribution determined by AFM, solid lines: simulation using elemental distributions.
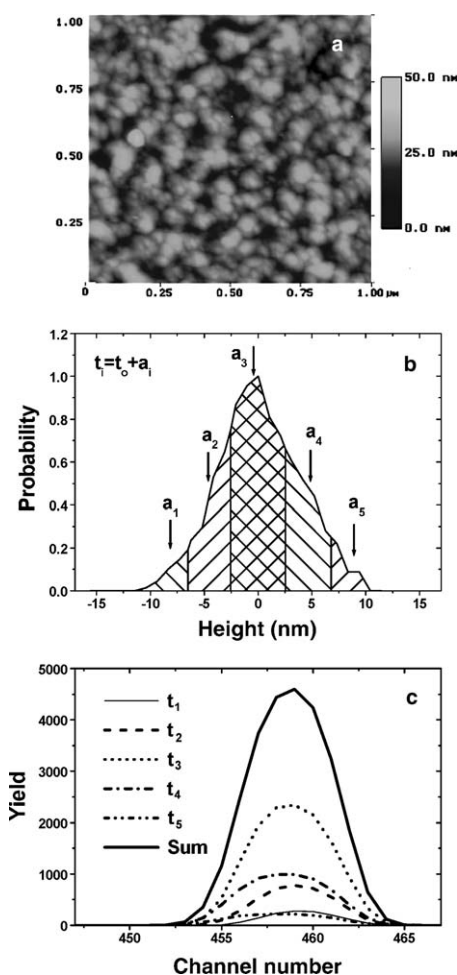

Fig. 2. (a) AFM image recorded on the Fe/Ag bilayer sample. (b) Height distribution extracted from the AFM image. The distribution is substituted by discrete thicknesses, t_i (indicated by arrows) and weight factors w_i. (c) Subspectra corresponding to the various thicknesses in the simulation.

low-energy tail at the interface. Conversely, for Ag/Fe an abrupt interface can be seen. Let us note that this result is in contradiction to those published in [8,9], where the Fe/Ag interface was found to be very sharp. Although the correct explanation of this contradiction and the nature of the Ag tail are missing, we can exclude the following possibilities. No impurities have been found in the single-layer samples (not shown), therefore we can exclude that, due to not properly closed shutters, co-evaporation occurred. Furthermore, the single Ag layer on the Si substrate has no tail, clearly confirming that the tail of the Fe/Ag sample cannot be the consequence of plural or double scattering.

We can exclude the Ag 'implantation' hypotheses, as well. The range of the 0.1-eV Ag atoms being about 0.1 nm [11] is much less than the tail length of the Ag layer. It is about 68 nm, i.e. comparable to the thickness of the iron layer (78 nm).

The surface roughness of the bilayers determined by AFM can be assigned mainly to the inhomogeneity of the top-layer, since the roughness of the single layers is much less than that of the bilayers. The FWHM of the height distributions of the single layers was about 2 nm, while for the bilayers it was 9 and 2.6 nm for Fe/Ag and Ag/Fe, respectively. Fig. 2 shows the surface of the Fe/Ag sample determined by AFM and the corresponding height distribution.

In RBX simulations, the roughness distributions were taken into account by superposing five different thicknesses with the corresponding weight factors (see Fig. 2). In Fig. 1, the simulation taking into account the roughness distribution fails to explain the tail of the Ag peak at the Fe/Ag interface. A better agreement can be reached only by assuming that Ag enters the Fe layer.

A large amount of oxygen has been found in the annealed samples, as shown in Fig. 3. This means that oxidation occurred during the annealing. In the case of Fe/Ag bilayer the layers are completely mixed to each other. A possible explanation might be that Fe first migrated into the Ag layer then the Fe–Ag mixture gets oxidized from the surface. The Ag/Fe sample can be described with well separated Fe and Ag oxide layers. In this case the surface Fe layer has been quickly oxidized, preventing further diffusion of Ag into Fe.

4. Conclusions

BS spectra of Fe/Ag and Ag/Fe interfaces indicate a difference in the interface shape. In case of Ag grown on Fe, the Ag peak shows a long low-energy tail at the interface, while for Fe grown on Ag, the interface is abrupt. This tail cannot be caused by double or plural scattering in the BS measurements. AFM measurements evidence that the surfaces of all the deposited single and double layers are too smooth to cause the observed asymmetry in the interfaces.

After annealing, both samples show mixing between the two layers; this mixing is much larger for the Fe/Ag bilayer again. The Ar atmosphere was not clean enough to prevent intensive oxidation; further annealing experiments are in progress under UHV conditions.
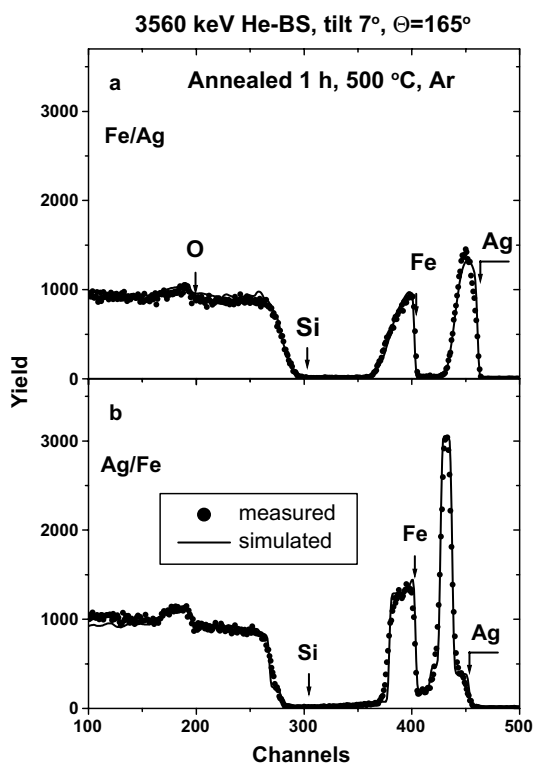
Fig. 3. Measured and simulated BS spectra taken on the bilayer-samples after annealing together with simulations: (a) for Fe/Ag and (b) for Ag/Fe. A large amount of oxygen is found in both samples.

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