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Effect of interface roughness on magnetic multilayers of Fe/Tb and Fe/Cr

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Abstract

The effect of systematic variation in the correlated interface roughness on perpendicular magnetic anisotropy (PMA) and giant magnetoresistance (GMR) has been studied in Fe/Tb and Fe/Cr multilayer systems, respectively. Multilayers for each system were deposited simultaneously on a set of float glass substrates pretreated with varying rms surface roughness. In both the systems the amount of intermixing at the interfaces and other morphological parameters are found similar, thus allowing one to separate out the effect of interface roughness only. X-ray reflectivity, diffuse scattering, conversion electron Möβbauer spectroscopy and superconducting quantum interference device magnetometry are used to characterise the systems. With the increase in σ, the PMA in Fe/Tb as well as the GMR in Fe/Cr shows a small decrease. The observed effects are mainly due to the changes in the correlated part of the roughness of the multilayers, while the uncorrelated part of the σ of different multilayers are expected to remain similar.

1. Introduction

Magnetic multilayers (MLs) showing properties like perpendicular magnetic anisotropy (PMA) in systems like Fe/Tb MLs or giant magnetoresistance (GMR) in Fe/Cr MLs are significantly affected by their interfacial structures [1,2].

Earlier studies on Fe/Tb MLs, to see the effect of interfacial modifications on PMA, were mainly done by post-deposition treatments like thermal annealing [3] or ion irradiation [1]. However, the induced effects include changes in geometrical roughness as well as intermixing/demixing at the interface. Therefore, it has not been possible to separate the effects of interface roughness (σ) from that of intermixing/demixing [1]. Experimental results on the effect of σ on GMR are also conflicting. It has been seen that depending upon the ratio of the spin asymmetry for the interface and bulk scattering and the various techniques used to modify the interfaces there is either an increase or decrease of GMR with roughness [2]. Therefore, in the present study, MLs for both the systems of Fe/Tb and Fe/Cr are deposited on substrates pretreated with varying surface roughnesses. It has been seen that except for the interface roughnesses, other microstructural features of the ML like grain size, coherence length, grain texture, intermixing at the interface, internal stresses etc. are similar, thus allowing one to selectively study the effect of interface structure (varied systematically) on PMA in Fe/Tb and on GMR in Fe/Cr MLs.

2. Experimental details

Substrates with varying surface roughness were prepared in two sets by etching the float glass (FG) substrates in dilute HF for different periods of time. Set1: Eight substrates with increasing etching times of 0, 15, 30, 60, 90, 120, 150 and 180 s, designated as S1–S8, respectively, were taken. The multilayer consisted of 20 bilayers of composition 3.0 nm Fe/2.0 nm Tb, were deposited on FG substrates. Set2: A set of substrates were prepared for 14 different etching times which are

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numbered as S1– S14 and which show similar results as that of Set1. In this set MLs consisted of the following deposition sequence: substrate/Cr (10.0 nm)/[Fe (3.0 nm)/Cr (1.2 nm)]$_{\times 20}$/Fe (5.0 nm). Deposition conditions are similar as reported in Refs. [2,3]

A powder X-ray diffractometer model D5000 of Siemens with Cu K_α radiation was used to measure the specular (XRR) and diffuse scattering geometry (XDS) [4]. ^{57}Fe conversion electron Möβbauer Spectroscopy (CEMS) was used to get information about the intermixing at the interface and the PMA at room temperature using a gas flowing (95% He, 5% CH_4) proportional counter. The spectral profiles were analysed by means of the NORMOS code developed by Brand [5]. The magnetic texture of the sample is revealed by the intensity of the 2nd and 5th peaks relative to the inner ones of the Möβbauer spectrum. RF SQUID measurements were done at 4.2 K (QUANTUM DESIGN model MPMSR2) with the field being in the film plane and the ratio of magnetic remanence M_r and the magnetic saturation M_s was used to infer the extent of antiferromagnetic coupling fraction (AFF) given by $(1 - M_r/M_s)$ [6].

3. Results and discussion

3.1. Fe/Tb MLs

The XRR pattern of the float glass substrates of Set1 and Set2 subjected to different etching time (see Ref. [2]) show distinct oscillatory variation of the substrate roughness with increasing etching time. The fitting of the reflectivity and rocking curve patterns for the substrates, done by simulations following theories [4,7], gives the value of σ, ξ (lateral correlation length) $\sim 450 \pm 50$ nm and h (Hurst parameter measuring jaggedness) $= 0.2 \pm 0.1$.

CEMS spectra for the MLs are fitted with two subspectra: one sharp (α-Fe) and other broad one corresponding to the Fe atoms at the interface. Intermixed layer thickness inferred from the area under the sharp sextet shows no significant increase (1.0 ± 0.3 nm) with etching time and is essentially being used as an input parameter in XRR curve fitting. The probability of hyperfine field distribution $P(B_{hf})$ and the average $\langle B_{hf} \rangle (T)$ was also similar. Fig. 1 shows the *specular* (subtracted off the off-specular) X-ray patterns for the specimens. The patterns clearly show the first-order Bragg peak due to ML periodicity and a distinct oscillatory variation of the σ with increasing etching time. The values of the substrate (σ_s) and the interface roughness (σ_i) for the MLs with sample nos. S1, S2, S4, S5 and S6 are also given with the figure. The similar increment in σ^2 ($\sigma_i^2 - \sigma^2$) from substrate to the ML interfaces signifies that the change in roughness by substrate roughness variation is only affecting the correlated part of the roughness of the MLs while the uncorrelated part of the roughness remains unaffected. The parameters could not be extracted for the sample nos. S3, S7 and S8 as the intensity of the off-specular scans is comparable to that of the specular scans, which also signifies that the peak at the first Bragg position for the specimens with higher substrate roughness is arising due to the correlated part of the roughness [8] only. The

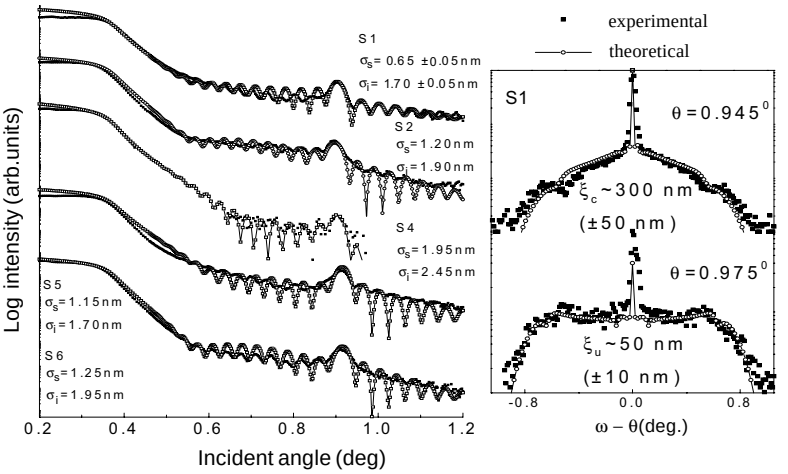

Fig. 1. XRR scans of [Fe(3.0 nm)/Tb(2.0 nm)]$_{\times 20}$ multilayers along with their fit deposited on FG substrates with different etching times. The substrate roughness (σ_s) and interface roughness (σ_i) are shown. The inset shows the transverse (ω) scan for S1 along with the fit at two different angles of θ corresponding to the position at the Bragg peak and at an off-set to it. At $\omega - \theta$ the specular peak is seen over a diffuse background. For clarity, various curves are shifted relative to each other along the y-axis.

fit to the rocking curve (inset for S1) shows the correlation length for the correlated and uncorrelated part of σ as $\xi_c \sim 300$ nm and $\xi_u \sim 50$ nm with $h = 0.5 \pm 0.02$ and do not change with σ· The polycrystalline nature of the Fe layer in the Fe/Tb MLs is confirmed for all the specimens from the XRD measurements.

The angle ϕ ($\pm 3.0^\circ$) (between the film normal and the average direction of the magnetic moments) as obtained from the fit of the room temperature CEMS spectra of four representative samples S1, S4, S5, and S8 is $\sim 42^\circ$ [9]. A small decrease in PMA though may be observed for sample no. S8 ($\phi \sim 54^\circ$) whose roughness is comparable with the thickness of the layer. It may be noted from an earlier study [1] that a small change in σ (~ 0.45 nm) by 80 MeV Si ion-irradiation causes the angle ϕ to decrease by 6.3°, whereas in the present case the roughness has been increased by 2.5 nm resulting in a similar change in ϕ. The intermixed layer thickness has also been found to increase (upon 150 MeV Ag ion-irradiation [1]) or decrease (by thermal annealing [3]) causing the PMA to decrease largely. Therefore, in both post-deposition treatments where the σ variation is expected to be uncorrelated is seen to be associated with a possible stress relaxation within the bulk of the layers causing the PMA to decrease but a correlated change in roughness does not affect the PMA significantly.

3.2. Fe/Cr MLs

In Set2 the substrate roughness variation, the σ and ξ (from XRR and XDS) and the intermixed layer thickness (from CEMS) of the MLs are found to behave similarly as in Set1. XRD measurements have shown that the structural coherence length (ζ), grain size, internal stresses and the texture (1 1 0) do not vary from sample to sample. Furthermore, since all the films were deposited simultaneously, the deposition conditions like deposition rate and substrate temperature are identical for all the specimens; therefore, the individual layer thicknesses as well as the density of defects in the bulk of the layers is expected to be similar.

Thus, the only difference between various MLs deposited on different substrates is in their σ, and the observed variation in GMR can solely be attributed to the variation in the σ. The GMR ratio is defined as $(R_0 - R_s)/R_s \times 100(\%)$, with R_0 and R_s being, respectively, the resistance values at zero and saturating fields. It is interesting to note that with increase in etching time as shown in Fig. 2, the variation in GMR is highly correlated with that in the roughness. The difference in the interfacial roughness in different MLs is essentially due to the difference in the roughness of their substrate which is transmitted to the successive layers. Thus, the difference among various MLs is expected to be in their correlated part of the interfacial roughness (similarly as in case of PMA in Fe/Tb MLs). The uncorrelated

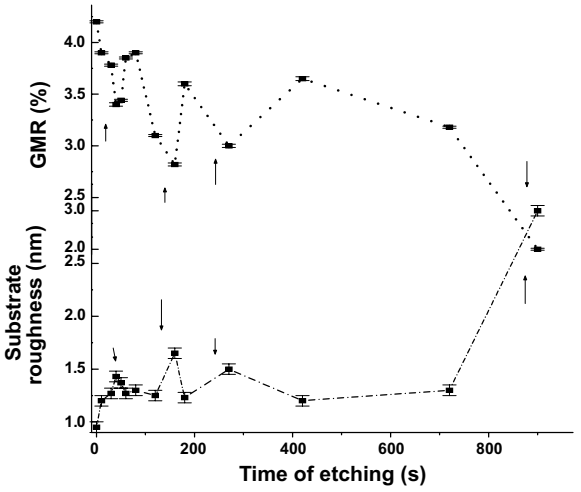

Fig. 2. The plot of surface roughness of the FG substrates and the corresponding GMR ratio as obtained from the fit to the XRR data and the magnetoresistance measured. The arrows indicate the points for variation maximum/minimum in roughness/GMR.

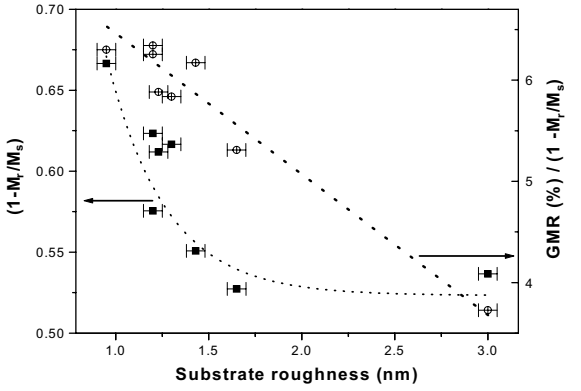

Fig. 3. The change in AFF (■) with increase in substrate roughness as obtained from SQUID measurements. Also shown is the GMR normalised to AFF (⊕) with increasing roughness.

roughness in all the MLs is expected to be similar in magnitude because of the identical conditions of deposition. The AFF showing a saturating behaviour with increase in roughness is plotted in Fig. 3. Normalising the GMR (%) ratio with AFF gives the contribution due to the interfacial scattering with the increase in roughness, which is also plotted. One may see from the figure that while the decrease in the AFF is $\sim 20\%$, the interfacial scattering alone can bring $\sim 40\%$ decrease in GMR ratio for a change of $\sim 70\%$ in correlated roughness in a range of few nm. This change in GMR

is smaller as compared to $\sim 65\%$ decrease due to 200 MeV Ag ion irradiation effects as observed in an earlier study [10]. The interface structure modification due to ion irradiation effects are expected to be uncorrelated and thus a small increase in roughness can cause a large decrease in GMR.

In conclusion it has been seen that keeping all other parameters unchanged a large change only in the correlated part of the interface roughness can be caused by etching the substrates for different periods of time. This correlated variation is expected to have a smaller effect on the RE–TM bonds at the interface of Fe/Tb systems or on the interfacial scattering in Fe/Cr MLs compared to the uncorrelated changes at the interfaces caused by other post-deposition treatments as ion irradiation, thermal annealing and in situ modification of roughness. Thus a small decrease in PMA and in GMR is observed with increase in roughness.

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