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Effect of interface structure correlation on magnetoresistance of Fe/Cr multilayers

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Abstract

Effect of interfacial roughness on giant magnetoresistance (GMR) in Fe/Cr multilayers has been studied. A set of samples is prepared by simultaneously depositing on a set of float-glass (FG) substrates with varying rms surface roughness. This causes the correlated part of the rms roughness to vary from sample to sample. Another set of specimen is irradiated with 200 MeV Ag ions in order to induce uncorrelated roughness at the interfaces. In both the cases morphological and other microstructural features of different multilayers remained similar, thus allowing one to separate the effect of interface roughness from that of morphological changes. GMR measurements on these multilayers show that increasing interfacial roughness causes GMR to decrease nonlinearly. It is found that the effect of uncorrelated part of the roughness is much stronger than that of the correlated part. © 2001 Elsevier Science Ltd. All rights reserved.

Keywords: Magnetic films and multilayers; Giant magnetoresistance; Interface structure and roughness

1. Introduction

Giant magnetoresistance (GMR) in metallic multilayers [1] continues to be a topic of great interest. A fairly good understanding of the basic phenomena, including the origin of the interlayer coupling and the spin-dependent electron scattering has been reached [2]. However, one aspect of the GMR phenomenon which is still not understood properly is the role of the interface roughness in determining the GMR. Experimentally the effect of interface roughness on GMR has been studied by varying the deposition conditions like sputtering pressure, sputtering power [1,3–6], substrate temperature [7] or by post-deposition treatments like ion irradiation [8] and thermal annealing [9]. But the neglect of the effects of associated changes in the morphological and other microstructural features of the films on GMR are responsible for the contradictory results in the above studies [1,3–12]. It may be noted that variations in the deposition conditions or the post-deposition treatments, besides affecting the interface quality, are also expected to affect other film properties like grain size and morphology, grain texture, internal stresses and defect concentration in the bulk of the layers, etc. which in turn affect the GMR in the multilayers [10–16]. Therefore, in the present work we have tried to separate the effect of interface roughness on GMR from that of morphological changes. The interface roughness σ can in general be written as the sum of

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two contributions $\sigma^2 = \sigma_c^2 + \sigma_u^2$, where σ_c is the correlated part and σ_u is the uncorrelated part of the interface roughness [13]. Therefore, two sets of multilayers are prepared in which either the correlated part or the uncorrelated part of the interface roughness is varied in a controlled manner. Characterization of the multilayers using X-ray reflectivity (XRR), X-ray diffraction (XRD), conversion electron Mossbauer spectroscopy (CEMS) and Atomic Force Microscopy (AFM) shows that except for the interface roughnesses, other microstructural features of the multilayer (ML) like grain size, coherence length, grain texture, intermixing at the interface, internal stresses etc. remain unchanged, thus allowing one to selectively study the effect of interface structure on the GMR.

2. Experimental

Float glass was used as substrate for depositing multilayers. Substrates with varying surface roughness were prepared by etching the float-glass substrates in dilute HF for varying periods of time. Six substrares with etching times of 0, 15, 60, 300, 600 and 1200 s, designated as specimens 1–6, respectively, were taken (set I). XRR measurements showed that the rms surface roughness varied non-monotonically with etching time. Multilayers were deposited on these substrates simultaneously in a UHV chamber using two e-beam guns (TELEMARK Model No. 528) at a rate of 0.01 nm/s. The base pressure in the chamber was 8.0×10^{-10} mbar. The source to substrate distance was kept at 60 cm, in order to ensure uniformity of layer thickness (within 0.5%) on different substrates. Thicknesses of individual layers were controlled during deposition using a standard quartz-crystal oscillator. Multilayers consisted of the following deposition sequence: substrate/Cr (10.0)/[Fe (3.0)/Cr (1.2)] $\times$ 20/Fe (5.0), where the numbers in brackets give the layer thickness in nm. Cr spacer layer thickness of 1.2 nm corresponds to the first peak in the antiferromagnetic coupling between Fe layers [1].

Another set of multilayers (set II) of composition [Fe (3.0 nm)/Cr (1.2 nm)] $\times$ 20/Fe (5.0 nm) samples deposited on FG and Si (1 1 0) substrates were irradiated with 200 MeV Ag ions up to fluences of 2.0×10^{11}, 5.0×10^{11}, 1.0×10^{12}, 5.0×10^{12}, 1.5×10^{13} and 3.0×10^{13} ions/cm^2 using 15 UD Pelletron at Nuclear Science Centre, New Delhi.

XRR and AFM were used to characterize the substrate as well as the multilayers. XRD was used to determine the grain texture and size of the multilayers. CEMS was used to gather information about the intermixing at the interface. For XRR a powder X-ray diffractometer model D5000 of Siemens with CuKα radiation was used. CEMS measurements were done using a gas flow proportional counter and a 50 mCi ^{57}Co source in Rh matrix. Magnetoresistance at room temperatures was measured using the standard four-probe technique with a constant current source and a nanovoltmeter in an external field upto 1 T. The field was applied parallel to the plane of the film and perpendicular to the electrical current which was also in the same plane.

3. Results

Table 1 summarizes the results of various measurements of the substrate as well as of the films in the case of set I. Column 3 of Table 1 gives the rms roughness of the substrates obtained by computer fitting of the X-ray reflectivity data on substrates etched for different periods of time. One may note that the surface roughness exhibits a non-monotonic variation with etching time: after reaching a maximum value of 1.25 nm for etching time of 300 s, it again decreases with further etching. This reflects some sort of layer by layer removal of the surface during etching.

XRD measurements showed that the films have a texture along the (1 1 0) direction. However the texture does not vary from sample to sample. The width of the (1 1 0) reflection was used to determine the structural coherence length ξ of grains along the momentum transfer vector q using the Scherrer method, and is reported in Table 1. It may be noted that ξ has several times the thickness of individual layers indicating a high-degree of coherency between adjacent Fe and Cr layers. Further, one finds that ξ does not vary with substrate roughness. The *d*-spacing of (1 1 0) planes as calculated from the

Table 1
Microstructural parameters of Fe/Cr multilayers on float glass (FG) substrates with different etching times T. σ is the rms roughness of the glass substrates after etching for different periods of time. The values of the lattice spacing d of (1 1 0) planes, structural coherence length ξ, and average grain size in x–y plane t is also reported. The relative area A under the broad hyperfine field component in the CEMS gives the fraction of total iron atoms located at the interfaces and is a measure of the thickness of the intermixed layer. The last column gives the saturation magnetoresistance of the multilayer

No.	T (s)	σ (nm)	d (nm)	ξ (nm)	t (nm)	A (%)	GMR (%)
1	0	0.67 ± 0.05	0.2025 ± 0.0005	16.3 ± 1.0	248	26	3.52 ± 0.01
2	15	0.77	0.2026	15.3	290	—	3.08
3	60	0.92	0.2025	15.6	295	—	2.94
4	300	1.25	0.2025	15.5	291	26	2.83
5	600	0.95	0.2028	15.9	266	25	3.19
6	1200	0.85	0.2029	16.1	261	—	3.22

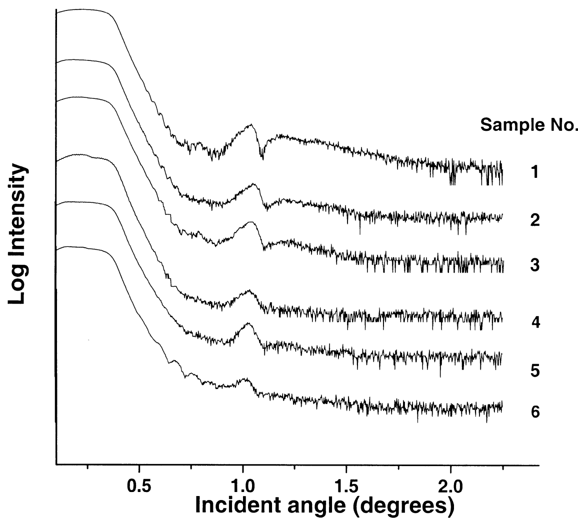

Fig. 1. X-ray reflectivity scans of Fe/Cr multilayers on float glass (FG) substrates with different etching times and on microscopic glass slide (SG). For clarity, various curves are shifted relative to each other along the y-axis.

position of (1 1 0) reflection is also reported in Table 1. The d-value and hence the internal stresses in the film also do not vary from sample to sample.

Fig. 1 shows the reflectivity pattern of the multilayers deposited on different substrates. The first Bragg peak due to multilayer periodicity is clearly visible. However beyond the first Bragg peak the reflectivity pattern becomes obscure due to strong diffuse scattering. The presence of the Cr seed layer, an electron density gradient in the substrate and a possible oxidation of the Fe capping layer made it difficult to obtain a good theoretical fit to the experimental data. However, the following information could be obtained: (i) from the position of the Bragg peak one can find that the bilayer periodicity is 4.4 nm instead of the designed value of 4.2 nm. This difference may be due to some error in the tooling factor of the thickness monitor, (ii) the height of the Bragg peak which is related to the average interface roughness, decreases with increasing substrate roughness, confirming that the roughness of the substrate is at least partly transferred to the successive layers.

The polycrystalline nature of the films is clearly visible from AFM pictures. For each sample 10 different frames of 1 μm × 1 μm were taken and the average grain size in the film plane was calculated. The results are reported in Table 1. CEMS measurements were done in specimens 1,4 and 5. The spectra were fitted with two distributions of hyperfine magnetic fields. The distribution in the range $28\,\text{T} < B_{\text{hf}} < 36\,\text{T}$ corresponds to the bulk of the iron layers while the broad distribution in the range $0\,\text{T} < B_{\text{hf}} < 30\,\text{T}$ corresponds to the iron atoms at the interfaces [17]. The fraction of total iron atoms located at the interfaces, which is proportional to the relative area under the broad sextet, is a measure of the thickness of the intermixed layer at the interface and is reported in Table 1. The GMR defined as $R_0 - R_s/R_s \times 100$ with R_0 and R_s being the resistance values at zero and saturating fields, respectively, is also reported in Table 1. Fig. 2 gives the variation of the GMR with the roughness σ.

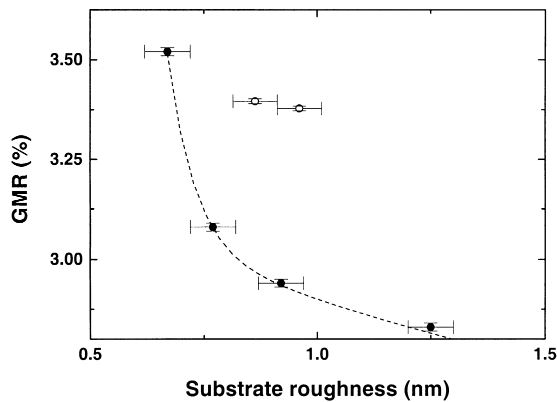

Fig. 2. Variation of percentage GMR with surface roughness of the substrates.

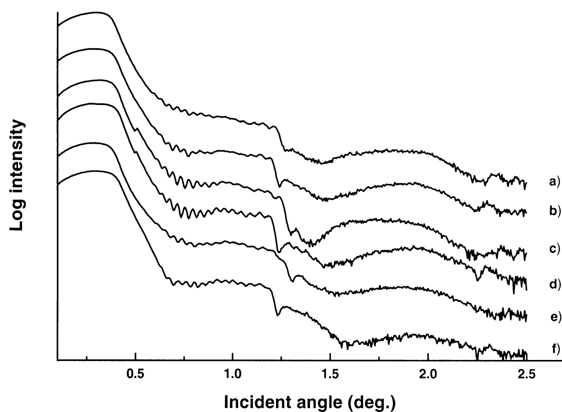

Fig. 3. XRR scans of [Fe(3.0 nm)/Cr(1.2 nm)] $\times$ 20 multilayers showing irradiated spectra with different irradiation fluences of (a) 2×10^{11} (b) 5×10^{11} (c) 1×10^{12} (d) 5×10^{12} (e) 1.5×10^{13} and (f) 3×10^{13} ions/cm^2 irradiated with 200 MeV Ag ion.

XRD measurements in the specimens of set II show that irradiation does not affect the morphological parameters like grain size, texture etc. Irradiation affects the XRR pattern indicating changes in the interface structure (Fig. 3). Because of the small contrast between Fe and Cr in their refractive indices it is not possible to fit the reflectivity data to get reliable information. However in some earlier studies it has been shown that in Fe/Tb multilayers, the interface roughness varies almost linearly with irradiation fluence [17]. Fig. 4 gives the variation of GMR as a function of irradiation fluence.

4. Discussions

Table 1 shows that grain size, grain texture, structural coherence length ξ, internal stresses and the thicknesses of the interface layers are similar for all the multilayers of set I grown on different substrates. 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 multilayers deposited on different substrates is in their interface roughness, and the observed variation in GMR can be attributed to variation in the interface roughness only.

The difference in the interfacial roughness in different multilayers is essentially due to the difference in the roughness of their substrate which is transmitted to the successive layers. Therefore, the difference among various multilayers is expected to be in their correlated part of the interfacial roughness. It is interesting to note that with increase in etching time, as the substrate roughness decreases for etching time beyond 300 s, the GMR of the corresponding multilayers also shows an increase. Thus, the observed variation in GMR in this set is due to the correlated part of the interface roughness.

From Fig. 4 one finds that the effect of 200 MeV Ag ion is again to cause a decrease in GMR. Since the modifications at various interfaces induced by irradiation are not expected to be correlated, the observed decrease in GMR is because of an increased uncorrelated part of the roughness. A comparison of Figs. 2 and 4 shows that while the effect of an increase in the correlated part of the roughness by almost 100% is to cause a decrease in GMR by only 20%, the effect of swift heavy ion irradiation is to decrease GMR by more than 60%. Thus the present study shows the effect of the uncorrelated part of the interface roughness is much stronger compared to the correlated part of the roughness.

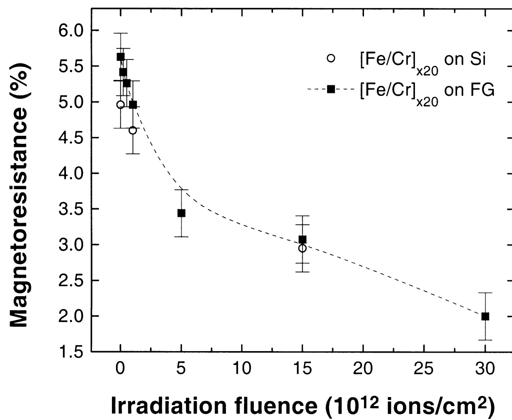

Fig. 4. GMR of the as-deposited and irradiated specimens of [Fe(3.0 nm)/Cr(1.2 nm)] $\times$ 20 multilayers deposited on float glass (- - -■- - -) and Si substrate (0) as a function of fluence irradiated with 200 MeV Ag ions.

5. Conclusions

In conclusion, the effects of correlated as well as uncorrelated interface roughness on GMR in Fe/Cr multilayers have been studied in specimens with similar morphological structure. The increasing interface roughness causes GMR to decrease. The effect of uncorrelated roughness is much stronger than the effect of correlated roughness.

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