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Journal of Magnetism and Magnetic Materials 286 (2005) 437–441
Journal of magnetism and magnetic materials
www.elsevier.com/locate/jmmm

Modification of microstructure and magnetic properties of Fe/Cr multilayers caused by ion irradiation

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Available online 22 October 2004

Abstract

The influence of Ar-ion irradiation on the microstructure, interlayer coupling and giant magnetoresistance (GMR) effect in the model Fe/Cr system was studied by the conversion electron Mössbauer spectrometry (CEMS), vibrating sample magnetometry (VSM), and magnetoresistivity, for the Fe-1.4 nm/Cr-t_{Cr} multilayers with t_{Cr} ranging from 0.73 to 1.85 nm. The loss of antiferromagnetic coupling and the simultaneous degradation of GMR, observed for increasing ion dose, was caused by the formation of pinholes due to the increase of interface roughness and at high ion doses, by the ion-beam mixing leading to the alloying of Fe and Cr at interfaces.

PACS: 68.35.−p; 75.47.De; 75.70.Cn; 76.80.+y

Keywords: Ion irradiation; Mössbauer spectroscopy; Giant magnetoresistance; Interlayer coupling; Fe/cr multilayers

1. Introduction

In thin film technologies thermal annealing and ion irradiation are the supplementary post deposition treatments enabling a modification of the microstructure and, consequently, the magnetic properties. A lateral resolution can be easily realized with ion irradiation. Therefore, a correlation between changes in the microstructure induced by irradiation, the interlayer coupling and the giant magnetoresistance (GMR) effect of multilayers is of a substantial interest. Recently, it was shown that ion irradiation may lead either to the increase of GMR effect or to the degradation of GMR, depending on an ion dose. Irradiation with 500 keV Xe-ions [1,2] induced initial increase of GMR, however, at higher ion doses the GMR was destroyed. Also the 200 MeV Ag-ion irradiation led to a decrease of GMR effect [3].

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0304-8853/$ - see front matter
doi:10.1016/j.jmmm.2004.09.061

Epitaxial Fe/Cr/Fe(0 0 1) trilayers with small thickness of Cr spacer ($t_{Cr} \leqslant 0.7$ nm) irradiated by 5 keV He ions showed a monotonic decrease of the antiferromagnetic coupling strength with increasing ion dose [4]. In the present experiment we have used 200 keV Ar-ion irradiation to modify the interface quality in Fe/Cr multilayers in a controlled way. The influence of the interface microstructure on the magnetic properties (interlayer coupling, GMR, hysteresis loop, etc.) is studied for model Fe/Cr multilayers.

2. Experiment

The Fe-1.4 nm/Cr-t_{Cr} ($0.97 \leqslant t_{Cr} \leqslant 1.85$ nm—the thicknesses correspond to the first antiferromagnetic coupling range for Fe/Cr multilayers [5–8]) polycrystalline multilayers were deposited on oxidized Si wavers using UHV magnetron sputtering (DC and RF for Fe and Cr, respectively). Total thickness of the Fe/Cr film was about 100 nm. Samples were irradiated at room temperature (RT) with 200 keV Ar ions and doses D_{Ar} ranging from 5×10^{12} to 2×10^{14} Ar/cm^2. A penetration range of ions matched well with the total film thickness. The as-deposited and irradiated samples were characterized at RT by the conversion electron Mössbauer spectroscopy (CEMS) and vibrating sample magnetometer (VSM) hysteresis loops. Magnetoresistance and resistivity were measured at RT using the four-probe technique in CIP geometry. The GMR(H) dependencies were determined as GMR(H) = $100 \times [R(H) - R(H = 2\,\mathrm{T})]/\mathrm{R}(H = 2\,\mathrm{T})$ (where H is the magnetic field); the maximal value of GMR(H) determines the GMR amplitude.

3. Results and discussion

The CEMS spectra for as-deposited and irradiated samples were fitted in terms of the model [9] in which four individual magnetic components are associated with different iron environments: component $H_1 \approx 33$ T, corresponds to the bulk Fe sites; components $H_2 \approx 30$ T and $H_3 \approx 24$ T, are related to the "step" sites at the Fe/Cr interfaces, and $H_4 \approx 20$ T, corresponds either to the "perfect" interface sites or to some other "step" positions. For Fe/Cr multilayers with Fe layer thickness $t_{Fe} = 1.4$ nm characterized by ideally smooth interfaces the expected relative fraction of H_1 and H_4 components should be 71% and 29%, respectively, and the contributions of H_2 and H_3 components should be zero [10]. In fact even for as-deposited samples, the measured fractions of H_1 and H_4 components are significantly smaller than expected and H_2, H_3 fractions show nonzero values. The relative fraction of H_1 component of about 40% indicates that only 0.6 nm of each Fe layer corresponds to bulk Fe sites. The remaining Fe atoms, corresponding to H_2, H_3, and H_4 components, together with Cr atoms form the interface regions.

Typical CEMS spectra recorded for Fe(1.4 nm)/Cr(1.4 nm) multilayers after ion irradiation with ion doses indicated are shown in Fig. 1. The significant changes in CEMS spectra of Fe/Cr multilayers corresponding to microstructure modification caused by Ar-ion irradiation are already detected at ion doses $D_{Ar} \geqslant 5 \times 10^{12}$ Ar/cm^2. Nearly the same increase of the fraction of Fe atoms at interfacial positions (spectral contributions of H_2–H_4) accompanied by the decrease of bulk positions of Fe atoms (H_1 component) is observed for all Fe/Cr multilayers. Summarizing the CEMS measurements we can conclude that the interface roughness is similar for all samples in the as-deposited state and that it increases with increasing D_{Ar} independently of t_{Cr}.

For multilayers with small thicknesses of Cr spacers and with an uncorrelated interface roughness caused by grain boundary diffusion during deposition, there is a certain probability of creating ferromagnetic bridges (pinholes) across Cr layers. Their existence in antiferromagnetically coupled structures leads to a strong ferromagnetic coupling localized in the vicinity of pinholes [11,12]. As a result, the antiferromagnetically coupled fraction F_{AF} ($F_{AF} = 1\text{-}M_R/M_S$, where M_R and M_S are the remanence and saturation magnetization, respectively) is smaller than one and the biquadratic component of interlayer coupling J_2 become significant (the relatively strong J_2 component is also observed for the

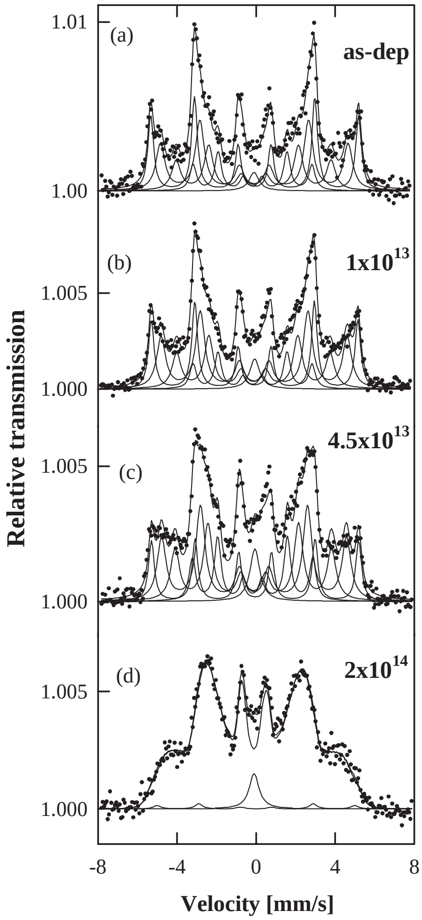

Fig. 1. CEMS spectra recorded for Fe(1.4 nm)/Cr(1.4 nm) multilayers in the as-deposited state (a) and after ion irradiation with doses indicated (b–d).

structures with spacer thickness corresponding to the transition between the antiferromagnetic and ferromagnetic coupling [13]). Such a behavior, typical for polycrystalline multilayers, is also observed in our as-deposited samples (Figs. 2a, c and 3). It should be noted that the GMR effect is correlated only with antiferromagnetically coupled regions (see Fig. 2 and Ref. [8]).

Simultaneously with subtle changes of CEMS spectra (Fig. 1) the distinct changes in magnetization reversal and magnetoresistance curves are observed for Fe/Cr multilayers with thin ($t_{Cr} <$ 1.4 nm) spacer layers (Fig. 2a, b). The increase of remanence, M_R, and decrease of saturation field,

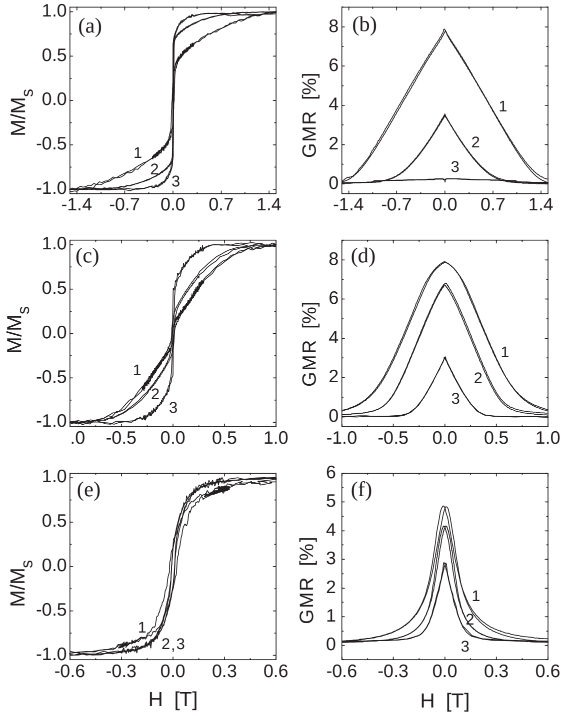

Fig. 2. Hysteresis loops and GMR(H) dependencies for as-deposited and ion irradiated Fe(1.4 nm)/Cr(t_{Cr}) samples with $t_{Cr} = 0.97$ nm (a,b), $t_{Cr} = 1.4$ nm (c,d) and $t_{Cr} = 1.55$ nm (e,f). The curves 1, 2 and 3 denote samples in the as-deposited state and irradiated with ion doses of 2×10^{13} and 8×10^{13} Ar/cm^2, respectively.

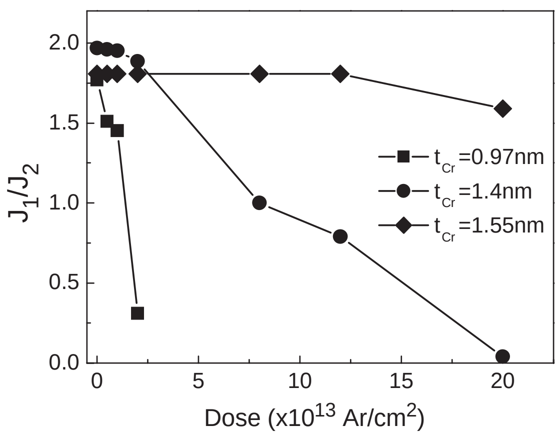

Fig. 3. Ratio of bilinear (J_1) and biquadratic (J_2) components of interlayer exchange coupling vs. ion dose, shown for t_{Cr} indicated.

H_S, can be recognized in the hysteresis loops after successive steps of irradiation. Simultaneously irradiation leads to the gradual disappearance of linearity of $M(H)$ dependence. We applied the procedure, proposed by Fullerton and Bader [15], of extracting the bilinear, J_1, and the biquadratic, J_2, terms of the exchange coupling from $M(H)$ dependences. Fig. 3 shows that the J_1/J_2 ratio decreases with increasing D_{Ar} (the possible changes of intrinsic magnetic properties of Fe layers, caused by irradiation, were not taken into account). Such a behavior provides the evidence for the growth process of existing pinholes and creation of the new ones [11]. In order to support this conclusion, the room temperature dependencies of H_S, GMR and F_{AF} versus t_{Cr} are shown in Fig. 4. The room temperature $H_S(t_{Cr})$ and GMR(t_{Cr}) dependencies of the as-deposited samples are related to the oscillatory behavior of interlayer exchange coupling that has the local maximum at $t_{Cr} \approx 1.1$ nm [5–8]. For multilayers with similar microstructures (see discussion of CEMS measurements), the maximum of the

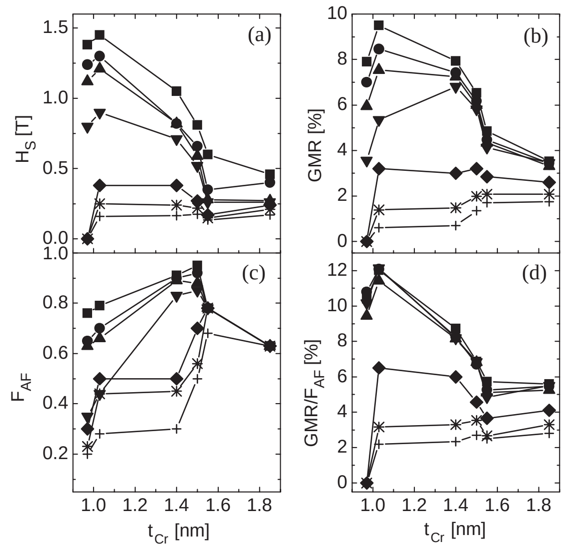

Fig. 4. The saturation field H_S (a), GMR amplitude (b), antiferromagnetically coupled fraction F_{AF} (c) and GMR/F_{AF} (d) of Fe(1.4 nm)/Cr(t_{Cr}) multilayers as a function of t_{Cr} for as-deposited samples (!) and after successive irradiation with ion doses 5×10^{12} Ar/cm^2 (,); 1×10^{13} Ar/cm^2 (7); 2×10^{13} Ar/cm^2 (B); 8×10^{13} Ar/cm^2 (Λ); 1.2×10^{14} Ar/cm^2 (Σ); 2×10^{14} Ar/cm^2 (+).

$F_{AF}(t_{Cr})$ dependence (Fig. 4c) is observed for higher Cr layers thickness ($t_{Cr} \approx 1.5$ nm). The spacer thickness is also responsible for the sensitivity of the changes of the magnetic properties due to the ion irradiation. A strong degradation of the antiferromagnetic coupling (decrease of H_S, GMR, F_{AF} with D_{Ar}) for low ion doses ($D_{Ar} \leqslant 2 \times 10^{13}$ Ar/cm^2) is observed only for samples with small thickness of the spacer layer ($t_{Cr} \leqslant 1.4$ nm). Such a behavior is related to the increase of the uncorrelated interface roughness independent of t_{Cr}. Therefore, the probability of pinhole creation is higher for multilayers with smaller t_{Cr}. As a consequence, also the changes of the J_1/J_2 ratio with D_{Ar} are stronger the thinner is the spacer layer. Moreover, the proportionality between GMR amplitude and F_{AF} parameter should be observed assuming that only antiferromagnetically coupled regions contribute to the GMR effect. Negligible changes in the ratio GMR/F_{AF} with D_{Ar} are observed for Fe/Cr multilayers in a whole range of t_{Cr} for ion doses smaller than 2×10^{13} Ar/cm^2 (Fig. 4d). This strongly supports our interpretation indicating creation of pinhole as the main source of degradation of antiferromagnetic coupling and GMR effect. However, the loss of the correlation between F_{AF} and GMR amplitude, observed for higher ion doses, indicates that the alloying at Fe/Cr interfaces induced by ion-beam mixing, clearly observed in CEMS spectra as a change in the shape of the spectra recorded for high ion doses (Fig. 1d), starts to dominate and causes a further decrease of H_S and GMR. Higher ion doses D_{Ar} increase both the fraction of the paramagnetic component observed in the CEMS spectra and the electrical resistance [15] due to enhanced alloying apart from the increase of interface roughness. Increasing number of defects in the multilayer structure shorten the mean free path of electrons reducing in this way the GMR effect. However, the influence of ion irradiation on intrinsic properties of the ferromagnetic layers (e.g., saturation magnetization, coercivity field) should be taken into account.

In summary, two dominant mechanisms responsible for the decrease of the antiferromagnetic interlayer exchange coupling and the GMR amplitude were detected. (i) At lower ion doses the increase of the interface roughness causes the formation of pinholes. (ii) At higher ion doses an efficient ion-beam mixing leads to the formation of the alloyed Fe–Cr layers at interfaces in addition to the pinholes creation.

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