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Journal of Alloys and Compounds 382 (2004) 174–178

Journal of ALLOYS AND COMPOUNDS

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Mössbauer spectroscopy, interlayer coupling and magnetoresistance of irradiated Fe/Cr multilayers

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Received 15 September 2003; received in revised form 29 December 2003; accepted 8 January 2004

Abstract

The influence of Ar-ion irradiation on the microstructure, interlayer coupling and magnetoresistance in Fe/Cr multilayers is studied. An increase of interface roughness of Fe/Cr multilayers caused by irradiation with 200 keV Ar ions and doses exceeding 5×10^{12} Ar/cm^2 is clearly seen in conversion electron Mössbauer spectroscopy (CEMS) measurements, while the small angle X-ray diffraction (SAXRD) technique hardly detects such changes in microstructure even at the higher ion doses. This subtle modification of the microstructure induces distinct changes in magnetization reversal (increase of the remanence magnetization, decrease of the saturation fields) and strongly decreases GMR effect with increasing irradiation dose. The most prominent changes are observed for the samples with a small thickness of Cr layers. The increasing immunity of GMR effect to ion irradiation with the increasing thickness of Cr layers as well as correlation between changes in GMR and antiferromagnetically coupled sample fraction suggest that the main effect responsible for the decrease of GMR is caused by the pinholes creation. For doses exceeding 2×10^{13} Ar/cm^2 the volume intermixing seems to be a dominating mechanism responsible for further degradation of GMR and antiferromagnetic interlayer exchange coupling.

Keywords: Magnetic multilayers; Interlayer coupling; Giant magnetoresistance; Mössbauer spectroscopy; Magnetic films and multilayers; Magnetic measurements; Irradiation; Argon; Electronic transport; Nuclear resonances

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1. Introduction

The antiferromagnetic interlayer exchange coupling [1,2] and giant magnetoresistance (GMR) effect was discovered in Fe/Cr structures more than a decade ago and the theoretical explanation of this phenomenon is now well established [3]. Presently, antiferromagnetically coupled multilayers are applied not only as magnetoresistance sensors but also as artificial antiferromagnets or antiferromagnetically coupled media. For certain applications, ion irradiation may be effectively used to laterally modify the magnetic properties of such structures after preparation [4]. Therefore, the correlation between the changes of microstructure related to implantation and interlayer coupling and/or GMR effect of multilayers seems to be of 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 [5,6] induced initial increase of GMR. However, at higher ion doses it destroyed GMR. Also the 200 MeV Ag-ion irradiation led to a decrease in GMR effect in Fe/Cr multilayers [7]. Epitaxial Fe/Cr/Fe(0 0 1) trilayers with small thickness of Cr spacer ($t_{\mathrm{Cr}} \leq 0.7$ nm) irradiated by 5 keV He ions showed a monotonic decrease in antiferromagnetic coupling strength $|J_{\mathrm{AF}}|$ with increasing ions dose [4]. However, for $t_{\mathrm{Cr}} \geq 0.7$ nm, $|J_{\mathrm{AF}}|$ initially slightly increases and then decreases. The increasing immunity of GMR and antiferromagnetic coupling to ion irradiation for the thicker Cr layers was also observed in our previous paper [8] and suggests that the main effect responsible for degradation of GMR is due to pinholes creation during irradiation.

In this contribution the preliminary experimental results presented in [8] are supplemented by the new ones. In

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doi:10.1016/j.jallcom.2004.01.069

particular, our investigations are extended to a new set of Fe/Cr multilayers with various thickness of Cr spacer and to wider range of Ar ion dose.

2. Experiment

The Fe-1.4 nm/Cr-t_{Cr} ($0.97 \leq t_{Cr} \leq 1.85$ nm) polycrystalline multilayers were deposited on naturally oxidized Si wavers using UHV magnetron sputtering (dc and rf for Fe and Cr, respectively). The deposition rate (0.04 nm/s for both materials) was monitored in situ with a quartz sensor. Additionally, the repetition period ($\lambda = t_{Fe} + t_{Cr}$) and thickness of iron and chromium layers were ex situ controlled by small angle X-ray diffraction (SAXRD) and X-ray fluorescence (XRF), respectively. The total thickness of the Fe/Cr film was about 100 nm. The samples were irradiated at room temperature (RT) with 200 keV Ar ions and doses D_{Ar} ranging from 5×10^{12} to 1.2×10^{14} Ar/cm^2. A penetration range of ions matched well the total thickness. The as-deposited and irradiated samples were characterized at RT by the conversion electron Mössbauer spectroscopy (CEMS), SAXRD and 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) = $100 \times [R(H) - R(H = 2T)]/R(H = 2T)$ (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 according to the model proposed by Landes et al. [9,10]. In the model four individual magnetic components (with different value of hyperfine field HF) are associated with different iron environments: $HF_1 \approx 33$ T, corresponds to the bulk Fe sites; $HF_2 \approx 30$ T and $HF_3 \approx 24$ T, contribute to the "step" sites at the Fe/Cr interfaces, and $HF_4 \approx 20$ T corresponds either to the "perfect" interface sites or to some other "step" positions. Thus, the analysis of CEMS spectra is helpful in investigations of microstructure changes of Fe/Cr multilayers during irradiation.

Fig. 1 presents the changes of the relative fraction of particular component of CEMS spectra versus D_{Ar} for the set of Fe-1.4 nm/Cr-t_{Cr} multilayers. For ideally smooth interfaces the expected relative fraction of HF_1, HF_4 components should be 71% and 29%, respectively, and the contributions corresponding to HF_2 and HF_3 components should be zero [11]. In fact even for as-deposited samples, the measured fractions of HF_1 and HF_4 components are significantly smaller and HF_2, HF_3 fractions show nonzero values (see Fig. 1, values for $D_{Ar} = 0$). The value of relative fraction of HF_1 component of about 40% indicates that only 0.6 nm of each Fe layer corresponds to bulk Fe sites. The other Fe atoms, corresponding to HF_2, HF_3, and HF_4 components, together with Cr atoms form the interface regions. It should be noted that for multilayers with $t_{Cr} \geq 1.4$ nm there is also a weak paramagnetic component (not included in Fig. 1) related to isolated Fe atoms in Cr, besides the ferromagnetic components. The intensity of this component increases almost linearly with D_{Ar}. Exemplary, for Fe-1.4 nm/Cr-1.85 nm multilayer in as-deposited state and after irradiation with $D_{Ar} = 1.2 \times 10^{14}$ Ar/cm^2 the paramagnetic component increases from about 3% to 8%, respectively. For multilayers with thinner Cr layer the amount of isolated Fe atoms (probably proportional to t_{Cr}) was too small to be detected in CEMS spectra.

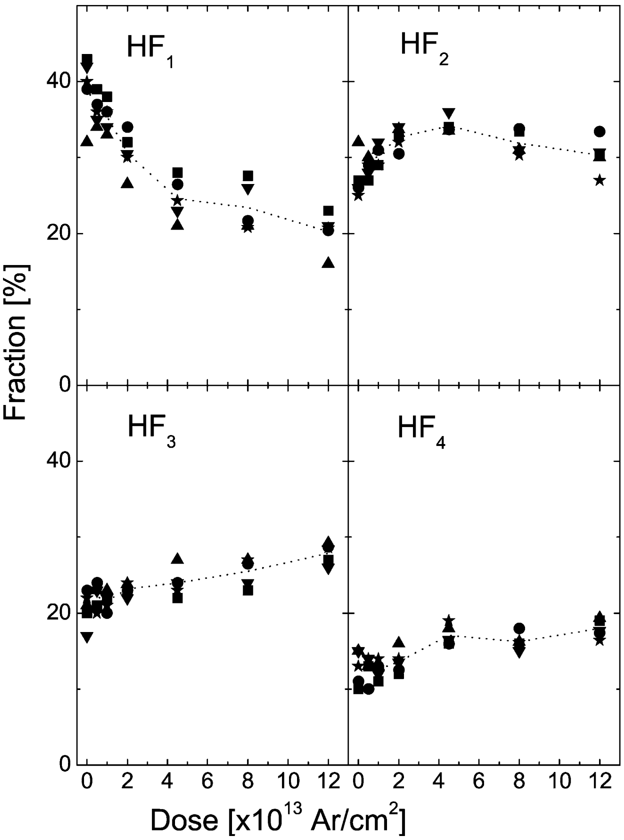

Fig. 1. Spectral fractions of HF_1–HF_4 components vs. ion dose for Fe-1.4 nm/Cr-t_{Cr} multilayers with $t_{Cr} = 0.97$ nm (■), $t_{Cr} = 1.03$ nm (●), $t_{Cr} = 1.4$ nm (▲), $t_{Cr} = 1.5$ nm (▼), $t_{Cr} = 1.85$ nm (★). The lines are guides to the eye.

For multilayers with similar roughness of interfaces the probability of the magnetic bridges (pinholes) creation across the Cr spacer increases with decreasing t_{Cr}. The existence of pinholes in antiferromagnetically (AF) coupled multilayers leads to a strong ferromagnetic coupling localized in their vicinity [12–14]. In result, the antiferromagnetically coupled fraction, F_{AF} (defined as $F_{AF} = 1 - M_R/M_S$, where M_R and M_S are the remanence and saturation magnetization determined from hysteresis loops) is smaller than one. The as-deposited Fe/Cr multilayers show nearly perfect antiferromagnetic coupling ($F_{AF} = 1$) at $t_{Cr} \approx 1.5$ nm (Fig. 2). For smaller t_{Cr} the F_{AF} factor decreases despite

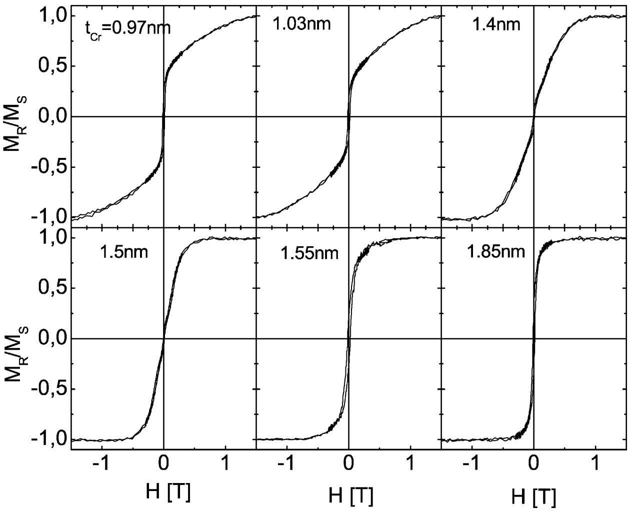

Fig. 2. Hysteresis loops of as-deposited Fe-1.4 nm/Cr-t_{Cr} multilayers with different thickness of Cr spacer.

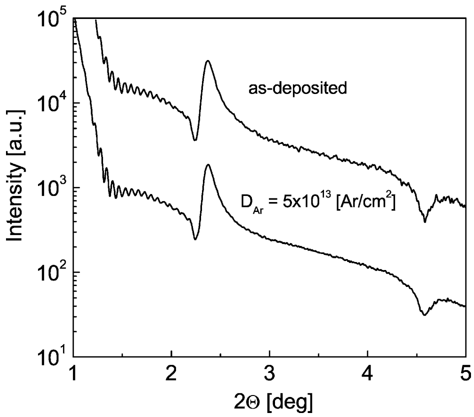

Fig. 3. Exemplary SAXRD spectra taken for as-deposited and irradiated sample (Fe-3 nm/Cr-1.1 nm). The curves are vertically shifted (by one decade) for clarity.

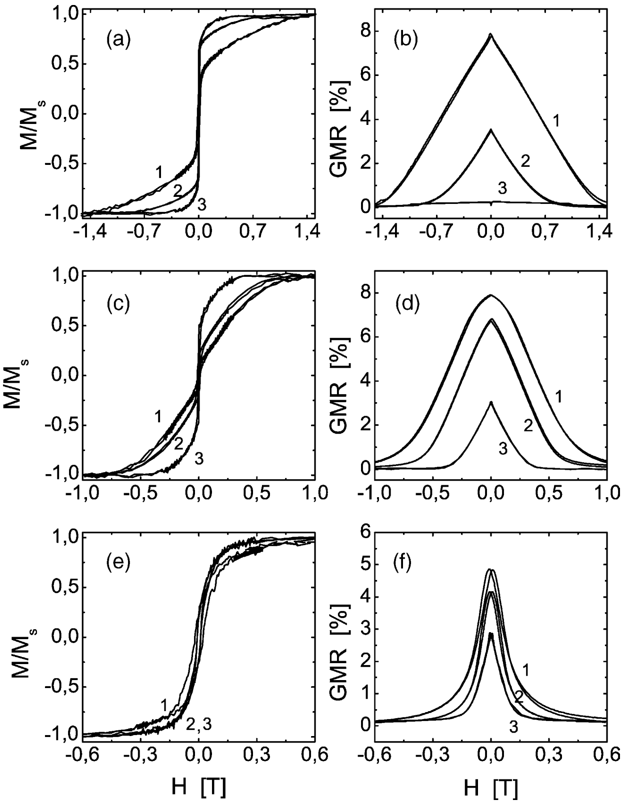

Fig. 4. Examples of hysteresis loops and GMR(H) dependences for as-deposited and irradiated Fe-1.4 nm/Cr-t_{Cr} samples with $t_{\mathrm{Cr}} = 0.97$ nm (a, b), $t_{\mathrm{Cr}} = 1.4$ nm (c, d), $t_{\mathrm{Cr}} = 1.55$ nm (e, f). The description of curves 1, 2 and 3 denote samples in as-deposited state and irradiated with doses 2×10^{13} and 8×10^{13} Ar/cm^2, respectively.

the increase in the antiferromagnetic coupling strengths and indicates an increasing role of the direct coupling through pinholes.

The changes in CEMS spectra corresponding to microstructure modification of Fe/Cr multilayers caused by Ar-ion irradiation are already detected at ion doses $D_{\mathrm{Ar}} \geq 5 \times 10^{12}$ Ar/cm^2. Nearly the same increase in the spectral contribution of HF$_2$ component accompanied by the decrease of HF$_1$ component is observed for all investigated samples. It suggests that the interface roughness (the number of Fe step sites) increases with increasing D_{Ar} independently of t_{Cr}. However, in the SAXRD spectra no changes caused by ion irradiation are detected (Fig. 3). The poor sensitivity of SAXRD method seems to be obvious taking into account a small contrast between Fe and Cr in their refractive indices. 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 ($t_{\mathrm{Cr}} < t_{\mathrm{Fe}}$ =1.4 nm) spacer layers (Fig. 4a and b). All the features characteristic for increasing density of pinholes [13], i.e., increase of M_{R} and decrease of H_{S} as well as gradual disappearance of linearity of $M(H)$ dependence (an increase of the biquadratic component of interlayer exchange coupling) can be recognized in the hysteresis loops measured after successive steps of irradiation. Obviously, due to the correlation between $M(H)$ and GMR(H) dependences (see, e.g. [15]) all the above mentioned changes in magnetic properties are also manifested in GMR(H) dependences (Fig. 4b). For a multilayer with $t_{\mathrm{Fe}} = t_{\mathrm{Cr}} = 1.4$ nm (Fig. 4c and d) the influence of irradiation is qualitatively similar. However, the modifications of particular parameters (F_{AF}, H_{S}, GMR) at $D_{\mathrm{Ar}} \leq 2 \times 10^{13}$ Ar/cm^2 are significantly smaller than for the samples discussed before. The increasing immunity of the F_{AF} parameter and GMR amplitude degradations to ion irradiation with $D_{\mathrm{Ar}} \leq 2 \times 10^{13}$ Ar/cm^2 is more distinct for multilayers with $t_{\mathrm{Cr}} > t_{\mathrm{Fe}}$ (see Fig. 4e and f). However, for multilayers with $t_{\mathrm{Cr}} \geq t_{\mathrm{Fe}}$ and $D_{\mathrm{Ar}} \geq 4.5 \times 10^{13}$ Ar/cm^2 a distinct decrease of GMR effect is accompanied by negligible changes in the shape of hysteresis loops.

To summarize the study concerning the influence of Ar-ion irradiation process on the magnetic properties of

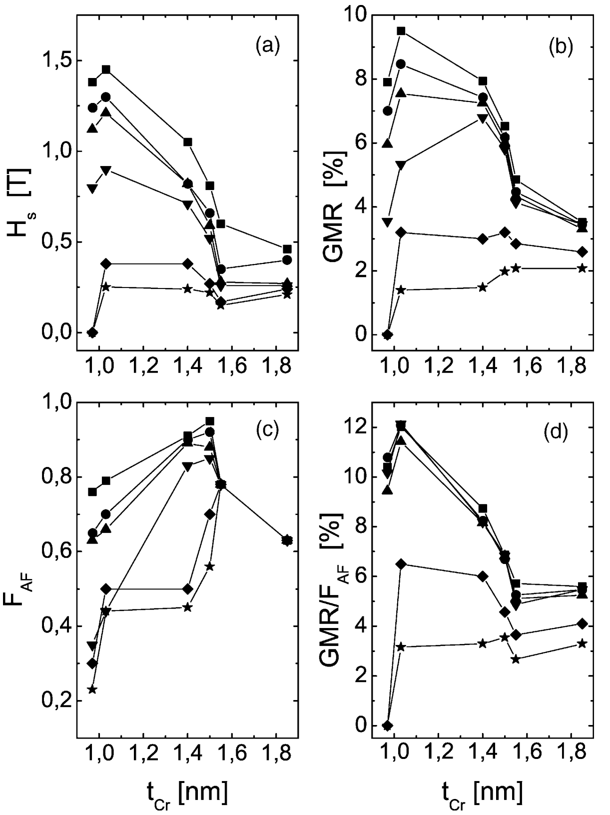

Fig. 5. The dependences of saturation field H_S, GMR value, antiferromagnetically coupled fraction F_{AF} and GMR/F_{AF} of Fe-1.4 nm/Cr-t_{Cr} multilayers as a function of t_{Cr} for the as-deposited samples (■) and after successive irradiation with doses 5×10^{12} Ar/cm^2 (●); 1×10^{13} Ar/cm^2 (▲); 2×10^{13} Ar/cm^2 (▼); 8×10^{13} Ar/cm^2 (◆); 1.2×10^{14} Ar/cm^2 (★).

Fe/Cr multilayers with different Cr layers thicknesses, the dependences of H_S, GMR and F_{AF} versus t_{Cr} are shown in Fig. 5 for the as-deposited samples and after successive steps of irradiation process. The room temperature $H_S(t_{Cr})$ and GMR(t_{Cr}) dependences of as-deposited samples are related to the oscillatory behavior of interlayer exchange coupling with the local maximum at $t_{Cr} \approx 1.1$ nm and are similar to those observed for other sputtered multilayers (see, e.g. [16]). As it was discussed before, for multilayers with a similar interface roughness, the maximum of the $F_{AF}(t_{Cr})$ dependence is observed for higher Cr layers thickness ($t_{Cr} \approx 1.5$ nm). The strong degradation of antiferromagnetic coupling (decrease of H_S, GMR, F_{AF} with D_{Ar}) for low doses of irradiated ions ($D_{Ar} \leq 2 \times 10^{13}$ Ar/cm^2) is observed only for the samples with the small thickness of spacer layers ($t_{Cr} \leq t_{Fe} = 1.4$ nm). Such a behavior can be explained as follows. As a result of irradiation, the uncorrelated interface roughness increases independently of t_{Cr} (see discussion of Fig. 1). Therefore, the probability of pinholes creation (leading to the decrease of F_{AF}) is larger for multilayers with smaller t_{Cr}. Moreover, the proportionality between GMR amplitude and F_{AF} parameter should be observed assuming that for antiferromagnetically coupled multilayers with different density of pinholes only the antiferromagnetically coupled regions contribute to the GMR effect. Such a behavior, i.e., negligible changes in the GMR/F_{AF} ratio with D_{Ar}, is also observed for studied Fe/Cr multilayers in a whole range of t_{Cr} for ion doses smaller than $\leq 2 \times 10^{13}$ Ar/cm^2 (Fig. 5d). This additionally supports our interpretation indicating the creation of pinholes 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 the higher doses ($D_{Ar} > 2 \times 10^{13}$ Ar/cm^2) indicates that another mechanism responsible for the decrease of GMR with D_{Ar} starts to dominate at $D_{Ar} > 2 \times 10^{13}$ Ar/cm^2. The higher irradiation dose D_{Ar} results in the increase of both the paramagnetic component seen in CEMS and the electrical resistance suggesting that there is some alloying besides the increase of interface roughness. Due to an increasing number of defects in the multilayer structure, the mean free path of electrons decreases reducing GMR effect. Moreover, with growing structural disorder of the spacer layers, the strength of the antiferromagnetic coupling decreases and its oscillatory behavior gradually vanishes.

4. Conclusions

The increase (independent of Cr layers thickness) of the interface roughness in Fe/Cr multilayers irradiated with 200 keV Ar ions and doses exceeding 5×10^{12} Ar/cm^2 was deduced from CEMS measurements. This results in the increase of pinholes density for multilayers with the thinner Cr spacer layers. Such a scenario is confirmed by the distinct correlation between changes in GMR effect and antiferromagnetically coupled fraction of investigated samples. For more heavily irradiated Fe/Cr multilayers, at doses exceeding 2×10^{13} Ar/cm^2, the volume intermixing is the dominating mechanism responsible for degradation of GMR and antiferromagnetic coupling.

Acknowledgements

This project was financially supported by Polish Committee for Scientific Research (KBN), grant number: PBZ/KBN-013/T08/23 and PBZ/KB/044/P03/2001.

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