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ION MODIFICATION OF THE MAGNETOTRANSPORT PROPERTIES OF Fe/Cr MULTILAYERS

M. KĄC[1,2], M. TOULEMONDE[2], J. JAWORSKI[1], J. JURASZEK[3], R. KRUK[1], S. PROTSENKO[4], V. TOKMAN[1], AND M. MARSZAŁEK[1]

[1]*The Henryk Niewodniczański Institute of Nuclear Physics PAN Radzikowskiego 152, 31-342 Kraków, Poland*

[2]*CIRIL (CEA-CNRS-ENSICAEN), BP5133, 14070 Caen Cedex 05, France*

[3]*Groupe de Physique des Matériaux, UMR6634-Université de Rouen, BP12 avenue de l'université 76801 Saint Etienne du Rouvray cedex, France*

[4]*Sumy State University, R.-Corsakova, 2, 40007 Sumy, Ukraine*

Abstract: The Cr thickness corresponding to the first antiferromagnetic maximum (1.2 nm) was chosen as a spacer in $(Fe/Cr)_{20}$ multilayers. Magnetotransport properties of as-deposited Fe/Cr multilayers measured by four probe method at room temperature shown magnetoresistance (MR) effect around 2.2%. Structural, magnetic and magnetotransport properties of Fe/Cr multilayers were modified by irradiations with 163 MeV Au ions with fluence ranging form 1×10^{11} to 5×10^{12} ions/cm^2. Conversion Electron Mössbauer Spectroscopy (CEMS) indicated increasing, with irradiation fluence, mixing at the interfaces. Irradiations resulted also in monotonic decrease of MR as a function of fluence.

1. INTRODUCTION

The magnetic properties of Fe/Cr multilayers have been a subject of intensive study due to their relevance in fundamental science and technology. The observation of antiferromagnetic coupling between two Fe layers separated by Cr spacer [1], oscillations of this coupling in magnitude (with a long ~1.2 nm and short period ~0.3 nm) as a function of the Cr spacer thickness [2-4], and giant magnetoresistance (GMR) effect [5, 6], were all first reported for Fe/Cr systems. The GMR is closely related to the antiferromagnetic exchange coupling of Fe layers through the non magnetic spacer layer. The transition from antiparallel (at remanence) to parallel alignment induced by externally applied magnetic field changes the scattering probabilities of spin-up and spin-down electrons and results in a decrease of the resistivity. The electronic transport in layered thin films is determined by combined effect of the electronic structure of the two metals (Fe, Cr in discussed samples), as well as by the microstructure of the interface [7]. Some theoretical descriptions show that interface roughness can enhance or reduce the GMR [8]. It was found experimentally that GMR increases substantially with increasing interfacial roughness [9]. However, in contrast to these results, it was also observed that increasing interfacial roughness causes GMR to decrease [10] and a very large GMR (220% at 1.5 K) has been found in Fe/Cr multilayers with smooth interfaces [11]. It is well known that interface roughness can be affected by deposition conditions of multilayer structure [9] or by post-deposition treatments like thermal annealing [12, 13] and ion irradiation [14, 15]. Ion irradiation may lead to the increase of the GMR [14] as well as to the degradation of the GMR [10] depending on the applied ions and fluences.

In this paper, we present study of the structural and magnetotransport properties of Fe/Cr multilayers irradiated at room temperature with 163 MeV Au ions.

2. EXPERIMENT

Fe/Cr/Fe trilayers and Fe/Cr multilayers were prepared by thermal evaporation in ultra high vacuum system on the silicon Si(100) substrate at room temperature. Before evaporation the substrate was annealed during 1 hour at 700 K. The chemical purity of the silicon surface was checked by Auger Electron Spectroscopy (AES). All layers were deposited on the 10 nm Cr buffer layer and covered by 5 nm of Cr protective layer. Thickness of the layers and evaporation rate were controlled during evaporation process by quartz balance. The Fe layer thickness varied between 0.5 and 2 nm while Cr layer thickness changed between 1-2.3 nm, the evaporation rate was about 0.6 nm/min for Fe layers and about 1 nm/min for Cr layers. The base pressure during evaporation was about 10^6 Pa. Irradiation experiments were performed at the Vivitron in IReS in Strasbourg with the initial energy 200 MeV Au^{15+}. The ion beam went through a 2 micrometer foil in order to be at an equilibrium charge state for energy of 163 MeV (0.82 MeV/u). The maximum flux used in the experiment was equal to 5×10^8 ions/(cm^2s) and the fluence changed from 1×10^{11} to 5×10^{12} ions/cm^2. The structure of the Fe/Cr interfaces was observed by Conversion Electron Mössbauer Spectroscopy (CEMS) measured at room temperature with Co(Rh) source. The spectra were analyzed numerically by fitting a hyperfine distribution using Voigt-based method of Rancourt and Ping [16]. Hysteresis loops were measured by SQUID. Magnetoresistance was measured at room temperature using standard four probe technique in transversal geometry in an external field up to 1 Tesla.

3. RESULTS AND DISCUSSION

In the first stage of the investigations the Fe/Cr/Fe trilayer systems were prepared to determine the Cr spacer thickness corresponding to antiferromagnetic coupling. The magnetic coupling between adjacent Fe layers through the Cr spacer layer was measured as a function of the Cr thickness changing from 1 to 2.3 nm. Magnetic measurements showed that the magnetization was aligned in the plane of the Fe layers. It was confirmed by CEMS spectra in which line intensity ratio (the intensity of line 2 with respect to line 3) was close to 4. Antiferromagnetic coupling characterized by remanence-less curve was found for Cr layer thickness of 1.2 nm (Fig. 1a). It is a typical value which corresponds to the first antiferromagnetic peak in the interlayer exchange coupling of Fe/Cr system [2]. For (2 nm Fe/1.2 nm Cr/2 nm Fe) sample in magnetic fields $H > H_2$ the Fe magnetic moments are aligned parallel to the external field and the magnetization is saturated (Fig. 1a). For $H_1 < H < H_2$ the Fe magnetic moments are noncollinear and they deviate from the external field direction. The antiferromagnetic coupling turns on for fields $H < H_1$. For the 1.3 and 1.4 nm of Cr thickness ferromagnetic coupling was dominating as shown in the Fig. 1b (example for 2 nm Fe/1.4 nm Cr/2 nm Fe). The sample with the Cr thickness of 1.5 nm showed large fraction of antiferromagnetic coupling (Fig. 1c). The system demonstrated again ferromagnetic coupling for thicker Cr layers (1.6, 1.9 and 2.3 nm) as shown in Fig. 1d for 1.6 nm of Cr.

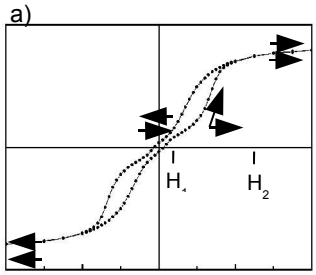
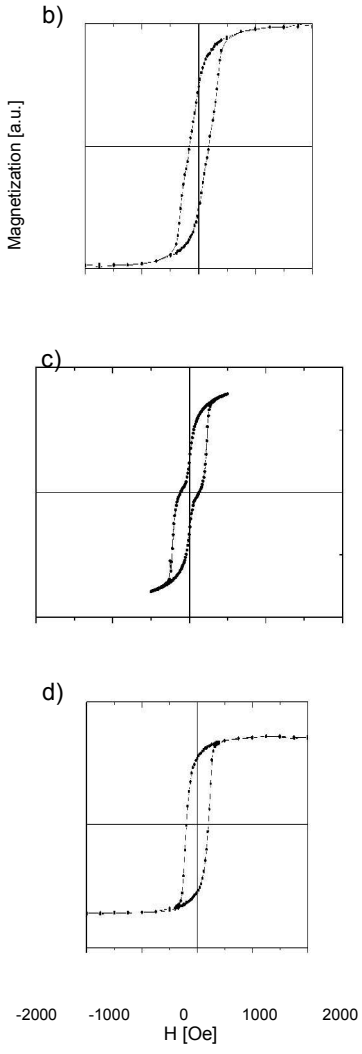


Fig. 1. The hysteresis loops measured by SQUID for (2 nm Fe/x nm Cr/2 nm Fe) trilyers where: a) x = 1.2, b) x = 1.4, c) x = 1.5, d) x = 1.6 nm. The arrows indicate the magnetization direction in adjacent Fe layers



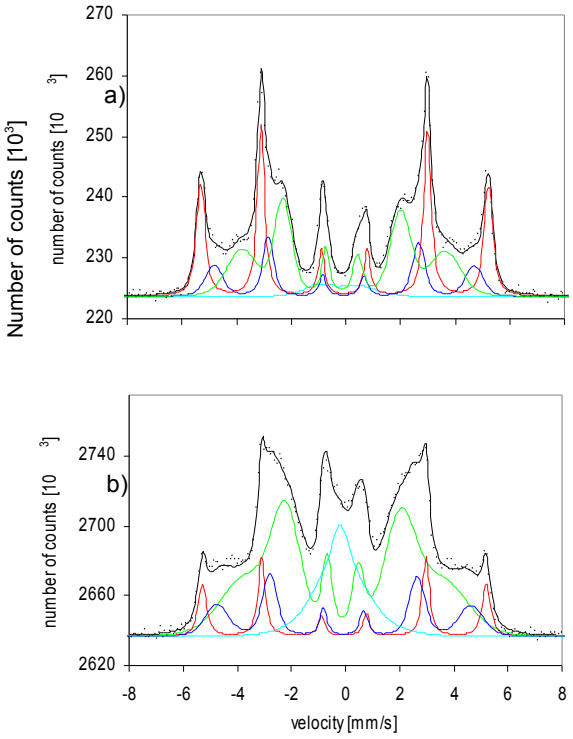


Fig. 2. CEMS spectra measured for (2 nmFe/1.2 nmCr)$_{20}$ multilayers: a) as deposited, and b) irradiated with 163 MeV Au ions at fluence 5 × 10^{12} ions/cm^2. The solid lines are simulated fits to the data

Unexpected large fraction of antiferromagnetic coupling observed for Fe/1.5 nm Cr/Fe may be interpreted as a presence of short period oscillation provided that very smooth Fe layers were obtained in deposition process. The structure of a single Fe layer of 2 nm thickness was observed by CEMS measured for the Cr/2 nm Fe^{57}/Cr trilayer system. The spectra were interpreted in terms of model which correlates four components characterized by hyperfine parameters with the different sites in Fe/Cr system. The component with the largest value of hyperfine magnetic field (~33 T) corresponds to Fe atoms in bulk sites. The components equal to 30 T and 24 T correspond to Fe atoms at interface with Cr atoms in second, and in both first and second coordination shells, respectively. Small field component (~7 T) is attributed to isolated Fe atoms which diffused into Cr matrix. For all spectral components the isomer shift parameters changed in the same way as the hyperfine magnetic fields, quadrupole splitting parameters were negligible beside of a small field component for which small value was measured. The detail analysis of 2 nm Fe layer sandwiched between Cr shows very large fraction of bulk-like component (63%), small fractions of 30 T and 24 T equal to 16% and 18%, respectively and a very small fraction of the component ~7 T equal to 3%. For ideally smooth interfaces only 3 components (33 T, 30 T and 24 T) should be observed in the assumed model, with fraction equal to 70%, 15% and 15%, respectively. The results obtained for described sample are in good agreement with above-mentioned values.

Presented analysis was performed to find the Cr spacer thickness for which in our experimental conditions antiferromagnetic coupling is observed. The further investigations were continued for Fe/Cr multilayers with Cr layer thickness of 1.2 nm chosen as the Cr spacer thickness. The interface structure was observed by CEMS separately for Fe-on-Cr (lower) and Cr-on-Fe (upper) interfaces. It was possible by introducing 0.5 nm ^{57}Fe probe layer at all lower (0.5 nm ^{57}Fe/1.5 nm Fe^{nat}/1.2 nm $Cr)_{20}$ and at all upper (1.5 nm Fe^{nat}/0.5 nm ^{57}Fe/1.2 nm $Cr)_{20}$ Fe interfaces. In the Figure 2a CEMS spectrum for the multilayers with Cr-on-Fe (upper) interface is presented. The analysis of the component fractions showed that these interfaces are slightly rougher as compared with Fe-on-Cr (lower) interfaces. Such an asymetry was postulated previously from CEMS experiment [17, 18] and also from STM studies [19, 20] and it was explained by binding energy between substrate and ad-atom material, which is proportional to the melting point of solids. The melting point of Cr is higher than melting point of Fe and, therefore, interface mixing for Fe-on-Cr is suppressed and this interface is smoother [21].

The Fe/Cr multilayers were modified by irradiation with 163 MeV Au ions. The electronic stopping power for Au ions in Fe target equal to $(dE/dx)_e$ = 39.7 keV/nm [TRIM-95] is closed to the value of electronic stopping power for Sn ions which were successfully used by Teillet *et al*. [22] in Fe/Tb irradiation, resulting in the smoothening of the interface structure. The samples were irradiated with swift heavy ions where interfacial modifications are produced *via* electronic stopping power $(dE/dx)_e$. CEMS spectrum measured for the same sample as in Fig. 2a but after Au irradiation with the fluence equal to 5×10^{12} ions/cm^2 is presented in the Fig. 2b. Large increase of small field component fractions (24 T, 7 T) related to roughening of the interface and diffusion of Fe atoms into Cr layer is seen. These changes evidence huge mixing at the interfaces. Similar changes were observed in the samples with ^{57}Fe probe layer

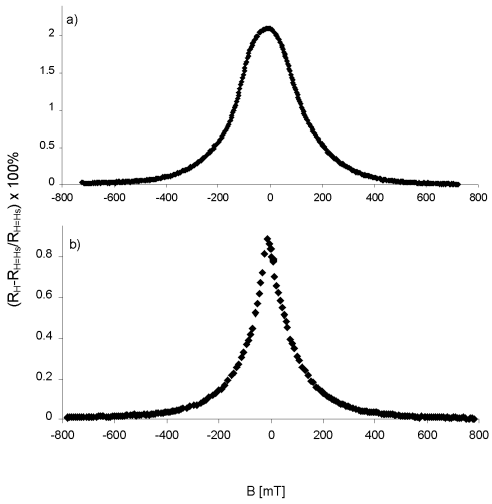


Fig. 3. Magnetoresistance *versus* applied magnetic field measured at room temperature for (2 nm Fe/1.2 nm Cr)$_{20}$ multilayers: a) as deposited, and b) irradiated with 163 MeV Au ions at fluence 4×10^{11} ions/cm^2

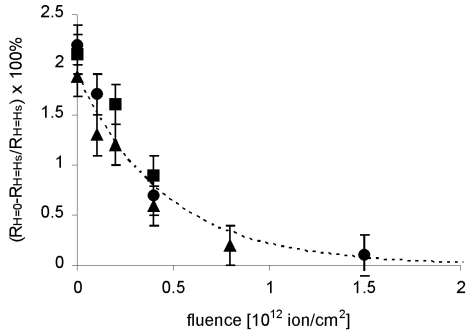


Fig. 4. MR of the as deposited and irradiated (163 MeV Au ions) multilayers of (2 nm Fe/1.2 nm Cr)$_{20}$ as a function of fluence. The results are presented for three identical samples. The broken line is a guide the eye

at lower interfaces. For all samples the fractions of small field components increased as a function of fluence. It means that after irradiations all interfaces became rougher than originally what could enhance GMR provided that an appreciable portion of the Fe layers is coupled antiferromagneticaly [7]. The GMR value was defined by: GMR = $(R_{H=0}\ R_{H=Hs}/R_{H=Hs}) \times 100$, where $R_{H=0}$ is the resistance measured at remanence and $R_{H=Hs}$ is the resistance measured at saturation field. Magnetoresistance effect measured for Fe/Cr multilayers at room temperature before irradiation (the typical curve is presented in the Fig. 3a) was around 2.2%. All irradiated samples demonstrated smaller values of MR than as-deposited. Magnetoresistance curves after irradiation became more pointed and narrow but were saturated nearly at the same field as the nonirradiated samples (Fig. 3). These changes can be attributed to decrease of antiferomagnetic coupling fraction. Figure 3b shows the MR of the sample from Fig. 3a irradiated with the fluence of 4×10^{11} ions/cm^2. The magnetoresistance decreased monotonically with irradiation fluence (Fig. 4), the maximum change of MR being as large as 95% at an irradiation fluence of 1.5×10^{12} ions/cm^2. The similar results were observed by Paul *et al*. [10] in Fe/Cr multilayers irradiated with 200 MeV Ag ions. The qualitatively different results were obtained by Korniewski [7] in earlier study of 500 keV Xe ion irradiation on Fe/Cr multilayers, where GMR was found to increase with fluence up to 1×10^{13} ions/cm^2 followed by a decrease for still higher irradiation fluences. From the literature it is known that the electronic stopping power threshold determined for high energy beam (around 20 MeV/u) for damage creation in bulk Fe is ~50 keV/nm [23]. Using the experimental results obtained at high energy beam and their description by the inelastic thermal spike model, one can calculate the threshold of defect creation in iron irradiated with energy beam of 0.8 MeV/u. This

threshold value decreases significantly reaching ~30 keV/nm. The $(dE/dx)_e$ = 39.7 keV/nm is clearly above the threshold what explains large mixing at the interfaces. These changes likely induced gradual loss of antiferromagnetic coupling and degradation of GMR effect. Further experiments considering magnetic properties of Fe/Cr multilayers are in progress.

4. CONCLUSIONS

Effect of 163 MeV Au ion irradiation on Fe/Cr multilayers has been studied. The electronic energy loss results in modifications of the interfaces. The magnetoresistance decreases monotonically with irradiation fluence, the maximum decrease in MR being as large as 95% at the fluence of 1.5×10^{12} ions/cm^2. Such large change in MR with irradiation may be the effect of the mixing at the interfaces and disappearance of antiferromagnetic coupling.

Acknowledgements

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References

[1] P. Grünberg, R. Schreiber, Y. Pang, M. B. Brodsky, and Sowers, Phys. Rev. Lett. **57**, 2442 (1986) .
[2] S. S. P. Parkin, N. More, and K. P. Roche, Phys. Rev. Lett. **64**, 2304 (1990).
[3] J. Unguris, R. J. Celotta, and D. T. Pierce, Phys. Rev. Lett **67**, 140 (1991).
[4] S. T. Purcell, W. Folkerts, M. T. Johnson, N. W. E. McGee, K. Jager, J. ann de Stegge, W. B. Zeper, W. Hoving, and P. Grünberg, Phys. Rev. Lett. **67**, 903 (1991).
[5] M. N. Baibich, J. M. Broto, A. Fert, F. Nguyen Van Dau, F. Petroff, P. Etienne, G. Creuzet, A. Friederich, and J. Chazelas, Phys. Rev. Lett. **61**, 2472 (1988).
[6] G. Binasch, P. Grünberg, F. Saurenbach, and W. Zinn, Phys. Rev. B **39**, 4828 (1989).
[7] V. Korenivski, K. V. Rao, David M. Kelly, Ivan K. Schuller, Kim K. Larsen, and J. Bottiger, J. Magn. Magn. Mater. **140-144**, 549 (1995).
[8] J. Barnaś and Y. Bruyenseraede, Europhys. Lett. **32**, 167 (1995).
[9] E. Fullerton, D. M. Kelly, J. Guimpel, I. Schuller, and Y. Bruynseraede, Phys. Rev. Lett. **68**, 859 (1992).
[10] A. Paul, A. Gupta, S. M. Chaudhari, and D. M. Phase, Vacuum **60**, 401 (2001).
[11] R. Schad, C. D. Potter, P. Belien, G. Verbanck, V. V. Moshchalkov, and Y. Bruynseraede, Appl. Phys. Lett. **64**, 3500 (1994).
[12] J. M. Colino, I. K. Schuller, V. Kornievski, and K. V. Rao, Phys. Rev. B **54** (1996) 13030.
[13] M. Kopcewicz, T. Luciński, F. Stobiecki, and G. Reiss, J. Appl. Phys. **85** (1999) 5039.
[14] D. M. Kelly, I. K. Schuller, V. Kornievski, K. V. Rao, K. K. Larsen, J. Bottiger, E. M. Gyorgy, and R. B. van Dover, Phys. Rev. B **50**, 3481 (1994).
[15] M. Kopcewicz, F. Stobiecki, J. Jagielski, B. Szamański, M. Schmidt, J. Dubowik, and J. Kalinowska, J. Appl. Phys. **93** 5514 (2003).
[16] D. G. Rancourt and J. Y. Ping, Nucl. Instr. and Meth. B **28**, 85 (1991).
[17] T. Shinjo and W. Keune, J. Magn. Magn. Mater. **200**, 598 (1999).
[18] M. Kubik, T. Ślęzak, M. Przybylski, W. Karaś, and J. Korecki, Vacuum **63**, 337 (2001).
[19] A. Davies, A. Joseph Stroscio, D. T. Pierce, and R. J. Celotta, Phys. Rev. Lett. **76**, 4175 (1996).
[20] Y. J. Choi, I. C. Jeong, J-Y Park, S-J Kahng, J. Lee, and Y. Kuk, Phys. Rev. B **59**, 10918 (1999).
[21] V. M. Uzdin, W. Keune, H. Schrör, and M. Walterfang, Phys. Rev. B **63**, 104407 (2001).
[22] J. Teillet, F. Richomme, A. Fnidiki, and M. Toulemonde, Phys. Rev. B **55**, 11560 (1997).
[23] A. Dunlop, D. Lesuer, P. Legrand, H. Dammak, and J. Dural, Nucl. Instr. and Meth. B **90**, 330 (1994).