Influence of the Ar-ion irradiation on the giant magnetoresistance in Fe/Cr multilayers

M. Kopcewicz[a)]
Institute of Electronic Materials Technology, 01-919 Warsaw, Poland

F. Stobiecki
Institute of Molecular Physics, Polish Academy of Sciences, 60-179 Poznań, Poland

J. Jagielski[b)]
Institute of Electronic Materials Technology, 01-919 Warsaw, Poland

B. Szymański, M. Schmidt, and J. Dubowik
Institute of Molecular Physics, Polish Academy of Sciences, 60-179 Poznań, Poland

J. Kalinowska
Institute of Electronic Materials Technology, 01-919 Warsaw, Poland

(Received 14 August 2002; accepted 17 January 2003)

The influence of 200 keV Ar-ion irradiation on the interlayer coupling in Fe/Cr multilayers exhibiting the giant magnetoresistance (GMR) effect is studied by the conversion electron Mössbauer spectroscopy (CEMS), vibrating sample magnetometer hysteresis loops, magnetoresistivity, and electric resistivity measurements and supplemented by the small-angle x-ray diffraction. The increase of Ar-ion dose causes an increase of interface roughness, as evidenced by the increase of the Fe step sites detected by CEMS. The modification of microstructure induces changes in magnetization reversal indicating a gradual loss of antiferromagnetic (AF) coupling correlated with the degradation of the GMR effect. Distinctly weaker degradation of AF coupling and the GMR effect observed for irradiated samples with a thicker Cr layer thickness suggest that observed effects are caused by pinholes creation. The measurements of temperature dependence of remanence magnetization confirm increase of pinhole density and sizes during implantation. Other effects which can influence spin dependent contribution to the resistance, such as interface roughness as well as shortening of mean-free path of conduction electrons, are also discussed.
 [DOI: 10.1063/1.1559640]

I. INTRODUCTION

The giant magnetoresistance (GMR) effect was discovered in Fe/Cr structures more then a decade ago[1] and the theoretical explanation of this phenomenon is now well established.[2] However, despite many studies, the influence of the interface quality in this structure on the GMR effect seems to still be puzzling and not well understood. Some theoretical descriptions show that the interface roughness can enhance or reduce the GMR generated by spin dependent scattering on structural defects inside the ferromagnetic layer.[3] It was found experimentally that the increase of interface roughness may enhance the GMR.[4] However, in contrast to these results, a very large GMR has been found in Fe/Cr multilayers (MLs) with sharp interfaces and low residual resistivity.[5] It is well known that the interface roughness can be affected by deposition conditions of multilayer structure, substrate temperature, and thermal annealing (see, e.g., Refs. 6 and 7). Moreover, it was shown recently that ion irradiation may lead to the increase of the GMR as well as to the degradation of the GMR depending on ion dose. Irradiation with 500 keV Xe ions[8,9] induced the initial increase in the GMR, however, at higher ion doses, destroyed the GMR. Also, the 200 MeV Ag-ion irradiation led to the decrease of the GMR in Fe/Cr MLs.[10] The observed reduction of the GMR and the loss of antiferromagnetic (AF) coupling were interpreted in terms of the creation of ferromagnetic pinholes, however, with no clear evidence.

We have studied the modification of the interface structure by conversion electron Mössbauer spectroscopy (CEMS) and small-angle x-ray diffraction (SAXRD) measurements, induced by Ar-ion irradiation, in an Fe/Cr system and its influence on the magnetization reversal and the GMR effect.

In particular, our studies should give answers on the following important questions:

(a) Are the pinholes created during ion irradiation?
(b) Is the degradation of the GMR and AF coupling caused mainly by pinhole creation?
(c) Are the changes in ML structure detectable by CEMS or SAXRD measurements?

According to the model proposed by Fulghum and Camley,[11] the existence of pinholes in as-deposited samples and the increase of their density and size during implantation

a)Electronic mail: kopcew_m@sp.itme.edu.pl
b)Also at: The Andrzej Soltan Institute of Nuclear Studies, 05-400 Świerk/Otwock, Poland.

can be confirmed by temperature measurements of remanence magnetization. The influence of Cr layer thickness (t_{Cr}) on the degradation of the GMR and AF coupling in Fe/Cr MLs subjected to ion irradiation is discussed. The increasing resistance of the GMR effect to ion irradiation in Fe/Cr MLs with increasing t_{Cr} as well as correlation between changes in the GMR and antiferromagnetically coupled fraction suggest that the main effect responsible for decrease of the GMR is caused by pinhole creation. The influence of shortening of the mean-free path (MFP) of electrons and the increase of interface roughness (caused by ion irradiation) on spin dependent scattering is discussed for structures with different Fe layer thicknesses (t_{Fe}).

II. EXPERIMENT

The Fe 3 nm/Cr 1.1 nm, Fe 1.4 nm/Cr 1.03 nm, and Fe 1.4 nm/Cr 1.5 nm MLs were deposited on SiO_x substrates using UHV magnetron sputtering (dc and rf for Fe and Cr, respectively). The modulation wavelengths ($\lambda = t_{Fe} + t_{Cr}$) and thickness of iron and chromium layers were controlled by SAXRD and x-ray fluorescence, respectively. The deposition rate was 0.04 nm/s for both materials. The total thickness of the Fe/Cr film was about 100 nm. Samples were irradiated at room temperature (RT) with 200 keV Ar ions with doses (D_{Ar}) ranging from 1×10^{12} to 5×10^{14} Ar/cm^2 for Fe 3 nm/Cr 1.1 nm and in the range of 5×10^{12}–2×10^{13} for two other samples. The range of ions matched the thickness of the multilayer film well. The as-deposited and irradiated samples were characterized at RT by CEMS, SAXRD, and vibrating sample magnetometer (VSM) hysteresis loops. Magnetoresistance and resistivity were measured at RT using the four-probe technique in current-in-plane (CIP) geometry. The GMR(H) dependencies were determined as $\mathrm{GMR}(H) = 100\times[R(H) - R(H=2T)]/R(H=2T)$ (H denotes the magnetic field), the maximal value of GMR(H) determine the GMR amplitude. The temperature dependence of remanence magnetization was determined from hysteresis loops measured by VSM in an N_2 atmosphere for temperatures ranging from 230 to 470 K.

III. RESULTS AND DISCUSSION

The CEMS spectra were interpreted in terms of the model[12,13] which correlates four magnetically split spectral components with the Fe sites in the Fe/Cr system: $H_1 \approx 33T$, corresponds to the bulk Fe sites; $H_2 \approx 30T$ and $H_3 \approx 24T$, correspond to the "step" sites at the Fe/Cr interfaces, and $H_4 \approx 20T$, corresponds either to the "perfect" interface sites or to some other step positions. The origin of the H_4 component is somewhat controversial. It was suggested recently[14] that this component corresponds to the Fe sites in Cr layer separated by at least two atomic Cr monolayers from the Fe layer. However, it was shown earlier that Fe atoms isolated in Cr layer are paramagnetic.[15]

For ideally smooth interfaces, the expected relative fraction of H_1, (H_4) components should be 87% (13%) and 71% (29%) for $t_{Fe} = 3$ nm and 1.4 nm, respectively, and the spectral contribution corresponding to H_2 and H_3 components should be zero.[15] In fact, even for as-deposited samples, the measured fractions of H_1 and H_4 are significantly smaller than those predicted for ideal interfaces and H_2, H_3 fractions show nonzero values (see Figs. 1 and 2). The simple calculation indicates that for the experimental value of the relative fraction of the H_1 component equal to 73% for as-deposited Fe 3 nm/Cr 1.1 nm MLs, only 2.2 nm correspond to bulk Fe sites and 0.4 nm to Fe at each interface. For both samples with $t_{Fe} = 1.4$ nm, the fitted value of

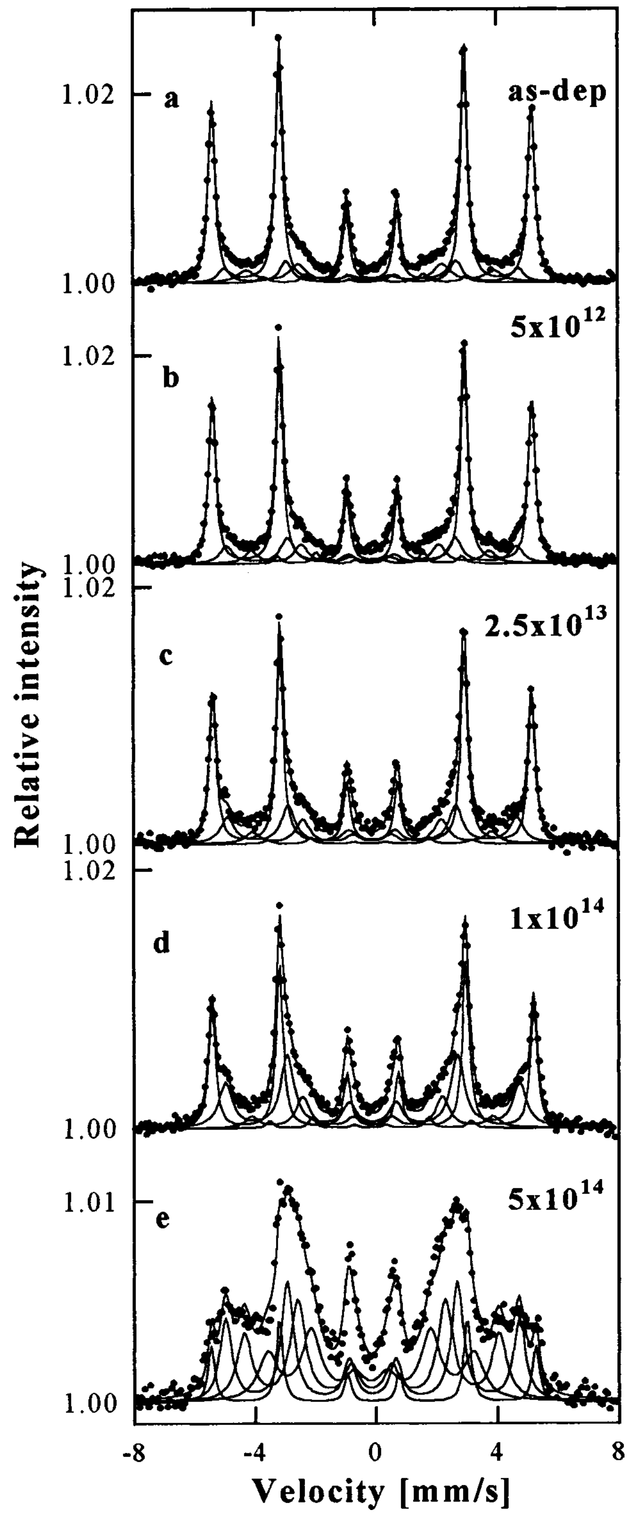

FIG. 1. CEMS spectra for as-deposited (a) and irradiated Fe 3 nm/Cr 1.1 nm MLs at indicated ion doses (b)–(e).

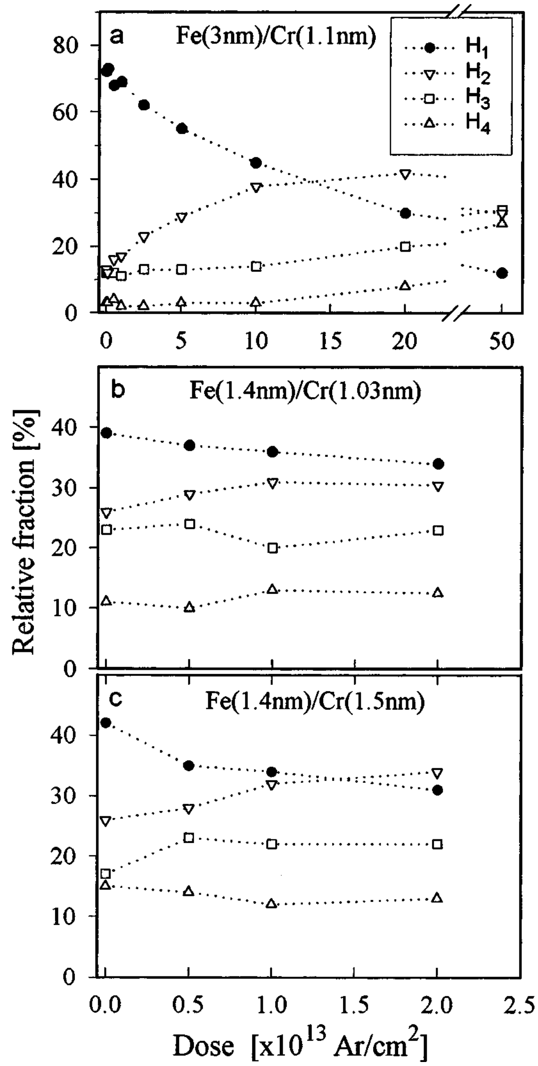

FIG. 2. Relative spectral fractions of H_1–H_4 components vs ion dose for Fe 3 nm/Cr 1.1 nm (a), Fe 1.4 nm/Cr 1.03 nm (b), and Fe 1.4 nm/Cr 1.5 nm (c).

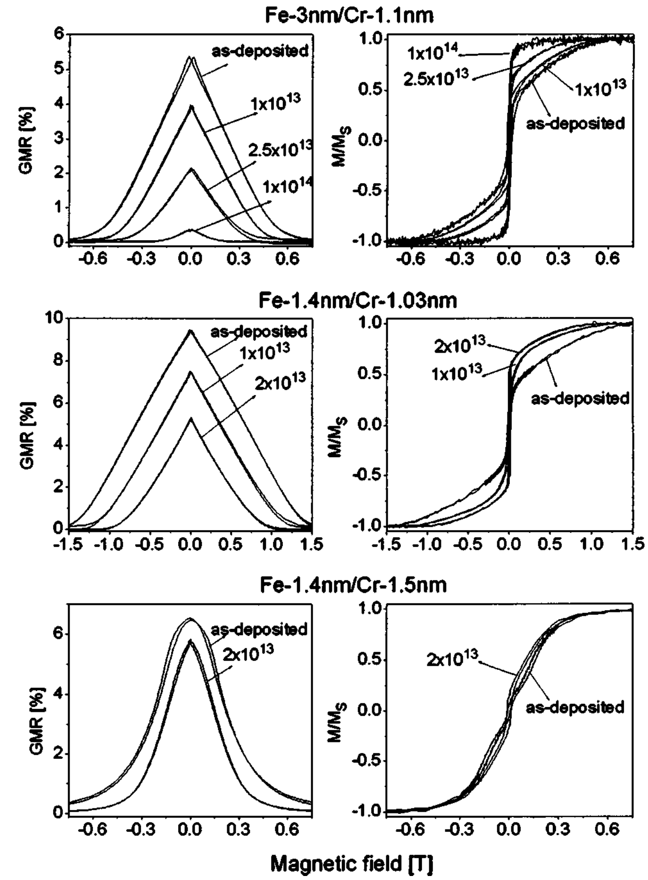

FIG. 3. Examples of the GMR(H) dependencies and hysteresis loops for as-deposited and irradiated Fe/Cr multilayers at indicated ion doses.

nominal thickness of Fe at the interface sites is also 0.4 nm. This indicates that the interface roughness is nearly the same for all as-deposited Fe/Cr MLs independently of t_{Cr} and t_{Fe}. For MLs with small thicknesses of spacer layers and with an uncorrelated interface roughness caused by grain-boundary diffusion during deposition process, there is a certain probability of creating ferromagnetic bridges (pinholes) across Cr layers. The existence of pinholes in antiferromagnetically coupled MLS leads to a strong ferromagnetic coupling localized in the vicinity of pinholes.[11,16–18] As a result, the antiferromagnetically coupled fraction, F_{AF}, (defined as $F_{AF} = 1\text{-}M_R/M_S$, where M_R and M_S are the remanence and saturation magnetizations determined from hysteresis loops) is smaller than one. Such a behavior, i.e., small F_{AF} for thin Cr layers ($F_{AF}=0.65$ and 0.79 for Fe 3 nm/Cr 1.1 nm and Fe 1.4 nm/Cr 1.03 nm MLs, respectively) and nearly perfect AF coupling for thicker spacer layers ($F_{AF}=0.95$ for Fe 1.4 nm/Cr 1.5 nm) are observed in our as-deposited samples. It should be noticed that the smaller value of F_{AF} for Fe 3 nm/Cr 1.1 nm MLs than for Fe 1.4 nm/Cr 1.03 nm seems to be related to the difference in t_{Fe} (difference in intralayer coupling) rather than to the different densities of pinholes. According to the model proposed in Ref. 11 for a given density of pinholes in antiferromagneticaly coupled MLs, the value of M_R increases with increasing thickness of ferromagnetic layers. The origin of imperfect AF coupling observed in our as-deposited samples with a small t_{Cr} was examined independently by measurements of the temperature dependence of M_R (see next).

Ar-ion irradiation induced clear changes in the CEMS spectra of irradiated samples. Already at low ion doses $5\times 10^{12} \leqslant D_{Ar} \leqslant 2.5\times 10^{13}$ Ar/cm^2 (but sufficiently high to induce distinct changes in magnetic properties and, therefore, most interesting for our study of MLs with various Cr thicknesses [Figs. 2(b) and 2(c)]), the spectral contribution of the H_2 component increases substantially at the expense of the H_1 component in a similar way for all investigated samples [Figs. 1(a)–1(e), and 2]. This suggests that the interface roughness increases (the number of Fe step sites increases) independently of t_{Cr}. However, in the SAXRD spectra, no changes caused by ion irradiation are detected. Simultaneously, for MLs with a thin Cr layer, i.e., for Fe 3 nm/Cr 1.1 nm and for Fe 1.4 nm/Cr 1.03 nm, distinct changes in the shape of the hysteresis loop are observed in contrast to Fe 1.4 nm/Cr 1.5 nm. As the remanence value increases (F_{AF} decreases), the saturation field (H_S) decreases, and the hysteresis loop becomes more rectangular (Fig. 3). Similar changes were observed for Fe 3 nm/Cr 1.2 nm MLs irradi-

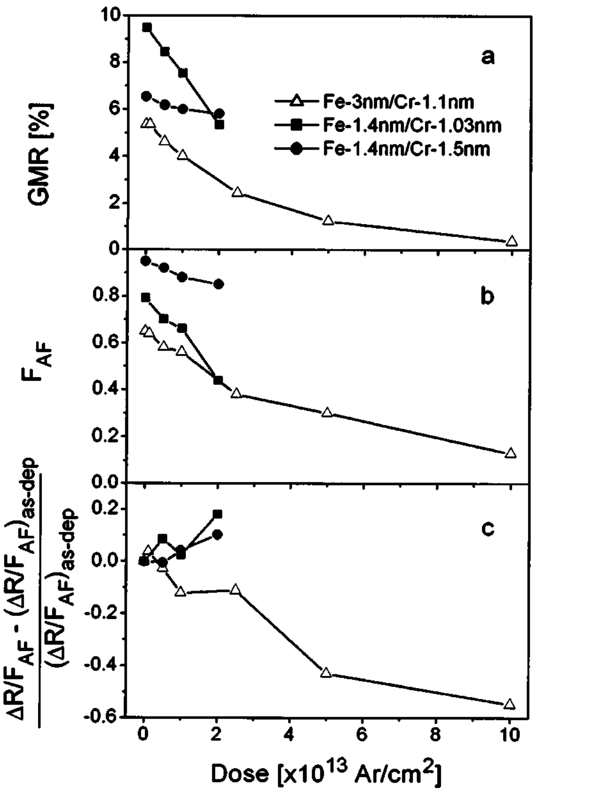

FIG. 4. GMR value (a), antiferromagnetically coupled fraction, F_{AF}, (b), and relative changes of $\Delta R/F_{AF}$ (c), vs ion dose for Fe 3 nm/Cr 1.1 nm (Δ), Fe 1.4 nm/Cr 1.03 nm (■), and Fe 1.4 nm/Cr 1.5 nm (●) MLs.

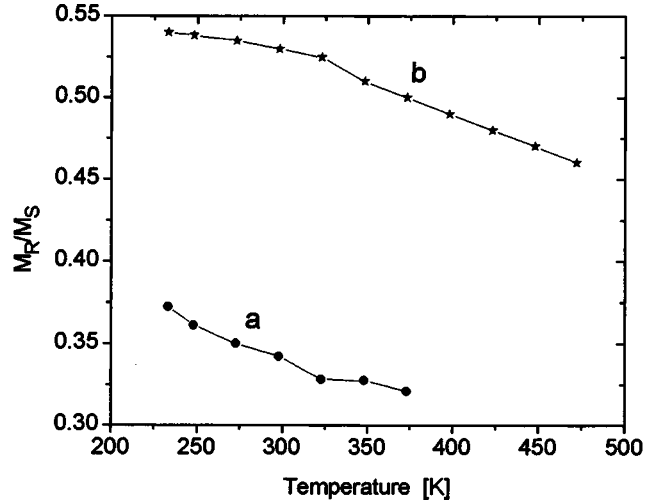

FIG. 5. Normalized remanence magnetization as a function of temperature for Fe 3 nm/Cr 1.1 nm MLs in as-deposited state (a) and for sample irradiated with 5×10^{13} Ar/cm^2 (b).

ated with Xe ions.[8,9] The electrical transport measurements reveal almost the same monotonic increase of electrical resistance (determined for magnetic field $H=2T$) with D_{Ar} for all our samples (for $D_{Ar}=2\times10^{13}$ Ar/cm^2 in Fe 3 nm/Cr 1.1 nm MLs resistance is about 13% higher than for the as-deposited sample). However, the degradation of the GMR is much stronger for MLs with smaller t_{Cr} (Fig. 3). The GMR and F_{AF} dependencies on D_{Ar} are plotted in Fig. 4. In order to separate the changes in the spin dependent contribution to the resistance from the loss of AF coupling[6] in Fig. 4, the relative changes of $\Delta R/F_{AF}(D_{Ar})$ [ΔR is the difference between the largest and the lowest values of resistance determined from $R(H)$ dependence] are also shown.

From CEMS measurements, we have concluded that for all investigated samples the interface roughness increases in the same way during irradiation. On the other hand, according to the results presented by Paul,[10] we can conclude that the ion irradiation increases the uncorrelated part of the interface roughness. Thus, the probability of pinhole creation in Fe/Cr MLs subjected to ion irradiation should be larger for MLs with a smaller thickness of the Cr layer. This explains a strong degradation of F_{AF} and the GMR with ion dose observed for Fe 3 nm/Cr 1.1 nm and Fe 1.4 nm/Cr 1.03 nm MLs. The relatively weak changes observed for the $\Delta R/F_{AF}(D_{Ar})$ dependencies (Fig. 4) indicate that decrease of the GMR with ion dose is mainly caused by the progressive loss of AF coupling correlated with the creation of pinholes. The other factors that can also influence the GMR effect are the changes in the MFP of conduction electrons and interface roughness. The increase of electric resistance (caused by ion-beam induced alloying at the interfaces) indicates a reduction of MFP of electrons and consequently, a reduction of the spin dependent scattering. A small increase of $\Delta R/F_{AF}(D_{Ar})$ observed for Fe 1.4 nm/Cr 1.03 nm and Fe 1.4 nm/Cr 1.5 nm is probably caused by the interface roughness enhancement induced by ion irradiation.[8,9] Which effect dominates depends, of course, on the multilayer structure. Thus, observed only in the structures with small modulation wavelengths ($\lambda=t_{Cr}+t_{Fe}$), the increase of $\Delta R/F_{AF}(D_{Ar})$ seems to be reasonable since, for such structures, electron spins are conserved traversing a large number of ferromagnetic layers.

For higher ion doses ($D_{Ar}>2\times10^{13}$ Ar/cm^2), the investigations performed for Fe 3 nm/Cr 1.1 nm MLs show that at doses exceeding 1×10^{14} Ar/cm^2, the AF coupling and GMR vanish.

Despite the fact that the creation of pinholes during ion irradiation seems to be well documented in the aforementioned results, we have performed an additional experiment which confirms this finding. Due to the small cross section area of pinholes, the size effects, typical of low dimensional magnetic entities, become important leading to a strong reduction of the local ferromagnetic coupling via the thermal fluctuations of magnetic moments. As a consequence, with the increasing density of pinholes and their cross section area, both the low-temperature value of M_R and the temperature at which ferromagnetic coupling vanishes increase.[11,18] Such a characteristic behavior can be seen in Fig. 5 for the Fe 3 nm/Cr 1.1 nm MLs indicating that the main cause responsible for the degradation of F_{AF} and the GMR effect is related to the increase of pinhole densities and their sizes during ion-beam mixing.

The ion irradiation induced microstructural modification of investigated Fe/Cr multilayers are responsible for observed changes in magnetization reversal, $M_R(T)$ dependencies, GMR effect, and CEMS spectra, which are, however, hardly detectable by SAXRD. The poor sensitivity of the SAXRD method seems to be obvious taking into account a small contrast between Fe and Cr in their refractive indices as well as the fact that a very small density of pinholes can completely destroy AF coupling.[11]

IV. CONCLUSIONS

We have demonstrated that the increase of interface roughness of Fe/Cr MLs caused by irradiation with 200 keV Ar ions and doses exceeding 5×10^{12} Ar/cm^2 is clearly seen in CEMS measurements, however, the SAXRD technique, even at higher ion doses, hardly detects such changes in the microstructure. The subtle modification in microstructure induces distinct changes in magnetization reversal (increase of remanence magnetization) and a strong decrease of the GMR effect with increasing irradiation dose in particular for samples with small thicknesses of Cr layers. The increasing immunity of the GMR effect to ion irradiation with an increasing thickness of Cr layers, as well as the correlation between changes in the GMR and antiferromagnetically coupled fraction, suggest that the main effect responsible for the decrease of the GMR is caused by pinhole creation. The characteristic changes in the temperature dependence of remanence magnetization measured for the as-deposited and irradiated samples confirm the increase of pinhole densities and sizes during the irradiation process.

ACKNOWLEDGMENTS

The authors are grateful to T. Nowak for technical support. This project was financially supported by the NEDO grant (Japan) ''Nanopatterned Magnets,'' and by the Polish Committee for Scientific Research (KBN) Grant No. PBZ/KBN-013/T08/23.

[1] M. N. Baibich, J. M. Broto, A. Fert, F. Nguyen Van Dau, F. Petroff, P. Eitenne, G. Creuzet, A. Friederych, and J. Chazelas, Phys. Rev. Lett. **61**, 2462 (1988).
[2] R. E. Camley and J. Barnaś, Phys. Rev. Lett. **63**, 664 (1989); see also J. Barnaś, A. Fuss, R. E. Camley, P. Grünberg, and W. Zinn, Phys. Rev. B **42**, 8110 (1990).
[3] J. Barnaś and Y. Bruyenseraede, Europhys. Lett. **32**, 167 (1995).
[4] E. Fullerton, D. M. Kelly, J. Guimpel, I. Schuller, and Y. Bruynseraede, Phys. Rev. Lett. **68**, 859 (1992).
[5] R. Schad, C. D. Potter, P. Belien, G. Verbanck, V. V. Moshchalkov, and Y. Bruynseraede, Appl. Phys. Lett. **64**, 3500 (1994).
[6] J. M. Colino, I. K. Schuller, V. Kornievski, and K. V. Rao, Phys. Rev. B **54**, 13030 (1996).
[7] M. Kopcewicz, T. Lucińsnki, F. Stobiecki, and G. Reiss, J. Appl. Phys. **85**, 5039 (1999).
[8] D. M. Kelly, I. Schuller, V. Korenivski, K. V. Rao, K. K. Larsen, J. Bøttiger, E. M. Gyorgy, and R. B. van Dover, Phys. Rev. B **50**, 3481 (1994).
[9] V. Korenivski, K. V. Rao, D. M. Kelly, I. K. Schuleller, K. K. Larsen, and J. Bøttiger, J. Magn. Magn. Mater. **140**, 549 (1995).
[10] A. Paul, A. Gupta, S. M. Chaudhari, and D. M. Phase, Vacuum **60**, 401 (2001).
[11] D. B. Fulghum and R. E. Camley, Phys. Rev. B **52**, 13436 (1995).
[12] J. Landes, C. Sauer, R. A. Brand, W. Zinn, S. Mantl, and Z. Kajcsos, J. Magn. Magn. Mater. **86**, 71 (1990).
[13] F. Klinkhammer, C. Sauer, E. Y. Tsymbal, S. Handschuh, Q. Leng, and W. Zinn, J. Magn. Magn. Mater. **161**, 49 (1996).
[14] V. M. Uzdin, W. Keune, H. Schörr, and M. Walterfang, Phys. Rev. B **63**, 104407 (2001).
[15] N. M. Rensing, B. M. Clemens, and D. L. Williamson, J. Appl. Phys. **79**, 7757 (1996).
[16] J. F. Bobo, M. Piecuch, and E. Snoeck, J. Magn. Magn. Mater. **126**, 440 (1993).
[17] J. F. Bobo, H. Kikuchi, O. Redon, E. Senoeck, M. Piecuch, and R. L. White, Phys. Rev. B **60**, 4131 (1999).
[18] F. Stobiecki, T. Luciński, C. Loch, J. Dubowik, B. Szymański, M. Urbaniak, and K. Röll, J. Magn. Soc. Jpn. **23**, 176 (1999).