phys. stat. sol. (a) **189**, No. 3, 705–709 (2002)

Interface Structure and Indirect Coupling in Annealed Fe/Cr/Fe Ultrathin Films

M. Kubik (a), B. Handke (a, b), W. Karaś (a), N. Spiridis (b), T. Ślęzak (a), and J. Korecki[1]) (a, b)

(a) Faculty of Physics and Nuclear Techniques, University of Mining and Metallurgy, al. Mickiewicza 30, PL-30-059 Kraków, Poland

(b) Institute of Catalysis and Surface Chemistry, Polish Academy of Sciences, ul. Niezapominajek 8, PL-30-239 Kraków, Poland

(Received May 1, 2001; accepted September 30, 2001)

Subject classification: 68.35.Ct; 68.60.Dv; 75.70.Ak; 75.70.Cn; 76.80.+y; S1.1; S1.2

Molecular beam epitaxy grown (001) oriented Cr/Fe/Cr and Cr/Fe/Cr/Fe/Cr sandwiches were characterized using the conversion electron Mössbauer spectroscopy (CEMS), which proved that the FeCr interface extended up to about 2.5 atomic layers. Analysis of the CEMS results was based on a simple alloy-model of the Fe/Cr interface, resulting in concentration profiles of Fe and Cr atoms. The derived interface model was then used to study the effect of thermal annealing on the film properties. The CEMS studies were correlated with the measurements of the indirect exchange coupling followed by the magneto-optic Kerr effect. Whereas CEMS revealed a measurable effect of annealing on the interface atomic structure for the annealing temperature $T_A = 200\,^\circ C$, the coupling character began to change at considerably higher temperature (about 400 °C).

Introduction Among numerous papers concerning coupling in Fe/Cr multilayers, only relatively few deal with Fe/Cr/Fe sandwiches [1–3]. They represent an important tri-layer system of ferromagnetic films coupled through a non-magnetic one, in which the indirect exchange is influenced by different extrinsic factors connected with the spacer and interface atomic and magnetic structure. It has been suggested [3] that the roughness and atomic interface intermixing between Fe and Cr are mainly responsible for suppressing the short-range coupling oscillations, with the period of two Cr(001) atomic layers (AL), and leaving the long-range ones, with the period of 12 Cr AL. Such observations were gathered mainly from studying model systems on single crystalline whisker substrates [3]. Recently, we have undertaken detailed studies of Fe/Cr sandwich structures grown on the commonly used MgO(001) substrates, to find the correlation between the interface structure and the magnetic properties [4]. The conversion electron Mössbauer spectroscopy (CEMS) allowed to characterize Fe/Cr interfaces in single Fe films sandwiched between Cr layers on the atomic scale. CEMS offers the unique possibility to locally analyze both the structural and the magnetic properties of buried interfaces. Moreover, the isotopic sensitivity of the method enables depth profiling of iron films using a ^{57}Fe probe layer embedded during the growth in a film consisting otherwise of ^{56}Fe. The probe layer concept is here especially useful for verifying an asymmetry suggested for the Fe/Cr and Cr/Fe interfaces [5]. In the present paper we use the derived interface model to follow properties of coupled Fe films in order to find the influence of the annealing induced modification of the interface structure on the type and strength of the coupling.

[1]) Corresponding author; Tel.: +4812 6172911; Fax: +4812 6341247;
e-mail: korecki@uci.agh.edu.pl

 0031-8965/02/18902-0705 $ 17.50+.50/0

Experimental The sample preparation and characterization procedure was described in detail in the previous paper [4]. Molecular beam epitaxy, ultrahigh vacuum (UHV) preparation [6] was essential for the sample perfection in view of the high reactivity of Fe and Cr layers and their structural sensitivity to impurities. The investigated coupled Fe/Cr/Fe structures were obtained as: Mg(001)/20 nm Cr(001)/^{57}Fe$_N$(001)/Cr$_M$(001)/^{57}Fe$_N$(001)/5 nm Cr(001), where Mg(001) is a single crystalline cleaved substrate, 20 nm Cr(001) is a buffer, ^{57}Fe$_N$(001) are two coupled ^{57}Fe films consisting of $N = 14$ AL, Cr$_M$(001) is a Cr spacer film consisting of $M = 8$ to 20 AL and 5 nm Cr(001) is a capping layer. All layers were grown at a substrate temperature $T_p = 50\,°C$. A nominal Fe(001) or Cr(001) monolayer is assumed to be 0.1435 nm thick. The growth process was *in situ* controlled with low energy electron diffraction (LEED) showing (001) epitaxial orientation across the entire structure.

The CEMS measurements could be performed *in situ*, at UHV condition [6] or *ex situ*, using a proportional conversion electron detector. Since the sample stability after the atmosphere exposure was verified, most of the CEMS experiments were done *ex situ* to obtain better spectra quality. However, the samples have been always introduced into an UHV preparation chamber for annealing.

The coupling was characterized performing longitudinal Kerr intensity measurements using a standard magnetometer with polarization modulation and lock-in detection.

Results CEMS studies of Fe$_N$(001) single films sandwiched between Cr(001) showed that the films with a nominal thickness $N \geq 5$ AL (about 0.7 nm) can be modeled as composed of a bulk-like core and an interface of a constant thickness. For comparison, if the interfaces were ideally flat, the central core part of the film would consist of $(N - 4)$ AL, since, according to alloy model [4, 7], only Fe atoms from the surface and sub-surface layers are distinguishable from the bulk in a Mössbauer spectrum. From this point of view, our Fe films should be considered as nearly ideally layered, as seen from the plotted contribution of the bulk-like spectral component as a function of the film thickness, shown in Fig. 1. Comparison with the dependence expected for an ideal film (broken line) indicates that only less than 1 AL contributes to the deviation from a perfect structure. However, depth profiling of iron films using a ^{57}Fe probe layer embedded during the growth in a film consisting otherwise of ^{56}Fe [4] revealed that the detailed interface structure is more complicated than assumed by the simple interface/core model. It has been found that the film material of nominal 5 AL is distributed over two non-equivalent interfaces spreading through seven intermixed Fe–Cr AL, in total for both interfaces. The interface resulting from deposition of Fe on Cr is nar-

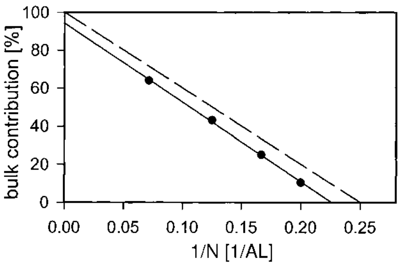

Fig. 1. Relative contribution of the bulk-like component in the CEMS spectrum of Cr(001/Fe$_N$(001)/Cr(001) films in the Fe film thickness ranging from $N = 5$ to 14 atomic layers as a function of $1/N$ (points and the solid line). The broken line shows the dependence expected for an ideally layered film, in which each interface consists of two atomic layers

rower and sharper than the one formed when Cr is deposited on Fe. Such enhanced Fe–Cr intermixing was reported for single crystalline substrates – Cr single crystals [8] or Fe whiskers [9] – only at elevated preparation [8] or annealing [9] temperatures. The presently observed interfacial alloying at room temperature is favored by the step-like structure of the Cr buffer layer seen distinctly from the LEED analysis [4], which led to the conclusion that about 20% of the surface Cr atoms in the buffer layer are located at step edges. This finding coincides with the STM observation by Choi at al. [8] that steps or island edges work as the reaction sites for incorporation of the Fe adatoms into the Cr substrate.

The $Fe_N/Cr_M/Fe_N$ sandwiches do not differ considerably from single Fe layers as seen by CEMS. It is clear however that the interfacial roughness increases for successive Fe layers in the stack. This effect is illustrated in Fig. 2, which shows the comparison of the CEMS results for the single iron $Cr/Fe_{14}/Cr$ film and for the $Fe_{14}/Cr_8/Fe_{14}$ sandwich. The both spectra are dominated by a narrow bulk-like component with a similar contribution amounting to 65% of the spectral area, which corresponds to nine bulk-like central layers. To visualize the interfacial effects, the bulk-like component has been subtracted from the spectra shown in Fig. 2a. The interfacial spectral component for the double layer is considerably broader than for the single Fe film, as seen also from the extracted interfacial distributions of the hyperfine magnetic field B_{hf} in Fig. 2b. The interface structure is also slightly sensitive to the thickness of the Cr spacer. Increasing M from 8 to 20, interfacial roughening is observed. Simultaneously, the coupling character in this M range changes between antiferromagnetic, ferromagnetic and non-collinear one.

Post-preparation annealing improves in some cases giant magnetoresistance (GMR) in Fe–Cr multilayers, which is accompanied by the increase of the interfacial roughness [10], but also by diffusion of Cr into the Fe film, as verified by CEMS analysis of sputtered polycrystalline samples. Certainly, the grain boundary diffusion dominated the above mentioned processes. We examined the influence of annealing on the interface structure and on the coupling character in the $Fe_{14}/Cr_8/Fe_{14}$ epitaxial sandwich, for

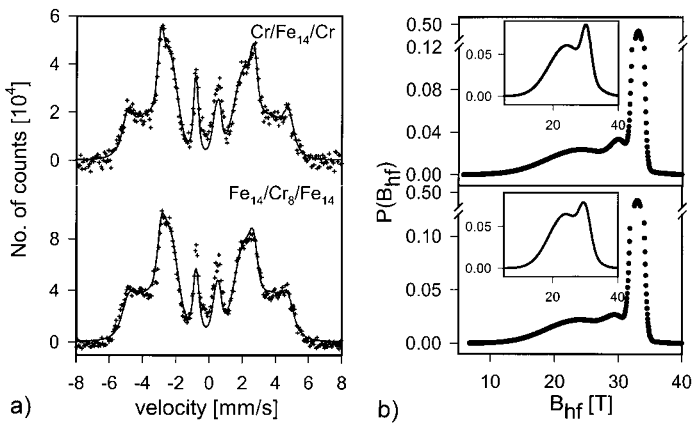

Fig. 2. Comparison of the CEMS spectra for the $Cr/Fe_{14}/Cr$ and $Fe_{14}/Cr_8/Fe_{14}$ sandwiches. a) The CEMS spectra after subtraction of the bulk-like contributions and b) B_{hf} distributions; the insets show extracted interfacial contribution

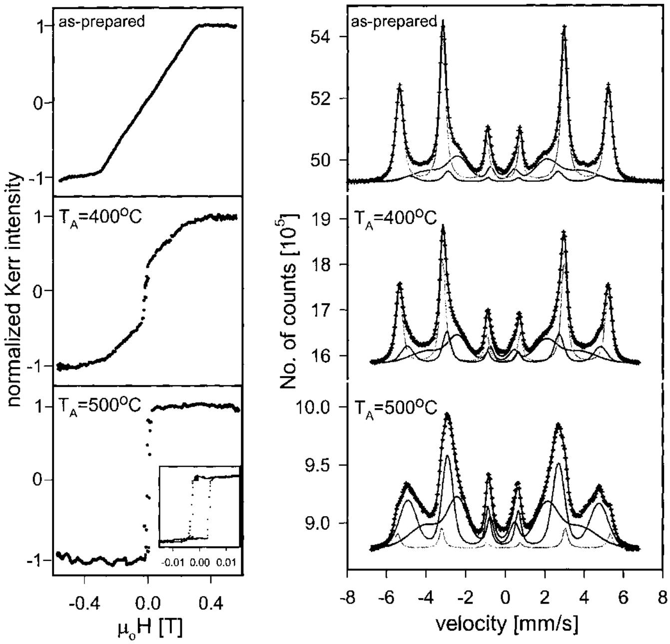

Fig. 3. Kerr loops (left) and CEMS spectra (right) of the as-prepared and annealed $Fe_{14}/Cr_8/Fe_{14}$ double-layer

which antiferromagnetic coupling, characterized by a remanence-less linear increase of the Kerr signal saturating at $\mu_0 H_s = 0.32$ T in the as-prepared state, was proved. The effect of annealing, negligible up to $T_A = 200\,°C$, is exemplified in Fig. 3. A remanence appears in the Kerr loops, going along with a decrease of the saturation field. This is accompanied by gradual changes of the CEMS spectra, which however begin to evolve at a slightly lower temperature than the Kerr loops. Also, whereas the changes in the Kerr loop parameters are homogeneous, the CEMS spectra change in two steps. At lower annealing temperatures, all spectral components become slightly sharper, with only a minor change of their relative intensity; then, at higher annealing temperatures, above $T_A = 300\,°C$, the intensity of the spectral component corresponding to the film center begins to decrease at the cost of the interfacial ones. Finally, after 500 °C annealing, the spectrum resembles the one of a FeCr alloy [11] whereas the Kerr loop becomes rectangular and the sample is fully saturated in the remanence state. Such behavior can be explained, similarly as it was done by Kopcewicz et al. [10] for polycrystalline Fe/Cr superlattices, by assuming an enhanced interfacial diffusion, which occurs at a relatively low temperature and leads to interface smoothening. Obviously, such process does not influence the interlayer exchange coupling but may lead to an enhancement of the GMR effect, as reported previously [10]. At higher annealing temperatures, a mass transport vertical to the interfaces occurs, which causes that the Fe/Cr interfaces become blurred and the ferromagnetic order appears in the intermixed Fe/Cr/Fe alloy-like layers. Rensing et al. [12] reported that adding Cr to the Fe layers in Fe/Cr multilayers dramatically enhances the GMR and lowers the saturation field, leaving however a completely antiferromagnetic coupling character. Because we observed that annealing induces a strong ferromagnetic contribution, it seems that it is Fe, which diffuses into the Cr spacer, making it able to form ferromagnetic bridges between Fe layers.

Acknowledgement This work was supported by the Polish State Committee for Scientific Research, grant no. 7 T08C 002 16 and 2 P03B 142 17.

References

[1] S. T. Purcell, W. Folkerts, M. T. Johnson, N. V. E. McGee, K. Jager, J. aan de Stegge, W. B. Zeper, and W. Howing, Phys. Rev. Lett. **67**, 903 (1991).
[2] D. T. Pierce, J. Unguris, R. J. Celotta, and M. D. Stiles, J. Magn. Magn. Mater. **200**, 290 (1999), and references therein.
[3] J. Unguris, R. J. Celotta, and D. T. Pierce, Phys. Rev. B **69**, 1125 (1992).
[4] M. Kubik, T. Ślęzak, M. Przybylski, W. Karaś, and J. Korecki, Vacuum **63**, 337 (2001).
[5] T. Shinjo and W. Keune, J. Magn. Magn. Mater. **200**, 598 (1999).
[6] J. Korecki, M. Kubik, N. Spiridis, and T. Ślęzak, Acta Phys. Polon. A **97**, 129 (2000).
[7] V. M. Uzdin, W. Keune, H. Schrör, and M. Walterfang, Phys. Rev. B **63**, 104407 (2001).
[8] Y. J. Choi, I. C. Jeong, J.-Y. Park, S.-J. Kahng, J. Lee, and Y. Kuk, Phys. Rev. B **59**, 10918 (1999).
[9] D. Venus and B. Heinrich, Phys. Rev. B **53**, R1733 (1995).
[10] M. Kopcewicz, T. Luciñski, F. Stobiecki, and G. Reiss, J. Appl. Phys. **85**, 5039 (1999).
[11] S. M. Dubiel and J. Żukrowski, J. Magn. Magn. Mater. **15–18**, 655 (1980).
[12] N. M. Rensing, B. M. Clemens, and D. L. Williamson, J. Appl. Phys. **79**, 7757 (1996).