



ELSEVIER

Journal of Magnetism and Magnetic Materials ■■■■■ ■■■■■ ■■■■■



www.elsevier.com/locate/jmmm

# Epitaxial growth, alloying and magnetic structure of interfaces in Fe/Cr (001) superlattices

V. Uzdin<sup>a</sup>, W. Keune<sup>b,\*</sup>, M. Walterfang<sup>b</sup>

<sup>a</sup>St. Petersburg State University, ICAPE, 14 linia V.O. 29, 199178, St. Petersburg, Russia

<sup>b</sup>Laboratorium für Angewandte Physik, Gerhard-Mercator-Universität Duisburg, Lotharstr. 65, D-47048 Duisburg, Germany

## Abstract

Fe/Cr(001) superlattices containing two-monolayers thick <sup>57</sup>Fe probe layers at the Fe/Cr (Fe-on-Cr) or Cr/Fe (Cr-on-Fe) interfaces were studied using conversion electron Mössbauer spectroscopy (CEMS). For the interpretation of the CEMS data of superlattices annealed at different temperatures, we performed theoretical modeling of their chemical and magnetic structure. Roughness and interface alloying were introduced to the model by algorithms of epitaxial growth, which included ballistic deposition with subsequent floating of some atoms on the surface. Self-consistent calculations of magnetic moments within the periodic Anderson model confirmed the proportionality between hyperfine fields and magnetic moments. For the explanation of the evolution of CEM spectra versus annealing temperature, the difference in the melting points of bulk Fe and Cr has to be taken into account. © 2001 Published by Elsevier Science B.V.

**Keywords:** Mössbauer spectroscopy; Multilayers; Annealing; Magnetic structure; Fe–Cr

In Fe/Cr multilayers antiferromagnetic interlayer exchange coupling and giant magnetoresistance (GMR) were discovered for the first time. Despite the large efforts which were undertaken for studying the correlation between their interface structure and macroscopic magnetic and transport properties, many aspects are still contradictory. In particular, the value of the GMR effect was found to correlate with the spectral contribution (spectral area) of the satellite Zeeman sextet in the conversion electron Mössbauer (CEM) spectra [1] that corresponds to a hyperfine field,  $B_{hf}$ , of 20 T. According to the traditional interpretation for Fe/Cr multilayers, the peak at 20 T in the hyperfine field (hff) distribution,  $P(B_{hf})$ , originates from Fe atoms at the ideally flat (001) interface with four nearest neighbours and one second neighbour Cr atoms [1–3]. This interpretation seems to allow the correlation of transport properties, interface roughness and the ratio between interface and bulk scattering [1]. However, our

recent calculations of magnetic moments in Fe/Cr multilayers with interface alloying [4] did not confirm the assumption that a large spectral intensity of the 20-T satellite means that the interface is atomically smooth. On the contrary, our results showed that this peak corresponds to short-range interdiffused Fe atoms inside the Cr spacer layers, but not far away from the interface. Therefore, its relative spectral intensity has to increase upon interface alloying. According to our recent findings, earlier conclusions about the role of interface and bulk scattering in the GMR effect should be revised. We emphasize in this context that the interpretation of experimental data is an ambiguous problem, and for the understanding of real mechanisms of epitaxial growth and interface alloying in Fe/Cr multilayers, experimental and theoretical studies have to be used together to achieve self-consistency in details.

Here, we present results of a CEMS investigation after isochronal annealing of Fe/Cr(001) superlattices with two-monolayers (ML) thick <sup>57</sup>Fe probe layers (95% enriched) placed either at Fe/Cr interfaces (Fe-on-Cr or “lower” interfaces) or at Cr/Fe interfaces (Cr-on-Fe or “upper” interfaces), and of theoretical modeling of

\*Corresponding author. Tel.: +49-203-379-2387; fax: +49-203-379-3601.

E-mail address: keune@uni-duisburg.de (W. Keune).

Table 1

Position of the fitted Gaussian peaks (satellites) in the hff distribution (first column) and their relative spectral area (in %) for the sample with the  $^{57}\text{Fe}$  probe layers at the upper (Cr-on-Fe) and lower (Fe-on-Cr) interfaces after isochronal annealing at different temperatures

$B_{\text{hff}}$ (T)	RT	200°C	300°C	400°C	450°C	500°C	550°C
<i>Upper interface: Cr on Fe</i>							
33.7(%)	17.8	21.6	21.9	22.2	23.3	21.4	22.5
31.3(%)	16.1	13.3	12.8	12.1	12.5	20.2	28.3
28.5(%)	13.9	13.9	14.7	16.3	14.0	17.1	30.4
25.6(%)	11.3	12.9	12.0	12.6	13.0	10.6	
23.1(%)	11.3	12.0	13.1	10.1	11.2	8.5	
20.3(%)	15.1	11.1	10.0	9.9	11.2	11.9	15.5
16.5(%)		12.1	12.4	13.9	12.2	8.2	
<i>Lower interface: Fe on Cr</i>							
33.8(%)	29.6	30.7	30.4	32.3	31.3	30.0	25.6
31.4(%)	15.8	13.9	15.3	14.5	14.9	18.1	21.9
28.7(%)	11.3	12.9	13.1	13.9	13.2	15.1	17.2
25.5(%)	10.9	11.6	10.5	9.8	9.7	10.1	13.6
23.0(%)	10.4	9.7	9.6	8.9	11.1	7.2	
20.0(%)	10.7	11.1	11.9	13.7	10.4	11.1	17.4

interface alloying in these systems during the sample preparation and annealing process.

Superlattices of composition  $\text{MgO}(001)/\text{Cr}(50 \text{ \AA})/[^{57}\text{Fe}(2 \text{ ML})/^{nat}\text{Fe}(12 \text{ ML})/\text{Cr}(8 \text{ ML})]_{15}$  (lower interfaces) or  $\text{MgO}(001)/\text{Cr}(50 \text{ \AA})/[^{nat}\text{Fe}(12 \text{ ML})/^{57}\text{Fe}(2 \text{ ML})/\text{Cr}(8 \text{ ML})]_{15}$  (upper interfaces) were epitaxially grown at  $T_s = 433 \text{ K}$  by ultrahigh-vacuum (UHV) deposition of the metals on epipolished  $\text{MgO}(001)$  substrates, as described in detail in Ref. [4]. ( $^{nat}\text{Fe} = \text{Fe}$  metal of natural  $^{57}\text{Fe}$  abundance, 2.1%). Characterization by low and high angle X-ray diffraction demonstrates the high-quality superlattice structure of our samples [4].

CEM spectra were measured at room temperature as described in Ref. [4]. Typical CEM spectra and hff distributions before annealing of samples with either “upper” or “lower” interfaces were shown in Fig. 2(a) of Ref. [5]. One-hour isochronal annealing of the samples was performed in UHV in steps of increasing temperatures between 200°C up to 550°C, when strong bulk diffusion starts. The CEM spectra and hff distributions (not shown here) are of similar statistical quality as those displayed in Refs. [4] (Figs. 4 and 5) and [5] (Fig. 2(a)). The obtained hff distributions have been decomposed by least-squares fitting into several (here: six or seven) Gaussian functions (satellites) with individual widths and spectral areas. Our results, i.e. the average position of the Gaussian satellite lines and the corresponding relative spectral areas (in %, relative to the total experimental spectral area), obtained after annealing both types of samples at different temperatures, are given in Table 1. The total area (corresponding

to the sum of the areas of the fitted Gaussian peaks) obtained for each annealing temperature is only slightly less than 100% of the total experimental spectral area according to Table 1.

The hff distributions for the two types of interfaces prove to be remarkably different (Table 1). For the Fe-on-Cr interface, the relative area of the “bulk” peak (near 33.8 T) is found to be about 30%, whereas for the Cr-on-Fe interface it is only about 20%. Other satellite peaks are narrower and generally yield less contribution (except for the peak near 31.4 T) to the total spectrum for the lower interface as compared with the upper interface. The largest difference was detected for the peak near 20 T: before annealing it contributes less than 11% of the total spectral area for the lower interface and more than 15% for the upper one. An additional peak corresponding to a hff of 16.5 T appears in  $P(B_{\text{hff}})$  of the upper interface after annealing at 200°C and above. In general, for the upper interface the low-field distribution was found to be essentially broader and the amplitude of the 20 T peak itself was smaller than that for the lower interface. Annealing of the samples up to a temperature of 450°C does not crucially modify the CEM spectra. A small increase of the “bulk” contribution (33.8 T peak) and of the low-field contribution (20 T peak for the lower interface, and the sum of the 20.3 and 16.5 T peaks for the upper interface) up to a temperature of 400°C reflects the weak (short-range) diffusion of  $^{57}\text{Fe}$  atoms from the interface towards the inside of the Fe and Cr slabs, respectively. A similar result concerning a weak increase in area of the 20 T satellite line after annealing at 300°C was reported by Kopcewicz et al. [3]. Their

1 interpretation is based on the assumption that this hff  
 3 corresponds to Fe atoms in the “flat” interface, and they  
 5 had to conclude that there is in-plane diffusion inside the  
 7 superlattice during annealing which leads to smoothing  
 9 of the interfaces. However, this is an unlikely process,  
 and our finding that the 20 T peak (as well as the 16.5 T  
 peak) originates from Fe atoms embedded in the Cr  
 spacer near the interface [4] gives a more natural  
 explanation of the annealing experiment.

11 Another problem is the explanation of the differences  
 13 in  $P(B_{hf})$  for lower and upper interfaces. This difference  
 15 was reported earlier [5,6], and it was assumed that  
 17 interface alloying is governed by the binding energies  
 19 between the substrate and ad-atom material, which, as a  
 21 first approach, are proportional to the melting points of  
 23 the solids [6]. The melting point of Cr is higher than for  
 25 Fe and, therefore, interface mixing for the lower  
 27 interface might be less significant as compared with the  
 29 upper one [6]. However, recent investigations of epitaxy  
 31 of Fe on Cr [7] and Cr on Fe [8] using scanning  
 33 tunneling microscopy demonstrate the occurrence of  
 35 alloying on both interfaces. Modeling of the epitaxial  
 37 growth with the algorithm of simple ballistic deposition  
 39 cannot reproduce the differences between interfaces [4].  
 41 Now, we developed a new algorithm [9] for interface  
 43 alloying, which includes ballistic deposition with con-  
 45 sequent rising up of some atoms on the surface. It allows  
 47 to reproduce the main differences between lower and  
 49 upper interfaces. We assume that site exchange of atoms  
 51 and their diffusion take place only at the surface during  
 53 the epitaxial growth and there is no internal bulk  
 55 diffusion. We start from the structure obtained by the  
 algorithm of simple ballistic deposition. Then in every  
 layer, we choose a definite fraction ( $\zeta$ ) of atoms using a  
 random procedure, and layerwise, starting from the  
 bottom, this fraction of atoms was exchanged in every  
 pair of neighbouring layers. The parameter  $\zeta < 1$   
 determines the intensity of interface alloying. Such a  
 scenario automatically leads to the asymmetry of the  
 interface: atoms could float up on several layers, but did  
 not move down due to suppression of diffusion in the  
 inner layers below the surface. For the probe layer at the  
 lower (Fe-on-Cr) interface,  $^{57}\text{Fe}$  atoms will float and  
 move into the  $^{nat}\text{Fe}$  slab, thus increasing the intensity of  
 the bulk-like peak in  $P(B_{hf})$ . At the upper (Cr-on-Fe)  
 interface, these  $^{57}\text{Fe}$  atoms will float and move into the  
 Cr spacer, thus increasing the low-field contribution in  
 $P(B_{hf})$ . Self-consistent calculations of the magnetic  
 moment distribution performed within the periodic  
 Anderson model for these structures show a consider-  
 able number of Fe atoms which have a magnetic  
 moment corresponding to the hff of about 20 T [9],  
 contrary to the case of structures obtained by a simple  
 ballistic deposition algorithm [4]. Note that such a  
 scenario of epitaxial growth is very general. It does not  
 assume any differences in the strength of interactions

between atoms of different chemical elements, and gives  
 a natural explanation of the change of the hff distribu-  
 tion on  $^{119}\text{Sn}$  atoms in V/Cr superlattices versus the  
 position of the  $^{119}\text{Sn}$  probe layer inside the Cr spacer [10].

We conclude that the difference between the hff  
 distributions from lower and upper interfaces in Fe/Cr  
 superlattices can be explained without consideration of  
 Fe and Cr melting points. However, the different melting  
 points play an important role in the evolution of  $P(B_{hf})$   
 versus annealing, especially at high temperature, when  
 bulk diffusion starts. After annealing at 500°C, we  
 observe a remarkable increase of the spectral area of the  
 second satellite line (31.3 T–31.4 T) in both types of  
 interfaces. This means that individual Cr atoms start to  
 penetrate into the Fe layers. There is no similarly  
 remarkable increase of the low-field contribution at  
 500°C, which would correspond to isolated Fe atoms in  
 the Cr spacer. The latter process starts only after  
 annealing at 550°C for the lower (Fe-on-Cr) interface  
 whereas for the upper (Cr-on-Fe) interface, the total  
 low-field contribution (at 20.3 and 16.5 T) decreases  
 strongly with annealing at 550°C. Consequently  $^{57}\text{Fe}$   
 atoms at the upper interface do not penetrate deeply (far  
 away from the interface) into the Cr spacer even at such  
 a high annealing temperature. Although the observed  
 starting temperature of 500–550°C for bulk diffusion is  
 remarkably lower than the melting points of bulk Fe and  
 Cr, it is closer to the Fe melting point. That is why Cr  
 atoms can diffuse into the Fe slabs, which are nearer to  
 the liquid state, but Fe atoms do not diffuse into the  
 solid Cr spacer. This conclusion is in agreement with  
 recent calculations of vacancy formation energies which  
 are found to be larger for Cr than for Fe [11].

V.U. appreciates financial support from the Alexan-  
 der von Humboldt foundation and the MML01  
 symposium organizers.

This work was partially supported by the Russian  
 Ministry of Higher Education (grant E00-3.4-547), the  
 program “Universities of Russia: fundamental re-  
 searches” (project 015.01.01.083) and the Deutsche  
 Forschungsgemeinschaft (SFB 491 Bochum/Duisburg).

## References

- [1] R. Schad, P. Belien, G. Verbanck, K. Temst, V.V. Moshchalkov, Y. Bruynseraede, B. Bahr, J. Falta, J. Dekoster, G. Langouche, Europhys. Lett. 44 (1998) 379.
- [2] F. Klinkhammer, Ch. Sauer, E.Yu. Tsybal, S. Handschuh, Q. Leng, W. Zinn, J. Magn. Magn. Mater. 161 (1996) 49.
- [3] M. Kopcewicz, T. Lucinski, F. Stobiecki, G. Reiss, J. Appl. Phys. 85 (1999) 5039.
- [4] V. Uzdin, W. Keune, H. Schrör, M. Walterfang, Phys. Rev. B 63 (2001) 104407.
- [5] T. Shinjo, W. Keune, J. Magn. Magn. Mater. 200 (1999) 598.

- 1 [6] B. Heinrich, J.F. Cochran, T. Monchesky, R. Urban, Phys.  
Rev. B 59 (1999) 14520. 11
- 3 [7] A. Davies, J.A. Stroscio, D.T. Pierce, R.J. Celotta, Phys.  
Rev. Lett. 76 (1996) 4175. 13
- 5 [8] Y.J. Choi, I.C. Jeong, J.-Y. Park, S.-J. Kahng, J. Lee,  
Y. Kuk, Phys. Rev. B 59 (1999) 10918. 15
- 7
- 9 [9] V. Uzdin, W. Keune, Phys. Metals, Metallurgy (2001) in  
press. 11
- [10] M. Almokhtar, K. Mibu, A. Nakanishi, T. Kobayashi,  
T. Shinjo, J. Phys.: Condens. Matter 12 (2000) 9247. 13
- [11] P.A. Korzhavyi, I.A. Abrikosov, B. Johansson,  
A.V. Ruban, H.L. Skriver, Phys. Rev. B 59 (1999) 11693. 15

UNCORRECTED PROOF