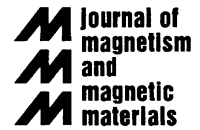




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Epitaxial growth, alloying and magnetic structure of interfaces in Fe/Cr (001) superlattices

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Abstract

Fe/Cr(001) superlattices containing two-monolayers thick ⁵⁷Fe 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. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Mössbauer spectroscopy; Multilayers; Annealing; Magnetic structure

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, $P(B_{\text{hf}})$, 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 ⁵⁷Fe 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

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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}Fe probe layers at the upper (Cr-on-Fe) and lower (Fe-on-Cr) interfaces after isochronal annealing at different temperatures. CEMS spectra were measured at room temperature

B_{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 $\text{MgO}(001)/\text{Cr}(50 \text{ \AA})/[^{57}\text{Fe}(2 \text{ ML})/^{54}\text{Fe}(12 \text{ ML})/\text{Cr}(8 \text{ ML})]_{15}$ (lower interfaces) or $\text{MgO}(001)/\text{Cr}(50 \text{ \AA})/[^{54}\text{Fe}(12 \text{ ML})/^{57}\text{Fe}(2 \text{ ML})/\text{Cr}(8 \text{ ML})]_{15}$ (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]. ($^{54}\text{Fe} = \text{Fe}$ metal of natural ^{57}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}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

interpretation is based on the assumption that this hff corresponds to Fe atoms in the “flat” interface, and they had to conclude that there is in-plane diffusion inside the superlattice during annealing which leads to smoothing 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.

Another problem is the explanation of the differences in $P(B_{\text{hf}})$ for lower and upper interfaces. This difference was reported earlier [5,6], and it was assumed that interface alloying is governed by the binding energies between the substrate and ad-atom material, which, as a first approach, are proportional to the melting points of the solids [6]. The melting point of Cr is higher than for Fe and, therefore, interface mixing for the lower interface might be less significant as compared with the upper one [6]. However, recent investigations of epitaxy of Fe on Cr [7] and Cr on Fe [8] using scanning tunneling microscopy demonstrate the occurrence of alloying on both interfaces. Modeling of the epitaxial growth with the algorithm of simple ballistic deposition cannot reproduce the differences between interfaces [4]. Now, we developed a new algorithm [9] for interface alloying, which includes ballistic deposition with consequent rising up of some atoms on the surface. It allows to reproduce the main differences between lower and upper interfaces. We assume that site exchange of atoms and their diffusion take place only at the surface during the epitaxial growth and there is no internal bulk diffusion. We start from the structure obtained by the algorithm of simple ballistic deposition. Then in every layer, we choose a definite fraction (ζ) 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}Fe atoms will float and move into the $^{\text{nat}}\text{Fe}$ slab, thus increasing the intensity of the bulk-like peak in $P(B_{\text{hf}})$. At the upper (Cr-on-Fe) interface, these ^{57}Fe atoms will float and move into the Cr spacer, thus increasing the low-field contribution in $P(B_{\text{hf}})$. Self-consistent calculations of the magnetic moment distribution performed within the periodic Anderson model for these structures show a considerable 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 distribution on ^{119}Sn atoms in V/Cr superlattices versus the position of the ^{119}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 $P(B_{\text{hf}})$ 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}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].

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