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Distribution of magnetic moments and hyperfine fields for Fe/Cr multilayers with different interface roughness

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Abstract

Distribution of d-electron magnetic moments has been calculated within the Periodic Anderson Model (PAM) for Fe/Cr superlattices with various interface roughness. Special random algorithms were developed for the modelling of stepped interfaces with different average size of the steps as well as for the modelling of interface alloying. Self-consistent calculation of magnetic moment distribution for alloyed interfaces show strong correlation with hyperfine fields (hff) on Fe nuclei, measured by the Mössbauer spectroscopy. It allows one to correlate the hff with specific environment of interfacial Fe atoms and leads to the essential correction of the empirical approach for the interpretation of Mössbauer spectra. New criterion for the testing of smoothness of the interface using Mössbauer data is suggested.

Keywords: Fe/Cr interface magnetism; Mössbauer spectroscopy

The modern state of investigations of magnetic nanostructures is characterized by increased attention to the role of the interface roughness, alloying, embedded clusters, pin-holes, etc. Such space defects cannot be avoided for real systems but they prove to be responsible for the novel and interesting properties important for fundamental magnetism and for the applications. Epitaxial growth and all the characteristics of complex magnetic nanostructures depend strongly on the details of the sample preparation.

In particular in Fe/Cr trilayers bilinear exchange coupling can be changed by as much as a factor of 5 by varying the substrate temperature during the growth of the first Cr atomic layer [1]. In situ magnetometry measurement demonstrated the very large decrease of the total moment during Cr evaporation on the smooth Fe surface whereas for the rough Fe surface no changes of total moment at all was observed [2,3]. All the theories on non-collinear magnetic ordering in the Fe/Cr systems [4] presuppose the existence of the spatial defects which are the real reasons of non-collinear structure formation. Therefore, investigation of the roughness, interdiffusion, their dependence on the growth conditions and the control of the interface structure using complementary experimental methods become a principal problem for understanding of the Fe/Cr interface magnetism.

Most of the experimental techniques give only indirect information about chemical and magnetic roughness of interfaces at the atomic scale level. Averaging over the whole interface region or even on several interfaces in the multilayers make it difficult to reconstruct the microscopic interface

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structure from experimental data. Mössbauer spectroscopy with ^{57}Fe-probe layers provide local information about interface magnetism via different hyperfine fields (hff) at different interface sites. However the interpretation of these data is a very complicated and ambiguous problem due to the nontrivial connection between the hff and the local magnetic moments localised on Fe atoms. Surprisingly but for the most of the Fe/Cr multilayer structures, the position of satellite lines in Mössbauer spectra does not differ essentially [5–9], although all other characteristics like GMR, parameters of exchange coupling etc., as a rule, are very different. Comparison of hff distributions for Fe/Cr interfaces in the multilayers and for Fe/Cr random alloys leads to the conclusion that as a first approach interface region can be considered as bulk alloy with varying concentration. For the treatment of Mössbauer spectra most of investigations follow the empirical procedure, which was suggested initially for description of random alloys [10]. The hff B_{hff} on Fe nuclei is assumed to decrease linearly with the number of the Cr atoms at nearest (n_1) and next nearest (n_2) positions

$$B_{\mathrm{hff}} = B_{\mathrm{hff}}(\mathrm{bulk}) + n_1 B_1 + n_2 B_2, \tag{1}$$

where B_1 (B_2) is the contribution to the hff from one Cr atom in the first (second) shell around Fe atom under consideration. Improvement of the resolution of the measurements and development of the epitaxial growth technique offers the opportunity to refine the alloy model. Klinkhammer et al. [5] suggested for the description of the set of hff the following relation:

$$B_{\mathrm{hff}} = B_{\mathrm{hff}}(\mathrm{bulk}) + n_1 B_1 + n_2 B_2 + \Delta B + \Delta B_{(i=2)}. \tag{2}$$

Besides fitting parameters $B_1 = 2.5$ T and $B_2 = 2.05$ T which change their values as compared to simple alloy approach (1), this relation contains two additional parameters $\Delta B = -1.75$ T which the authors [5] correlate with the broken spatial symmetry in the transverse direction and $\Delta B_{(i=2)} = -1.2$ T which exists only for the atoms next nearest from the interface Fe layer. Taking into account that $B_{\mathrm{hff}}(\mathrm{bulk})$ is negative we obtain for these atoms from (2) and enhancement of hff relatively to the bulk value.

Fig. 1 illustrates the distributions of hff obtained for Fe/Cr interfaces by different groups using Conversion Electron Mössbauer Spectroscopy (CEMS). Interpretation of CEMS data on the basis of empirical approach (1) or (2) leads for the same set of hff to different conclusions about local environment of Fe atoms. Therefore microscopic analysis of Fe/Cr interface magnetism taking into account roughness and interdiffusion is very important for the understanding of the physical information which can be extracted from CEMS data.

Most of the calculations of magnetic moment distributions in mixed Fe/Cr layers of multilayered systems were restricted to ordered structure at the interface and on the very thin interface region, which include usually 1–2 monolayers [11,12]. Here we present the results of calculations of the magnetic structure on atomic scale for disordered Fe/Cr interface within collinear Periodic Anderson Model (PAM). Self-consistent calculations of d-electron moments on each atomic site were performed in Hartree–Fock approximation using modification of zero and pole method [13,14]. The d–d interaction is taken into account inside first shell of the atom under consideration. Calculation of magnetic moments were performed to obtain self-consistency both on every site and between magnetic moments localized on different sites. One more procedure is used to determine the Fermi energy, which fixes the total number of d-electrons during self-consistency.

Interface roughness was introduced into the model by two kinds of random procedure. The first produces the steps at the interface, whereas another one simulates the interface alloying and diffusion. For modeling the stepped interfaces we started from the ideal superlattice Fe(N_1)/Cr(N_2), ($N_1 = 14,\ N_2 = 8$), where all (1 0 0)-layers contain only Fe or Cr atoms. In each plane we used 64×1 cell with periodic boundary conditions. Total length of the cell (64 sites) was divided into even number of equal intervals L_0, where L_0 is the parameter of the algorithm. Then for every pair of neighboring intervals we choose random number

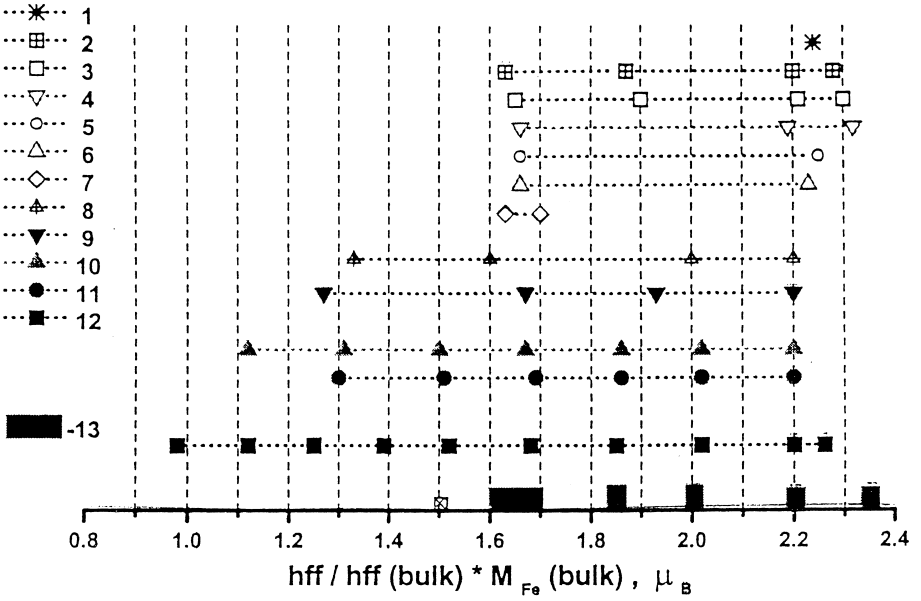

Fig. 1. Distribution of hff in Fe/Cr structures. ^{57}Fe layers are undelayered:
1. W(1 1 0)/Cr(1 1 0): 40 ML/Fe(1 1 0): 3 ML + 2 ML + 21 ML + 2 ML + 3 ML/Cr(1 1 0), [6];
2. W(1 1 0)/Cr(1 1 0): 40 ML/Fe(1 1 0): 3 ML + 25 ML + 3 ML/Cr(1 1 0), [6];
3. W(1 1 0)/Cr(1 1 0): 40 ML/Fe(1 1 0): 3 ML + 4 ML + 3 ML/Cr(1 1 0), [6];
4. W(1 1 0)/Cr(1 1 0): 40 ML/Fe(1 1 0): 6 ML/Cr(1 1 0), [6];
5. W(1 1 0)/Cr(1 1 0): 40 ML/Fe(1 1 0): 4 ML/Cr(1 1 0), [6];
6. W(1 1 0)/Cr(1 1 0): 40 ML/Fe(1 1 0): 3.3 ML/Cr(1 1 0), [6];
7. W(1 1 0)/Cr(1 1 0): 40 ML/Fe(1 1 0): 2 ML/Cr(1 1 0), [6];
8. Si/Fe: 60 Å [Cr: 11/Fe: 30 Å]$_{60}$/Cr: 11 Å, [9];
9. MgO/Fe(0 0 1)/Cr(0 0 1), [8];
10. MgO/Cr: 50 Å [Fe(1 0 0): 3 ML + 8 ML + 3 ML/Cr(1 0 0): 8 ML]$_{10}$, [7];
11. MgO/Cr: 50 Å/[Fe(1 0 0): 0.7 ML + 25 ML + 0.7 ML/Cr(1 0 0): 8 ML]$_{10}$, [7];
12. MgO/Cr: 400 Å/Fe: 2 ML/Cr: 10 Å, [5]; GaAs/Fe: 10 Å/Ag: 1500 Å/Fe: 40 Å + 2 ML/Cr: 10 Å, [5];
13. calculations with PAM.

$l < L_0$ and l successive interface Fe atoms from the first interval were exchanged with l successive interface Cr atoms from the second one. Position of l atoms inside the intervals L_0 was taken as random. This procedure produces stepped interface where one monolayer (ML) height up steps alternate with one ML height down-steps and the lateral size of every step do not exceed L_0. Decreasing the value of the parameter L_0 leads to the formation of the interfaces with larger density of steps per unit of length. In the perpendicular in-plane direction the system stays spatially homogeneous. Modeling of each superlattice was repeated 20 times to obtain the distributions, which does not depend on particular input structure.

Alloying and interdiffusion have been simulated in ballistic regime using algorithm "epitaxy". This algorithm fills the prism with a cross-section of 8×8 atomic sites by Fe and Cr atoms. Outside the prism the structure was repeated periodically. The height of the prism was taken equal to 30 ML. Initially the bottom layer of the prism was filled by Fe atoms and all the other sites inside the prism were empty. Then new atoms are thrown at the top level of the prism with random procedure and epitaxy routine provided their descent through empty sites in bcc lattice until the sliding is blocked. Transfer of atoms from one layer to the next layer occurs with equal probability to any of the nearest neighbor sites. Probability of this

transfering P_{tr} and probability for atom to stop in a given layer $1 - P_{tr}$ depends on the number of availible sites in the next layer. Set of the probabilities P_{tr} are input parameters of the epitaxy algorithm. Different P_{tr} leads to distinct surface and interface structure [13,14]. Successive deposition of Fe and Cr atoms leads to the layered structure with rough interfaces. It is also possible to model the sample with different kinds of Fe atoms and to study the moment distribution only for the Fe atoms of one kind, similar to the probing the interface magnetism using ^{57}Fe layers in experiments. Taking into account that surface layer is not fully filled by the atoms we delete few top and bottom layers and put periodic boundary conditions for the remaining layers in the prism to obtain the same multilayers structure Fe(14)/Cr(8). Such modeling was repeated 20 times like in the case of stepped interfaces.

Calculations of magnetic moments for the stepped interface with $L_0 = 4$ and $L_0 = 16$ show very similar distributions where deviation from the bulk moments take place only for 3 interface layers. Distribution function have few sharp maxima, one of which corresponds to the bulk moment, and other to the enhanced moment up to $3.4\mu_B$. There are also two extended maxima with less amplitudes in the region $1.5–1.8\mu_B$ and $1.95–2.05\mu_B$. Existence of atoms with enhanced moments most of which have only one second neighbor Cr is in accordance with empirical expression (2), which give enhanced hff on such atoms. This enhancement was detected only for the epitaxial samples with smooth interfaces [5,6] and was never reported for sputtered multilayers.

Alloying and interdiffusion at the interface leads to the distribution of the moments with larger number of maxima. Some of them, however, merge with increasing of the roughness. Fig. 2 shows such distribution for 3 interface ML in the case of "smooth" interface [13,14], where only 2–3 layers contain both Fe and Cr atoms. Such inter-

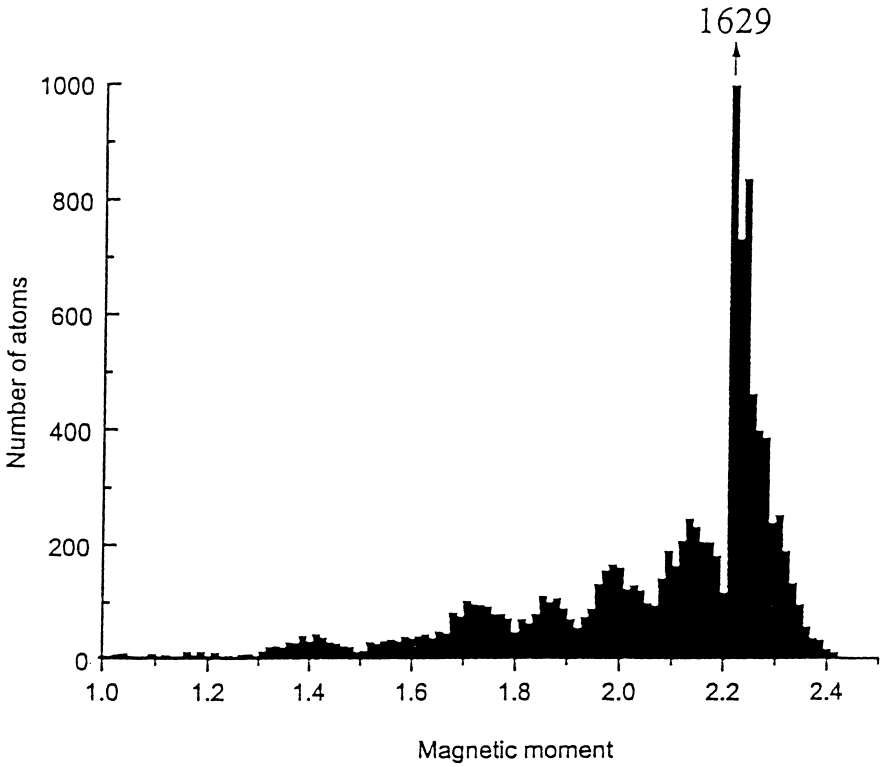

Fig. 2. Distribution of magnetic moments for Fe atoms in 3 interface ML. Interdiffusion is modelled by epitaxy algorithm.

face come out when atoms are forced to slide into the next layer if there is at least one empty place among its nearest neighbors. The distribution function contains well-separated maxima, which give most probable values of Fe moments. Positions of these maxima strongly correlate with hff measured by CEMS as seen in Fig. 1. It allows one to say that in Fe/Cr layered structures localized magnetic moments scale with hff on ^{57}Fe atoms.

In our calculations we do not reproduce the peaks in the distribution of the magnetic moments corresponding to the small hff less than 20 T, because only the structures where Fe atoms do not penetrate far from the interface to the Cr spacer were considered. For the rougher interfaces we obtain however peak around ($1.5\mu_B$), which is shown in Fig. 1 as small square with cross. Such moments ordered antiparallel to the direction of magnetisation in Fe slabs have Fe atoms embedded into the Cr spacer near interface.

The satellite with $H = 19$–20 T ($M \sim 1.3\mu_B$) was associated in previous studies [5–8] with Fe atoms at the "flat" interface. Correspondingly the increase of its amplitude in the Mössbauer spectra which take place, in particular, with the increase of the substrate temperature during sample preparation [7] was interpreted as smoothing of the interface. Calculated values of Fe moments on the flat interface (about $1.7\mu_B$) prove to be essentially higher than the moment $M^* = (H^*/H_{Bulk})M_{Bulk}$, which corresponds to the hff H^*. If we accept the hypothesis about proportionality of the magnetic moments and hff, the field H^* should be located on atoms which have more Cr neighbors and, therefore lie inside Cr spacer similar to the "loose spins" in the models of non-collinear ordering [4]. Large contribution of the satellite with hff H^* cannot be the signature of the smooth interface but on the contrary, indicate alloying and interdiffusion.

Our calculations give new criterion for the determination of the interface roughness. It is the onset of the hff enhanced relatively to the bulk Fe. Its observation was reported only on the specially prepared samples where interdiffusion was artificially suppressed [6].

Conclusion that hff H^* is associated with Fe atoms inside Cr spacer leads to some important consequences about mechanisms of the epitaxial growth of Fe on Cr and Cr on Fe. Mössbauer study shows that for probe ^{57}Fe layer at the Fe/Cr interface the amplitude of the H^* satellite enhanced in comparison with Cr/Fe interface [7]. It was interpreted in [7] as a proof of alloying suppression during Fe growth on Cr substrate in accordance with simple thermodynamic consideration based on different melting points for bulk Fe and Cr [1]. Our calculations show that interdiffusion however exists and that the number of Fe atoms which penetrate into the Cr spacer is even greater than for Cr on Fe substrate. Recent STM-study of Fe on Cr epitaxial growth [15] reveals surface alloy formation which confirms our conclusions.

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