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Interface roughness/intermixing and magnetic moments in a Fe/Co(001) superlattice

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Abstract

The superiority of a local probe method over diffraction methods in determining the interface details is demonstrated by a comparison between Fe/Co and Fe/V superlattice data. From Mössbauer spectra, the higher interface quality in the Fe/Co superlattice was evident although the x-ray diffraction data were similar in the two cases. In fact, by comparison of the details of the Mössbauer spectrum for the Fe/Co case with published values of the magnetic hyperfine field for iron in cobalt no detectable roughness/intermixing was found. From the variation of the iron magnetic hyperfine field as a function of location of the iron atoms, with respect to the interface, the individual iron magnetic moments could be derived. The magnitudes of the magnetic moments thus obtained correlate well with recent calculated values.

1. Introduction

Thin layers of bcc Co can be grown in superlattices together with thin bcc layers of Fe as shown earlier [1]. By introducing a radioactive probe of ^{57}Co Dekoster *et al* [1] were also able to determine the magnetic hyperfine field at room temperature for dilute Fe in bcc Co. This experimental value, 31.2(1) T, which can be compared with 33.0 T for Fe in bcc Fe, has in the present study been used as a means to obtain detailed information on the atomic distribution close to the interface. For Fe close to the interface layer an average field of 35.4(1) T was observed [1], in good agreement with the value of 35.5(5) T from a recent study in our group of the magnetic anisotropy in Co/Fe superlattices. The Leuven group also studied the lattice relaxation of an Fe/Co superlattice using the PAC technique [2] and found evidence for sharp interfaces from studies of the transferred hyperfine field at the probe Cd [3]. The PAC studies do, however, rely on the observations from an impurity atom in the superlattice, which could affect the actual physical situation in the material. In the present study, using ^{57}Fe Mössbauer spectroscopy, the probe atoms are regular constituents of the system under study and should thus deliver unperturbed information. Similarly, one can from the Co hyperfine field distribution, as determined from NMR studies, obtain local information from the Co side of the superlattice as has been discussed by Panissod *et al* [4].

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The advances in the calculation of magnetic properties using the local density approximation allows for predictions also of magnetic moment distributions in metals. Thus, layer resolved magnetic moments in a perfect interfaced bcc crystal consisting of Co and of Fe, grown in the (001) direction, have been calculated for a sample at 0 K [5]. Against this background, the present study was undertaken with two things in mind: firstly to examine the local distribution of Co and Fe in the layer by layer growth, and secondly to investigate the validity of the calculated magnitude of the magnetic moments. The method used was ^{57}Fe conversion electron Mössbauer spectroscopy measuring the local magnetic hyperfine field, which is directly related to the local surrounding and to the local magnetic moment. The structural results are compared with data for Fe/V multilayers to stress the importance of the Mössbauer method in determining the interface structure.

2. Experiment

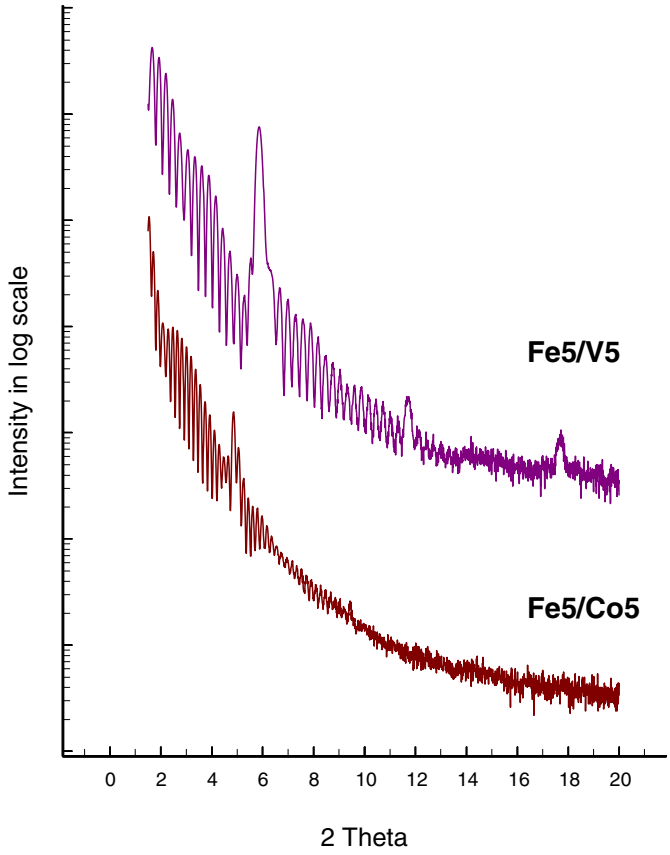
The superlattices were grown on annealed (30 minutes at 900 K) single crystal wafers of MgO ($20 \times 20 \times 1$ mm^3) in a three target, ultra-high vacuum based sputtering equipment. The samples were grown at 300 K using targets of Co (99.5%), V (99.7%) and isotopically enriched (>95%) ^{57}Fe and had the nominal structure MgO(001) [^{57}Fe (5ML)/X (5ML)]$_{20}$ with X = Co or V and capped with 10 ML V. Typical deposition rates were 0.5 Å s^{-1}.

The sample quality was checked by recording x-ray diffraction (XRD) data both at low angles ($2\theta = (1.5^\circ$–$20^\circ)$) and high angles ($2\theta = (50^\circ$–$80^\circ)$) using Cu Kα radiation ($\lambda = 1.540\,562$ Å).

The conversion electron Mössbauer spectra were obtained using a spectrometer working in constant acceleration mode with a 25 mCi ^{57}Co Rh source and with the samples at room temperature. The detector used was a gas flow detector using a (He–CH_4) mixture as counting gas. Calibration spectra were recorded simultaneously with the main measurement using a α-Fe reference at room temperature.

3. Results

In figure 1 the low and high angle XRD scans for the Fe5/Co5 and the Fe5/V5 samples are displayed. The low angle reflectivity curve shows well defined peaks arising from the chemical modulation and the high angle diffraction pattern shows a well defined (002) Bragg peak with satellites up to the first order as is expected for a symmetric superlattice. The average monolayer thickness m in the multilayer and the periodicity Λ for Fe5/Co5 and Fe5/V5 samples were found to be $(m, \Lambda) = 1.420(1)$ Å, 18.54(1) Å and 1.447(1) Å, 15.19(1) Å, respectively. From these values the number of atomic layers per period are calculated as 13.1(1) ML and 10.5(1) ML for the two samples. These values deviate from the nominal 10 ML especially for the Fe/Co multilayer. If 2.866 and 2.82 Å [6] are used as lattice parameters for α-Fe and bcc Co respectively, the averaged monolayer thickness for equal amount of Fe and Co would give $m \approx 1.42$ Å in good agreement with the found experimental value. We thus conclude that the Fe/Co multilayer sample, nominally denoted Fe5/Co5, has a periodicity of ≈6.5 ML of Fe and ≈6.5 ML of Co. From a corresponding analysis for the Fe/V multilayer sample using the result presented in [7] we can conclude that the individual Fe and V layer thicknesses are within error equal. The out-of-plane structural coherence length ξ is calculated from the equation $\xi = \lambda/[2 \sin\theta(002)\,\Gamma(002)]$ where $\lambda = 1.540\,562$ Å and $\Gamma(002)$ is the full width at half maximum of the (002) Bragg peak. The values found are 143 Å and 154 Å for the Fe5/Co5 and Fe5/V5 sample respectively. The roughness of the interface could not be reliably



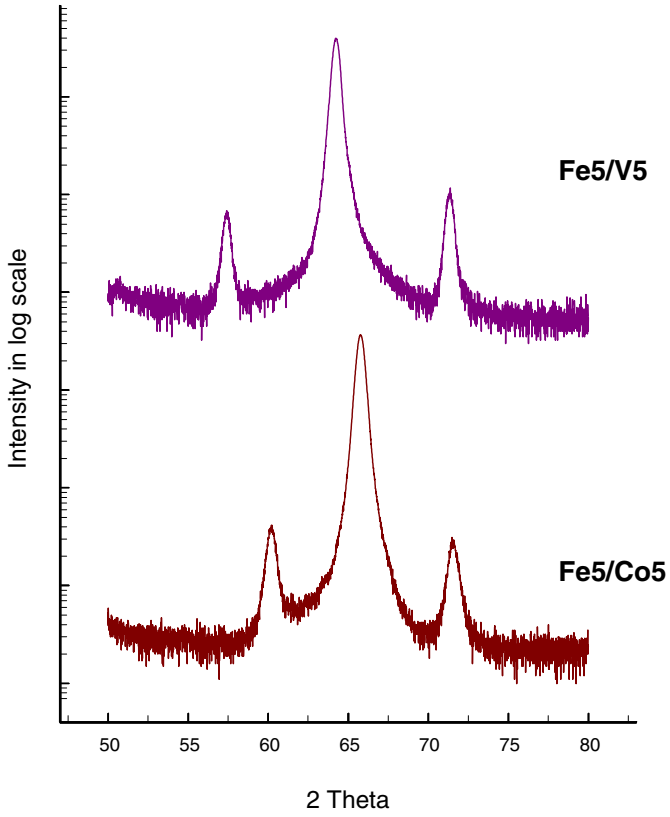
(a)

Figure 1. Low (a) and high (b) angle XRD scans for the two samples using $\lambda = 1.540\,562$ Å. The small number of satellites is due to the limited thickness of the sample. The poor contrast of the low angle data in the Fe/Co case is due to the similarity of electron numbers between Fe and Co.

determined due to the complexity in the parameter space and the rather low resolution in the present low angle x-ray data. However from other Fe/Co and Fe/V multilayer samples with larger number of repetitions, but prepared in the same way, the roughness parameter has been found to be around 2 Å [8].

The recorded Mössbauer spectra are shown in figure 2 and the least squares fitting results for Fe5/Co5 are given in table 1. (The data for the Fe/V sample have been presented earlier [7].) In the fitting three six line patterns were used. The Lorentzian line width W was kept the same for all three patterns and also the intensity ratios of the sub-spectral six lines. The best fit value for the sample linewidth was $W = 0.141(5)$ mm s^{-1} and the line intensity ratios were found to be 3:3.51:1:1:3.51:3 indicating that the magnetic moments are, within $\pm 15^{\circ}$, confined to the sample plane as expected from the shape anisotropy. Taking into consideration line intensity saturation, this value would be further reduced.

The non-zero value found for the electric quadrupole splitting ε (table 1), although very small, corroborates the slightly tetragonal distortion of the cubic lattice due to the mismatch (1.6%) between the Fe and Co lattice constants.



(b)

Figure 1. (Continued)

Table 1. Results of the fitting of the Mössbauer spectrum of the Fe/Co sample. B is the magnetic hyperfine field, I is the spectral intensity, δ is the isomer shift versus Fe metal at room temperature, ε is the first order electric quadrupole splitting. The estimated experimental errors are $B(\pm 0.5$ T), $I(\pm 1\%)$, $\delta(\pm 0.005$ mm s$^{-1})$, $\varepsilon(\pm 0.005$ mm s$^{-1})$.

	Pattern	B (T)	I (%)	δ (mm s^{-1})	ε (mm s^{-1})
1	Fe IL	37.2	34	0.043	−0.034
2	IL + 1	35.4	37	0.029	0.012
3	IL + 2	33.0	29	0.023	0.027

4. Comparison between Fe/V and Fe/Co samples

It is instructive to compare superlattices involving Fe prepared in the same way in the same set-up but with one magnetic spacer (Co) and with one non-magnetic spacer (V). As seen in figure 1, the XRD diffractograms are rather similar for the two samples but the Mössbauer spectra are markedly different (figure 2) with a much broader magnetic hyperfine field distribution in the Fe/V case as compared to the Fe/Co case. For an ideal layer-by-layer growth there would exist just three different Fe surroundings as given in table 2 represented by three different sub-spectra in the recorded Mössbauer spectrum. In the Fe/V case one requires more than three subsets to fit the data while for Fe/Co only three different environments

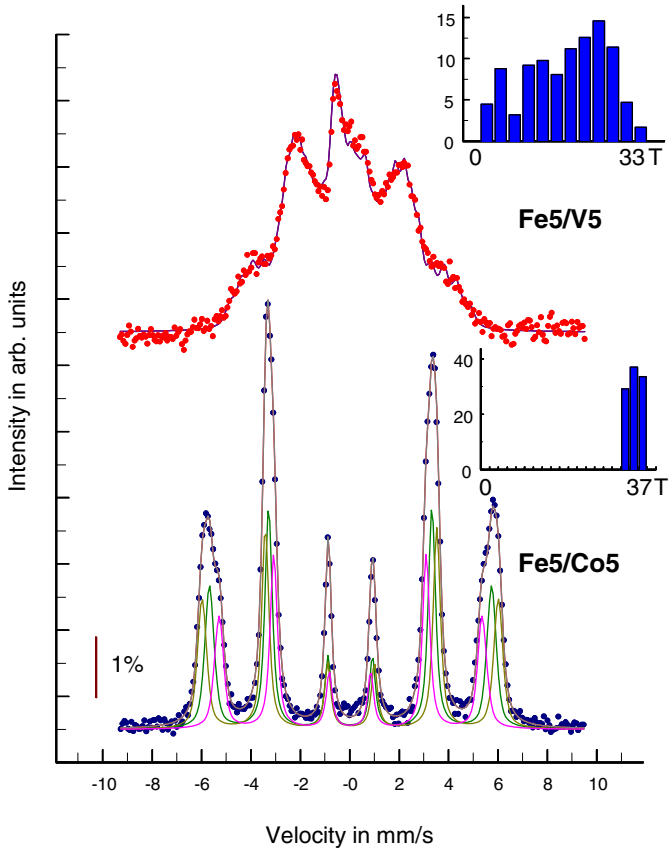


Figure 2. The CEMS spectra for the Fe5/V5 and Fe5/Co5 superlattices recorded at room temperature. The insets show the magnetic field distributions as found from the fittings.

for iron are needed in the fit. V being a non-magnetic nearest neighbour atom around Fe should have a larger influence on the Fe magnetic hyperfine field than a magnetic Co atom. The field distributions found in the V case cannot, however, be explained on this basis only: one has to introduce a relatively large roughness/intermixing of the elements at the interface as well [5]. Mössbauer spectroscopy, however, being a local probe technique, does not distinguish between roughness and intermixing since these defects do not differ in the local atomic structure but only in the arrangements of those local structures. The Mössbauer spectra reveal a marked difference in the atomic distributions in the interface area for the two cases, which is not apparent from the diffraction data. Judging the sample quality and the sharpness of the interface from x-ray diffractograms should, therefore, be done with caution.

5. Comparison between NMR and Mössbauer data

The Fe/Co multilayers are particularly attractive for local probe studies of the interface characteristics since both constituents can be studied using either NMR (Co) or the Mössbauer techniques (Fe). The two methods observe the interface region from different sides and, ideally, the results should be conformal. In most cases, however, the variations between samples are

Table 2. Summary of the experimental and theoretical result for the Fe/Co superlattice together with atomic surroundings for different layers. The layer notation represents the different distances (in number of monolayers) of the monolayers counted from the Fe interface layer denoted IL.

Atom in layer	Nearest neighbours	Next nearest neighbours	Experimental magnetic field (T)	Experimental magnetic moment (μ_B)	Theoretical magnetic moment [5] (μ_B)
Co IL − 2	8 Co	6 Co	31.2 (Fe) [1]		1.75 (Co)
Co IL − 1	8 Co	5 Co + 1 Fe			1.76 (Co)
Co IL	4 Co + 4 Fe	5 Co + 1 Fe			1.75 (Co)
Fe IL	4 Co + 4 Fe	5 Fe + 1 Co	37.2 (Fe)	2.48	2.57 (Fe)
Fe IL + 1	8 Fe	5 Fe + 1 Co	35.4 (Fe)	2.36	2.33 (Fe)
Fe IL + 2	8 Fe	6 Fe	33.0 (Fe)	2.20	2.25 (Fe)

such that a direct comparison is only possible when using the same sample. Such studies are planned but no results are available so far. All the same, it might be of interest to compare the available data.

The NMR data of Panissod *et al* [4] show Co resonance intensity for the system [Fe (24 Å)/Co (2–42 Å)] over a fairly large frequency range although with some predominant features and the data are analysed in a model involving alloy formation. The results most relevant for the present comparison are the ones for low Co thickness. In this range, essentially all intensity is attributed to an Fe/Co alloy. This is in marked contrast to the Mössbauer data presented above and discussed further below, and the results, taken together, are very strong evidence for large differences in sample properties resulting from differences in growth conditions. The samples in [4] were MBE grown on GaAs(110) without specification of the growth temperature (the authors do also mention MgO as substrate but those results seem not to have been published) whereas the samples in the present study were sputter deposited on MgO(100) at an elevated, controlled temperature.

6. Magnetic moments

The lowest observed magnetic field is 33.0 T which is markedly higher than the reported field for Fe in a bcc Co matrix of 31.2 T [1]. The line width is too small to hide an additional six-line pattern, coming from Fe in a Co matrix, with any reasonable intensity. It is, therefore, obvious that the layer roughness or intermixing in the Fe/Co sample is very small. Assuming a symmetry in the layer roughness between Co and Fe, i.e. that growing cobalt on iron gives the same local structure for Fe as growing iron on cobalt, our experimentally found roughly six layers of Fe in the superlattice can be reduced to three different layer types with different occurrences: an interface layer (IL) with occurrence 2, an intermediate layer (IL + 1) with occurrence 2 and the layer (IL + 2) with occurrence 2. In the Mössbauer spectrum these layers would show up as three different six line patterns with equal spectral intensities. From the fitting result as presented in table 1 it is not easy to assign patterns 1, 2 and 3 to Fe in the different layers IL, IL + 1 and IL + 2. Using the conversion factor of 15 T μ_B^{-1}, valid for bulk Fe at room temperature, the moments would be 2.48 μ_B, 2.36 μ_B and 2.20 μ_B for pattern 1, 2 and 3 respectively. Relying on the theoretical results for the layer resolved magnetic moments [5] we may however propose an assignment of pattern 1, 2, 3 to Fe in IL, IL + 1 and IL + 2, respectively as written in the tables and depicted in figure 3. The isomer shift, δ, approaches zero when moving from the interface layer deeper into Fe layer a trend consistent with the assignment of the different components.

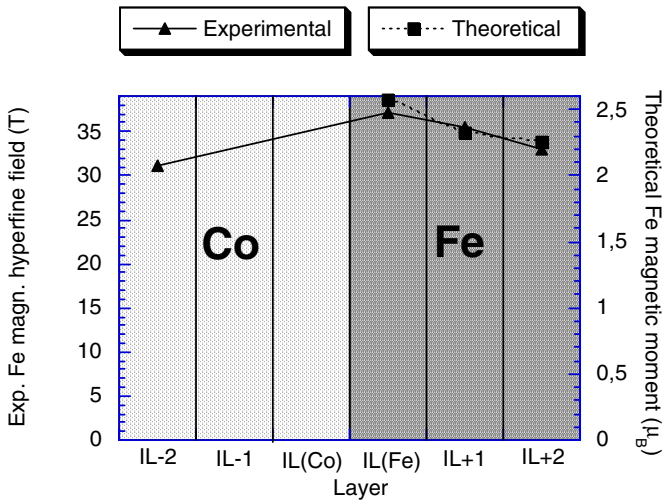


Figure 3. Experimental Fe magnetic hyperfine fields and theoretical moments as a function of layer position in an ideal Fe/Co interface structure (values from table 2).

In table 2 we present the experimental Fe hyperfine fields, theoretical magnetic moments and nearest atomic surroundings valid for different layers in the Fe/Co superlattice. The connection between hyperfine field and magnetic moment is not obvious for interfaces between magnetic and non-magnetic elements, as discussed e.g. by Przybylski [9], but in the present case, involving two similar magnetic materials, the ratio between magnetic moment and hyperfine field can be approximated to be constant (figure 3). This is also borne out in the MCXD study by Pizzini *et al* [10]. Comparing the data from theory and experiment shows that they follow the same trend but deviate somewhat from each other in absolute values. The values derived also show that the conversion factor to obtain the magnetic moments is appropriately chosen.

7. Conclusions

The present study has shown that Mössbauer spectroscopy, and by comparison also NMR, is a valuable tool in studying the sample quality and the magnetic properties of multilayers. In particular, the marked difference in sensitivity to the interface structure for Mössbauer spectroscopy and x-ray diffraction is illustrated by the comparison with the Fe/V case. For a high quality Fe/Co superlattice the interface quality was ascertained by comparison of the details of the spectrum with the published value of the magnetic hyperfine field for iron in cobalt. No indication of the presence of iron in a cobalt surrounding was detected and, hence, no detectable roughness/intermixing was found. From the analysis of the Mössbauer spectrum, the different components could be associated with only three different iron surroundings as would be expected for a perfect superlattice. It is estimated that the experimental uncertainty corresponds to a deviation from the ideal structure of less than 10%. From the variation of the iron magnetic hyperfine field as function of the location of the iron atoms, with respect to the interface, the individual iron magnetic moment could be derived and found to correlate well with recent calculated values.

Considering the large difference in derived parameters between the Mössbauer and the NMR data this unambiguously shows the sensitivity and applicability of the local probe

techniques in the study of the details of the atomic arrangements at interfaces in multilayers and superlattices.

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