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Conversion electron Mössbauer spectroscopy studies of the magnetic moment distribution in Fe/V multilayers

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Abstract. The Fe hyperfine field distribution in Fe(x ML)V(y ML) as function of x and y has been determined. The samples were prepared in a ultra-high vacuum sputtering system with ^{57}Fe as a probe. Low- and high-angle x-ray diffraction and Mössbauer spectroscopy were used to determine the monolayer structure of the samples. The multilayer growth is not layer-by-layer, as found from the magnetic hyperfine field distributions. No Fe magnetic ordering is found above 133 K for multilayers with $x \approx 3$ and $y \approx 14$ in contrast to recently reported antiferromagnetic ordering. The average Fe magnetic moments as deduced from the average magnetic hyperfine fields changes in the sequence 1.2, 1.5, 1.7 to 2.0 μ_B layerwise in going towards the centre of a 10 ML film of Fe.

1. Introduction

The magnetic properties of multilayers are of particular interest from a fundamental physics point of view, as well as various technological applications. In a recent paper by Isberg *et al* [1] the preparation and properties of Fe(x ML)V(y ML) (where x and y are the number of Fe and V layers, respectively) superlattices were studied. By the use of the ultra-high vacuum (UHV) sputtering technique and optimal growth conditions, superlattices with good interface and crystalline order were obtained. An interesting initial magnetic phase diagram as a function of x and y was derived. Notably, antiferromagnetic ordering was observed for $x \approx 3$ and $y \approx 14$. Conversion electron Mössbauer spectroscopy (CEMS) studies of Fe/V multilayers prepared by the sputtering technique revealed an interfacial microscopic roughness of $\sim$3 Å and, by depositing ^{57}Fe as a monolayer probe at different distances from the Fe/V interface, average magnetic moments were deduced for a 10 ML Fe/10 ML V multilayer [2]. The average room-temperature Fe moment changed from 1.1 μ_B at the interface layer to 1.5 μ_B and 1.7 μ_B for the monolayers deeper into the Fe layer (the bulk value at saturation being 2.2 μ_B). The present study is an extension of the earlier work which covers a larger part of the Fe/V magnetic phase diagram and also reports the first low-temperature data. A new analysis of the CEMS spectra for 10 ML Fe/10 ML V has also been performed.

2. Experimental details

The samples were prepared by deposition of the atoms in a UHV environment by the sputtering technique. The substrate was MgO (001) of dimensions 20 × 20 × 1 mm^3. The wafers were annealed for 1 h at about 700 °C before preparing the sample. Samples

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were grown at 300 °C using a rotating substrate holder to avoid thickness gradients in the multilayer structure. The metal targets used were V (99.7%) and Fe enriched to 95% in ^{57}Fe. The samples had the compositions: [^{57}Fe(10 ML)/V(5 ML)] × 20, [^{57}Fe(5 ML)/V(5 ML)] × 20, [^{57}Fe(2.7 ML)/V(14 ML)] × 22, [^{57}Fe(3.4 ML)/V(13.9 ML)] × 22 and [^{57}Fe(4.5 ML)/V(14.3 ML)] × 17. In the following we will use the abbreviations 10/5, 5/5 etc for the samples to represent the layers of Fe and V, respectively.

To check the structure of the multilayers, a SIEMENS D 5000 x-ray diffractometer was used and low- and high-angle data were recorded ($\lambda = 1.5416$ Å). The experiments were carried out using the CEMS technique. The room-temperature data were obtained with a gas flow He and CH_4 detector and the low-temperature data were obtained in a cryostat with a cold finger sample holder cooled by liquid nitrogen. In the cryostat, the electrons are detected by two microchannel plates. The velocity calibration spectrum was collected with a ^{57}Fe foil as a scatterer.

3. Results

Two different sets of x-ray diffraction (XRD) measurements were made, the low-angle scanning was in the range $1.5^\circ \leqslant 2\theta \leqslant 10^\circ$ and high-angle scanning covered the range $50^\circ \leqslant 2\theta \leqslant 80^\circ$. In figure 1 the XRD scans for one of the samples are displayed as an illustration of the crystal quality. The limited number of satellites seen is due to the small thickness of the sample. The average lattice parameters are 2.875 Å, 2.894 Å, 2.989 Å, 2.976 Å and 2.962 Å for 10/5, 5/5, 2.7/14, 3.4/13.9 and 4.5/14.3, respectively, while the spacings between two Fe/V layers are 23.62 Å, 25.187 Å, 25.54 Å, 25.88 Å and 28.16 Å, respectively.

The Mössbauer spectra at 295 K show magnetic splitting for samples 10/5, 5/5 and 4.5/14.3 while the spectra for samples 2.7/14 and 3.4/13.9 show no magnetic splitting, only a partly resolved asymmetric doublet (figure 2). For the spectra of the samples with $y \approx 14$ measured at 133 ± 5 K, no magnetic splitting was observed for the samples with $x \approx 3$.

The 'non-magnetic' spectra were analysed using two single Lorentzian lines with the same line width and with free intensities and centroid shifts. The 'magnetic' spectra were analysed as a superposition of 11 sextets with an equidistant magnetic field step of 3 T, a Gaussian standard deviation of 0.75 T and one or two single lines with zero field. The Lorentzian line widths for all of the individual lines were kept the same. The intensity ratio for the peaks in the sextets were fixed to 3:z:1:1:z:3. In the fittings the resulting z-value was very close to four, indicating that the magnetic hyperfine field directions are in the sample plane and thus perpendicular to the γ-ray direction. A linear correlation between the magnetic field, B, and the centroid shift CS was assumed in these samples [2]. The intensities of each subspectrum and the correlation factor were, in fact, the only parameters in the fittings. The results of the fittings are presented in table 1. Also presented are the re-analysed spectra from [2] using the same model as above. The corresponding magnetic hyperfine field distributions found are presented in figure 3. For the 10/5 sample, where a resolution of the outer lines was clearly seen, another fit was performed to obtain the precise fields. The results were 33.6 (1.0) T, 30.8 (1.0) T, 27.7 (1.0) T and 24.4 (1.0) T for the four resolved outer lines, showing that the step length of 3 T chosen above is realistic.

The fitting of the correlation factor gave a rather flat minimum in χ^2, but with almost constant intensity values for a rather broad region of the correlation factor. It turned out that the centroid shift CS varied between 0.0 mm s^{-1} and $-0.2(1)$ mm s^{-1} for the subspectral components. Since the 'non-magnetic' spectra gave a spectral component with $CS = -0.28(1)$ we adopted that value in all fittings of the 'magnetic' spectra as an end value for the centroid shift.

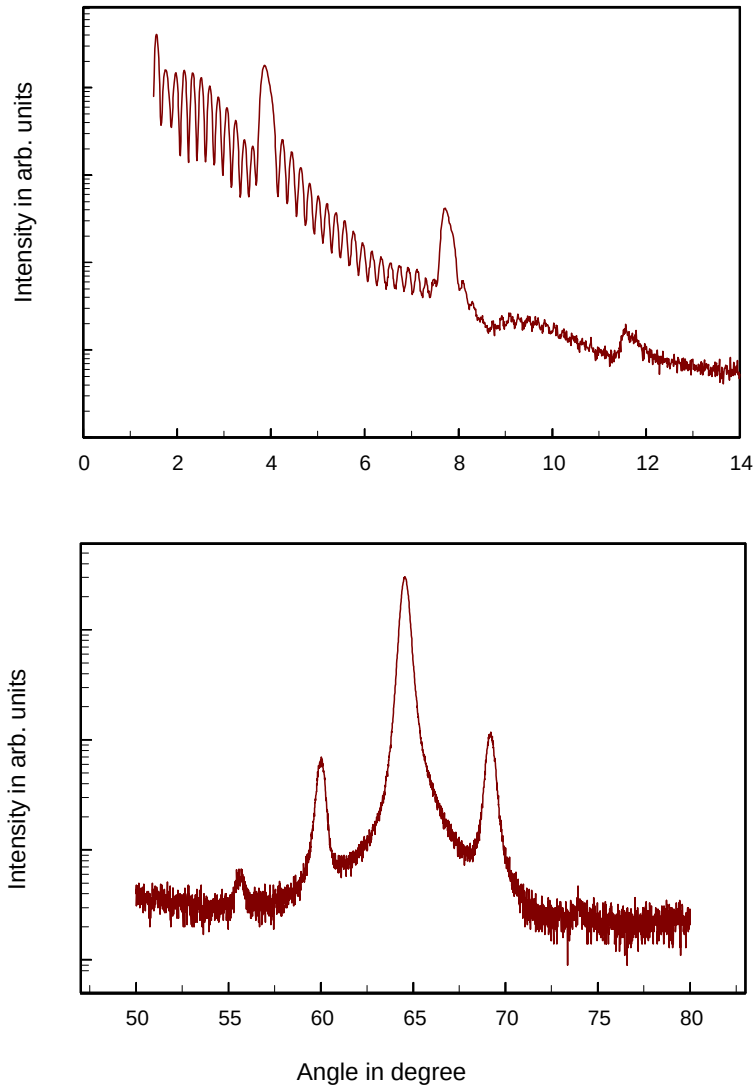

Figure 1. Characteristic low- and high-angle XRD spectra of the 10/5 multilayer. The upper figure clearly shows the interference peaks from the chemical modulation of Fe and V. The lower figure shows a well defined (002) Bragg peak at $\theta \approx 65^\circ$ with well defined satellites on each side. Cu Kα radiation with wavelength $\lambda = 1.5416$ Å was used.

4. Discussion

4.1. Samples with $x \approx 3$ and $y \approx 14$

The two samples (2.7/14 and 3.4/13.9) gave almost the same results and they are, therefore, dealt with simultaneously. From the data it is quite clear that the 'bulk' magnetic transition point, if any, is below 133 ± 5 K for this multilayer. From the paper of Granberg *et al* [3], in which 3/14 multilayers are studied by SQUID magnetometry, the conclusion was drawn that antiferromagnetic coupling between the ferromagnetically coupled Fe layers existed.

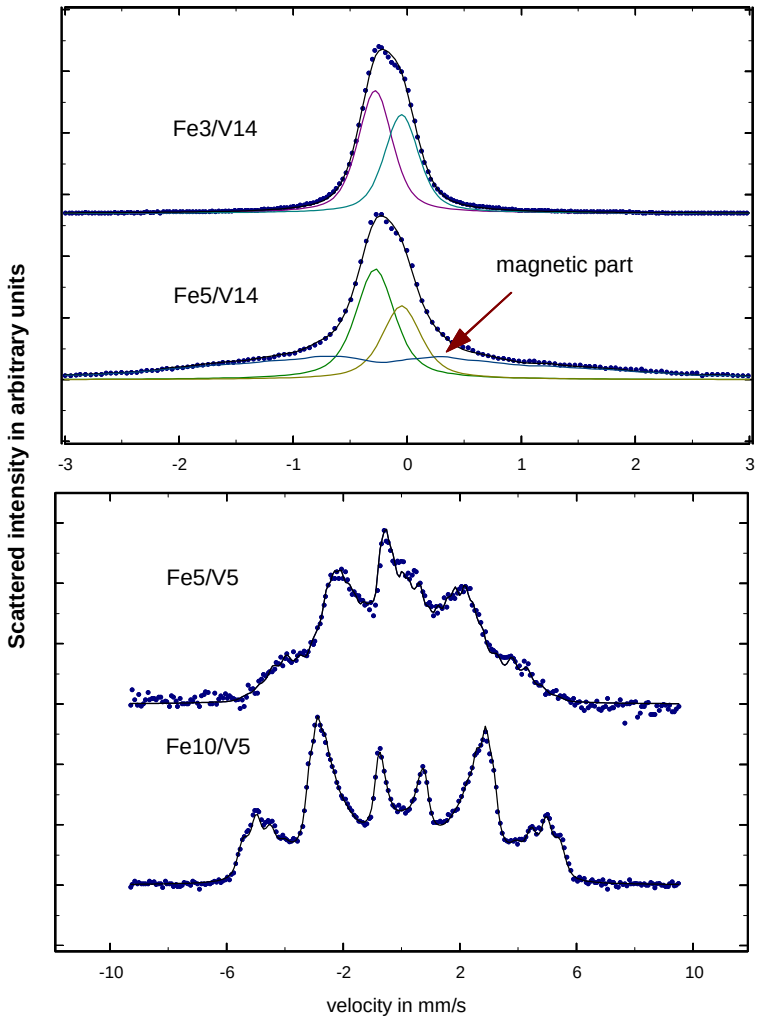

Figure 2. CEMS spectra of various samples at room temperature.

No magnetic transition temperature was given, but by assuming that the V magnetic moment is zero, the Fe saturation moment would be about 0.4 μ_B at 300 K, about 0.8 μ_B at 100 K and saturating at about 0.9 μ_B at 10 K. This is the saturation moment arrived at for a relatively high magnetizing field of about 300 kA m^{-1} in the plane of the layers. The slow approach to saturation and the low saturation moment could be due to an inhomogenous sample, but the fact that non-zero GMR is observed even at 300 K is a clear indication of antiferromagnetic coupling. However, even by assuming that only a part of the sample is magnetically ordered, this fraction would have to be large enough to be visible in the Mössbauer spectra. The differences between the results of the earlier study [3] and the present study must, therefore, be due to variations between the samples used. For the thin Fe layers of interest, slight variations in actual composition will have a dramatic influence on, for example, T_C.

The two-line feature of the Mössbauer spectra is most easily interpreted as being due to the structure of the samples. Since there are about three monolayers in each Fe layer, two of

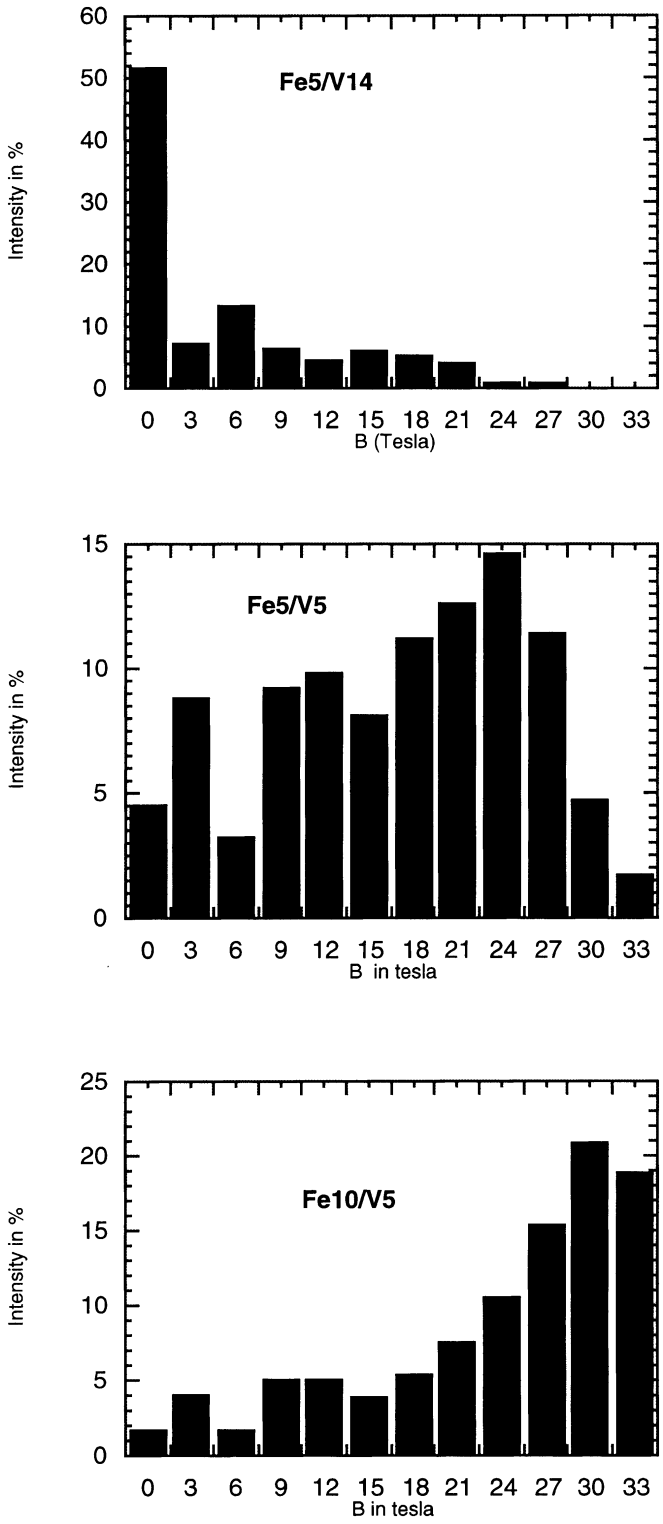

Figure 3. Hyperfine field distributions at room temperature.

Table 1. The magnetic field distribution for samples 5/5, 10/5 and 10/10. The error in intensity is ±1%. IL + 1, IL + 2 and IL + 3 represent a ^{57}Fe layer as the first, second and third layers next to the V layers in the Fe/V superlattice, respectively.

	B	Spectral intensity in samples				
	T	5/5	10/5	10/10 IL + 1	10/10 IL + 2	10/10 IL + 3
1	33	1.7	18.9	5.5	11.8	19.2
2	30	4.7	20.9	8.7	17.6	23.4
3	27	11.4	15.3	11.5	16.4	18.0
4	24	14.6	10.5	12.3	13.2	12.4
5	21	12.6	7.6	11.0	11.5	9.0
6	18	11.2	5.4	10.5	6.2	4.3
7	15	8.1	3.9	7.7	5.7	2.1
8	12	9.8	5.0	9.0	4.0	3.6
9	9	9.2	5.0	7.7	6.9	2.9
10	6	3.2	1.8	5.3	0	0.1
11	3	8.8	4.0	6.2	4.9	3.6
12	0	4.5	1.8	4.7	1.7	1.3

the monolayers are in direct contact with the V layers on one side and one is in direct contact with the Fe layers on both sides. The Fe layers of the samples can therefore be divided into two parts: one 'close to' V with an intensity of 66% and one 'close to' Fe with an intensity of 33%. In the fitting, the relative intensities were 56(2)% and 44(2)%. This crude interpretation thus gives a good picture of the situation. We interpret the strongest line as emanating from Fe sitting in a V-rich surrounding with $CS = -0.28(1)$ mm s^{-1} at 295 K and the weaker line as emanating from Fe sitting in an Fe-rich surrounding with $CS = -0.04$ mm s^{-1} at 295 K. These values correspond rather well with those observed for Fe in a pure V matrix and in a pure Fe matrix, respectively [4]. The temperature change in the centroid shift between 295 K and 133 K is $-0.092(5)(1)$ mm s^{-1} for both of the single lines, which is completely compatible with a second-order Doppler shift.

4.2. Sample with $x \approx 5$ and $y \approx 14$

The room-temperature and 133 K spectra gave similar results (figure 4), the main difference being a broadening for the 133 K spectra. Here, there are also two 'non-magnetic' single lines with a centroid shift in line with that found for the samples with $x = 3$. The amount of 'magnetic' signal is approximately 45% at 295 K. Since, in this case there are five Fe layers, two of these layers, in a perfect multilayer growth, are in direct contact with the V layer while the other three layers have more Fe-rich surroundings. Exchange coupling is expected to be observed for the Fe-rich part of the sample. We interpret, therefore, the result as follows: (i) the single line with $CS = -0.28$ mm s^{-1} and an intensity of 34% as emanating from Fe in a V-rich matrix, (ii) the single line with $CS = -0.05$ mm s^{-1} and an intensity of 21% as emanating from Fe with an Fe-rich surrounding, but of dimensions that are not large enough to have a static spin orientation (as observed during the Mössbauer observation time of about 0.1 μs) and (iii) the magnetic six-line patterns with the centroid shift between -0.05 mm s^{-1} and -0.28 mm s^{-1} and a total intensity of 45% from Fe with varying numbers of V and Fe in the near neighbour surrounding, but situated in larger clusters.

The ferromagnetic transition temperature obviously depends on the thickness of the Fe layer and for this multilayer it is above 295 K. The magnetic hyperfine field distribution for this sample at 295 K is presented in figure 3. As can be observed the Fe nuclei are exposed to

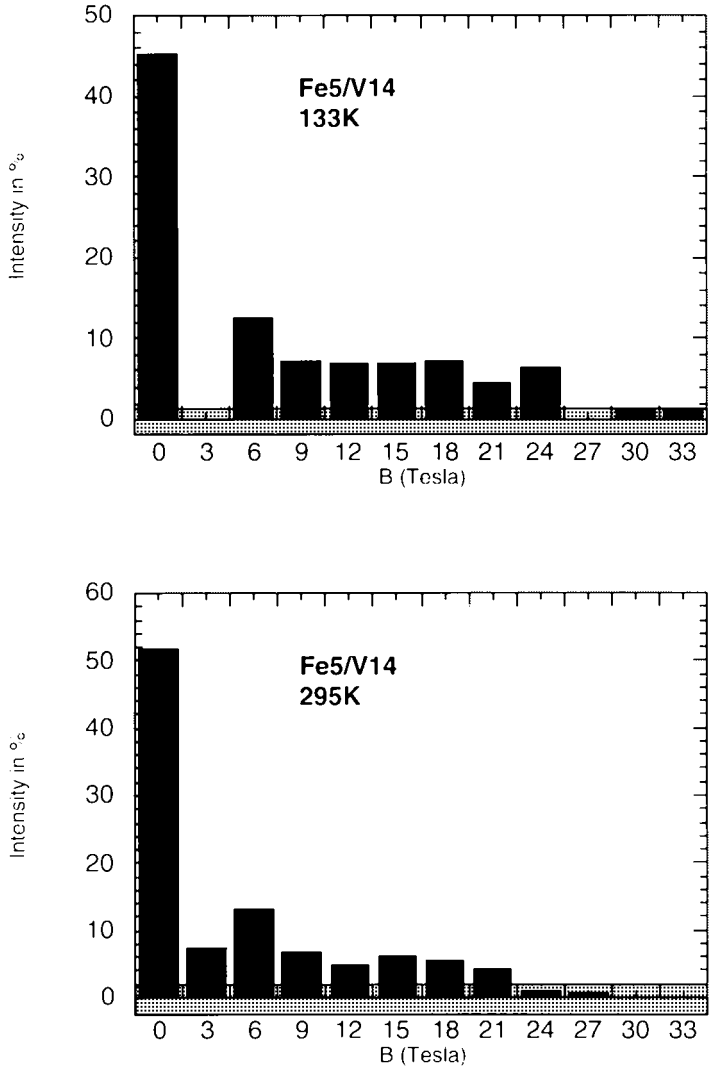

Figure 4. Hyperfine field distributions for 5/14 at 295 K and at 133 K. Shaded region represents the error in intensity.

a wide range of fields ranging from about 25 to 0 T. Using the conversion factor 15 T μ_B^{-1}, the average Fe magnetic moment would be 0.36 μ_B at 295 K and 0.53 μ_B at 133 K.

4.3. Samples with $x \approx 5$ and 10 and $y \approx 5$

The magnetic hyperfine field distributions for these samples (figure 3) are rather broad, ranging from 33 to 0 T. The distribution for the 5/5 sample is more even than that for the 10/5 sample, where a more peaked form is seen towards higher fields. This is natural since the wider the Fe layers, the more bulk-like (closer to 33 T) the magnetic field value will become. For a strict layer-by-layer growth and only taking the near and next neighbour surroundings into account, a limited number of Fe patterns, Fe(i, j), would be observed. Here i represents the number of V

Table 2. Probabilities $P(i, j)$ in per cent of finding Fe in different nearest atomic neighbour surroundings for ideal layer growth. Here i and j represent the number of V atoms as nearest and next nearest neighbours.

Sample	$P(0, 0)$	$P(0, 1)$	$P(4, 1)$
5/5	20%	40%	40%
10/5	60%	20%	20%

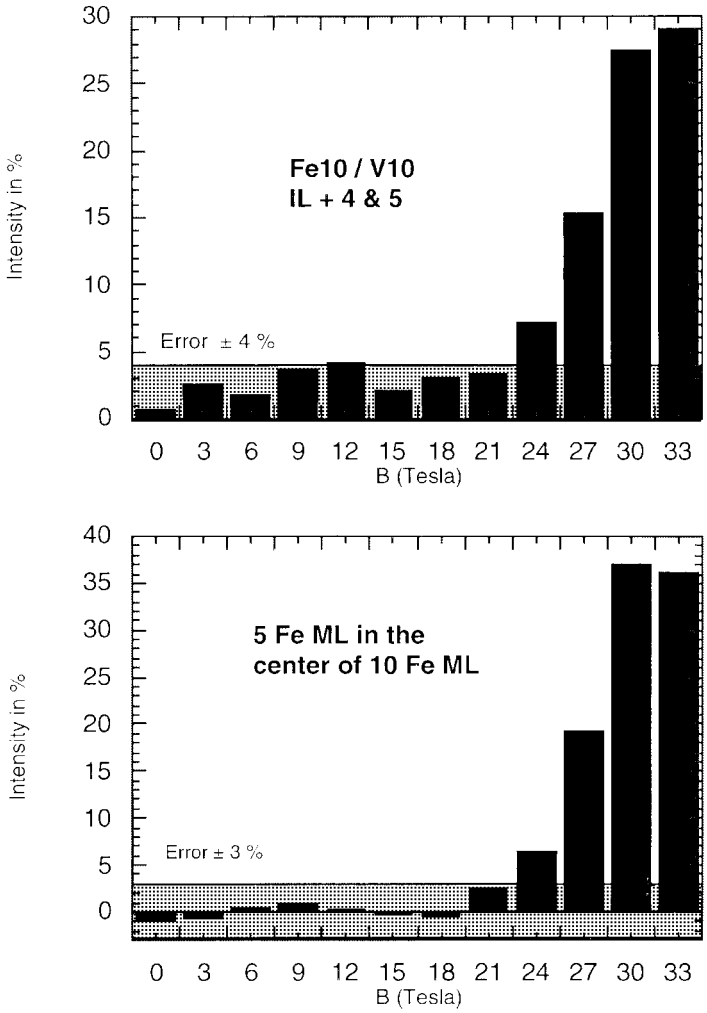

Figure 5. Calculated Fe magnetic field distribution for the innermost layers in the multilayer Fe 10 ML/V 10 ML from (above) the experimental distributions 10/5 and 10/10 (IL + 1, 2 and 3) (below) the experimental distributions 5/5 and 10/5 from table 1.

atoms as near neighbour and j for the number of V atoms as next nearest neighbour. In the case of 5/5 and 10/5 samples, the calculated intensities for such patterns are presented in table 2.

The Mössbauer spectra would, in this model, be a superposition of three patterns each attributed to a certain nearest neighbour surrounding. This is not the case for our spectra.

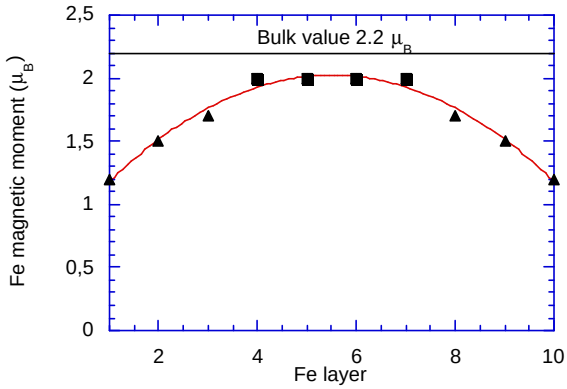

Figure 6. Experimental Fe magnetic moments (▲) at different monolayers in a 10 ML film of Fe interfoliated by 5 to 10 ML of V in a multilattice. The values (■) are deduced from the average hyperfine fields found from figure 5 for the four innermost layers. It is not possible, from our data, to separate the values for the four innermost layers. The moments are calculated using the conversion factor 15 T μ_B^{-1} between the magnetic fields and magnetic moments.

Häggström *et al* [2] found that the growth is not strictly layerwise, but spread out over at least five successive layers. Using this, it is possible to explain the wide distributions found for the magnetic hyperfine fields.

The thickness of the V layers seems to be of limited importance concerning the Fe magnetic hyperfine field distribution of the samples studied. Assuming this distribution to be independent of the number of V layers in the superlattice, it is possible to deduce the summed Fe field distribution of the IL + 4 and IL + 5 layers (see table 1 for definitions) from the results of the 10/5, 10/10 IL + 1, 10/10 IL + 2 and 10/10 IL + 3 spectra. This result is presented in figure 5. Another way to obtain information about the layers in the centre of the 10 ML Fe sample is to subtract the field distributions obtained for the 5/5 sample from those for the 10/5 sample, taking into account that the intensities should add up to 100%. This result is also presented in figure 5. These two different approaches give nearly the same result. The errors in spectral intensities in this type of analysis are, of course, rather large, being ±4% and ±3% respectively. Within these errors we do not observe hyperfine fields below 24 T and we arrive at an average field and magnetic moment for the four to five innermost layers in a 10 ML film of Fe of about 30.1 T and 2.0 μ_B, respectively, using the factor 15 T μ_B^{-1}.

This could be compared with the corresponding values from the renewed analysis of the IL + 1, IL + 2 and IL + 3 spectra, giving 18.1 T (1.2 μ_B), 22.4 T (1.5 μ_B) and 25.0 T (1.7 μ_B) respectively (figure 6). This analysis shows that the bulk value (33 T or 2.2 μ_B) is not found even as an average value in the middle of a 10 ML thick Fe layer at 295 K, indicating that the Curie temperature must be substantially lower than the bulk value.

5. Conclusions

The main conclusions follow.

(1) Changes in the magnetic field distribution for the 5/14 sample as a function of temperature were determined. For samples with $x < 5$ no magnetic part was observed in the temperature range 133–300 K.
(2) The magnetic field distribution for the innermost layers in 10/5 was estimated. The

average field was found to be markedly lower than the bulk value.

(3) A strong dependence of the average magnetic hyperfine field with the Fe layer thickness was observed.

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