Hyperfine Interact (2006) 169:1371–1377
DOI 10.1007/s10751-006-9453-8

# Mössbauer spectroscopical investigation of the exchange biased $Fe/MnF_2$ interface

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Published online: 27 October 2006


**Abstract** Two different $Fe/MnF_2$ samples have been prepared by e-beam evaporation on MgO(001) substrates. The Fe layer in the samples includes a 10 Å thick $^{57}Fe$ probe layer either at the $Fe/MnF_2$ interface (interface sample) or 35 Å away from the interface (center sample). The samples are characterized by X-ray diffraction,

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conversion electron Mössbauer spectroscopy (CEMS) and SQUID magnetometry. $^{57}Fe$ CEMS has been employed to study the depth dependent hyperfine interactions in $Fe/MnF_2$ as a function of temperature between 18 K to 300 K. The hyperfine field $B_{hf}$ has been obtained for the interfacial and off-interfacial $^{57}Fe$ layers. At the interface, besides $B_{hf}$ of bcc-Fe, the presence of a component with a distribution $P(B_{hf})$ is observed. The latter is assigned to interfacial $^{57}Fe$ atoms, indicating some (∼15%, equivalent to ∼1 Fe atomic layer) intermixing at the $Fe/MnF_2$ interface and a decrease of the average $<B_{hf}>$ by 21%. The influence of the interface disappears as the $^{57}Fe$ probe layer is placed away from the interface. The temperature dependence of the average $<B_{hf}>$ of the interface has been measured. The Fe spins, at remanence, are found to lie in the film plane.

**Key words** conversion electron Mössbauer spectroscopy · exchange bias · $Fe/MnF_2$ · interfacial properties · spin structure

## 1 Introduction

Bilayers of $Fe/MnF_2$ are an archetype system showing exchange bias effect [1–15] evidenced by the shift of the magnetic hysteresis loop. This effect originates from the exchange coupling at the interface of the ferromagnet (Fe) and antiferromagnet ($MnF_2$). The roughness and the intermixing at the interface are among the important parameters which can control the interfacial coupling and hence the exchange bias [4–6]. Therefore, for the understanding of the exchange bias effect, information on interface parameters such as magnetic moment or hyperfine field, interdiffusion, etc. are highly desirable along with the interfacial spin structure. $^{57}Fe$ conversion electron Mössbauer spectroscopy (CEMS) in combination with the $^{57}Fe$ probe layer method is a unique isotope selective technique in order to explore depth-dependent physical properties such as spin structure [7], metallurgical and magnetic phases present in the sample, temperature dependent magnetic ordering and local magnetic moment at the interface. The change in the remanent spin structure in this exchange biased systems below and above the Néel temperature ($T_N = 67$ K) has been reported earlier [7]. In this work the CEMS investigation of the interfacial parameters of $Fe/MnF_2$ bilayers will be discussed.

## 2 Sample preparation and characterisation

Two kinds of exchange biased $Al/Fe/MnF_2/ZnF_2/MgO(001)$ heterostructures were prepared by sequential electron beam evaporation [8]. In both kinds of samples a 160 Å $ZnF_2$ buffer layer was deposited in order to relax the large (8%) lattice mismatch between the MgO substrate and $MnF_2$. The typical thicknesses of the Al, Fe and $MnF_2$ layers are 30, 80 and 520 Å, respectively. The samples differ only by their ferromagnetic Fe layer. Out of the 80 Å Fe layer, a 10 Å $^{57}Fe$ probe layer was deposited just at the interface between the AFM ($MnF_2$) and the FM (Fe) layer in the first kind of sample called 'interface sample' (labeled MFEMF01). Hence, the FM layer for the interface sample contains 70 Å $^{nat}Fe$/10 Å $^{57}Fe$. The other kind of sample is called 'center sample' (labeled MFEMF03). In this sample the $^{57}Fe$ probe

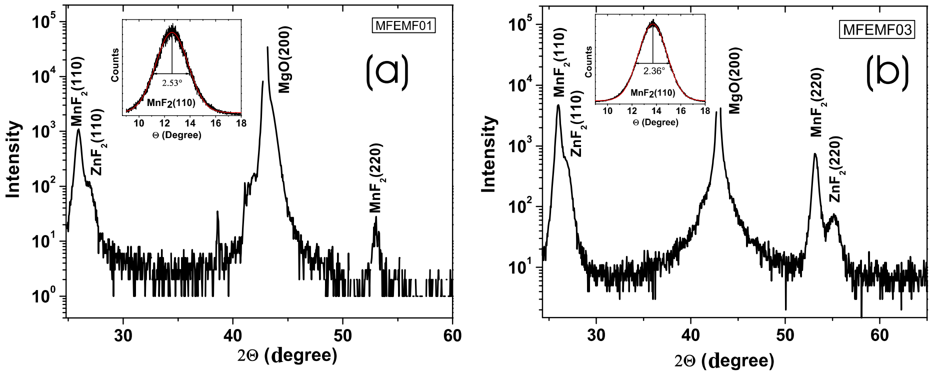


**Figure 1** XRD pattern of Fe/$MnF_2$ interface sample (**a**) and center sample (**b**). *Inserts* show the corresponding *rocking curves* taken at the $MnF_2(110)$ peak. (Cu–$K_\alpha$ radiation).

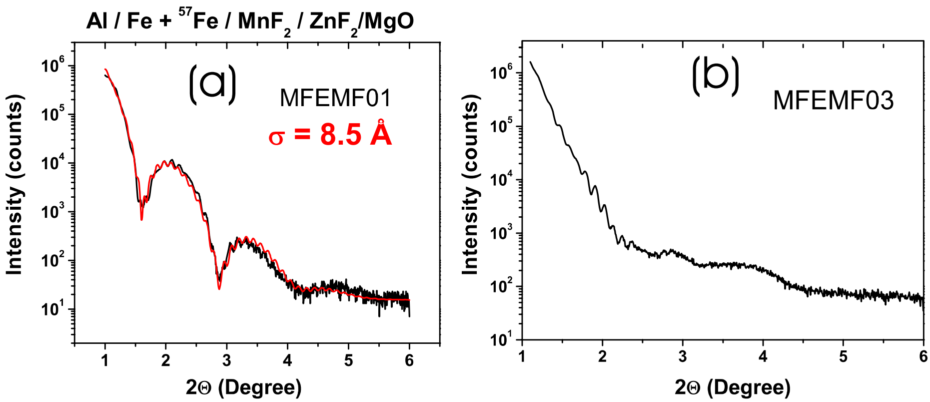


**Figure 2** Small-angle XRD scans of Fe/$MnF_2$ interface sample (**a**) and center sample (**b**). The *light-gray curve* (*red, in color*) is the fitting to the measured curve in (**a**). (Cu–$K_\alpha$ radiation).

layer was placed at the center of the Fe layer (35 Å$^{nat}$Fe/ 10 Å$^{57}$Fe/35 Å$^{nat}$Fe), i.e. 35 Å away from the interface. Prior to deposition the MgO(001) substrate was heated to 450°C for 15 min and then cooled to 200°C for $ZnF_2$ deposition. The base pressure of the system was $3 \times 10^{-8}$ Torr and the pressure during $MnF_2$ deposition was around $6 \times 10^{-7}$ Torr. The deposition temperatures, and deposition rates for Al, Fe, $MnF_2$ and $ZnF_2$ layers were 150, 150, 325 and 200°C, and 0.5, 1, 2 and 2 Å/s, respectively. The thickness of the fluoride layers was monitored by calibrated quartz crystal oscillators and the Fe layer thickness was monitored by optical sensors.

After deposition the structural and magnetic characterizations of the samples were performed by high and small angle XRD (Cu–$K_\alpha$ radiation, $\lambda$ = 1.5418 Å), CEMS and SQUID magnetometry. The high-angle and small-angle XRD patterns for the interface sample are shown in Figures 1a and 2a, respectively. The high-angle XRD pattern confirms the epitaxial nature of the film with $MnF_2(110)$ in the sample plane.

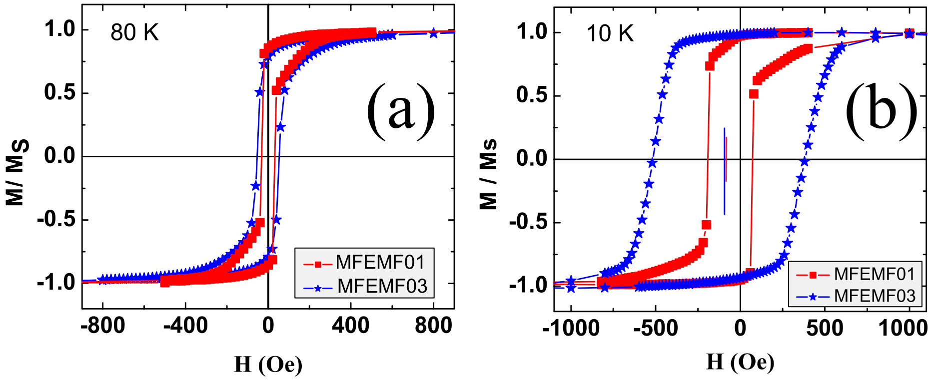


**Figure 3** The normalized SQUID hysteresis loops for the interface sample (*squares*) and center sample (*asterisks*) taken at 80 K (**a**) and 10 K (**b**).

The relatively sharp rocking curve (insert in Figure 1a) of the $MnF_2(110)$ reflection has a full width at half maximum (FWHM) of about 2.5°. The small-angle XRD suggests good homogeneity of the $MnF_2$ and Fe layer. The high frequency (low frequency) oscillations are due to the fluoride layer (Fe layer). The light-gray curve is the least-squares fitting to the measured black curve. The fitting gives the interface roughness $\sigma$ of about 8.5 Å. The epitaxial nature of the center sample has also been verified by the high-angle XRD pattern shown in Figure 1b. The relatively sharp rocking curve (insert, Figure 1b) has a FWHM of about 2.4°. In comparison to the interface sample, the small angle scattering (Figure 2b) indicates a higher roughness for the center sample. It is worth mentioning that the $MnF_2$ layers (for interface and center sample) are growing as twinned epitaxial layers [10–13].

## 3 SQUID magnetometry results

SQUID magnetometry has been used to measure the exchange bias field, $H_E$, and the coercivity, $H_c$ for the two samples. The results are shown in Figure 3a and b. Prior to the measurement the sample was cooled from 150 K to either 80 or 10 K in an in-plane applied field of 2 kOe. Clearly the samples show no exchange bias above $T_N$ (at 80 K). However, below $T_N$ (at 10 K), $H_E$ of about 55 Oe has been observed for both samples. It should be noticed that $H_c$ of the two samples is different, which is associated with the different microstructure (roughness and intermixing) of the two samples.

## 4 CEMS results and discussion

The samples were also characterized by $^{57}$Fe CEMS with the $\gamma$-ray either normal to the sample plane ($\Phi = 90°$) or at an angle $\Phi = 45°$. Typical Mössbauer spectra for the interface sample (MFEMF01) taken at different temperatures are given in Figure 4. Each spectrum clearly shows a dominant six-line pattern superimposed to

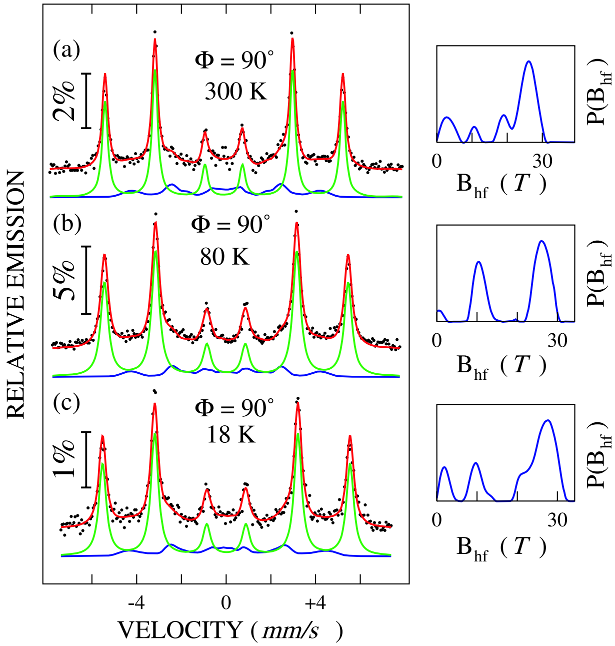


**Figure 4** CEM spectra of the Fe/$MnF_2$ interface sample taken at 300 K (**a**), 80 K (**b**) and 18 K (**c**). The spectra have been fitted by a dominant sextet assigned to bcc-Fe, and a component with a distribution of hyperfine fields, $P(B_{hf})$ (*right-hand side*) which accounts for the chemical intermixing at the interface.(Angle between $\gamma$-ray direction and film plane $\Phi = 90°$).

a weak distribution of magnetic hyperfine fields. Each spectrum has been fitted by a dominant sextet ($B_{hf} = 32.8$ T) and a subspectrum with distribution of hyperfine fields $P(B_{hf})$. The dominant sextet is unambiguously assigned to the bcc-Fe layer, and the distribution of hyperfine fields, which has a dominant peak at about 26 T, is attributed to the chemical intermixing at the Fe/$MnF_2$ interface. The typical relative spectral area of the hyperfine field distribution is about 15% of the total area. Considering the $^{57}$Fe layer thickness (10 Å), 15% intermixing corresponds about 1.5 Å of the $^{57}$Fe layer, i.e., about one monolayer (ML) of Fe is chemically intermixed at the interface. It should be noticed that the roughness of 8.5 Å obtained from the simulation of the small-angle XRD results (Figure 2) is larger than the 1.5 Å intermixing at the interface. The CEM spectra (not shown) measured with the $\gamma$-ray incident angle of $\Phi = 45°$ with respect to the film plane resulted in similar hyperfine parameters (isomer shift, magnetic hyperfine field, and spectral area) as those obtained at $\Phi = 90°$. The isomer shifts, magnetic hyperfine fields and the spectral area contribution for both, the dominant sextet and the distribution $P(B_{hf})$, are shown in Figure 5. It is worth mentioning that in all cases the average isomer shifts of the $P(B_{hf})$ distribution are very small and similar to that of the dominant sextet.

For the center sample, the CEM spectra (Figure 6) are measured in the same way as that for the interface sample. The spectra have been least-squares fitted by using a single sextet typical of bcc Fe. No interfacial intermixing has been observed because the $^{57}$Fe-probe layer is about 35 Å away from the AFM/FM interface. It is interesting to note that the intensity ratio between the second and third line ($R_{23}$) of the dominant Mössbauer sextet (for $\Phi = 90°$) is 4.0 for both samples. This indicates that the Fe spins are in the plane of the sample at all temperatures between 300 K and 18 K.

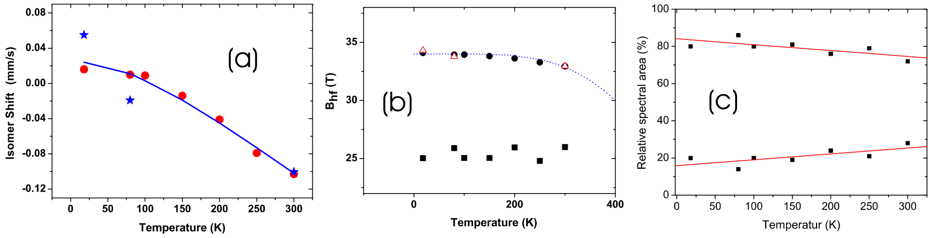


**Figure 5** Temperature dependence of: (**a**) isomer shift of the bcc-Fe phase relative to the $^{57}Co(Rh)$ source (*full dots*: $\Phi = 45°$, *asterisks*: $\Phi = 90°$). The *solid line* is a Debye model fit to the data yielding a Debye temperature $\Theta_D = 466$ K.; (**b**) magnetic hyperfine field of the bcc-Fe phase (*circles*: $\Phi = 45°$ and *triangles*: $\Phi = 90°$) and of the component with the $P(B_{hf})$ distribution (*average hyperfine field, squares*). The *dotted curve* is a Brillouin function (J = 1/2) fit to the data points yielding $T_C = 627$ K for the Fe layer; (**c**) relative spectral area of the bcc-Fe sextet (*top*) and of the component with the $P(B_{hf})$ distribution (*bottom*). The *straight lines* are linear fits to the data points.

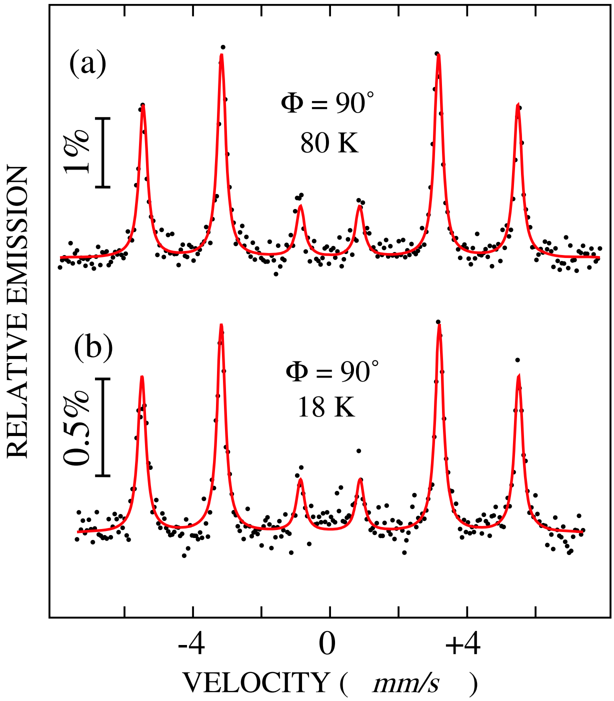


**Figure 6** CEM spectra of the $Fe/MnF_2$ center sample taken at 80 K (**a**) and 18 K (**b**). The spectra have been fitted by a sextet assigned to bcc-Fe. (Angle between $\gamma$-ray direction and film plane $\Phi = 90°$).

## 5 Summary

In summary, we have prepared two different $Fe/MnF_2$ samples with an $^{57}Fe$-probe layer at and away from the AFM/FM interface, respectively. The structure, magnetic properties and hyperfine parameters have been obtained by using XRD, SQUID magnetometry and CEMS. Our CEMS results indicate very small interfacial intermixing, equivalent to ∼1 ML of Fe, a reduced interfacial hyperfine field, and fully in-plane magnetization of the Fe films.

**Acknowledgements** Work supported by the DFG (SFB 491), the US-DOE, the US–Israel BSF, the CNPq (Brazil), the AvH Foundation, the Spanish CICYT (MAT2001-2555), the Alfred P. Sloan Foundation, and the Catalan DGR (2001SGR00189). W.A.A.M. gratefully acknowledges the hospitality during his stay at the UCSD, and thanks J. Santamaria and R. Paniago for assistance with GIXR simulations.

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