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Spin configurations in exchange coupled magnetic phases studied by Mössbauer spectroscopy

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The ability of the ^{57}Fe Mössbauer technique to reveal the spin structures in exchange coupled magnetic phases containing iron is emphasized. Experimental results obtained on layered exchange-bias systems are presented. The influence of the ferromagnetic top layer on the out-of-plane spin component of the pinning antiferromagnetic-like layer, in both Fe/FeSn$_2$ and Fe/Fe-Gd-B exchange bias systems, is discussed.

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1. Introduction

The pinning effects at the interface of magnetic phases with different magnetic anisotropy give rise to interesting magnetic phenomena. Among them, due to both theoretical and practical challenges, a key attention is presently paid to exchange-bias and exchange-spring phenomena.

Exchange bias phenomena are related to the unidirectional anisotropy induced at the interface between a soft ferromagnetic (FM) and an antiferromagnetic (AF) material with high magnetic anisotropy, when the system is prepared in a magnetic field or cooled down in an applied magnetic field below the Néel temperature, T_N, of the AF phase. The effect was discovered more than four decades ago, in 1956 [1]. Recent applications of exchange biased layered systems, especially as “spin valve” devices [2-5] have renewed the interest in this effect and its underlying physics. Intensive experimental and theoretical work was done in the last years, but due to its complexity, both the microscopic origin and the role of the different parameters involved in the exchange-bias mechanism (anisotropy, roughness, magnetic domains, etc.), is far from being clarified. The knowledge of the real spin structure at the interface between the exchange coupled magnetic phases represents an important issue for the correct exploitation of the theoretical models and the final understanding of the involved microscopic mechanisms. Unfortunately, up to now, due to the low number of suitable experimental techniques, there is a lack of experimental studies evidencing the interfacial local spin structure in such systems.

In the present work we emphasize the capabilities of ^{57}Fe Mössbauer spectroscopy to reveal the interfacial spin structure of the exchange coupled magnetic phases containing iron. Some exemplifications on layered exchange-bias systems are presented.

2. Interfacial spin structure revealed by ^{57}Fe Mössbauer spectroscopy

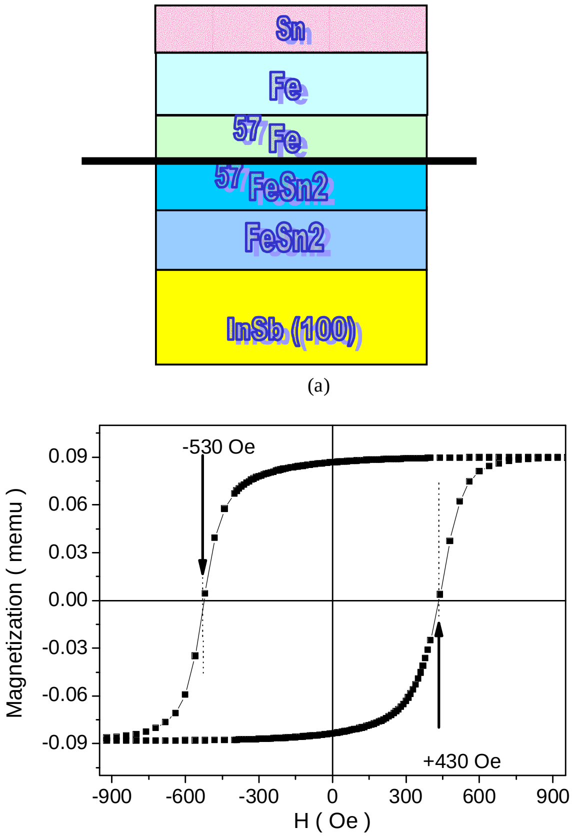
Among other information, ^{57}Fe Mössbauer spectroscopy provides direct information about the iron phase composition of the sample. This is related to the number and the relative contributions of the different components (subspectra assigned to various phases or non-equivalent Fe sites) in the experimental Mössbauer pattern. Compared to other investigation methods, the Mössbauer spectroscopy is able to give simultaneous information about the local interactions and symmetry as well as on the local spin structure on each of the constituent metallurgical phases of the sample. A magnetically ordered phase is revealed in the Mössbauer spectrum by a Zeeman split six-line pattern (sextet). The magnetic splitting (quantified by the magnitude of the hyperfine magnetic field) is proportional to the magnitude of the Fe magnetic moment, whereas the relative intensity of the second and fifth Mössbauer line is influenced by the direction of the local magnetic moment (Fe spin) with respect to the direction of the γ- radiation. Therefore, the only limitation for a suitable study of the spin structure by Mössbauer spectroscopy is that the analyzed samples should contain iron atoms with reasonably large effective (time averaged) magnetic moments, giving rise to a reasonably resolved sextet in order to reveal properly the ratios of the line intensities in the Mössbauer sextet. Quantitatively, the spin configuration may be analyzed starting from the intensity ratio between the second (or fifth line) and the third (or the forth) line of the sextet component: $R_{23}=4\sin^2\theta/(1+\cos^2\theta)$ with θ being the angle between the Fe spin direction and the γ-ray direction. In a perpendicular geometry, the above relation can be used also in the form $R_{23}(\alpha)=4\cos^2\alpha/(2-\cos^2\alpha)$, with α being the angle between the spin direction and the sample plane. The case of $R_{23}=4$ shows fully in plane orientated spins, whereas for R_{23}=0 the spins are oriented perpendicular to the sample plane. In the real case, there is often a wide spread of the spin orientation. Theoretical angular distribution functions for the spin orientation may be proposed as convenient approximations for the real situations. The unknown parameters of the theoretical distributions can be estimated from the suitable computations of the intensity ratio R_{23} and their fit to the experimental values [6]. However, such analyses are beyond the aim of this paper, which deals only with the demonstration of the Mossbauer capability to reveal changes in the angular spin distribution. Briefly speaking, the modifications in the orientation spin distribution can be followed for each magnetic phase by the observation of the intensity ratio between the second and the third line of the sextet: a value of 2 for the ratio R_{23} points to a random spin orientation while $0<R_{23}<2$ points to a preferred out-of-plane and $2<R_{23}<4$ for a preferred in-plane spin orientation. Such modifications can be analyzed also with high depth selectivity by using the ^{57}Fe tracer layer technique [7]. This technique is mainly used in thin films or multilayers, where the Mössbauer events are counted by conversion electrons (the corresponding surface sensitive characterization tool is called Conversion Electron Mössbauer Spectroscopy). The tracer layer technique consists in high enrichment of the layer of interest in the ^{57}Fe Mossbauer isotope. A layer with the same phase composition as the main layer, but enriched in ^{57}Fe (natural abundance of 2%), is artificially deposited at the desired depth or at the interface. The Mössbauer signal is provided mainly from the enriched layer, resulting in a depth selective information with a resolution limit of about 1 nm. This confers to Mössbauer spectroscopy excellent capabilities in revealing the surface and interface properties through the intensity ratio of the sextet components in the Mössbauer spectra. Some examples are presented in the following.

3. Experimental

Two different multilayer systems presenting exchange bias effects at low temperatures were prepared and subsequently analyzed by Conversion Electron Mössbauer Spectroscopy (CEMS).

The first system is a multilayer grown by Molecular Beam Epitaxy (MBE) on an InSb substrate, in the sequence: 3 nm Sn / 4.8 nm Fe / 1.2 nm ^{57}Fe / 5 nm $^{57}FeSn_2$ / 20 nm $FeSn_2$ / InSb(001) (sample A1). In principle, it consists in an Fe/$FeSn_2$ bilayer (Fe is the ferromagnetic (F) layer and $FeSn_2$ the antiferromagnetic (AF) one). A tracer layer of 1.2 nm of ^{57}Fe was introduced in the ferromagnetic part and a tracer layer of 5nm of $^{57}FeSn_2$ in the antiferromagnetic part of the ferromagnetic/antiferromagnetic interface (Fig. 1 (a)). The preparation conditions of such systems were presented elswere [8,9]. The low temperature hysteresis loop at 5 K shows an exchange bias field (the shift of the hysteresis loop relative to H=0 Oe) of about -50 Oe, as shown in Fig. 1 (b). In order to study the influence of the Fe top layer on the spin structure of the antiferromagnetic phase, another system was prepared, consisting only in a Sn capped antiferromagnetic phase with a 5 nm $^{57}FeSn_2$ tracer layer, in the following geometry: 3 nm Sn / 5 nm $^{57}FeSn_2$ / 28 nm $FeSn_2$ / InSb(001) (sample A2).

A second exchange bias system was prepared by r.f. sputtering. A 9 nm Fe layer was used as a top ferromagnetic layer and a 30 nm amorphous phase was used as a quasi-antiferromagnetic pinning layer (sample A3 with geometry 15 nm SiO_2/9 nm Fe/30 nm $(^{57}Fe_{77}Gd_{23})_{83}B_{17}$/glass substrate). In this case, the pinning layer consists of a ferrimagnetic amorphous compound close to the compensation point, namely $(Fe_{77}Gd_{23})_{83}B_{17}$. From the magnetic point of view it behaves similarly to an antiferromagnetic phase (no contribution to the system magnetization), but in view of the pinning effects it could be interesting because of the opposite effects of high interfacial Gd atomic moments and low anisotropy constant of the amorphous compound. This system presents an exchange bias field of about -20 Oe at 5 K.



(a)

(b)

Fig. 1. The geometry of the Fe/FeSn$_2$ exchange bias system (a) and the corresponding hysteresis loop, obtained at 5 K after cooling the sample through the Neel temperature of the antiferromagnetic phase in a field of 1000 Oe (b).

Only the amorphous antiferromagnetic-like layer was uniformly enriched in ^{57}Fe. To see the influence of the Fe top layer on the spin structure of the amorphous pinning layer, another sample consisting only in the SiO_2 capped amorphous layer (30 nm thickness) at the compensation point was also prepared (sample A4, with geometry 15 nm SiO_2/30 nm ($^{57}Fe_{77}Gd_{23})_{83}B_{17}$/glass substrate). Both samples A3 and A4 were covered by a 15 nm SiO_2 cap layer.

The spin structure of all samples was analyzed at room temperature (RT) by CEMS.

4. Results and discussion

The Conversion Electron Mössbauer (CEM) spectra of the first two samples with $FeSn_2$ as antiferromagnetic phase (the F/AF and the AF type, respectively) are shown in Figure 2.

The four different Mössbauer components (three sextets and a broad paramagnetic singlet) of the F/AF system (sample A1) originates mainly from the two interfacial tracer layers at the F/AF interface. On the other hand, the two components (a sextet and a broad singlet) in the Mössbauer pattern of the AF system (sample A2), has to originate mainly from the $^{57}FeSn_2$ tracer layer at the surface of the AF phase. According to our previous studies [8,9], the unique sextet in the Mössbauer spectrum of the AF sample and the inner sextet in the spectrum of the F/AF sample presents hyperfine parameters specific to a well ordered $FeSn_2$ phase, whereas the broad singlet in each sample is attributed to pathways of a defects containing phase, with an inverse occupation of the Fe and Sn sites in the tetragonal structure of the $FeSn_2$. Taking into account these observations and the different geometrical sequences of the two samples, the outer sextets in the Mössbauer spectrum of the F/AF sample have to be assigned to the 1 nm thick interfacial ^{57}Fe layer of this structure.

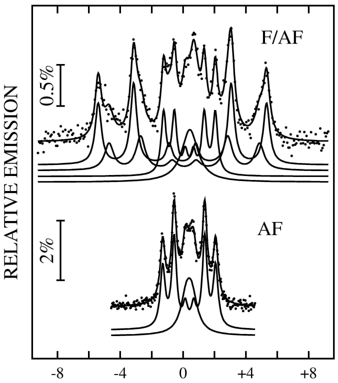


Fig. 2. The CEM spectra collected at RT on layered systems with $FeSn_2$ as antiferromagnetic phase (F/AF = ferromagnetic/antiferromagnetic exchange coupled structure (sample A1), AF = only the antiferromagnetic structure (sample A2)). Tracer layers enriched in ^{57}Fe are introduced at the F/AF interface or at the surface of the AF layer.

Indeed, the values of the hyperfine parameters confirm the above assignation and show that the outer sextet with lower intensity can be assigned to an about 0.4 nm thick interfacial (and defects-containing) layer of metallic Fe, whereas the outer sextet with a higher intensity corresponds to the following 0.6 nm thick defect-free, nearly bulk-like bcc interfacial layer of Fe. As observed from the above data, the interfacial spin distribution can be obtained in this situation with very good depth selectivity for each of the involved phases. The values of the R_{23} ratios for the two outer sextets (R_{23} close to 4) shows that the Fe spins in the ferromagnetic top layer are almost in the film plane, even at the very interface with the antiferromagnetic phase. Concerning the antiferromagnetic spins, a much lower value of measured intensity ratio (R_{23} about 2.8) corresponds to the interfacial (5 nm thick) $FeSn_2$ layer in the F/AF system. Therefore, the spin configuration of the AF phase in the F/AF system (sample A1), shows a pronounced out-of-plane component. A different situation appears in sample A2 (only AF as magnetic phase), where the sextet corresponding to the surface $FeSn_2$ layer shows an intensity ratio R_{23} close to 4, in agreement with an in-plane configuration of the Fe spins in the antiferromagnetic layer. That is, the presence of the top Fe layer induces a reorientation of the interfacial spins of the antiferromagnetic layer, with expected influence on the exchange coupling effect.

The Mössbauer spectra of the layered systems with $(Fe_{77}Gd_{23})_{83}B_{17}$ as pinning antiferromagnetic-like phase are presented in Fig. 3. In both cases, the Mössbauer signal should come predominantly from the 30 nm amorphous pinning phase, the only one which was partially (30%) enriched in the ^{57}Fe Mössbauer isotope.

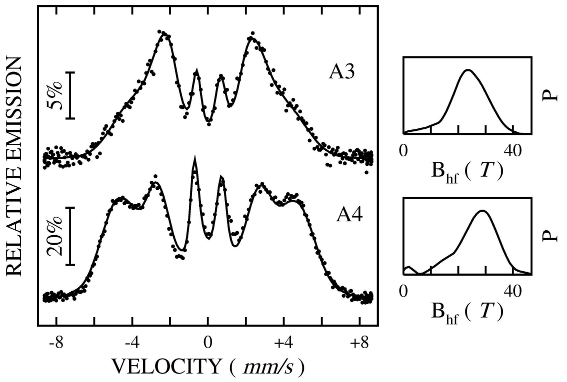


Fig. 3. The CEM spectra collected at RT on layered systems with $(Fe_{77}Gd_{23})_{83}B_{17}$ as pinning antiferromagnetic-like phase (A3 = ferromagnetic /amorphous antiferromagnetic-like structure, A4 = only amourphous antiferromagnetic-like structure). On the right side are presented the corresponding hyperfine magnetic field distributions.

The Mössbauer patterns are specific to an amorphous phase and therefore the spectra were least-squares fitted with a distribution of hyperfine magnetic fields, by keeping the common R_{23} ratio for the elementary sextets as free parameter. The fitted R_{23} value changes from about 3.0 in sample A3 to about 1.4 in sample A4 (a drastic decrease of the relative intensity of the second line in the Mössbauer spectrum of sample A4 is also observed with the naked eye). The presence of the top Fe layer in this system induces a reorientation of the spins in the whole amorphous pinning layer toward the in-plane direction, contrary to the previous case (samples A1 and A2).

Such reorientation processes of the interfacial spins, induced by the presence of the second component of an exchange coupled system might take place very often. They should influence the exchange coupling strength. Tentatively, the spin reorientation processes might be attributed to magetoelastic effects exerted at the interface between phases with different lattice constants.

4. Conclusions

The surface and interface spin configuration of iron-containing phases may be obtained with excellent depth selectivity by Mössbauer spectroscopy. In optimal situations, the depth selectivity can decrease well below 1 nm. In addition, the spin reorientation process in layered systems can be also observed by varying the temperature or the applied magnetic fields. This technique was successfully applied for the study of the interfacial spin structure in exchange coupled magnetic layers.

The top ferromagnetic layer has been shown to change the interfacial spin configuration of the antiferromagnetic layer in exchange-bias F/AF systems. Both, out-of-plane or in-plane spin reorientation processes can take place, depending on the type of the pinning layer. Magnetoelastic effects might be involved in this microscopic behavior.

The real spin structure at the ferromagnetic /antiferromagnetic interface has to be considered in material-specific theoretical models for exchange-bias.

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