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LETTER TO THE EDITOR

Temperature dependence in the magnetization reversal process of $DyFe_2/YFe_2$ exchange-coupled superlattices

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Abstract

X-ray magnetic circular dichroism (XMCD) measurements have been performed to study the magnetization reversal of a $[DyFe_2(5\ nm)/YFe_2(20\ nm)]_{13}$ superlattice that exhibits a negative interface exchange coupling. By recording compound-specific hysteresis loops, we have shown a strong temperature dependence in this magnetization reversal process: below 150 K interface magnetic twists develop as expected predominantly in the softer YFe_2 layers, whereas above 200 K the magnetization reversal first affects the harder $DyFe_2$ compound. The sharp transition between these two regimes accounts for the variation of the coercive field, from a positive to a negative value. Different possible mechanisms for the first reversal of the hard compound are discussed.

The magnetic study of the $REFe_2$ Laves phase superlattices is of great interest for potential applications in permanent magnets [1] or magnetic sensors [2], as well as for the theoretical understanding of the magnetic coupling in these materials. These superlattices are among the few single-crystalline superlattices that exhibit spring magnet behaviour [3–5], most of the other systems being either textured polycrystalline [6, 7] or amorphous [8] materials, or consisting of randomly oriented magnetically hard grains embedded in a soft matrix [9]. The Laves phase superlattices constitute therefore a model system for the study of magnetic springs, of exchange bias phenomena, and of the magnetization reversal process in exchange coupled systems. This latter process is still a debated issue, as proved by the number of recent studies reported on magnetization reversal of in-plane- [7] and perpendicularly biased systems [10, 11].

The main magnetic characteristics of the $DyFe_2/YFe_2$ system are the following. (i) $DyFe_2$ is a ferrimagnet with a resultant magnetization along the dominant Dy subnet magnetization.

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Because of the shape of the 4f electron cloud of Dy, $DyFe_2$ is a hard magnetic material. (ii) YFe_2 is also ferrimagnetic, but the yttrium site only has a small induced moment [12]. Its anisotropy, which originates from iron moments, is weak and YFe_2 is a soft magnetic material. (iii) Finally, the magnetic coupling at the $DyFe_2/YFe_2$ interfaces occurs mainly through the ferromagnetic coupling between iron spins, which results in a global antiparallel coupling between the magnetizations of the $DyFe_2$ and YFe_2 layers. $DyFe_2/YFe_2$ superlattices are thus composite systems of hard and soft magnetic layers, antiferromagnetically coupled at their interfaces.

The simplest scenario to describe the magnetization reversal in such hard/soft coupled systems is usually the formation of exchange springs in the soft material, before the irreversible switch of the hard material [4]. Such a reversal mechanism, referred to as 'soft first' (SF) in the following, has been previously observed [3, 5] in several $DyFe_2/YFe_2$ superlattices. However, some recent studies, based on the chemical selectivity of resonant soft x-ray magneto-optical Kerr effect and of x-ray magnetic circular dichroism (XMCD) [6, 10, 13], have shown that this oversimplified scenario of the magnetization reversal in coupled systems is likely too crude a description. On the other hand, Bentall *et al* [14] recently showed by neutron scattering that canted states could occur in superlattices when the total net moment of the $DyFe_2$ layer is very close to the total moment of the YFe_2 layers.

In this letter, we focus on a $[DyFe_2(5\ nm)/YFe_2(20\ nm)]_{13}$ superlattice for which macroscopic magnetization measurements suggest a strong temperature dependence of the magnetization reversal. In this sample, the YFe_2 magnetization is dominant at all temperatures since the magnetic moment per formula unit decreases from 6.4 μ_B at 10 K to 3.9 μ_B at room temperature in $DyFe_2$ [14], while it remains close to 2.8 μ_B in YFe_2 [15]. Field variations of the $DyFe_2$ and YFe_2 magnetization in the 10–300 K temperature range have been probed selectively by XMCD at the Dy and Y L_3 absorption edges, respectively. Although XMCD only provides depth-averaged information and not a layer-resolved magnetic profile, it nevertheless allows one to separate magnetic contributions from the two kinds of layers and thus provides deeper insight into the magnetization reversal mechanism.

The results show that the magnetization reversal process changes drastically with temperature. It evolves from a 'soft first' mechanism below 150 K to a very unusual mechanism above 200 K, where the magnetization reversal *first* affects the *hard* $DyFe_2$ layers, while the soft YFe_2 magnetization remains aligned with the applied field direction. This specific mechanism will be referred to as 'hard first' (HF) in the following. A sharp transition between these two processes has been observed, that accounts for the thermal evolution of coercivity from a positive to a negative value.

The sample was prepared by molecular beam epitaxy on a $(11\bar{2}0)$ sapphire substrate. A 500 Å thick (110) niobium buffer layer was first deposited and covered by a very thin iron film (15 Å) to initiate the subsequent RFe_2 epitaxy (R = Y or Dy) [16]. The $DyFe_2$ and YFe_2 layers were obtained by alternative co-deposition of Fe and Dy, and of Fe and Y, respectively. The sample was finally coated with a 200 Å thick yttrium layer to prevent oxidation.

In situ RHEED analysis reveals that the $REFe_2$ compounds grow along the [110] direction of the expected Laves phase structure, with a high crystal quality and a rather smooth surface [3]. X-ray scattering experiments and high resolution transmission electron microscopy observations confirm the periodicity of the stacking, the high degree of chemical modulation and rather flat interfaces.

The XMCD experiments were performed at the European Synchrotron Radiation Facility (ESRF) in Grenoble (France) on the ID12 beamline [13]. A grazing incident geometry has been

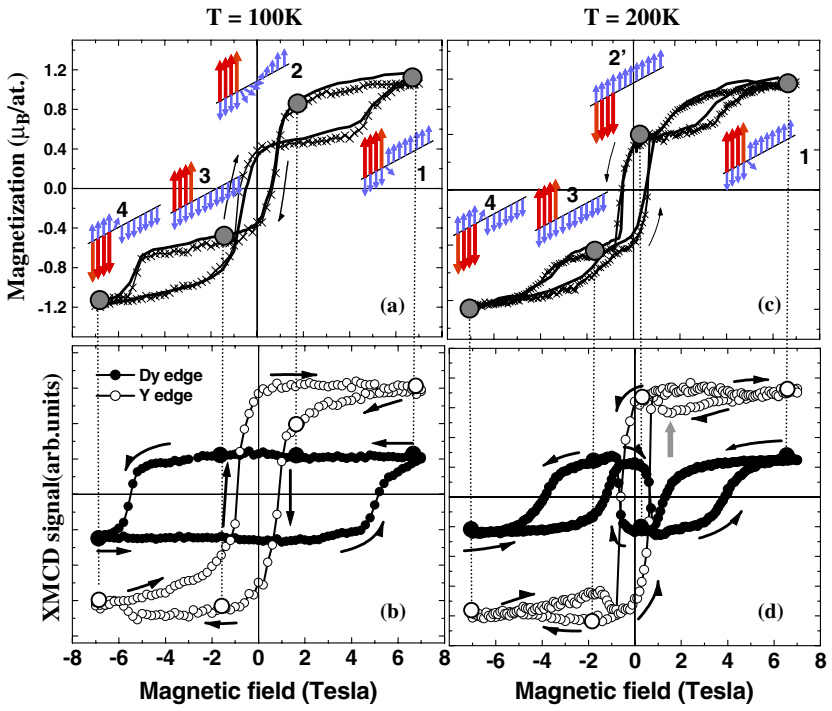

Figure 1. Measurements performed at 100 and at 200 K for the superlattice $[DyFe_2(5\ nm)/YFe_2(20\ nm)]_{13}$. Top curves ((a) and (c)) are macroscopic magnetization measurements (solid curves) superimposed on a weighted sum of the XMCD loops measured at the Y and Dy L_3 edges (crosses). Bottom loops ((b) and (d)) are XMCD results measured at the Dy (filled circles) and Y (empty circles) edges. The schemes represent the magnetic configurations of one bilayer at the stages marked with large dots on the loops. (Long arrows correspond to Dy moments, short ones to Fe moments. For clarity, Y moments, antiparallel to Fe ones in YFe_2, are not sketched.)

(This figure is in colour only in the electronic version)

used with the external magnetic field (up to ± 7 T) parallel to the incident x-ray beam, both at 5° from the in-plane $[1\bar{1}0]$ direction. In $DyFe_2/YFe_2$ superlattices, this direction has been shown to be magnetically easier than the in-plane [001], which is the easy magnetization direction in bulk $DyFe_2$ [15]. Such a difference can be related to epitaxial strains and magnetostrictive effects, as is the case for $DyFe_2$ epitaxial films [17]. The XMCD spectra were recorded at the Dy and Y L_3 absorption edges, by flipping the helicity of incoming x-rays and keeping the direction of the magnetic field fixed, which enables the measurements of compound-selective hysteresis loops. The spectra were recorded in total fluorescence detection mode with a Si photodiode detector, which is not sensitive to the external applied magnetic field, at least in the range of interest.

Figure 1 illustrates the strong difference between the magnetization reversals in the $[DyFe_2(5\ nm)/YFe_2(20\ nm)]_{13}$ superlattice at 100 and at 200 K. Net magnetizations, collected by SQUID measurement after cooling the sample from room temperature under a +7 T external field, are shown in the top panels (continuous lines), and the compound-selective magnetizations, collected by XMCD at both the Dy (filled symbols) and Y (open symbols) edges, are presented in the bottom panels.

At 100 K (figure 1(a)), the net magnetization reversal occurs in two steps, the coercive field is positive and the remanent state thus corresponds to a net negative magnetization. At 200 K (figure 1(c)), three steps are obvious in this more complex reversal process; the coercive field is negative and the remanent state now corresponds to a net positive magnetization.

At 100 K, the XMCD loops reveal a behaviour in agreement with the usual scheme for exchange springs (figure 1(b)). The $DyFe_2$ layers act as a hard magnetic material, with a nearly square hysteresis loop and a large coercive field of 5.5 T. In contrast, the YFe_2 layers behave as a soft magnetic material subjected to a negative exchange field at the interfaces. Under the maximum applied field of +6 T, both magnetizations are mainly along the field direction with presumably a very narrow interface domain wall, due to the interface negative coupling (scheme 1). The interface domain walls expand inside the softer YFe_2 layers when the field is lowered (scheme 2), resulting in the decrease of the average YFe_2 magnetization, and thus of the Y XMCD signal. Below -1 T and down to the coercive field of the $DyFe_2$ layers, the sample exhibits the equilibrium ferrimagnetic structure (scheme 3). This remanent state results in a negative net magnetization because the YFe_2 magnetization is dominant. The initial high field situation is finally recovered below -6 T when the negative field overcomes the $DyFe_2$ anisotropy (scheme 4).

The compound-selective loops measured at 200 K (figure 1(d)) exhibit considerable changes compared with the previous case. Despite a similar initial magnetic configuration under the maximum applied field (scheme 1), a continuous decrease of the Dy XMCD signal is now observed while reducing the field. The $DyFe_2$ magnetization goes through zero at +1.3 T and reaches its highest negative value at +0.5 T, which means that there is a complete reversal of the $DyFe_2$ magnetization under *positive* field. As a matter of fact, a small variation of the Y signal is simultaneously observed: this decreases by approximately 15% from the saturation to +1.5 T (grey arrow in figure 1(d)) and then increases back rapidly to recover its saturation value when $DyFe_2$ becomes negatively saturated. In this 0/+0.5 T field range, the giant ferrimagnetic configuration is stabilized (scheme 2′), as expected for small field when exchange contributions are dominant. Moreover, at this temperature, the dominant magnetization (that of YFe_2) is along the field direction and the remanent state therefore corresponds to a positive magnetization.

As the external applied field becomes negative, the global magnetization is reversed as a block (both XMCD signals reverse simultaneously for $H = -0.7$ T) to minimize the Zeeman energy while keeping the ferrimagnetic profile (scheme 3). The increased negative field finally breaks the giant ferrimagnetic structure in forcing the $DyFe_2$ net magnetization to align progressively with the external field (scheme 4).

The curve presented with crosses in figures 1(a) and (c) is the weighted sum of the two compound-selective loops recorded with XMCD (presented in figures 1(b) and (d)): it is the sum of the loop recorded at the Dy edge and of the loop recorded at the Y edge, multiplied by a factor of -0.95. Because the units are totally different, this factor of 0.95 is meaningless. It nevertheless becomes evident that the XMCD measurements nicely reproduce the macroscopic magnetization loop, using exactly the same factor for the weighted sum, for both temperatures.

The above-reported results thus evidence a strong temperature dependence for the magnetization reversal process, although the reduction of the external magnetic field tends, in both cases, to stabilize the equilibrium ferrimagnetic structure. At 100 K, where the $DyFe_2$ anisotropy is large, the reversal essentially occurs in the dominant soft YFe_2 layers. At 200 K, the dominant YFe_2 magnetization remains aligned with the external field and the $DyFe_2$ magnetization reverses first. The direct and separate observation of those two contributions to the reversal mechanism explains the temperature dependence of the remanent state and of the coercive field, which are both governed by the dominant YFe_2 layers.

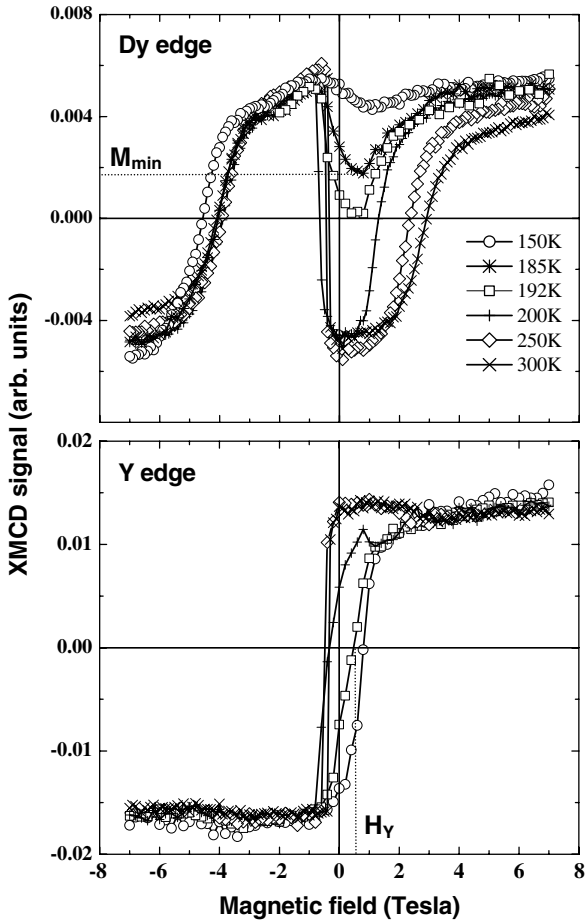

Figure 2. XMCD half hysteresis curves (from +7 to −7 T) measured for various temperatures after a +7 T field-cooling process at the Dy and Y L_3 edges for the superlattice $[DyFe_2(5\ nm)/YFe_2(20\ nm)]_{13}$.

The transition between these two reversal mechanisms has been studied in further detail by recording at various temperatures the Dy and Y XMCD half loops from +7 to −7 T after the +7 field-cooling process (figure 2). As seen in figure 2(a), at intermediate temperature of 192 K (squares), the Dy XMCD signal exhibits only a partial reduction under positive field and reaches a minimum value M_{min}. The rate of the transition can be quantified by M_{min} that, as shown in figure 3 (filled circles), decreases very rapidly between 175 and 200 K. In the same temperature range, the reversal of the Y XMCD signal (figure 2(b)) evolves from smooth at 150 K (circles) to much sharper at 250 K (diamonds), and can also be quantified by the reversal field H_Y where the Y signal becomes zero. The temperature dependence of H_Y (empty circles in figure 3) follows that of the minimum Dy signal M_{min} and H_Y changes sign from positive to negative when there is a complete reversal of the $DyFe_2$ magnetization under a positive external field ($M_{min} \approx -M_{max}$). These results allow us to define three different regimes: (i) the low temperature regime (SF below 150 K), where the $DyFe_2$ magnetization remains aligned with the field ($M_{min} \approx +M_{max}$) and the negative interface exchange field drives the YFe_2 reversal under positive field ($H_Y > 0$); (ii) the high temperature regime (HF above 200 K), where the

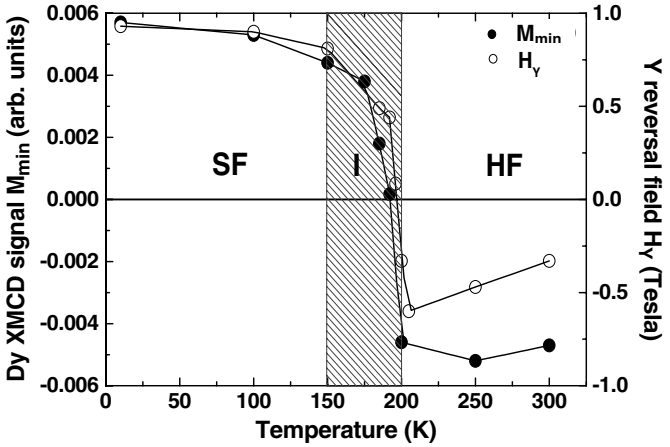

Figure 3. Compared temperature dependence of the Dy XMCD signal ($M_{\rm min}$) at the deep position (filled circles) and of the reversal field $H_{\rm Y}$ measured at the Y edge (empty circles) for the superlattice $[DyFe_2(5\ nm)/YFe_2(20\ nm)]_{13}$. 'SF' is the low temperature regime where the soft layers reverse first; 'HF' the high temperature regime where the hard layers reverse first; 'I' the intermediate regime.

exchange field first drives the hard $DyFe_2$ layer reversal ($M_{\rm min} \approx -M_{\rm max}$) while the YFe_2 magnetization remains aligned with the applied field ($H_{\rm Y} < 0$); (iii) the intermediate regime (hatched I area), where the magnetization of both compounds is affected by the external field reduction. In this case, the interface domain walls partially expanding in the YFe_2 layers drive the complete reversal of the YFe_2 magnetization before that of $DyFe_2$($H_{\rm Y} > 0$), which causes the switching of the $DyFe_2$ net magnetization back along the field direction for positive fields.

Let us now discuss the possible origin for this temperature dependent magnetization reversal. As already mentioned previously, the YFe_2 compound is the dominant one in this superlattice, since the YFe_2 layers are four times thicker than the $DyFe_2$ ones. The stable configuration expected under small field, and at any temperature, is therefore ferrimagnetic with the YFe_2 net magnetization pointing along the field and the $DyFe_2$ net magnetization opposite, as actually observed at 300 K. However, having saturated the sample under +7 T at a given temperature, the question is whether this low energy configuration can be reached, i.e. via the $DyFe_2$ reversal, or if the energy required for this $DyFe_2$ reversal is too high.

The results presented in this letter show that the energy required for the $DyFe_2$ reversal is obviously too high at 100 K and the YFe_2 thus reverses under positive field, despite its dominant character. But the $DyFe_2$ reversal becomes easier when increasing temperature, which enables the YFe_2 dominant magnetization to stay aligned with the external field.

The easier $DyFe_2$ reversal in increasing temperature may be attributed to the simultaneous thermal reduction of the magnetization density and of the anisotropy in that compound: in bulk $DyFe_2$, the anisotropy constants are reduced by an order of magnitude between 0 and 300 K and the net moment decreases by 17% between 100 and 200 K, from 5.25 to 4.5 $\mu_{\rm B}$/fu. respectively. This smaller magnetization density makes it easier to reverse the $DyFe_2$ magnetization opposite to the applied field, since the cost in Zeeman energy is lower. The lower anisotropy at higher temperature also reduces the cost for the shift away from a given easy magnetization direction.

Finally, subtle changes of easy magnetization axis in the $DyFe_2$ layers in increasing temperature may not be completely ruled out, since such temperature-dependent effects have been evidenced in thin $DyFe_2$ films [17]. It has been proved that the lattice strains in thin

$DyFe_2$ films play a key role in determining the easy magnetization direction. However, the strain state of the $DyFe_2$ layers embedded in the superlattices is likely different from the case of thin films alone; this does not enable us to infer the easy magnetic axis in the superlattice, from previous results obtained for thin films. Much more elaborate experiments should be planned to address this issue.

As these XMCD measurements do not provide depth-resolved or laterally resolved information, complementary experiments should be also performed to clarify the exact magnetic profile during the reversal process, especially in the intermediate and high temperature regimes. Nevertheless, different possible schemes, consistent with the experimental results reported, can be proposed.

(i) A first possible configuration is a canting-like structure, where the $DyFe_2$ magnetization would rotate almost homogeneously with the interface magnetic twist inside (or almost completely inside) the soft material. Note that this structure is different from the canted one reported by Bentall *et al* [14], since in their case the magnetization of both compounds is almost perpendicular to the field direction. In this study, starting from 180° narrow interface walls in the YFe_2 layers, the field reduction would induce a canting of the homogeneous $DyFe_2$ magnetic blocks by an angle θ with respect to the field direction. At 200 K and under 1.3 T, θ would reach 90° and 4 nm of the YFe_2 layers would be involved in each 90° interface domain wall. The main point that makes this hypothesis unlikely is however the cost in magnetocrystalline anisotropy of the homogeneous $DyFe_2$ magnetic blocks, which are not aligned along the easy magnetization direction.

(ii) Another possibility is that the interface magnetic twists, which take place and expand in the soft YFe_2 layers at low temperature, translate towards the $DyFe_2$ layers on increasing the temperature. Such an effect can be likely explained by the simultaneous thermal reduction of the magnetization density and of the anisotropy mentioned previously: in increasing temperature, the smaller anisotropy especially reduces the domain wall energy in that layer.

(iii) Finally, the reduction of the $DyFe_2$ magnetization could also be interpreted as the result of nucleation of lateral domains with opposite magnetization direction and growth of these reversed magnetic domains.

In conclusion, element specific XMCD measurements provide unique information to unravel the complex magnetization reversal in exchange coupled systems. We have shown in the $[DyFe_2(5\ nm)/YFe_2(20\ nm)]_{13}$ superlattice that the harder magnetic layers can exhibit the first reorientation process to a direction antiparallel to the field. This process is completely different from the classical description of the spring magnet, which is observed at lower temperature. A strong and rather abrupt temperature dependence of the reversal mechanism allows a clarification of the evolution of the coercivity and of the remanent state as a function of temperature. However, the magnetic configuration during reversal in the intermediate and high temperature regime still has to be determined, by performing depth-resolved measurements, such as polarized neutron and x-ray magnetic reflectivity, and laterally resolved experiments, using off-specular scattering. The temperature and thickness range where the HF reversal mechanism is favoured, as well as the influence of the field direction, also remains to be explored in further detail.

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