Decoupling of the magnetic sublattices at the compensation point in *R*-Fe compounds

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The behavior of the *R*-Fe interaction at the magnetic compensation point of $Ho_{0.5}Lu_{0.5}Fe_2$ and $Er_{0.5}Y_{0.5}Fe_2$ compounds has been studied by means of x-ray magnetic circular dichroism (XMCD). The results point out that at the compensation point, the rare-earth sublattice loses completely the magnetic order whereas the Fe sublattice still presents local magnetic order. In addition, the XMCD data show that close below to the compensation point the system is in an intermediate state in which the magnetization of both magnetic sublattices are parallel to the applied magnetic field, and that the ferrimagnetic coupling is progressively restored again by decreasing the temperature.

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Since the first indication by Néel,[1] a wealth of ferrimagnets showing a magnetic compensation point have been extensively studied both experimentally and theoretically.[2,3] Recently, the study of the magnetic properties of materials near the magnetic compensation temperature has received renewed interest due to the discovery of zero-moment $Sm_{1-x}Gd_xAl_2$ ferromagnets.[4] These systems show a compensation temperature at which long-range ferromagnetic order persists in spite of the zero net magnetization, opening the possibility of developing spin-resolved devices without the interference of the applied magnetic field.[5–8] This behavior is due to the peculiar nature of the Sm^{3+} ion in which the spin and orbital moments are almost compensated. By contrast, the magnetic compensation in *R*-Fe intermetallic compounds (*R*=heavy rare earth) originates from the cancellation of the net magnetization resulting from the antiparallel coupling of both *R* and Fe magnetic sublattices. Despite that this simple scheme accounts for the observed macroscopic properties,[9,10] little is known regarding the behavior of the individual magnetic sublattices and their coupling through the compensation point. Aimed to obtain such a microscopic picture, we have performed a XMCD study in the case of $Ho_{0.5}Lu_{0.5}Fe_2$ and $Er_{0.5}Y_{0.5}Fe_2$ compounds.

In previous works, we have shown that the combined study of the Fe *K* and *R* L_2 edges XMCD provides the disentanglement of the magnetic behavior of both sublattices at the microscopic level.[11,12] In this work, we will apply this disentangling procedure in order to obtain an insight in the behavior of the individual magnetic moments as well as the spin polarization of the hybridized *R*(5*d*) states, which mediate the magnetic coupling of both sublattices, through the magnetic compensation transition. Our results lead to a new scenario to describe the magnetic behavior of the system at the compensation point. Moreover, the disentangling procedure allows us to explore the transient regime between the compensation point and the ferrimagnetic state.

$Ho_{1-x}Lu_xFe_2$, $Er_{1-x}Y_xFe_2$ (x=0.50 and 1), $Ho_{0.5}Lu_{0.5}Al_2$, and $ErAl_2$ compounds were prepared by arc-melting the pure elements under Ar protective atmosphere. The ingots were annealed at 850 °C for one week. X-ray diffraction characterization indicates that all the samples show a single C15 Laves phase. The presence of secondary phases is overall less than <2%. For both $M(T)$ and XMCD measurements the samples were zero-field cooled (ZFC). Magnetization measurements were performed by using a commercial superconducting quantum interference device magnetometer (Quantum Design MPMS-5S model). The curves were recorded upon warming up while the magnetic field was kept constant. XMCD experiments were performed at the beamline BL39XU of the SPring-8 facility.[13] XMCD spectra were recorded in the transmission mode at the Fe *K* edge and at the rare-earth $L_{2,3}$ edges by using the helicity-modulation technique.[14] For the measurements, homogeneous layers of the powdered samples were made by spreading fine powders of the material on adhesive Kapton tape. The detailed description of the experimental setup and the measurement mode can be found elsewhere.[15] In all the cases, the origin of the energy scale (E_0) was chosen at the inflection point of the absorption edge and the x-ray absorption spectra were normalized to the averaged absorption coefficient at high energy.

The temperature dependence of the magnetization of both $Er_{0.5}Y_{0.5}Fe_2$ and $Ho_{0.5}Lu_{0.5}Fe_2$ compounds under a magnetic field of H=50 kOe is shown in Fig. 1. By contrast to the flat curve of YFe_2 and $LuFe_2$, the magnetization passes through a minimum at T_{Comp}=230 and 300 K for the Er and Ho compounds, respectively. This thermal dependence reflects the existence of a magnetic compensation point at T_{Comp}: the Fe sublattice dominates the overall magnetization of the system at temperatures above T_{Comp} but, upon cooling, the magnetization of the rare-earth sublattice increases and surpass the Fe one below T_{Comp}. Because the Fe *K* XMCD spectra reported in Fig. 1 are referred to the direction of the total magnetization of the system, the change in sign of the signal for temperatures above and below T_{Comp} directly reflects the change in the dominant magnetic sublattice. It is worth to note that the compensation temperature for the studied compounds does not change for magnetic fields equal or higher than H=20 kOe.

As it is shown in panels (b) and (c) of Fig. 1, the Fe *K* XMCD signals recorded on each compound at temperatures below and above T_{Comp} are similar and resembling the char-

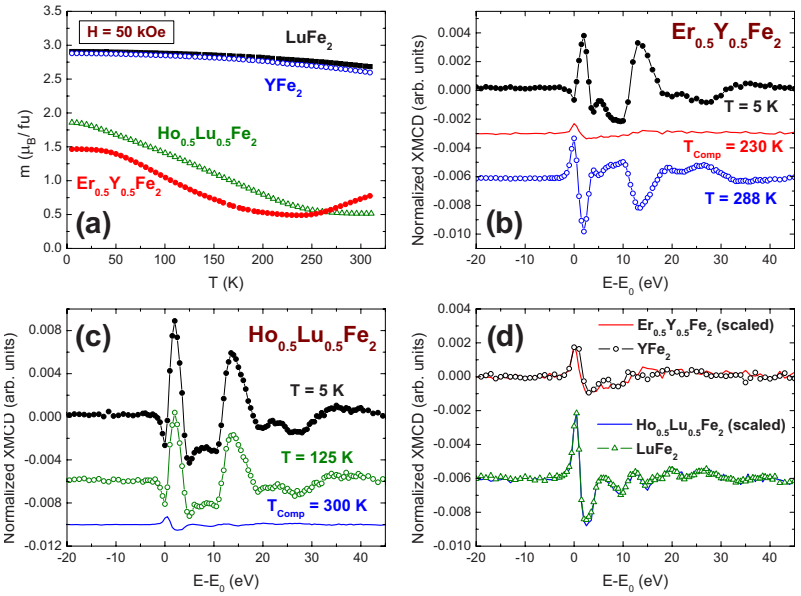

FIG. 1. (Color online) (a) Temperature dependence of the ZFC magnetization measured upon warming on $Er_{1-x}Y_xFe_2$ and $Ho_{1-x}Lu_xFe_2$ compounds under an applied magnetic field $H=50$ kOe. Temperature dependence of Fe K-edge XMCD spectra of $Er_{0.5}Y_{0.5}Fe_2$ [panel (b)] and $Ho_{0.5}Lu_{0.5}Fe_2$ [panel (c)]. (d) Comparison of the XMCD signals recorded at the compensation temperature in both $Er_{0.5}Y_{0.5}Fe_2$ and $Ho_{0.5}Lu_{0.5}Fe_2$ scaled to those of YFe_2 and $LuFe_2$ recorded at room temperature (see text for details). Some spectra have been vertically shifted for the sake of clearness.

acteristic Fe K XMCD of R-Fe intermetallics in which R is a magnetic rare earth.[16–18] For these compounds, the Fe K XMCD spectrum is originated from the addition of two contributions associated to both the Fe and rare-earth sublattices.[15,19,20] Surprisingly, this characteristic absorption profile is lost at the compensation point and the XMCD spectra are similar to those of YFe_2 and $LuFe_2$ (compounds in which no $4f$ magnetic moments are present). In fact, the dichroic signals at T_{Comp} match perfectly to those of YFe_2 and $LuFe_2$ [see Fig. 1(d)]. The rare-earth contribution to the Fe K-edge XMCD stems from the hybridization of the conduction states and its magnitude depends on the strength of the exchange interaction between the rare-earth and the Fe magnetic moments.[15] The fact that the dichroic signal at T_{Comp} is similar to that observed in the absence of magnetic rare earth indicates that at T_{Comp} the rare-earth sublattice is fully magnetically disordered whereas there is still some magnetic order in the Fe sublattice. Indeed, if the rare-earth sublattice was locally ordered the magnetic moments of the rare earths would create a molecular field at the Fe sites and, consequently, the rare-earth contribution to the Fe K-edge XMCD should be present, contrary to the present results.

Aimed to verify these results, we have studied the behavior of the XMCD spectra recorded at the rare-earth L_2 edge through the compensation temperature. Previous works have shown that, as in the case of Fe K edge, the XMCD at the rare-earth L_2 edge is made of the addition of two components,[21,22] one due to the rare-earth sublattice, mainly reflecting the $4f$-$5d$ intra-atomic polarization, and a second one in which the magnetic state of Fe is reflected through the hybridization of the Fe($3d$,$4p$) and $R(5d)$ states.[23,24] The Fe contribution yields a characteristic positive peak in the dichroic signal at the threshold energy, E_0. As shown in the comparison of the Er L_2-edge XMCD spectra of $Er_{0.5}Y_{0.5}Fe_2$ and $ErAl_2$, and that of the Ho L_2 XMCD of $Ho_{0.5}Lu_{0.5}Fe_2$ and $Ho_{0.5}Lu_{0.5}Al_2$ reported in Fig. 2, it is clear that this peak is not present in absence of Fe. Indeed, by subtracting the $Ho_{0.5}Lu_{0.5}Al_2$ and $ErAl_2$ XMCD spectra from the

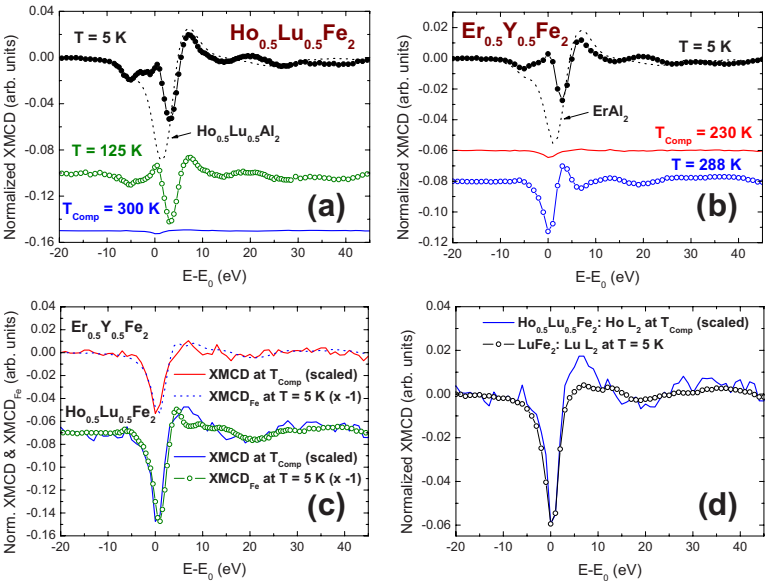

FIG. 2. (Color online) Temperature dependence of Ho L_2 and Er L_2-edges XMCD spectra of $Ho_{0.5}Lu_{0.5}Fe_2$ [panel (a)] and $Er_{0.5}Y_{0.5}Fe_2$ [panel (b)], respectively. The dotted lines at panels (a) and (b) corresponds to XMCD spectra of $Ho_{0.5}Lu_{0.5}Al_2$ and $ErAl_2$, both measured at $T=5$ K. (c) Comparison of the R L_2 XMCD signals of $Er_{0.5}Y_{0.5}Fe_2$ and $Ho_{0.5}Lu_{0.5}Fe_2$ at T_{Comp} and the extracted contribution of the Fe sublattice to the XMCD spectrum at $T=5$ K (see text for details). For a better comparison the signals measured at T_{Comp} have been scaled to match the extracted $XMCD_{Fe}$ ones which have been multiplied by -1. (d) Comparison of the Ho L_2 XMCD in $Ho_{0.5}Lu_{0.5}Fe_2$ ($T=T_{Comp}$) and Lu L_2 XMCD in $LuFe_2$ ($T=5$ K). For a better comparison the signal at the Ho L_2 has been scaled to the one at Lu L_2 one.

$Ho_{0.5}Lu_{0.5}Fe_2$ and $Er_{0.5}Y_{0.5}Fe_2$ ones, respectively, we find in both cases a similar difference signal that corresponds to the Fe contribution, $XMCD_{Fe}$. The sign of this Fe contribution is opposite to that of the rare-earth itself, and its relative weight decreases as temperature diminishes because the Fe magnetization remains nearly constant while the rare-earth one significantly increases [~100% (Ref. 11)] as the temperature decreases from room temperature down to 5 K.

At temperatures far from T_{Comp} the Ho L_2 and Er L_2 XMCD spectra of $Ho_{0.5}Lu_{0.5}Fe_2$ and $Er_{0.5}Y_{0.5}Fe_2$, respectively, are as described above [see panels (a) and (b) of Fig. 2]. However, the XMCD signals exhibit a dramatic change at T_{Comp}. In both cases, the XMCD spectra only show a negative peak at the absorption threshold, i.e., at the energy region in which the Fe sublattice contributes to the rare-earth L_2 edge. As shown in panel (c) of Fig. 2, this signal matches with the $XMCD_{Fe}$ extracted from the data at $T=5$ K. This result indicates that, at T_{Comp}, only Fe is contributing to the XMCD recorded at the rare-earth L_2 edge. Additionally, its negative sign indicates that the Fe sublattice governs the direction of the total magnetization of the system. A final confirmation is given by the comparison of the signal of $Ho_{0.5}Lu_{0.5}Fe_2$ at Ho L_2 edge at T_{Comp} with that of $LuFe_2$ at Lu L_2 edge at $T=5$ K. In the latter case, the polarization of the Lu $5d$ states is undoubtedly due to the action of the Fe magnetic moments. As shown in Fig. 2(d), both signals match which corroborates that at the compensation point the Ho $5d$ states are only polarized by the Fe sublattice. These results are in agreement with those obtained at the Fe K-edge XMCD study: at the compensation point the Fe sublattice is locally ordered and the Fe conduction states are polarized. As these states are hybridized with the $5d$ states of the rare earth, there is also a polarization of the conduction states projected at the rare-earth sites due to the local order of the Fe sublattice.

These results suggest that at the compensation point, the R and the Fe magnetic sublattices behave in a different way. The XMCD data demonstrate that the R sublattice is completely magnetically disordered while some magnetic order is still present in the Fe sublattice. This different behavior might be ascribed to the hierarchy of the magnetic interactions in R-Fe intermetallics: Fe-Fe $\gg$ R-Fe $\gg$ R-R. At the compensation point, both R and Fe sublattices would be magnetically disordered since there is no preferred magnetic direction. However, the local Fe-Fe exchange interaction is strong enough to maintain a certain local order among the Fe magnetic moments while the R-Fe interaction is not strong enough and the R magnetic moments remain magnetically disordered.

In order to verify this proposition, we have explored the transient regime between the anomalous magnetically disordered state at the compensation point and the ferrimagnetic state. To this end we have slightly decreased the temperature of the $Ho_{0.5}Lu_{0.5}Fe_2$ compound from $T_{Comp}=300$ K down to $T=288$ K. On the one hand, as shown in Fig. 3(a), new spectral features appear in the $T=288$ K spectra. The intensity of these features (labeled as B, C, and D) is enhanced by increasing the magnetic field from $H=20$ to 100 kOe, up to resemble the XMCD signal recorded at low temperature.[25] Peaks B, C, and D have the same sign in both cases, indicating that at $T=288$ K, the Ho moments are orientated parallel to the magnetic field, becoming the dominant magnetic sublattice. On the other hand, the peak at the threshold (peak A) surprisingly has the same sign in both the $T=T_{Comp}$ and $T=288$ K spectra. This peak should reverse its sign if both sublattices were ferrimagnetically coupled. Hence, this result suggests that the Fe moments remains oriented parallel to the net magnetization of the system and the magnetic sublattices are still decoupled at $T=288$ K. Similar results are found at the Ho L_2-edge XMCD. As shown in Fig. 3(b), the slightly reduction in the temperature changes the shape of the XMCD spectrum as to resemble the one measured at low temperature. However, the characteristic peak associated to Fe (A′ peak in Fig. 3) is not positive, as occurring at temperatures far below T_{Comp}, but negative, indicating that both the Fe and Ho moments are parallel to the net magnetization of the system.

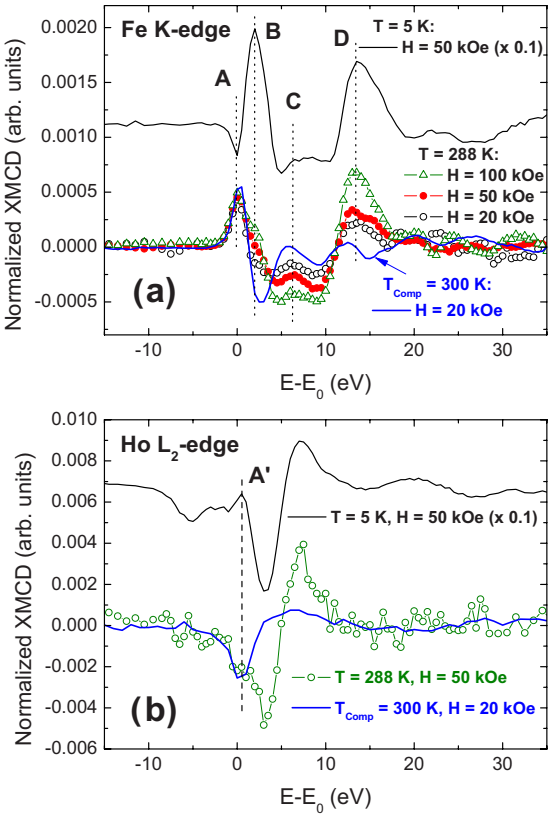

FIG. 3. (Color online) Evolution with the magnetic field of the XMCD signals recorded at the Fe K-edge [panel (a)] and Ho L_2-edge [panel (b)] in $Ho_{0.5}Lu_{0.5}Fe_2$ compound when the system is driven out the compensation condition by decreasing the temperature. The signals recorded at $T=5$ K and $H=50$ kOe are also included for the sake of clearness (see text for details).

Another interesting result can be inferred from a closer inspection of the data displayed in Fig. 3(a). As the Ho sublattice become more ordered, by increasing the magnetic field, two different Fe contributions to the XMCD, parallel and antiparallel to the net magnetization of the system, are evidenced. Moreover, the antiparallel contribution grows at expenses of the parallel one as the magnetic field increases. This is exemplified in the Fig. 4(a) in which the $XMCD_{Ho}$

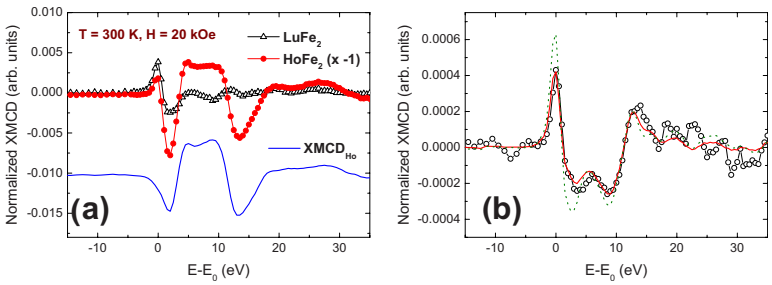

FIG. 4. (Color online) (a) $HoFe_2$ and $LuFe_2$ Fe K-edge XMCD signals recorded at $T=300$ K and $H=20$ kOe and the respective extracted $XMCD_{Ho}$ contribution. (b) Composed signal by using parallel $XMCD_{Fe}$ contribution (green dotted line) and both, parallel and antiparallel (red line) to match the experimental dichroic signal of $Ho_{0.5}Lu_{0.5}Fe_2$ measured at $T_{Comp}=300$ K and $H=20$ kOe (black ○) (see text for details).

contribution has been obtained by following the procedure described at Ref. 11. Then to recover the XMCD signal at $T=288$ K by composition of $XMCD_{Ho}$ and $XMCD_{Fe}$ components it is needed to consider both, parallel and antiparallel, contributions of $XMCD_{Fe}$ [see Fig. 4(b)]. These results indicate that both Fe and R magnetic sublattices are decoupled at T_{Comp} and as the compound is driven out from the compensation point the ferrimagnetic coupling is progressively recovered.

Summarizing, we have studied the behavior of the rare-earth and Fe magnetic sublattices of $Ho_{0.5}Lu_{0.5}Fe_2$ and $Er_{0.5}Y_{0.5}Fe_2$ at the compensation temperature by means of XMCD. Our results indicate that the R sublattice is completely magnetically disordered at T_{Comp} while the Fe sublattice remains locally ordered. This scenario is in contrast to that observed in the Sm-Gd-Al compounds which preserve the long-range ferromagnetic order at the compensation point. Moreover, our XMCD data show that close to the compensation point both the rare-earth and Fe sublattices are magnetically decoupled and their magnetic moments are parallel to the applied magnetic field. As the compounds are driven out from the compensation point, the rare-earth moments begin to be orientated in the direction of the applied magnetic field and the R-Fe ferrimagnetic coupling of both magnetic sublattices is progressively recovered.

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