Competing effects in the magnetic polarization of *non-magnetic* atoms.

R. Boada^{1,2‡}, M. A. Laguna-Marco³ C. Piquer^{1,2} and J. Chaboy^{1,2}

¹Instituto de Ciencia de Materiales de Aragón, Consejo Superior de Investigaciones Científicas, CSIC-Universidad de Zaragoza, 50009 Zaragoza, Spain.

²Departamento de Física de la Materia Condensada, Universidad de Zaragoza, 50009 Zaragoza, Spain.

³Instituto de Ciencia de Materiales de Madrid, CSIC, Cantoblanco, Madrid, 28049, Spain.

E-mail: rboada@unizar.es

Abstract.

The magnetic polarization of the Lu $5d$ states through the $\text{Ho}_{1-x}\text{Lu}_x(\text{Fe}_{1-y}\text{Al}_y)_2$ series has been studied by means of X-ray magnetic circular dichroism. The combined study of the dichroic signals performed at the Fe K-, Ho L₂- and Lu L_{2,3}-edges gives a complete picture of the polarization scheme at the conduction band. The results point out that in the presence of competing localized magnetic moments, $\mu_{\text{Fe}}(3d)$ and $\mu_{\text{Ho}}(4f)$, the dichroic signal at the Lu site is mainly due to the Fe atoms, being negligible the effect of the magnetic rare-earth. Estimation of the spin and orbital components of the Lu($5d$) induced magnetic moment have been obtained by applying the magneto-optical sum rules derived for X-ray magnetic circular dichroism.

(Some figures in this article are in colour only in the electronic version)

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‡ Present address: Diamond Light Source Ltd, Harwell Science and Innovation Campus, Didcot, Oxfordshire OX11 0DE, UK

1. Introduction

The magnetic polarization of *non-magnetic* atoms, plays an important role into determining the magnetic properties of many systems [1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12]. However, the exact nature of the induced magnetic moments remains an open problem, whose clarification is a key step for the understanding of the magnetic interactions in these systems. This is the case of the rare-earth $5d$ states in rare-earth – transition metal (R-T) intermetallics which mediate the R-T interaction via de $R(5d)$ - $T(3d)$ hybridization [13, 14, 15].

Previous works have demonstrated that, in R-T intermetallics, these $R(5d)$ states can be explored not only by means of XMCD measurements at the R $L_{2,3}$ -edges, but also by measuring at the T K-edge [16, 17, 18, 19, 20, 21, 22, 23, 24]. It has been shown that the dichroic spectra at the T K- and R $L_{2,3}$ -edges are a simultaneous fingerprint of the magnetism of both transition-metal and rare-earth, even when only one atomic element is probed. More specifically, in these systems the transition metal contributes to the rare-earth $L_{2,3}$ -edges XMCD, spectra and, conversely, there is a contribution of the lanthanide metal to the XMCD recorded at the K-edge of the transition metal [16, 17, 18, 19, 20, 21, 22, 23, 24]. The proper disentanglement of these crossed contributions, XMCD_R at the Fe K- and XMCD_{Fe} at the R L_2 -edges, provides selective magnetic information from the study of the dichroic signals at the conduction band [25, 26, 27]. **So far, these dichroic signals have been studied for magnetic absorbing atoms in which either $4f$ or $3d$ localized magnetic moments were present.** However, little is known about both the magnetic polarization and the XMCD behaviour of *non-magnetic* atoms as Lu in the presence of localized magnetic moments.

In this work, we have faced the problem of determining which localized magnetic moment, $R(4f)$ and/or $Fe(3d)$, is responsible for the induced magnetic polarization of the Lu $5d$ states. To this end, we have synthesized two series, $\text{Ho}_{1-x}\text{Lu}_x\text{Fe}_2$ and $\text{Ho}_{0.5}\text{Lu}_{0.5}(\text{Fe}_{1-y}\text{Al}_y)_2$, in which the relative contribution of both $4f$ and $3d$ moments is controlled. In the former case, for a fixed Fe contribution, the magnetization of the rare-earth sublattice is progressively depleted by increasing the Lu content. Therefore, the polarization of the Lu $5d$ states due to the Fe $3d$ ones is maintained fixed, while that of the Ho $4f$ states decreases. Whereas in the latter series, the polarization of the Lu $5d$ states due to Ho atoms is fixed, while the polarization due to Fe atoms decreases by increasing the concentration of *non-magnetic* Al.

Therefore we have studied how the modification of the magnetization of each magnetic sublattice as well as the R-Fe hybridization affect the magnetic polarization of the Lu $5d$ states by recording the XMCD at the Lu $L_{2,3}$ -edges. Our results point out that in the presence of competing localized magnetic moments, $\mu_{Fe}(3d)$ and $\mu_{Ho}(4f)$, the dichroic signal at the Lu site is mainly due to the Fe atoms, being negligible the effect of the magnetic rare-earth.

2. Experimental

$\text{Ho}_{1-x}\text{Lu}_x\text{Fe}_2$ ($x = 0, 0.25, 0.5, 0.75, 1$) and $\text{Ho}_{0.5}\text{Lu}_{0.5}(\text{Fe}_{1-y}\text{Al}_y)_2$ ($y = 0, 0.25, 0.5, 0.75, 1$) series 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 (XRD) patterns were recorded to determine the crystallographic structure and the degree of crystallinity of all the synthesized samples. Measurements have been performed on powdered samples at room temperature by using a Cu K_α radiation in Bragg-Brentano geometry, $2\theta = 15 - 80^\circ$ (Rigaku RTO 500RC diffractometer). Magnetization measurements, $M(H)$, were performed at $T = 5$ K by using a commercial superconducting quantum interference device magnetometer (Quantum Design MPMS-5S).

XMCD experiments were performed at the beamline BL39XU of the SPring-8 facility [28]. XMCD spectra were recorded in the transmission mode at the Fe K- and at the rare-earths $L_{2,3}$ -edges by using the helicity-modulation technique [29]. Circularly polarized X-rays were generated using a diamond phase plate 0.73 mm-thick. For the measurements, homogeneous layers of the powdered samples were made by spreading fine powders of the material on adhesive Kapton tape. The thickness and homogeneity of the samples were optimized to obtain the best signal-to-noise ratio. The XMCD spectra were recorded at $T = 5$ K and room temperature under the action of a $H = 50$ kOe magnetic field.

The dichroic signal was obtained as the difference of the absorption coefficient $\mu_c = \mu^- - \mu^+$ for antiparallel μ^- and parallel μ^+ orientation of the photon helicity and sample magnetization. For the sake of accuracy the direction of the applied magnetic field is reversed and the XMCD, now $\mu_c = \mu^+ - \mu^-$, is recorded again by switching the helicity. The subtraction of the XMCD spectra recorded for both field orientations cancels, if present, any spurious signal. In all the cases, the origin of the energy scale (E_0) was chosen at the inflection point of the absorption edge and the XAS spectra were normalized to the averaged absorption coefficient at high energy.

3. Results and Discussion

3.1. Structural and magnetic characterization

Figure 1 shows the evolution of the XRD patterns through the $\text{Ho}_{1-x}\text{Lu}_x\text{Fe}_2$ and $\text{Ho}_{0.5}\text{Lu}_{0.5}(\text{Fe}_{1-y}\text{Al}_y)_2$ series. X-ray diffraction characterization indicates that all the samples show a single C15 Laves phase, with the exception of $\text{Ho}_{0.5}\text{Lu}_{0.5}(\text{Fe}_{0.5}\text{Al}_{0.5})_2$ which shows the C14 structure as expected for intermediate Fe concentrations [30]. The patterns of the compounds with C15 structure are nearly identical and only a small shift upon dilution is found. This shift is due to the modification of the crystal cell parameters: lattice expansion (contraction) occurs upon replacing Fe by Al (Ho by Lu) leading to a displacement of the peaks towards lower (higher) 2θ angle.

The structural information derived from the Rietveld refinement is summarized in

Figure 1. Comparison of the XRD diffraction patterns through the $\text{Ho}_{1-x}\text{Lu}_x\text{Fe}_2$ (left panel) and $\text{Ho}_{0.5}\text{Lu}_{0.5}(\text{Fe}_{1-y}\text{Al}_y)_2$ (right panel) series.

Table 1. Structural information derived from Rietveld refinement of XRD patterns: crystal structure, lattice parameter ($\pm 0.5 \times 10^{-3}$), reliability Bragg factor associated to the main phase and percentage of secondary phases (see text for details).

Sample	Structure	a(Å)	c(Å)	R_{Bragg}	Secondary Phases
HoFe_2	C15	7.294	-	9.44	<2% R_2O_3
$\text{Ho}_{0.75}\text{Lu}_{0.25}\text{Fe}_2$	C15	7.278	-	9.83	<1% RFe_3
$\text{Ho}_{0.5}\text{Lu}_{0.5}\text{Fe}_2$	C15	7.254	-	10.5	<1% R_2O_3
$\text{Ho}_{0.25}\text{Lu}_{0.75}\text{Fe}_2$	C15	7.241	-	9.78	<1% R_2O_3
LuFe_2	C15	7.217	-	7.51	<2% R_2O_3
$\text{Ho}_{0.5}\text{Lu}_{0.5}(\text{Fe}_{0.75}\text{Al}_{0.25})_2$	C15	7.372	-	7.83	<2% R_2O_3
$\text{Ho}_{0.5}\text{Lu}_{0.5}(\text{Fe}_{0.5}\text{Al}_{0.5})_2$	C14	5.320	8.658	1.04	<2% R_2O_3
$\text{Ho}_{0.5}\text{Lu}_{0.5}(\text{Fe}_{0.25}\text{Al}_{0.75})_2$	C15	7.657	-	10.1	<3% R_2O_3
$\text{Ho}_{0.5}\text{Lu}_{0.5}\text{Al}_2$	C15	7.773	-	10.5	<4% RAl_3 + <2% R_2O_3

table 1: crystal structure, lattice parameters and the reliability Bragg factor associated to the main phase. Good crystallinity of the samples is indicated by the low Bragg factors obtained. In all the cases, as indicated in table 1, the presence of secondary phases has been proved to be **almost negligible**.

Figure 2(a) shows the magnetic field dependence of the magnetization recorded at $T = 5$ K for $\text{Ho}_{1-x}\text{Lu}_x\text{Fe}_2$ series. In all cases, the magnetization is nearly saturated at $H \gtrsim 10$ kOe. Hence, the saturation magnetization will be taken as the magnetization measured at $T = 5$ K and $H = 50$ kOe.

In the $\text{Ho}_{1-x}\text{Lu}_x\text{Fe}_2$ compounds the Fe magnetic moments are collinear and anti-

Figure 2. Isothermal magnetization curves measured at $T = 5$ K for (a) $\text{Ho}_{1-x}\text{Lu}_x\text{Fe}_2$: $x = 0$ (black, ∇), 0.25 (red, $\bullet$), 0.5 (green, $\blacktriangle$), 0.75 (blue, $\square$) and 1 (cyan, $\blacklozenge$); and (b) $\text{Ho}_{0.5}\text{Lu}_{0.5}(\text{Fe}_{1-y}\text{Al}_y)_2$: $y = 0$ (green, $\blacktriangle$), 0.25 (magenta, $\circ$), 0.5 (black, $\blacksquare$), 0.75 (orange, $\bullet$) and 1 (dark yellow, solid line).

ferromagnetically coupled to those of Ho, resulting in a ferrimagnetic behaviour. This ferromagnetic coupling can lead to a reversal in the dominant magnetic sublattice in the case of $\text{Ho}_{1-x}\text{Lu}_x\text{Fe}_2$ compounds, which has to be taken into account when analysing the XMCD data.

The magnetization measured at $T = 5$ K and $H = 50$ kOe $\text{Ho}_{1-x}\text{Lu}_x\text{Fe}_2$ can be accounted for by applying a two sublattice model. Within this approach, the total magnetization, M_{Tot} , corresponds to the simple addition of the magnetization of both magnetic sublattices: $\vec{M}_{\text{Tot}} = \vec{M}_{\text{Ho}} + \vec{M}_{\text{Fe}}$ and, therefore, the direction of the total magnetization of the system stems from the competition of the two magnetic sublattices. In this way, the absolute value of the total magnetization of the diluted $\text{Ho}_{1-x}\text{Lu}_x\text{Fe}_2$ compounds can be expressed as: $|M_{\text{Tot}}| = |(1-x)\mu_R - 2\mu_{\text{Fe}}|$. In the two sublattice model, it is usually assumed that M_{Fe} corresponds to the magnetization of the RFe_2 compounds in which R is *non-magnetic*, LuFe_2 , that is $\mu_{\text{Fe}} \approx 1.41 \mu_B$ [31]. Similarly, μ_{Ho} is taken as the magnetization value of the $\text{Ho}_{0.5}\text{Lu}_{0.5}\text{Al}_2$ compounds measured at the same experimental conditions ($\mu_{\text{Ho}} \sim 9.25 \mu_B$). This procedure reproduces well the values of the magnetization measured at $T = 5$ K and $H = 50$ kOe. It is important to remark that there is a critical concentration of the *non-magnetic* rare-earth, $x_c \sim 0.7$, for which the total magnetization of the system becomes zero at $T = 5$ K. Consequently, for concentrations below x_c the magnetization of the rare-earth sublattice dominates over the Fe one and it determines the direction of the total magnetization of the system, whereas the contrary holds for $x > x_c$.

The $M(H)$ curves recorded at $T = 5$ K on the $\text{Ho}_{0.5}\text{Lu}_{0.5}(\text{Fe}_{1-y}\text{Al}_y)_2$ compounds are displayed in figure 2(b). In this case, since the Lu content is always lower than x_c for all the members of the $\text{Ho}_{0.5}\text{Lu}_{0.5}(\text{Fe}_{1-y}\text{Al}_y)_2$ series, the Ho magnetic sublattice always dominates the direction of the total magnetization. The saturation magnetization

increases when Fe is progressively diluted by Al due to the antiferromagnetic coupling between Fe and Ho magnetic sublattices. In contrast to the behaviour found for the $\text{Ho}_{1-x}\text{Lu}_x\text{Fe}_2$, the saturation magnetization through the $\text{Ho}_{0.5}\text{Lu}_{0.5}(\text{Fe}_{1-y}\text{Al}_y)_2$ series does not follow a linear variation, as it would be expected from a simple dilution effect [31]. These magnetization measurements confirm the results obtained from X-ray absorption spectroscopy (XAS) studies performed on the $\text{Ho}_{1-x}\text{Lu}_x(\text{Fe}_{1-y}\text{Al}_y)_2$ series. The substitution of the rare-earth by Lu acts as a simple magnetic dilution effect whereas the Fe-Al substitution leads to a decrease of the $R(5d)$ -Fe(3d) hybridization and induces magnetic disorder into the Fe sublattice [32, 33].

3.2. XMCD measurements

To get a deeper insight into the origin of these two different magnetic trends we will study how the polarization of the conduction band states changes through the $\text{Ho}_{1-x}\text{Lu}_x(\text{Fe}_{1-y}\text{Al}_y)_2$ series depending on which magnetic atom, Ho or Fe, is substituted. As a first step, we will determine if the behaviour observed in the magnetization and XAS measurements is also reflected in the XMCD signals recorded at the Fe K- and Ho L_2 -edges. Additionally, studying the variation of the XMCD signal at Lu $L_{2,3}$ -edges will be crucial to discern which magnetic atom mainly determines the magnetic polarization of the Lu 5d states.

Fe K-edge and Ho L_2 -edge

Figure 3 shows the Fe K- and Ho L_2 -edge XMCD signals through the $\text{Ho}_{1-x}\text{Lu}_x(\text{Fe}_{1-y}\text{Al}_y)_2$ series recorded at $T = 5$ K and $H = 50$ kOe. In panels (a) and (d), the dichroic signals have been displayed with the sign obtained from the experimental measurements. As commented above, LuFe_2 and $\text{Ho}_{0.25}\text{Lu}_{0.75}\text{Fe}_2$, $x > x_c$, undergo a change of the dominant magnetic sublattice and, accordingly, the characteristic features of the dichroic signal have opposite sign than the rest of the compounds [27]. In order to be able to discern the evolution of the different dichroic contributions, the XMCD spectra has been referred to the direction of the magnetization of the Ho sublattice in the rest of the panels of figure 3, i.e., the sign of the dichroic signals for $x = 0.75$ and 1 has been reversed.

The Fe K-edge spectra are composed by two different magnetic contributions, the Fe one, XMCD_{Fe} , is more intense close to the absorption threshold (peak A) while the contribution due to the magnetic rare-earth, XMCD_{Ho} , extends over a wider energy range and dominates the shape of the XMCD spectra (peaks B, C_1 , C_2 and D), see figure 3(b) and (c).

In the case of the $\text{Ho}_{1-x}\text{Lu}_x\text{Fe}_2$, see figure 3(b), i.e., when only Ho is substituted by Lu, both the shape and the energy position of the main spectral features remain unvaried. However, the amplitude of the peaks associated to XMCD_{Ho} (B, C_1 , C_2 and D) decreases as the Lu content increases. The uniformly decrease of these XMCD_{Ho}

Figure 3. XMCD spectra at the Fe K- [panels (a), (b) and (c)] and at the Ho L_2 -edges [panels (d), (e) and (f)] for $\text{Ho}_{1-x}\text{Lu}_x(\text{Fe}_{1-y}\text{Al}_y)_2$ series recorded at $T = 5$ K and $H = 50$ kOe. Upper panels (a) and (d) display the dichroic signals with their signs obtained experimentally and shifted for clarity. Dichroic signals of $\text{Ho}_{1-x}\text{Lu}_x\text{Fe}_2$ are displayed in the panels (a), (b), (d) and (e): $x = 0$ (black, ∇), 0.25 (red, $\bullet$), 0.5 (green, $\blacktriangle$), 0.75 (blue, $\square$) and 1 (cyan, $\blacklozenge$); and $\text{Ho}_{0.5}\text{Lu}_{0.5}(\text{Fe}_{1-y}\text{Al}_y)_2$ ones in the lower panels: $y = 0$ (green, $\blacktriangle$), 0.25 (magenta, $\circ$), 0.5 (black, $\blacksquare$), 0.75 (orange, $\bullet$) and 1 (dark yellow, solid line). Insets in panels (e) and (f) shows the XMCD_{Fe} contribution extracted from the Ho L_2 -edge XMCD spectra of $\text{Ho}_{1-x}\text{Lu}_x\text{Fe}_2$ and $\text{Ho}_{0.5}\text{Lu}_{0.5}(\text{Fe}_{1-y}\text{Al}_y)_2$, respectively. See text for details.

features with the Ho concentration indicates that the substitution of Ho by Lu acts as a simple magnetic dilution effect.

By contrast, the substitution of Fe by Al, see figure 3(c), affects the shape, energy position and amplitude of the XMCD spectra. More specifically, the increase of the Al content diminishes the intensity of the main dichroic features (B , C_1 , C_2 and D) which shift towards lower energy.

It has been previously determined that the magnetic moment remains close to its free-ion value through the $\text{Ho}_{0.5}\text{Lu}_{0.5}(\text{Fe}_{1-y}\text{Al}_y)_2$ series [16]. Consequently, the observed reduction of the XMCD_{Ho} contribution to the Fe K-edge is in agreement with previous findings reporting a progressive reduction of the R-Fe exchange induced by the Al substitution through the reduction of the $\text{Ho}(5d)\text{-Fe}(3d)$ hybridization [32].

The results obtained at the Fe K-edge are confirmed by the XMCD measurements performed at the Ho L_2 -edge through the $\text{Ho}_{1-x}\text{Lu}_x(\text{Fe}_{1-y}\text{Al}_y)_2$ series.

Ho L_2 -edge XMCD spectra recorded at $T = 5$ K and $H = 50$ kOe through both series are shown in panels (d)-(f) of figure 3. In the case of compounds without Fe, $\text{Ho}_{0.5}\text{Lu}_{0.5}\text{Al}_2$, the dichroic signal is composed by a main negative peak A and a positive one B at higher energies. As the amount of Fe increases, the intensity of peak A decreases and, in several cases a weak positive peak A_1 arises. This A_1 peak is associated to the Fe contribution at the Ho L_2 -edge XMCD. Indeed, its intensity remains constant through the $\text{Ho}_{1-x}\text{Lu}_x\text{Fe}_2$, i.e., when the Fe content is maintained fixed, while it decreases through the $\text{Ho}_{0.5}\text{Lu}_{0.5}(\text{Fe}_{1-y}\text{Al}_y)_2$ as the Fe content does. By contrast, the intensity of peak B does not vary with the Fe content but with the Ho one, which demonstrates that this peak is mainly due to the Ho contribution.

The influence of the Fe sublattice in the Ho L_2 -edge XMCD spectra can be evaluated by extracting the XMCD_{Fe} contribution. To this end we have subtracted the dichroic signal of $\text{Ho}_{0.5}\text{Lu}_{0.5}\text{Al}_2$, i.e., of a homologous compound which does not contain Fe, from the Ho L_2 -edge XMCD spectra through the series. The results indicate that the XMCD_{Fe} extracted from the $\text{Ho}_{1-x}\text{Lu}_x\text{Fe}_2$ spectra almost does not vary through the series. In fact, the intensity of the main peak matches for all the $\text{Ho}_{1-x}\text{Lu}_x\text{Fe}_2$ compounds, see inset of figure 3(e). By contrast, the intensity of the XMCD_{Fe} signal decreases through the $\text{Ho}_{0.5}\text{Lu}_{0.5}(\text{Fe}_{1-y}\text{Al}_y)_2$ series upon substitution of Fe by Al [see inset of figure 3(f)]. This amplitude reduction cannot only be accounted for in terms of the dilution of the Fe sublattice since it does not vary linearly with the Fe content (as it will be shown in the inset of figure 5(b)). Indeed, the substitution of Fe by Al induces magnetic disorder in the $3d$ magnetic sublattice as well as the modification of the electronic structure, as confirmed by magnetization and XAS measurements [32, 33]. Consequently, the observed decrease of the XMCD_{Fe} contribution can be ascribed to the reduction of the $\text{Ho}(5d)\text{-Fe}(3d)$ hybridization associated with the modification of the magnetic properties of the $3d$ magnetic sublattice.

Lu $L_{2,3}$ -edges

So far we have shown that the dilution of the Fe and Ho magnetic sublattices affects differently the XMCD spectra recorded at both Fe K- and Ho L_2 -edges. In

Figure 4. Lu $L_{2,3}$ -edges XMCD signals measured at $T = 5$ K and $H = 50$ kOe for $\text{Ho}_{1-x}\text{Lu}_x\text{Fe}_2$ series: 0.25 (red, $\bullet$), 0.5 (green, $\blacktriangle$), 0.75 (blue, $\square$) and 1 (cyan, $\blacklozenge$). Upper panels (a) and (c) display the dichroic signals with their signs obtained experimentally and shifted for clarity. Inset in panel (b) displays the intensity difference of peaks B, D and F respect to the LuFe_2 signal. See text for details.

brief, the Ho-Lu substitution through the $\text{Ho}_{1-x}\text{Lu}_x\text{Fe}_2$ series acts as a simple magnetic dilution and does not modify the Ho(5d)-Fe(3d) hybridization. By contrast, the Fe-Al substitution through the $\text{Ho}_{0.5}\text{Lu}_{0.5}(\text{Fe}_{1-y}\text{Al}_y)_2$ series yields the depletion of the Ho(5d)-Fe(3d) hybridization and induces disorder into the Fe magnetic sublattice. Now, we will apply the previous results to the study of the magnetic polarization of the Lu 5d states. Our aim is to determine which magnetic species, Ho or Fe, is responsible for the appearance of a magnetic moment at the Lu sites.

To this end, we have performed a Lu $L_{2,3}$ -edges XMCD study of the $\text{Ho}_{1-x}\text{Lu}_x(\text{Fe}_{1-y}\text{Al}_y)_2$ series. Figure 4 shows the Lu $L_{2,3}$ -edges XMCD spectra of the $\text{Ho}_{1-x}\text{Lu}_x\text{Fe}_2$ series recorded at $T = 5$ K and $H = 50$ kOe. Panels (a) and (c) display the dichroic signals with the sign obtained from the experimental measurements and shifted vertically for clarity. Panels (b) and (d) the sign of the compounds with $x = 0.75$ and 1 has been reversed for better comparison. The Lu L_3 -edge dichroic signal, see figure 4(b), is composed by a main negative peak A located at $E-E_0 \sim 1$ eV and a less intense positive peak B at ~ 10 eV. The shape and intensity of the spectral feature A exhibit subtle

Figure 5. Lu L_{2,3}-edges XMCD signals measured at T = 5 K and H = 50 kOe for Ho_{0.5}Lu_{0.5}(Fe_{1-y}Al_y)₂ series: y = 0 (green, ▲), 0.25 (magenta, ○), 0.5 (black, ■), 0.75 (orange, ●) and 1 (dark yellow, solid line). Inset in panel (a) is an enlarged view of peak B. Inset in panel (b) compared the intensity of XMCD signal of peaks A and C with that of XMCD_{Fe} at the Ho L₂-edge. See text for details.

Figure 6. Comparison of the Lu L_{2,3}-edges XMCD spectra for Ho_{0.5}Lu_{0.5}(Fe_{0.25}Al_{0.75})₂ (orange, ●) and Ho_{0.5}Lu_{0.5}Al₂ (dark yellow, line) at T = 5 K and H = 50 kOe.

changes through the series, whereas the intensity of peak B decreases upon increasing the Lu content up to almost vanishing for LuFe₂. In the case of the Lu L₂-edge XMCD spectra, see figure 4(d), the dichroic signal is composed by a main peak C located at E-E₀ ~ 0 eV and two other less intense peaks, D and F, located at E-E₀ ~ 4 eV and ~ 7 eV, respectively. Peak C remains nearly unvaried upon dilution of the Ho sublattice whereas the intensity of both D and F peaks markedly decreases upon increasing the Lu content. In other words, the shape of the XMCD spectra at both Lu L_{2,3}-edges is mostly retained upon substitution of Ho by Lu and only minor changes are observed at both edges through the Ho_{1-x}Lu_xFe₂ series. Since the Fe content is kept constant through the Ho_{1-x}Lu_xFe₂ series and only the Ho content is varied, this suggests that the magnetic polarization at the Lu sites is mainly due to the Fe magnetic moments.

On the other hand, the intensity of the less intense peaks B (Lu L_3 -edge), D and F (Lu L_2 -edge) varies upon increasing the Lu content. The intensity difference respect to the LuFe_2 dichroic spectrum, see inset in figure 4(b), shows a linear decrease as the Ho content decreases which indicates that the intensity of these peaks is proportional to the Ho concentration. Therefore, this observation confirms that the effect of the magnetic polarization of the Lu $5d$ states due to the Ho atoms is clearly smaller than that due to the Fe ones.

Similar comparisons performed on the $\text{Ho}_{0.5}\text{Lu}_{0.5}(\text{Fe}_{1-y}\text{Al}_y)_2$ series are shown in figure 5. **The Lu L_3 -edge XMCD spectrum is composed by** a main negative peak A and a less intense peak B . In the present case, the intensity of the main peak A decreases notably as the Al content increases. The intensity of peak B is clearly less affected by the substitution and only a slight narrowing together with a shift to lower energies is observed. Similar results are found in the Lu L_2 -edge XMCD spectra [see panel (b) of figure 5]. The main peak C decreases and the peaks above the absorption edge, D and F , are less affected.

The observed reduction of the intensity of the main peaks A and C at the Lu L_3 - and L_2 -edge, respectively, indicates that the Fe polarization of the Lu $5d$ states is dramatically affected by the Al substitution. As shown in the inset of figure 5(b), the amplitude of the main dichroic peak at both Lu $L_{2,3}$ -edges follows the same dependence with the Fe content, a trend which also coincides with that of the XMCD_{Fe} contribution at the Ho L_2 -edge. Contrary to the case of the Ho-Lu substitution, this reduction does not scale with the Fe content. Moreover, the evolution of the dichroic signal when Al concentration increases suggest that for compounds with Al concentration equal or higher than 75% the Fe contribution has almost vanished. Indeed, as it is clearly shown in the enlarged view displayed in figure 6, the Lu $L_{2,3}$ -edges XMCD spectrum of $\text{Ho}_{0.5}\text{Lu}_{0.5}(\text{Fe}_{0.25}\text{Al}_{0.75})_2$ overlap with that of $\text{Ho}_{0.5}\text{Lu}_{0.5}\text{Al}_2$, which does not contain Fe. This implies that the magnetic polarization by the Fe atoms becomes negligible for compounds with low Fe concentration and only the polarization due to the Ho ones remains, which is clearly less intense. These results confirm our previous findings regarding the electronic impact of the Fe-Al substitution that cannot be regarded as a simple dilution effect in the $3d$ magnetic sublattice.

Further confirmation that the Fe sublattice is mainly responsible for the magnetic polarization of the Lu $5d$ states instead of the rare-earth one can be obtained by studying the temperature dependence of the Lu $L_{2,3}$ -edges XMCD signals. In these compounds, the magnetization of the Fe sublattice undergoes a decrease of $\sim 7\%$ from $T = 5$ K to ambient while the R magnetization is markedly reduced, around 50%, in the same temperature range. Accordingly, if Fe sublattice is mainly responsible for the magnetic polarization of the Lu $5d$ states, the intensity of the XMCD spectra should not vary appreciably between $T = 5$ K and room temperature. By contrast, if this polarization is mainly due to the Ho sublattice this intensity should be strongly depressed at high temperature.

By this reason, we have recorded the Lu $L_{2,3}$ -edges XMCD spectra of $\text{Ho}_{1-x}\text{Lu}_x\text{Fe}_2$

Figure 7. Comparison of Lu L_3 - (a) and L_2 -edges (b) XMCD signals at $T = 5$ K (dotted black line) and 288 K (solid red line) for $\text{Ho}_{0.75}\text{Lu}_{0.25}\text{Fe}_2$ and $\text{Ho}_{0.25}\text{Lu}_{0.75}\text{Fe}_2$. Dichroic spectra for each different compound have been shifted for clarity and those of the $x = 0.75$ have been displayed with the sign reversed for sake of comparison.

at $T = 288$ K. The comparison displayed in figure 7 shows that the amplitude of the XMCD spectra remain nearly unvaried and only a slight decrease is observed when the temperature increases. A detailed analysis of the evolution of the dichroic features yields that upon warming, from $T = 5$ K to 288 K, the intensity of the main peaks exhibits a decrease of $\sim 7.5\%$ which is in agreement with the decreasing of the magnetization of the Fe sublattice. By contrast, the relative variation of the intensity of the less intense peaks, associated to the Ho sublattice (see above), is about $\sim 76\%$, in agreement with the variation of the Ho sublattice magnetization. Thus, these results confirm that the appearance of a magnetic polarization at the Lu sites in the $\text{Ho}_{1-x}\text{Lu}_x(\text{Fe}_{1-y}\text{Al}_y)_2$ series is mainly due to the Fe magnetic moments.

Finally, we have obtained a quantitative determination of the magnetic moment of Lu through the studied series by applying the so-called XMCD sum rules [34, 35] to the Lu $L_{2,3}$ -edges XMCD spectra. These sum rules have been derived by connecting the integrated XMCD spectra at the Lu $L_{2,3}$ -edges with the ground-state expectation value of both $\langle L_z \rangle$ and $\langle S_z \rangle$ of the $5d$ states [36]:

$$\langle L_z \rangle = 2 (A_{L_3} + A_{L_2}) \frac{n_h}{\mu} \quad (1)$$

$$\langle S_z \rangle + \frac{7}{2} \langle T_z \rangle = \frac{3}{2} (A_{L_3} - 2A_{L_2}) \frac{n_h}{\mu}, \quad (2)$$

where A_{L_3} and A_{L_2} are the integrals over the dichroic signal at the L_3 and L_2 edges respectively, n_h is the number of holes in the $\text{Lu}(5d)$ band, $\langle T_z \rangle$ is expected value of the z component of the magnetic dipole operator and μ is the unpolarized $L_{2,3}$ -edges cross

Figure 8. LuFe₂ Lu L_{2,3}-edges XAS (black, solid line) and XMCD (green, ●) and the respective integrated curves. (see text for details).

section after subtraction of a double step function that ideally models the contribution of the continuum states (see figure 8) [37, 36]. No normalization to the absorption jump has been done either in the L₃ or the L₂ edge spectra in order to preserve the direct applicability of the sum rules.

The analysis of the XMCD signals has been performed under the following assumptions: i) μ is approximated by $\frac{3}{2}(\mu^+ + \mu^-)$; ii) $\langle T_z \rangle$ is assumed to be negligible in the spin sum rule; iii) estimates of both the orbital and spin moments, $\mu_{orb} = -\langle L_z \rangle \mu_B$ and $\mu_{spin} = -2\langle S_z \rangle \mu_B$ respectively, have been derived from the sum-rule by considering $n_h = 9$. It is well known that the sum rules cannot be applied to account for the L_{2,3}-edge XMCD spectra of those lanthanides showing a localized (4*f*) magnetic moment. However, in the case of La and Lu it has been largely demonstrated that the sign of the XMCD is the "right" one as there is no breathing effect. In this way the sum rules are commonly applied to determine the magnetic moment (of 5*d* origin) of Lu [36, 38].

Figure 9 show the results obtained after applying the sum rules procedure. For sake of comparison the magnetic moments have been displayed with their absolute value. In the case of Ho_{1-x}Lu_xFe₂ series, the magnetic moment of Lu, mainly of spin character (see table 2), does not vary appreciably whereas it decreases abruptly through the Ho_{0.5}Lu_{0.5}(Fe_{1-y}Al_y)₂ when the aluminum content increases. The decrease of the Lu magnetic polarization upon Fe substitution is in agreement with the previous discussion regarding the intensity variation of the dichroic signals. This confirms that the magnetic polarization at the Lu site is mainly due to the Fe atoms and that, in addition, not only the Fe content but also both the hybridization and the magnetic disorder affect the value of the magnetic moment induced at the Lu site.

Figure 9. Orbital (blue, $\circ$), spin (red, $\blacksquare$) and total (black, $\star$) magnetic moments of the Lu 5d states for $\text{Ho}_{1-x}\text{Lu}_x\text{Fe}_2$ and $\text{Ho}_{0.5}\text{Lu}_{0.5}(\text{Fe}_{1-y}\text{Al}_y)_2$ derived from the sum rules procedure. For sake of comparison absolute values of the values displayed in table 2 have been represented.

Table 2. Orbital and spin magnetic moments (in μ_B) of the Lu 5d states derived from the XMCD sum-rules analysis at T = 5 K. The uncertainty of these values is estimated to be of 15 %.

compound	μ_{orb}	μ_{spin}	$\mu_{\text{Lu}}(5d)$
$\text{Ho}_{1-x}\text{Lu}_x\text{Fe}_2$			
x = 0.25	-0.002	0.118	0.116
x = 0.50	-0.005	0.115	0.110
x = 0.75	0.006	-0.112	-0.106
x = 1	0.005	-0.116	-0.111
$\text{Ho}_{0.5}\text{Lu}_{0.5}(\text{Fe}_{1-y}\text{Al}_y)_2$			
y = 0	-0.005	0.115	0.110
y = 0.25	-0.004	0.056	0.052
y = 0.50	-0.002	0.024	0.022
y = 0.75	-0.002	0.008	0.006
y = 1	-0.001	0.008	0.007

4. Summary and Conclusions

Summarizing, we have presented here a systematic XMCD measurement on $\text{Ho}_{1-x}\text{Lu}_x(\text{Fe}_{1-y}\text{Al}_y)_2$ series to study the Lu 5d states magnetic polarization in the presence of competing polarizations, Fe and Ho localized magnetic moments. The analysis of the Fe K- and Ho L₂-edge XMCD spectra have proved that the substitution of

Fe by Al leads to a depletion of the Ho(5d)-Fe(3d) hybridization and induces magnetic disorder in the Fe sublattice, whereas the dilution of Ho by Lu behaves as a simple magnetic dilution. The evolution of the Lu L_{2,3}-edges XMCD spectra through the Ho_{1-x}Lu_x(Fe_{1-y}Al_y)₂ series shows that the magnetic polarization at the Lu site is constant through the Ho_{1-x}Lu_xFe₂ series and decreases upon increasing the Al content through Ho_{0.5}Lu_{0.5}(Fe_{1-y}Al_y)₂. This demonstrates that the appearance of a magnetic moment at the Lu sites is mainly due to the Fe magnetic moments. Estimation of the spin and orbital components of the Lu(5d) induced magnetic moment have confirmed these results and that the magnetic moment of Lu is mainly of spin character.

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