ESRF

Experiment title: **Moment Collapse in $U_xLa_{1-x}S$ investigated by X-ray magnetic circular dichroism at the uranium M edges**	Experiment number: HE-538

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Shifts:	**Local contact(s):**	*Received at ESRF:*
15	A. Rogalev	

Names and affiliations of applicants (*indicates experimentalists):

*A. Bombardi, CEA-Grenoble,
*P. Dalmas de Réotier, CEA-Grenoble
*N. Kernavanois, CEA-Grenoble
J.-P. Sanchez, CEA-Grenoble
P. Burlet, CEA-Grenoble
*A. Yaouanc, CEA-Grenoble
G.H. Lander, ITU-Grenoble
A. Rogalev, ESRF-Grenoble

Report:

The uranium monochalcogenides UX (X=S, Se, Te) compounds with FCC NaCl-structure are ferromagnets with easy axes $\langle 111 \rangle$. Magnetic properties of U compounds are generally ascribed to the magnetic moment associated to the $5f$ electrons at the U site. In UX compounds, the ordering temperature decreases whereas the magnetic moment, strongly reduced compared to the free ion value, increases with the X ion size. This clearly indicates that the features of the $5f$ electrons in these systems are strongly modified by hybridization with band and valence electrons. The role of the hybridization decreases going from US to UTe [1]. Since the magnetic ordering onset in these compounds results from a fine balance between hybridization and mediated exchange, it can be easily modified by applying an external perturbation, e.g. pressure or dilution by diamagnetic ions. Dilution was found to induce dramatic effects on magnetism. According to neutron diffraction measurements [2], replacing U by diamagnetic La in US and USe compounds, leads to the collapse of the magnetic ordering in zero field for a U concentration ($x_U \approx 0.57$) far above the percolation limit for the NaCl structure ($x_U = 0.14$). These measurements established an upper limit for the magnetic moment (m) of $\approx 0.2\mu_B$/U. Muon spin rotation experiments states that magnetic ordering is still present up to $x = 0.15$, but the magnetic moment collapses ($\approx 0.05\mu_B$) [3]. The collapse of the magnetic ordering is quantitatively described by an ab-initio model [4], based on the hypothesis that the f electron spectral density must be divided into two parts, a localized one that contributes to magnetic ordering and a delocalized one that is not effective from the magnetic point of view.

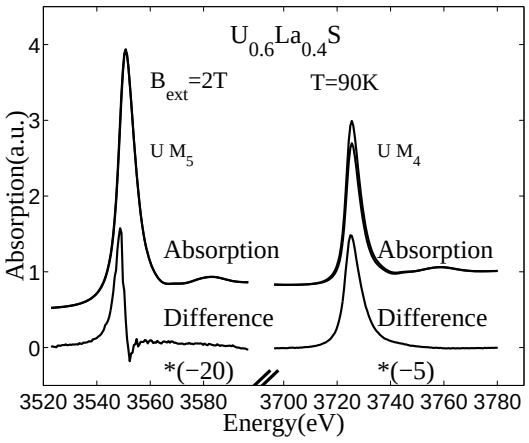

Figure 1: Absorption and dichroic signal at the M_5 and M_4 edges in $U_{0.6}La_{0.4}$.

One way to check the variation of the spatial extent of the $5f$ electrons wavefunction is to measure the ratio between the orbital, μ_L, and spin, μ_S, moments [5]. In principle X-ray magnetic circular dichroism (XMCD) experiments should be able to observe such a variation. For example in UFe_2 [6], where $5f$ electrons are almost completely delocalized, $-\frac{\mu_L}{\mu_S} \simeq 1$ whereas in US $-\frac{\mu_L}{\mu_S} \simeq 2.4$. For this experiment we have chosen three samples with x_U = 0.60, 0.40 and 0.30. $U_{0.6}La_{0.4}S$ behaves like pure US with a Curie temperature $T_C \simeq 100$ K and $m \simeq 1.3\mu_B$/U. Crystals with x_U = 0.40 and 0.30 do not show any magnetic ordering in neutron diffraction experiments. Their magnetic anisotropy is weak but their induced magnetic moments are considerable ($\approx 1\mu_B$/U at saturation in $x_U = 0.4$). In order to avoid crystal breaking we have chosen to perform the measurements on $U_{0.6}La_{0.4}S$ at T = 90 K close to T_C. On the other samples the measurements were performed at low temperature. The shape of the observed signals (see Figs. 1, 2 and 3) is similar to the one observed on pure US [7] and does not change along the series, whereas, as expected, the magnetic moment is reduced upon dilution. The XMCD $\langle L_z \rangle$ values poorly agree with $\mu_L \equiv -\langle L_z \rangle \mu_B$ obtained from neutron magnetic form factor measurements [8]. For instance, after normalisation to the saturation values to acount for the different experimental temperature, field and orientation conditions, XMCD on the $x_U = 0.60$ sample gives $\langle L_z \rangle \simeq -2.2(2)$ whereas neutrons give $\mu_L = 2.9(1)\mu_B$. The discrepancy is well beyond the error bars. The uncertainties given above are the uncertainties of the experiments. They do not take into account the normalisation procedure, based on bulk magnetisation measurements, which is needed to compare the XMCD and neutron results. Another origin for the difference between the two techniques arises from the fact the X-rays penetrate only the first 3000 Å of the sample whereas neutrons probe the bulk of the crystals. We also notice that the XMCD measurements may be affected by coherence effects which are not yet under control [9].

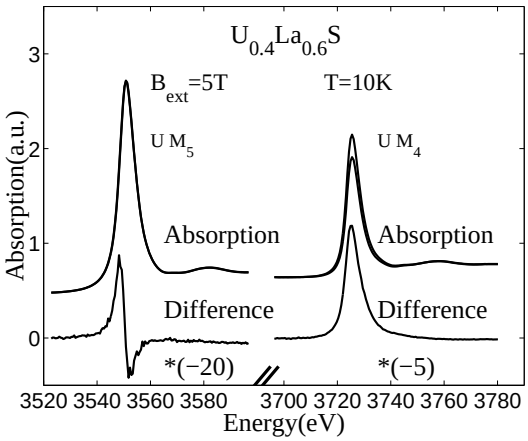

Figure 2: Absorption and dichroic signal at the M_5 and M_4 edges in $U_{0.4}La_{0.6}$

However part of the systematic uncertainties in XMCD compensate when the ratio $\frac{\langle L_z \rangle}{\langle S_e \rangle}$ (where S_e is the effective spin operator [7]) is considered. Comparing this ratio for the different examinated samples is meaningful as the experiments were performed in the same way. We find $\frac{\langle L_z \rangle}{\langle S_e \rangle}$ = $-1.72(8)$, $-1.44(7)$ and $-1.43(7)$ for the compounds with x_U = 0.6, 0.4 and 0.3 respectively. This abrupt change between x_U = 0.6 and 0.4 supports well the idea that when decreasing the U content, the localized $5f$ electron fraction falls abruptly below a critical value around $x_U = 0.57$ [4]. For comparison we give the $\frac{\langle L_z \rangle}{\langle S_e \rangle}$ values available for US (Collins et al [7] $[-1.65(8)]$ and Kernavanois et al [10] $[-1.56(6)]$).

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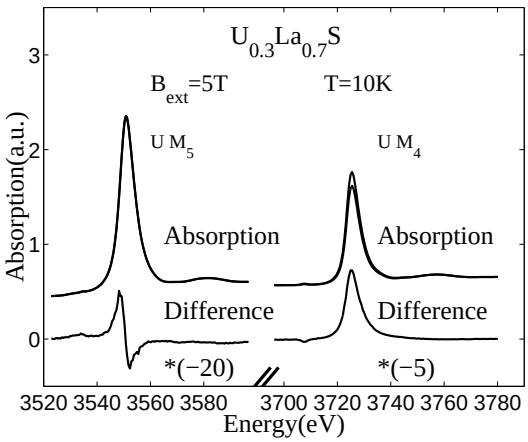

Figure 3: Absorption and dichroic signal at the M_5 and M_4 edges at $x = 0.3$.