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| ESRF | **Experiment title:** Possible input of the higher J=7/2 multiplet on ground state properties in the Kondo insulators $CeRu_2Al_{10}$ and $CeOs_2Al_{10}$ | **Experiment number**: HC-679 |
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## Report:

The orthorhombic Kondo insulators $CeM_2Al_{10}$ with M = Ru, Os and Fe have attracted much interest because of their unusual magnetic properties. $CeRu_2Al_{10}$ and $CeOs_2Al_{10}$ order antiferromagnetically below $T_0$ = 27 and 29 K, which cannot be explained by RKKY-type interactions because the Ce-Ce spacings are large and de-Gennes-scaling implies ordering temperatures below 1 K. On the contrary $CeFe_2Al_{10}$ does not exhibit any magnetic order at all.

It is known that the Kondo screening becomes increasingly important from M = Ru to Os to Fe. Detailed neutron diffraction and μSR experiments revealed the long range nature of the magnetic order and also that the moments order along the *c*-axis and not along the easy axis *a* [1]. The general anisotropy of the static susceptibility and the small size of the ordered magnetic moment can be largely explained within a crystal electric field (CEF) model, which we have determined via linearly polarized x-ray absorption spectroscopy experiments at the Ce $M_{4,5}$ edge [2]. However, deviations from the measured susceptibility occur. This can be due to various reasons since anisotropic molecular fields, anisotropic 4*f*-conduction electron hybridization (*c*-*f* hybridization) and/or contributions from the higher multiplet are not considered.

From full multiplet simulations (XTLS program by A. Tanaka, see Ref. [2]) we know that the linear dichroism (LD) is sensitive to contributions from the J=5/2 multiplet, but it is not to contributions from the J=7/2 multiplet. Usually, in Ce compounds the CEF splitting is much smaller than the spin-orbit splitting ($\Delta E_{so} \approx 280$ meV) and one can work in the so-called Stevens approximation, neglecting the higher multiplet. Yet, in $CeM_2Al_{10}$ the CEF splittings are known to be fairly large (about 30 and 46 meV), as can be seen in inelastic neutron scattering experiments [2]. When modeling the magnetic susceptibility with our CEF results from Ref. [2] we found indications for a non-negligible contribution of the J=7/2 multiplet in the CEF ground-state wave function. Namely, it turned out that there are clear changes in the static susceptibility when taking into account the higher multiplet, especially for the b direction, and that these changes might explain the differences between the CEF-only and the measured magnetic susceptibility.

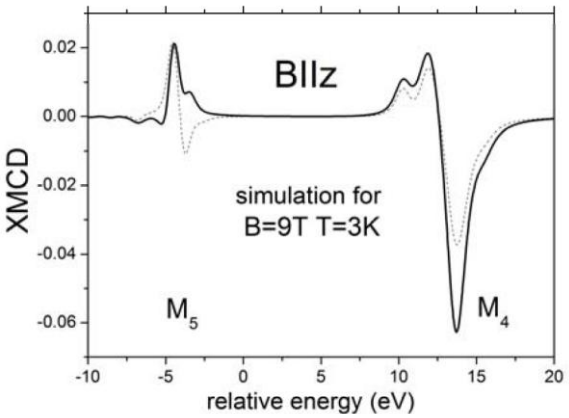


**Fig. 1:** Simulated XMCD for two CEF models. Solid lines show the Stevens approximation and dotted lines show the case where contributions from the higher J=7/2 multiplet are included.

The polarization originating from the J=7/2 states is too small to be detected in an LD experiment, however, preliminary simulations showed that the presence of J=7/2 in the ground state should become visible in the XMCD signal: The line shape for the ground-state wave function including contributions from the higher multiplet differs clearly from the line shape for the wave function neglecting the higher multiplet (see Fig. 1).

Single crystals of $CeRu_2Al_{10}$ and $CeOs_2Al_{10}$ were grown by an Al self-flux method and their quality and orientation were confirmed by Laue x-ray diffraction. All XMCD spectra were recorded in the total electron yield (TEY) mode at a pressure of $3 \cdot 10^{-3}$mbar and with a magnetic field of B = 9 T. The TEY was normalized to the incoming photon flux as measured on the Au mesh before the entrance of the experimental chamber. Clean sample surfaces were obtained by cleaving the samples *in situ*. Entrance and exit slit were set to 30µm. Both polarizations (left/right circular polarized) were recorded on the same sample spot guaranteeing a reliable comparison of the spectral line shapes.

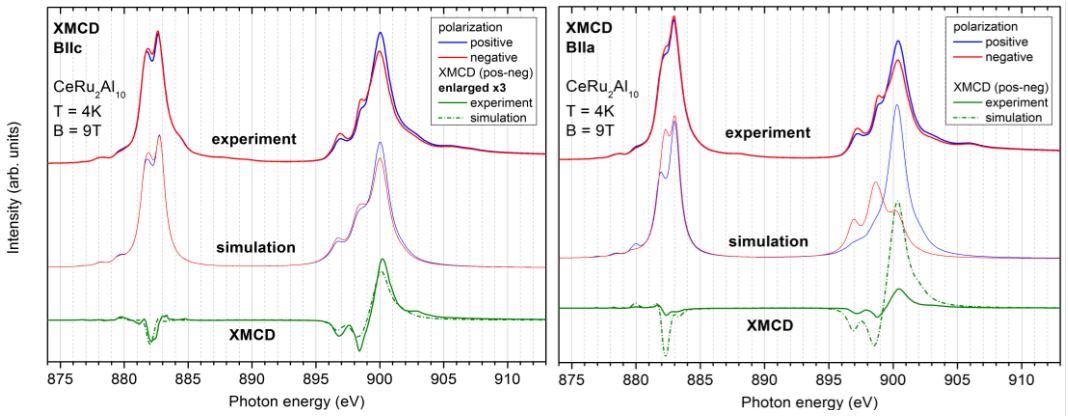


**Fig. 2:** Two sets of XMCD spectra of $CeRu_2Al_{10}$, measured at 4 K and with the magnetic field B||*c* (left panel) and B||*a* (right panel) and B = 9 T.

In Fig. 2 two sets of XMCD spectra of $CeRu_2Al_{10}$ are shown, namely for B||*c* (left) and for B||*a* (right). All spectra were taken at 4 K, thus only the ground state is probed. In each panel the full multiplet simulation is plotted (middle) below the experimental data (top). **The simulations use the CEF parameters exactly as determined in Ref. [2]** without including any contribution from the J=7/2 multiplet. At the bottom the XMCD signal (the difference spectra) expected from the simulation (dashed) and the measured one (solid line) are plotted on top of each other. In this preliminary analysis the agreement between experiment and simulation for B||*c* is very good. The line shape and even the intensity of the XMCD signal are nicely reproduced. This is also the case for B||*b* (not shown), but here the magnitude of the difference signal deviates a bit more from the theoretical expectation. However, for B||*a* the measured XMCD is much smaller than the one expected from the CEF model, although the general line shape is somehow reproduced. In Fig. 3 the experimental and simulated difference spectra for $CeOs_2Al_{10}$ are shown for B||*a*, B||*b* and B||*c*. Here we observe the same behavior as for $CeRu_2Al_{10}$: The agreement with the theoretical expectation from the CEF model [2] is very good for B||*c*, diminishes slightly for B||*b* and is worst for B||*a*.

One might conclude that the suspected influence of the J=7/2 multiplet is minor since the **general line shape** of the XMCD spectra in all three direction corresponds to the one expected from the CEF model determined in Ref. [2] without J=7/2 contributions (see also Fig. 1). Then the question remains why the **size** of the polarization dependence deviates that much for B||*a* (and to some extent also for B||*b*). At this point one might speculate that the anisotropic *c*-*f* hybridization plays an essential role: It is supposed to be strongest along the *a*-direction, thereby making the *c*-axis the preferred direction for the ordered moment. A detailed analysis, whether including J=7/2 in the initial state can explain the deviations to some extent or whether the Kondo effect is responsible, is in progress.

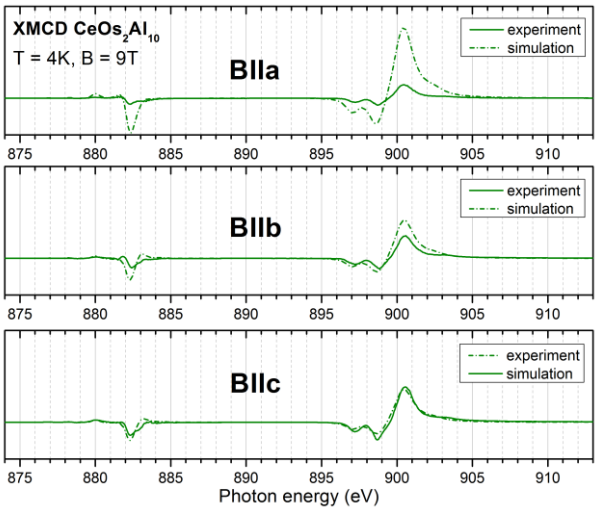


**Fig. 3:** Measured and simulated XMCD difference spectra of $CeOs_2Al_{10}$, measured at 4 K and with the magnetic field B = 9 T.


References:

[1] Robert *et al.*, PRB 82, 100404(R) (2010); Khalyavin *et al.*, PRB 82, 100405(R); Adroja *et al.*, PRB 104405 - all (2010).

[2] Strigari *et al.*, PRB 86, 081105(R) (2012) and PRB 87, 125119 (2013).