Experimental report - Beamtime HC 4147

Ce M5-edge RIXS study on the giant crystal field compound $CeRh_3Si_2$

The samples (3 single crystals of $CeRh_3Si_2$) have been cleaved in vacuum just prior to their insertion in the measuring chambers. The quality of the cleaved surfaces was checked with a x ray absorption (TEY-XAS) measurement, which showed a that cerium was not oxidized, and the linear dichroism confirmed the single crystallinity of the samples.

RIXS spectra have been acquired with about 40meV resolution for several different scattering geometries, polarizations and incident photon energies (a selection of spectra in Fig. 1).
Five inelastic peaks (+ the elastic line at 0meV) were clearly visible in the spectra, and the variation of the intensity ratio of the different excitations with the experimental settings allowed us to better define the position of each peak.

The spectra for T=20K were fit with 6 Lorentzian functions (plus a Gaussian at 0meV for the elastic signal), and this provided us a good estimate of the peak positions (Fig. 1 and 2).

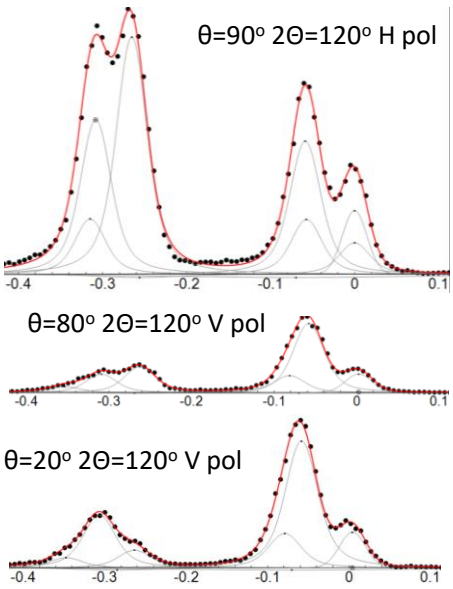

Figure 1: Some RIXS data acquired at 20K, and Voigt fit. The sample was oriented so that *c* was normal to the cleaved surface and *a* in the scattering plane

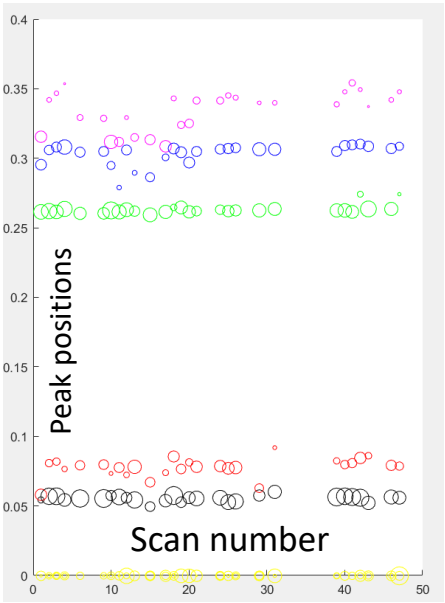

Figure 2: Peak positions resulting from the Voigt fit of the spectra acquired at T=20K, each spectrum having different experimental settings (polarization, geometry, photon energy…)

Average position

345 meV
307 meV
263 meV

77 meV
56 meV
0 meV

We analyzed the RIXS data using the full multiplet code Quanty.
To ease the analysis and reduce the number of parameters, we approximated the crystal field acting on Ce with that of an hexagonal lattice (hexagonal axis = *a*).
This approximation, which was considered reasonable due to pseudo-hexagonal crystal structure of $CeRh_3Si_2$ (Fig. 3), was further confirmed by the good fit of the XAS spectra obtained with an hexagonal crystal field simulation, with ground state Jx=1/2 (note, we chose the notation Jx instead of Jz to be consistent with our choice of the quantization axis as the *a* crystallographic axis).

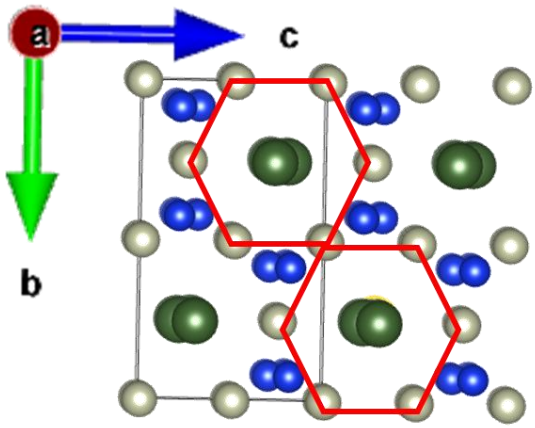

Fig. 3: Crystal structure of $CeRh_3Si_2$, with the pseudo-hexagonal lattice highlighted around the Ce ions (green spheres)

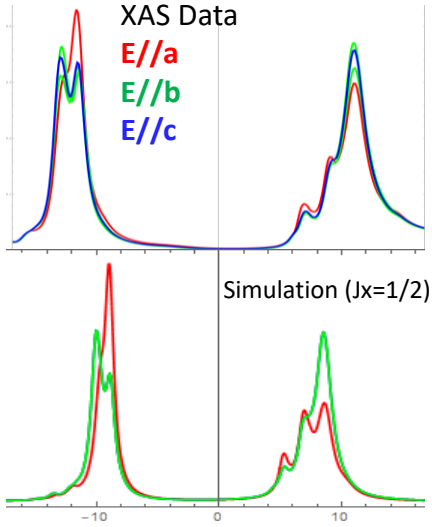

Fig. 4: XAS data (top) acquired during the beamtime and simulation (bottom) supposing an hexagonal crystal field and a Jx=1/2 ground state. The experimental E//b spectrum was slightly different on different cleaved surfaces (see two green lines in the top panel), probably due to surface problems.

The XAS spectra acquired on different spots or different cleave of the sample are slightly different (see green line) due to the surface sensitivity of TEY-XAS, but the same variation was not observed in the RIXS spectra, much more bulk sensitive.

The measured crystal field splittings and the Jx=1/2 symmetry of the ground state limited the possible sets of (A20, A40, A60) crystal field parameters to a specific region of the phase space (Fig. 5) .
In Fig. 6 we show some RIXS spectra and the corresponding fit with a crystal field simulation (A20=-0.13 eV, A40=0.18 eV, A60=0.035 eV) for both T=20K and 300K data. These parameters also offer a good fit of the magnetic susceptibility data (Fig. 7).

To summarize, our experiment found a Jx=1/2 ground state symmetry and rather large crystal field splittings of the order of 60meV. This size of the crystal field causes a non-zero, although negligible, mixing of the two spin-orbit split multiplets. The crystal field scheme didn't show big changes with temperature in the 20K-300K range.

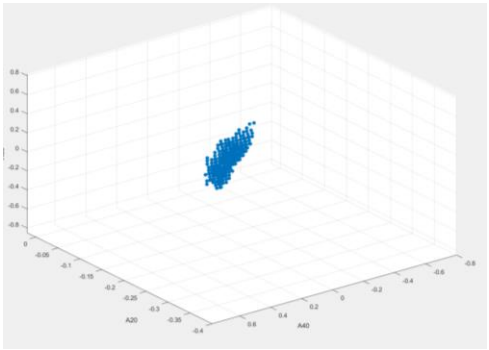

Fig. 5: Region of the A20, A40, A60 crystal field parameter space which corresponds to the measured splittings (obtained from RIXS) and ground state symmetry (obtained from XAS)

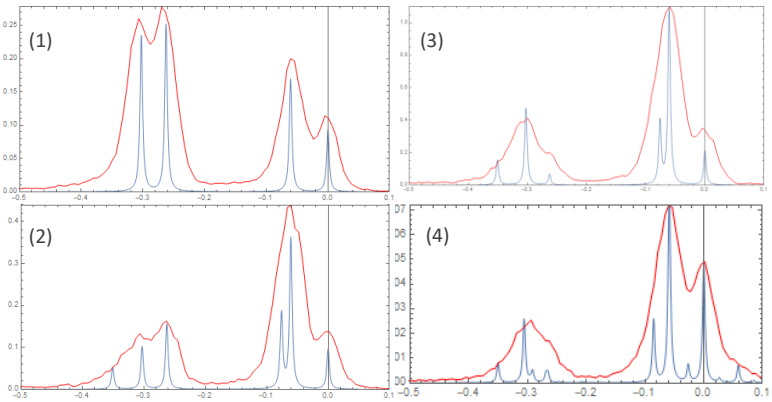

Fig. 6: RIXS spectra (red) and corresponding crystal field simulation with A20=-0.13, A40=0.18, A60=0.035 eV. (1-3) T=20K spectra already shown in Fig.1, (4) T=300K spectrum with same scattering geometry as spectrum (3), note the appearance of anti-Stokes peaks at high temperature.

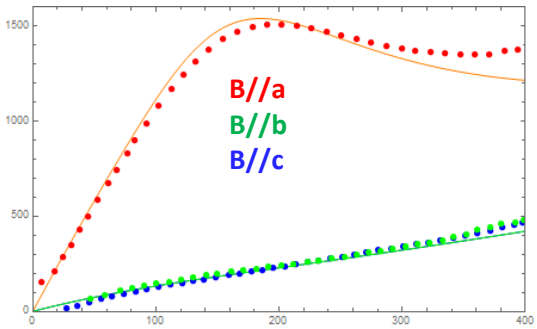

Fig. 6: Magnetic susceptibility data (dots) and fit (lines) with the crystal field parameters obtained from the present experiment

These results shall be compared with the parent compound $CeRh_3B_2$, which is characterized by reduced moments and high T_C=115K ferromagnetism, opposed to the T_N~4K antiferromagnetic behavior of $CeRh_3Si_2$.
In $CeRh_3B_2$ we found the same ground state symmetry (Jz=1/2), but with much larger overall splitting of the 4f levels and broader peaks, which are signs of stronger influence of the ligands on the 4f levels (experiment HC3582) .
We want to use the information gained with the two experiments to fully characterize the 4f levels and investigate the underlying reasons for the two very different magnetic behaviors.

We also dedicated few hours to perform a feasibility test for a future experiment on Ce metal. The test showed that Ce metal is almost impossible to post-cleave in vacuum in the ID32 loadlock. Moreover, we found out that a fresh surface degrades quickly when irradiated by X-rays in the 10^{-9} mbar atmosphere of the RIXS sample chamber. These tests were very useful because provided us with the needed knowledge to plan a Ce metal experiment with improved experimental methods. This will be the topic of a future proposal.