Experimental report beamtime HC 3582

Crystal field, intermultiplet mixing and magnetic excitations in $CeRh_3B_2$ probed with Ce M_5 edge RIXS

The samples (2 single crystals of $CeRh_3B_2$) 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 sample.

At first, a relaxed resolution (about 40 meV) was used to quickly acquire the first RIXS spectra. We then decided to optimize the resolution to better distinguish each one of the many features in the spectrum.

The spectra (Fig.1) showed the presence of 6 excitations (+ the elastic line at 0meV), already providing much more information compared to the published Inelastic Neutron Scattering data, which consisted of only one peak at about 150meV (Fig. 2). These excitations persisted in the whole temperature range explored, from 20K to 320K, i.e. from well below to well above the T_{Curie}=115K, so we attributed them to electronic excitations within the 4f shell (*ff* excitations, or crystal field excitations) and not to collective magnetic excitations, which are only possible in the ordered state(T< T_{Curie}).

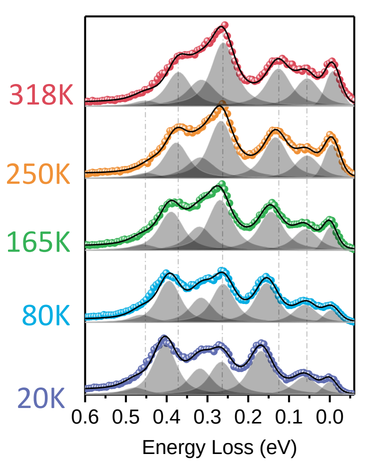

Figure 1: ID32 RIXS data from 20K to 320K and Voigt fit

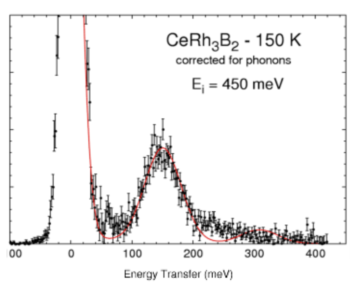

Figure 2: Inelastic Neutron Scattering data from A. Givord *et al.* J. Phys.: Condens. Matter 19 (2007) 506210

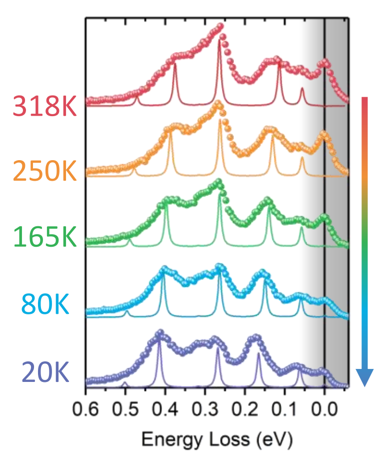

Figure 3: ID32 RIXS data and best crystal field simulation (thin ines)

The presence of the first crystal field excitation at about 60meV is of great importance because it was not visible in the previously available Inelastic Neutron Scattering spectra, due to limitations in the range of excitations reachable by neutron spectroscopy (ΔJ_z^{INS}=0,1 while ΔJ_z^{RIXS}=0,1,2). This peak indicates that the crystal electric field in $CeRh_2B_2$ is much less strong than what thought before (the first INS excitation detected was at 150meV). As a consequence, the spin orbit splitting of about 300meV between the two multiplets $^2F_{5/2}$ and $^2F_{7/2}$ is still the larger energy scale, and the crystal field is not strong enough to generate a mixing of the two multiplets. This is already a great result, because a multiplet mixing driven by the “giant crystal field” in $CeRh_2B_2$ was considered in literature as one of the possible reasons to explain the unusual magnetic properties.

The energies of each crystal field excitation were obtained from a 6-Voigt fit of the 320K spectrum and subsequently compared with simulations to find the best set of crystal field parameters describing the system (Fig. 3). The crystal field scheme (Fig. 4) found confirmed that the first excitation was not reachable by the INS cross section (ΔJ_z=2). The ground state wavefunction turns out to be the “J_z=1/2”, which is elongated along the *c* direction, i.e. elongated towards the closest Ce ion. This suggests a direct Ce-Ce interaction along the *c* axis.

The analysis of the temperature dependence showed a strong variation of the peaks’ positions with temperature. This must be related to the large (and anomalous) changes in the lattice constants with temperature. Repeating the crystal field analysis for each temperature allowed us to study the change of the crystal field potential with temperature (Fig. 3,4,5). Such analysis revealed that at low T the potential becomes stronger along *c*, further confirming the previous idea of interactions happening along the *c* axis.

Then we checked for possible dispersions of the strongly hybridized 4f states, but the spectra were not showing clear signs of dispersion (Fig. 5).

The acquired data are of great importance for the understanding of the physics of $CeRh_3B_2$, since they allow us to finally map the full Ce 4f crystal field scheme. We are now performing DFT+DMFT simulations to further understand how this new information on the crystal field scheme relates with the tentative explanation previously provided by theoreticians about the causes leading to the large T_{Curie} ferromagnetism with strongly reduced magnetic moments present in $CeRh_3B_2$. Overall, we consider this experiment successful and we foresee a publication of this study soon.

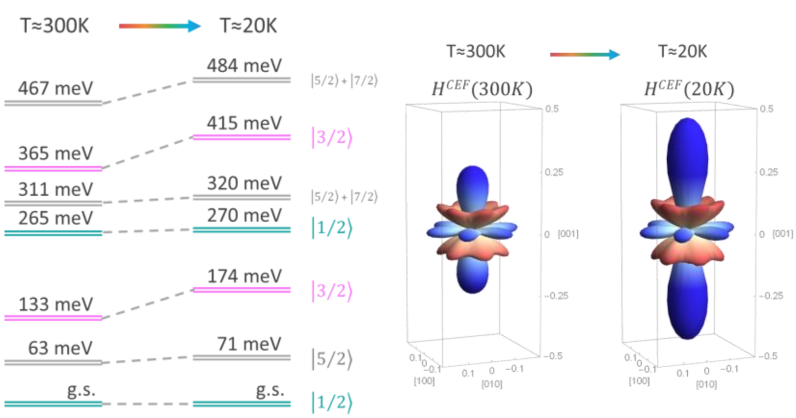

Figure 4: Crystal field scheme and its evolution with temperature (left). Shape of the crystal field potential seen by Ce ions, according to our best fits (right)

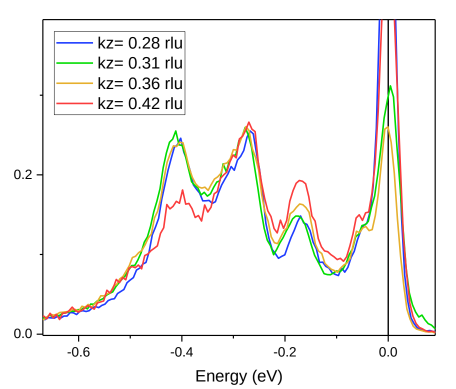

Figure 5: ID32 RIXS data acquired with different scattering geometries to vary the momentum transfer. No clear dispersion is visible.