ESRF	**Experiment title: Determining the in-plane orientation of the crystal-field ground state orbital of the heavy fermion compound CeCoIn5 with non resonant inelastic scattering.**	**Experiment number**: HC-992
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Shifts:	**Local contact(s)**: Marco Moretti	

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The in-plane orientation of the *4f* ground state orbital of the heavy fermion compound $CeCoIn_5$ [1] has been determined with vector **q**-dependent non resonant inelastic scattering (NIXS) at the Ce^{3+} $N_{4,5}$ edge. Here the vector **q**-dependence gives access to the initial state symmetry in analogy to the polarization dependence in an x-ray absorption experiment.

Report

In a tetragonal crystalline electric field the Hund's rule ground state of Ce^{3+} with J=5/2 splits into three Kramers doublets which can be represented in the basis of $|J_z\rangle$. Two doublets have Γ_7 symmetry, $\Gamma_7^1=\alpha|^{+}/_{-}5/2\rangle + \sqrt{(1-\alpha^2)}\, |^{-}/_{+}3/2\rangle$ and $\Gamma_7^2 = \sqrt{(1-\alpha^2)}\, |^{+}/_{-}5/2\rangle - \alpha\, |^{-}/_{+}3/2\rangle$, and one is a $\Gamma_6 = |^{+}/_{-}1/2\rangle$. The ground state of CeCoIn5 is a Γ_7. The absolute size of the mixing parameter $|\alpha|$ characterizes the anisotropy between the crystallographic *c* axis and the *ab*-plane and it has been measured for $CeCoIn_5$ with inelastic neutron scattering and linear polarized x-ray absorption spectroscopy [2,3]. Figure 1 shows the ground state orbital of $CeCoIn_5$ as determined from XAS [2]. However, soft XAS and also neutron scattering are dipole methods and therefore not able to detect anisotropies with a higher than twofold rotational symmetry. This has the consequence that the sign of α cannot be determined with these techniques. Since α determines the orientation of the orbital within the lattice, the latter is still unknown. Theories trying to explain ground state properties should take the CEF ground state orbital into account, however, this makes only sense when the entire information – including the orbital orientation - is available.

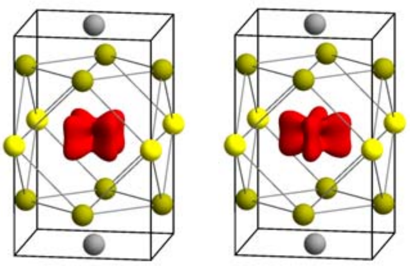

Fig. 1 Crystal structure of $CeCoIn_5$ and the two possible orientations of the cerium Γ_7 crystal-field ground state. Both 4*f* orbitals have the same |+-5/2> admixture |α| but different signs of α.

At the example of $CeCu_2Si_2$ [4] we could show that the inelastic scattering functions S(**q**,ω) at the cerium $N_{4,5}$ edge in a non-resonant inelastic x-ray scattering experiment (NIXS) exhibits differences at large momentum transfers between the two directions **q**||[100] and **q**||[110]. We could show further that these differences are due to the anisotropy of the crystal-field ground state in the (001) plane on the basis of calculations with multipole selection rules, in particular higher multipole contributions.

Here we applied NIXS at the cerium $N_{4,5}$ edge to $CeCoIn_5$. We used the Si(111) monochromator and Si(660) anaylzers, yielding incident energies of about 9.8 eV. The corresponding resolution was 1.5eV. High momentum transfers are crucial for such an experiment, so that we used the horizontal geometry where the highest scattering angles can be reached. The high angle analyser box was set such that the analyser column (A1, A2, and A3) at the highest scattering angles was at 2θ=152.8°. This corresponds to a momentum transfer of |**q**| = 9.5 Å^{-1}. Two samples were mounted in the beam, one with a [100] surface and another one with a [110] surface so that S(**q**,ω) could be measured in specular geometry for **q**||<100> and **q**||<110>. The samples were cooled down to 6 K with a closed cycle cooler in order to assure only the ground state is populated. The closed cycle cooler was fitted with a double Be dome which is important to mention because the beryllium K edge (111.5 eV) appears at the same energy as the cerium $N_{4,5}$ edge (109 eV). However, thanks to the position sensitive detectors the signals from sample and Be dome could be separated.

Below the $CeCoIn_5$ NIXS data are shown for the two in-plane **q** directions <100> (blue) and <110> (green). Only a linear background has been subtracted. The left of Fig. 2 shows the sum of the analyser column at the highest accessible angle of 2θ = 152.8°. These data correspond to the sum of three analysers. The right of Fig. 2 shows the sum of the analyser columns at 152.8°, 146.4°, and 140.2°, respectively, i.e. the sum of nine analysers. The statistics is obviously better but the differences of the two directions are less pronounced.

The Fig. 3 shows the simulations of the scattering function S(**q**,ω) [5] for the highest possible angle. The simulations correspond to an orientation of the 4*f* orbital with the loops along <110> (left of Fig. 1) so that the experiment has answered the key question of the proposal. Figure 4 also shows simulations. Here the differences of the S(**q**||<100>,ω)-S(**q**||<110>,ω) are shown for the three highest angles. It shows that the vector **q** dependent effect diminishes with decreasing |**q**| and that this is already visible when going from 9.5 to 9.18 Å^{-1}, thus showing the necessity to work at momentum transfers as high as possible.

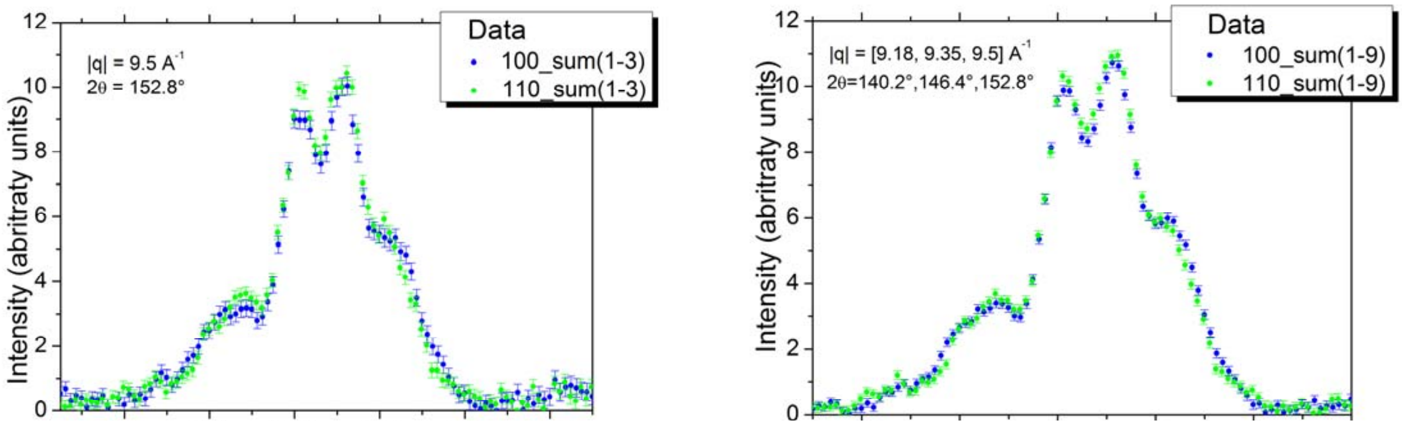

Fig. 2: NIXS data of two CeCoIn5 single crystals, blue for **q**||[100] and green for **q**||[110]. Right: Sum of **<u>three</u>** analyzers at the highest possible scattering angle of 152.8°. Left: Sum of **<u>nine</u>** analyzers at 140.2°, 146.4° and 152.8°:

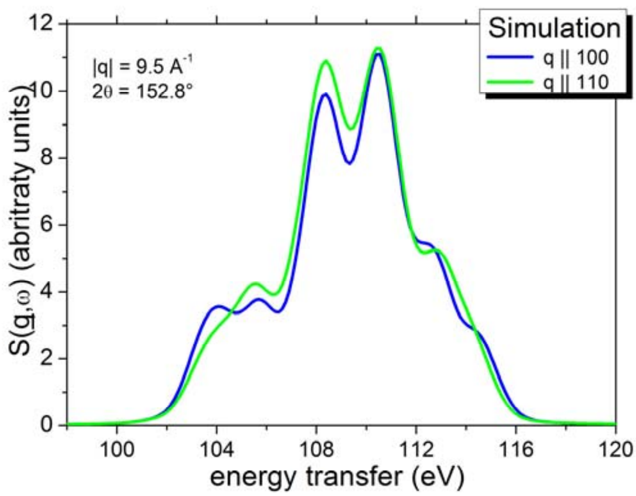

Fig. 3: Simulation of the scattering function S(**q**,ω) for **q**||<100> and **q**||<110> and |**q**| = 9.5 Å^{-1} which corresponds to the highest scattering angle.

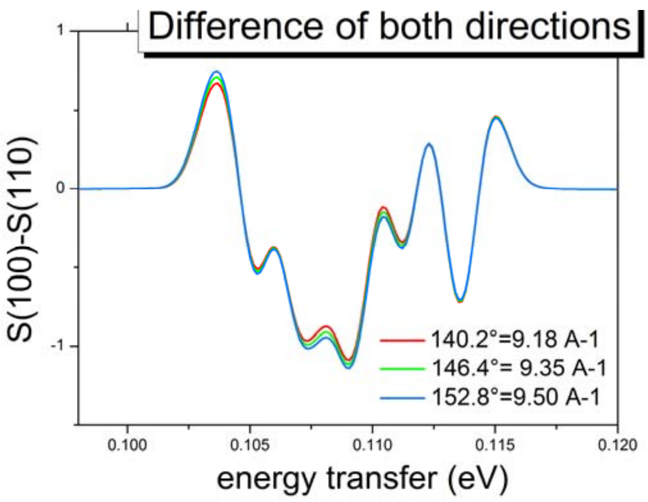

Fig. 4: Difference plot of S(**q**,ω) for **q**||<100> and q||<110> calculated for the three highest angle analyzer columns showing the trend of the vector **q** effect with the size of |**q**|..

<u>References:</u> [1] see e.g. J.L. Sarrao and J.D. Thompson, J. Phys. Soc. Japan **76** (2007) 051013, H. Hegger *et al.* PRL **84** (2000) 4986, C. Petrovic *et al.* Europhys. Lett. **53** (2001) 354, C. Petrovic *et al.* J. Phys. Cond. Matter **13** (2001) L337T, [2] A. Christiansen, PRB **70**, 134505 (2004). [3] T. Willers *et al.* , PRB **81**, 195114 (2010), [4] T.Willers *et al*., PRL. **109**, 046401 (2012), [5] Code by M.W. Haverkort