Orbital orientation of the *4f* ground state in $CeCu_2Si_2$

Heavy fermion compounds are rare earth or actinide materials where the *f* electrons hybridize with the conductions electrons. Due to the hybridization the charge carriers have enhanced effective masses which can be up to a thousand times larger than the free electron mass, giving the name to this class of materials. The hybridization of *f* and conduction electrons competes with the RKKY interaction so that quantum critical transitions from local magnetism to itinerant *f* electron behavior take place when the exchange interaction is increased with external parameters such as pressure, field or doping. Often unconventional superconductivity occurs in the vicinity of the quantum critical point. $CeCu_2Si_2$ was the first heavy fermion compound where this proximity of magnetism and superconductivity was observed [1].

The key to understanding heavy fermion compounds is linked to the ground state wave function of the *f* electrons. In a crystalline environment an ion experiences a so-called crystalline electric field (CEF) caused by the charges of the surrounding ions and the degeneracy of the Hund's rule ground state is lifted. The resulting CEF 4*f* states are highly anisotropic (see Fig. 1). However, as the wave functions cannot be calculated *ab initio*, they have to be determined experimentally. Single crystal magnetization measurements, inelastic neutron scattering and more recently also soft X-ray absorption spectroscopy at the $M_{4,5}$ edges ($3d \rightarrow 4f$) have been very successful in determining the 4*f* wave function. However, none of the established methods is able to determine the *orientation* of the 4*f* orbital in the tetragonal (a=b≠c) structure of $CeCu_2Si_2$. Figure 1 shows the ground state orbital of $CeCu_2Si_2$ for two possible orientations with loops in [100] direction (left) or [110] direction (right). The established techniques rely on dipole transitions, thus cannot distinguish these two fourfold orbitals. Hence an experiment which goes beyond the dipole limit is required.

Simulations of the scattering function $S(q,\omega)$ have shown that Inelastic X-ray Scattering (IXS) with hard X-rays has the potential to reach non-dipole allowed transitions when working at large momentum transfers. In order to mark the difference to resonant techniques the method is called non-resonant IXS, in brief NIXS. The left panel of Fig. 2 shows at the example of the cerium $N_{4,5}$ edge ($4d \rightarrow 4f$) the different multipole contributions to the radial parts of $S(q,\omega)$ as function of the momentum transfer $|\vec{q}|$. Already at 10 $Å^{-1}$ the scattering due to quadrupolar transitions (k=3) is quite substantial with respect to the dipole transitions (k=1). The simulation of the angular parts of $S(q,\omega)$ shows further that extra transitions appear, which are not allowed in the dipole approximation (see Fig. 2(b)).

We adapted these simulations of $S(q,\omega)$ at the cerium $N_{4,5}$ edge to our crystal-field problem by implementing the vector $\vec{q}$ dependence in addition to the absolute size of the momentum transfer $|\vec{q}|$. The direction of $\vec{q}$ gives the sensitivity to the shape of the orbital. Our simulation shows that NIXS is first of all, like inelastic neutron scattering and other methods, sensitive to the orbital shape out-of-plane. On top of this, and this is the intriguing novelty, the same simulation for two different directions within the tetragonal *ab*-plane ($\vec{q} \parallel [100]$ and $\vec{q} \parallel [110]$) shows that the NIXS intensity also depends on the $\vec{q}$ directions within the plane with fourfold rotational symmetry (Fig. 3, top graph). Hence NIXS should be able to tell us how the orbital is oriented.

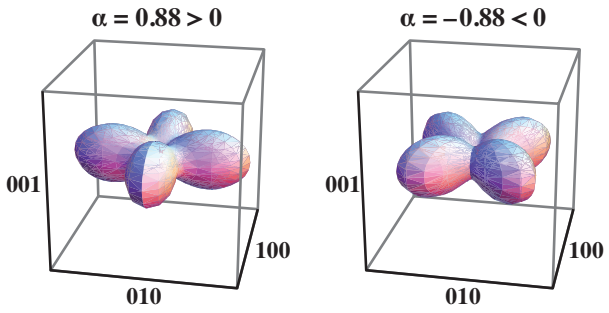

Fig. 1. Angular distribution of the *4f* electron in $CeCu_2Si_2$ corresponding to the wave function $\alpha\,|\pm 5/2> + \sqrt{(1-\alpha^2)}\,|\pm 3/2>$ with $\alpha = |0.88|$; on the left for $\alpha > 0$, on the right for $\alpha < 0$. Both orbitals are identical but rotated by 45° around the *c*-axis.

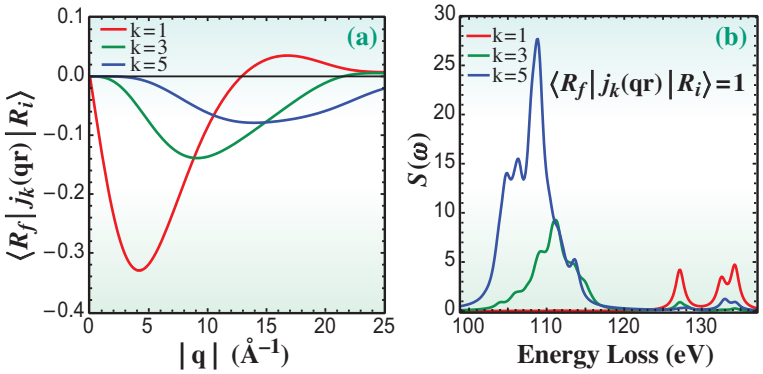

Fig. 2. **(a)** k^{th} order term of the radial part of the scattering function versus momentum transfer. **(b)** k^{th} order contribution of angular part of the scattering function as function of energy transfer.

Such a NIXS experiment was realized in a collaboration of the University of Cologne, Germany, the Max-Planck Institute for Chemical Physics of Solids in Dresden, Germany, and the National Synchrotron Radiation Research Center in Hsinchu, Taiwan. The experiment was carried out at the Taiwan beamline **BL12XU** at SPring-8 with 9.8 keV incident photons so that at a scattering angle of about 135° momentum transfers of 9.3 Å^{-1} could be achieved. This is well beyond the dipole limit. The bottom graphs of Fig. 3 show the NIXS data for momentum transfers in the two different in-plane directions, $\bar{q} \| [100]$ (blue dots) and $\bar{q} \| [110]$ (green dots). The error bars reflect the statistical error. Only a linear background has been subtracted. The comparison of simulation (top) and data (bottom) shows that the simulation for an orbital orientation with the loops along the [110] direction (see inset) reproduces the experimental data very well. We therefore conclude that we solved the long standing problem of the orbital orientation in the tetragonal heavy fermion system $CeCu_2Si_2$ and also opened up a new direction for studying the 4*f* orbitals of other heavy fermion systems. This work has been published in reference [2].

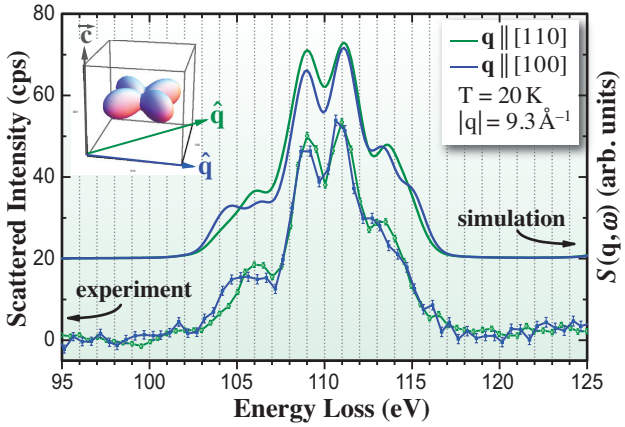

Fig. 3. Top: Simulation of $S(q,\omega)$ for the two in-plane directions assuming the orbital orientation as in the inset. The calculations are convoluted with a Lorentzian of FWHM = 0.3 eV and a Gaussian of FWHM=1.32 eV to account for lifetime broadening and instrumental resolution. Bottom: NIXS data of $CeCu_2Si_2$ single crystals for the two in-plane directions $\bar{q} \| [100]$ and $\bar{q} \| [110]$.

Andrea Severing[a,*], Liu Hao Tjeng[b] and Nozomu Hiraoka[c]

[a] Institute of Physics II, University of Cologne, Germany
[b] Max-Planck Institute for Chemical Physics of Solids, Germany
[c] National Synchrotron Radiation Research Center, Taiwan

*E-mail: severing@ph2.uni-koeln.de

References

[1] O. Stockert *et al.*: Nat. Physics **7** (2011) 119; and references therein.
[2] *T. Willers, F. Strigari, N. Hiraoka, Y.Q. Cai, M.W. Haverkort, K.-D. Tsuei, Y.F. Liao, S. Seiro, C. Geibel, F. Steglich, L.H. Tjeng and A. Severing: Phys. Rev. Lett.* ***109*** *(2012) 046401.*