A New Way to Explore 4f-Orbital Orientation in a Heavy Fermion System

This report features the work of Thomas Willers and his co-workers published in Phys. Rev. Lett. ***109****, 046401 (2012).*

A determination of the orientation of the quantum state of 4f electrons in cubic heavy fermion systems is of the utmost importance to understand the crystal-field ground state. Because the 4f orbital is highly symmetric, the crystal-field ground state has been incompletely determined. A research team from Germany, USA and NSRRC in Taiwan has revealed the orientation of the ground-state orbital of a cubic 4f system using non-resonant inelastic X-ray scattering.[1] Heavy fermion systems exhibit intriguing phenomena such as unconventional superconductivity and non-Fermi liquid behavior emerging near quantum critical points. $CeCu_2Si_2$, the first superconducting heavy fermion compound, was discovered in 1979.[2] The peculiar phenomena that occur in these rare-earth (4f) or actinide (5f) intermetallic compounds reflect the strong interactions of the f electrons with the conduction electrons. The charge carriers are heavy because the strong interaction impedes their motion, as if they acquired a large mass: which can be up to a thousand times that of a free electron.

When an ion is introduced into a crystalline environment, it experiences a crystalline electric field (CEF) caused by the charges of the surrounding ions. The CEF lifts the degeneracy of the ground state according to Hund's rule; the resulting CEF 4f states of a rare-earth ion are highly anisotropic, as Fig. 1 [adopted from Fig. 1 of Ref. 1] shows. To improve an understanding of the hybridization between these anisotropic 4f-orbitals and the conduction electrons, the 4f ground-state orbital must be determined.[3] Measurement of magnetization

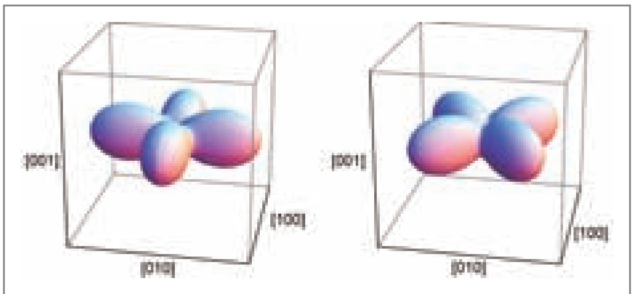

Fig. 1: Angular distribution of a 4f orbital in $CeCu_2Si_2$. The orbitals are identical but their orientations differ within the tetragonal ($a = b \neq c$) ab-plane. They are rotated by 45° about axis c. [reproduced from Ref. 1]

and inelastic neutron scattering in combination has been applied for this purpose, and recently also soft X-ray absorption spectra at the cerium $M_{4,5}$-edges (3d → 4f), but no established method has determined the orientation of the 4f orbital of $CeCu_2Si_2$ in its tetragonal structure ($a = b \neq c$). Willers *et al.* presented the ground-state orbital of $CeCu_2Si_2$, as Fig. 1 clearly shows. The 4f orbital is oriented with lobes in either direction [100] (left) or direction [110] (right). As the established techniques that rely on dipole transitions cannot distinguish these two four-fold orbitals, an experiment that transcends the dipole limit is required.

Interesting results were obtained on the simulated scattering function. Willers *et al.* presented simulations and indicated 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. To distinguish from resonant techniques, the method is called non-resonant IXS, in brief NIXS. The left side of Fig. 2 shows an example of the cerium $N_{4,5}$-edge (4d → 4f) with the contributions of various multipoles to the radial parts of the scattering function as a function of momentum transfer $|q|$. Already at 10 Å^{-1}, the scattering due to quadrupolar transitions (k = 3) is substantial relative to dipole transitions (k = 1). The simulation of the angular parts of the scattering cross section shows further that, in the spectra as intensity versus energy transfer, extra transitions, which are not allowed in the dipole approximation, become visible (see the right side of Fig. 2).

The research team adapted these simulations of the scattering function at the cerium $N_{4,5}$- edge to the crystal-field problem by implementing the dependence on vector q in addition to the magnitude $|q|$ of the momentum transfer.[3] The direction of q gives the sensitivity to the shape of the orbital. The simulation shows that NIXS is primarily, like inelastic neutron scattering and other methods, sensitive to the orbital shape out of the plane. Furthermore -- and this is the intriguing novelty, the same simulation for two directions ($q||$<100> and $q||$<110>) within the tetragonal *ab*-plane shows that the NIXS intensity depends also on the q directions within the plane with four-fold rotational symmetry (top graphs in Fig. 3). The research team hence proposed that NIXS might determine how the orbital is oriented.

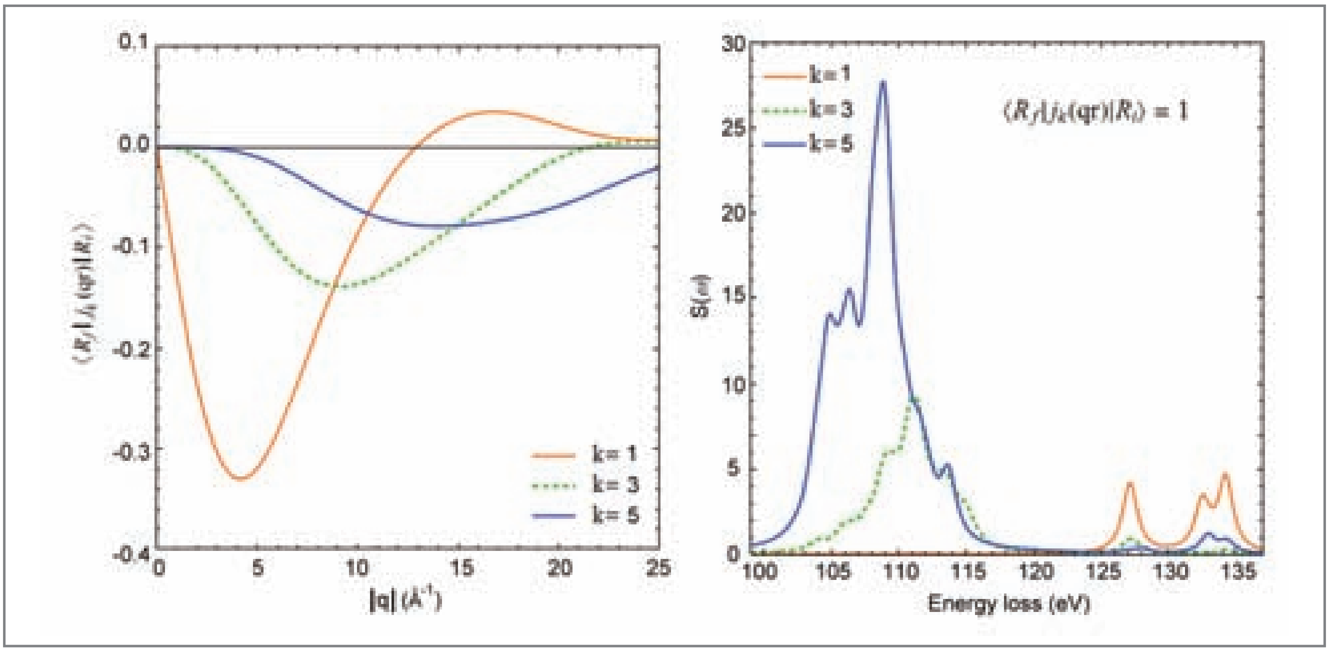

Fig. 2: Left: k^{th}-order term of the radial part of the scattering function versus momentum transfer. Right: k^{th} order contribution of the angular part of the scattering function as a function of energy transfer. [reproduced from Ref. 1]

Such a NIXS experiment has been realized in an international collaboration of the University of Cologne, the Max-Planck Institute for Chemical Physics of Solids in Germany and NSRRC in Taiwan. The research team performed an experiment at Taiwan beamline **BL12XU** at SPring-8 with incident photons of energy 10 keV so that, at a scattering angle about 135°, momentum transfer of 9.3 Å^{-1} was achieved -- well beyond the dipole limit. The bottom graphs of Fig. 3 show the NIXS data recorded for momentum transfers in two in-plane directions q|| [100] (blue dots) and q|| [110] (yellow dots). The error bars reflect the statistical error. Only a linear background has been subtracted. A comparison of the simulation (top) and the data (bottom) shows that the simulation of the orbital orientation as shown in the inset reproduces the experiment satisfactorily. The research team hence concluded that they both solved an enduring problem of the orbital orientation in the tetragonal heavy fermion system $CeCu_2Si_2$ and opened a new direction to study the 4f orbital orientation of other heavy fermion systems.

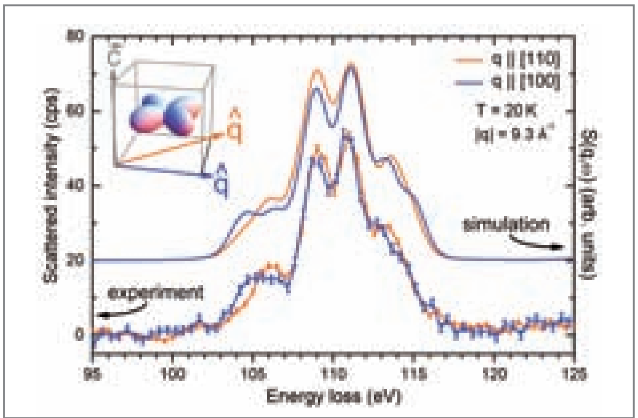

Fig. 3: Top: Simulation of the scattering function for two in-plane directions, q||[100] and q|| [110], assuming an orbital orientation as shown in the inset. The calculations are convoluted with a Lorenzian component of FWHM = 0.3 eV to account for life-time broadening and a Gaussian component of FWHM = 1.32 eV to account for the instrumental resolution. Bottom: NIXS data of $CeCu_2Si_2$ single crystals. The blue dots correspond to the setup for q|| [100] and the green ones to q|| [110]. [reproduced from Ref. 1]

References

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