Experimental report beamtime HC 4911

UPt$_2$Si$_2$, UOs$_2$Si$_2$, and of UAu$_2$Si$_2$ were studied with non-resonant inelastic x-ray scattering (NIXS), also called x-ray Raman. This is part of a bigger project to elucidate the symmetry of the ground states within the UM$_2$Si$_2$ family. The dependence of the cross-section from the direction of the momentum transfer $\vec{q}$ gives this insight to the ground state symmetry in analogy to the linear dichroism in XAS. Here, we focus on large momentum transfers and multipole scattering (in contrast to dipole as in XAS).

The mixture of multipoles that contribute to the scattering cross-section depends on the length of $|\vec{q}|$, and the modelling of the data with full multiplet calculations is based on atomic values for the radial wave function. This, however, must not be correct. Actually, we noticed that we have to correct the radial extension of the wave function by artificially adapting the length of $|\vec{q}|$ in the simulation. This is a *loose* way of doing it, so that we set out to determine the multipole contributions by measuring the isotropic scattering cross-section of the above compounds for several $|\vec{q}|$.

The UM_2Si$_2$ crystallize in the I4/mmm space group with tetragonal point group symmetry for the U atoms so that two crystal orientations had to be measured to construct the isotropic spectrum. In particular, two crystals each (one with the 001, the other with the 100 direction perpendicular to sample holders) were measured. The measurement was conducted so that the momentum transfer $\vec{q}$ was parallel to the relevant crystallographic directions mentioned above. The incident energy was fixed at about 10 keV, with a resolution of around 0.6 eV and 10K so that only the ground state was probed. Different values of the momentum transfer were achieved my utilizing both the horizontal and vertical scattering planes. Grouping only six analyzers of a given box, values of 9.52 $Å^{-1}$ and 9.17 $Å^{-1}$ momentum transfer were achieved with backscattering geometry in the horizontal scattering plane. Values of 8.85 $Å^{-1}$, 8.3 $Å^{-1}$, 7.64 $Å^{-1}$, 6.88 $Å^{-1}$ and 3.48 $Å^{-1}$ momentum transfer were achieved in the vertical scattering plane.

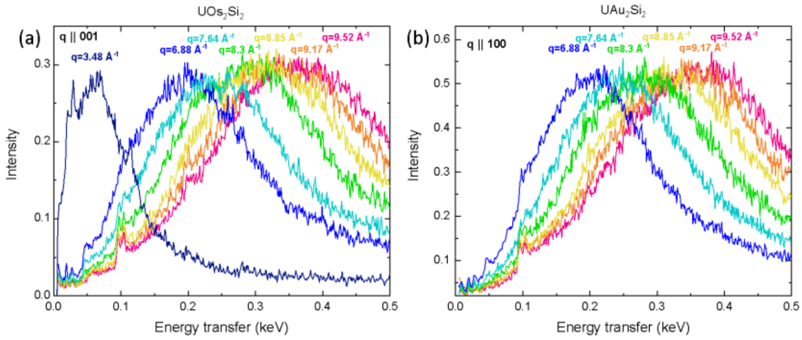

Fig. 1:
$\overrightarrow{|q|}$-dependence of the Compton scattering, normalized to the peak height.

Fig. 1 shows the $|\vec{q}|$-dependence of the Compton scattering of UOs$_2$Si$_2$ (a) and UAu$_2$Si$_2$(b). The long scans are used to normalize the spectra to the height of the Compton profile. We focus of the feature appearing at 85-125 eV energy transfer, namely the U O$_{4,5}$ edge.

Fig. 2 shows the $|\vec{q}|$-dependence of the U O$_{4,5}$ edge of UOs$_2$Si$_2$ (a) and UAu$_2$Si$_2$(b). The measurement was performed with $\vec{q}$ parallel to the 100 direction for UAu$_2$Si$_2$, since we know from previous NIXS experiment performed at the P01 beamline (DESY, Hamburg) that there is no $\vec{q}||001 - \vec{q}||100$ directional dependence. For UOs$_2$Si$_2$ two samples were measured, with the 100 and 001 directions parallel to $\vec{q}$, to probe a possible directional dependence of the spectra due to crystal-field anisotropy. It turns out the directional dependence is, if anything, very subtle. We then concentrate on the trend seen in the spectra when the size of $|\vec{q}|$ varies.

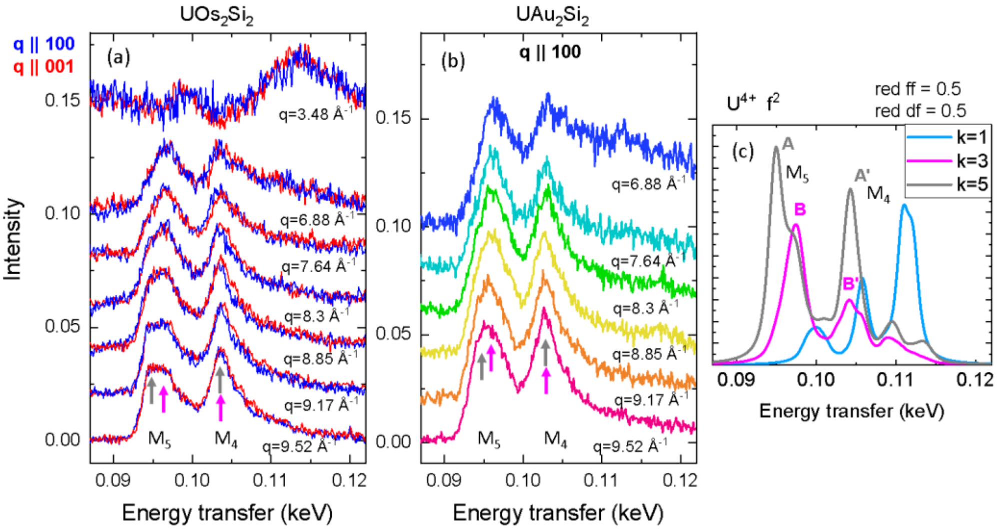

Fig. 2: $|\vec{q}|$-dependence of the U $O_{4,5}$ edge of (a) UOs_2Si_2, with $\vec{q}$ ||100 and 001 and (b) of UAu_2Si_2, with $\vec{q}$ ||100. (c) Angular part of the scattering function as resolved into multipole contributions.

In order to analyze the $|\vec{q}|$-dependence we plot the angular part of the NIXS scattering function for a Uf^2 configuration in Fig. 2(c), as resolved into multipoles contributions (k=1 dipole, k=3 octupole, k=5 triacontadipole). It can be seen that the M_5 intensity is mainly given by a k=5 feature (A) and a k=3 feature (B), inducing the formation to a double peak structure, while the corresponding A’ and B’ features of the M_4 edge are rather aligned in energy. The radial part of the scattering function consists of an integral over the radial wave function and is dependent on $|\vec{q}|$. As $|\vec{q}|$ varies, the multipole contributions of Fig. 2 (c) acquire different weights in the total spectrum. Looking at the experimental data of Fig. 2 (a) and (b), we first of all notice the double peak structure of the M_5 edge and the single peak structure of the M_4 edge (gray and magenta arrows). Furthermore, it is evident how at high $|\vec{q}|$, 9.52 $Å^{-1}$ and 9.17 $Å^{-1}$, the k=5 contribution is emphasized, as compared to the k=3 one. As $|\vec{q}|$ decreases, the k=5 contribution decreases and the k=3 becomes dominant ($|\vec{q}|$=6.88 $Å^{-1}$). The k =1 contribution is appreciably visible only in the $|\vec{q}|$ = 3.48 $Å^{-1}$ data of Fig 2(a) acquired for the UOs_2Si_2 sample.

As discussed above, the shape of the spectra is given by the multipole admixture. The resulting $|\vec{q}|$-dependent trend of the multipolar contributions is shared among UOs_2Si_2 and UAu_2Si_2. On the other hand, the shape itself of the UOs_2Si_2 and UAu_2Si_2 spectra is different, with the M_5 edge of the UAu_2Si_2 sample being narrower and with the A and B features closer to each other. It should be noted that the calculations of Fig. 2(c) are based on atomic values. We can capture the common $|\vec{q}|$-dependence and the differences in shape by extending the radial wave function in the calculations of about 10 to 20%. This may not be completely physical, since it is not needed in non-metallic and more localized samples, such as UO_2. The aim of the analysis of the present data is then to develop an intuition on the behavior of this parameter. We intend to complement the acquired data with measurements of other members of the UM_2Si_2 family (with M being now a 3d and 4d transition metal) in order to attempt a systematic modelling of the NIXS data of this family of compounds.