ESRF	**Experiment title:** UPd$_3$: temperature dependence of the quadrupolar order parameters	**Experiment number**: HE2417
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Report:

Numerous studies of UPd$_3$, both macroscopic and microscopic, have indicated 4 phase transitions below T = 8 K, see [1] for a summary and full references. These have been attributed to a series of antiferromagnetic quadrupolar orderings of the localised U $5f^2$ electrons. A detailed understanding of the origin of these transitions has proved a challenge for many years, but a new proposed crystal field scheme [1] has been able to explain the existence of the four transitions and our recent X-ray resonant scattering experiments (e.g. HE1936, HE2051) have revealed the nature of the first quadrupolar phase as being described by the $\boldsymbol{Q_{zx}}$ order parameter, and suggested a more complex combination of order parameters in the third phase [2,3]. The aim of this experiment was to further investigate the nature of the quadrupolar order in the lower temperature phases of UPd$_3$ by investigating the azimuthal dependence of the scattered intensity.

In our experiment we used the low temperature displex cryostat and the standard gold analyser for polarisation analysis. Initially we used a single crystal sample cut and polished with a face normal to [207], since this would allow us to measure the whole 360^o azimuthal dependence for the two quadrupolar superlattice reflections at (103) and (104). However, the surface quality of the sample had degraded severely since the previous experiment and the mosaicity was large, such that the observation of low intensity peaks, such as quadrupolar reflections, was made very difficult. We therefore changed to a [001] face crystal, which gave much sharper peak profiles. Using this sample, after orientation at 9 keV, we were able to measure the azimuthal dependence of the (104) reflection over 360^o for E = 3.72 keV at 2.1 K, i.e. in the lowest temperature phase. By fitting lorentzian peak shapes to theta rocking curves, the integrated intensity was obtained in both σσ and σπ channels as a function of the azimuth angle Ψ, where the zero is defined by the azimuthal reference vector [010]. A fit to the two data sets was then made simultaneously using different models for the quadrupolar order in this phase. As shown in figure 1, both data sets are in excellent agreement with the calculation for the $\boldsymbol{Q_{xy}}$ order parameter.

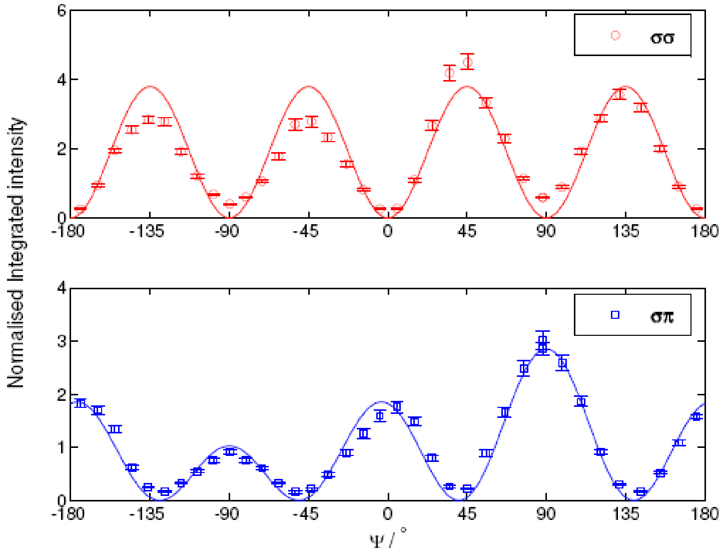

Figure 1: Azimuthal dependence of the (104) σσ and σπ scattered intensity in UPd_3 at $T = 2.1$ K. The solid lines are fits based on a model for the $\boldsymbol{Q}_{xy}$ order parameter.

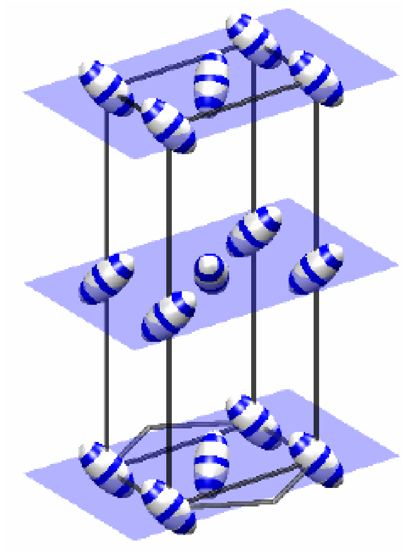

Figure 2: Unit cell showing the quadrupolar ordering of the $5f^2$ electrons on the quasi-cubic sites for $\boldsymbol{Q}_{zx}$ antiparallel and $\boldsymbol{Q}_{xy}$ parallel stacking along the c-axis

The same measurements were repeated at $T = 5.2$ K, and an identical agreement with the $\boldsymbol{Q}_{xy}$ order parameter was observed, with the only difference being the reduction in the magnitude of the integrated intensities, which was by a factor of ~2.5 in both scattering channels. It is interesting to compare the σπ data with that measured in HE2051, which showed an azimuthal variation which could not be simply fitted using a combination of the symmetry allowed quadrupolar order parameters for the quasi-cubic uranium sites. The excellent quality of the data arising from this latest experiment, the reduced mosaicity and increased count rate leads us to believe that this is an improvement on the previous measurements, and as such should be seen as the definitive data for the (104) reflection.

Unfortunately the restrictions in chi imposed by the diffractometer meant that it was impossible to repeat the measurements over the full 360° for the (103) reflection, however data recorded at accessible azimuth angles showed a sinusoidal variation in the unrotated σσ channel with a much more complicated variation in σπ. Time limitations meant we were unable to expand this data set.

The (103) and (104) reflections reveal different information about the stacking of the quadrupolar moments along the c-axis: an antiferro-type and a ferro-type stacking respectively. The observation of the $\boldsymbol{Q}_{zx}$ order parameter for (103) corresponds to a rotation of the quadrupole moments in the zx plane, which alternates moving up the c-axis, while the observation of the $\boldsymbol{Q}_{xy}$ order parameter for (104) corresponds to the same rotation of the quadrupole moments in the xy plane for each layer, see Figure 2 for a schematic representation of the quadrupoles.

References

[1] K A McEwen, J-G Park, A J Gipson and G A Gehring, *J. Phys.: Condens. Matter* **15** (2003) S1923
[2] H C Walker, K A McEwen, D F McMorrow et al., *Phys. Rev. Lett.* **97** (2006) 137203
[3] K A McEwen, H C Walker, M D Le et al., *J. Magn. Magn. Mater.* **310** (2007) 718