ESRF	**Experiment title:** Quadrupolar order parameters in UPd_3	**Experiment number**: HE-1696
Beamline: ID20	**Date of experiment**: from: 16 June 2004 to: 22 June 2004	**Date of report**: August 2004
Shifts: 18	**Local contact(s)**: Pascale Deen	*Received at ESRF:*

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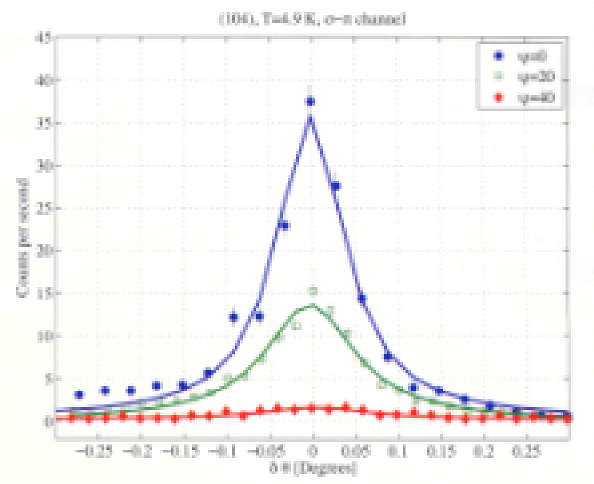
Report:

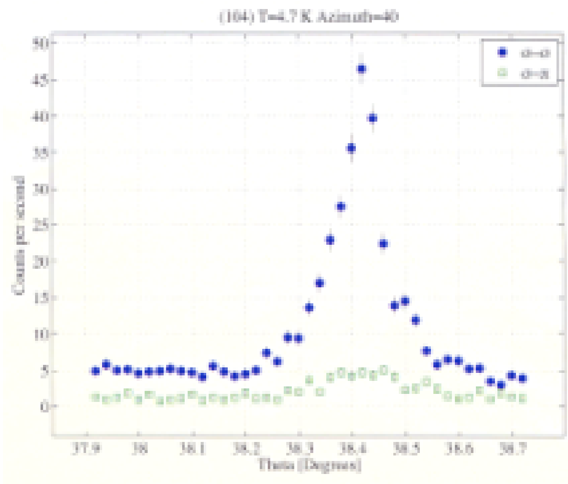
UPd_3 is a particularly interesting system because it is one of the small number of metallic materials that exhibit long range quadrupolar ordering. Moreover, it has no less than *four phase transitions* associated with *different quadrupolar order parameters*, which have been seen by both macroscopic and microscopic measurement techniques (see [1] for full references). The uranium 5f electrons in UPd_3 are well localized, with the $5f^2$ configuration. The large orbital moment (L=5) gives rise to a strong coupling to the lattice, and hence it is not so surprising that quadrupolar effects are dominant in this system.

UPd_3 exhibits the double-hexagonal close-packed crystal structure, with uranium ions at sites of local hexagonal and local quasi-cubic symmetry. The transitions occur at T_0 = 7.8 K, T_{+1} = 6.9 K, T_{-1} = 6.7 K, and T_2 = 4.4 K. Polarised neutron diffraction measurements [2] indicated that the phase transition at T_0 is to an antiferroquadrupolar (AFQ) structure of the U 5f electrons that is accompanied by periodic lattice distortions and a doubling of the crystallographic unit cell. The unit cell in the phase below T_0 is orthorhombic, with the ordered quadrupole moments predominantly on the quasi-cubic sites, and stacked in anti-phase along the c-axis. *Definitive proof of this was provided in our earlier experiment on ID20, reported in Phys. Rev. Letters [3].* Using the Orange cryostat, with horizontal scattering geometry, we observed resonant scattering of the (103) reflection [we index the reflections using the orthorhombic unit cell] at the uranium M_{IV} edge. Polarisation analysis showed that the scattering was predominantly □-□ between T_0 and T_1. We concluded that the transition at T_0 is second order with order parameter Q_{x2-y2}. Below T_1, a new reflection at (104) was observed, which was predominantly □□□. At the same time, the (103) reflection developed a □□□ component. Because of time limitations and the weak intensities of the peaks, we were not able to distinguish whether these developments took place below T_{+1} or T_{-1}. Below T_2, the (103) peak displayed a sharp reduction in intensity, whilst the (104) intensity increased abruptly. We interpret these results in terms of a rotation of the quadrupole moments around the x and y axes below T_1, and a component with uniform stacking along the c-axis.

In [1] we developed a new model for the crystal field states and quadrupolar transitions in UPd_3. It explains why there are four phase transitions, and also our earlier inelastic neutron scattering studies of the excitations in UPd_3 at 2 K. In our model, the order parameter below T_0 is Q_{x2-y2}, as observed. We believe that the order parameter below T_{+1} has an additional Q_{yz} component with uniform stacking along the c-axis. Below T_{-1}, we propose a composite phase where Q_{yz} and Q_{xz} are both present with uniform stacking, but on *different sites* in the unit cell. There may also be a Q_{xy} component with antiphase stacking. Below T_2, we think that Q_{x2-y2}, Q_{yz}, Q_{xz} and Q_{xy} are all present but the Q_{xz} ordering wave vector has moved to q=0.

The aim of this experiment was to test these detailed predictions for the order parameters of the AFQ structures in UPd_3 below T_{+1}, T_{-1} and T_2. The constraints of working at the uranium M_{IV} resonance energy (3.728 keV) severely limit the volume of reciprocal space that can be studied. However, the two key reflections characterizing the AFQ ordering in UPd_3 are (103) and (104). We were allocated time in June 2004, when the new ID20 cryostat became operational. However, whilst we were able to cool the sample to base temperature, control at temperatures above 5K was unstable, and we were unable to study the temperature dependence of the intensities in the crucial range between 6 and 8K. Moreover, alignment and other instrumental problems meant that we had only a fragmented total of 2 days useful beam time. Nevertheless, we were able to demonstrate a dramatic dependence of the (104) intensities, at the uranium resonance energy of 3.724 keV, as a function of azimuthal angle □. The figures below show that the (104) □□□ channel intensity, in the phase between T_{-1} and T_2, decreases rapidly as □ increases from 0^o to 40^o. In contrast, the □□□ channel intensity, which has no resonant intensity at □ $=0^o$, becomes the dominant channel at □ $=40^o$.





Our recent development of a new microscopic model for UPd_3 has provided a major breakthrough in understanding the four quadrupolar phases in this canonical, localized-moment, uranium system. Further time is needed to complete this experiment, but the results will provide a vital test of our predictions for the order parameters in this fascinating compound.

References

[1] K A McEwen, J-G Park, A J Gipson and G A Gehring, *J.Phys.: Condens. Matter* **15** (2003) S1923

[2] K A McEwen, U Steigenberger, K N Clausen, J Kulda, J-G Park and M B Walker, *J.Magn. Magn. Mater.* **177-181** (1988) 37 and refs therein

[3] D F McMorrow, K A McEwen, U Steigenberger, H M Rønnow and F Yakhou, *Phys. Rev. Lett*. **87** (2001) 057201