

Quadrupolar interactions in UPd₃ observed by inelastic neutron scattering

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Low temperature phase transitions in condensed matter systems are usually driven by the exchange interactions between the spins of electrons around neighbouring ions. However, in systems where the orbital moment is not quenched, such as in the rare earths or actinides, the exchange interaction between the orbital angular momentum of nearby electrons may be stronger than the spin dipolar interaction. This can lead to the ordering of higher order multipoles, of which the electric quadrupole and octupole order is most commonly observed, for example in NpO₂[1].

UPd₃ was one of the first compounds to be shown to exhibit quadrupolar ordering and has been extensively studied since. It has a double-hexagonal close-packed crystal structure, shown in fig. 1, with the U⁴⁺ ion on sites with two different point symmetry, hexagonal and quasi-cubic (trigonal). The ground spin orbit multiplet with $J=4$ is split by the crystal field giving the hexagonal sites a singlet ground state and the quasi-cubic sites a doublet, as shown in fig. 2. In the absence of exchange interactions, excitations between these crystal field states are dispersionless, however recent inelastic neutron scattering measurements show that this is not the case, and that the dispersion is best explained by quadrupolar exchange interactions, rather than the usual dipolar.

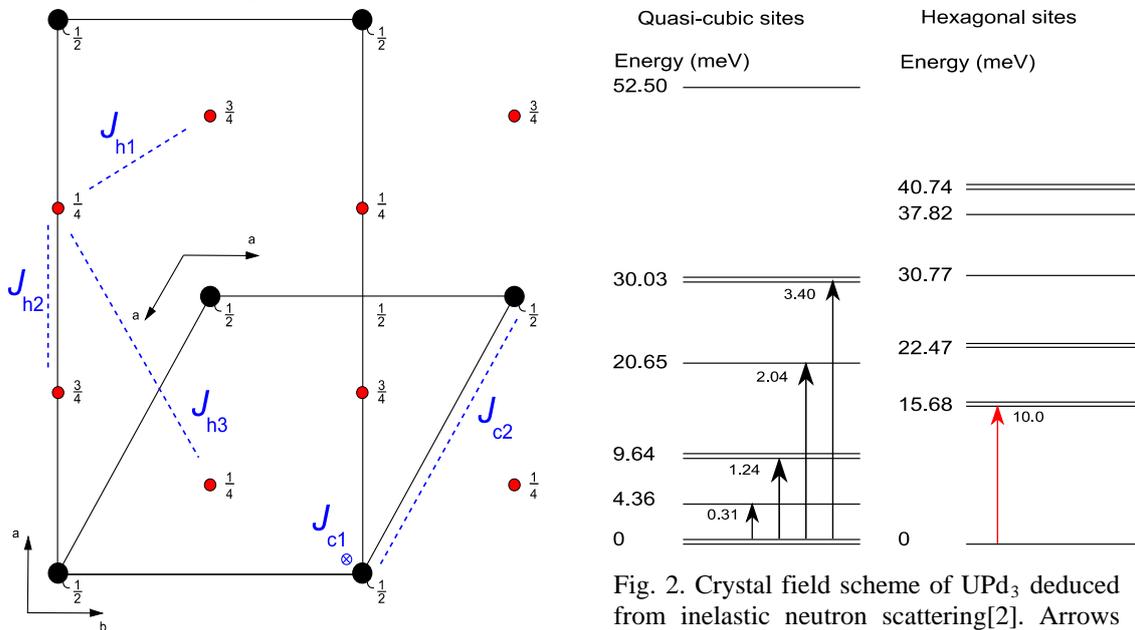


Fig. 1. Crystal structure of UPd₃ showing the hexagonal and (ordered phase) orthorhombic cells. The quasicubic site U⁴⁺ ions are shown as large black circles, whilst the hexagonal site ions are small red circles. Fractions next to the ions show height above the $z=0$ plane. Blue dashed lines show exchange interactions, as

Fig. 2. Crystal field scheme of UPd₃ deduced from inelastic neutron scattering[2]. Arrows indicate dipole allowed transitions, with the square of the transition matrix element, which is proportional to the intensity of the excitation, shown next to the arrowhead. The main excitation observed, shown in red, is the only allowed transition on the hexagonal site.

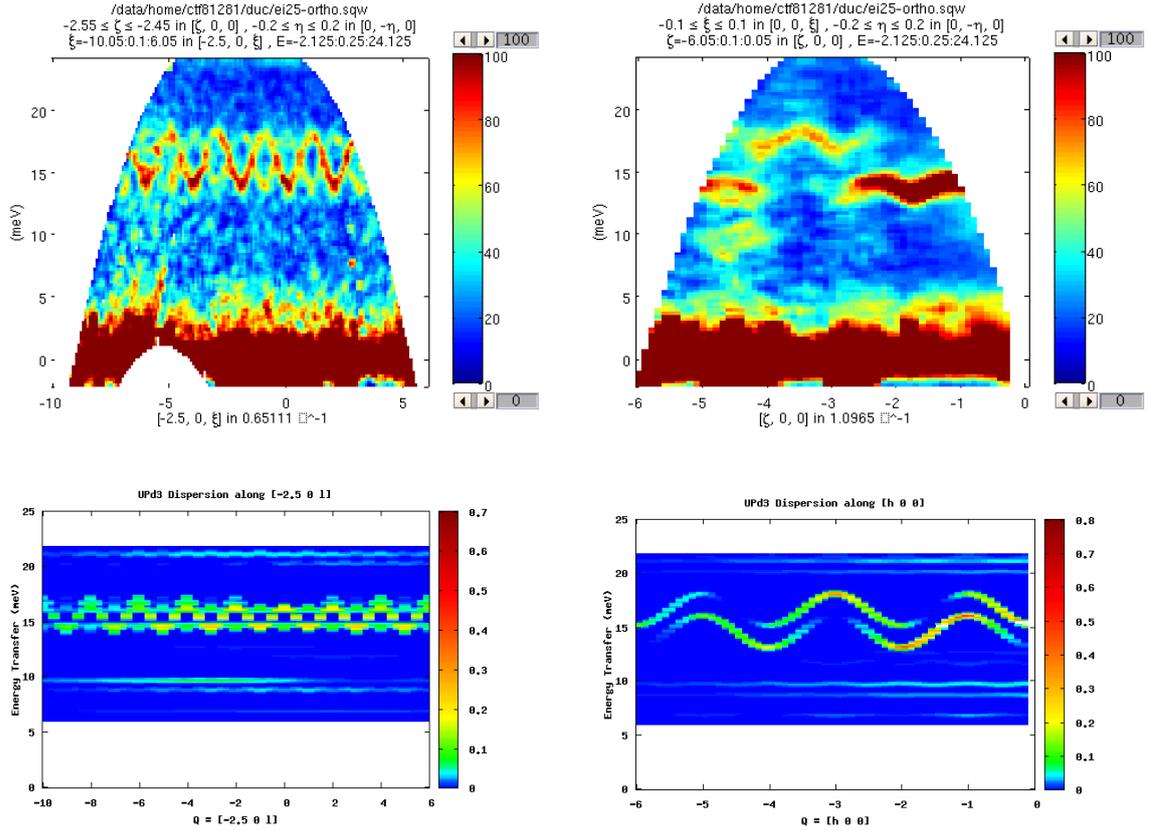


Fig. 3. Inelastic neutron scattering spectra measured on Merlin (top) and calculated in McPhase (bottom), with incident energy of 25meV, and showing the dispersion along the $[-2.5\ 0\ L]$ (left) and $[h\ 0\ 0]$ (right) directions. The exchange parameters for the hexagonal sites used to calculate the dispersion are, in the notation of fig 1. in meV, $J_{h1}^{zx} = -0.2$, $J_{h3}^{zx} = 0.15$, $J_{h1}^{yz} = 0.05$, $J_{h2}^{yz} = 0.15$.

The measurements were performed on a 25g single crystal sample grown by the Czochralski method using the Merlin time-of-flight spectrometer at the ISIS facility. Cuts through the dataset for incident neutron energy of 25 meV, showing dispersion along the h and l directions are in fig. 3. These show clearly the dispersion of the 15 meV levels on the hexagonal sites.

The ordered quadrupole moments reside entirely on the quasi-cubic sites, and using previous resonant-xray scattering and magnetostriction data a mean-field model involving two exchange parameters (an out-of-plane one, labeled J_{c1} and an in-plane one, J_{c2}) for each of the five quadrupolar moment components was deduced[2].

The new data, however show the dispersion of the hexagonal sites very well, and allows us to deduce the exchange parameters on the hexagonal sites. As there are four sites in the ordered phase (orthorhombic) unit cell, we should expect four modes. However, due to the magnetic structure factor the intensity of two of these modes is near zero. Also the anti-phase modulation at the same energy of the modes along $[-2.5\ 0\ l]$ show that the hexagonal sites are not ordered, for otherwise the crystal field doublet at 15 meV would be split, and the two excitations would be offset in energy. Meanwhile the simplest set of exchange parameters which yields a calculated dispersion in the *random phase approximation* close to that observed along $[h\ 0\ 0]$ involves ferro-quadrupolar coupling of the Q_{zx} and Q_{yz} quadrupoles.

References

- [1] P. Santini et al., *Rev. Mod. Phys.* **81**, 807–863 (2009)
- [2] M.D. Le., *Magnetism and quadrupolar order in f-electron systems*, PhD Thesis, University of London

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Short Abstract

UPd₃ is a rare example of a compound where the low temperature ordered phase involves the alignment of electric quadrupoles of the f-electrons on neighbouring ionic sites, rather than their magnetic dipole moments. We show inelastic neutron spectra of the dispersion of crystal field excitations in UPd₃, which are consistent with RPA calculations assuming quadrupolar exchange interactions between the f-electrons on the hexagonal U⁴⁺ sites.