Anisotropic phonon line-broadening in UO_2 below room temperature

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The interaction between the lattice and magnetic excitations in UO_2 was first observed in 1966 [1]. Many neutron studies have been reported over the years, with the latest in 2011 [2]. In 2014 [3] there was a report from bulk property measurements showing that the thermal conductivity of UO_2 below 300 K is anisotropic, which is formally forbidden in a cubic system. The interactions in UO_2 are extremely complex [4], but to understand the thermal conductivity we need a close examination of the phonon linewidths at low temperature.

We have undertaken X-ray inelastic scattering using ID28 to measure the phonons in UO_2 from a single crystal. X-rays have the advantage that they are insensitive to magnetic and quadrupole effects (providing the energy is far removed from resonances) and are sensitive only to the vibrational spectra, and any broadening of them caused by their interactions with the electronic system.

The experiments show large line-broadening effects in the transverse acoustic phonons in the [100] direction due to the electronic-lattice interactions, and no measurable effects in the [110] direction. This anisotropy is consistent with the lower thermal conductivity measured in the [100] direction [3]. The effects extend to at least 200 K, consistent with short-range magnetic correlations that extend to at least this temperature [5]. These measurements also help to understand the large differences between the thermal conductivity of ThO_2 and UO_2. However, by 300 K any line-broadening is negligible, consistent with the recent work of Pang *et al*. [6].

In addition, we have observed a quasi-elastic signal (i.e. at zero energy transfer) that appears to peak at ~ 40 K (*above* T_N = 31 K), and, in contrast with the phonon line-broadening, appears isotropic. The origin of this quasi-elastic is currently uncertain, but may be a signal of polaron formation due to the local oxygen displacements and the quadrupoles coupling to the lattice. An initial effort to observe possible diffuse scattering at low temperature did not succeed.

References

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