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Temperature-dependent anisotropy in bond length of UO$_2$ as a result of phonon-induced correlations

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Previous experiments on cubic UO$_2$ have suggested that the temperature dependences of the nearest-neighbour U–O and U–U distances are different. We have acquired total-scattering neutron diffraction patterns out to Q=23.5 Å$^{-1}$ for 50 < T < 1023 K and produced via Fourier transform a pair-distribution function PDF(r). The PDF(r) shows quite clearly that r(U–O), defined by the maximum of the U–O peak in the PDF(r), does in fact decrease with increasing temperature, whereas r(U–U) follows the lattice expansion, as expected. We also observe that the r(U–O) contraction accelerates continuously above T $\approx$ 400 K, consistent with earlier experiments by others. Furthermore, by analysing the eigenvectors of the phonon modes, we show that the $\Delta$(TO) phonon tends to separate the eight equivalent U–O distances into six shorter and two longer distances, where the longer pair contribute to a high-r tail observed in the U–O distance distribution becoming increasingly anisotropic at higher T. These results have significance for a wide range of materials in which heavy and light atoms are combined in a simple atomic structure [1].

Neutron PDF(r) results from 50 to 1023 K. The r(U-U) follows the thermal expansion, as it must. However, the r(U-O) decreases with increasing temperature as a result of the phonon correlations. The r-space resolution is about 0.16 Å (FWHM) for a Q$_{\text{max}}$ = 23.5 Å$^{-1}$. Taken from Ref. [1]