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Temperature-dependent anisotropy in bond length of UO_2 as a result of phonon-induced correlations

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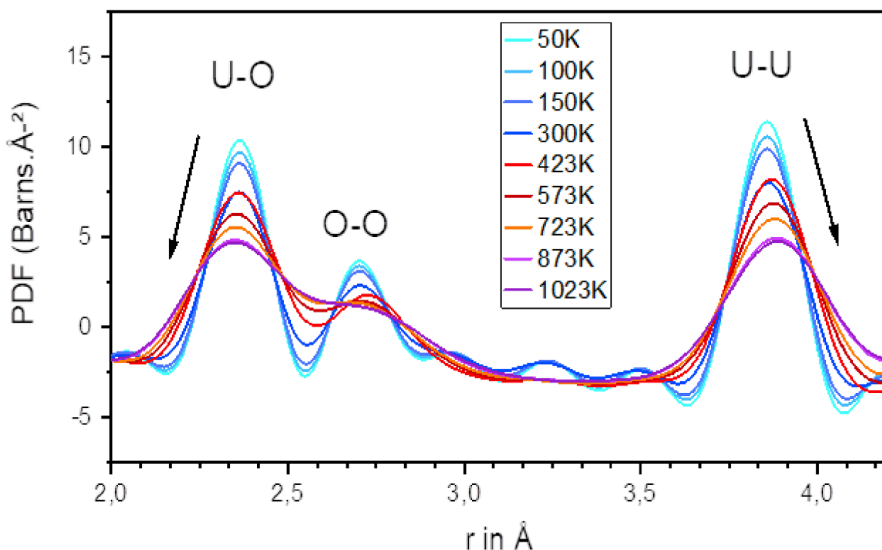
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Previous experiments on cubic UO_2 have suggested that the temperature dependences of the nearest-neighbour U–O and U–U distances are different. We have acquired total-scattering neutron diffraction patterns out to $Q=23.5 \text{ \AA}^{-1}$ for $50 < T < 1023 \text{ K}$ and produced via Fourier transform a pair-distribution function $\text{PDF}(r)$. The $\text{PDF}(r)$ shows quite clearly that $r(\text{U–O})$, defined by the maximum of the U–O peak in the $\text{PDF}(r)$, does in fact decrease with increasing temperature, whereas $r(\text{U–U})$ follows the lattice expansion, as expected. We also observe that the $r(\text{U–O})$ contraction accelerates continuously above $T \approx 400 \text{ K}$, consistent with earlier experiments by others. Furthermore, by analysing the eigenvectors of the phonon modes, we show that the $\Delta_5(\text{TO}_1)$ 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 $\text{PDF}(r)$ results from 50 to 1023 K. The $r(\text{U–U})$ follows the thermal expansion, as it must. However, the $r(\text{U–O})$ decreases with increasing temperature as a result of the phonon correlations. The r -space resolution is about 0.16 \AA (FWHM) for a $Q_{\text{max}} = 23.5 \text{ \AA}^{-1}$. Taken from Ref. [1]

[1] L. Desgranges *et al.* J. Physics: Condensed Matter **35** 10LT01 (2023)

