



HAL
open science

Temperature-dependent anisotropy in bond length of UO₂ as a result of phonon-induced correlations

Lionel Desgranges, Gianguido Baldinozzi, Henry Fischer, Gerard H Lander

► **To cite this version:**

Lionel Desgranges, Gianguido Baldinozzi, Henry Fischer, Gerard H Lander. Temperature-dependent anisotropy in bond length of UO₂ as a result of phonon-induced correlations. 52nd Journées des Actinides (JdA) and 14th School on the Physics and Chemistry of the Actinides (SPCA), The Max Planck Institute for Chemical Physics of Solids, Dresden, Apr 2023, Dresden, Germany. hal-03956840

HAL Id: hal-03956840

<https://cnrs.hal.science/hal-03956840v1>

Submitted on 25 Jan 2023

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



Distributed under a Creative Commons Attribution 4.0 International License

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

L. Desgranges¹, G. Baldinozzi², H. E. Fischer³, and G. H. Lander^{3,4}

¹ IRESNE, CEA Cadarache, 13108 Saint-Paul-lez-Durance, France

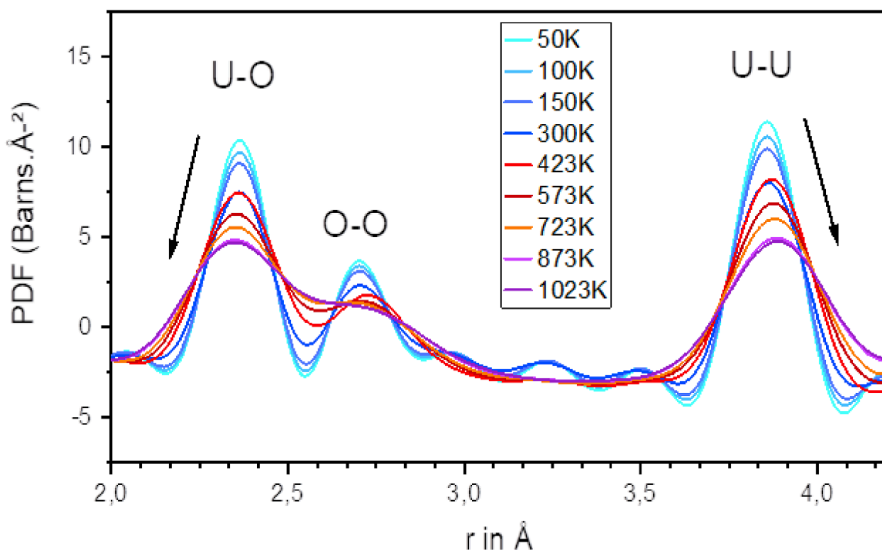
² CentraleSupélec, CNRS, SPMS, Université Paris-Saclay, 91190 Gif-sur-Yvette, France

³ Institut Laue Langevin, 38042 Grenoble, France

⁴ European Commission, Joint Research Centre, 76125 Karlsruhe, Germany

e-mail: lander@ill.fr

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 PDF(r). The PDF(r) shows quite clearly that $r(\text{U–O})$, defined by the maximum of the U–O peak in the 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 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)

