Experimental Report

Proposal: **5-24-506** **Council:** 4/2012

Title: Neutron powder diffraction study of the zero birefringence points in congruent and stoichiometric lithium tantalate

This proposal is resubmission of: 5-24-481

Researh Area: Physics

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Samples: Li0.954Ta1.046O3
LiTaO3

Instrument	Req. Days	All. Days	From	To
D2B	3	3	03/12/2012	06/12/2012

Abstract:

The birefringence of lithium tantalate varies as a function of lithium content and temperature, resulting in a range of temperatures at which lithium tantalate becomes zero birefringent. We would like to investigate the precise structural changes across this zero birefringence point as a function of temperature for 7Li-enriched congruent lithium tantalate and stoichiometric lithium tantalate. By using neutron diffraction, the position of lithium in the structure, and critically, how this position changes across the zero-birefringence point, will be observed for the first time. These measurements will aid in understanding how lithium tantalate can be simultaneously electrically polar and optically isotropic.

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Neutron powder diffraction study of the zero-birefringence points in congruent and stoichiometric lithium tantalate

The structural changes in stoichiometric and congruent lithium tantalate (LT) around their zero-birefringence points have been investigated between 10 and 445 K using powder diffraction measurements on D2B. Rietveld refinements of the data were performed using TOPAS academic[1]. For the stoichiometric sample, diffraction patterns were collected every 15 K between 300 and 445 K with the expected zero-birefringence point at 373 K[2].

The refinement of the measured diffraction data at 300 K for the stoichiometric sample is shown in figure 1. The calculated Ta displacement, Li displacement, octahedral distortion and octahedral tilt for the stoichiometric sample are shown in figure 2. The Ta displacement and Li displacement both decrease over the temperature range studied. The octahedral distortion is small and negative, but because of the large uncertainties on the calculated value, a trend with temperature cannot be determined. The octahedral tilt decreases as the temperature increases. These structural changes are continuous and linear across the temperature range investigated, without any abrupt changes at the expected zero-birefringence point. This confirms the zero-birefringence point occurs as a result of the linear changes in the Li displacement, Ta displacement and octahedral tilt.

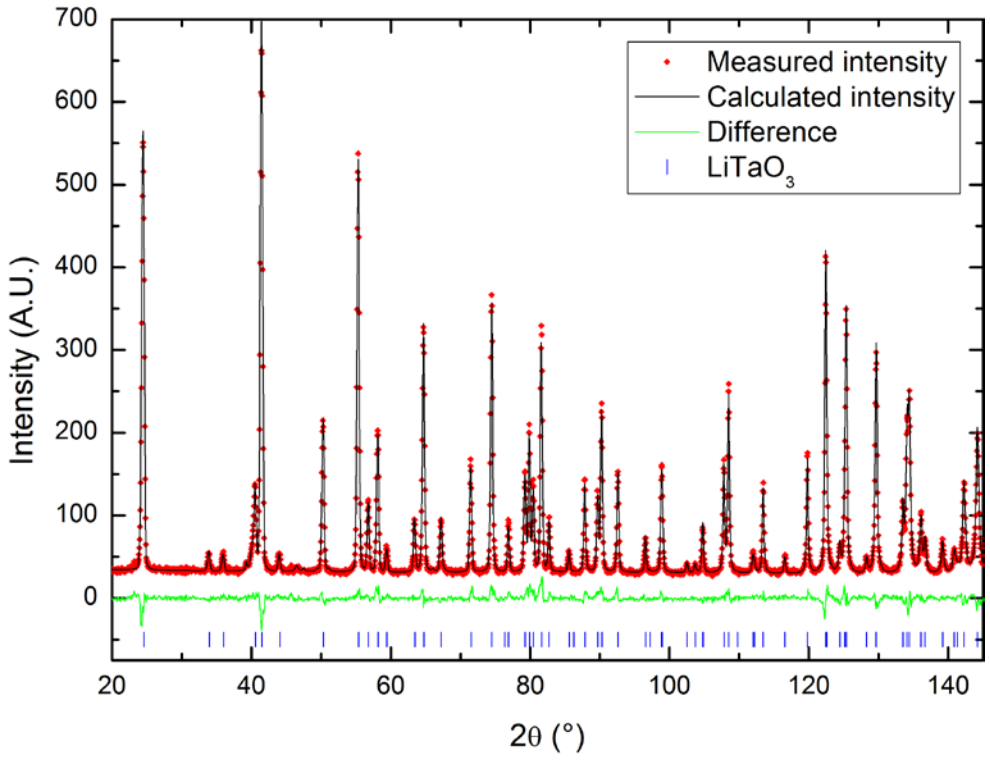

Figure 1: Rietveld refinement of data measured at 300 K.

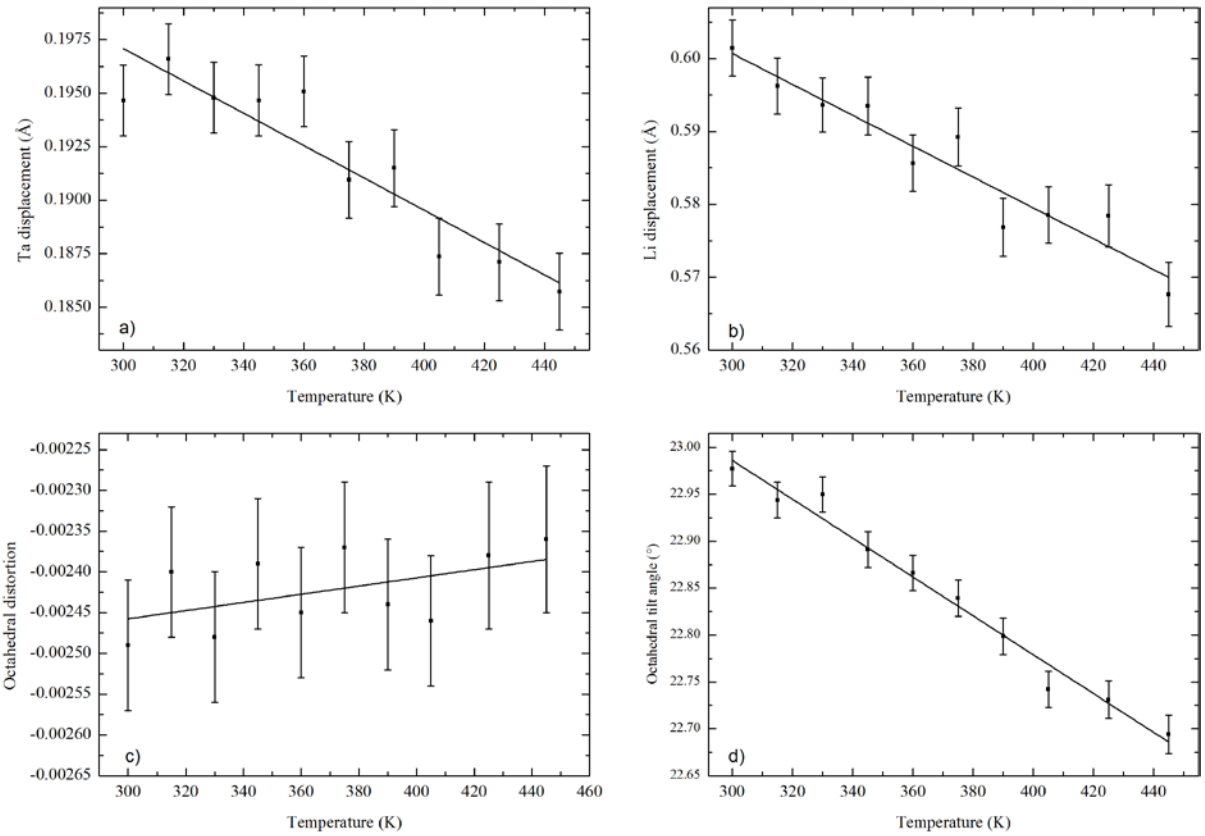

Figure 2: The a) Ta displacement, b) Li displacement, c) octahedral distortion and d) octahedral tilt angle as a function of temperature.

The stoichiometric LT structures determined from the neutron diffraction measurements have been used for a series of DFT calculations focusing on the optical properties. The structures used for the calculations are based on the linear fits to the Ta displacement, Li displacement and octahedral tilt, with the octahedral distortion kept constant. Calculations where only the Ta atom, Li atom, O atom or the lattice parameters have been varied have also been performed to investigate the effect of each atom on the optical properties.

The birefringence determined from these calculations has been plotted in figure 3. There is a good agreement between the calculated birefringence when all the parameters are varied and when only the Ta position is varied, which suggests that the birefringence is heavily dependent on the displacement of the Ta atom from the centre of the O octahedra. The change when only the Li position and O position are varied, both result in a small increase in the birefringence, while the increasing lattice parameters result in a small decrease.

A manuscript for publication is in preparation.

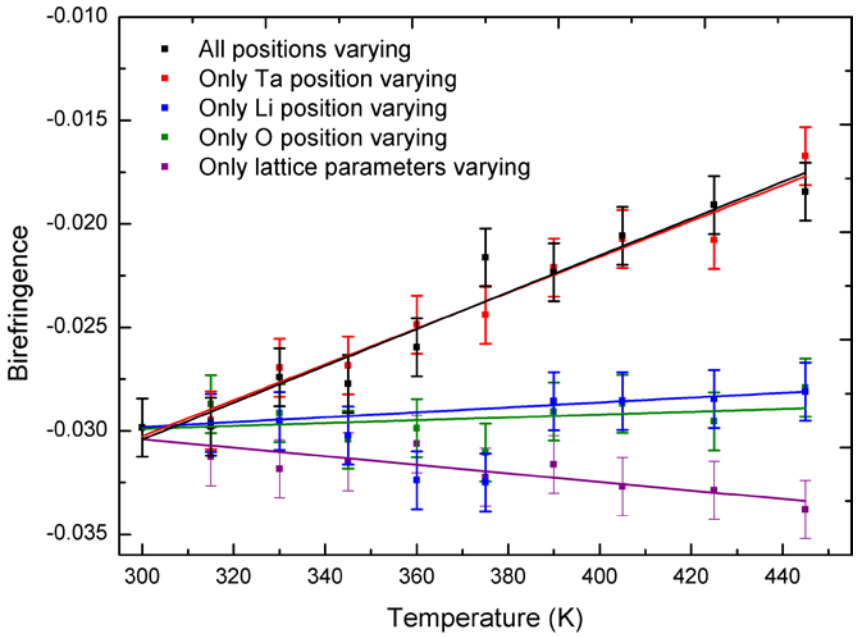

Figure 3: The calculated birefringence using the structures determined form the neutron diffraction measurements.

References

[1] A. Coelho, (2007) http://www.topas-academic.net/.

[2] C. Bäumer, D. Berben, K. Buse, H. Hesse, and J. Imbrock, Appl. Phys. Lett. **82**, 2248 (2003).