Experimental report

19/06/2024

Proposal: **7-01-603**

Council: 10/2023

Title: Search for lamellar fluctuations in incipient ferroelectrics

Research area: Materials

This proposal is a new proposal

Main proposer: **Frank WEBER**

Experimental team:

Local contacts: Paul STEFFENS
Alexandre IVANOV

Samples: $KTaO_3$

Instrument	Requested days	Allocated days	From	To
IN8	6	0		
THALES	6	6	23/11/2023	29/11/2023

Abstract:

$KTaO_3$ (as well as $SrTiO_3$) has long been considered a prototypical paraelectric, lying at or extremely close to a quantum critical point such that slight compositional substitutions induce low temperature ferroelectricity or even superconductivity to emerge as ground-states. However, recently Guzman-Verri et al. performed calculations including coupling of the ferroelectric order to strain gradients (flexoelectric coupling)1 and predict an incommensurately modulated phase which can be suppressed to zero temperature by thermal fluctuations. Thus, the behavior of $KTaO_3$ is not dominated by quantum fluctuations but rather can be understood by classical thermal fluctuations. Here, we propose to measure the softening of the TA (hybridized TA-TO) mode close to the zone center in $KTaO_3$ with high momentum and energy resolution and look for signatures of incommensurate correlations in the (quasi-)elastic scattering. Phonon spectroscopy on the cold TAS THALES can provide decisive experimental evidence for (or against) this new scenario for an alleged quantum paraelectric.

	Experiment title: Search for lamellar fluctuations in KTaO3	**Experiment number**: 7-01-603
Beamline: ThALES, ILL	**Date of experiment**: from: 2023-11-23 to: 2023-11-29	**Date of report**: 08.12.2023
Shifts:	**Local contact(s)**: Paul Steffens	
MONO – PG002, ANA – PG002, fixed k_F = 2.0 Å^{-1} (≈8 meV) Collimation: open – 40 – 20 – open Most data were taken with no horizontal focusing (due to collimation) but keeping the vertical focusing; some data were taken with fully flat Mono and Ana – labelled "HR"		

We performed a study of the transverse acoustic (TA) phonon mode in $KTaO_3$ dispersing along the [100] and [110] directions. Measurements were done at T = 2K – 300K starting close to the zone center, $\boldsymbol{q}$ = (0.025,0,0) and (0.025,0.025), and investigating the dispersions up to q = (0.3,0,0) and (0.4,0.4,0). Measurements were done close to the reciprocal lattice point $\boldsymbol{\tau}$ = (0,0,2).

% Motivation

The idea for the experiment was based on a recent publication[1]. Here, Guzman-Verri *et al.* performed calculations of the phase space of a ferroelectric in which the order parameter couples to strain gradients (flexoelectric coupling)[1]. The key result of their work is that the flexoelectric coupling leads to an incommensurately modulated phase which can be suppressed to zero temperature by thermal fluctuations and that the behavior of $KTaO_3$ and $SrTiO_3$ is not dominated by esoteric quantum fluctuations but rather can be understood by classical thermal fluctuations. In Guzman-Verri's model, there is a structural instability at a finite, incommensurate wave-vector $\boldsymbol{q}_{mod}$. Essentially, the flexoelectric coupling will lead to a local minimum in the dispersion of the hybridized transverse acoustic and transverse optic (TO) modes.

The aim of the experiment was to search for this phonon anomaly in $KTaO_3$. Previous experiments reported already on the dispersion of the TA modes. However, the spacing of measured points in wave vector was quite large.

% experimental setup

Thus, we put a special emphasize on using the best possible resolution with the following points:

- Scattering plane 100-001, the dispersion along the [110] direction was investigated by tilting the Orange Croystat out of the scattering plane (up to 10°).
- Low final energy of k_f = 2.0 Å^{-1} (≈ 8 meV); a lower value is not possible since we have to close the scattering triangle with momentum transfers around the Brillouin zone center $\boldsymbol{\tau}$ = (0,0,2); that is the smallest scattering wave vector with reasonable phonon intensities

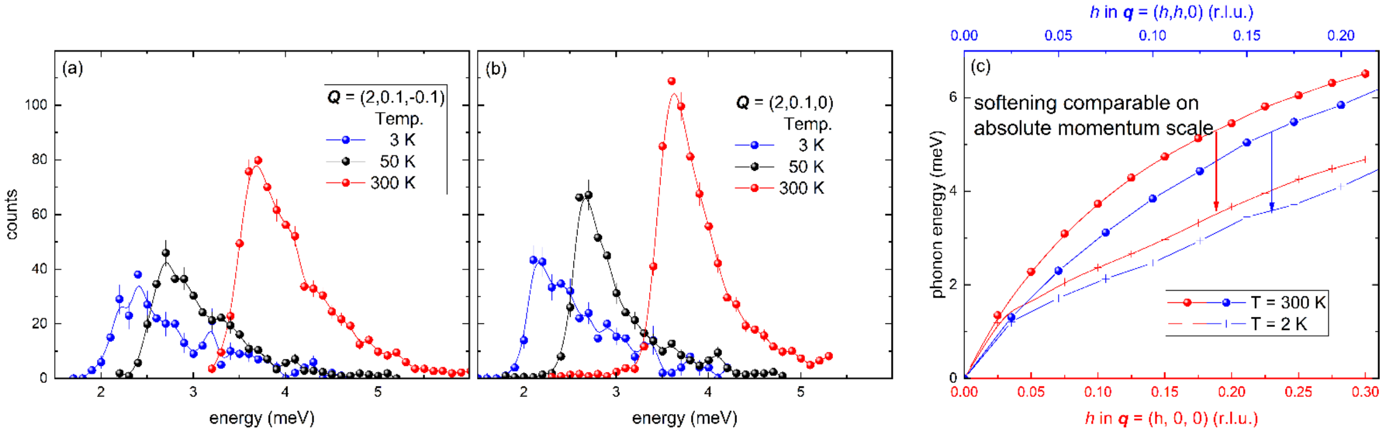

Fig. 1. Raw data and overall softening. (a)(b) Raw data from inelastic neutron scattering at (a) $\boldsymbol{Q}$ = (2,0.1,-0.1) and (b) $\boldsymbol{Q}$ = (2,0.1,0) for three temperatures. Data were normalized by the Bose factor of the corresponding phonon energy. (c) Dispersions at room (spheres) and base temperature (crosses) for the TA phonon dispersing along $\boldsymbol{q}$ || [100] (red, bottom axis) and $\boldsymbol{q}$ || [110] (blue, top axis). The arrows mark the temperature dependence at one wave vector. Wave vectors are scaled a factor of $\sqrt{2}$ to put the dispersion on a comparable scale.

- Horizontally flat monochromator and analyzer; collimation "open – 40 – 20 – open"
- Additionally, the vertical focusing was switched off for some scans

The resulting resolution was measured on a Quartz sample:

- ΔE = 0.35 meV at zero energy transfer
- ΔQ was 0.01 r.l.u. for a Bragg peak; we estimate to have ~0.02 r.l.u. for diffuse scattering

% Results

Some raw data and the most important results are summarized in Figure 1. The TA modes along the high-symmetry direction are at a valley of the dispersion. Since the walls of these valleys are steep, our resolution function picks up intensities of the corresponding lattice excitations at energies larger than the phonon energy at the high-symmetry position. This is visible as a high-energy tail of the observed neutron scattering intensities (Fig. 1(a,b)). The observed dispersions along both directions for room and base temperature are ploted in Figure 1(c). The horizontal axes (top: **q** II (110); bottom: **q** II (100)) are scaled by a factor of sqrt(2) such that the absolute values of the wave vectors are the same at a given position.

We conclude that there is no minimum of the phonon dispersion at a non-zero wave vector at any temperature. The dispersions at room temperature follow typical behavior. The softening is very small or even absent at small wave vectors $\boldsymbol{q}$ = (h,0,0) and (h,h,0) with h = 0.025. The softening is clear for h ≥ 0.05. The dispersions at base temperature seem to be almost linear for h ≥ 0.05.

% Conclusion and outlook

I think we have shown conclusively that there is no phonon anomaly in KTaO3. It will be interesting to compare this to the recent reports on SrTiO3,[2] where authors claim that there results are in reasonable agreement with the predictions by Guzman-Verri et al.

1 Guzmán-Verri, G. G., Liang, C. H. & Littlewood, P. B. Lamellar Fluctuations Melt Ferroelectricity. *Physical Review Letters* **131**, 046801, doi:10.1103/PhysRevLett.131.046801 (2023).

2 Fauqué, B., Bourges, P., Subedi, A., Behnia, K., Baptiste, B., Roessli, B., Fennell, T., Raymond, S. & Steffens, P. Mesoscopic fluctuating domains in strontium titanate. *Physical Review B* **106**, L140301, doi:10.1103/PhysRevB.106.L140301 (2022).