Experimental report

17/02/2025

Proposal:	**7-02-219**	**Council:**	10/2022
Title:	Low-temperature dynamics of the quantum critical soft-phonon mode in SrTiO3		
Research area:	Physics		

This proposal is a continuation of 7-02-212

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Samples: SrTiO3

Instrument	Requested days	Allocated days	From	To
IN8	6	6	12/04/2023	18/04/2023
ORIENTEXPRESS	1	1	12/04/2023	13/04/2023

Abstract:

SrTiO3 (STO) forms the textbook example of a quantum paraelectric and of a quantum critical ferroelectric system ¿ it naturally lies very close to a quantum critical point (QCP) where long-range ferroelectric order is suppressed to zero temperature. The relevant physics of STO are dictated only by the phonons making it an extremely clean model system, and the behaviour of the soft optical mode which would give rise to ferroelectricity has previously been characterised by inelastic neutron experiments down to 4.5 K.
Our recent dielectric measurements show that below 2 K, the inverse dielectric susceptibility displays an unexpected upturn which is attributed to the coupling of the soft optical mode with another acoustic phonon. This theoretical coupling of these phonon modes remains to be directly explored by inelastic neutron scattering below the temperature of this new dielectric behaviour. We propose to perform an inelastic scattering measurement using IN8 to measure the optical and acoustic mode in STO down to 50 mK. Direct observation of these phonon modes is necessary for developing this theory, which will impact also on the observation of superconductivity in doped STO.

Experimental Report – 7-02-219

Low-temperature dynamics of the quantum critical soft-phonon mode in $SrTiO_3$ (as continuation of 7-02-12)

We aimed to study the ground state dynamics of the phonon modes in the perovskite $SrTiO_3$ (STO) down to the novel "quantum polar-acoustic" regime below 2 K. This regime was first observed by high-precision dielectric measurements an upturn in the inverse dielectric constant below ~2 K which has been attributed to a coupling of the soft optical mode which would give rise to ferroelectricity with an acoustic phonon [1-2]. We sought to track the low-temperature behaviour of these modes to further develop the theory of this dielectric behaviour.

Our first trial (7-02-12) initially proposed to use Cu/Cu as monochromator/ analyser but ended up with using PG/PG during the experiment. The sample was successfully cooled down to ~0.04 K with dilution refrigerator. We managed to obtain the phonon energy dispersion in the desired temperature range where our dielectric data suggested novel quantum order [3], and compared with that in the quantum paraelectric regime by Yamada and Shirane [4]. However, the resolution in both E and q was limited to tackle the hybridization between two phonon modes at finite q, where the excitation energy of the transverse acoustic (TA) and the lowest transverse optic (TO), the "soft mode", was very close and the observed intensity showed broad peak feature instead of two clearly separate peaks as being observed and fitted at higher q, as shown in **Fig.1**. In addition, the transverse acoustic phonon demonstrated substantial intensity along [110] whereas exhibited extremely low intensity along [100] direction.

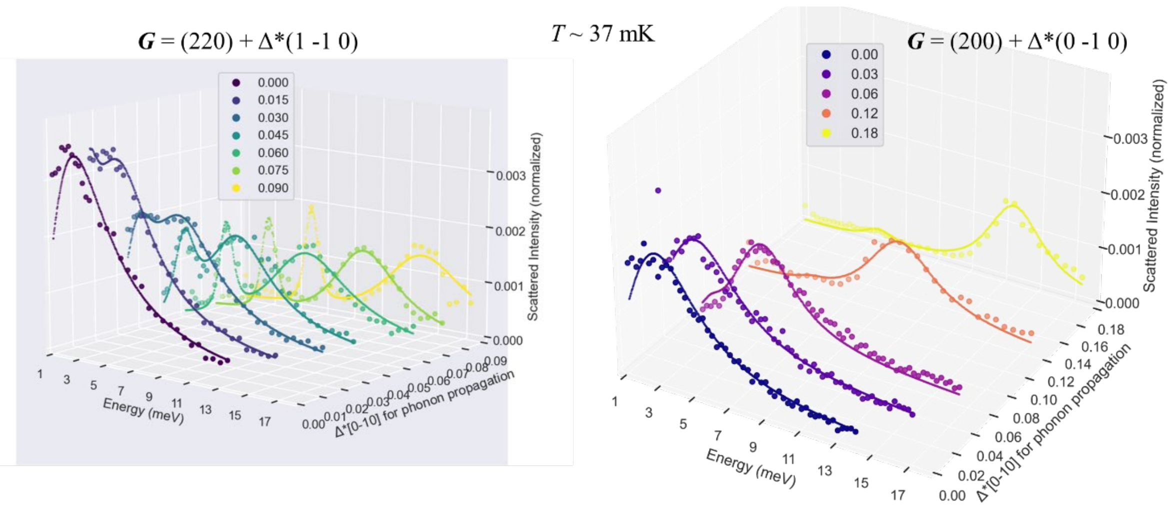

Fig. 1 Inelastic neutron scattering intensity energy scans from the zone centre (***q*** = 0) to increasing ***q*** up to 0.18 r.l.u. along the transverse direction of (a) [200] and (b) [220] in the reciprocal space. The data were collected using PG/PG as monochromator/analyser at 37 mK.

All these concerns pushed us to insist utilizing Cu/Cu as monochromator/ analyser at the expense of losing a great amount of observed neutron flux and focused on phonon modes along [110] direction. We used the same sample as before and did quick check at base temperature to ensure the data consistency. Then we let the sample to stabilize at 0.3 K to collect data from Brillion zone centre (ZC) to finite q. The data was analysed and plotted following the same procedure as described in experimental report 7-02-212, shown in **Fig. 2**.

It can be seen that the gained resolution managed to give a definite single peak at Brillion zone centre (Z.C.) out of the intensity from quasi-elastic tail, which was not available in 7-02-212.

We observe clearly two modes for Δ larger than 0.030 at ***G*** = (220) + Δ*(1 -1 0). Our observation at Δ=0.015, however, cannot be explained with the two phonon modes scenario, as shown on the right of Fig. 2. We believe this observation can be potentially explained by the hybridization between the transverse acoustic and optical phonon modes, as expected in the novel "quantum polar-acoustic" regime below 2 K. However, to solidify the arguments, we need to focus on specifically this finite and small ***q*** point and fully understand how the hybridization evolved across distinctive quantum ordering region.

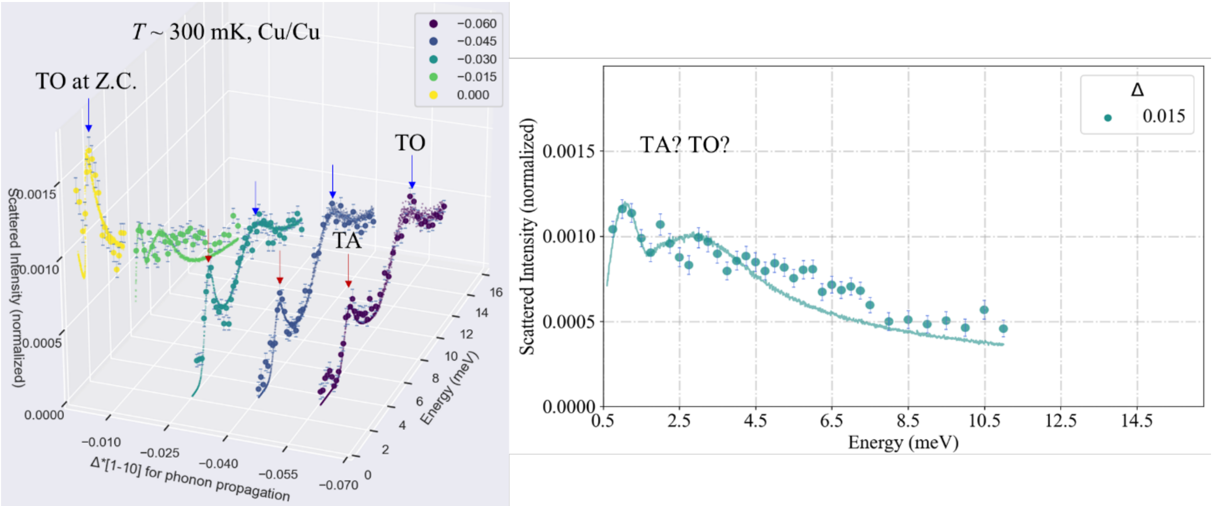

Fig. 2 Inelastic neutron scattering intensity energy scans from the zone centre (***q*** = 0) to increasing ***q*** up to 0.12 r.l.u. along the transverse direction of [220] in the reciprocal space. The data were collected using Cu/Cu as monochromator/analyser at 300 mK.

References

[1] S. E. Rowley et al., Nat Phys **10**, 367 (2014).
[2] M. J. Coak et al., PNAS **117**, 12707 (2020).
[3] ILL experimental report 7-02-212
[4] Y. Yamada and G. Shirane, J. Phys. Soc. Jpn. **26**, 396 (1969).