Experimental report

07/09/2022

Proposal: **7-02-213**

Council: 4/2021

Title: Dynamics of the quantum critical soft-phonon mode in SrTiO3 under pressure

Research area: Physics

This proposal is a new proposal

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Samples: SrTiO3

Instrument	Requested days	Allocated days	From	To
IN8	5	5	13/09/2021	18/09/2021

Abstract:

The insulating cubic perovskite SrTiO3 (STO) is an excellent example of a quantum critical paraelectric, lying naturally in close vicinity to a ferroelectric quantum critical point (QCP). Being free from the complications of free electrons or magnetism, it is an excellent system in which to study clean quantum criticality.

Recent studies have shown that pressure is an excellent parameter with which to cleanly tune STO away from this quantum critical point, and revealed new behaviour in the dielectric properties associated with the QCP. Relatively small pressures act to dramatically suppress the dielectric susceptibility across a wide temperature range by directly tuning the phonon modes which may be inferred from the dielectric data.

We propose to directly measure this soft phonon mode as a function of pressure by inelastic scattering using IN8. Observation of the stiffening of this soft mode with pressure (as inferred from dielectric data) is vital to verify and expand the current theoretical description, which is important for fully understanding the lowest temperature dielectric behaviour as well as the unusual superconductivity in doped STO.

Experimental Report – 7-02-213
Dynamics of the quantum critical soft-phonon mode in $SrTiO_3$ under pressure

At ambient pressure, $SrTiO_3$ (STO) exists close to a ferroelectric quantum critical point (QCP) in its phase diagram. Recent dielectric studies have shown that pressure is an ideal parameter with which to tune STO away from this QCP, with comparatively small pressures dramatically supressing the dielectric susceptibility across a wide temperature range [1]. From dielectric measurements, the energy of the soft phonon mode as a function of temperature and pressure has been inferred. We aimed to measure directly this soft mode using IN8 with a gas pressure cell up to 6 kbar.

The single crystal STO sample was loaded in the gas cell at 0.5 kbar and cooled to 2 K. Scans were performed at the zone centre ($\boldsymbol{q}$ = 0) and at $\boldsymbol{q}$ = -0.12 $Å^{-1}$ around the (200) and (220) Bragg peaks.

Following the measurements at 0.5 kbar, the pressure was increased to 5 kbar with the aim of maximising the observable difference from the lower pressure scan. Scans were taken at the zone centre for (200) and (220) positions, and at $\boldsymbol{q}$ = -0.12 $Å^{-1}$ from the (200). Similar scans were performed at 2.0 kbar and attempted at 6.8 kbar following discussion with the instrument scientist and sample environment technicians but the pressure here was found to be unstable, falling to below 5.0 kbar and effective measurements were not possible. Despite continued pressure instability, useful data was taken at 6.0 kbar. Finally, measurements were repeated at 2 kbar to explore potential pressure hysteresis effects.

Fig.1 summarizes the original scans for STO being loaded with the gas gate pressure cell at the (200) Bragg peak (red) and the background scans to capture the intensity contribution from the background of the pressure cell (blue). We rotate the A3 by 10° away from the Bragg peak and perform the energy scan following the same parameters as the scan at the Bragg peak. We expect ignorable contribution from any lattice points of STO and the scattering from the pressure cell is nearly uniform considering the amorphous nature of the aluminium alloy. The inelastic scattering from the phonon modes at the zone centre ($\boldsymbol{q}$ = 0) can thus been achieved by subtracting the counts of the red with the blue data. Once the background has been removed, we can continue to consider the convolution of the triple-axis resolution following the latest Eckold–Sobolev approach [2] as implemented in Takin2.0 software [3].

Fig.2 summarizes the energy scans for STO under a series of pressures 0.5, 2.0, 5.0 and 6.0 kbar around 2.45 K at the (200) and (220) Bragg peaks in the Brillouin zone. We fit the phonon energy intensity with the damped harmonic oscillator model [4] and overplot the fitting in Fig.2 as well. There is a clear indication of the pressure effect that the frequency (or equivalently energy) of the transverse optical phonon mode has been shifted to higher value. This is consistent with the previous high-precision dielectric measurement observing that pressure opens up the phonon energy gap and thus tunes the phonon spectrum away from criticality. However, the difficulty with regard to the stable control of the pressure and temperature during the energy scans prohibits further analysis of "clean" data out of the background. Whether there exist coupling with another phonon mode and how pressure tunes the phonon modes would require more efforts.

Initial results have been presented in oral presentations at The International Conference on Strongly Correlated Electron Systems 2022. (WedPA1:2.04)

<u>References</u>

[1] M. J. Coak et al., Phys. Rev. B 100, 214111 (2019).
[2] G. Eckold, O. Sobolev, Nucl. Instrum. Methods Phys. Res. A, 752, 54–64. 2014
[3] Weber T. Takin version 2 (Software)
[4] B. Fak, B. Dorner, Physica B 234-236 (1997) pp. 1107-1108

[2 0 0]

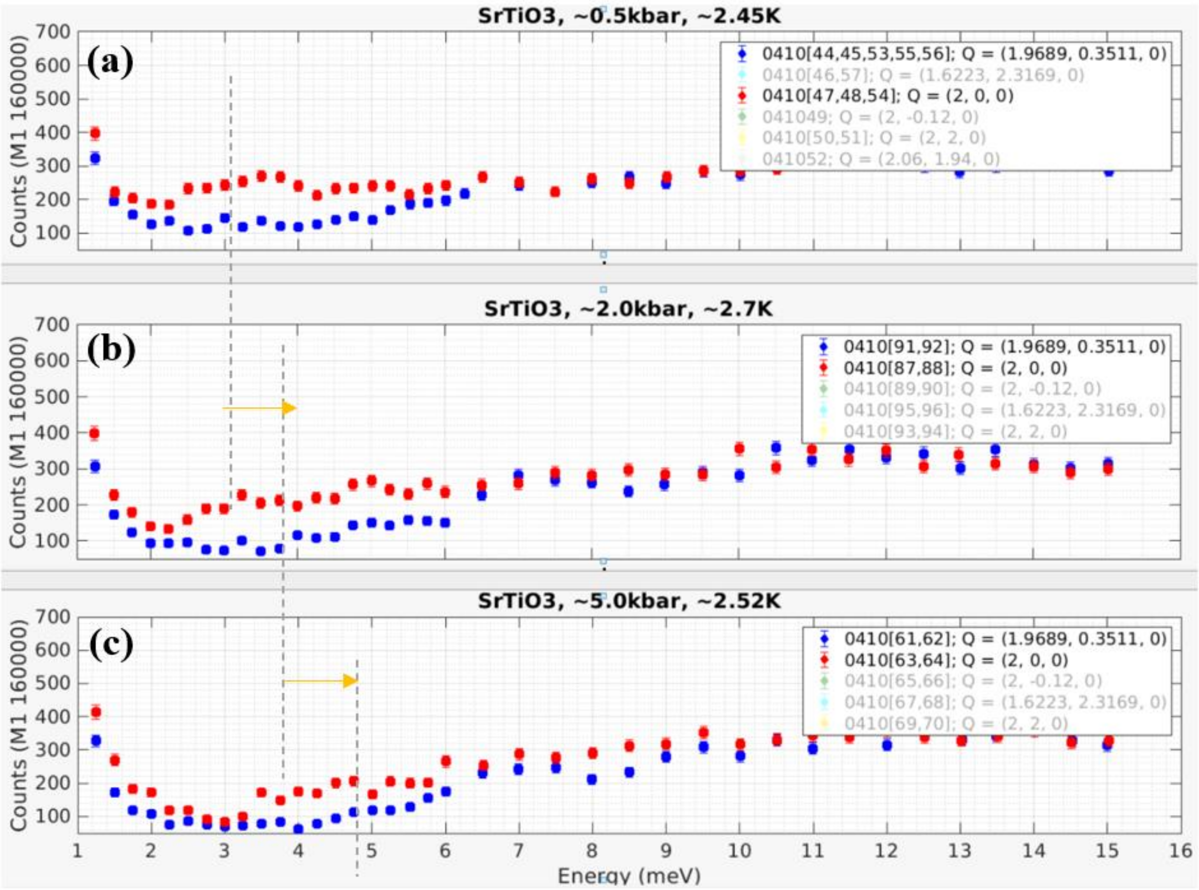
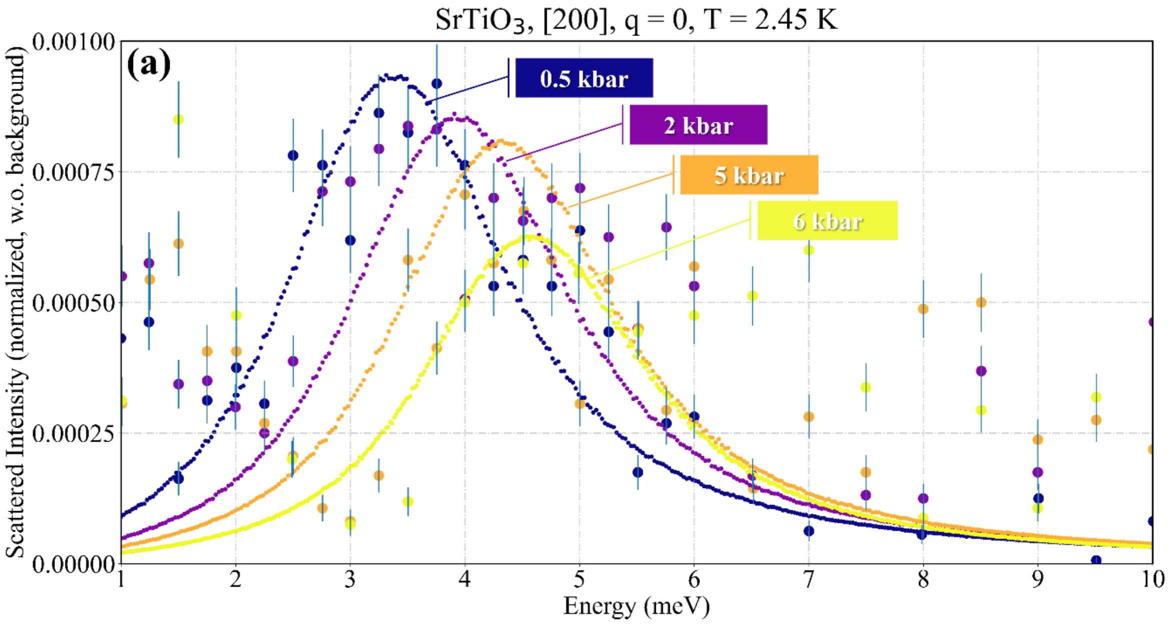


Fig.1 The Brillouin zone centre (***q*** = 0) energy scans under pressure at 0.5 kbar (a), 2.0 kabr (b) and 5.0 kbar (c) at (200) Bragg peaks. The red dots represent the STO sample together with the background from the gas gate pressure cell. The blue ones represent the corresponding background scans.



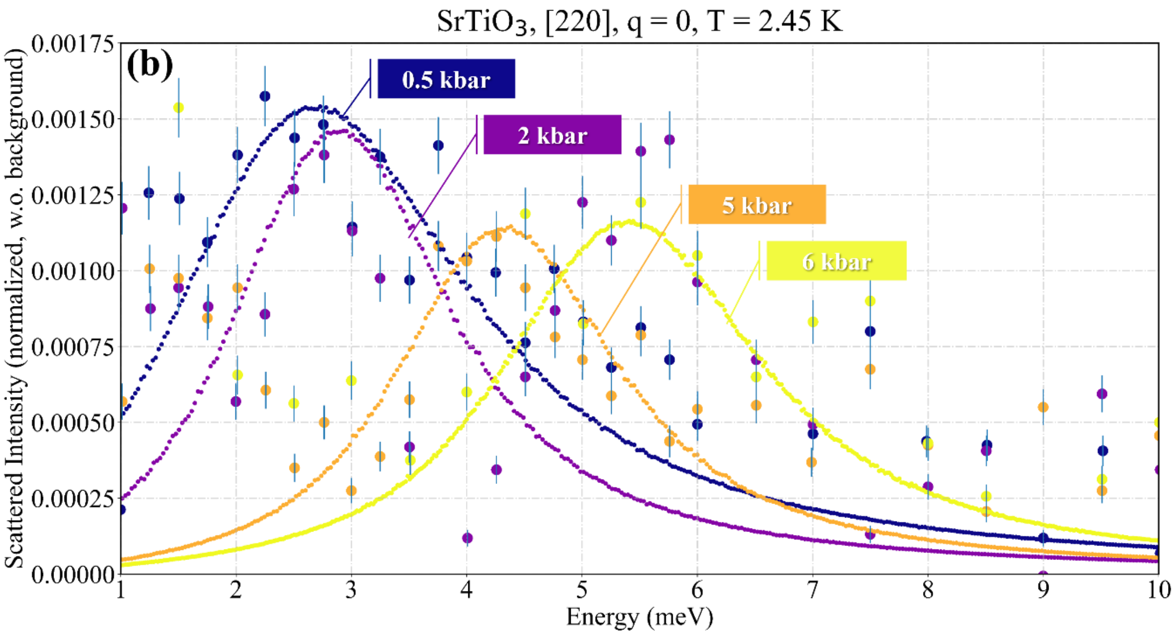


Fig.2 The Brillouin zone centre ($\boldsymbol{q}$ = 0) energy scans under pressure from 0.5 up to 6 kbar at ~2.45 K around the (200) and (220) Bragg peaks. The background contribution from the pressure cell has been subtracted. And the intensity from the sample itself has been fitted with *Takin2.0* to deconvolute the triple-axis resolution on IN8, shown in the dense dotted lines.