EUROPEAN SYNCHROTRON RADIATION FACILITY

INSTALLATION EUROPEENNE DE RAYONNEMENT SYNCHROTRON

Experiment Report Form

The double page inside this form is to be filled in by all users or groups of users who have had access to beam time for measurements at the ESRF.

Once completed, the report should be submitted electronically to the User Office via the User Portal:
https://wwws.esrf.fr/misapps/SMISWebClient/protected/welcome.do

Deadlines for submission of Experimental Reports

Experimental reports must be submitted within the period of 3 months after the end of the experiment.

Experiment Report supporting a new proposal ("relevant report")

If you are submitting a proposal for a new project, or to continue a project for which you have previously been allocated beam time, you must submit a report on each of your previous measurement(s):

- even on those carried out close to the proposal submission deadline (it can be a *"preliminary report"),*
- even for experiments whose scientific area is different form the scientific area of the new proposal,
- carried out on CRG beamlines.

You must then register the report(s) as "relevant report(s)" in the new application form for beam time.

Deadlines for submitting a report supporting a new proposal

- 1st March Proposal Round - **5th March**
- 10th September Proposal Round - **13th September**

The Review Committees reserve the right to reject new proposals from groups who have not reported on the use of beam time allocated previously.

Reports on experiments relating to long term projects

Proposers awarded beam time for a long term project are required to submit an interim report at the end of each year, irrespective of the number of shifts of beam time they have used.

Published papers

All users must give proper credit to ESRF staff members and proper mention to ESRF facilities which were essential for the results described in any ensuing publication. Further, they are obliged to send to the Joint ESRF/ ILL library the complete reference and the abstract of all papers appearing in print, and resulting from the use of the ESRF.

Should you wish to make more general comments on the experiment, please note them on the User Evaluation Form, and send both the Report and the Evaluation Form to the User Office.

Instructions for preparing your Report

- fill in a separate form for each project or series of measurements.
- type your report in English.
- include the experiment number to which the report refers.
- make sure that the text, tables and figures fit into the space available.
- if your work is published or is in press, you may prefer to paste in the abstract, and add full reference details. If the abstract is in a language other than English, please include an English translation.

Experiment title: Lattice dynamics and phason modes of the incommensurate composite [Sr]1+x[TiS3]		Experiment number: HC 4311
Beamline: ID 28	**Date of experiment**: from: 09/09/2020 to: 15/09/2020	**Date of report**: 26/02/2020
Shifts: 18	**Local contact(s)**: Alexei Bossak	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

Dr. Marc de Boissieu, Laboratoire SIMaP, CNRS Grenoble (France)

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Prof. Dr. Sander Van Smaalen, laboratory of crystallography, Bayreuth University (Germany)

Report:

Aperiodic crystals are long range ordered structure lacking lattice translation. Their structure is well described by the superspace approach which introduces additional degrees of freedom [1]. However, the behaviour and atomic realisations of the excitation modes associated to these new degrees of freedom are still debated. In the long wavelength limit, diffuse excitations called phason modes are predicted and have been observed as damped phonon modes in a few incommensurately modulated phases and as purely diffusive excitations in quasicrystals [1; 2]. However, their existence and characterisation incommensurate composites is controversial.

In this experiment, we proposed to measure phason modes in the well characterised incommensurate composite crystal $Sr_{[1+x]}TiS_3$. It is composed of two intermodulated sublattices of Sr and TiS_3 forming parallel chains along the c axis with incommensurate periodicities [3]. Phason modes would correspond to the relative ‘sliding’ of the two sublattices along c. Few theoretical predictions are available (e.g. [4; 5]). It has been suggested that the two sublattices might give rise to two different velocities, although this has not been proven rigorously. The phason mode or sliding mode should be measurable around the satellites reflections associated to the incommensurability and located along the c* axis.

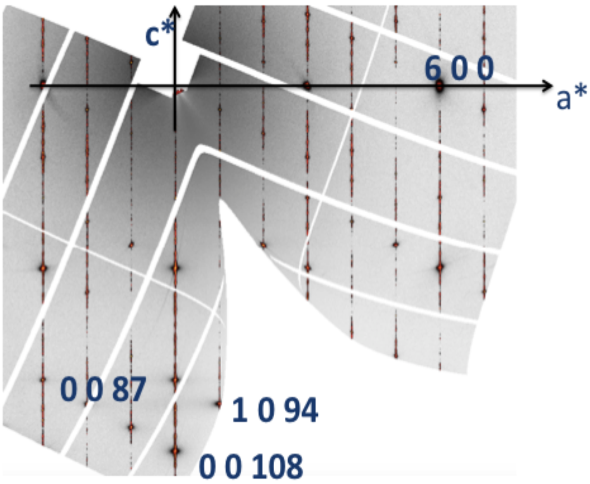

Figure 1 – Imaging the (h0l) plane.

We used a needle shaped sample of section 20 x 10 µm and length 200µm grown in the laboratory of crystallography, University of Bayreuth, by the group of Sander van Smaalen. First, we performed diffraction experiments at room temperature and at 100K using the ID28 side station. We recorded full maps of reciprocal space by a 360° phi rotation and 0.1° steps. The main plane in reciprocal space have been extracted as shown for the H0L plane the figure 1. We observed many satellite reflection along the incommensurability axis c* and were able to index the reflections as a 54/47 approximant, with c*= 2.990 $Å^{-1}$. We also observed diffuse scattering lines along c*, accounting for some correlations close to a tripling of the cell along (110).

The sample was then mounted on a goniometer head and oriented to work in the k=0 plane in order to perform inelastic X-ray scattering measurements at room temperature. We used the Si (9 9 9) setup and focussing optics with an energy resolution of the order 3 meV. We made our measurements on the (6 0 0) reflection which served as reference, the (0 0 108) which is a Sr sub-lattice contribution, the (1 0 94) which is a TiS3 sub-lattice contribution, and the (0 0 87) which is a mixed contribution of both sub-lattices. For each, we measured both longitudinal and transverse acoustic phonon dispersion. Energy scans were about 4 h long. In order to get enough points in the dispersions curves of the phonons, TA phonons required scans for at least four detectors positions, whereas TA phonons required only two.

We were not able to evidence phason modes, but acoustic phonon measurements do show some differences in the dynamic from a sublattice to another. Phonons were fitted with dumped harmonic oscillator in order to recover the dispersion curve as displayed on the figure 3. No major differences were found for the TA phonons in the acoustic regime with a similar slope of about 16meV/ $Å^{-1}$. On the other hand, the longitudinal phonon measurements are indicating a different sound velocity for the (0 0 108) reflection compared to (1 0 94) or (0 0 87) reflections, although we were not able to measure the phonon at low q. This indicates that the longitudinal modes do propagate differently along the Sr and TiS_3 chains of the lattice.

Although no indication of phason modes was found, the data analysis is still in progress and the preliminary results do suggest an actual differences in the dynamics relative to the two sublattices at room temperature. This results will be completed by more recent experiments on the diffuse scattering and low q phonon measurements that are supporting this difference in sound velocities. Further experiments would be necessary to compare with higher temperatures were sliding modes might be expected to show up.

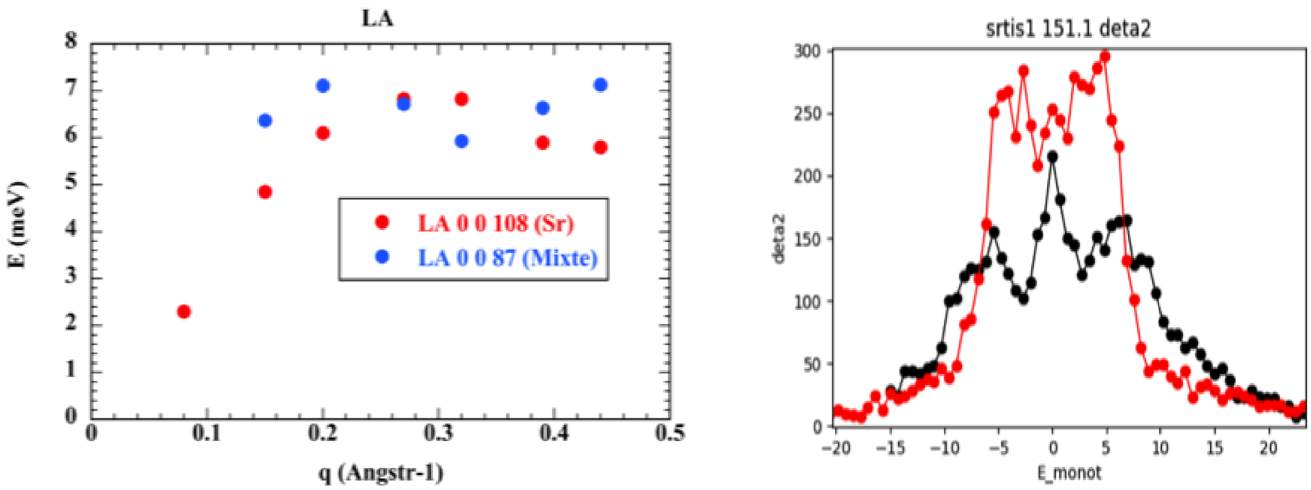

Figure 3 – Left: measured dispersion of the LA phonon around the (0 0 108) and (0 0 87) reflections. The q axis gives the distance to the Bragg reflection in $Å^{-1}$and the E axis gives the position in energy of the mode.
Right: comparison of a LA phonon measured around the (0 0 108) (red) and the (0 0 87) (black) reflection at q= 0,15$Å^{-1}$

References:

1 . Janssen T, Chapuis G and de Boissieu M, Aperiodic Crystals. From modulated phases to quasicrystals (second edition), 560 pages (Oxford University Press, Oxford,2018)
2 . de Boissieu M, Currat R and Francoual S 2008 in Handbook of Metal Physics: Quasicrystals (eds. T. Fujiwara and Y. Ishii) 107 (Elsevier Science)
3 . van Smaalen S, Incommensurate Crystallography, pages (Oxford University Press, 2012)
4 . Janssen T, Radulescu O and Rubtsov A N 2002 European Physical Journal B 29 85.
5 . Radulescu O and Janssen T 1999 Phys. Rev. B 60 12737.