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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

Reports supporting requests for additional beam time

Reports can be submitted independently of new proposals – it is necessary simply to indicate the number of the report(s) supporting a new proposal on the proposal form.

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.

Deadlines for submission of Experimental Reports

- 1st March for experiments carried out up until June of the previous year;
- 1st September for experiments carried out up until January of the same year.

Instructions for preparing your Report

- fill in a separate form for each project or series of measurements.
- type your report, in English.
- include the reference number of the proposal 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.

ESRF	**Experiment title:** Lattice dynamics of organic aperiodic composite	**Experiment number**: **HS 4656**
Beamline:	**Date of experiment**: from: 30/05/2012 to: 05/06/2012	**Date of report**: 11/07/2012
Shifts:	**Local contact(s)**: Alexey Bosak	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

L. Guérin*, C. Mariette*, B. Toudic*, P. Rabiller*, L. Khadeeva*, C. Ecolivet*

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Report:

Proposal summary:
Incommensurate composite structures can be considered as a new state of matter with specific structure, properties and dynamics. The goal of the experiment was to measure this dynamics in the prototype organic host-guest nonadecane-urea inclusion compound. In particular, gap and anomalous damping of pseudo-acoustic collective modes predicted by theoretical papers was investigated.

Experimental conditions:
A high energy resolution was reached with (Si 12,12,12) analysers configuration. The energy resolution was announced equal to 1.7meV full width at half maximum.
Due to the low interaction of X-ray with light atoms, organic materials have penetration depth of the order of 10mm; thus the samples few mm long are required. A large nonadecane-urea single crystal of ~10 mm long and ~8 mm large was successfully grown and was used. The obtained IXS spectra presented rather good statistics within few hours scans. Despite the large crystal size, an exceptionally good mosaicity was observed (less than 0.1 degree).

As proposed, the first part of the experiment was performed at room temperature. Supplementary low temperature measurements were performed in order to reduce phonon damping and to analyse the process in the other crystallographic superspace phases. This was done using a displex cryostat.

Technical problem encountered and possible improvement:
Temperature environment did not allow attaining very low temperature and neither to make precise temperature dependence measurements. Solutions for future experiments at low temperature exist using a low T He flow cryostat for example.

Results:

The main obtained results are the following:

1. Due to the accessibility to large value of Q (fig.1), we could for the first time measure the host longitudinal-acoustic (LA) phonons along the direction of the incommensurate wave vector c*(around 0 0 6 0 and 0 0 12 0 Bragg peaks) (fig. 2). Such Bragg peaks are not accessible using cold neutrons. We could by this way extract the urea dispersion branch and the associated damping confirming the very original features that we recently published measuring along a non pure longitudinal direction [1] (fig. 3).

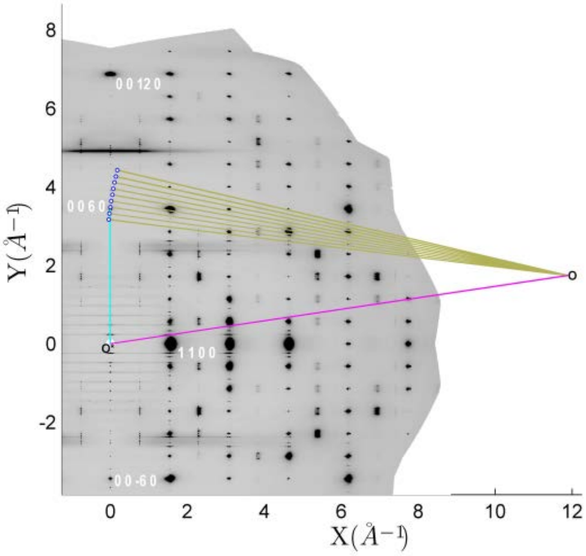

*Figure 1: Measured (**a*+b*, c***) reciprocal space used as the scattering plane in the IXS experiment. Schematic calculated positions of the nine analysers are shown in the case of the LA and sliding mode measurements around 0060 and 0061.*

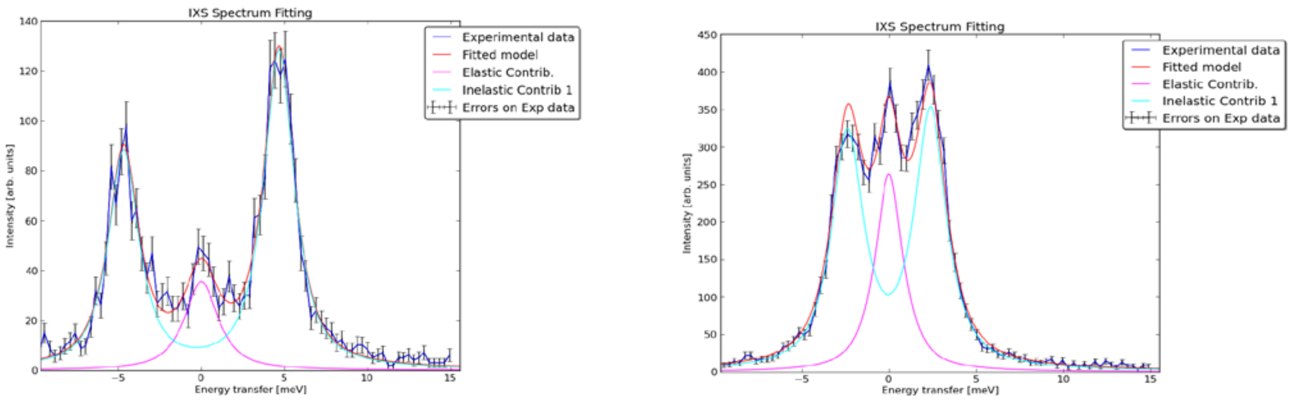

Figure 2 : inelastic LA phonon mode propagating along the incommensurate direction, measured at the (0 0 12.2 0) position(left) and (0 0 12.1 0) position (left) . Spectra obtained in 30 minutes.

2. Elasticity theory of periodic crystals require that transverse phonons propagating along a direction and polarized along another direction perpendicular to the propagation direction propagate at the same velocity as the transverse phonons obtained by exchanging the directions of propagation and polarization. This symmetry rule is broken for aperiodic structures due to the existence of an intermodulation related to an additional dimension. In that case, different slopes would be related to the absence of restoring forces between the two sublattices along the aperiodic direction. This fundamental question has been investigated here, measuring the phonon dispersion branches propagating and polarized along the (2 2 0 0) and (0 0 12 0) directions. Data analyses are in progress.

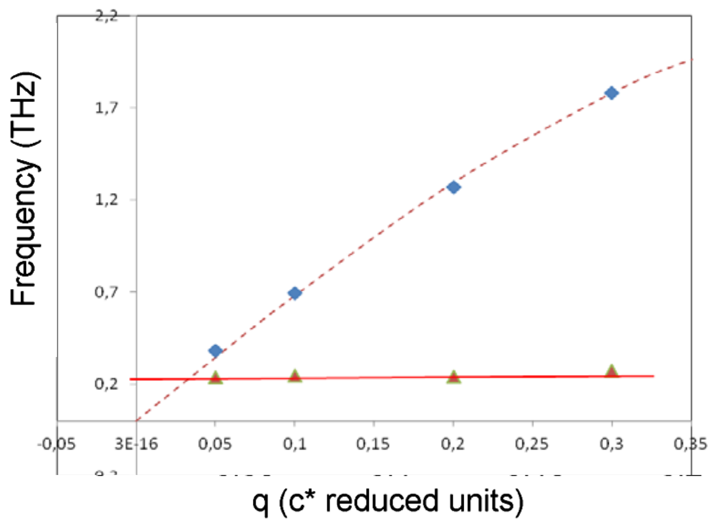

Figure 3 : Frequency and damping of a pure LA mode propagating along the incommensurate direction as measured around the (0 0 12 0) bragg peak at 117K. Damping appears constant in agreement with [1].

3. Several other exploratory studies were performed around previously determined interesting diffuse scattering positions in the reciprocal space. By this way, different information will be extracted concerning the translational and rotational molecular disorder of the guest and on the pretransitional critical phenomena.

4. The attempt to measure phonons around the real center of the Brillouin zone was unsuccessful due to the low inelastic structure factor around Q=0. Further calculations will be done to try to explain this observation.

5. Due to the relatively high intensity of the satellite Bragg peaks on this spectrometer, we decided to tackle the problem of the collective dynamics at a intermodulation satellite position. Due to limited time, this study was done at a single temperature in phase II. An overdamped low frequency mode seems to emerge from the (0 0 6 1) Bragg peak. If this is confirmed, this could be interpreted as the sliding mode and would constitute the very first unambiguous observation of this theoretically predicted excitation. Major information concerning interaction will be extracted from the obtained spectrum. Therefore a complete temperature analysis is strongly required.

Conclusion:

This first experiment of inelastic X-ray scattering on an aperiodic organic crystal was quite successful. The inelastic signal was strong enough and the energy resolution appears to be sufficient to answer many of our questions. The possibility of a direct observation of the sliding mode has to be confirmed. This would open a very exciting field of research in this family of materials. Systematic and precise studies with temperature will be needed to understand the original dynamics of this aperiodic crystal. The contribution of Alexey Bosak as local contact was essential in the success of the experiment.

[1] B. Toudic, R. Lefort, C. Ecolivet, L. Guérin, R. Currat, P. Bourges, and T. Breczewski, Phys. Rev. Lett. 107, 205502 (2011)