EUROPEAN SYNCHROTRON RADIATION FACILITY

INSTALLATION EUROPEENNE DE RAYONNEMENT SYNCHROTRON

ESRF

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.

ESRF	**Experiment title:** Revealing the nature of magnetic phase transitions in langasite-type materials	**Experiment number**: HC-5259
Beamline: ID15B	**Date of experiment**: from: 07-06-2023 to: 10-06-2023	**Date of report**:
Shifts: 9	**Local contact(s)**: Dr Michael Hanfland	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

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Report:

1. Technical description

   Two DACs with rhenium gaskets were prepared to conduct high-pressure (HP), and low-temperature–high-pressure (LT–HP) experiments. Each sample chamber contained two natural single crystals of Al-rich and Fe-rich langasite-type structures. Additionally, ruby spheres were loaded for pressure measurements. He gas was loaded into the DACs as a pressure-transmitting medium using the high-pressure gas loading system at ESRF. After gas loading, the pressure inside DACs was 17 GPa and 0.6 GPa for HP and LT–HP, respectively. The HP experiment was performed at room temperature in a pressure range of 17–47.5 GPa with a step of 4 GPa. Decompression was carried out with 10 GPa steps. SCXRD data was collected after each pressure step. The LT–HP experiment consisted of two parts. Firstly, LT measurements were conducted in the range of 230–13.55 K with a step of around 50 K. Subsequently, the temperature was increased to 130 K and the compression started up to 37 GPa. At the end of the compression, the temperature was decreased to 60.5 K. Single-crystal data collection was performed at every P–T point.
2. Object of investigation

   Compounds of the langasite family are well known due to a wide range of applications. Recently, Fe-bearing langasite-type structures have attracted great interest due to their multiferroic properties. According to Mössbauer spectroscopic studies, Fe-bearing langasites change magnetic properties upon compression or cooling. It has been envisaged that magnetic transitions are associated with structural changes. Despite that, the single-crystal XRD data is still missing. The conducted study aimed to reveal the structural nature of changes in magnetic properties. Fe-bearing qeltite and Al-bearing paquite, belonging to the langasite family, were investigated for this purpose.

3. Results of the high-pressure experiment

Conducted high-pressure measurements have not revealed structural phase transitions. At all P points, the crystal structure was solved and refined in the initial trigonal space group *P*321. The difference between Fe-rich crystal and Al-rich crystal has not been observed. However, Fe^{3+} coordination has changed from tetrahedral to octahedral (Fig. 1). The most significant modification considers oxygens, which become highly disordered above 19 GPa. Moreover, their atomic displacement parameters are remarkably elongated. Above 35 GPa, oxygens cannot be determined reliably. In conclusion, the pressure-induced changes of magnetic properties in Fe-bearing langasite are not associated with structural phase transition.

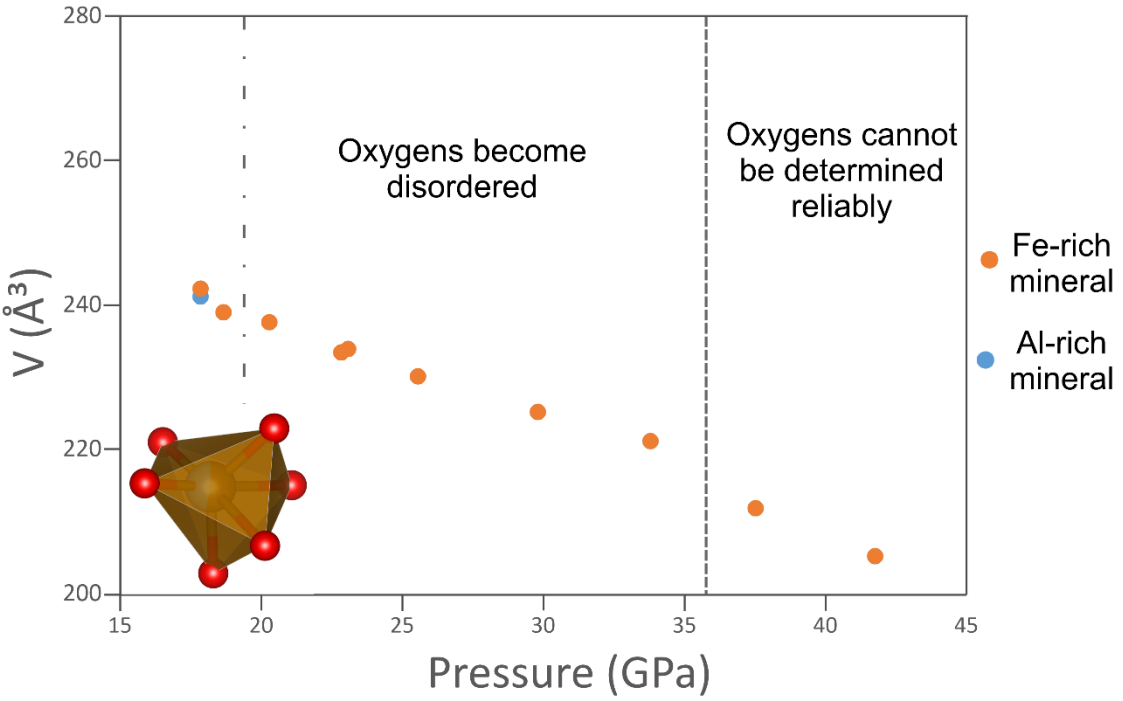

Fig. 1 Dependence of unit-cell volume on pressure at room temperature

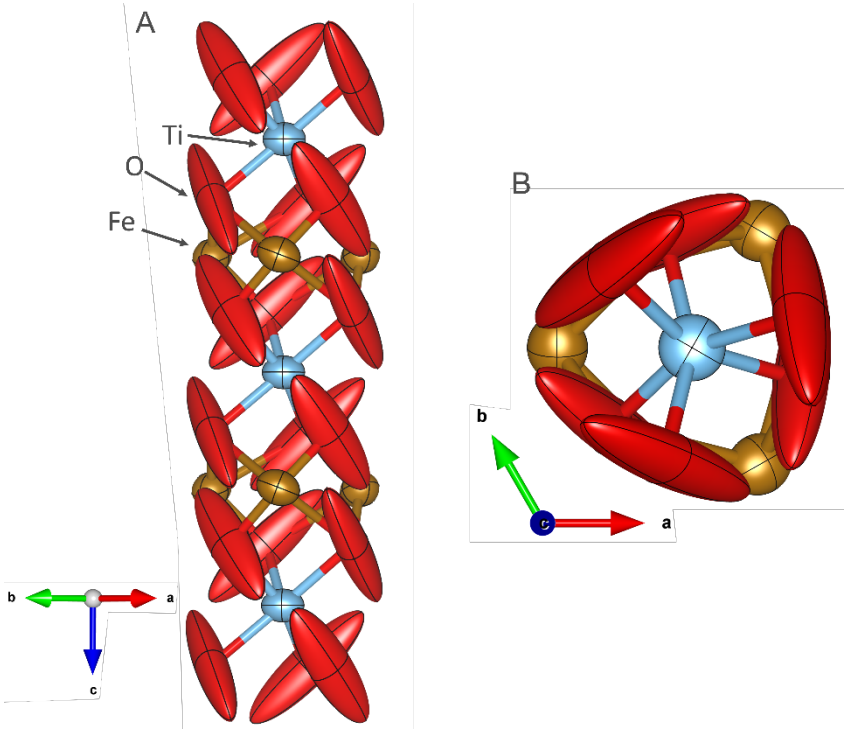

Fig. 2 (A) Fragment of langasite-type structure along [101] direction; (B) part of structure present in Fig. 2A along [001] direction.

4. Results of high-pressure and low-temperature experiment

Similar to high-pressure experiment, the low-temperature measurements did not reveal structural phase transition. At all P-T points, the structure was solved and refined in the *P*321 space group. However, the change of Fe^{3+} coordination has not been observed. Nevertheless, the dependence of the unit-cell volume on the temperature is noteworthy (Fig. 3). The volume of both crystals decreased with the temperature dropping up to 180 K. Below this temperature the unit-cell volume increases with the cooling. According to the literature, this phenomenon has not been noted in other langasite-type compounds. Probably, in our experiment, it was caused by technical problems and needs further investigation. Results of the experiments with compression at low temperature of 140 K are comparable to the high-pressure experiment at room temperature (Fig. 4). The Fe^{3+} coordination increased to VI. Oxygen sites split at 18 GPa.

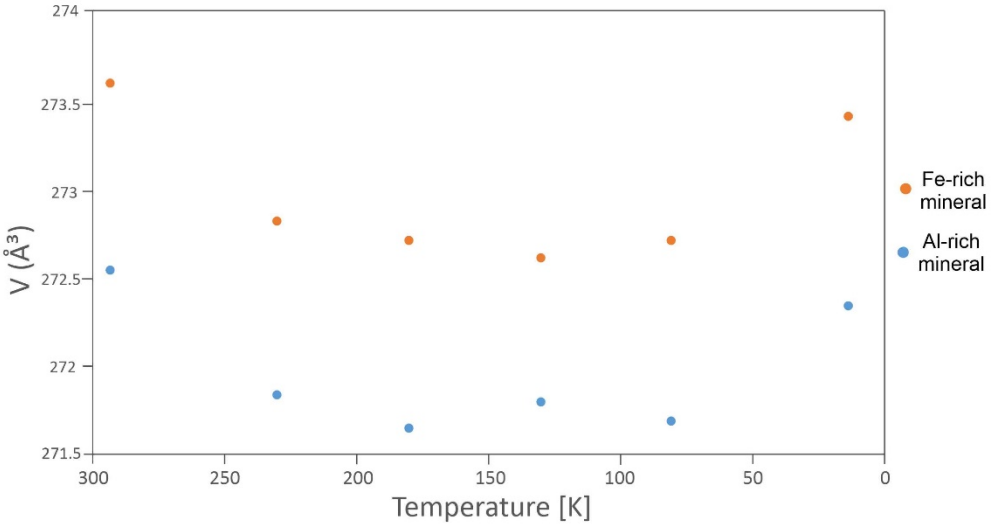

Fig. 3 Temperature-induced evolution of unit cell volume

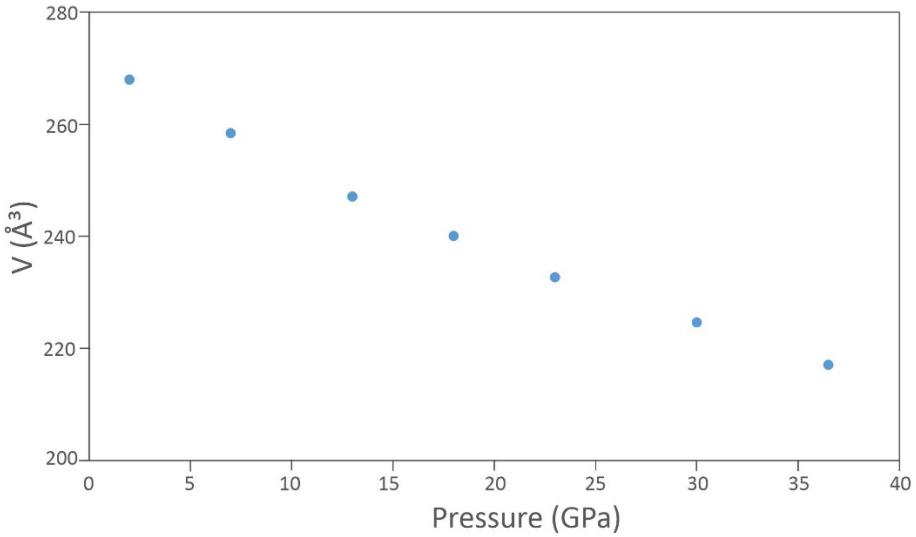

Fig. 4 The volume dependence on pressure at 130 K