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.

<table>
<tr><td>ESRF</td><td>Experiment title:
Origin of the Large Electromechanical Properties</td><td>Experiment number:
MA 3134</td></tr>
<tr><td>Beamline:
ID31</td><td>Date of experiment:
from: 26/08/2016 to: 28/08/2016</td><td rowspan="2">Date of report:
12/12/2016
Received at ESRF:</td></tr>
<tr><td>Shifts:
6</td><td>Local contact(s):
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<tr><td colspan="3">Names and affiliations of applicants (* indicates experimentalists):
Dr, Jurij Koruza [1], Prof. Dr. John Elliott Daniels [2,*], M. Sc. Hairui Liu [1,3,*]
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</table>

Report:

Introduction

Electric field-induced phase transitions have been proposed to play an important role in electromechanical response of many perovskite piezoelectric systems in the vicinity of structural phase transitions. *In-situ* electric field dependent high-energy X-ray diffraction data has been employed as a direct evidence of phase transition under electric field in both lead-based (example $Pb(Zr,Ti)O_3$ [1]) and lead-free (example $(Na_{0.5}Bi_{0.5})TiO_3$-$BaTiO_3$ [2,3]) piezoelectric systems. However, the direct evidence of electric field-induced phase transition and its possible contribution to electric field-induced strain was missing in $(Na,K)NbO_3$-based single crystals, primarily due to the great challenge in obtaining high-quality crystals with low leakage currents. Prior to this experiment, Mn-doped $(Li,Na,K)(Ta,Nb)O_3$ single crystals with enhanced electric field-induced strain in the vicinity of the orthorhombic–tetragonal (O–T) phase transition (about 100 ^oC) were grown [4], providing the opportunity to explore the underlying relationship between electric field-induced phase transition and enhanced strain. The direction of the applied electric field, generally along the crystallographic orientation of the investigated single crystal samples, is expected to have a strong influence on ferroelectric polarization rotation. Therefore, strain response along different electric field direction is expected to be different.

The objective of this experiment was to explore the field-induced phase transition of two $(Li,Na,K)(Ta,Nb)O_3$ single crystalline samples with different crystallographic orientations using high-energy X-ray diffraction under *in-situ* electric field. This is believed to indicate the origin of the large electromechanical properties observed in the vicinity of the orthorhombic–tetragonal (O–T) phase transition in perovskite lead-free piezoelectric Mn-doped $(Li,Na,K)(Ta,Nb)O_3$ single crystals. Two crystallographic orientations were selected to be investigated at the ID 31 beamline of the European Synchrotron Radiation Facility.

Experimental produce

Mn-doped (Li,Na,K)(Ta,Nb)O_3 single crystal were grown by the submerged-seed solution growth method. Two rectangular-shaped samples with different crystallographic orientations ($[001]_{PC}$ and $[110]_{PC}$), were cut and polished. In addition, an undoped (Na,K)(Ta,Nb)O_3 single crystalline sample with $[001]_{PC}$ orientation was prepared. The sample size was 0.8x0.8x0.5 mm^3. The two parallel faces with the size of 0.8x0.8 mm^2 were sputtered with silver. Prior to the measurements, the samples were immersed into silicone oil. Detailed description of the designed sample cell can be found elsewhere [5]. The sample cell was then mounted on a rotation table, in order to realize monochromatic rotational sweep collection strategy [6], for recording as many reflection signals as possible, which are later used for calculating the electric field-induced lattice strain. X-ray diffraction data were collected perpendicular to the samples' lateral side before and during the electric field loading.

The incident X-ray beam with the energy of approximately 78.40 keV, width of 300 μm and height of 300 μm was used. The scattering patterns were collected using a Pilatus3 X CdTe 2M detector. The sample cell was rotated along the axis of electric field direction with data collected in 0.1 degree rotation integrations. The samples were measured at various temperatures: room temperature, 100 ^{o}C and 120 ^{o}C, which provided pure orthorhombic, orthorhombic–tetragonal and pure tetragonal phase structures, respectively. The applied maximum electric field was 3 kV/mm.

The data were resampled into reciprocal space coordinates for visualisation and analysis.

Results

In the first shifts of the beamtime we measured the Mn-doped (Li,Na,K)(Ta,Nb)O_3 single crystals. Unfortunately, the collected data failed to show any obvious change of individual diffraction spots upon application of the electric field. Moreover, the multi-domain structure made it difficult to resolve the strain response under field loading process. It should be noted that some of the collected data are still being analysed.

In the second part we measured the (Na,K)(Ta,Nb)O_3 single crystal and selected data obtained at room temperature are presented below. Figure 1 presents 3D surface around a (330) diffraction spot. Figure 2(a) reveals the existing superstructure with half order peaks obvious in the principal reciprocal lattice planes before the loading of electric field. Figure 2(b) shows the diffuse features of the reciprocal lattice and how they change orientation under the maximum field. This preliminary results give evidence that lattice strain response could be induced by electric field and is dependent on the crystallographic direction. The observed superstructures might contribute to enhanced properties and were demonstrated to be changed by electric field loading.

Further refinements and comparison of the data with macroscopic electromechanical response are needed for an in-depth understanding of the electric field-induced lattice strain behaviour in (Na,K)(Ta,Nb)O_3 single crystal.

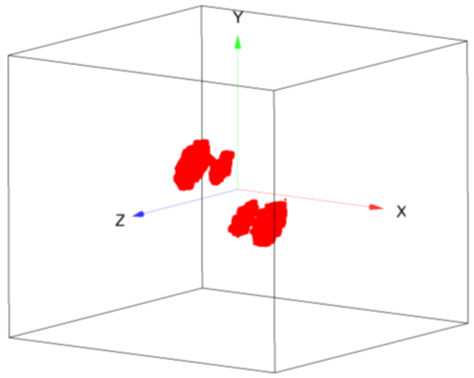
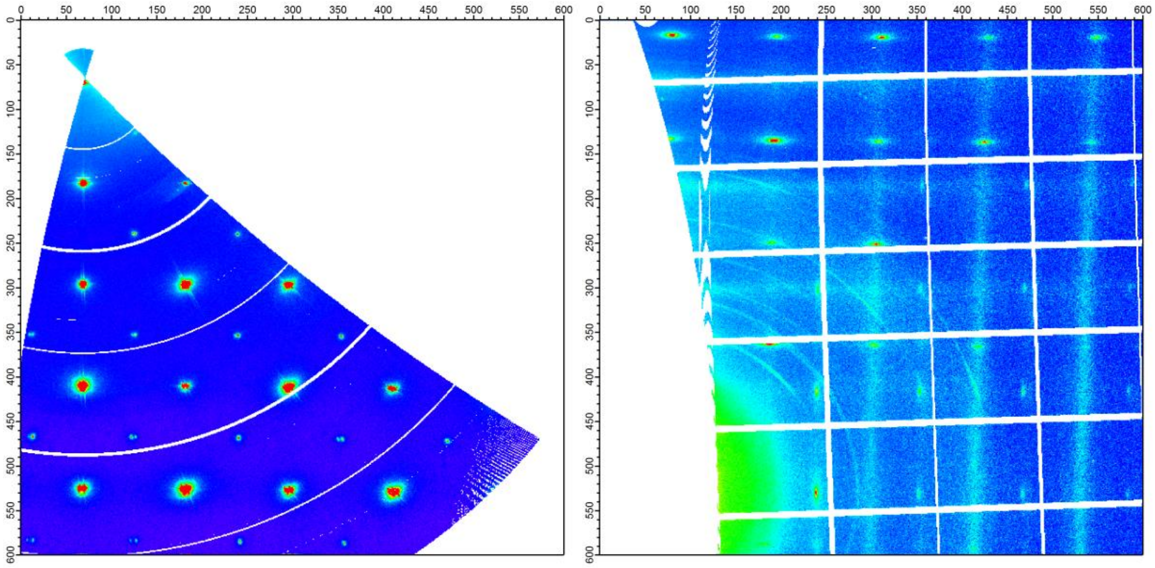


Figure 1. 3D lattice remanent strain response of a $(Na,K)(Ta,Nb)O_3$ single crystal along the [330] orientation.



(a) **(b)**

Figure 2. Diffraction patterns from the $(Na,K)(Ta,Nb)O_3$ single crystals (a) along [hk3] before electric field loading and (b) along [5kl] at maximum field.

References

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