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Neutron Diffraction Study of Piezoelectric Material under Cyclic Electric Field using Event Recording Technique

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Abstract. The stroboscopic data reduction technique for pulsed neutron diffraction was used in order to investigate the behaviours of the crystal lattice and the ferroelectric domains of the piezoelectric material in the multilayer-type piezoelectric actuator under a cyclic electric field. The division of neutron intensity data based on the condition of external fields recorded as event data can be performed by this technique. The peak shift and the intensity variations depending on the strength of the field were successfully observed in the divided diffraction patterns. The microscopic deformation estimated from the lattice strain and the domain switching agreed with the macroscopic behaviour of the actuator. The usefulness of the technique for the study of responses of materials to cyclic electric fields was demonstrated.

1. Introduction

Recently, time-resolved *in-situ* measurement is attracting great interest in order to observe transient phenomena of materials under external fields. Event-recording system of Materials and Life science experimental facility (MLF) at J-PARC has an advantage for time-resolved measurements, because of "event" characteristic of the data. For this purpose, new techniques for the measurement and the data reduction were developed and tried to adopt for the evaluation of the steel during the deformation [1].

Here, the responses of the crystal structure and the crystal lattice of piezoelectric materials to a cyclic electric field were chosen as the target to be investigated using this technique. Since piezoelectric materials are used in a variety of devices including sensor, actuator and the others, they are one of the important materials to be investigated. Generally, the performance of the materials is expressed by the parameter which shows the macroscopic deformation called piezoelectric *d* parameter. On the other hand, piezoelectric effect originates from the microscopic phenomena that are lattice strains, domain switching and ionic displacements. To understand the macroscopic characteristics of the material, the microscopic characteristics should be investigated in detail. The responses of these characteristics to the applied electric fields and mechanical stresses of lead zirconate titanate (PZT)-based and other types of piezoelectric ceramics or single crystals were evaluated using *in-situ* neutron and X-ray diffraction experiments [2, 3, 4].

In this study, the first trial of application of the event-recoding technique to the neutron diffraction experiment in MLF to investigate the behaviour of the material under the electric field was performed. The microscopic responses of the PZT-based piezoelectric material in a multilayer-type actuator under driving by a cyclic electric field were investigated using the engineering neutron diffractometer TAKUMI at BL19 of MLF at J-PARC. The obtained data were reduced to the field-condition-resolved (time-resolved) diffraction patterns with the specific field conditions. The lattice strain and the domain characteristics of the material in each condition are investigated.

2. Experimental

A commercially obtained multilayer piezoelectric actuator (PSt150, Piezomechanik GmbH) shown in figure 1 was used as the sample for the neutron diffraction measurement. The schismatic drawing of the experiment is shown in the figure 2. The sample was mounted horizontally on the sample stage of TAKUMI with the axial (layer stacking) direction to be 45° to the incident neutron beam. The neutron diffraction intensities in the directions of parallel and perpendicular to the applied electric field were measured simultaneously by ±90° detector banks. The gauge volume for the measurement was restricted to be 2×2×8 mm^3 by the beam slit and the radial collimators. High-resolution mode that has the resolution $\Delta d/d$ of 0.2 % was used for the measurement.
The sinusoidal wave signal with the frequency of 0.5 Hz from a function generator (33210A, Agilent Technologies Inc.) was amplified to the voltage of 0 ~ 130 V by a high-voltage amplifier (HEPOT-5B20, MATSUSADA Precision Inc.) and applied to the sample. The signal applying to the sample during the measurement was recorded as event data by a TrigNET [5] module and DAQ-middleware [6] together with neutron diffraction intensity and T_0 event which is a sync signal with generation of a neutron pulse. A TrigNET module has two types of analog-to-digital converter (ADC) inputs, high-speed and low-speed. Here, a low-speed ADC input was used for recording the signal. In the experiment, the diffraction intensity was collected in 12 hours, and the accelerator power of J-PARC was 300 kW.
Referring to the signal event data, collected neutron event data, which has the information about the detected position on the detectors and the time-of-flight from the neutron source, was divided by the voltage condition of every 10 V in the range from 0 V to 130 V and converted to the diffraction patterns using the software Utsusemi [7] and Emaki [8] developed based on Manyo-library [9]. Note that very flexible data reduction based on various analog or digital signals and time can be done by using these module and software after the measurement. Further, the condition window for the data division can be set freely. The diffraction patterns were analysed using the software Z-Rietveld [10] in order to refine the peak positions, the lattice parameters and the integrated intensities of the peaks.

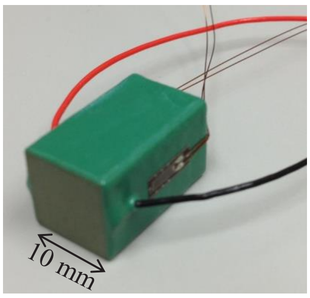

Figure 1. A multilayer piezoelectric actuator. A strain gauge is stuck on the side of the actuator in order to measure the macroscopic strain.

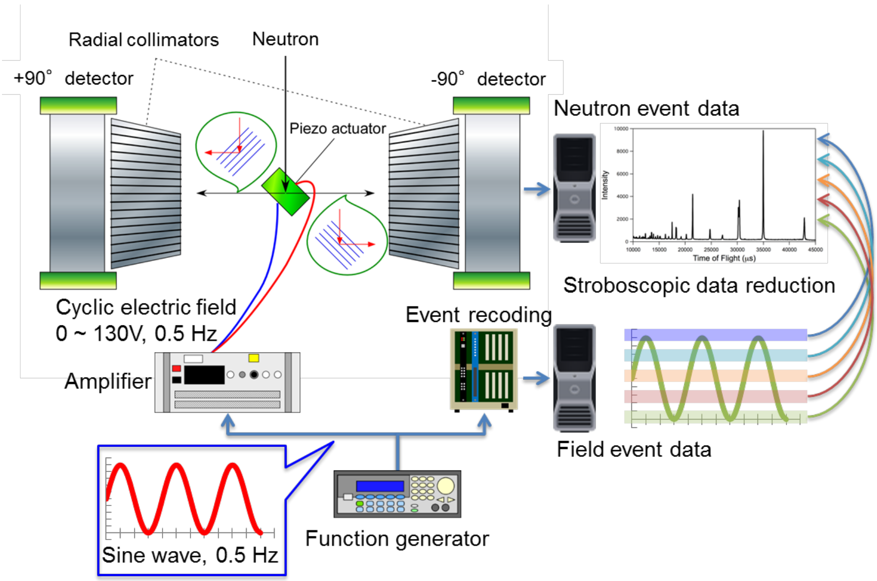

Figure 2. The schismatic drawing of the experimental setting.

3. Results and Discussions

All of diffraction peaks in the diffraction pattern without the field were characterized by the tetragonal unit cell with the space group of P4mm and the lattice parameters were refined to be a = 4.0251(1) Å and c = 4.0481(1) Å. These are close to the parameters of PZT that has a perovskite-type crystal structure. Figure 3 shows the peak profiles of 111, 200 and 002 reflections measured at axial direction of the actuator. The gradual peak shift and intensity variation depending on the electric field can be seen. The shift of 111 peak on the larger d side means an increasing of the lattice spacing by the applied electric field, and is a so-called inverse-piezoelectric effect. The decreasing of the intensity of 200 reflection and opposite variation in that of 002 reflection indicate the increase of the domains of which c-axis lies to the direction of the applied field. Both of these phenomena cause macroscopic elongation in this direction, and are agree with the nature of the actuator well. Thus the evolution of diffraction patterns is consistent with the macroscopic nature such as strain and electric field.

The electric field dependence of the lattice spacing of (111), (200) and (002) planes are shown in figure 4 (a). Although all the spacing increases with the rise of the field, there is crystal orientation dependence in the inclination. The deformation in [111] direction was larger than other directions, and its value was 0.09% at the field of 125 V in the present sample. The integrated intensities of 200 and 002 reflections in the axial direction at the various fields were obtained and the ratio of these two reflections was plotted in figure 4 (b). The ratio of I_{002}/I_{200} increased from 1.4 to 3.1 with the rise of the field by 90° domain switching. The deformation caused by this switching in the axial direction at 125 V is estimated from the variation of the volume fractions of the a-axis and the c-axis domains and the lattice parameters to be 0.09%. The macroscopic deformation of the actuator measured by a strain gauge was about 0.1 % at 125 V. The actuator consists of a polycrystalline material and includes various orientations of crystal grains. Because the behaviors of both the lattice strain and the domain switching have crystal orientation dependence, the contribution

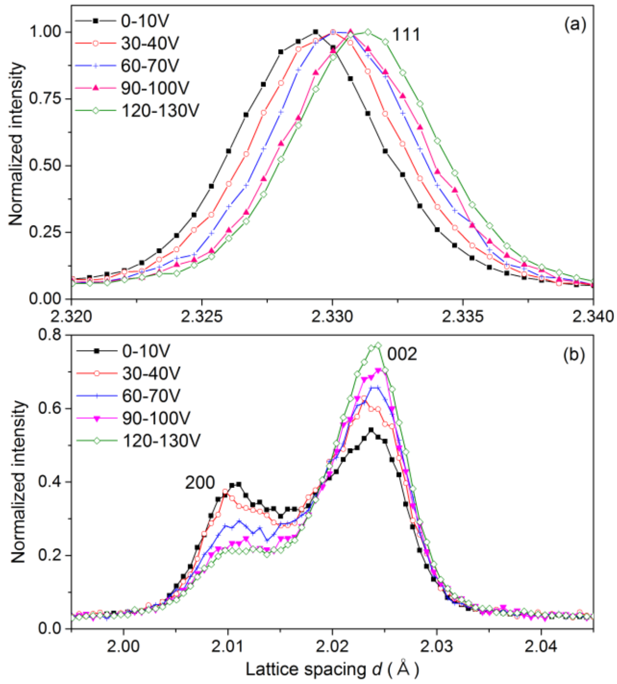

Figure 3. The diffraction peak profiles of (a) 111, (b) 200 and 002 reflections in the axial direction.

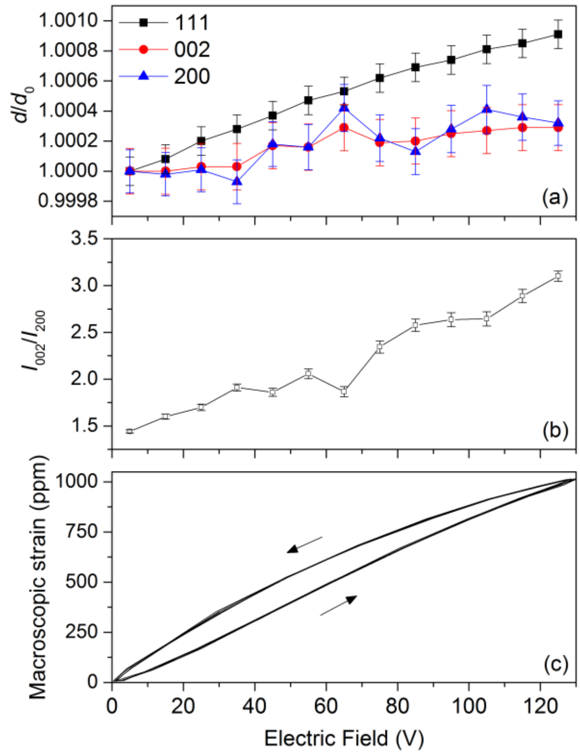

Figure 4. The electric field dependencies of (a) the strain in [111], [200] and [002] directions, (b) the intensity ratio of I_{002}/I_{200} and (c) the macroscopic strain.

of the microscopic deformation to the macroscopic one would be evaluated more precisely by adopting multi-peak analyses.
The measurement and the data reduction techniques which are used in the present study select and summed up the neutron events in the particular condition. Therefore, faster phenomena can be also observed if it has cyclic nature. On the other hand, the present data reduction is dividing data based on only the strength of the electric field, and it isn't classified whether the field is rising or declining. Because the hysteresis loop was found in the electric field-strain curve shown in figure 4 (c), the state of the materials may be different in these two situations. The data reduction based on the more complex condition should be done as the next step.

4. Summary

The behaviours of the lattice and the domain in the piezoelectric material under the electric field were investigated using event recording technique developed for time-resolved neutron diffraction experiment in MLF. Neutron diffraction intensities from the piezoelectric actuator were collected together with the waveform signal of the applied electric field as event data. The measurement and the data reduction were successfully finished and the stroboscopic diffraction patterns based on the strength of the field were obtained. The microscopic deformation which was estimated from the lattice strain and the domain switching of the material agreed with the macroscopic strain of the actuator. Further development of the measurement and the data handling techniques is in progress in order to analyse the behaviour more precisely. The extensive study of responses of materials to dynamical external fields is expected by applying the technique to neutron scattering experiments for various dielectric materials, ionic conductors and other materials which reply to an electric field.

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