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Author(s)	Kawamura, S.; Kaneko, J.H.; Fujimoto, H.; Otake, Y.; Fujita, F.; Homma, A.; Sawamura, T.; Mikula, P.; Furusaka, M.
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# Possibility of using a PMN-PT single-crystal as a neutron optical device.

S. Kawamura<sup>1</sup>, J. H. Kaneko<sup>1</sup>, H. Fujimoto<sup>2</sup>, Y. Otake<sup>3</sup>, F. Fujita<sup>1</sup>, A. Homma<sup>1</sup>,  
T. Sawamura<sup>1</sup>, P. Mikula<sup>4</sup>, M. Furusaka<sup>1</sup>

<sup>1</sup>Hokkaido University, North 13, West 8, Kita-ku, Sapporo 060-8628, Japan

<sup>2</sup>AIST, Umezono 1-1-1, Tsukuba 305-8563, Japan

<sup>3</sup>RIKEN, 2-1 Hirosawa, Wako, Saitama 351-0198, Japan

<sup>4</sup>Nuclear Physics Institute and Research Centre Rez Ltd., 250 68 Rez near Prague, Czech Republic

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## Abstract

Variations of the Bragg diffraction angle induced by electric fields on piezoelectric single-crystal were investigated with the aim of exploiting such crystals as novel neutron devices. A  $(1-x)(\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3)\text{-}x(\text{PbTiO}_3)$  (PMN-PT) single crystal with strong piezoelectric characteristics was used in the experiment. The sample was poled along  $\langle 001 \rangle$  direction before the experiment. In the course of diffraction measurements, a  $0.3^\circ$  change was observed in the diffraction angle at the maximum induced by the electric field. The diffraction angle variation shows time dependent behavior, which can be well described using a single exponential decay limited to a saturated value. This effect is considered a result of a change of the domain structure brought about through long-term application of an electric field for several hours. The obtained preliminary results confirmed the possibility of using a ferroelectric crystal as a neutron optical device.

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Key word : Neutron diffraction, Piezoelectric single-crystal, Optical devices

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## 1. Introduction

Silicon bent perfect crystals (BPCs) have been used widely as advanced neutron-monochromators and other novel optical-elements. The BPC elements are especially useful in neutron scattering devices when combined with position-sensitive detectors [1–2]. In the latter case, it is possible to carry out measurements at high resolution without losing luminosity [2]. However, this kind of device usually has very high resolution, sometimes too high; it is difficult to change the resolution. In some cases, variations of the bending radius within a piece of BPC would be desirable.

For that reason, we are trying to develop a new device using a piezoelectric-crystal. With such a crystal, we would be able to apply a varying electrical field along the crystal

to i) bend the crystal itself or ii) have a gradually changing d-spacing along the crystal. Other possibilities might include vibrating the crystal itself and exploiting the Doppler effect to use the device as a timing-chopper [3] or as a phase-space transforming device using Doppler shift.

Our literature searches have revealed very few papers describing the use of piezoelectric-crystals for a neutron optical device [4]. Therefore, we attempted first to elucidate the basic characteristics of such crystals; the change in d-spacing and corresponding Bragg angle as well as the effective mosaicity as a function of the value of the applied electric field. Using the neutron diffraction technique, we also tried to obtain information about the effect of a poling process (raising the temperature of a crystal above Curie temperature and subsequently cooling it under an applied electrical field), hysteresis, and time-dependent variation of d-spacing when the electric field is changed.

For the first experiment test, we chose a PMN-PT single-crystal ( $(1-x)\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-x(\text{PbTiO}_3)$ ) because it has superior piezoelectric characteristics and low hysteresis [5]. We were able to obtain a large amount of basic information regarding this crystal, which is applied widely to industrial applications.

## 2. Experimental set-up

A PMN-PT single-crystal used in the experiment was purchased from MTI Corp. Its dimensions were  $10 \times 8 \times 0.5 \text{ mm}^3$  and the face was parallel to the planes (001). The piezoelectric coefficient  $d_{33}$  was 2000 pC/N; the concentration of PT in the sample was estimated from the Curie temperature as 33% [6]. Aluminum electrodes of about  $1 \mu\text{m}$  thickness were evaporated on both faces of the crystal and 25- $\mu\text{m}$ -diameter gold wires were attached to the electrodes. The sample was poled at  $180^\circ\text{C}$  for 30 min under a loaded electric field (E) of 14 kV/cm. It was then field-cooled to room temperature [7].

Fig. 1 Experimental layout of ULS. Monochromatic neutrons of  $4.70 \text{ \AA}$  were used and rocking curves of PMN-PT were measured before and after applying the electric fields.

A diffraction experiment was carried out using the Ultra Small-Angle Scattering instrument (ULS) of the Institute for Solid State Physics (ISSP), University of Tokyo, installed at the JRR-3 reactor of the Japan Atomic Energy Research Institute (presently, Japan Atomic Energy Agency-JAEA). The schematic layout of the experimental setup is shown Fig. 1. The entire setup was situated on the optical bench of the ULS device accommodated in the hatch, which was temperature-controlled for  $24^\circ\text{C}$ . Monochromatized neutrons of  $\lambda=4.70\text{\AA}$  were extracted from the C-1 cold neutron guide tube using a PG (002) monochromator. Rocking curves (RC) of the crystal were obtained under various applied voltages from 100 V to 1 kV, which correspond to electric-fields between 2 kV/cm and 20 kV/cm. Full width at half maximum of these RC were about  $0.3^\circ$ , which is almost at the instrumental resolution limit. Two  $^3\text{He}$  proportional detectors were used: one for counting the diffracted neutrons and the other for transmitted beams. The statistical errors at each measuring point near the peak were about 2.4%.

## 3. Results and discussion

As shown in Fig. 2, the angular position of the diffraction peaks was changed by the electric field. Changes in its full width at half maximum were observed. The angular position of the diffraction peaks related to the rocking

curve measurements showed time-dependent variation after the applied electric field was changed from one value to another. Each variation was well described by a single exponential decay function limiting to a saturated value. The time constants had different values when applied electric fields were changed, but they were on the order of hours. Time constants are summarized in Table 1. The origin of this time-dependent variation of the diffraction angle is considered to occur through a slow relaxation of the domain configuration induced by the electric field applied to the crystals.

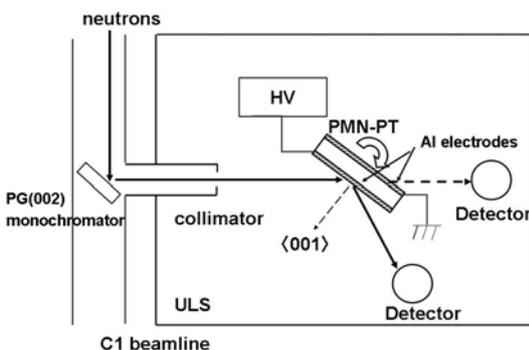
Saturation values of the angular position of the peaks of the rocking curves that were obtained from curve-fitting are shown in Fig. 3. A maximum diffraction-angle change of about  $0.3^\circ$  was obtained.

It is noteworthy that the saturated angular positions of the peaks of the rocking curves shifted toward one direction when the applied field was less than 7 kV/cm, but shifted to the opposite direction when a higher field was applied. That effect is attributable to the difference in the polarization direction in relation to the direction and amplitude of the applied field. However, an additional diffraction angle variation was not observed for a field higher than 15 kV/cm.

## 4. Conclusion

Dramatic changes of the d-spacing in the piezoelectric single-crystal (PMN-PT) were confirmed in this experiment. The Bragg-angle variations attributable to the applied electric field are sufficiently large and present the possibility of the crystal's use as a novel neutron optical device. This experiment is considered as a first-step; further experiments aimed at improving the characteristics of the device are in progress. First, the form factor of the (001)-reflection is intrinsically small because of the titanium it contains, which has a negative scattering length. The following experiments will be concentrated on the (111)-reflection, which has a much larger form factor. Furthermore, some dynamic experiments that apply a high-frequency electric field of megahertz order are planned for the near future.

Fig. 2 (upper) Rocking curves at each applied electric field. (middle) Time-dependent variations of the peak position on the rocking curve after changing the electric field of 0–14 kV/cm. The curve shows the result of fitting using a single exponential decay to a saturation value. (lower) Early behavior of the peak shift after applying different electric-field values.



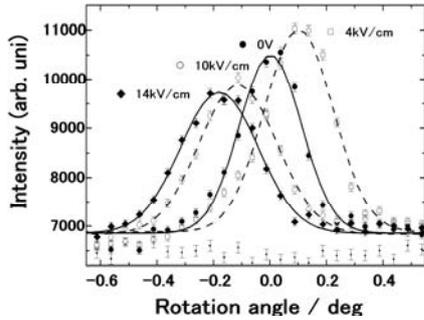
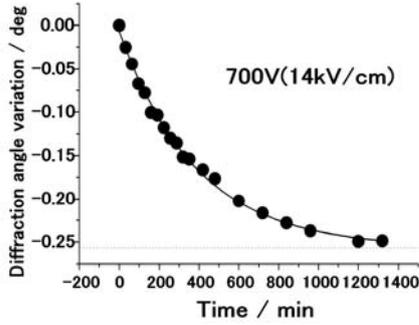


Fig. 3 Saturated value of diffraction angle variation for different values of the applied electric field.



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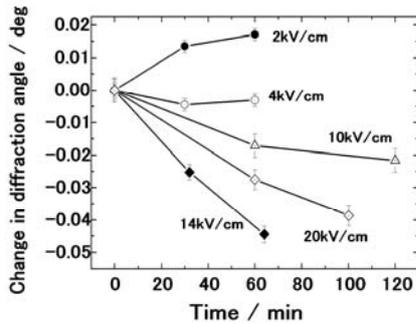


Table 1. Decay-time constants obtained from the time dependent variation of the diffraction peak angular position. They were obtained by fitting when assuming a single exponential decay to a saturated value.

E / (kV/cm)	Time constant / min
2	33.3
4	54.9
6	40.6
10	85.9
14	387
20	165

