ESRF	**Experiment title:** X-ray diffraction study of piezoelectric crystal response on the nanosecond time scale	**Experiment number**: HS-3789
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Report:

Interaction of a crystal with a permanent external electric field is well known in the form of the macroscopic phenomena such as dielectric polarization and converse piezoelectric effect. Deeper insights into the nature of the corresponding physical properties of solids (dielectricity and piezoelectricity) may be obtained by investigating the time dependence of the various processes that are initiated in a crystal by an ultra-fast change of an external electric perturbation state. In order to realize this experiment we applied periodically to single crystal plates a 2-step modulated high voltage (HV), consisting of U_+ = 1 kV and U_- = -1 kV HV states (see Fig. 1), with a frequency up to 3 kHz. The switching time between the both different HV states was of the order of 200 ns. The time-resolved scanning of the diffraction intensity for each single angle step of a ω-rocking curve (see Fig. 2) was performed using a commercial FPGA-board. In detail, the incoming digital signals of a scintillation counter were assigned to successive up to 20 ns short single time channels. The smallest time window of 20 ns that can be used is limited by the board clock frequency of 100 MHz, whereas the SRAM implemented on the FPGA restricts the number of time channels to 2^{18}. All measurements were carried out on a single crystal diffractometer at the BM01A (Swiss-Norwegian) beamline. To make visible the relaxation process of a crystal the time resolution was chosen in the range between 100 ns and 20 ns. Especially in the 20ns-mode we had severe counting statistics problems. Therefore each conducted measurement had to be repeated several times, so that by far not all relevant aspects of the experiment could be considered.

In addition to our measurements in June 2008 performed at $Li_2SO_4 \cdot H_2O$ crystal (see user report HS3355 [1]), during this beamtime we focussed on the piezoelectric response of BiB_3O_6 crystal. Compared to $Li_2SO_4 \cdot H_2O$ BiB_3O_6 exhibits a more than two times larger piezoelectric effect. We used a 0.5 mm thick (010) crystal plate and applied HV via thin gold contacts exactly sputtered on top of each other. In addition, this time we investigated the influence of the HV frequency on the observed relaxation process. The corresponding information was extracted from the shift of the Bragg peak position of single reflections, Δω,

recorded as a function of time and the HV state (see Figs. 3, 4 and 5). Furthermore, the FPGA-based data acquisition system was adjusted to a maximum time resolution of 20 ns (see Fig. 6), which is by a factor of 5 better than in June 2008. As already seen in previous experiment the sample responds by oscillation behaviour to the fast switch of HV. These oscillations are interpreted by piezoelectric-generated elastic waves propagating through the crystal area illuminated by the X-ray beam. As shown in Figs. 3 and 4 there are at least three different frequencies. At this, the recorded response function, Δω(t,HV(t)), was fitted by the following equation (see fitted red curves):

$$\sum_{i=1}^{3} A_i \exp(-\gamma_i t)\cos(\omega_i t - \delta_i) + a_0 . \quad (1)$$

Using this approach we could describe quite well the measured oscillation behaviour of single Bragg peak positions (see Figs. 3 and 6) that is related to the piezoelectric-induced macroscopic strains in BiB_3O_6. At the same time the HV frequency seems to be crucial for the validity of our model, compared to the measurements presented in Fig. 3 in case of Fig. 4 the HV frequency was slightly changed from 1 kHz to 975 Hz. By changing the HV frequency to 3 kHz we obtained the curves displayed in Figures 5 and 6.

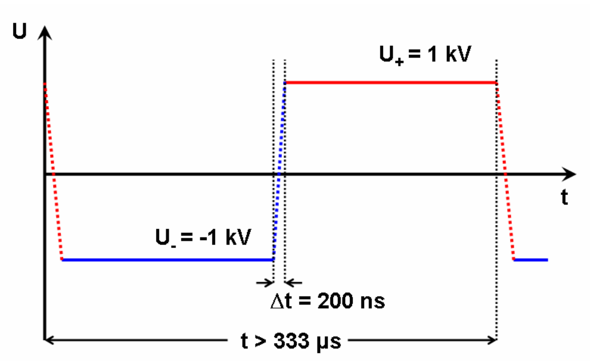


Fig. 1 Schematic view of the 2-step modulated HV, applied to single crystal plates. The both different HV states, U_- and U_+, were of the order of ±1 kV. The minimum HV period length of about 333 µs corresponds to a frequency of 3 kHz. The switching time Δt from U_- to U_+ was not greater than 200 ns.

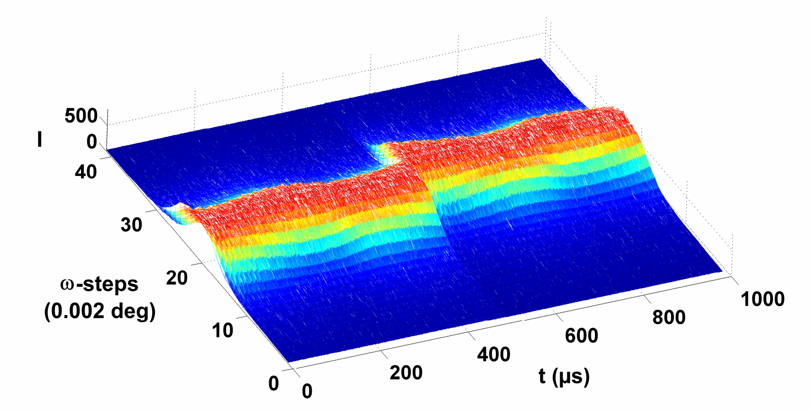


Fig. 2 Representation of the time-resolved scanning of the diffraction intensity of a (3 9 2) reflection of a (010) BiB_3O_6 crystal plate carried out with the ω-scan method. In this case the time resolution was 100 ns and due to the low counting statistics the measurements were repeated 5 times and then merged together. The width of the ω-steps was fixed to $2x10^{-3}$ deg and the frequency of the HV was 1 kHz.

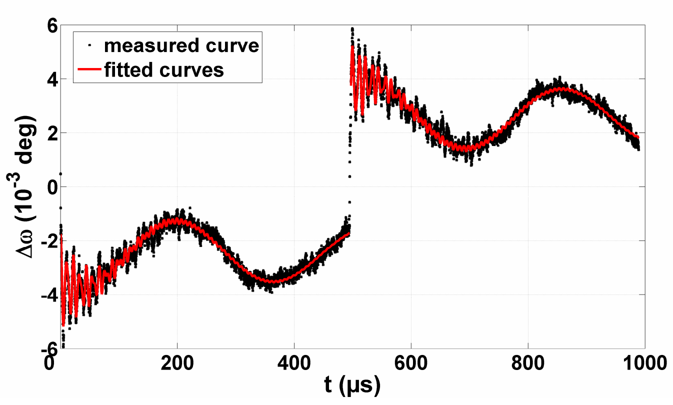


Fig. 3 Shift of the Bragg peak position, Δω, of a (3 9 2) reflection measured as a function of time and HV state (the HV frequency was 1 kHz). The both red curves were fitted to the experimental data using the approach described by equation (1).

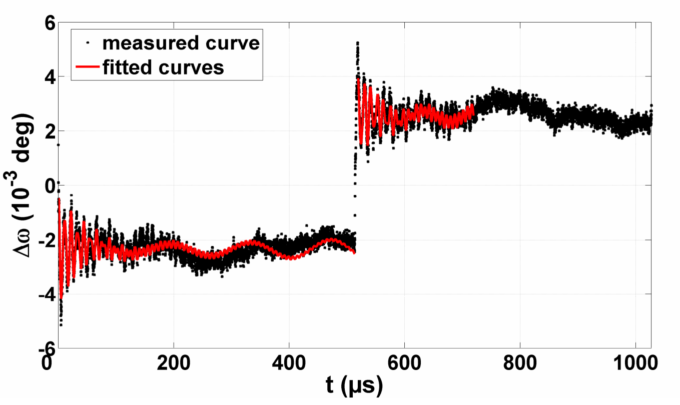
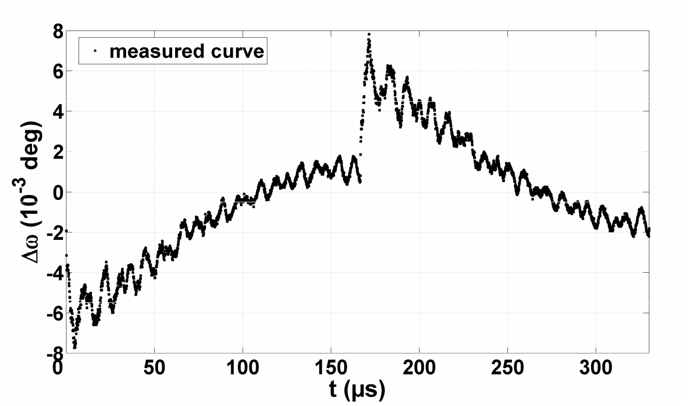


Fig. 4 Compared to the Figure 3 in the presented case the HV frequency was slightly changed to 975 Hz. As a result the measured oscillations in the shift of the Bragg peak position, Δω, do not show any more an exactly periodical time behaviour.



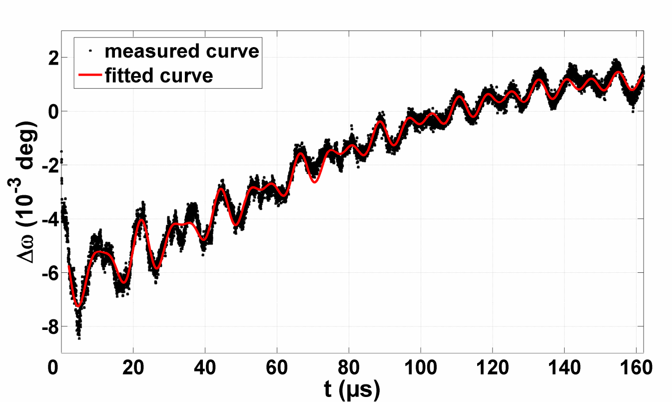


Fig. 5 In comparison with Figs. 3 and 4 this figure represents the Δω measurements of a (3 9 2) reflection performed applying the HV with a frequency of 3 kHz to the (010) BiB_3O_6 crystal plate. As a result the time dependence of the recorded response function changes dramatically.

Fig. 6 As distinguished from Fig. 5 the time behaviour of Δω in the first 62 µs of the HV period was resolved with 20 ns and not with 100 ns. Using the approach given by equation (1) one can describe quite well the measured response of BiB_3O_6 to an applied external electric perturbation.

Conclusion

Our time-resolved studies could successfully probe the response of BiB_3O_6 to an ultra-fast switch of an applied external electric field. Measuring the time dependence of the Bragg peak position of selected reflections on a time scale between 20 ns and 100 ns and using the approach described by equation (1) three different piezoelectric-induced elastic waves could be extracted propagating in the (010) BiB_3O_6 crystal plate:

$$\nu_1 = 135.7(2)\text{ kHz},\ \nu_2 = 90.4(2)\text{ kHz},\ \nu_3 \approx 3\text{ kHz}. \quad (2)$$

As shown in Figure 6, a time resolution of 20 ns gives a much better fit to the measured data. Therefore in future this time resolution will be used as a standard setting as soon as the counting statistics problems are overcome. The first both frequencies (2) are in the same range as those measured in June 2008 for $Li_2SO_4{\cdot}H_2O$ (see user report HS3355 [1]) and reported by Reuwijk (2002) [2] in case of KD_2PO_4. Considering the spatial extension of the used BiB_3O_6 crystal plate these frequencies can be associated with elastic waves that are generated at one edge of the plate and hereafter propagate throughout the crystal media until they are reflected by another crystal edge. In contrast, the value of the third frequency (2) is far too small for a propagating elastic wave. According to the performed measurements (see Figs. 3, 4 and 5) this mode of the electric-field-induced elastic waves in BiB_3O_6 is highly dependent on the frequency of the applied HV and shows resonance behaviour close to the HV frequency of 1 kHz. This observation has to be investigated in a more precise way in further experiments.

Finally, we have to note that the experimental data can also be evaluated in terms of the change of diffraction intensities and FWHMs of Bragg peaks. This data treatment is still in progress.

[1]. S. Gorfman, O. Schmidt, M. Ziolkowski, U. Pietsch. Experimental user report HS3355 (June, 2008).
[2]. Reeuwijk, S.J. van, PhD thesis, University of Twente, The Netherlands, (2002).