ESRF	**Experiment title:** Time-resolved study of the internal strains relaxation process in $Li_2SO_4 \cdot H_2O$ single crystals induced by an applied external electric field - II	**Experiment number**: HS-3355
Beamline: BM01A	**Date of experiment**: from: 23.06.2008 to: 01.07.2008	**Date of report**: 27.08.2008
Shifts: 18	**Local contact(s)**: Dr. Dmitriy Chernyshev	*Received at ESRF:*

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Report:

The physical properties of crystals are defined as their macroscopic responses to any kind of external perturbation. In particular the responses of a crystal to an applied external electric field are known as dielectric polarization and converse piezoelectric effect. Although these phenomena are highly demanded for various technical applications, their microscopic physics is not well understood yet and still extensively investigated by different experimental techniques. During the last few years we have been working on X-ray diffraction studies of atomic movements in piezoelectric crystals induced by an applied external electric field. These investigations of the microscopic crystal response to a ***static*** electric field were mostly performed at HASYLAB. (See for example [2])

In January 2008 we initiated the first experiments at ESRF, where the high flux of the beam was exploited for the ***time-resolved*** X-ray diffraction studies of the crystal response to the fast alternating external electric field. The details of the original experimental technique as well as the first results were already described in the preliminary report [1]. In the first test experiments external high voltage (HV) was applied to a 0.5 mm thick crystal plate and periodically switched between the values of $-U$, 0 and U (with $U \sim 3$ kV) with the shortest switching time of 50 μs. The corresponding structural responses were studied with the minimal time resolution of 1 μs. In July 2008 we expanded the above studies to the case where the external electric field was switched during 60 ns, in addition the time resolution was improved by the factor of 10. Developed at Siegen University the HV setup provided a periodic signal as schematically shown in Fig. 1. The modulation frequency of the applied voltage was 1-10 kHz. The rise-time of the strike pulses with the maximum amplitude of 50 kV had a width of about 100 ns. After the strike pulse the HV sloped down to a rest value of approximately 1 kV.

We used our own FPGA-based data acquisition system for the separation of incoming detector signal into successive time intervals. The width of the single time blocks (i.e. the time resolution) was fixed to 100 ns.

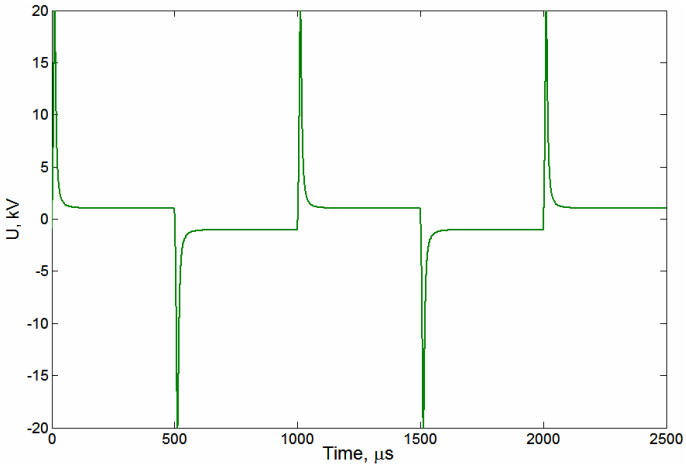

Fig. 1 Schematic view of the HV-signal, applied to a crystal. The HV-cycle consists of positive and negative voltages parts. The switches between different polarities are performed via intermediate pulse-like strikes with the short rise time (~100 ns) and high amplitude (~ 20-50 kV).

The measurements of few selected Bragg reflections were performed at $Li_2SO_4 \cdot H_2O$ single crystals in the ω-step scan mode (ω-step was 0.001 degree) using a KUMA four-circle diffractometer (BM01A@ESRF, Swiss-Norwegian beamline). The rocking curves were used to analyze the time dependence of the respective peak positions. The corresponding angular shifts of two representative reflections are shown in Figures 2 and 3.

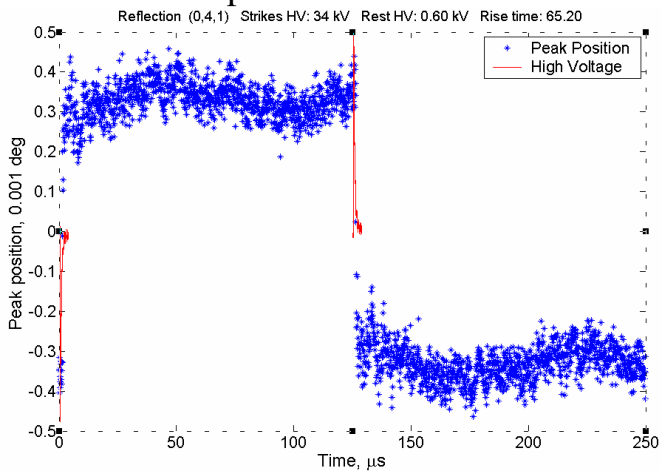

Fig. 2a Change of the *(041)* peak position during a complete high-voltage cycle. The rise-time of the HV pulses (U~ 34 kV) was ~ 65 ns. The rest voltages on the tails of the pulses are 600 V.

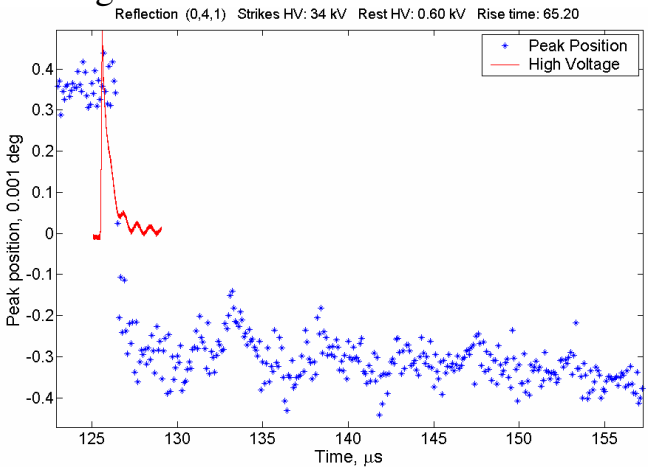

Fig. 2b Change of the peak position of the *(041)* reflection straight after the positive pulse of the high voltage has been switched. The normalized high voltage close to the switch is shown in red.

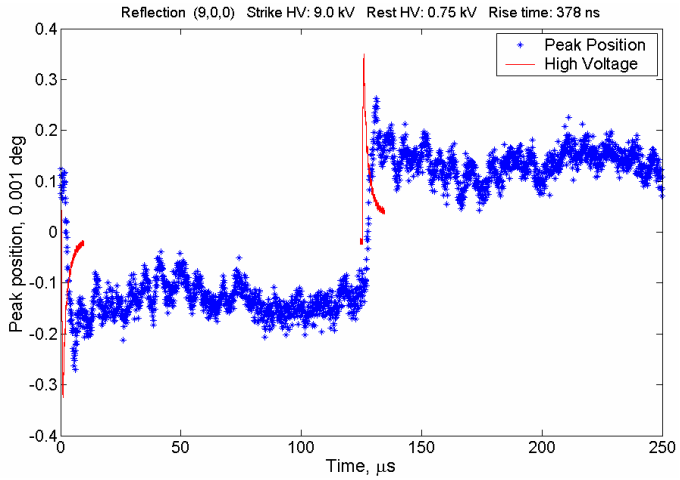

Fig. 3a Change of the *(900)* peak position during a complete HV-cycle. The rise time of the HV-pulses (U~ 9 kV) was ~ 378 ns. The rest voltages on the tails of the pulses are 750 V.

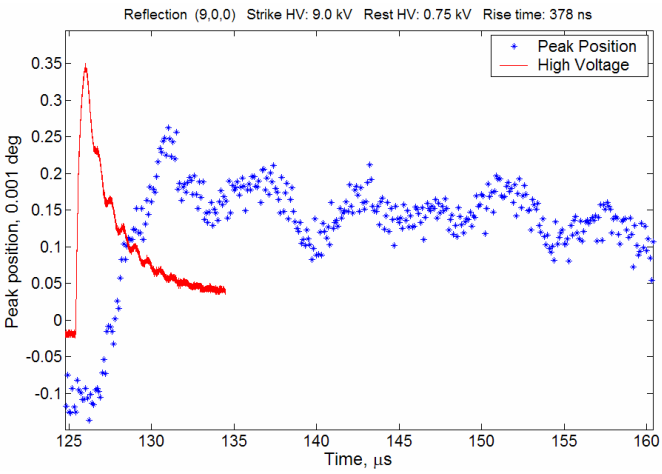

Fig. 3b Change of the peak position of the *(900)* reflection straight after the switching of a positive pulse of the high voltage. The normalized high voltage close to the switch is shown in red.

The change of a Bragg peak position is related to the magnitude of the macroscopic strains in the area illuminated by an X-ray beam. As visible from Fig. 2 and Fig. 3 the detected strain shows oscillatory behaviour and is related to an elastic wave propagating through the crystal. The measured vibration frequencies may be analyzed by means of the Fourier transform of the signal taken in the second half of the HV-cycle, where the electric field is constant. The Fourier transform of the peak position of the (900) reflections in the time interval from 130 till 250 μs is presented in Fig. 4.

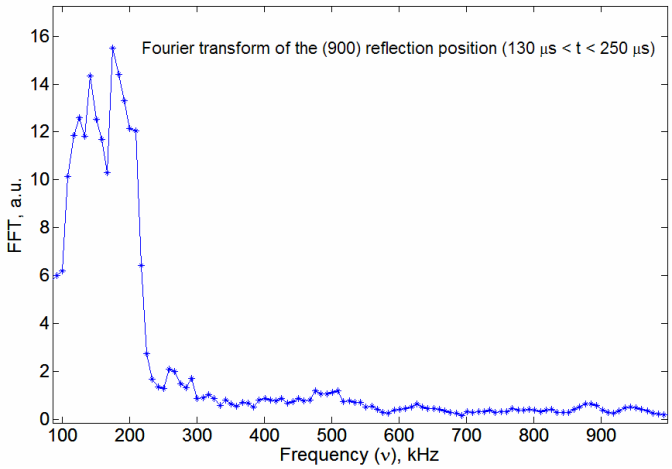

Fig 4. Fourier transform of the (900) peak position during the second half of the high voltage cycle (130 μs < t < 250 μs). The maximum frequency values span the range from 100 to 200 kHz, which corresponds well to the oszillations visible in Fig. 3b.

As follows from Fig. 4, the frequencies of the observed oscillations are mostly concentrated in the band between 100 and 200 kHz. This corresponds to an oscillation period between 5 and 10 μs.

Conclusion

We performed time-resolved X-ray diffraction studies of the piezoelectric crystal response to the alternations of an external electric field. In the first part of our work (January, 2008 see [1]) we investigated the deformations in $Li_2SO_4 \cdot H_2O$ crystals due to the slow switch of the electric field (with the rise-time of 50 to 5000 μs). As follows from the time behaviour of the several peak widths, the reorientation of the domain blocks takes place about 1000 μs after the switch of the high voltage. In the second part of our work (July, 2008) we updated the experimental equipment for the studies of the crystal response to the fast pulse-like HV switches with the rise-time of approximately 100 ns and succeeded in observing oscillations of the peak positions. The dominating frequencies were found to be ν=100 - 200 kHz (T=5-10 μs). The period of these oscillations is in quantitative agreement with the time T=2d/c an elastic wave, generated at one edge of a crystal, needs to propagate to another one and back, where d is the lateral size of the crystal (d~0.01 m) and c the sound velocities of the elastic wave (c~3800 m/s). In addition the onset of the generated strain at the X-ray probe point of the crystal is delayed by ~ 1 μs compared to the electric field switch. Considering that the X-ray beam hits approximately the centre of the crystal plate we conclude that the strain wave is created at the edges of the crystal. Note that these results are in agreement with the previous time-resolved X-ray studies of the strain waves in a crystal performed by S. V. Reeuwijk [3] by means of the much more time consuming pump-probe technique.

[1]. S. Gorfman, O. Schmidt, M. Ziolkowski, U. Pietsch. Preliminary experimental user report HS3355 (2008).
[2] S. Gorfman, V. Tsirelson, A. Pucher, W. Morgenroth, U. Pietsch. Acta Cryst A62, 1-10 (2006).

[3]. Reeuwijk, S.J. van, PhD thesis, University of Twente, The Netherlands, (2002).