

7.1 Microscopic mechanism of electrostriction

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Project description and motivation:

Electrostriction (the inverse piezo-electric effect) describes the deformation of a crystal due to an external electric field. In tensor notation this reads $\epsilon = \mathbf{d}^{-1} \mathbf{E}$, where \mathbf{d} is the piezo-electric tensor, \mathbf{E} the electric field vector and ϵ the tensor of deformation.

The macroscopic deformation of the crystal can be reduced to a microscopic deformation of the unit cell. But this does not help for the understanding of the effect because the origin of electrostriction is located on the atomic level. The electric field acts on the atomic cores and on the valence charge density. Compared to the internal field, the external one (which can reach several kVmm^{-1} , only) is a small perturbation. The atoms respond by small displacements, eventually hindered by chemical bonds. This structural change can be subdivided into an external effect, the electrostriction, and an internal distortion (within the unit cell).[1]

A theoretical description of a crystal in an external electric field is not easy. The external field leads to an additional potential and thus to a non-periodic Hamiltonian. Only recently a concept was developed to calculate the properties of a crystal in a homogeneous electric field *ab initio*. [2]

Unlike the Hamiltonian (and thus the wavefunction), the electron density and the nuclear positions are still periodic. This allows to determine the structural changes by conventional diffractive methods. The changes in the nearest neighbour distance are of the order of $10^{-5} \text{ \AA/kVmm}^{-1}$. This means that the Bragg peaks shift only slightly and their intensity varies in the order of 10^{-2} or less. So the challenge is to measure with high precision and to exclude as much external influences as possible to get reliable data to be interpreted with a model.

Earlier investigations in this field started with the semiconductors GaAs and ZnSe [3, 4] and later expanded to $\alpha\text{-SiO}_2$, GaPO_4 , KH_2PO_4 (KDP) and other ferroelectrics. Some of these were also measured at low temperatures.

For GaAs, ZnSe and SiO_2 the internal distortion could be determined and structural changes were modelled. But also problems showed up with the other materials. Some measurements could not be reproduced or instable results were obtained for the first hours of the crystal being in the external electric field. All of these measurements have been performed with x-rays or synchrotron radiation. Thus a possible explanation is that only the region close to the surface is probed and this is dominated by space charges.

Recent test measurements with KDP on TOPSI showed a stable signal from the beginning and an almost parabolic dependence of the relative intensity variation. This is in opposite to the findings of Reeuwijk et al.[5].

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- [2] F. BERNARDINI, V. FIORENTINI and D. VANDERBILT. Accurate calculation of polarization-related quantities in semiconductors. *Phys. Rev. B* **6319**, (19) 193201, 2001.
- [3] J. STAHN, A. PUCHER, U. PIETSCH, J. ZELLNER and E. WECKERT. Experimental determination of electric-field-induced differences in structure-factor phases of the order of 2%. *Acta Cryst. A* **55**, 1034–1037, 1999.
- [4] J. STAHN, U. PIETSCH, P. BLAHA and K. SCHWARZ. Electric-field-induced charge-density variations in covalently bonded binary compounds. *Phys. Rev. B* **63**, (16) 165205 (10 p.), 2001.
- [5] S.J. REEUWENIK, A. PUIG-MOLINA and GRAAFSMA H. Electric-field-induced structural changes in deuterated potassium dihydrogen phosphate. *Phys. Rev. B* **62**, 6192–6197, 2000.

Medium term goals:

Within the FP6 project *Design and improvement of complex materials for optical data storage and processing* (the proposal will be submitted beginning of April) the interaction of space charge fields with the crystal structure will be investigated. The electric fields caused by space charges are of the same order as externally applied fields. Thus the microscopic structural changes can be studied with an external field.

This will be done in co-operation with Niels Hansen, Nancy, who is responsible for the x-ray measurements. The substances we want to investigate are holographic data storage materials of the classes $\text{LiNbO}_3:\text{Fe}$ and $\text{Sr}_x\text{Ba}_{1-x}\text{Nb}_2\text{O}_6:\text{Cr}$.

Specific goals for the year 2003:

To check the comparability of x-ray and neutron measurements a series of test measurements is planned on TOPSI, using the crystals (and the high voltage equipment) already measured at the ESRF by Niels Hansen.

In addition the measurements on KDP will be repeated and continued to check the reproducibility of the observed non-linear behaviour of the relative intensity variation.

Collaborating scientists:

- Niels Hansen, Université Nancy 1
- Ullrich Pietsch, Universität Potsdam
- Theo Woike, Universität zu Köln