Investigation of the converse flexoelectric effect in KTaO₃ using the interferometric microscopy method

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Abstract. The inhomogeneous strain induced by an external electric field (the converse flexoelectric effect) has been studied in thin KTaO₃ single crystal plates by a scanning interferometric microscopy method. It has been shown that the type of inhomogeneous strain (spherical and cylindrical bending) is determined by the direction of the applied field along [001] and [011] crystallographic directions.

1. Introduction
The converse flexoelectric effect is an electrical and mechanical effect in which an inhomogeneous strain is induced by an external electric field. The flexoelectric effect is largely manifested in crystals and films of submicron and nanoscopic sizes that is important for elements of integrated electronics and micro-electromechanical systems [1]. Ferroelectrics are most promising material due to their high dielectric susceptibility favoring maximum flexoelectricity [1]. One of the important tasks in flexoelectricity is estimating of the flexoelectric coefficients and establishing their dependence on the crystalline structure, geometry of samples, thermodynamic parameters, etc. Our investigation of the converse flexoelectric effect is devoted to determine the type of inhomogeneous strain (torsion, cylindrical, and spherical bending) in model dielectric single-crystals, for example, in a cubic perovskite KTaO₃.

According [2, 3], an external field applied along [001] crystallographic direction induces the distortion of the perovskite crystal unit cell with a symmetry 4/mmm into a point group with 4mm; i.e., an initially tetrahedral (or cubic) cell is transformed and takes the form of a truncated pyramid. For an extended crystal, such geometry is expected to cause strain more complex than the cylindrical bending, i.e., spherical bending. On the other hand, it can be assumed that the external field applied along the crystallographic direction [011] with the rotational symmetry of the 2nd order for the cubic crystal cell, induces a cylindrical bending.

In the study of the converse flexoelectric effect in liquid crystals [4] and in ceramic Ba₀.₆₇Sr₀.₃₃TiO₃ samples of trapezoidal prism shapes [5], an interferometric technique has already been used, but the type of inhomogeneous strain has not been studied. This paper present the bending profiles of the KTaO₃ single crystal plates studied by scanning interferometric microscopy method.

2. Materials and method
A set of single crystal plates of KTaO₃ with a thickness of 150 ± 10 µm and an area of 10 x 10 mm² were used. The surfaces (001) and (011) of the plates were oriented by X-ray diffraction technique and polished for optical quality and were covered with gold electrodes with a diameter of 5 ÷ 6 mm by
thermal sputtering. The electrodes also served as mirrors. Triangular-shape pulses of the both polarity with amplitude up to 150 V (10 kV/cm) with period of 100 s were applied.

The induced inhomogeneous strain of thin single-crystal plates was measured by a modified interferometer microscope based on serial LOMO MII-4. The interferometer microscope was equipped with He-Ne laser ($\lambda = 633$ nm) and with a high resolution CCD camera. The interference maxima and minima had the form of concentric rings and their motion depended on the deformation of the mirror surfaces. Changes in the interference pattern were analyzed using a computer program “blink-comparator” based on a method of comparing two images alternately imposed on each other. The method made it possible to estimate the local strain up to 0.01 of the wavelength $\lambda$, i.e. 10 nm. The interferometer microscope also provided scanning the entire crystal surface along, which made it possible to directly evaluate the profile of inhomogeneous strain as a function $\delta z(x,y)$ (figure 1).

![Figure 1](image.png)

**Figure 1.** The KTaO$_3$ single-crystal plates with two different surfaces a) (001) and b) (011) and their crystallographic directions. The direction of the applied external field, the scanning directions $X$ and $Y$, and the induced inhomogeneous strain (spherical, and cylindrical bending) are shown.

3. Results

In thin KTaO$_3$ plates we observe the typical bending of the concave surface toward the positively charged electrode. Figures 2a and b show the profiles of the deformed surface (001) for scanning directions $X \parallel [100]$ and $Y \parallel [010]$ (figure 1a). Each point on the graphs corresponds to the maximum of the local strain shifting at the point on the plate surface for applied field 10 kV/cm. In the profiles, despite some scatter of the points, it can be seen that the strain increases from the periphery of the electrode to its center. The lines drawn near the experimental points make it possible to estimate the bending radii. In the case of scanning along the $X$-direction the bending radii are $55 \pm 15$ and $65 \pm 20$ m, for positive and negative polarities respectively and approximately the similar values $55 \pm 10$ and $65 \pm 10$ m are obtained for the $Y$-scanning direction. Since the radii for two mutually perpendicular directions are the same, the type of the inhomogeneous strain can be defined as a spherical bending (figure 1a). The differences between radii observed in the positive and negative polarity can be explained by the electrostriction and injection of charge carriers due to the high voltage 10 kV/cm. Our studies of the hysteresis and remanent inhomogeneous strain in SrTiO$_3$ and KTaO$_3$ due to the space charge injection will be published. The scatter of the points and the asymmetry of the profiles can be explained by the inhomogeneity of the crystal subsurface structure and the polarization gradients in it. According [1] the gradient of polarization in subsurface regions of thin films and plates mainly determines the magnitude of the converse effect.
Figures 3a and b show the profiles of the deformed surface (011) for scanning directions X || [100] and Y || [011] (figure 1b). It can be seen that the bending is observed only for scanning along the Y-direction, and the radii of curvature of the surface can be estimated 150 ± 50 and 180 ± 50 m for the both polarity. As for the X-direction, in this case no bending is detected. The obtained profiles show a cylindrical bending with the rotational symmetry of the 2nd order (figure 1b).

In the work [3] has been proposed the model of the crystal unit cells distortion of the BaTiO₃ thin films (figure 4). In barium titanate the energy of interaction between Ba²⁺ and O²⁻ ions changes due to overlap of electronic shells Ti⁴⁺ and O²⁻ ions, which causes relatively large distortion of the cell (the change in the interaction energy of barium and titanium ions is insignificant). The external field applied along [001] crystallographic direction induces the displacement of the Ti⁴⁺ ion in the oxygen octahedron and the distortion of the cell from cubic m3m (or tetragonal 4/mmm) into tetragonal 4mm point group symmetry (figure 4a). In thin plates and films the cell takes the form of a truncated pyramid with the rotational symmetry of the 4th order (figure 4b), which has to lead to a spherical bending of the entire crystal surface. The same model is applicable to KTaO₃ with the similar perovskite structure. Based on this model, we assume that the polarization of the crystal in the [011] direction causes a cell distortion, which has a rotational symmetry of the 2nd order (figure 4c). In this case, the cylindrical bending with the same kind of symmetry also shows that there is not the flexoelectric distortion in crystallographic direction [100] and the cell takes the form of a right rhombic prism.

Figure 2. Profiles of the surface (001) in the field 10 kV/cm. a) X- and b) Y- scanning directions (see figure 1). Lines for estimating the bending radius of the surface are shown. The vertical lines are electrode boundaries.

Figure 3. Profiles of the surface (011) in the field 10 kV/cm. a) X- and b) Y- scanning directions (see figure 1).
We can compare the magnitude of the converse effect in different crystals of equal geometry sizes \([2, 3, 6]\) if we use an effective 4th rank tensor, which relates the inhomogeneous strain and the electric field:

\[
\nu_{kl} = \frac{\partial (\frac{du_{kl}}{dx_i})}{\partial E_j},
\]

Where \(du_{kl}/dx_i\) – strain gradient, \(E_j\) - the electric field. According to the experimental data for thin plates with a thickness of 150 \(\mu\)m and an area 1 \(\times\) 1 cm\(^2\), the following effective tensors are estimated:

- \(\nu_{12} = 1 \times 10^{-5} \text{ V}^{-1}\) [2, 3], \(\nu_{12} = 1.5 \times 10^{-8} \text{ V}^{-1}\) [6].
- \(\nu_{12} = 3.4 \times 10^{-7} \text{ V}^{-1}\) (Here, we used the Voigt's two-suffix tensor notation).

The tensor \(\nu_{12}\) for BaTiO\(_3\) - thin plates from 20 to 500 \(\mu\)m depends on the sample thickness [3], but for SrTiO\(_3\) - plates of thickness from 150 to 300 \(\mu\)m does not [6]. The dependence of the tensor on the sample geometry requires a special investigation. In the case of geometry in figure 1b when the field is applied along \([011]\), the both tensors \(\nu_{42}\) and \(\nu_{44}\) must be taken into account, but their values require additional complex measurements.

**Figure 4.** The perovskite crystal cell: a) distortion in infinite crystal, b) the flexoelectric distortion is a truncated pyramid, and c) flexoelectric distortion is a right rhombic prism (no flexoelectric distortion in \([001]\)-direction). Arrows shows the directions of applied field and shifting of Ta\(^{5+}\) ion a), b) – along \([001]\), and c) - along \([011]\) crystallographic direction.

4. Conclusion

The inhomogeneous strain induced by a homogeneous external electric field (the converse flexoelectric effect) has been studied in a thin KTaO\(_3\) single crystal plates by the scanning interferometric microscopy method. It has been determined the type of inhomogeneous strain of spherical and cylindrical bending induced by the field applied along \([001]\) and \([011]\) crystallographic directions respectively.

**References**

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