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Anisotropy of electromechanical properties in KNbO_3 crystals with S-type domain boundaries

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Abstract. In this paper a large anisotropy of piezoelectric moduli of d_{ij}^p KNbO_3 crystals with S-type boundaries separating 60° (120°) domain regions in the ferroelectric $Bmm2$ phase is established for the first time. The sensitivity of this effect to changes in initial dielectric, piezoelectric and elastic constants of the corresponding single-domain crystal is discussed.

At present investigations of ferroelectric crystals with complex domain structures focus on crystallographic description of different domain patterns [1, 2] and physical properties [3, 4] of corresponding crystal samples. In particular, much attention is paid to the problem of determining the conditions for the formation and the stability of S-type domain boundaries [1, 5] and the prediction of physical properties important to different applications. As is known, such unusual domain boundaries [6] are characterized by orientations [1, 5] which depend on the temperature, unit-cell parameters and volume concentrations of adjacent domains, etc. For example, direct observation of S-type domain boundaries in some ferro- and antiferroelectric perovskite-type crystals was carried out by Wiesendanger [7], Chen Jun *et al* [8] (KNbO_3 , $Bmm2$ phase), Balyunis *et al* [1, 9] (PbHfO_3 , $Pba2$ and $P2221$ phases; PbZrO_3 , $Pbam$ and $R3m$ phases), Bah *et al* [5] ($\text{Pb}(\text{Zr}_{1-x}\text{Sn}_x)\text{O}_3$, $Cm2m$, $P2221$, $Pbam$ or $Pba2$ phases).

Our previous attempt to determine elastic, dielectric and piezoelectric constants of lamellar domain structures with non-collinear orientations of spontaneous polarization vectors was applied to LiNbO_3 and LiTaO_3 crystals in the ferroelectric $R3m$ phase. These polydomain crystals as mechanical twins may be characterized by the large anisotropy of the piezoelectric moduli d_{ij}^p within some temperature ranges [4]; e.g., for different tensor elements calculated in a coordinate system of principal axes of the polydomain sample, $|d_{ij}^p/d_{ik}^p| \gtrsim 10$ where $i = 1, 2, 3$; $j = 1, 2, 3$; $k = 1, 2, 3$; $j \neq k$. The problems associated with determination of electromechanical constants in low-symmetry phases of perovskite-type crystals having laminated domain structures with S-type boundaries have not yet been solved.

The present work is aimed at study of possibilities for achieving the large anisotropy of piezoelectric moduli d_{ij}^p in KNbO_3 crystals with S-type domain boundaries. Orientations of such boundaries dividing two domain regions (90° domains in each region and 60° (120°) domains in adjacent regions) in the ferroelectric $Bmm2$ phase may be determined by using results in [1] and [9]. If one takes into account four domain types with orientations of their spontaneous polarization vectors $P_{s1}(P_s; 0; P_s)$, $P_{s2}(P_s; 0; -P_s)$

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(region 1), $P_{s3}(0; -P_s; P_s)$, $P_{s4}(0; P_s; P_s)$ (region 2) and with volume concentrations t , $1 - t$ (region 1), $1 - u$, u (region 2), respectively, the orientation of the S-type boundary $n(hkl)$ between the regions 1 and 2 is described by formulae [1]

$$h = k \quad l/h = 4\eta\eta_a(2t - 1)/[\eta_a^2 + \eta_b^2 + \eta^2(2t - 1)^2] \quad (1)$$

in a coordinate system $(X_1X_2X_3)$ with axes (OX_i) parallel to axes of the perovskite unit cell. In equation (1) $\eta_a = (a \cos \omega)/a_c$, $\eta_b = b/a_c$, $\eta = (a \sin \omega)/a_c$ are the perovskite unit-cell distortions, where a , b and a_c are the unit-cell dimensions in the orthorhombic and cubic phases, respectively, while ω is the unit-cell shear angle in the orthorhombic phase. The S-type boundary dividing the regions mentioned above is a zero-net-strain plane by equality of domain volume concentrations $t = u$. In the simplest case we take into consideration two domain types (P_{s1} ; P_{s4}) with the S-type boundary and $t = u = 1$ as the basic elements of the lamellar domain structure occupying the crystal sample as a whole. An averaging procedure for determination of physical constants of the crystal having the domains with volume concentrations m (the first type, P_{s1}) and $1 - m$ (the second type, P_{s4}) is realized by using the method developed in [10, 11]. It is assumed that the volume concentration m of the first domain type in this crystal sample is varied† within the range of $0 < m < 1$. The equations corresponding to boundary conditions for electric and mechanical fields of these domains are derived from relations [12] between electromechanical constants of the separate domain type

$$\begin{pmatrix} \|\varepsilon^\sigma\| & \|d\| \\ \|d\|^T & \|S^E\| \end{pmatrix} \begin{pmatrix} E \\ \sigma \end{pmatrix} = \begin{pmatrix} D \\ \xi \end{pmatrix} \quad (2)$$

by taking into account the orientations of domain crystallographic axes. In equation (2) $\|\varepsilon^\sigma\|$, $\|d\|$ and $\|S^E\|$ are matrices of dielectric permittivities, piezoelectric moduli and elastic compliances of the single-domain crystal; $\|d\|^T$ is the transposed matrix $\|d\|$. The columns $(E; \sigma)$ and $(D; \xi)$ contain the components of the vector of the electric field strength $E(E_1; E_2; E_3)$, and the tensor of the mechanical stress field $\sigma(\sigma_1; \sigma_2; \dots; \sigma_6)$ and the components of the vector of the electric induction $D(D_1; D_2; D_3)$, and the tensor of the mechanical strain field $\xi(\xi_1; \xi_2; \dots; \xi_6)$, respectively. Finally, the averaged electromechanical constants are transformed to the coordinate system $(X_1^p X_2^p X_3^p)$ of the polydomain crystal with the axis OX_3^p being parallel to the vector of the averaged spontaneous polarization $P_{s,\Sigma} = mP_{s1} + (1 - m)P_{s4}$.

For our calculations one can use two known sets of initial electromechanical constants d_{ij} , ε_{kk}^σ and S_{im}^E of single-domain orthorhombic KNbO_3 crystals. These constants are taken from experimental data of Wiesendanger [13] and Zgonik *et al* [14] for temperatures $T = 25^\circ\text{C}$ and $T = 22^\circ\text{C}$, respectively. Both sets are characterized by an inessential anisotropy of piezoelectric moduli d_{ij} with $i, j = 1, 2, 3$ despite significant differences between the values of these and other (shear, $j > 3$) piezoelectric moduli (e.g., as follows from data [13], $d_{33}/d_{31} \approx 1.8$ and $|d_{33}/d_{32}| \approx 1.1$ while $d_{15}/d_{33} \approx 11$; according to data [14], $d_{33}/d_{31} \approx 3.1$ and $|d_{15}/d_{32}| \approx 1.6$ in comparison with $d_{15}/d_{33} \approx 6.8$). The averaging procedure carried out for determination of corresponding constants of polydomain crystals with S-type domain boundaries allows one to obtain other ratios between d_{ij}^p with $i, j = 1, 2, 3$. The results of our calculations for d_{ij}^p ($i, j = 1, 2, 3$) belonging to the coordinate system $(X_1^p X_2^p X_3^p)$ are shown in figure 1. The concentration dependences d_{ij}^p pass through zero values for different domain volume concentrations in many cases, which leads to

† Note that the possibility for variations of the volume concentration m belonging to the 60° (120°) domain types in the whole crystal sample is independent on the volume concentrations $u = p$ mentioned above and related to the 90° domains in the small regions 1 and 2 only.

the considerable enlargement of the piezoelectric anisotropy $|d_{ij}^p/d_{ik}^p|$ ($j \neq k$) of such polydomain crystals.

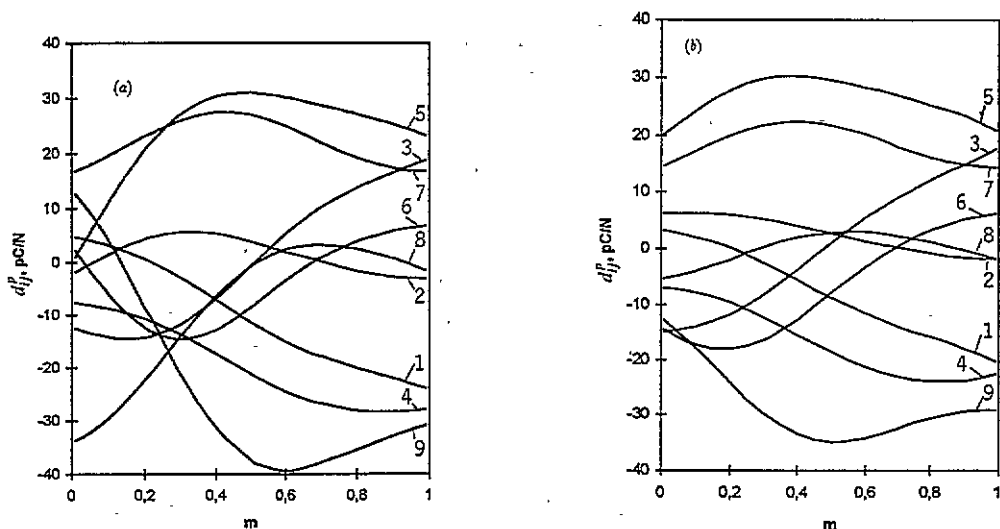


Figure 1. Concentration dependences of averaged piezoelectric moduli d_{ij}^p of polydomain KNbO_3 crystals with S-type boundaries in the $Bmm2$ phase: (a) calculated by using experimental data [13] for $T = 25^\circ\text{C}$; (b) calculated by using experimental data [14] for $T = 22^\circ\text{C}$. The curves 1–9 correspond to the following indices ij of calculated piezoelectric moduli d_{ij}^p : 1: $ij = 11$, 2: $ij = 12$, 3: $ij = 13$, 4: $ij = 21$, 5: $ij = 22$, 6: $ij = 23$, 7: $ij = 31$, 8: $ij = 32$ and 9: $ij = 33$. The volume concentration of the first domain type is varied within the range $0.01 \leq m \leq 0.99$.

The principal discrepancies in the behaviour of several curves (compare, e.g., curves 2, 5, 6, 8, and 9 in both parts of figure 1) are associated with different values of initial piezoelectric moduli d_{ij} (especially d_{33} and d_{24}) and other constants. It should be also noted that dielectric permittivities ε_{ii}^σ of the single-domain crystal are varied within a considerable range, which leads to the anisotropy [14] $\varepsilon_{11}^\sigma/\varepsilon_{33}^\sigma \approx 1.4$ and $\varepsilon_{22}^\sigma/\varepsilon_{23}^\sigma \approx 23$. In previous cases of PbTiO_3 [15], $(\text{Pb}_x\text{Ca}_{1-x})\text{TiO}_3$ [3], LiNbO_3 , and LiTaO_3 [4] single-domain crystals we accentuated only the small anisotropy of ε_{ii}^σ , i.e. $\varepsilon_{11}^\sigma/\varepsilon_{33}^\sigma \sim 1$ ($\varepsilon_{22}^\sigma = \varepsilon_{11}^\sigma$), but it is also characteristic of the phases with other (tetragonal or rhombohedral) symmetry. In the present case of the orthorhombic KNbO_3 single-domain crystal we see significant contrasts in ε_{ii}^σ values that finally may also influence the anisotropy of d_{ij}^p because of relations [12, 13] for single-crystal piezoelectric moduli d_{ij} written in the general form as $d \sim Q P_s \varepsilon^\sigma$, where Q and ε^σ correspond to matrix elements of electrostrictive constants Q_{ij} and ε_{ii}^σ , respectively, and where P_s is the spontaneous polarization of the separate domain. The electrostrictive constants Q_{ij} determined by using a connection [12] for the unit-cell spontaneous strain $\xi^s \sim Q P_s^2$ may have some influence on d_{ij} values. The slight differences in the initial elastic constants S_m^E taken from [13] and [14] cannot be responsible for changes in dependences from $d_{ij}^p(m)$ figure 1(a) to figure 1(b).

Different intervals of $d_{ij}^p(m)$ dependences (figure 1) contain interesting information on the piezoelectric anisotropy and its variation due to changes in the domain structure. As follows from figure 1(a), significant ratios of $|d_{21}^p/d_{22}^p|$ and $|d_{21}^p/d_{23}^p|$ and for $0 < m < 0.05$, $|d_{13}^p/d_{11}^p|$ and $|d_{13}^p/d_{12}^p|$ for $0.1 < m \lesssim 0.2$, $|d_{11}^p/d_{12}^p|$ and $|d_{11}^p/d_{13}^p|$ for $0.5 < m < 0.7$ are possible. The data from figure 1(b) testify to analogous possibilities for several piezoelectric

moduli $d_{ij}^p \rightarrow 0$ for the limited anisotropy of other moduli (d_{ik}^p, d_{il}^p). For example, $d_{13}^p \rightarrow 0$ and $|d_{11}^p/d_{12}^p| \approx 3.1$ for $m \approx 0.5$, $d_{12}^p \rightarrow 0$ and $|d_{11}^p/d_{13}^p| \approx 1.5$ for $m \approx 0.7$, $d_{32}^p \rightarrow 0$ and $|d_{31}^p/d_{33}^p| \approx 0.5$ for $m \approx 0.9$. It is important to note that not all the values of the domain volume concentration m may be regarded as optimal (m_{opt}), i.e. corresponding to the equilibrium 60° (120°) domain structure arising at the structural first-order phase transition $P4mm \rightarrow Bmm2$. The values of m_{opt} depend on the volume concentration of 90° domains which form at the structural first-order phase transition $Pm3m \rightarrow P4mm$. If one uses the corresponding calculations [16, 17] for domain structures in KNbO_3 crystals, one can conclude that the concentrations $m_{opt} \approx 0.75$ and $m_{opt} \approx 0.25$ are preferable because of fulfilment of conditions for effective internal stress relief at both the phase transitions mentioned above. The points of m_{opt} determined are situated near the concentration ranges (figure 1) where the large anisotropy of $|d_{ij}^p/d_{ik}^p|$ is possible. Of course, the results shown in figure 1 may be modified to take into account a possible presence of 90° domains in regions 1 and 2 (see text before equation (1)) as well as incomplete stress relief at the S-type domain boundary.

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