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Orientation relationships between electromechanical properties of monoclinic $0.91Pb(Zn_{1/3}Nb_{2/3})O_3$–$0.09PbTiO_3$ single crystals

V.Yu. Topolov [a,b,*]

[a] *Forschungszentrum Karlsruhe GmbH in der Helmholtz-Gemeinschaft, Institut für Materialforschung II, Postfach 3640, D-76021 Karlsruhe, Germany*
[b] *Department of Physics, Rostov State University, Zorge St. 5, 344090 Rostov-on-Don, Russia*

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Abstract

The relaxor-based ferroelectric $0.91Pb(Zn_{1/3}Nb_{2/3})O_3$–$0.09PbTiO_3$ single crystal provides remarkable piezoelectric and dielectric properties in the monoclinic phase near the morphotropic phase boundary that is important for various actuator and sensor applications. This paper first describes orientation dependencies of piezoelectric coefficients d_{3j} and g_{3j} ($j = 1, 2, 3$) and dielectric permittivity ε_{33}^T which have been calculated and analyzed for the single-domain monoclinic crystal sample. Results from calculations of these parameters at arbitrary orientations of the single-domain sample have been first systematized on the basis of a group of criteria and compared with known experimental data. The criteria introduced in this work enable to give the clear description of periodical and other changes in the piezoelectric and dielectric properties of the low-symmetry single-domain sample in terms of Euler angles. Some features of the dielectric anisotropy and high piezoelectric activity in the single-domain state have been considered. It has been shown that the dielectric anisotropy influences the piezoelectric sensitivity of the studied single crystal at various orientations.

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1. Introduction

Perovskite-type relaxor-ferroelectric solid solutions of $(1-x)Pb(Zn_{1/3}Nb_{2/3})O_3$–$xPbTiO_3$ (PZN–xPT) and $(1-x)Pb(Mg_{1/3}Nb_{2/3})O_3$–$xPbTiO_3$ (PMN–xPT) are characterized by extra high dielectric and piezoelectric constants, electromechanical coupling factors [1–6] as well as various domain/heterophase structures near the morphotropic phase boundary [7–12]. These prominent physical properties of the above-mentioned solid solutions and their non-linear behaviour under electric or mechanical loading are important for modern smart-materials applications, e.g., as elements of electromechanical transducers, piezoelectric actuators, medical ultrasonic imaging, hydrophones, highly effective components of piezoactive composites [13,14], etc. Recently, in both the PZN–xPT and PMN–xPT systems close to the morphotropic phase boundary, intermediate ferroelectric phases were revealed and described (see, e.g., [15–17]). These phases were identified as monoclinic [15–20] or orthorhombic [21,22] in single crystals (SCs) prepared under different conditions, after poling, thermal or mechanical treatments of samples, etc.

Despite a considerable interest in the study of the intermediate phases, heterophase states and piezoelectric response of PZN–xPT and PMN–xPT SC cuts poled in certain directions [4,5,23], there is very limited data on electromechanical (i.e., piezoelectric, elastic and dielectric) constants of these single-domain SCs. The first paper [24] dealing with the complete set of the electromechanical constants in the single-domain state is related to the rhombohedral (3 m) ferroelectric phase of PMN–0.33PT. In recent works authors [25,26] considered

* Tel.: +7 863 2975127; fax: +7 863 2434044.
E-mail address: topolov@phys.rsu.ru.

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a few examples of orientation dependencies of piezoelectric coefficients d_{3j} in the 3 m phase. Recently, Dammak et al. [6] reported a series of tensor components of dielectric, elastic and piezoelectric constants measured on single-domain monoclinic PZN–0.09PT $(\bar{1}\,0\,1)_c$ cuts where the Miller indices of the plane are written with respect to the perovskite unit-cell axes. Based on these electromechanical constants, piezoelectric coefficients d_{33} and d_{31} were calculated [6] for the $[0\,0\,1]_c$ oriented PZN–0.09PT SC. The aim of the present paper is to describe all the orientation dependencies of the piezoelectric coefficients d_{3j} and g_{3j} ($j = 1, 2, 3$) and dielectric permittivity ε_{33}^T of the stress-free single-domain monoclinic PZN–0.09PT SC.

2. Orientation dependencies of piezoelectric and dielectric properties in the single-domain state

It is assumed that the spontaneous polarization vector $\mathbf{P}_s$ in the single-domain state is oriented along the $[\bar{1}\,0\,1]_c$ direction and interconnections between the perovskite and principal (monoclinic or orthorhombic) unit-cell axes are determined in accordance with results from [6]. The coordinate axes of a rectangular system $(X_1X_2X_3)$ are parallel to the unit-cell vectors $(\mathbf{a}_m, \mathbf{b}_m, \mathbf{c}_m)$ of the single-domain SC so that $\mathbf{a}_m||OX_1||[1\,0\,1]_c$, $\mathbf{b}_m||OX_2||[0\,1\,0]_c$ and $\mathbf{c}_m||OX_3||[\bar{1}\,0\,1]_c$ by neglecting the very small monoclinic distortion.[1] The arbitrary SC orientation caused by a transformation of the coordinate axes $(X_1X_2X_3) \rightarrow (X'_1X'_2X'_3)$ is described in terms of Euler angles φ, ψ and θ (Fig. 1). Dielectric permittivities and piezoelectric coefficients of this SC are written in the coordinate system $(X'_1X'_2X'_3)$ as components of tensors of second and third ranks, respectively, as follows:

$$\varepsilon'^{T}_{mn} = \alpha_{mp}\alpha_{nq}\varepsilon^T_{pq} \quad \text{and} \quad d'_{efg} = \alpha_{ej}\alpha_{fk}\alpha_{gl}d_{jkl}. \tag{1}$$

In Eq. (1), α_{mp} are elements of the rotation matrix [28]

$$||\alpha|| = \begin{pmatrix} \cos\psi\cos\varphi - \sin\psi\cos\theta\sin\varphi & \cos\psi\sin\varphi + \sin\psi\cos\theta\cos\varphi & \sin\psi\sin\theta \\ -\sin\psi\cos\varphi - \cos\psi\cos\theta\sin\varphi & -\sin\psi\sin\varphi + \cos\psi\cos\theta\cos\varphi & \cos\psi\sin\theta \\ \sin\theta\sin\varphi & -\sin\theta\cos\varphi & \cos\theta \end{pmatrix}$$

written in terms of the above-mentioned Euler angles. After this transformation, we use the conventional two-index form [27,28] of notation of the electromechanical constants. The piezoelectric coefficients g'_{3j} are derived from a matrix equation [27]

$$||g'|| = ||\varepsilon'^{T}||^{-1}||d'|| \tag{2}$$

and calculated for comparison.

Our calculations are carried out by using the experimental values of dielectric and piezoelectric constants [6] determined by Dammak et al. (Table 1). Paper [6] contains the incomplete set of elastic constants for the same single-domain state of PZN–0.09PT SCs, and this circumstance enables us to analyze the orientation dependencies of two groups of the piezoelectric coefficients (see formulas (1) and (2)) as well as the orientation behaviour of dielectric permittivity only.

The orientation $d'_{3j}(\varphi, \psi, \theta)$, $g'_{3j}(\varphi, \psi, \theta)$ and $\varepsilon'^{T}_{33}(\varphi, \psi, \theta)$ dependencies being considered in this paper characterize piezoelectric activity, piezoelectric sensitivity and dielectric response, respectively. All these parameters are connected with the poling axis and calculated starting from the principal crystallographic axes of the single-domain SC (i.e., from Euler angles $\varphi = \psi = \theta = 0$, see Fig. 1). Main features of the calculated orientation dependencies of piezoelectric coefficients $d'_{3j}(\varphi, \psi, \theta)$, $g'_{3j}(\varphi, \psi, \theta)$ and dielectric permittivity $\varepsilon'^{T}_{33}(\varphi,$

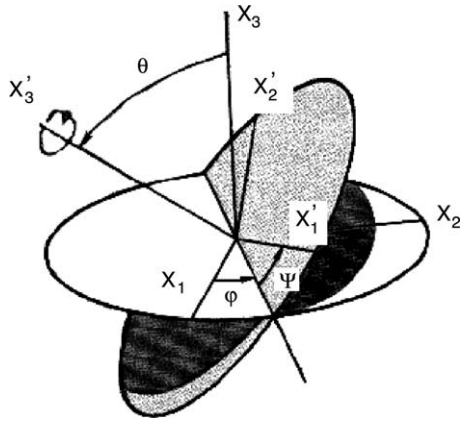

Fig. 1. Schematic representation of the rectangular coordinate systems $(X_1X_2X_3)$ and $(X'_1X'_2X'_3)$. φ, ψ and θ are Euler angles [28].

Table 1
Tensor components of dielectric permittivity ε_{ff}^T and piezoelectric coefficients d_{ab} which were measured [6] on single-domain monoclinic $0.91Pb(Zn_{1/3}Nb_{2/3})O_3$–$0.09PbTiO_3$ single crystals

$\varepsilon_{11}^T/\varepsilon_0$	9000
$\varepsilon_{22}^T/\varepsilon_0$	21000
$\varepsilon_{33}^T/\varepsilon_0$	800
d_{31} (pC/N)	120
d_{32} (pC/N)	−270
d_{33} (pC/N)	250
d_{24} (pC/N)	950
d_{15} (pC/N)	3200

Note: The PZN–0.09PT SCs were grown by the flux method [19], cut and polished with the orientation [1 0 1]/[0 1 0]/[1 0 1] related to the perovskite axes. The single-domain state with the monoclinic distortion of the unit cell was attained by poling the samples at room temperature. The tensor components of dielectric permittivity ε_{ff}^T were measured with the impedance analyzer at frequency 10 kHz, the piezoelectric coefficients d_{ab} were derived from the IRE method using the impedance spectra near resonance where the resonance frequencies were 100 kHz < f_r < 700 kHz.

[1] Using the same approximation for the single-domain state of PZN–0.09PT SC, Dammak et al. [6] introduced the matrices of dielectric, piezoelectric and elastic constants that have forms [27] similar to those in the orthorhombic mm2 phase of the ferroelectric single-domain $KNbO_3$ SC.