NON-CONVENTIONAL METHODS OF POLARIZATION SWITCHING IN UNIAXIAL FERROELECTRICS

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Thermodynamic potentials of pure uniaxial ferroelectrics, namely, triglycine sulphate and lead germanate, have been considered taking into account their secondary ferroic properties. It has been shown that the polarization switching in those crystals can occur under the action of definite combinations of external mechanical stresses and electrical fields, which may be applied normally to the ferroelectric axis.

Uniaxial ferroelectrics involve a plenty of crystals where spontaneous polarization can be oriented along only one of the crystallographic directions, and the polarization switching occurs under the action of an external electric field applied along this direction. It is this class of crystals that includes triglycine sulphate $(NH_3CH_2COOH)_3{\times}H_2SO_4$ (TGS) and lead germanate $Pb_5Ge_3O_{11}$ (LG). According to Aizu's classification [1,2], all ferroelectrics are primary ferroics. However, the theoretical and experimental researches carried out by us in works [3—8] have allowed a conclusion to be drawn that all ferroelectrics should be simultaneously secondary ferroics as well, i.e. crystals where the polarization switching may occur under the action of certain combinations of an electric field and a mechanical stress, or under the action of two combined mechanical stresses. The fact that the spontaneous polarization in TGS and LG ferroelectrics can be oriented along only one direction, together with the absence of ferroelastic properties of those crystals at temperatures above and below the phase transition point, makes them very convenient objects for studying secondary ferroic properties. Moreover, the $\bar{6} \to 3$ phase transition in an LG crystal is accompanied by the emerging of spontaneous components not only in the tensor of piezoelectric moduli but also in the tensor of elastic constants, contrary to the $2/m \to 2$ phase transition in a TGS crystal, which is characterized by the emergence of only new components in the tensor of piezoelectric moduli. That is why a ferroelectric LG crystal is simultaneously both a ferroelastoelectric and a ferrobielastic, while an TGS one is only a ferroelastoelectric [6–8].

Consider the $2/m \to 2$ phase transition in a TGS crystal. Provided that there are external electric fields and external mechanical stresses, the Gibbs elastic energy F for this proper ferroelectric transition can be written down in the form

$$F = F_0 + \frac{\alpha}{2}D_2^2 + \frac{\beta}{4}D_2^4 + \frac{\gamma}{2}D_1^2 + \frac{\mu}{2}D_3^2 - \frac{1}{2}s^D_{1111}\sigma_{11}^2 -$$
$$-s^D_{1122}\sigma_{11}\sigma_{22} - s^D_{1133}\sigma_{11}\sigma_{33} - 2s^D_{1113}\sigma_{11}\sigma_{13} -$$
$$-\frac{1}{2}s^D_{2222}\sigma_{22}^2 - s^D_{2233}\sigma_{22}\sigma_{33} - 2s^D_{2213}\sigma_{22}\sigma_{13} -$$
$$-\frac{1}{2}s^D_{3333}\sigma_{33}^2 - 2s^D_{3313}\sigma_{33}\sigma_{13} - 2s^D_{2323}\sigma_{23}^2 -$$
$$-4s^D_{2312}\sigma_{23}\sigma_{12} - 2s^D_{1313}\sigma_{13}^2 - 2s^D_{1212}\sigma_{12}^2 +$$
$$+q_{2211}D_2^2\sigma_{11} + q_{2222}D_2^2\sigma_{22} + q_{2233}D_2^2\sigma_{33} +$$
$$+2q_{2213}D_2^2\sigma_{13} + 4q_{2323}D_2D_3\sigma_{23} + 4q_{2312}D_2D_3\sigma_{12} +$$
$$+4q_{1223}D_1D_2\sigma_{23} + 4q_{1212}D_1D_2\sigma_{12} +$$
$$+q_{1111}D_1^2\sigma_{11} + q_{1122}D_1^2\sigma_{22} + q_{1133}D_1^2\sigma_{33} +$$
$$+2q_{1113}D_1^2\sigma_{13} + q_{3311}D_3^2\sigma_{11} + q_{3322}D_3^2\sigma_{22} +$$
$$+q_{3333}D_3^2\sigma_{33} + 2q_{3313}D_3^2\sigma_{13} + 2q_{1311}D_1D_3\sigma_{11} +$$
$$+2q_{1322}D_1D_3\sigma_{22} + 2q_{1333}D_1D_3\sigma_{33} +$$

$$+4q_{1313}D_1D_3\sigma_{13} + \cdots \quad (1)$$

where α, β, γ, and μ are certain coefficients, D_i the electrostatic induction components, σ_{kl} the mechanical stresses, s^D_{ijkl} the elastic constants measured at a constant induction, and q_{ijkl} the electrostriction constants.

The center of symmetry in a ferroelectric TGS crystal disappears at the phase transition, and the crystal becomes piezoelectric. Although expansion (1) remains valid at that, the induction D_2 stands now for the sum of the spontaneous induction D_{2S} and induction D_{2I} that arises owing to the application of an external field. With respect to this circumstance, the relation

$$u_{11} = \left(\frac{\partial F}{\partial \sigma_{11}}\right) = q_{2211}(D_{2S} + D_{2I})^2 =$$

$$= q_{2211}D^2_{2S} + 2q_{2211}D_{2S}D_{2I} + q_{2211}D^2_{2I}, \quad (2)$$

where u_{11} is a deformation, can be written down for a mechanically free crystal. Thus, it follows from Eq. (2) that the piezoelectric coefficient g_{211} equals

$$g_{211} = 2q_{2211}D_{2S}. \quad (3)$$

In the same manner, one can find the other piezoelectric coefficients:

$$g_{222} = 2q_{2222}D_{2S}, \quad (4)$$

$$g_{233} = 2q_{2233}D_{2S}, \quad (5)$$

$$g_{213} = 2q_{2213}D_{2S}. \quad (6)$$

Taking into account that $q_{1223} = q_{2123}$, one can also write down the equation

$$u_{23} = \left(\frac{\partial F}{\partial \sigma_{23}}\right) =$$

$$= q_{1223}D_1(D_{2S} + D_{2I}) + q_{2123}(D_{2S} + D_{2I})D_1 =$$

$$= 2q_{1223}D_{2S}D_1 + 2q_{1223}D_1D_{2I} \quad (7)$$

From Eq. (7), it follows that

$$g_{123} = 2q_{1223}D_{2S}. \quad (8)$$

In the same manner, we obtain

$$g_{112} = 2q_{1212}D_{2S}, \quad (9)$$

$$g_{323} = 2q_{2323}D_{2S}, \quad (10)$$

$$g_{312} = 2q_{2312}D_{2S}. \quad (11)$$

Therefore, the piezoeffect in the ferroelectric phase can be regarded as the electrostriction linearized by the spontaneous induction D_{2S}.

Let a mechanical stress σ_{12} and an electric field E_3 be applied to the crystal. Then, the induction

$$D_3 = \frac{1}{\mu}E_3 \quad (12)$$

arises. Using the relation

$$\left(\frac{\partial F}{\partial D_2}\right) = E_2, \quad (13)$$

and taking into account that $E_2 = 0$, we obtain

$$\alpha D_2 + \beta D^3_2 = -4q_{2312}D_3\sigma_{12}, \quad (14)$$

$$\alpha D_2 + \beta D^3_2 = -4q_{2312}\frac{E_3}{\mu}\sigma_{12}, \quad (15)$$

or

$$\alpha' P_2 + \beta' P^3_2 = -4q_{2312}\frac{E_3\sigma_{12}}{\mu}. \quad (16)$$

Thus, a nonlinear dependence of P_2 on $E_3\sigma_{12}$ takes place (its plot should look like a hysteresis loop). Analogously, one can obtain the relations

$$\alpha' P_2 + \beta' P^3_2 = -4q_{2323}\frac{E_3\sigma_{23}}{\mu}, \quad (17)$$

$$\alpha' P_2 + \beta' P^3_2 = -4q_{1212}\frac{E_1\sigma_{12}}{\gamma}, \quad (18)$$

$$\alpha' P_2 + \beta' P^3_2 = -4q_{1223}\frac{E_1\sigma_{23}}{\gamma}. \quad (19)$$

Generalizing the dependences quoted above, we can say that the sign of spontaneous polarization in a TGS crystal can be changed by applying to it not only the electric field E_2 but also the following combinations of the electric field and the mechanical stress: $E_1\sigma_{23}$, $E_1\sigma_{12}$, $E_3\sigma_{23}$, and $E_3\sigma_{12}$, where the electric field is directed perpendicularly to the spontaneous polarization direction (ferroelastoelectric polarization switching) [6]. It should be noted that the polarization switching in a TGS crystal can be induced also by applying the $E_2\sigma_{11}$, $E_2\sigma_{22}$, $E_2\sigma_{33}$, and $E_2\sigma_{13}$ combinations of the electric field and the mechanical stress, but in those cases, taking into account the values of coercive fields for a ferroelectric and a ferroelastoelectric, it will occur practically due to the presence of the electric field E_2 only.

Let us write down the difference between the thermodynamic potentials (Gibbs functions) for two kinds of domains that arise during the phase transition in the TGS crystal, confining the relevant expansions into series by those terms that describe secondary ferroic properties and taking into account the form of the tensor of spontaneous piezoelectric coefficients [3—6]:

$$\Delta\Phi = -2P_{(S)2}E_2 - 4d_{123}E_1\sigma_{23} - 4d_{112}E_1\sigma_{12} -$$
$$-2d_{211}E_2\sigma_{11} - 2d_{222}E_2\sigma_{22} - 2d_{233}E_2\sigma_{33} -$$
$$-4d_{213}E_2\sigma_{13} - 4d_{323}E_3\sigma_{23} - 4d_{312}E_3\sigma_{12}..., \quad (20)$$

where P_i are the components of the polarization vector and d_{ikl} are the piezoelectric coefficients. The subscript S denotes a spontaneous effect.

Therefore, if the TGS crystal undergoes the action of the electric field E_2 or one of the following combinations of the electric field and the mechanical stress $E_1\sigma_{23}$, $E_1\sigma_{12}$, $E_2\sigma_{11}$, $E_2\sigma_{22}$, $E_2\sigma_{33}$, $E_2\sigma_{13}$, $E_3\sigma_{23}$, or $E_3\sigma_{12}$, there emerges an energy difference between domains, and the crystal will passes to a state with minimal energy.

A procedure that was connected to piezoeffect measurements by the static method, as well as the result of experimental verifications of theoretical conclusions, was described in work [6]. The experiments showed that the application of the combination $E_3\sigma_{12}$ of the electric field and the mechanical stress, where the electric field was directed perpendicularly to the polar axis, to a monodomain TGS crystal resulted in a change of the sign of the piezoelectric response, i.e. in a change of the spontaneous polarization direction, which can be explained by the availability of ferroelastoelectric properties of a TGS crystal. In this case, the electric field strength was 4×10^5 V/m, and the mechanical stress was 1.5×10^7 Pa. Various specimens had being undergone such a combined action of the electric field and the mechanical stress for 30—90 min. When the electric field and mechanical stress, each possessing the same corresponding amplitude, had been applied to the same specimen for the same time interval but separately, there was no change in the sign of the piezoelectric response.

For the proper ferroelectric phase transition $\bar{6} \to 3$ which occurs in lead germanate, the elastic Gibbs energy can be written down as

$$F = F_0 + \frac{\alpha}{2}D_3^2 + \frac{\beta}{4}D_3^4 + \frac{\gamma}{2}(D_1^2 + D_2^2) - \frac{1}{2}s^D_{1111}\sigma_{11}^2 -$$
$$-s^D_{1122}\sigma_{11}\sigma_{22} - s^D_{1133}\sigma_{11}\sigma_{33} - \frac{1}{2}s^D_{1111}\sigma_{22}^2 -$$
$$-s^D_{1133}\sigma_{22}\sigma_{33} - \frac{1}{2}s^D_{1111}\sigma_{33}^2 - 2s^D_{2323}\sigma_{23}^2 -$$
$$-2s^D_{2323}\sigma_{13}^2 - 2s^D_{1212}\sigma_{12}^2 + g_{111}D_1\sigma_{11} -$$
$$-g_{111}D_1\sigma_{22} - 2g_{222}D_1\sigma_{12} - g_{222}D_2\sigma_{11} +$$
$$+g_{222}D_2\sigma_{22} - 2g_{111}D_2\sigma_{12} + q_{3311}D_3^2\sigma_{11} +$$
$$+q_{3311}D_3^2\sigma_{22} + q_{3333}D_3^2\sigma_{33} + 4q_{2323}D_2D_3\sigma_{23} -$$
$$-4q_{2313}D_2D_3\sigma_{13} + 4q_{2313}D_1D_3\sigma_{23} +$$
$$+4q_{2323}D_1D_3\sigma_{13} + q_{1111}D_1^2\sigma_{11} +$$
$$+q_{1122}D_1^2\sigma_{22} + q_{1133}D_1^2\sigma_{33} +$$
$$+2q_{1112}D_1^2\sigma_{12} + q_{1122}D_2^2\sigma_{11} + q_{1111}D_2^2\sigma_{22} +$$
$$+q_{1133}D_2^2\sigma_{33} - 2q_{1112}D_2^2\sigma_{12} - 2q_{1112}D_1D_2\sigma_{11} +$$
$$+2q_{1112}D_1D_2\sigma_{22} + 4q_{1212}D_1D_2\sigma_{12} +$$
$$+4\vartheta_{31123}D_3\sigma_{11}\sigma_{23} - 4\vartheta_{32213}D_3\sigma_{11}\sigma_{13} -$$
$$-4\vartheta_{31123}D_3\sigma_{22}\sigma_{23} + 4\vartheta_{32213}D_3\sigma_{22}\sigma_{13} +$$
$$+8\vartheta_{32213}D_3\sigma_{23}\sigma_{12} + 8\vartheta_{31123}D_3\sigma_{13}\sigma_{12} + \cdots \quad (21)$$

where α, β, and γ are certain coefficients, D_i the components of the electrostatic induction, σ_{kl} the mechanical stresses, g_{ijk} the piezoelectric coefficients, s^D_{ijkl} the elastic constants measured at a constant induction, q_{ijkl} the electrostriction constants, and ϑ_{mijkl} is the tensor of the fifth rank with internal symmetry $V[[V^2]^2]$, whose components were included into the expansion series of F in order to describe the emergence of new components in the tensor of elastic stiffnesses.

An LG crystal is a piezoelectric one above the phase transition point, but below it, there apprear several new components of its tensor of piezoelectric coefficients. If the induction D_3 in Eq. (21) is considered as the sum of the spontaneous induction D_{3S} and the induction D_{3I} that arises owing to the application of the external field,

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the spontaneous piezoelectric coefficients can be found from the equations

$$g_{311} = g_{322} = 2q_{3311}D_{3S}, \tag{22}$$

$$g_{333} = 2q_{3333}D_{3S}, \tag{23}$$

$$g_{123} = -g_{213} = 2q_{2313}D_{3S}, \tag{24}$$

$$g_{113} = g_{223} = 2q_{2323}D_{3S}. \tag{25}$$

An LG crystal differs essentially from a TGS one by the appearance of the spontaneous components of the tensor of piezoelectric coefficients at the phase transition together with the spontaneous components of the tensor of elastic constants. If $D_3 = D_{3S} + D_{3I}$, the following equations are valid for the latter:

$$s_{1123} = -s_{2223} = s_{1312} = \vartheta_{31123}D_{3S}, \tag{26}$$

$$s_{1113} = -s_{2213} = -s_{2312} = -\vartheta_{32213}D_{3S}. \tag{27}$$

Let the mechanical stress σ_{13} and the electric field E_1 be applied to an LG crystal. In this case, the induction

$$D_1 = \frac{1}{\gamma}E_1 \tag{28}$$

appears. Using the relation

$$\left(\frac{\partial F}{\partial D_3}\right) = E_3 \tag{29}$$

and taking into account that $E_3 = 0$, we obtain

$$\alpha D_3 + \beta D_3^3 = -4q_{2323}D_1\sigma_{13}, \tag{30}$$

$$\alpha D_3 + \beta D_3^3 = -4\frac{q_{2323}}{\gamma}E_1\sigma_{13} \tag{31}$$

or, converting from the induction to the polarization,

$$\alpha P_3 + \beta P_3^3 = -4\frac{q_{2323}}{\gamma}E_1\sigma_{13}. \tag{32}$$

Similar equations can be written down as well for other combinations of the electric field and the mechanical stress:

$$\alpha P_3 + \beta P_3^3 = -4\frac{q_{2313}}{\gamma}E_1\sigma_{23}, \tag{33}$$

$$\alpha P_3 + \beta P_3^3 = 4\frac{q_{2313}}{\gamma}E_2\sigma_{13}, \tag{34}$$

$$\alpha P_3 + \beta P_3^3 = -4\frac{q_{2323}}{\gamma}E_2\sigma_{23}. \tag{35}$$

Equations (32)—(35) testify to that the sign of spontaneous polarization in the LG crystal can be changed by applying not only the electric field E_3 but also the combinations $E_1\sigma_{23}$, $E_1\sigma_{13}$, $E_2\sigma_{23}$, and $E_2\sigma_{13}$ of the electric field and the mechanical stress, in which the electric field is directed perpendicularly to the direction of spontaneous polarization (ferroelastoelectric polarization switching). It should be noted that the combinations $E_3\sigma_{11}$, $E_3\sigma_{22}$, and $E_3\sigma_{33}$ of the electric field and the mechanical stress can also induce the polarization switching in the LG crystal, but in this case, it will occur practically at the expense of the electric field E_3 only.

Owing to the fact that the phase transition in the LG crystal is accompanied by the appearance of the spontaneous components of the tensor of elastic constants, the polarization switching in this crystal can be also provoked by applying a combination of two mechanical stresses. Let the mechanical stresses σ_{11} and σ_{23} be applied to the crystal. Using Eq. (29), taking into account that $E_3 = 0$, and neglecting the electrostriction terms $q_{33kl}D_3^2\sigma_{kl}$, we obtain

$$\alpha P_3 + \beta P_3^3 = -4\vartheta_{31123}\sigma_{11}\sigma_{23}. \tag{36}$$

Analogously,

$$\alpha P_3 + \beta P_3^3 = 4\vartheta_{32213}\sigma_{11}\sigma_{13}, \tag{37}$$

$$\alpha P_3 + \beta P_3^3 = 4\vartheta_{31123}\sigma_{11}\sigma_{23}, \tag{38}$$

$$\alpha P_3 + \beta P_3^3 = -4\vartheta_{32213}\sigma_{22}\sigma_{13}, \tag{39}$$

$$\alpha P_3 + \beta P_3^3 = -8\vartheta_{32213}\sigma_{12}\sigma_{23}, \tag{40}$$

$$\alpha P_3 + \beta P_3^3 = -8\vartheta_{31123}\sigma_{12}\sigma_{13}. \tag{41}$$

It follows from the dependences quoted above that the sign of spontaneous polarization in the LG crystal can be changed without the application of the electric field but under the influence of the following combinations of two mechanical stresses (ferrobielastic polarization switching) [7, 8]: $\sigma_{11}\sigma_{23}$, $\sigma_{11}\sigma_{13}$, $\sigma_{22}\sigma_{23}$, $\sigma_{22}\sigma_{13}$, $\sigma_{23}\sigma_{12}$, and $\sigma_{13}\sigma_{12}$.

Relations (32)—(35) and (36)—(41) demonstrate the nonlinear dependence of the polarization P_3 on the combinations $E_1\sigma_{23}$, $E_1\sigma_{13}$, $E_2\sigma_{23}$, $E_2\sigma_{13}$, $E_3\sigma_{11}$, $E_3\sigma_{22}$, and $E_3\sigma_{33}$ of the electric field and the mechanical stress, and the dependence of P_3 on the combinations $\sigma_{11}\sigma_{23}$, $\sigma_{11}\sigma_{13}$, $\sigma_{22}\sigma_{23}$, $\sigma_{22}\sigma_{13}$, $\sigma_{23}\sigma_{12}$, and $\sigma_{13}\sigma_{12}$ of two mechanical stresses, respectively. The plots of those dependences have to look like a hysteresis loop.

The difference between the thermodynamic potentials of two 180° domains that arise during the phase transition in the LG crystal, taking into account the form of the tensor of piezoelectric coefficients d and the tensor of elastic compliances s for each domain is

$$\Delta\Phi = -2P_{(S)3}E_3 - 4d_{123}E_1\sigma_{23} - 4d_{113}E_1\sigma_{13} -$$
$$-4d_{113}E_2\sigma_{23} + 4d_{123}E_2\sigma_{13} - 2d_{311}E_3\sigma_{11} -$$
$$-2d_{311}E_3\sigma_{22} - 2d_{333}E_3\sigma_{33} - 4s^E_{1123}\sigma_{11}\sigma_{23} +$$
$$+4s^E_{2213}\sigma_{11}\sigma_{13} + 4s^E_{1123}\sigma_{22}\sigma_{23} - 4s^E_{2213}\sigma_{22}\sigma_{13} -$$
$$-8s^E_{2213}\sigma_{23}\sigma_{12} - 8s^E_{1123}\sigma_{13}\sigma_{12}\ldots, \qquad (42)$$

where P_i are the components of the polarization vector, d_{ikl} the piezoelectric coefficients, and s^E_{ijkl} the coefficients of elastic compliance measured at a constant electric field E.

Therefore, provided that a $Pb_5Ge_3O_{11}$ crystal undergoes the action of the electric field E_3 or one of the $E_1\sigma_{23}$, $E_1\sigma_{13}$, $E_2\sigma_{23}$, $E_2\sigma_{13}$, $E_3\sigma_{11}$, $E_3\sigma_{22}$, or $E_3\sigma_{33}$ combinations of the electric field and the mechanical stress, or one of the $\sigma_{11}\sigma_{23}$, $\sigma_{11}\sigma_{13}$, $\sigma_{22}\sigma_{23}$, $\sigma_{22}\sigma_{13}$, $\sigma_{23}\sigma_{12}$, or $\sigma_{13}\sigma_{12}$ combinations of two mechanical stresses, there appears a difference between the energies of domains, and the crystal will pass to a state with minimal energy.

The experimental optical researches, the method and the results of which were reported in works [7, 8], established that if the combination $\sigma_{11}\sigma_{13}$ of the mechanical stresses with a value of 8×10^{14} Pa^2 was applied to the LG specimen in the course of its cooling from above to below its Curie point, the specimen became composed of a single domain, although, following all the rules, ferroelectrics have a polydomain structure below their Curie points if no constant external electric field is applied. At the same time, if the same specimen was cooled under the action of the $-\sigma_{11}\sigma_{13}$ combination of stresses, it also became monodomain, but with the opposite orientation of the vector of spontaneous polarization. The direction of spontaneous polarization is easily controlled by using the polarization-optical method, because domains with different directions P_S rotate the polarization plane of light that transmits through a crystal into opposite sides. The same results were also observed for the stress combination $\sigma_{22}\sigma_{23}$. Thus, the ferroelectric LG crystal is simultaneously a ferrobielastic.

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НЕТРАДИЦІЙНІ СПОСОБИ ПЕРЕПОЛЯРИЗАЦІЇ ОДНООСЬОВИХ СЕГНЕТОЕЛЕКТРИКІВ

О.Ф. Дуднік, В.М. Дуда, О.І. Кушнерьов

Р е з ю м е

Розглянуто термодинамічні потенціали чистих одноосьових сегнетоелектриків тригліцинсульфату та германату свинцю із урахуванням їх вторинних фероїдних властивостей. Показано, що переполяризація у цих кристалах може проходити під дією певних комбінацій зовнішніх механічних напружень та електричних полів, які можуть бути спрямовані перпендикулярно до сегнетоелектричної осі.