Transverse Piezoelectric Resonance in KH$_2$PO$_4$ Type Crystals

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December 31, 2013

Within the framework of proton model with taking into account the piezoelectric interaction with the shear strain $\varepsilon_4$, a dynamic dielectric response of KH$_2$PO$_4$ family crystals to the electric field perpendicular to the axis of spontaneous polarization is considered. Piezoelectric resonance frequencies of rectangular thin plates of the crystals cut in the (100) plane ($0^\circ$ X-cut) are calculated.

Key words: ferroelectrics, KH$_2$PO$_4$, piezoelectric resonance.

PACS: 77.22.Ch, 77.22.Gm, 77.84.Fa, 77.65.Fs

1. Introduction

In our previous paper [1] we explored the dynamic dielectric response of square thin plates, cut from the KH$_2$PO$_4$ family ferroelectric and antiferroelectric crystals in the planes (001), to the a.c. electric field applied along the axis [001]. This is the axis of a spontaneous polarization in the KH$_2$PO$_4$ type ferroelectrics, and we call this field longitudinal.

Using the modification of the proton ordering model [2] that includes the piezoelectric coupling with the shear strain $\varepsilon_6$, within the framework of the Glauber approach [3] and the four-particle cluster approximation, we obtained expressions for the dynamic dielectric permittivity of the crystals, which took into account the dynamics of the shear strain $\varepsilon_6$. The found expressions for the resonant frequencies of the longitudinal dielectric permittivity are in a good agreement with experiment.

Useful information can be obtained also by investigating the transverse dielectric response of ferroelectric crystals, and especially the proton glass systems of the Rb$_{1-x}$(NH$_4$)$_x$PO$_4$ type, when the applied electric field is perpendicular to the axis of spontaneous polarization. For the crystals of the KH$_2$PO$_4$ family that would be the axis [100]. The field $E_1$ induces the shear strain $\varepsilon_4$ via the piezoelectric coefficient $d_{14}$, and the corresponding dielectric permittivity exhibits a piezoelectric resonant dispersion. Frequencies of these resonances will be determined in the present paper. In our calculations we shall use the models presented in [4-6], which take into account the piezoelectric coupling to the shear strain $\varepsilon_4$.

2. Transverse dynamic permittivity of KH$_2$PO$_4$ type crystals

We shall consider shear mode vibrations of a thin $L_y \times L_z$ rectangular plate of a KH$_2$PO$_4$ crystal, cut in the (100) plane, with the edges along [010] and [001] ($0^\circ$ X-cut). The vibrations are induced by time-dependent electric field $E_{1t} = E_1 e^{i\omega t}$.

Dynamics of pseudospin subsystem will be considered in the spirit of the stochastic Glauber model [3], using the four-particle cluster approximation. The system of equations for the time-dependent deuteron...
Solution of (4) with the boundary conditions (6) is

\[ -\alpha \frac{d}{dt} \left( \prod_{f} \sigma_{af} \right) = \sum_{f'} \left( \prod_{f} \sigma_{af} \right) \left[ 1 - \sigma_{af} \tanh \frac{1}{2} \beta \epsilon_{af}(t) \right], \]

where \( \epsilon_{af}(t) \) is the local field acting on the \( f' \)th deuteron in the \( af \)th cell, which can be found from the system Hamiltonian \([4, 5, 6]\). \( \alpha \) is the parameter setting the time scale of the dynamic processes in the pseudospin subsystem.

Dynamics of the deformational processes is described using classical Newtonian equations of motion of an elementary volume, which for the relevant to our system displacements \( u_1 \) and \( u_2 \) (\( \epsilon_4 = \frac{\partial u_1}{\partial x} + \frac{\partial u_2}{\partial y} \)) read

\[ \rho \frac{\partial^2 u_2}{\partial t^2} = \frac{\partial \sigma_4}{\partial z}, \quad \rho \frac{\partial^2 u_3}{\partial t^2} = \frac{\partial \sigma_4}{\partial y}. \]

Here \( \rho \) is the crystal density, \( \sigma_4 \) is the mechanical shear stress, which, being the function of \( \eta^{(4)}, E_1 \), and \( \epsilon_4 \), is found from the constitutive equations derived in \([7]\).

Following the usually used procedure \([1, 4, 7, 8]\), at small deviations from the equilibrium, we derive the following equations for the displacements

\[ \frac{\partial^2 u_1}{\partial y^2} + k_4^2 u_1 = 0, \quad \frac{\partial^2 u_2}{\partial x^2} + k_4^2 u_2 = 0, \]

where \( k_4 \) is the wavevector

\[ k_4 = \frac{\omega \sqrt{\rho}}{\sqrt{c_{44}^E(\alpha \omega)}}. \]

Expressions for the frequency dependent elastic constants \( c_{44}^E(\alpha \omega) \) for the KH_2PO_4 type ferroelectrics and NH_4H_2PO_4 type antiferroelectrics are presented in \([4, 6]\).

Differentiating the first and second equations of (5) with respect to \( z \) and \( y \), correspondingly, and adding the two obtained equations, we arrive at the single equation for the strain \( \epsilon_4 \)

\[ \frac{\partial^2 \epsilon_4(y, z)}{\partial y^2} + \frac{\partial^2 \epsilon_4(y, z)}{\partial z^2} + k_4^2 \epsilon_4(y, z) = 0. \]

Boundary conditions for \( \epsilon_4(y, z) \) follow from the assumption that the crystal is traction free at its edges (at \( y = 0, y = L_y, z = 0, z = L_z \), to be denoted as \( \Sigma \))

\[ \sigma_{4|\Sigma} = 0. \]

Solution of (4) with the boundary conditions (6) is

\[ \epsilon_4(y, z) = \epsilon_{40} + \epsilon_{40} \sum_{k, l=0}^{\infty} \frac{16}{(2k+1)(2l+1)\pi^2} \frac{\omega^2}{(c_{44}^E(\alpha \omega))^2} \frac{1}{L_x^2} \sin \frac{\pi(2k+1)y}{L_y} \sin \frac{\pi(2l+1)z}{L_z}, \]

where

\[ \epsilon_{40} = \frac{\epsilon_{14}(\alpha \omega)}{c_{44}^E(\alpha \omega)} E_1; \]

\( \omega_{kl}^0 \) is given by

\[ \omega_{kl}^0 = \sqrt{\frac{\rho (c_{44}^E(\alpha \omega))^2 \pi^2}{L_x^2} \left( \frac{(2k+1)^2}{L_y^2} + \frac{(2l+1)^2}{L_z^2} \right)}, \]

\( \epsilon_{14}(\alpha \omega) \) is the piezoelectric coefficient, the expressions for which for the KH_2PO_4 and NH_4H_2PO_4 type crystals, have been derived in \([4, 5, 6]\).

The transverse dynamic dielectric susceptibility of a free crystal has been obtained in the following form \([4]\)

\[ \chi_{11}^\alpha(\omega) = \chi_{11}^E(\omega) + R_4(\omega) \frac{\epsilon_{14}^2(\alpha \omega)}{c_{44}^E(\alpha \omega)}, \]
where

\[ R_4(\omega) = 1 + \sum_{k,l=0}^{\infty} \frac{64}{(2k+1)^2(2l+1)^2\pi^4} \beta_{kl} \frac{\omega^2}{(\omega^2 - \omega_0^2)^2} \]

\( \chi_{11}^E(\alpha \omega) \) is the dynamic dielectric susceptibility of a clamped crystal. The corresponding expressions can be found in [4–6].

The static and the high frequency limits of (9) are the static susceptibility of a free crystal [2] and the dynamic susceptibility of a mechanically clamped crystal, exhibiting relaxational dispersion in the microwave region. Thus, eq. (9) explicitly describes the effect of crystal clamping by high-frequency electric field.

In the intermediate frequency region, the susceptibility has a resonance dispersion with numerous peaks at frequencies where \( \text{Re}[R_4(\omega)] \to \infty \). Frequency variation of \( c_{44}^E(\alpha \omega) \) is perceptible only in the region of the microwave dispersion of the dielectric susceptibility. Below this region it is practically frequency independent and coincides with the static elastic constant \( c_{44}^E \). Since the resonance frequencies are expected to be in the \( 10^4 - 10^7 \) Hz range, depending on temperature and sample dimensions, the equation for the resonance frequencies (8) is reduced to an explicit expression by putting in it \( c_{44}^E(\alpha \omega) \to c_{44}^E \).

In figure 1 we plotted the frequency dependences of the transverse dynamic dielectric permittivity for \( \text{KH}_2\text{PO}_4 \) and \( \text{NH}_4\text{H}_2\text{PO}_4 \) crystals. The used values of the model parameters can be found in [4–6]. For the other ferroelectric and antiferroelectric crystals of the \( \text{KH}_2\text{PO}_4 \) family, obtained by isomorphic replacement of K and P ions, these dependences are totally analogous. Evolution of the permittivity with increasing frequency from the free crystal value through the piezoelectric resonances to the clamped crystal value and then to the relaxational dispersion in the microwave region is observed for all crystals of the family.

Since the elastic constant \( c_{44}^E \) shows no significant anomalies at the transition point or temperature variation in these crystals [4, 11], the first resonant frequency of the transverse permittivity, according to (8) at \( k = l = 0 \), is temperature independent as well. This is illustrated in fig. 2 where these frequencies are given for different crystals of the \( \text{KH}_2\text{PO}_4 \) family.
Figure 2. The temperature dependences of the first resonant frequency of the transverse dynamic dielectric permittivity for different crystals of the KH₂PO₄ family. \( L_y = 2 \text{ mm}, L_z = 1 \text{ mm} \).

3. Conclusions

Within the proton ordering model with taking into account the shear strain \( \varepsilon_4 \) we explored a dynamic response of ferroelectric and antiferroelectric crystals of the KH₂PO₄ family to a transverse external harmonic electric field \( E_1 \). Corrected expressions for the piezoelectric resonance frequencies of simply supported rectangular \( 0^\circ \) X-cuts of these crystals are obtained. The ultimate goal of the present studies will be to generalize the obtained expression for the dynamic permittivity to the case of the Rb\(_{1-x}(\text{NH}_4)_x\)PO₄ type proton glasses, in order to explore their dynamic dielectric response.

Acknowledgement

The authors acknowledge support from the State Foundation for Fundamental Studies of Ukraine, Project “Electromechanical nonlinearity of mixed ferro-antiferroelectric crystals of dihydrogen phosphate family” No F53.2/070.

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