

Dynamic properties of $\text{NH}_3\text{CH}_2\text{COOH}\cdot\text{H}_2\text{PO}_3$ ferroelectric

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Using a modified pseudospin model of $\text{NH}_3\text{CH}_2\text{COOH}\cdot\text{H}_2\text{PO}_3$ ferroelectric taking into account the piezoelectric coupling with strains ε_i , ε_4 , ε_5 and ε_6 within Glauber method in two-particle cluster approximation, we have calculated components of dynamic dielectric permittivity tensor and relaxation times of the model. At the proper set of theory parameters, frequency and temperature dependences of the components of permittivity and temperature dependences of the relaxation times are studied. A satisfactory agreement of the theoretical results with experimental data for longitudinal permittivity is obtained.

Key words: *ferroelectrics, cluster approximation, dynamic dielectric permittivity, relaxation time*

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1. Introduction

The problem of investigation of physical properties of ferroelectric materials has occupied one of the central places in condensed matter physics for a long time. The presence of different classes of these materials with different crystal structure and chemical composition requires elaboration of universal methods for investigation of phase transition mechanisms. It is also necessary to develop concrete microscopic theories for them, which could explain the observed experimental data for thermodynamic and dynamic characteristics and anomalies in the behaviour of these characteristics in the phase transition region.

Granting this, glycinium phosphite $\text{NH}_3\text{CH}_2\text{COOH}\cdot\text{H}_2\text{PO}_3$ (GPI) is of special interest due to the combination of structure elements typical of different classes of ferroelectric crystals. In [1–3] basing on the analysis of structural data [4] it was determined that the main role in the phase transition in GPI is played by two structurally nonequivalent types of O-H...O hydrogen bonds of different length, which connect phosphite groups HPO_3 in the chains along the crystallographic c -axis. As a result, in [1, 3] there was proposed a model of GPI crystal with proton ordering, within which the main peculiarities of its dielectric permittivity were explained qualitatively. Later, this model was supplemented by taking into account the piezoelectric coupling of proton and lattice subsystems [5], which made it possible to calculate thermal, piezoelectric and elastic characteristics of GPI. At the proper set of theory parameters, a good agreement of the obtained theoretical results with corresponding experimental data for the crystals of this type was obtained.

In order to better understand the mechanism of phase transition in these crystals and their physical properties, the effects of transverse electric fields [6] and uniaxial pressures [7] on the static physical properties of GPI were calculated within the model proposed in [5]. A good agreement of the obtained theoretical results with the available experimental data was obtained. This confirms the key role of proton ordering on the above mentioned bonds. It should be noted that several results obtained in these papers may be interpreted as predictions which will be a stimulus for further experimental investigations.

The aim of this paper is to study the relaxation phenomena in GPI and explain the available experimental data [8–11] for longitudinal dynamic characteristics within the proton ordering model proposed in [5].

2. Model of GPI crystal

The pseudospin model proposed in [5] considers the system of protons in GPI, localized on O-H...O bonds between phosphite groups HPO_3 , which form chains along the crystallographic c -axis of the crystal (figure 1). Dipole moments $\mathbf{d}_{qf} = \boldsymbol{\mu}_f \frac{\sigma_{qf}}{2}$ are ascribed to the protons on the bonds. Here, q is a primitive cell index, $f = 1, \dots, 4$; $\frac{\sigma_{qf}}{2}$ are pseudospin variables that describe the changes connected with reorientation of the dipole moments.

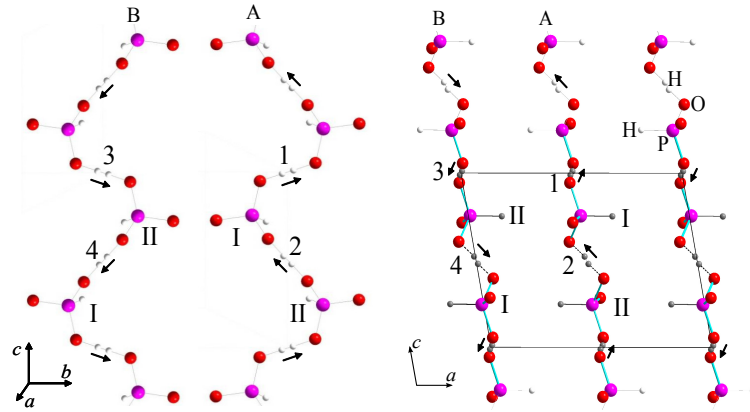


Figure 1. (Colour online) Orientations of vectors \mathbf{d}_{qf} in the primitive cell in the ferroelectric phase [5, 6].

The Hamiltonian of a proton subsystem of GPI, which takes into account the short-range and long-range interactions and the applied electric fields E_1, E_2, E_3 along the positive directions of the Cartesian axes X, Y and Z ($X \perp (b, c), Y \parallel b, Z \parallel c$) can be written in such a way:

$$\hat{H} = NU_{\text{seed}} + \hat{H}_{\text{short}} + \hat{H}_{\text{long}} + \hat{H}_E, \quad (2.1)$$

where N is the total number of primitive cells. The first term in (2.1) is the “seed” energy, which relates to the heavy ion sublattice and does not explicitly depend on the configuration of the proton subsystem. It includes elastic, piezoelectric and dielectric parts expressed in terms of electric fields E_i and strains ε_i :

$$\begin{aligned} U_{\text{seed}} = v & \left[\frac{1}{2} \sum_{i,i'=1}^3 c_{ii'}^{E0}(T) \varepsilon_i \varepsilon_{i'} + \frac{1}{2} \sum_{j=4}^6 c_{jj}^{E0}(T) \varepsilon_j^2 + \sum_{i=1}^3 c_{i5}^{E0}(T) \varepsilon_i \varepsilon_5 + c_{46}^{E0}(T) \varepsilon_4 \varepsilon_6 \right. \\ & - \sum_{i=1}^3 e_{2i}^0 \varepsilon_i E_2 - e_{25}^0 \varepsilon_5 E_2 - e_{14}^0 \varepsilon_4 E_1 - e_{16}^0 \varepsilon_6 E_1 - e_{34}^0 \varepsilon_4 E_3 - e_{36}^0 \varepsilon_6 E_3 \\ & \left. - \frac{1}{2} \chi_{11}^{\varepsilon 0} E_1^2 - \frac{1}{2} \chi_{22}^{\varepsilon 0} E_2^2 - \frac{1}{2} \chi_{33}^{\varepsilon 0} E_3^2 - \chi_{31}^{\varepsilon 0} E_3 E_1 \right]. \end{aligned} \quad (2.2)$$

Parameters $c_{ij}^{E0}(T), e_{ij}^0, \chi_{ij}^{\varepsilon 0}$ are the so-called “seed” elastic constants, piezoelectric stresses and dielectric susceptibilities, respectively; v is the volume of a primitive cell.

The second term in (2.1) is the Hamiltonian of short-range interactions:

$$\hat{H}_{\text{short}} = 2w \sum_{qq'} \left(\frac{\sigma_{q1}}{2} \frac{\sigma_{q2}}{2} + \frac{\sigma_{q3}}{2} \frac{\sigma_{q4}}{2} \right) (\delta_{\mathbf{R}_q \mathbf{R}_{q'}} + \delta_{\mathbf{R}_q + \mathbf{R}_c, \mathbf{R}_{q'}}). \quad (2.3)$$

In (2.3), σ_{qf} is the z -component of the pseudospin operator that describes the state of the f -th bond ($f = 1, 2, 3, 4$) in the q -th cell. The first Kronecker delta corresponds to the interaction between protons in the chains near the tetrahedra HPO_3 of type “I” (figure 1), where the second one near the tetrahedra HPO_3 of type “II”, \mathbf{R}_c is the lattice vector along the crystallographic c -axis. Contributions into the energy of interactions between protons near the tetrahedra of different type, as well as the mean values of the pseudospins $\eta_f = \langle \sigma_{qf} \rangle$, which are related to the tetrahedra of different type, are equal. Parameter w , which describes the short-range interactions within the chains, is expanded linearly into the series over strains ε_i :

$$w = w^0 + \sum_{i=1}^6 \delta_i \varepsilon_i. \quad (2.4)$$

The third term in (2.1) describes the long-range dipole-dipole interactions and indirect (through the lattice vibrations) interactions between protons, which are taken into account in the mean field approximation:

$$\hat{H}_{\text{long}} = \frac{1}{2} \sum_{qq'ff'} J_{ff'}(qq') \frac{\langle \sigma_{qf} \rangle}{2} \frac{\langle \sigma_{q'f'} \rangle}{2} - \sum_{qq'ff'} J_{ff'}(qq') \frac{\langle \sigma_{q'f'} \rangle}{2} \frac{\sigma_{qf}}{2}. \quad (2.5)$$

Fourier transforms of interaction constants $J_{ff'} = \sum_{q'} J_{ff'}(qq')$ at $\mathbf{k} = 0$ are linearly expanded over the strains ε_i :

$$J_{ff'} = J_{ff'}^0 + \frac{\partial J_{ff'}}{\partial \varepsilon_i} \varepsilon_i = J_{ff'}^0 + \sum_{i=1}^6 \psi_{ff'i} \varepsilon_i. \quad (2.6)$$

Thus, (2.5) can be written as:

$$\hat{H}_{\text{long}} = NH^0 - \sum_q \sum_{f=1}^4 \mathcal{H}_f \frac{\sigma_{qf}}{2}, \quad (2.7)$$

where

$$H^0 = \frac{1}{8} J_{11}(\eta_1^2 + \eta_3^2) + \frac{1}{8} J_{22}(\eta_2^2 + \eta_4^2) + \frac{1}{4} J_{13} \eta_1 \eta_3 + \frac{1}{4} J_{24} \eta_2 \eta_4 + \frac{1}{4} J_{12}(\eta_1 \eta_2 + \eta_3 \eta_4) + \frac{1}{4} J_{14}(\eta_1 \eta_4 + \eta_2 \eta_3). \quad (2.8)$$

In (2.7) the following notations are used:

$$\begin{aligned} \mathcal{H}_1 &= \frac{1}{2} J_{11} \eta_1 + \frac{1}{2} J_{12} \eta_2 + \frac{1}{2} J_{13} \eta_3 + \frac{1}{2} J_{14} \eta_4, \\ \mathcal{H}_2 &= \frac{1}{2} J_{22} \eta_2 + \frac{1}{2} J_{12} \eta_1 + \frac{1}{2} J_{24} \eta_4 + \frac{1}{2} J_{14} \eta_3, \\ \mathcal{H}_3 &= \frac{1}{2} J_{11} \eta_3 + \frac{1}{2} J_{12} \eta_4 + \frac{1}{2} J_{13} \eta_1 + \frac{1}{2} J_{14} \eta_2, \\ \mathcal{H}_4 &= \frac{1}{2} J_{22} \eta_4 + \frac{1}{2} J_{12} \eta_3 + \frac{1}{2} J_{24} \eta_2 + \frac{1}{2} J_{14} \eta_1. \end{aligned} \quad (2.9)$$

The fourth term in (2.1) describes the interactions of pseudospins with an external electric field:

$$\hat{H}_E = - \sum_{qf} \boldsymbol{\mu}_f \mathbf{E} \frac{\sigma_{qf}}{2}. \quad (2.10)$$

Here, $\boldsymbol{\mu}_1 = (\mu_{13}^x, \mu_{13}^y, \mu_{13}^z)$, $\boldsymbol{\mu}_3 = (-\mu_{13}^x, \mu_{13}^y, -\mu_{13}^z)$, $\boldsymbol{\mu}_2 = (-\mu_{24}^x, -\mu_{24}^y, \mu_{24}^z)$, $\boldsymbol{\mu}_4 = (\mu_{24}^x, -\mu_{24}^y, -\mu_{24}^z)$ are the effective dipole moments per one pseudospin.

The two-particle cluster approximation for short-range interactions is used for the calculation of thermodynamic characteristics of GPI. In this approximation, thermodynamic potential is given by:

$$G = NU_{\text{seed}} + NH^0 - k_B T \sum_q \left[2 \ln \text{Sp} e^{-\beta \hat{H}_q^{(2)}} - \sum_{f=1}^4 \ln \text{Sp} e^{-\beta \hat{H}_{qf}^{(1)}} \right] - Nv \sum_{i=1}^6 \sigma_i \varepsilon_i. \quad (2.11)$$

Here, $\beta = 1/k_B T$, k_B is Boltzmann constant, $\hat{H}_q^{(2)}$, $\hat{H}_{qf}^{(1)}$ are two-particle and one-particle Hamiltonians:

$$\hat{H}_q^{(2)} = -2w \left(\frac{\sigma_{q1} \sigma_{q2}}{2} + \frac{\sigma_{q3} \sigma_{q4}}{2} \right) - \sum_{f=1}^4 \frac{y_f \sigma_{qf}}{\beta}, \quad (2.12)$$

$$\hat{H}_{qf}^{(1)} = -\frac{\bar{y}_f \sigma_{qf}}{\beta}, \quad (2.13)$$

where such notations are used:

$$y_f = \beta(\Delta_f + \mathcal{H}_f + \boldsymbol{\mu}_f \mathbf{E}), \quad (2.14)$$

$$\bar{y}_f = \beta \Delta_f + y_f. \quad (2.15)$$

The symbols Δ_f are the effective cluster fields created by the neighboring bonds from outside the cluster. Minimizing the thermodynamic potential (2.11) with respect to the cluster fields Δ_f and to the strains ε_i , and expressing Δ_f through the equilibrium order parameters $\tilde{\eta}_1 = \tilde{\eta}_3 = \tilde{\eta}_{13}$, $\tilde{\eta}_2 = \tilde{\eta}_4 = \tilde{\eta}_{24}$, we have obtained a system of equations for the equilibrium order parameters and strains for the case of zero mechanical stresses and fields:

$$\begin{aligned} \tilde{\eta}_{13} &= \frac{1}{\tilde{D}} [\sinh(\tilde{y}_{13} + \tilde{\eta}_{24}) + a^2 \sinh(\tilde{y}_{13} - \tilde{\eta}_{24}) + 2a \sinh \tilde{y}_{13}], \\ \tilde{\eta}_{24} &= \frac{1}{\tilde{D}} [\sinh(\tilde{y}_{13} + \tilde{\eta}_{24}) - a^2 \sinh(\tilde{y}_{13} - \tilde{\eta}_{24}) + 2a \sinh \tilde{\eta}_{24}], \\ 0 &= c_{l1}^{E0} \varepsilon_1 + c_{l2}^{E0} \varepsilon_2 + c_{l3}^{E0} \varepsilon_3 + c_{l5}^{E0} \varepsilon_5 - \frac{2\delta_l}{v} + \frac{2\delta_l}{v\tilde{D}} M_\varepsilon \\ &\quad - \frac{\psi_{1l}^+}{4v} \tilde{\eta}_{13}^2 - \frac{\psi_{2l}^+}{2v} \tilde{\eta}_{13} \tilde{\eta}_{24} - \frac{\psi_{3l}^+}{4v} \tilde{\eta}_{24}^2, \quad (l = 1, \dots, 6), \end{aligned} \quad (2.16)$$

where such notations are used:

$$\begin{aligned} \tilde{y}_{13} &= \frac{1}{2} \ln \frac{1 + \tilde{\eta}_{13}}{1 - \tilde{\eta}_{13}} + \beta v_1^+ \tilde{\eta}_{13} + \beta v_2^+ \tilde{\eta}_{24}, & \tilde{y}_{24} &= \beta v_2^+ \tilde{\eta}_{13} + \frac{1}{2} \ln \frac{1 + \tilde{\eta}_{24}}{1 - \tilde{\eta}_{24}} + \beta v_3^+ \tilde{\eta}_{24}, \\ v_l^\pm &= v_l^{0\pm} + \sum_{i=1}^6 \psi_{li}^\pm \varepsilon_i, & v_1^{0\pm} &= \frac{1}{4} (J_{11}^0 \pm J_{13}^0); & v_2^{0\pm} &= \frac{1}{4} (J_{12}^0 \pm J_{14}^0); & v_3^{0\pm} &= \frac{1}{4} (J_{22}^0 \pm J_{24}^0); \\ \psi_{li}^\pm &= \frac{1}{4} (\psi_{11i} \pm \psi_{13i}), & \psi_{2i}^\pm &= \frac{1}{4} (\psi_{12i} \pm \psi_{14i}), & \psi_{3i}^\pm &= \frac{1}{4} (\psi_{22i} \pm \psi_{24i}), \\ \tilde{D} &= \cosh(\tilde{y}_{13} + \tilde{y}_{24}) + a^2 \cosh(\tilde{y}_{13} - \tilde{y}_{24}) + 2a \cosh \tilde{y}_{13} + 2a \cosh \tilde{y}_{24} + a^2 + 1, \\ M_\varepsilon &= 2a^2 \cosh(\tilde{y}_{13} - \tilde{y}_{24}) + 2a \cosh \tilde{y}_{13} + 2a \cosh \tilde{y}_{24} + 2a^2. \end{aligned}$$

3. Theoretical calculations of dynamic dielectric permittivity of mechanically clamped GPI crystal

To calculate the dynamic properties we use an approach based on the ideas of a stochastic Glauber model [12]. Using the methods developed in [13], we obtain the following system of Glauber equations for time dependent correlation functions of the pseudospins:

$$-\alpha \frac{d}{dt} \left\langle \prod_f \sigma_{qf} \right\rangle = \sum_{f'} \left\langle \prod_f \sigma_{qf} \left[1 - \sigma_{qf'} \tanh \frac{1}{2} \beta \varepsilon_{qf'}(t) \right] \right\rangle, \quad (3.1)$$

where parameter α determines the time scale of dynamic processes, $\varepsilon_{qf'}(t)$ is the local field acting on the f' -th pseudospin in q -th cell. We use a two-particle cluster approximation in order to obtain a closed system of equations. In this approximation, local fields $\varepsilon_{qf}(t)$ are coefficients at $\sigma_{qf}/2$ in two-particle and one-particle Hamiltonians (2.12), (2.13). Correspondingly, these fields are presented in a two-particle approximation:

$$\varepsilon_{q1} = w\sigma_{q2} + \frac{y_1}{\beta}, \quad \varepsilon_{q2} = w\sigma_{q1} + \frac{y_2}{\beta}, \quad \varepsilon_{q3} = w\sigma_{q4} + \frac{y_3}{\beta}, \quad \varepsilon_{q4} = w\sigma_{q3} + \frac{y_4}{\beta}, \quad (3.2)$$

and in a one-particle approximation:

$$\varepsilon_{qf} = \frac{\bar{y}_f}{\beta}. \quad (3.3)$$

As a result, from (3.1) we obtain a system of equations for mean values of pseudospins $\langle \sigma_{qf} \rangle = \eta_f$ in a two-particle approximation:

$$\begin{aligned} \alpha \frac{d}{dt} \eta_1 &= -\eta_1 + P_1 \eta_2 + L_1, & \alpha \frac{d}{dt} \eta_3 &= -\eta_3 + P_3 \eta_4 + L_3, \\ \alpha \frac{d}{dt} \eta_2 &= P_2 \eta_1 - \eta_2 + L_2, & \alpha \frac{d}{dt} \eta_4 &= P_4 \eta_3 - \eta_4 + L_4 \end{aligned} \quad (3.4)$$

and in a one-particle approximation:

$$\alpha \frac{d}{dt} \eta_f = -\eta_f + \tanh \frac{\bar{y}_f}{2}, \quad (3.5)$$

where the following notations are used:

$$\begin{aligned} P_f &= \frac{1}{2} \left[\tanh \left(\frac{\beta w}{2} + \frac{y_f}{2} \right) - \tanh \left(-\frac{\beta w}{2} + \frac{y_f}{2} \right) \right], \\ L_f &= \frac{1}{2} \left[\tanh \left(\frac{\beta w}{2} + \frac{y_f}{2} \right) + \tanh \left(-\frac{\beta w}{2} + \frac{y_f}{2} \right) \right]. \end{aligned}$$

Let us restrict ourselves to the case of small deviations from equilibrium state to solve the equations (3.4) and (3.5). For this case we write η_f and effective fields y_f, \bar{y}_f in the form of a sum of equilibrium values and their deviations from equilibrium values (a mechanically clamped crystal):

$$\begin{aligned} \eta_{1,3} &= \tilde{\eta}_{13} + \eta_{1,3t}, & \eta_{2,4} &= \tilde{\eta}_{24} + \eta_{2,4t}, \\ y_1 &= \tilde{y}_{13} + y_{1t} = \beta \left[\Delta_{13} + 2v_1^+ \tilde{\eta}_{13} + 2v_2^+ \tilde{\eta}_{24} + \Delta_{1t} + v_1^+ (\eta_{1t} + \eta_{3t}) + v_2^+ (\eta_{2t} + \eta_{4t}) \right. \\ &\quad \left. + v_1^- (\eta_{1t} - \eta_{3t}) + v_2^- (\eta_{2t} - \eta_{4t}) + \mu_{13}^x E_{1t} + \mu_{13}^y E_{2t} + \mu_{13}^z E_{3t} \right], & E_{it} &= E_t e^{i\omega t}, \\ y_3 &= \tilde{y}_{13} + y_{3t} = \beta \left[\Delta_{13} + 2v_1^+ \tilde{\eta}_{13} + 2v_2^+ \tilde{\eta}_{24} + \Delta_{1t} + v_1^+ (\eta_{1t} + \eta_{3t}) + v_2^+ (\eta_{2t} + \eta_{4t}) \right. \\ &\quad \left. - v_1^- (\eta_{1t} - \eta_{3t}) - v_2^- (\eta_{2t} - \eta_{4t}) - \mu_{13}^x E_{1t} + \mu_{13}^y E_{2t} - \mu_{13}^z E_{3t} \right], \\ y_2 &= \tilde{y}_{24} + y_{2t} = \beta \left[\Delta_{24} + 2\beta v_2^+ \tilde{\eta}_{13} + 2\beta v_3^+ \tilde{\eta}_{24} + \Delta_{2t} + v_2^+ (\eta_{1t} + \eta_{3t}) + v_3^+ (\eta_{2t} + \eta_{4t}) \right. \\ &\quad \left. + v_2^- (\eta_{1t} - \eta_{3t}) + v_3^- (\eta_{2t} - \eta_{4t}) - \mu_{24}^x E_{1t} - \mu_{24}^y E_{2t} + \mu_{24}^z E_{3t} \right], \\ y_4 &= \tilde{y}_{24} + y_{4t} = \beta \left[\Delta_{24} + 2\beta v_2^+ \tilde{\eta}_{13} + 2\beta v_3^+ \tilde{\eta}_{24} + \Delta_{2t} + v_2^+ (\eta_{1t} + \eta_{3t}) + v_3^+ (\eta_{2t} + \eta_{4t}) \right. \\ &\quad \left. - v_2^- (\eta_{1t} - \eta_{3t}) - v_3^- (\eta_{2t} - \eta_{4t}) + \mu_{24}^x E_{1t} - \mu_{24}^y E_{2t} - \mu_{24}^z E_{3t} \right], \\ \bar{y}_f &= \beta \Delta_f + \tilde{y}_f + \beta \Delta_{ft} + y_{ft}, & \tilde{y}_1 &= \tilde{y}_3 = \tilde{y}_{13}, & \tilde{y}_2 &= \tilde{y}_4 = \tilde{y}_{24}. \end{aligned} \quad (3.6)$$

Here, $\Delta_{13} = \Delta_1 = \Delta_3$, $\Delta_{24} = \Delta_2 = \Delta_4$ are equilibrium effective cluster fields, and Δ_{ft} are their deviations from equilibrium values. Parameters v_i^\pm describe long-range interactions. We decompose the coefficients P_f and L_f in a series of $\frac{y_{ft}}{2}$ limited by linear items:

$$\begin{aligned}
 P_{1,3} &= P_{13}^{(0)} + \frac{y_{1,3t}}{2} P_{13}^{(1)}, & L_{1,3} &= L_{13}^{(0)} + \frac{y_{1,3t}}{2} L_{13}^{(1)}, \\
 P_{2,4} &= P_{24}^{(0)} + \frac{y_{2,4t}}{2} P_{24}^{(1)}, & L_{2,4} &= L_{24}^{(0)} + \frac{y_{2,4t}}{2} L_{24}^{(1)},
 \end{aligned} \tag{3.7}$$

where the following notations are used:

$$P_{13}^{(0)} = \frac{1-a^2}{Z_{13}}, \quad P_{13}^{(1)} = -\frac{4a(1-a^2) \sinh \tilde{y}_{13}}{Z_{13}^2}, \quad L_{13}^{(0)} = \frac{2a \sinh \tilde{y}_{13}}{Z_{13}}, \quad L_{13}^{(1)} = \frac{4a[2a + (1+a^2) \cosh \tilde{y}_{13}]}{Z_{13}^2},$$

$$P_{24}^{(0)} = \frac{1-a^2}{Z_{24}}, \quad P_{24}^{(1)} = -\frac{4a(1-a^2) \sinh \tilde{y}_{24}}{Z_{24}^2}, \quad L_{24}^{(0)} = \frac{2a \sinh \tilde{y}_{24}}{Z_{24}}, \quad L_{24}^{(1)} = \frac{4a[2a + (1+a^2) \cosh \tilde{y}_{24}]}{Z_{24}^2},$$

$$Z_{13} = 1 + a^2 + 2a \cosh \tilde{y}_{13}; \quad Z_{24} = 1 + a^2 + 2a \cosh \tilde{y}_{24},$$

$$a = e^{-\frac{w}{k_B T}}, \quad w = w^0 + \sum_{i=1}^3 \delta_i \varepsilon_i + \sum_{j=4}^6 \delta_j \varepsilon_j.$$

Substituting (3.6), (3.7) into (3.4), (3.5) and excluding parameter Δ_{ft} , we obtained the following differential equations for sums and differences of proton unary distribution functions:

$$\frac{d}{dt} \begin{pmatrix} (\eta_{1t} - \eta_{3t})_1 \\ (\eta_{2t} - \eta_{4t})_1 \end{pmatrix} = \begin{pmatrix} m_{11}^- & -m_{12}^- \\ -m_{21}^- & m_{22}^- \end{pmatrix} \begin{pmatrix} (\eta_{1t} - \eta_{3t})_1 \\ (\eta_{2t} - \eta_{4t})_1 \end{pmatrix} - \beta E_{1t} \begin{pmatrix} m_1 \mu_{13}^x \\ -m_2 \mu_{24}^x \end{pmatrix}, \tag{3.8}$$

$$\frac{d}{dt} \begin{pmatrix} (\eta_{1t} + \eta_{3t})_2 \\ (\eta_{2t} + \eta_{4t})_2 \end{pmatrix} = \begin{pmatrix} m_{11}^+ & -m_{12}^+ \\ -m_{21}^+ & m_{22}^+ \end{pmatrix} \begin{pmatrix} (\eta_{1t} + \eta_{3t})_2 \\ (\eta_{2t} + \eta_{4t})_2 \end{pmatrix} - \beta E_{2t} \begin{pmatrix} m_1 \mu_{13}^y \\ -m_2 \mu_{24}^y \end{pmatrix}, \tag{3.9}$$

$$\frac{d}{dt} \begin{pmatrix} (\eta_{1t} - \eta_{3t})_3 \\ (\eta_{2t} - \eta_{4t})_3 \end{pmatrix} = \begin{pmatrix} m_{11}^- & -m_{12}^- \\ -m_{21}^- & m_{22}^- \end{pmatrix} \begin{pmatrix} (\eta_{1t} - \eta_{3t})_3 \\ (\eta_{2t} - \eta_{4t})_3 \end{pmatrix} - \beta E_{3t} \begin{pmatrix} m_1 \mu_{13}^z \\ m_2 \mu_{24}^z \end{pmatrix}, \tag{3.10}$$

where

$$m_{11}^\pm = \frac{1}{\alpha} (1 - \beta v_1^\pm r_{13} K_{13}), \quad m_{12}^\pm = \frac{1}{\alpha} \left[(1 + K_{13}) P_{13}^{(0)} + \beta v_2^\pm r_{13} K_{13} \right],$$

$$m_{21}^\pm = \frac{1}{\alpha} \left[(1 + K_{24}) P_{24}^{(0)} + \beta v_2^\pm r_{24} K_{24} \right], \quad m_{22}^\pm = \frac{1}{\alpha} (1 - \beta v_3^\pm r_{24} K_{24}),$$

$$m_1 = \frac{1}{\alpha} K_{13} r_{13}, \quad m_2 = \frac{1}{\alpha} K_{24} r_{24},$$

$$K_{13} = \frac{P_{13}^{(1)} \tilde{\eta}_{13} + L_{13}^{(1)}}{2r_{13} - [P_{13}^{(1)} \tilde{\eta}_{13} + L_{13}^{(1)}]}, \quad r_{13} = 1 - (\tilde{\eta}_{13})^2,$$

$$K_{24} = \frac{P_{24}^{(1)} \tilde{\eta}_{24} + L_{24}^{(1)}}{2r_{24} - [P_{24}^{(1)} \tilde{\eta}_{24} + L_{24}^{(1)}]}, \quad r_{24} = 1 - (\tilde{\eta}_{24})^2.$$

Solving the equations (3.8)–(3.10), we obtained time-dependent unary distribution function of protons. The components of dynamic susceptibility of GPI clamped crystal can be written as:

$$\chi_{11}(\omega) = \chi_{11}^0 + \lim_{E_{1t} \rightarrow 0} \frac{1}{\nu} \left[\mu_{13}^x \frac{d(\eta_{1t} - \eta_{3t})_1}{dE_{1t}} - \mu_{24}^x \frac{d(\eta_{2t} - \eta_{4t})_1}{dE_{1t}} \right],$$

$$\chi_{22}(\omega) = \chi_{22}^0 + \lim_{E_{2t} \rightarrow 0} \frac{1}{\nu} \left[\mu_{13}^y \frac{d(\eta_{1t} + \eta_{3t})_2}{dE_{2t}} - \mu_{24}^y \frac{d(\eta_{2t} + \eta_{4t})_2}{dE_{2t}} \right],$$

$$\chi_{33}(\omega) = \chi_{33}^0 + \lim_{E_{3t} \rightarrow 0} \frac{1}{\nu} \left[\mu_{13}^z \frac{d(\eta_{1t} - \eta_{3t})_3}{dE_{3t}} + \mu_{24}^z \frac{d(\eta_{2t} - \eta_{4t})_3}{dE_{3t}} \right].$$

The obtained susceptibilities consist of the “seed” part and two relaxational modes:

$$\chi_{ii}(\omega) = \chi_{ii}^0 + \sum_{l=1}^2 \frac{\chi_l^i}{1 + i\omega\tau_l^i}, \quad i = 1, 2, 3 \rightarrow x, y, z, \tag{3.11}$$

where

$$\chi_l^i = \frac{\beta}{2v} \frac{\tau_1^i \tau_2^i}{\tau_2^i - \tau_1^i} \{(-1)^{l-1} [(\mu_{13}^i)^2 m_1 + (\mu_{24}^i)^2 m_2] + (-1)^l \tau_1^i [(\mu_{13}^i)^2 m_1 m_{22}^\gamma + (\mu_{24}^i)^2 m_2 m_{11}^\gamma - \mu_{13}^i \mu_{24}^i (m_1 m_{21}^\gamma + m_2 m_{12}^\gamma)]\}, \quad (3.12)$$

$\tau_{1,2}^i$ are relaxation times of the following form:

$$(\tau_{1,2}^i)^{-1} = \frac{1}{2} \left[(m_{11}^\gamma + m_{22}^\gamma) \pm \sqrt{(m_{11}^\gamma + m_{22}^\gamma)^2 - 4(m_{11}^\gamma m_{22}^\gamma - m_{12}^\alpha m_{21}^\gamma)} \right]. \quad (3.13)$$

In (3.12), (3.13) $\gamma = "+"$ for $i = y$ and $\gamma = "-"$ for $i = x, z$.

Components of dynamic dielectric permittivity of proton subsystem of GPI is as follows:

$$\varepsilon_{ii}(\omega) = 1 + 4\pi\chi_{ii}(\omega). \quad (3.14)$$

4. Comparison of numerical calculations with the experimental data. Discussion of the obtained results

To calculate the temperature dependence of dielectric, elastic, piezoelectric and thermal characteristics of GPI we need to set certain values of the following parameters:

- parameters of the short-range interactions w^0 ;
- parameters of the long-range interactions $v_f^{0\pm}$ ($f = 1, 2, 3$);
- deformational potentials δ_i, ψ_{fi}^\pm ($f = 1, 2, 3; i = 1, \dots, 6$);
- effective dipole moments $\mu_{13}^a; \mu_{24}^a; \mu_{13}^b; \mu_{24}^b; \mu_{13}^c; \mu_{24}^c$;
- "seed" dielectric susceptibilities χ_{ii}^0 ;
- "seed" coefficients of piezoelectric stress e_{ij}^0 ;
- "seed" elastic constants c_{ij}^{E0} .

The values of the present theory parameters are determined while studying the static properties of GPI [5]. The optimal values of long-range interactions $v_f^{0\pm}$ are as follows: $\tilde{v}_1^{0+} = \tilde{v}_2^{0+} = \tilde{v}_3^{0+} = 2.643$ K, $\tilde{v}_1^{0-} = \tilde{v}_2^{0-} = \tilde{v}_3^{0-} = 0.2$ K, where $\tilde{v}_f^{0\pm} = v_f^{0\pm}/k_B$. The determined parameter w^0 of the GPI crystal is $w^0/k_B = 820$ K. The optimal values of the deformational potentials δ_i are $\tilde{\delta}_1 = 500$ K, $\tilde{\delta}_2 = 600$ K, $\tilde{\delta}_3 = 500$ K, $\tilde{\delta}_4 = 150$ K, $\tilde{\delta}_5 = 100$ K, $\tilde{\delta}_6 = 150$ K; $\tilde{\delta}_i = \delta_i/k_B$. The optimal values of the ψ_{fi}^\pm are as follows: $\tilde{\psi}_{f1}^+ = 87.9$ K, $\tilde{\psi}_{f2}^+ = 237.0$ K, $\tilde{\psi}_{f3}^+ = 103.8$ K, $\tilde{\psi}_{f4}^+ = 149.1$ K, $\tilde{\psi}_{f5}^+ = 21.3$ K, $\tilde{\psi}_{f6}^+ = 143.8$ K, $\tilde{\psi}_{fi}^- = 0$ K, where $\tilde{\psi}_{fi}^\pm = \psi_{fi}^\pm/k_B$. The effective dipole moments in the paraelectric phase are equal to $\mu_{13}^x = 0.4 \cdot 10^{-18}$ esu-cm; $\mu_{13}^y = 4.02 \cdot 10^{-18}$ esu-cm; $\mu_{13}^z = 4.3 \cdot 10^{-18}$ esu-cm; $\mu_{24}^x = 2.3 \cdot 10^{-18}$ esu-cm; $\mu_{24}^y = 3.0 \cdot 10^{-18}$ esu-cm; $\mu_{24}^z = 2.2 \cdot 10^{-18}$ esu-cm. In the ferroelectric phase, the y -component of the first dipole moment is $\mu_{13\text{ferro}}^y = 3.82 \cdot 10^{-18}$ esu-cm.

In [9] the transition temperature is $T_c = 223.6$ K, and one should multiply the parameters $w^0, v_f^{0\pm}, \delta_i, \psi_{fi}^\pm, \mu_{13}^i, \mu_{24}^i$ by the coefficient 0.994.

The volume of a primitive cell of GPI is $v = 0.601 \cdot 10^{-21}$ cm³.

Parameter α is determined from the condition of an agreement of theoretically calculated and experimentally obtained frequency dependences of $\varepsilon_{22}(\omega)$. We consider that parameter α slightly changes with temperature:

$$\alpha = [1.6 - 0.011(\Delta T)] \cdot 10^{-14} \text{ s}, \quad \Delta T = T - T_c.$$

The “seed” coefficients of piezoelectric stress, dielectric susceptibilities and elastic constants are as follows:

$$e_{ij}^0 = 0.0 \frac{\text{esu}}{\text{cm}^2}; \quad \chi_{11}^0 = 0.1, \quad \chi_{22}^0 = 0.403, \quad \chi_{33}^0 = 0.5;$$

$$c_{11}^{0E} = 26.91 \cdot 10^{10} \frac{\text{dyn}}{\text{cm}^2}, \quad c_{12}^{E0} = 14.5 \cdot 10^{10} \frac{\text{dyn}}{\text{cm}^2}, \quad c_{13}^{E0} = 11.64 \cdot 10^{10} \frac{\text{dyn}}{\text{cm}^2}, \quad c_{15}^{E0} = 3.91 \cdot 10^{10} \frac{\text{dyn}}{\text{cm}^2},$$

$$c_{22}^{E0} = [64.99 - 0.04(T - T_c)] \cdot 10^{10} \frac{\text{dyn}}{\text{cm}^2}, \quad c_{23}^{E0} = 20.38 \cdot 10^{10} \frac{\text{dyn}}{\text{cm}^2}, \quad c_{25}^{E0} = 5.64 \cdot 10^{10} \frac{\text{dyn}}{\text{cm}^2},$$

$$c_{33}^{E0} = 24.41 \cdot 10^{10} \frac{\text{dyn}}{\text{cm}^2}, \quad c_{35}^{E0} = -2.84 \cdot 10^{10} \frac{\text{dyn}}{\text{cm}^2}, \quad c_{55}^{E0} = 8.54 \cdot 10^{10} \frac{\text{dyn}}{\text{cm}^2},$$

$$c_{44}^{E0} = 15.31 \cdot 10^{10} \frac{\text{dyn}}{\text{cm}^2}, \quad c_{46}^{E0} = -1.1 \cdot 10^{10} \frac{\text{dyn}}{\text{cm}^2}, \quad c_{66}^{E0} = 11.88 \cdot 10^{10} \frac{\text{dyn}}{\text{cm}^2}.$$

Other components $c_{ij}^{E0} \equiv 0$.

From expression (3.11) we can see that there are two contributions into the components of dielectric permittivity tensor of GPI. Numerical analysis shows that only one contribution to the permittivities is determinative ($\chi_2^i \gg \chi_1^i$).

Let us first consider the longitudinal dynamic dielectric characteristics. They are predetermined by the behaviour of static dielectric characteristics χ_1^y, χ_2^y and relaxation times τ_1^y, τ_2^y in the system. Relaxation time τ_2^y is connected with some relaxation frequency (soft relaxation mode) typical of this crystal $\nu_s = (2\pi\tau_2^y)^{-1}$, which conventionally separates the regions of low-frequency and high-frequency dispersion. In figure 2 there are presented temperature dependences of the relaxation frequencies ν_s^y , taken from [9–11], and the calculated temperature dependences of the longitudinal relaxation times $\tau_2^y = (2\pi\nu_s^y)^{-1}$ [9, 10]. Relaxation frequency, taken from [11], greatly differs from the frequencies taken from [9, 10]. One can see from these figures that theoretical results satisfactorily agree with experimental data [9, 10], except the phase transition region. Relaxation frequency ν_s^y decreases at approaching to the phase transition temperature and tend to zero at the temperature $T = T_c$. The calculated relaxation time τ_2^y has a singularity at $T = T_c$, but experimental values of τ_2^y are finite at this temperature.

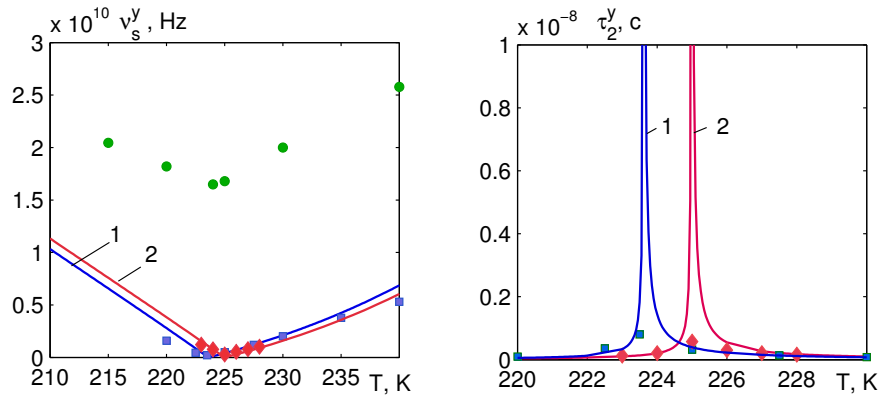


Figure 2. (Colour online) The temperature dependence of relaxation frequency ν_s^y : 1, ■ — [9]; 2, ◆ — [10]; ● — [11] and relaxation time τ_2^y : 1, ■ — [9]; 2, ◆ — [10].

At the frequencies $\nu \ll \nu_s^y$ the real part of the dynamic dielectric permittivity ε'_{22} behaves as static, but the imaginary part ε''_{22} is close to zero at all temperatures excepting the narrow region near T_c . One can see this on the frequency dependences $\varepsilon_{22}(\nu)$ at different $\Delta T = T - T_c$ in the frequency region $\nu < 10^7$ Hz (figure 3), as well as on the temperature dependences $\varepsilon_{22}(T)$ at low frequencies (10^4 Hz, 10^5 Hz, 10^6 Hz) (figure 4).

At the frequencies $\nu \approx \nu_s$ we observe a relaxation dispersion, which reveals itself in the steep decreasing of the real part of dielectric permittivity ε'_{22} with an increasing frequency and in the large values of imaginary part ε''_{22} ; the peak of ε''_{22} corresponds to the frequency ν_s . One can see it on the frequency dependences $\varepsilon_{22}(\nu)$ at different $\Delta T = T - T_c$ in the frequency region $10^7 < \nu < 10^{10}$ Hz (figure 3), as well as on the temperature dependences $\varepsilon_{22}(T)$ at the frequencies 1 MHz–27000 MHz (figure 5).

At the frequencies $\nu \gg \nu_s$, the dielectric permittivity behaves as a purely lattice contribution. It corresponds to the frequency region $\nu > 10^{10}$ Hz on the frequency dependences $\varepsilon_{22}(\nu)$ in figure 3.

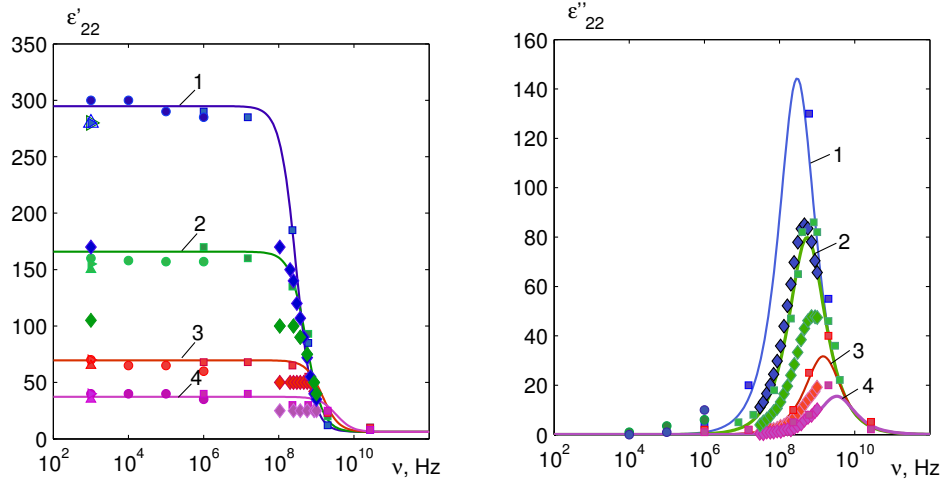


Figure 3. (Colour online) The frequency dependences of real ϵ'_{22} and imaginary ϵ''_{22} parts of dielectric permittivity of GPI at different ΔT (K): 1.0 — 1; 2.0 — 2; 5.0 — 3; 10.0 — 4; ● [8]; ■ [9]; ◆ [10]; ► [14]; ▲ [15].

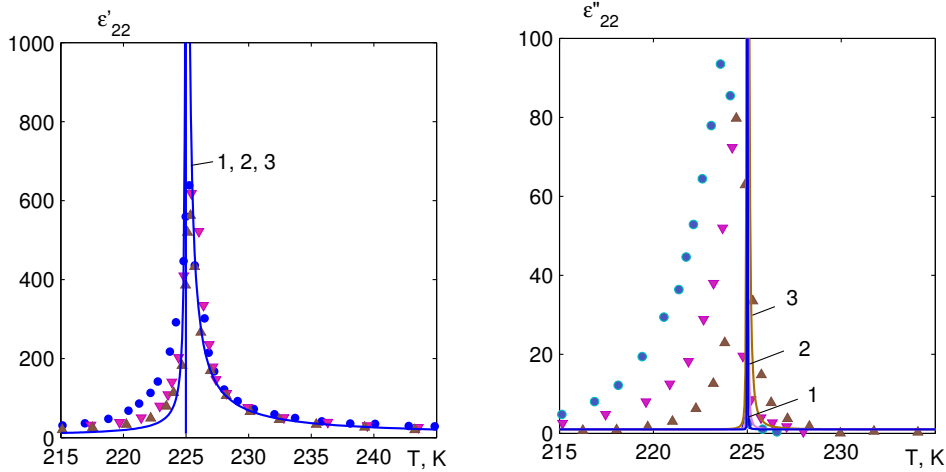


Figure 4. (Colour online) Temperature dependences of ϵ'_{22} and ϵ''_{22} of GPI at different frequencies ν (MHz): 0.01 — 1, ● [8]; 0.1 — 2, ▼ [8]; 1.0 — 3, ▲ [8].

An increase of the relaxation time τ_2^y and a decrease of the relaxation frequency ν_s^y at approaching the temperature $T = T_c$ manifests itself in the shift of the region of dispersion to lower frequencies in the frequency dependence $\epsilon_{22}(\nu)$ (figure 3) at approaching the temperature $T = T_c$, as well as in the availability of depression near $T = T_c$ on the temperature dependence $\epsilon'_{22}(T)$, and of a sharp peak on the curve $\epsilon''_{22}(T)$ (figures 4, 5). Since $\nu_s^y \rightarrow 0$ at $T = T_c$, then a depression of $\epsilon'_{22}(T)$ and a peak of $\epsilon''_{22}(T)$ appears at all frequencies; they are very narrow at low frequencies and widen with an increase of frequency. The value of permittivity in the minimum point (at $T = T_c$) is equal to the lattice contribution ϵ_{22}^0 . Since the experimental value is $\nu_s^y \neq 0$ at $T = T_c$, one can observe a low-frequency maximum in the experimental temperature dependence $\epsilon'_{22}(\nu, T)$ at low frequencies. Starting from frequency $\nu_s \approx 10^7$, a depression-minimum appears instead of a maximum of $\epsilon'_{22}(\nu, T)$, and this minimum decreases with an increase of frequency.

From figures 3–5 one can see that the proposed theoretical model satisfactorily describes the experimental data for the frequency and temperature dependences $\epsilon'_{22}(\nu, T)$ and $\epsilon''_{22}(\nu, T)$ of GPI crystal in the paraelectric phase, with the exception of [10], and less satisfactorily in the ferroelectric phase.

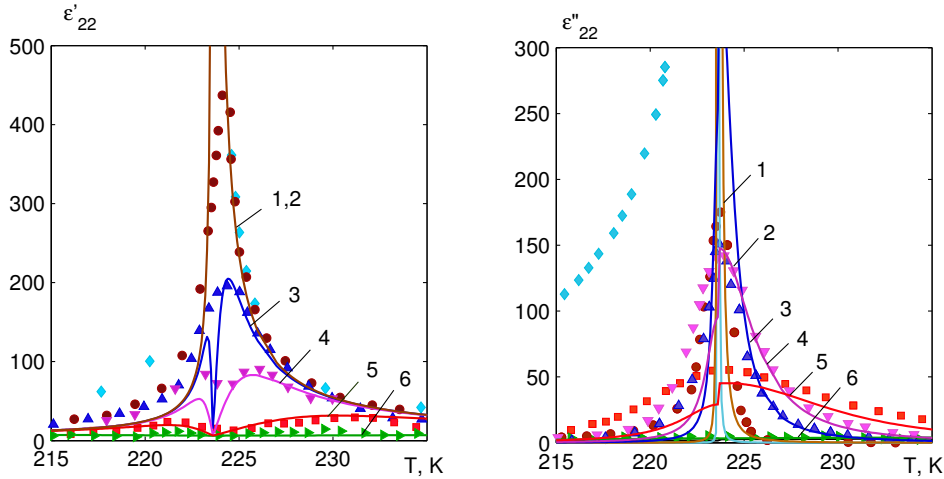


Figure 5. (Colour online) Temperature dependences of ε'_{33} and ε''_{33} of GPI crystal for various frequencies ν (MHz): 1.0 — 1, \blacklozenge [9]; 15.0 — 2, \bullet [9]; 230.0 — 3, \blacktriangle [9]; 610 — 4, \blacktriangledown [9]; 2000 — 5, \blacksquare [9]; 27000 — 6, \blacktriangleright [9].

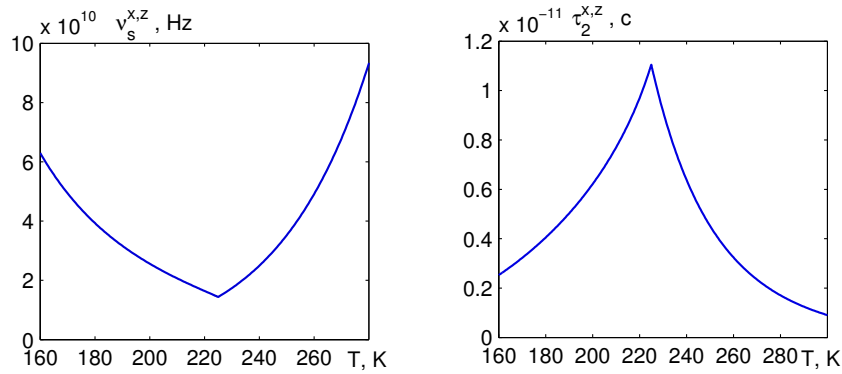


Figure 6. (Colour online) The temperature dependences of relaxation frequencies $\nu_s^{x,z}$ and relaxation times $\tau_2^{x,z}$.

A disagreement of the theoretical curves with the experimental data in the low-frequency region in the ferroelectric phase is connected with an essential role of domain processes in this region [16], which are not taken into account in the proposed theory.

Let us discuss the transverse dynamic characteristics. Transverse relaxation frequencies $\nu_s^{x,z}$ and transverse relaxation times τ_2^x and τ_2^z are calculated at the same α as longitudinal ν_s^y and τ_2^y . The frequencies $\nu_s^{x,z}$ are higher than ν_s^y and they also decrease at approaching the phase transition temperature (figure 6), and take on a nonzero value at $T = T_c$. The transverse relaxation times $\tau_2^{x,z}$ in contrast to τ_2^y are finite at $T = T_c$. This results in the frequency dependences of $\varepsilon_{11}(\nu)$ (figure 7) and $\varepsilon_{33}(\nu)$ (figure 8) at different ΔT that are qualitatively similar to the frequency dependences of $\varepsilon_{22}(\nu)$, but the region of dispersion exists at higher frequencies and at weaker changes with temperature.

However, in the temperature dependences of ε'_{11} and ε'_{33} , only the angle of the curve fracture in the point T_c changes (figures 9, 10) instead of a depression near the phase transition temperature. The maximum value of $\varepsilon'_{11,33}(T, \nu)$ at $T = T_c$ decreases with an increase of frequency. Values of $\varepsilon'_{11,33}(T, \nu)$ at $T = T_c$ increase with an increase of frequency up to $1.5 \cdot 10^{10}$ Hz. At higher frequencies, the maximum values of $\varepsilon'_{11,33}(T, \nu)$ decrease and shift to the region of higher temperatures. Experimental investigations of transverse dynamic characteristics of GPI are very important to verify the obtained theoretical results in this regard. It is necessary to note that experimental data in figures 9 and 10 are measured at frequency 1 kHz. They are close to static permittivities at such a small frequency.

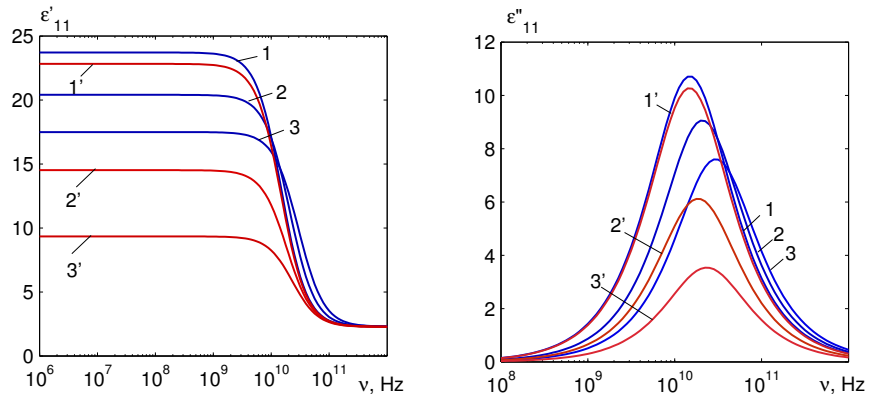


Figure 7. (Colour online) Dispersion of real ϵ'_{11} and imaginary ϵ''_{11} parts of dielectric permittivity of GPI at different ΔT (K): 1 — 1; 10 — 2; 20 — 3; -1 — 1'; -5 — 2'; -10 — 3'.

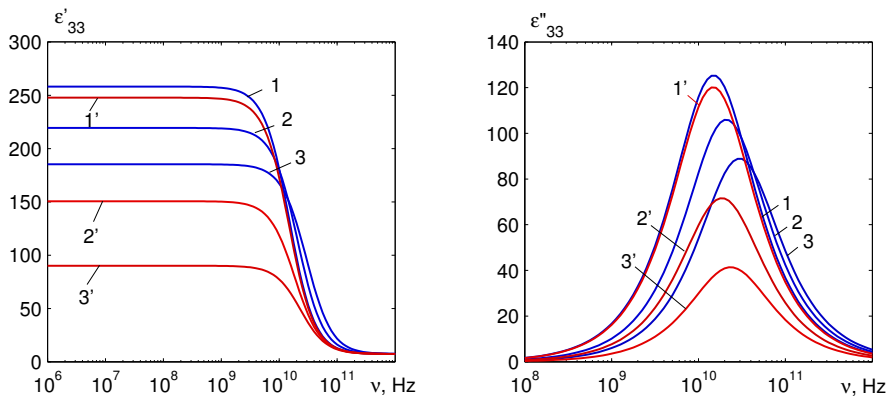


Figure 8. (Colour online) Dispersion of real ϵ'_{33} and imaginary ϵ''_{33} parts of dielectric permittivity of GPI at different ΔT (K): 1 — 1; 10 — 2; 20 — 3; -5 — 2'; -10 — 3'.

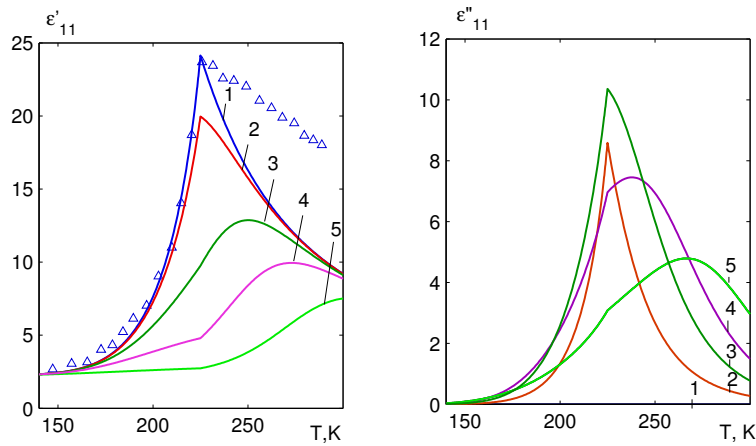


Figure 9. (Colour online) Temperature dependences of ϵ'_{11} and ϵ''_{11} for GPI crystal for various frequencies ν (GHz): 0.0 — 1, Δ [15] (1 kHz); 7 — 2; 20 — 3; 40 — 4; 100 — 5.

The results of calculation of Cole-Cole curves (figure 11) witness for monodispersivity of dielectric permittivity in the crystals studied. The results of measurements of Cole-Cole curves for the longitudinal permittivity, presented in [9–11], disagree with each other. The calculated curves well agree with the results of [9] for longitudinal permittivity.

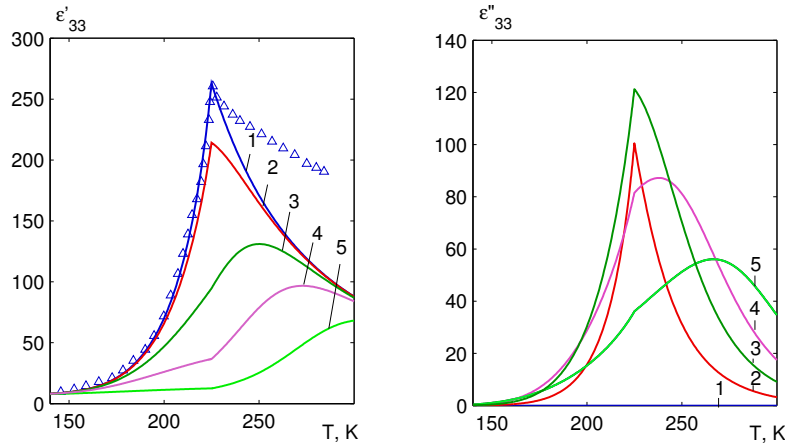


Figure 10. (Colour online) Temperature dependences of ϵ'_{33} and ϵ''_{33} for GPI crystal for various frequencies ν (GHz): 0.0 — 1, Δ [15] (1 kHz); 7 — 2; 20 — 3; 40 — 4; 100 — 5.

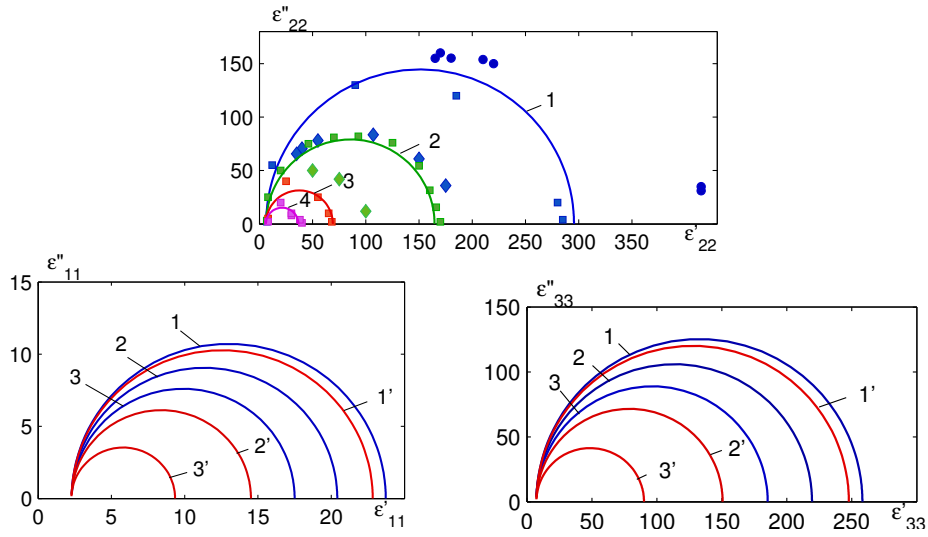


Figure 11. (Colour online) Cole-Cole (22) plot for GPI crystal at ΔT (K): 1 — 1, \blacksquare [9]; \blacklozenge [10]; \bullet [11]; 2 — 2, \blacksquare [9]; \blacklozenge [10]; 5 — 3, \blacksquare [9]; \blacklozenge [10]; 10 — 4, \blacksquare [9] and Cole-Cole (11) and Cole-Cole (33) plot at different ΔT (K): 1 — 1; 10 — 2; 20 — 3; -1 — 1'; -10 — 2'; -20 — 3'.

5. Conclusions

Using the modified GPI model, the components of dynamic dielectric permittivity tensor and relaxation times are calculated in a two-particle cluster approximation. A satisfactory agreement of the theoretical results with experimental data for longitudinal permittivity is obtained, with the exception of low-frequency region in the ordered phase, inasmuch as the proposed theory does not take the domain processes into account, which can give a contribution into the above mentioned frequency region.

It is determined that the dynamic dielectric permittivity at low frequencies behaves as static; at the frequencies comparable with an inverse relaxation time, a relaxational dispersion is observed; at high frequencies, only a lattice contribution to permittivity reveals itself. The region of longitudinal dispersion in GPI shifts to the low frequencies at temperature approaching the phase transition point, which is connected with a considerable increase of relaxation time at approaching the temperature T_c . The region of transverse dispersion lies at higher frequencies and weakly depends on temperature.

The obtained results for transverse characteristics bear the character of predictions and can be a stimulus for further experimental investigations.

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Динамічні властивості сегнетоелектрика $\text{NH}_3\text{CH}_2\text{COOH}\cdot\text{H}_2\text{PO}_3$

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Використовуючи модифіковану псевдоспінову модель сегнетоелектрика $\text{NH}_3\text{CH}_2\text{COOH}\cdot\text{H}_2\text{PO}_3$ шляхом врахування п'єзоелектричного зв'язку з деформаціями ε_i , ε_4 , ε_5 , ε_6 в рамках методу Глаубера в наближенні двочастинкового кластера розраховано для неї компоненти тензора комплексної діелектричної проникності і часи релаксації. При належному виборі параметрів теорії вивчено частотні та температурні залежності компонент сприйнятливості та температурні залежності часів релаксації. Отримано задовільну згоду теоретичних результатів з експериментальними даними для поздовжньої проникності.

Ключові слова: сегнетоелектрики, кластерне наближення, динамічна діелектрична проникність, час релаксації
