UDC 537.266

DISPERSION OF DIELECTRIC PERMEABILITY IN POLYDOMAIN FERROELECTRICS

© 2002 г. A. S. Sidorkin, A. S. Sigov

Voronezh State University
Moscow Institute of Radioelectronic Engineering

The parameters of the equation of the domain wall motion for its translational movement have been calculated: the effective mass of the wall connected with involving in motion of the bulk of ferroelectric through the piezoeffect, the coefficient of the quasielastic force effecting on the wall and connected with the change of charging state on the surface of the material at the displacement of domain boundary and the effective resistance to the motion of domain boundary in crystal with defects. On the basis of the equation of motion for the domain wall the value of its shift is determined in dependence on the amplitude and frequency of external electrical field. The contribution of domain boundaries into the dielectric permeability and the frequency dispersion of the latter are calculated. The obtained results are compared with the experimental data. It has been shown, that the dispersion of the dielectric permeability ε in KH_2PO_4 crystal at the frequencies $10^7 \div 10^8$ Hz can be explained by inertial properties of domain boundaries. Rather low-frequency dispersion ε at the frequencies $\sim 10^3$ Hz in γ-irradiated crystals KH_2PO_4 is due to the decrease of mobility of the domain boundaries in crystals with defects.

According to numerous experimental data the dispersion of dielectric permeability and tangent of the dielectric losses angle in ferroelectrics besides the so-called soft mode [1] is connected with the motion of domain boundaries [2]. Dispersion of a resonant type observed in ferroelectrics with perovskite-type structure in the megaherz range of frequencies [3], and dispersion of relaxation type noted in the kiloherz range in numerous crystals of KH_2PO_4 group, exposed to γ- or electron irradiation [4] is also related to the same class of phenomena.

For theoretical description of the above mentioned dispersion the study of the motion regularities of domain boundaries in a wide range of frequencies is necessary. They depend on the inertial properties of the boundaries [5] as on the materials themselves [6, 7], on the forces arising at the displacement of domain boundaries from their equilibrum positions [8—10], and, finally, on the mobility of the boundary determined by dissipative processes in the ideal material [11, 12] as well as by the interaction of the moving boundary with defects of the crystal lattice [13—15]. A particular character of the motion of domain boundaries depends on the relative role of terms in the equation of motion of the domain wall.

Let's specify the physical reasons of appearance of different terms in the equation of motion of the domain wall. In the initial state in the absence of external field the charges on the surface of ferroelectric are compensated as a rule at the expense of the bulk or surface conductivity. The applying of external electric field results in appearance of the pressure on the domain walls. Being displaced under this pressure the domain boundaries break the balance of charges on the surface of ferroelectric. It results in the increase of electrostatic energy of the crystal and, hence, the appearance of quasielastic returning force effecting on the displaced boundary. In the crystal of ferroelectric-ferroelastic and also in the case of ferroelectric with 90-degree domain boundaries besides of the electrical returning force there also arises a returning force of the elastic nature related to the appearance of the elastic stresses under displacement of domain boundaries in the place of contact of ferroelectric with the nonferroelectric layer [9, 10].

Any domain wall has its local effective mass connected with the inertial properties of the particles, composing the wall, since due to their motion the vector of polarization is overturned in the region of the moving boundary. Besides [7], even in the process of translational motion of domain walls there appears also their effective nonlocal mass. It occurs due to involving in the motion through piezoeffect of the whole layer of the ferroelectric. The source of the corresponding fields is the above mentioned non-compensated charges on the surface of the ferroelectric. The thickness of the layer involved in motion and, hence, the value of nonlocal effective mass depend on the wave length of translational displacements. For ultimately long-wave shifts all the bulk of ferroelectric can be considered as such a layer.

The presence of defects of the crystal lattice interacting with the domain wall, depending on the mobility of such defects is the reason, in a general case, of appearance of additional as quasielastic as dissipative terms in the equation of the motion of domain wall.

Let's formulate now the equation of motion of the domain wall in ferroelectric in an external field. For definiteness we shall begin the consideration with the case of pure ferroelectric with 180-degree domain structure. As the latter one we shall choose an infinite ferroelectric plate with thickness of L_x along the polar axis. We shall place the origin of coordinate system into the middle of its thickness and choose it as coinciding with one of the walls along Z axis. Let's begin with the case of a non-defect crystal. Taking into account all the above mentioned facts, the equation of motion of the domain wall in an external field E here takes the following form:

$$(m_0 + m_1)\ddot{U} + \mathcal{K} U = 2P_0 E\,. \tag{1}$$

Here m_0 and m_1 are local and nonlocal effective mass of the domain wall, respectively. $\mathcal{K}$ is the coefficient of the quasielastic force effecting on the displaced domain wall, $2P_0E$ is the pressure on this wall from the external field. Let's note, that the equation (1) will be of the same form for all the domain walls since the field E varying in time, will be identical at any moment of time for all the points of the sample.

In order to calculate the coefficient of the quasielastic force K the equation (1) it should be completed by the equation of electrostatics

$$\frac{\partial D_i}{\partial x_i} = 0\,, \tag{2}$$

the equation of motion of the elastic medium

$$\rho\,\ddot{u}_{ij} = \frac{\partial \sigma_{ij}}{\partial x_j} \tag{3}$$

and by the corresponding material equations in a crystal with piezoeffect:

$$\begin{cases}\sigma_{ij} = c_{ijkl}u_{kl} + \beta_{kij}E_k,\\ D_i = \varepsilon_{ij}E_{ij} + 4\pi P_{0i} - 4\pi\beta_{ijk}u_{jk}.\end{cases} \tag{4}$$

Here D_i is the vector of electrostatic induction, P_{0i} and E_i are vectors of spontaneous polarization and strength of the electrical field, u_i is the vector of elastic shift of the medium points, u_{kl} and σ_{ij} are tensors of deformation and elastic stresses, respectively, c_{ilkl}, β_{ijk}, ε_{ij} are tensors of modules of elasticity, piezocoefficients and dielectric permeability, ρ is the density of medium.

In order to calculate the coefficient $\mathcal{K}$ and mass m_1 we shall calculate the electrical field arising at the displacement of domain boundaries. For the considered small displacements of domain walls $U \ll d$ (d — average width of the domain) the source of the indicated fields is the charges arising with the surface density

$$\sigma_1(z) = \sum_n \gamma_n \delta(z - nd) \tag{5}$$

on the surfaces of ferroelectric, where $\gamma_n = 2P_0U_n$ at $x = L_x/2$ and correspondingly is equal to $-2P_0U_n$ at $x = -L_x/2$; n — is the number of the domain wall. Substituting the relationship (4) into (2) and (3) with the account of expressions for the tensor of deformation and of the connection between the strength of the electrostatic field and its potential for determination of the field arising at the displacement of the one domain wall we shall obtain the following system of the equations [16]:

$$\begin{cases}-\varepsilon_{ij}\dfrac{\partial^2\varphi}{\partial x_i \partial x_j} - 4\pi\beta_{ijk}\dfrac{\partial^2 u_j}{\partial x_k \partial x_i} = 4\pi\gamma_n\delta(x)\delta(z),\\[2ex] \rho\ddot{u}_i = c_{ijkl}\dfrac{\partial^2 u_l}{\partial x_l \partial x_j} - \beta_{kij}\dfrac{\partial^2\varphi}{\partial x_k \partial x_j}.\end{cases} \tag{6}$$

Let's present $\varphi(x,z)$ and $u_i(x,z)$ as a Fourier-expansions

$$\varphi(x,z) = \int \varphi_z(t)\cdot e^{-ik_x x}\cdot e^{-ik_z z}\,\frac{dk_x dk_z}{(2\pi)^2},$$

$$u_i = \int u_{i\bar{k}}(t)\cdot e^{-ik_x x}\cdot e^{-ik_z z}\,\frac{dk_x dk_z}{(2\pi)^2}. \tag{8}$$

For elastically-isotropic material the equation of dynamics for the elastic medium looks as following

$$\rho \ddot{u}_i = (\lambda + \mu)\frac{\partial^2 u_l}{\partial x_l \partial x_i} + \mu \frac{\partial^2 u_i}{\partial x_k^2} - \beta_{kij}\frac{\partial^2 \varphi}{\partial x_k \partial x_j}, \quad (9)$$

where λ and μ are Lame coefficients. Determining with the account of (9)

$$div\vec{u} = -\frac{\partial \Lambda_i/\partial x_i}{\rho(c_l^2 \tilde{k}^2 - \omega^2)}, \tilde{k}^2 = k_x^2 + k_z^2, \quad (10)$$

where $\Lambda_i = \beta_{kij}\partial^2\varphi/\partial x_k \partial x_j$ and $c_1 = \sqrt{(\lambda + 2\mu)/\rho}$ is the velosity of longitudinal sound wave we obtain instead of (9)

$$\rho \ddot{u}_i = -\frac{(\lambda+\mu)\partial^2 \Lambda_l/\partial x_l \partial x_i}{\rho(c_l^2\tilde{k}^2 - \omega^2)} + \mu\frac{\partial^2 u_i}{\partial x_k^2} - \Lambda_i \quad (11)$$

The substitution in (11) and in the equation of electrostatics in (7) of Fourier-expansions (8) gives us now the system of equations for determination of Fourier-coefficients $u_{i\tilde{k}}$ and $\varphi_{\tilde{k}}$

$$\begin{cases} u_{ik} = \frac{1}{(c_l^2\tilde{k}^2 - \omega^2)}\left[\beta_{kij}k_k k_j - \frac{(c_l^2 - c_t^2)}{(c_l^2\tilde{k}^2 - \omega^2)}\beta_{klj}k_k k_j k_l k_i\right]\varphi_{\tilde{k}} \\ \varepsilon_{ij}k_i k_j \varphi_{\tilde{k}} + 4\pi\beta_{ijk}k_k k_i u_{j\tilde{k}} = 4\pi\gamma \end{cases} \quad (12)$$

($c_t = \sqrt{\mu/\rho}$ — is the velocity of the transvers sound wave). Then

$$\varphi_{\tilde{k}}(t) = 4\pi\gamma_n\left\{\varepsilon_{ij}k_i k_j + \frac{4\pi\beta_{ijk}k_k k_i}{\rho(c_t^2\tilde{k}^2 - \omega^2)}\times \right.$$
$$\left.\times\left[\beta_{pjm}k_p k_m - \frac{(c_l^2 - c_t^2)}{(c_l^2\tilde{k}^2 - \omega^2)}\beta_{plm}k_p k_j k_l k_m\right]\right\}^{-1}. \quad (13)$$

The expression (13) contains the contributions as into the coefficient of quasielastic force $\mathcal{K}$, as into effective mass of the domain wall m_1 arising by means of piezoelectric interactions. For their determination we shall expand $\varphi_{\tilde{k}}$ in a sieries over ω^2. Thus, with respect of static contribution into $\varphi_{\tilde{k}}$ we shall take into account that in a view of real relationship $4\pi\beta^2/\rho c^2\varepsilon \ll 1$ the contribution of the piezoeffect into $\varphi_{\tilde{k}}(\omega = 0)$ can be neglected. As a result we shall obtain

$$\varphi_{\tilde{k}} = \varphi_{\tilde{k}}(\omega = 0) + \left.\frac{\partial\varphi_{\tilde{k}}}{\partial\omega^2}\right|_0 \cdot \omega^2 + \ldots \quad (14)$$

where $\varphi_{\tilde{k}}(\omega = 0) = 4\pi\gamma/\varepsilon_{ij}k_i k_j$,

$$\left.\frac{\partial\varphi_{\tilde{k}}}{\partial\omega^2}\right|_0 = \frac{4\pi\gamma_n}{(\varepsilon_{ij}k_i k_j)^2}\cdot\left\{\frac{4\pi\beta_{ijk}\beta_{pjm}k_k k_i k_p k_{jm}}{\rho c_t^4\tilde{k}^4}\right.$$
$$\left. - \frac{(c_l^4 - c_t^4)}{c_t^4 c_l^4}\cdot\frac{4\pi\beta_{ijk}\beta_{plm}k_k k_i k_p k_j k_l k_m}{\rho\tilde{k}^6}\right\}. \quad (15)$$

The bulk density of the charge corresponding to (5) and distributed on both surfaces of the ferroelectric plate is

$$\rho(x,z) = \sum_n \gamma_n \delta(z - nd)\left[\delta\left(x - \frac{L_x}{2}\right) - \delta\left(x + \frac{L_x}{2}\right)\right]. \quad (16)$$

According to properties of the Green's function the potential of these charges

$$\varphi_1(x,z) = \int \rho_{\tilde{k}}\varphi_{\tilde{k}}e^{-i\vec{k}\vec{\rho}}\frac{d\vec{k}}{(2\pi)^2} =$$
$$= \sum_{n'}\gamma_{n'}\int\varphi_{\tilde{k}}e^{-i\vec{k}_z(z-n'd)}[e^{-ik_x(x-\frac{L_x}{2})} - e^{-ik_x(x+\frac{L_x}{2})}]\frac{dk_x dk_z}{(2\pi)^2}, \quad (17)$$

where $\varphi_{\tilde{k}} = \varphi_{\tilde{k}}/\gamma_n, \gamma_n = 2P_0U_n$ and $\gamma_{n'} = 2P_0U_{n'}(-1)^{n-n'}$. Appearance in the last expression of the factor $(-1)^{n-n'}$ takes into account that at the identical signs of U_n and $U_{n'}$ the charge arising on the surface of the crystal near the neighbouring domain walls has the opposite sign.

The energy of interaction of charges (16) between themselves with the account of (5) and (17) in a general case is

$$\mathcal{F} = L_y\int\sigma_1(x = \tfrac{L_x}{2}, z)\cdot\varphi_1(x = \tfrac{L_x}{2}, z)dz =$$
$$= L_y\sum_{n,n'}\gamma_n\gamma_{n'}\cdot\varphi_{\tilde{k}}\cdot e^{-ik_z d(n-n')}[1 - e^{-ik_x L_x}]\frac{dk_x dk_z}{(2\pi)^2}. \quad (18)$$

While calculating the sum over n and n' in (18) in the case of the displacement of the domain boundaries in external field it is necessary to take into account the following. In this case the neighbouring domain walls are displaced towards each other by equal distances. Therefore, while calculating here all the summands except for $n = n'$ the coefficient $(-1)^{n-n'}$ in $\gamma_{n'}$ should be rejected and we should take all the terms with one and the same sign. Expression (18) can not be used for the case of $n = n'$, because it does not allow to separate the self-interaction effect of the charges arising in the field of the displaced domain wall. In this case it is necessary to take into account that the contribution in $\mathcal{F}$ corresponding to the displacement of only one domain wall is equivalent to the displacement of all the walls except the given one by the same distances in an opposite direction. Thus the contribution in $\mathcal{F}$ at $n = n'$ is obtained by summation in (18) of all the terms with alternating (with the account of the multiplier $(-1)^{n-n'}$) signs. As a result, the total contribution into $\mathcal{F}$ corresponding to the displacement of all the domain walls can be obtained under merging of the mentioned contributions where the odd terms are doubled while the even ones are neglected. Thus, with the account of the fact that all $\gamma_n \equiv \gamma = 2P_0U$ we have

$$\mathcal{F} = 16P_0^2U^2NL_y\sum_{n=1}^{\infty}\varphi_{\tilde{k}}e^{-ik_zd(2n-1)}[1-e^{ik_xL_x}]\frac{dk_xdk_z}{(2\pi)^2}, \quad (13)$$

where N is the real number of domain walls in the material of the ferroelectric plate. The substitution of the expression for $\varphi_{\tilde{k}}$ (14) in (19) allows to write down $\mathcal{F}$ as

$$\mathcal{F} = NL_yL_x\left\{\frac{\mathcal{K}U^2}{2}+\frac{m_1\omega^2U^2}{2}\right\}, \quad (20)$$

where the effective coefficient of quasielastic force $\mathcal{K}$ effecting on some domain wall, and its effective mass m_1 are equal, respectively, to:

$$\mathcal{K} = \frac{32P_0^2}{L_x}\sum_{n=1}^{\infty}\int\frac{4\pi}{\varepsilon_{ij}k_ik_j}e^{-ik_zd(2n-1)}[1-e^{-ik_xL_x}]\frac{dk_xdk_z}{(2\pi)^2}, \quad (21)$$

$$m_1 = \frac{32P_0^2}{L_x}\sum_{n=1}^{\infty}\int\frac{4\pi}{(\varepsilon_{ij}k_ik_j)^2}\left\{\frac{(c_l^4-c_t^4)}{c_t^4c_l^4}\times\right.$$
$$\left.\times\frac{4\pi\beta_{ijk}\beta_{plj}k_kk_ik_pk_jk_ek_m}{\rho\tilde{k}^6}-\frac{4\pi\beta_{ijk}\beta_{pjm}k_kk_ik_pk_m}{\rho c_t^4\tilde{k}^4}\right\}\times \quad (22)$$
$$\times e^{-ik_zd(2n-1)}[1-e^{-ik_xL_x}]\frac{dk_xdk_z}{(2\pi)^2},$$

Here, as usual, the summation by the repeating indexes is assumed and $k_y = 0$ with the account of symmetry of the task.

In the particular case of tetragonal symmetry of the polar phase [17]

$$\varepsilon_{ij} = \begin{pmatrix}\varepsilon_c & 0 & 0\\ 0 & \varepsilon_a & 0\\ 0 & 0 & \varepsilon_a\end{pmatrix} \quad (23)$$

the calculations of the coefficient $\mathcal{K}$ provides the following expression

$$\mathcal{K} = \frac{32P_0^2}{\sqrt{\varepsilon_a\varepsilon_c}}\cdot\frac{1}{L_x}\sum_{n=1}^{\infty}\ln[1+\frac{\varepsilon_a}{\varepsilon_c}\frac{L_x^2}{d^2}\frac{1}{(2n-1)^2}] =$$
$$= \frac{32P_0^2}{L_x\sqrt{\varepsilon_a\varepsilon_c}}\cdot\ln ch(\frac{\pi}{2}\sqrt{\frac{\varepsilon_a}{\varepsilon_c}}\frac{L_x}{d}). \quad (24)$$

In this case the matrix of piezomodules $\beta_{imk} = \beta_{ikm}$ has the following non-equal to zero coefficients $\beta_{111} \equiv \beta_1, \beta_{122} = \beta_{133} \equiv \beta_2, \beta_{221} = \beta_{313} \equiv \beta_3$ [17]. The estimation of the value of m_1 gives [18] here

$$m_1 = \frac{16\pi^2P_0^2\beta^2}{\varepsilon_c^2\rho c_t^4}\cdot\frac{L_x^2}{d} \quad (25)$$

Note, that in the case of the crystal of ferroelectric–ferroelastic and also for ferroelectric with 90-degree domains in addition to the introduced coefficient $\mathcal{K}$ it is necessary to add the coefficient $\mathcal{K}_1$ caused by the elastic interaction of the displaced spontaneously deformed material with the surface nonferroactive layer. In the case of the enough large thickness of the ferroelectric plate $\mathcal{K}_1 = 4\mu\varepsilon_0^2/d$, where μ is the the elastic module and ε_0 is the spontaneous deformation or the corresponding deformation accompanying the appearance of P_0 for 90-degree domain structure [10].

Let's consider the influence of defects of the crystal lattice on the motion of domain wall. We shall confine to the case of point defects. Depending on the mobility of the defects their influence on the motion of the domain wall can take place in two qualitatively different ways — through the forces of dry and viscous friction respectively. In the first case the motion of domain wall is an advance through the system of stationary obstacles consisting of the consecutive acts of dispinning of the boundary from those stoppers and its further capture by new defects. The second case takes place if the domain wall interacts with the system of mobile defects accompanying its motion.

These both types of the motion can be described within the frameworks of one-dimensional model. Thus, in spite of a general scheme of consideration due to the difference in terms their particular description is convenient to perform separately. Let's consider the first of the mentioned type of motion, when the domain wall in external field advances through the system of immobile stoppers.

Let's determine the expression for the force acting on the moving boundary from the defects interacting with it. The power per unit of area of domain wall spent by the external electrical field to overcome the resistance of defects to the motion of any domain wall is equal to

$$F\cdot\dot{U}=\int\frac{\partial W}{\partial U}\dot{U}\cdot n(z,t)dz=-\dot{U}\int\frac{\partial W}{\partial z}n(z,t)dz. \quad (26)$$

Here $W(z-U(t))$ is the increase of energy related to the deviation of the wall from its equilibrum position in the system of points of its pinning by defects, U is the coordinate of the plane of average orientation of the domain wall interacting with defects, $\dot{U}$ is its speed, $n(z,t)$ is the bulk concentration of points of pinning for the boundary by defects.

The time evolution of the function $n(z,t)$ is described by kinetic equation with one relaxation time τ:

$$\frac{dn}{dt}=-\frac{n-n_\infty}{\tau}, \quad (27)$$

where the equilibrum distribution of the pinning points in the given place of the crystal [7]

$$n_\infty=n\cdot\theta(\tilde{U}-|z|). \quad (28)$$

Here $\theta(z)$ is the Heaviside function, n is the bulk concentration of defects, $\tilde{U}$ is the maximum distance of a defect from the plane of the average orientation of the boundary, at which the boundary is still captured by a defect.

The solution of (27) is

$$n(z,t)=\int_{-\infty}^{t}\exp[-(t-\xi)/\tau]\cdot n_\infty\xi\frac{d\xi}{\tau}. \quad (29)$$

This pressure on the boundary from defects is

$$F=-\int_{-\infty}^{\infty}\tfrac{\partial W}{\partial z}n(z,t)dz=\int_{-\infty}^{t}\exp(-\tfrac{t-\xi}{\tau})\cdot\int_{-\infty}^{\infty}\tfrac{\partial W(z)}{\partial z}\times$$
$$\times n_\infty[U(t)-U(\xi)]\cdot dz\frac{d\xi}{\tau}. \quad (30)$$

In the case of linear response the difference $U(t)-U(\xi)$ can be considered to be small. In a view of the specific form of $n_\infty(z)$ the spatial integral in (30) can be presented here as $\bar{\mathcal{K}}\cdot(U(t)-U(\xi))$ where $\bar{\mathcal{K}}=nk\tilde{U}$ and k is the coefficient of quasielasticity describing the interaction of the boundary with an isolated defect [7]. As a result the pressure F looks as

$$F=\int_{-\infty}^{t}\exp(-\frac{t-\xi}{\tau})\bar{\mathcal{K}}[U(t)-U(\xi)]\frac{d\xi}{\tau}. \quad (31)$$

The obtained expression enables us to write down the complete equation of motion of the domain wall in a crystal with defects. Substituting (31) into (1) we obtain

$$(m_0+m_1)\ddot{U}(t)+(\mathcal{K}+\bar{\mathcal{K}})U(t)-$$
$$-\bar{\mathcal{K}}\int_{-\infty}^{t}\exp\left(-\frac{t-\xi}{\tau}\right)U(\xi)\frac{d\xi}{\tau}=2P_0E(t). \quad (32)$$

Now we find the solution of the equation (32). Let the external electrical field alters in time under the harmonic law $E(t)=E_0 exp(i\omega t)$. Let's search for solution of the equation (32) for the steady motion as $U(t)=U_0 exp[(i(\omega t+\alpha)]$. Substituting it into (32), in a general case we obtain

$$U(t)=\frac{2P_0E(t)/(m_0+m_1)}{[(\omega_0^2-\omega^2)-\omega_{01}^2/(1+i\omega\tau)]}, \quad (33)$$

where $\omega_0^2=(\mathcal{K}+\bar{\mathcal{K}})/(m_0+m_1)$ and $\omega_{01}^2=\bar{\mathcal{K}}/(m_0+m_1)$ is the square of the fundamental frequency of the ultimately longwave translational vibrations of domain boundaries in crystals with defects and purely defect contribution into it, respectively.

Being displaced in an external field the domain boundaries carry out a repolarization of the ferroelectric material and thus provide the contribution to its dielectric permeability ε. The value of the latter with the account of the determination of the dielectric constant is

$$\varepsilon=\frac{4\pi\Delta P}{E}=\frac{8\pi P_0}{E}\cdot\frac{U}{d}. \quad (34)$$

Substituting of the expression (33) in (34) gives the general expression for the domain contribution into ε:

$$\varepsilon(\omega)=\frac{16\pi P_0^2}{d(m_0+m_1)[(\omega_0^2-\omega^2)-\omega_{01}^2/(1+i\omega\tau)]}, \quad (35)$$

which can be rewritten in a routine form

$$\varepsilon(\omega)=\frac{16\pi P_0^2}{d(m_0+m_1)[\tilde{\omega}^2-\omega^2+i\beta\omega]}, \quad (36)$$

where

$$\tilde{\omega}^2=\omega_0^2-\frac{\omega_{01}^2}{1+\omega^2\tau^2},\ \beta=\frac{\omega_{01}^2\tau}{1+\omega^2\tau^2}. \quad (37)$$

According to (35—37) the type of dielectric dispersion of the domain origin depends on the ratio between the frequencies $\tilde{\omega}^2$ and ω^2. Thus, the value of $\tilde{\omega}^2$ depends on ω (or on τ). For

$\tau \to 0$ (highly mobile defects) $\tilde{\omega}^2 = \omega_0^2 - \omega_{01}^2$ and it is controlled only by the coefficient $\mathcal{K}$. In the other ultimate case for $\tau \to \infty, \tilde{\omega}^2 = \omega_0^2$.

The dispersion of dielectric permeability of a resonant type is realized at $\tilde{\omega} \sim \omega$. In the other practically important case of relatively small frequencies ω the inertial term in (33) and in (35) can be neglected. In this case we have dispersion of a relaxation type where

$$U(t) = \frac{2P_0E}{K + \tilde{K}} \left\{ \left[1 + \frac{\tilde{\mathcal{K}}/\mathcal{K}}{1 + \omega^2 \tau_c^2} \right] \cos \omega t + \right.$$

$$\left. + \frac{\omega \tau_c \tilde{\mathcal{K}}/\mathcal{K}}{1 + \omega^2 \tau_c^2} \right\} \sin \omega t, \tag{38}$$

$$\tau_c = \tau \frac{(\mathcal{K} + \bar{\mathcal{K}})}{\mathcal{K}}.$$

and

$$\varepsilon(\omega) = \varepsilon_1 + \frac{\varepsilon_0 - \varepsilon_1}{1 + i\omega\tau_c} \tag{39}$$

Here $\varepsilon_0 = 16\pi P_0^2/\mathcal{K}d$ and $\varepsilon_1 = 16\pi P_0^2/d(\mathcal{K} + \bar{\mathcal{K}})$ are static (i.e. measured at $\omega = 0$) and high-frequency ($\omega \to \infty$) dielectric permeabilities, respectively, controlled by the charges on the surface of ferroelectric or by another interaction of the boundary with the nonferroelectric layer and by the interaction of the boundaries with defects, respectively.

The height of the maximum $\operatorname{tg}\delta$ is

$$(\operatorname{tg}\delta)_{max} = \frac{\varepsilon_0 - \varepsilon_1}{2\sqrt{\varepsilon_0 \varepsilon_1}} = \frac{nk\tilde{U}}{\sqrt{K(\mathcal{K} + \bar{\mathcal{K}})}}. \tag{40}$$

Taking into account the concentration dependence of the coefficient $\tilde{\mathcal{K}} \sim n$ one can see that in the case of immobile defects the dielectric permeability $\varepsilon_1 \sim 1/n$. With increase of concentration of defects the originally linear uprise of the height of a maximum $\operatorname{tg}\delta$ (where $\mathcal{K} > \bar{\mathcal{K}}$) is replaced then with root dependence, i.e. the growth rate of the height of a maximum $\operatorname{tg}\delta$ with increase of n gradually slows down.

The description of motion of the domain wall interacting with mobile defects is performed similarly with the only difference that the initial equilibrum distribution here is

$$n_\infty(z) = n + n \cdot [\exp(W_0/T) - 1] \cdot a \cdot \delta(z - U(t)), \tag{41}$$

where a is the size of the elementary cell, W_0 is the energy of interaction of the boundary with a defect. As a result, all the final expressions (35—40) here should be kept, where $\bar{\mathcal{K}} = nk\tilde{U}$ should be replaced with $\bar{\mathcal{K}} = nka[exp(W_0/T) - 1]$ and τ here is the relaxation time of the defect atmosphere.

Let's compare the obtained theoretical dependences with the experimental data. In the experiment the domain contribution to dielectric permeability was observed practically in all basic families of ferroelectric crystals - in the crystals of TGS, $BaTiO_3$, KH_2PO_4 groups etc. [19—21].

The domain contribution into ε in the megaherz range of frequencies was observed, in particular, in barium titanate [3] and, apparently, in kalium dihydrophosphate [22], where in a nominally pure crystal the dependence of $\varepsilon(\omega)$ in the «plateau» region was displayed only in the field of frequencies $10^7 \div 10^8$ Hz. At these frequencies of the measuring field the values of ε in the area of «plateau» sharply decreased, while the maximum of $\operatorname{tg}\delta$ from area of «freezing» was abruptly shifted into the area of high temperatures.

The reason of these phenomena in a defectless crystal should be, obviously, the inertial exclusion of domain boundaries from the switching processes at $\omega > \omega_0$. In order to confirm this we shall estimate the value of the latter. For usual $P_0 \sim 10^4$ CGSE units, $\varepsilon_c \sim 10^3, \varepsilon \sim 10, L_x \sim 10^{-1}$, $L_y, L_z \sim 1$ the value of $\mathcal{K}$ proves to be $\sim 10^{10}$, and the value of m_1 at $\beta \sim 10^6, d \sim 10^{-4}, \rho \sim 5, c_t \sim 10^5$ of the order of 10^{-4}, respectively. It provides $\omega_0 = \sqrt{\mathcal{K}/m_1} \sim 10^7$ s^{-1}, i.e. just that value of ω, at which the mentioned peculiarity has been observed in experiment.

Low-frequency peculiarities of the domain contribution in ε were investigated especially well with the example of the family of kalium dihydrophosphate. In numerous experiments it was shown that in the «plateau» region (where below T_c the high values of ε are realized) at the frequencies of $\sim 10^3$ Hz the quasistatic contribution into ε is realized here [23]. Its value is controlled (can be reduced) by applying to a sample of a constant electrical field [24]. Introduction into the crystal of the defects under its doping with chromium ions during the process of growth [25], and also under irradiation of the sample either by γ-rays, or by electron beam [26], or by neutrons [27] also result in a decrease of the values of ε in the «plateau» region.

All these features can be described with the help of expressions (39, 40). In particular, experimentally observed contribution into ε in nominally pure crystals can be described by the quasistatic expression for ε_0. It is also favoured by observing in the experiment of the thickness dependence (growth along with L_x) of the value of ε, which is predicted by the expression for ε_0.

The defects appearing in KH_2PO_4 crystal under its doping are immobile. Therefore, the dielectric constant of such a material can be described by the expression for ε_1. The expected inverse proportional dependence of ε on n according to the above-stated formulas agrees well enough with the experimental results on the measurement of ε values in the «plateau» range in KH_2PO_4 crystal with a various degree of its doping by ions of chromium.

The numerical estimation of ε also coincides well with the experiment in the considered cases. With the above-obtained value of $\mathcal{K}$ the quasistatic dielectric permeability ε_0 in a defectless crystal proves to be of $\sim 10^4$. The ions of chromium in the doped crystals of KH_2PO_4 can be considered as charged defects, and their energy of interaction with domain boundaries is of $\sim 10^{-13}$ erg. The calculated value of $\mathcal{K}$ at this value of W_0 proves to be of $\sim 10^{11}$ [14] while the value of ε_1 itself proves to be $\sim 10^3$, which also corresponds to the experimental data [24].

Finally, the formulas (39, 40) are well described the experimental dependences on defects concentration of components of dielectric permeability and maximum $\mathrm{tg}\,\delta$ in crystals KH_2PO_4 exposed to γ- or electron irradiation: in particular, the fact of delay of the height growth of the maximum $\mathrm{tg}\,\delta$ in γ-irradiated crystal with the increase of defects concentration [4].

REFERENCES

1. *Smolensky G. A., Bokov V. A., Isupov V. A., Kraynik N. N., Pasinkov R. E., Sokolov A. I., Yushin N. K.* Fizika segnetoelektricheskih yavleniy, L., 1985, Nauka, 396 p.

2. *Strukov B. A., Levanyuk A. P.* Fizicheskiye osnovi segnetoelectricheskih yavleniy v kristallah, M. Nauka, Fizmatlit, 1995, 301 p.

3. *Yao X., McKinstry H., Cross L. E.* J. Am. Ceram. Soc., 66, 637 (1983).

4. *Kamysheva L. N., Burdanina N. A., Zhukov O. K. et.al.*, Kristallografiya, 15, 834(1970).

5. *Sannikov D. G.* Izv. Akad. Nauk SSSR, Ser fiz., 28, 703 (1964).

6. *Nechaev V. N., Roschupkin A. M.* Fiz. tverd. tela, 30, 1908 (1988).

7. *Darinskii B. M., Sidorkin A. S., Kostsov A. M.* Ferroelectrics, 160, 35 (1994).

8. *Fedosov V. N., Sidorkin A. S.* Fiz. tverd. tela, 18, 1632 (1976).

9. *Roytburd A. L.* Fiz. tverd. tela, 28, 3051 (1986).

10. *Arlt G., Pertsev N. A.*, J. Appl. Phys., 70, 2283 (1991).

11. *Darinsrii B. M., Parshin A. V., Turkov S. K.* Izv. Akad. Nauk SSSR, Ser fiz., 39, 4, 77 (1975).

12. *Dumachev V. N., Nechaev V. N., Roschupkin A. M.* Izv. Akad. Nauk SSSR, Ser fiz., 10, 14 (1992)

13. *Darinskii B. M., Nechaev V.N., Fedosov V. N.* Phys. St. Sol. (a), 59, 701 (1980).

14. *Sidorkin A. S., Darinsrii B. M.* Fiz. tverd. tela, 28, 285 (1986).

15. *Sidorkin A.S., Shevchenko A. A.* Ferroelectrics, 141, 235 (1993).

16. *Sidorkin A. S.* Ferroelectrics, 186, 239 (1996).

17. *Novatsky V.* Electromagnitnie effekti v tverdih telah. Mockva, Mir, 1986, 159 p.

18. *Sidorkin A. S., Nesterenko L. P.* Fiz. tverd. tela, 37, 3747, (1995).

19. *Fousek J., Brezina B.* J. Phys. Soc. Japan, 19, 830 (1964).

20. *Shilnikov A. V.* Ferroelectrics, 98, 3 (1989).

21. *Bornarel J.* J. Appl. Phys., 71, 255 (1987).

22. *Shuvalov L. A., Zheludev I. S., Mnakatsanyan A. V., Ludupov Ts.-Zh.* Izv. Akad. Nauk SSSR, Ser fiz., 31, 1919 (1967).

23. *Malek Z., Shuvalov L. A., Fiala I., Straiblova Y.* Kristallografiya, 13, 825 (1968).

24. *Kamysheva L. N., Burdanina N. A., Zhukov O. K., Bespamyatnova L. A.* Kristallografiya, 14, 162 (1969).

25. *Kamysheva L. N., Burdanina N. A., Zhukov O. K.* Kristallografiya, 14, 940 (1969).

26. *Kamysheva L. N., Burdanina N. A., Zhukov O. K., Darinskii B. M., Sizova L. N.* Izv. Akad. Nauk SSSR, Ser fiz., 34, 2612 (1970).

27. *Peshikov E. V.* Radiatsionnie effekti v segnetoelektrikah. Tashkent, 1986: FAN, 139 p.