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The impact of the nonlinear effects on thermally stimulated depolarization currents in ion dielectrics

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ABSTRACT

In this paper, the methods described the calculation of thermally stimulated depolarization currents in materials of the HBC class. In the identification of nonlinear processes of thermally stimulated depolarization, at this stage of the research, as defining criteria the authors consider the calculation of the dielectric initial polarization in the infinite approximation of perturbation theory (at the fundamental frequency fields) and calculation of kinetic coefficients in functions of the polarizing field intensity. Generalized nonlinear expressions for the complex dielectric constant and polarization are formulated, which are performed at the fundamental frequency of the alternating polarizing field. The generalized equations that are nonlinear by the field for kinetic coefficients of the kinetic equation are formulated. The obtained theoretical results are of current interest from the perspective of further development of analytical and computer methods of research and prediction of HBC properties as perspective nonlinear materials for a number of branches of modern industry.

Keywords: Nonlinear polarization effect, Polarization of dielectric, Thermally stimulated depolarization, Kinetic theory.

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1. Introduction

When studying the electrophysical properties of various semiconducting and dielectric materials with a complex crystal structure (ceramics; layered crystals (mica, crystalline hydrates); perovskites; vermiculites, etc.), the most effective method is dielectric spectroscopy, which is reduced to precision measurements and calculations of the parameters of dielectric loss spectra and thermally stimulated polarization (TSP) and depolarization currents [1-4]. The results of these studies are relevant for modern electric materials science, which constitutes the theoretical basis for the development and search for the most effective methods and schemes for the use of solid-state composite materials (MIS, CPM-structures, etc.) as functional elements of electrical circuits of

various installations and systems (control-and-measuring, electronically-controlled, electronic-computing, diagnostic), operating in real industrial production.

As an object of research, the authors have selected widely used in modern technologies (microelectronics and radio electronics; optoelectronics and laser technology; insulation and cable technology; electrochemical technologies and hydrogen energy, etc.) hydrogen-bonded crystals (HBC), classified as proton semiconductors and dielectrics. The theoretical concepts of the mechanism of thermally stimulated depolarization, at the present time, are based on the phenomenological kinetic theory built on the solution of the system of Fokker-Plack and Poisson equations for a given model of electrodes, in the linear approximation of the perturbation theory in a small parameter [1; 4]. According to M. P. Tonkonogov as a physical relaxation oscillator (the most mobile particle that relaxes the electric field) in the HBC, far from the breakdown, a hydrogen ion (proton) is picked, moving with the activation energy determined in the vicinity of the temperature of the experimental maximum of the thermally stimulated current. Measurements of temperature density spectra of TSP in the work of M.P. Tonkonogov [1] were carried out at the polarizing field strengths and temperature $\mathrm{E}_0 \approx \left(10^5 \div 10^6\right) \frac{\mathrm{B}}{\mathrm{M}}$ and temperature T = 50-550 K. From the results of studies carried out in [5-7], it is obvious that expanding the experimental range of variation of field and temperature parameters will deepen the theoretical understanding of the mechanism of thermally stimulated polarization and depolarization, especially in the region of anomalously high nonlinearities. These nonlinearities are exhibited in the range of weak fields (100-1000 kV/m) at ultra-low temperatures (1-10 K) and strong fields (100-1000 kV/m) at ultra-low temperatures (1-10 K).

Mensionless parameter [6; 8; 9] on the kinetics of bulk-charge polarization and conductivity in the area of abnormally high polarization nonlinearities [4; 5]. Quantum-mechanical nonlinearities associated with proton tunneling transitions will be reflected in the calculation of the generalized diffusion and mobility coefficients, also in an infinite approximation at a small dimensionless parameter, which has the meaning of the ratio of the potential energy in an electric field to the energy of its thermal motion. The developed methodology for calculating the depolarization (TSDC) density spectra is associated with the procedure for optimizing the calculated values of the parameters of relaxers performed by method of comparison function minimization.

In this paper, the methods described in works of V.A. Kalytka [10], will be applied to the calculation of thermally stimulated depolarization currents in materials of the HBC class. Issues related to categorization, practical application and analysis of electrophysical properties of HBC are considered in publications of V.A. Kalytka [10], et al., a comparative analysis of different models of thermally stimulated depolarization currents in HBC was performed from the perspective of comparability of the results of theoretical and experimental studies.

In the identification of nonlinear processes of thermally stimulated depolarization, at this stage of the research, as defining criteria the authors consider the calculation of the dielectric initial polarization in the infinite approximation of perturbation theory (at the fundamental frequency fields) and calculation of kinetic coefficients in functions of the polarizing field intensity.

2. Materials and methods

In this paper, the study of polarization processes of a model crystal of the HBC class will be conducted by the methods of quasi-classical kinetic theory [4; 6; 10; 11], using the results of analytical solutions of system of nonlinear equations of the phenomenological proton conductivity model that was previously constructed by methods of perturbation theory and Fourier transform methods [4; 6]. The Fokker-Planck kinetic equation is assumed to be linearized, i.e. it does not contain nonlinear degrees of the sought-for concentration of charge carriers (protons). The electrodes are blocking. The external perturbation is reduced to the effect on the dielectric of a harmonically time-varying uniform electric field:

$$\mathrm{E}(t) = \mathrm{E}_0 \exp(\mathrm{i}\omega t). \quad (1)$$

In order to reduce the bulkiness of mathematical transformations, we will operate on the first approximation on the frequency of the alternating field. Based on the results of the research of [6], we assume the polarization of the crystal in an infinite approximation at a small parameter of the perturbation theory, at the fundamental frequency of the alternating field:

$$P^{(\omega)}(t)=\frac{8aqn_0\gamma}{\pi^2\left(1-\frac{8n_0\phi\Lambda_0\gamma}{\pi^2}\right)}\times\sum_{n=1}^{+\infty}\left[\frac{\sin^2\left(\frac{\pi n}{2}\right)}{n^2\left(\frac{1}{\tau_n}+i\frac{\omega}{W^{(0)}}\right)}\right]\times\exp(i\omega t)\cdot \tag{2}$$

In (2) the complex dimensionless parameter (3):

$$\Xi_0=\frac{8n_0\phi\Lambda_0\gamma}{\pi^2} \tag{3}$$

is determined by a number of other fundamental parameters of the kinetic theory of proton relaxation (4) [6]:

$$\phi=\frac{aq}{\varepsilon_0\varepsilon_\infty E_0}- \tag{4}$$

is a value having the dimension that is inverse of the equilibrium concentration of relaxers (protons) n_0:

$$\gamma=\frac{\mu_{mob}^{(1)}aE_0}{D_{diff}^{(0)}}=\varsigma_0\frac{W^{(1)}}{W^{(0)}}- \tag{5}$$

(5) is a small dimensionless parameter of the perturbation theory:

$$D_{diff}^{(0)}=a^2W^{(0)}, \tag{6}$$

$$\mu_{mob}^{(1)}=\frac{qa^2W^{(1)}}{k_BT}- \tag{7}$$

(6), (7) is diffusion and mobility coefficients for protons calculated by the “zero” approximation of the theory of successive approximations [6]. The small dimensionless parameter (8):

$$\varsigma_0=\frac{qE_0a}{k_BT} \tag{8}$$

has the meaning of the parameter of comparison of potential (in an electric field) energy and energy of proton thermal motion:

$$\Lambda_0=\sum_{n=1}^{\infty}\frac{\sin^2\left(\frac{\pi n}{2}\right)}{n^2\left(\frac{1}{\tau_n}+i\frac{\omega}{W^{(0)}}\right)}- \tag{9}$$

(9) is a parameter of interaction of relaxation modes, at the fundamental frequency ω [6; 11]. The dimensionless relaxation time (10):

$$\tau_n=\frac{\tau_{n,D}\tau_M}{\tau_{n,D}+\tau_M} \tag{10}$$

corresponding to the nth relaxation mode of the bulk charge density is revealed through the diffusion relaxation time of the nth mode (11):

$$\tau_{n,D} = \frac{\tau_D}{n^2} \tag{11}$$

and the Maxwell relaxation time (12) [6]:

$$\tau_M = \frac{\varepsilon_0 \varepsilon_\infty W^{(0)}}{q n_0 \mu_{mob}^{(1)}}, \tag{12}$$

where (13):

$$\tau_D = \left(\frac{d}{\pi a}\right)^2 \tag{13}$$

is determined by the crystal thickness d and the lattice parameter a; ε_∞ – is the high-frequency dielectric permittivity; q – is the proton charge.
The kinetic coefficients are calculated in the approximations l=0, l=1, at a small parameter (14):

$$\zeta(x;t) = \left|\frac{qE(x;t)a}{2k_B T}\right| < 1, \tag{14}$$

with regard to both thermally activated (classical) and tunneled (quantum) proton transitions through the parabolic potential barrier (15)-(18) [11]:

$$W^{(0)}(T) = \frac{\nu_0}{2}\left(\exp(-X) + \left\langle D^{(0)}\right\rangle\right), \tag{15}$$

$$\left\langle D^{(0)}\right\rangle = \frac{\exp(-\Lambda) - \exp(-X)}{1 - \frac{\Lambda}{X}}, \tag{16}$$

$$W^{(1)}(T) = \frac{\nu_0}{2}\left(\exp(-X) + \left\langle D^{(1)}\right\rangle\right), \tag{17}$$

$$\left\langle D^{(1)}\right\rangle = \frac{\frac{\Lambda}{X}\exp(-\Lambda) - \exp(-X)}{1 - \frac{\Lambda}{X}}, \tag{18}$$

where:

$$X = \frac{U_0}{k_B T}, \ \Lambda = \frac{\pi \delta_0 \sqrt{m U_0}}{\hbar\sqrt{2}}, \tag{19}$$

m – is the mass of the proton; U_0 – is the height of the potential barrier (the hydrogen-bonded proton activation energy); δ_0 – is the width of the potential barrier; ν_0 – is the linear frequency of the self-oscillations of the proton in the potential well [11]. Noticing that:

$$\tau_{\mathrm{M}} = \frac{1}{\theta}, \tag{20}$$

where:

$$\theta = \phi \gamma n_0 \tag{21}$$

and taking

$$\Xi_0 = \frac{8\Lambda_0}{\pi^2 \tau_{\mathrm{M}}}, \tag{22}$$

and then moving on to dimensional relaxation time:

$$T_{\mathrm{M}} = \frac{\tau_{\mathrm{M}}}{W^{(0)}}, \tag{23}$$

because of:

$$\Xi_0 = \frac{8\Lambda_0}{\pi^2 T_{\mathrm{M}} W^{(0)}}, \tag{24}$$

and given equality:

$$T_{\mathrm{n}} = \frac{\tau_{\mathrm{n}}}{W^{(0)}}, \tag{25}$$

we rewrite (2) as:

$$P^{(\omega)}(t) = \varepsilon_0 \varepsilon_\infty E(t) \times \frac{\Gamma^{(\omega)}}{1 - \Gamma^{(\omega)}}. \tag{26}$$

In (15)-(16) the complex function is introduced:

$$\Gamma^{(\omega)} = \frac{8}{\pi^2 \tau_{\mathrm{M}}} \sum_{\mathrm{n}=1}^{\infty} \left[\frac{\sin^2\left(\frac{\pi \mathrm{n}}{2}\right)}{\mathrm{n}^2 \left(\frac{1}{\tau_{\mathrm{n}}} + \mathrm{i}\frac{\omega}{W^{(0)}}\right)} \right] = \frac{8}{\pi^2 \mathrm{T}_{\mathrm{M}}} \sum_{\mathrm{n}=1}^{\infty} \left[\frac{\sin^2\left(\frac{\pi \mathrm{n}}{2}\right)}{\mathrm{n}^2 \left(\frac{1}{T_{\mathrm{n}}} + \mathrm{i}\omega\right)} \right] \tag{27}$$

Take (26) as

$$\Gamma^{(\omega)} = \Gamma_1^{(\omega)} - \mathrm{i}\Gamma_2^{(\omega)} \tag{28}$$

and denote (29)-(30):

$$\Gamma_1^{(\omega)}(\mathrm{T}) = \frac{8}{\pi^2}\sum_{n=1}^{\infty}\left[\frac{\frac{\tau_n}{\tau_M}\sin^2\left(\frac{\pi n}{2}\right)}{n^2\left(1+\omega^2\tau_n{}^2\right)}\right] = \frac{4}{\pi^2}\sum_{n=1}^{\infty}\left[\frac{\frac{T_n}{T_M}\left(1-(-1)^n\right)}{n^2\left(1+\omega^2 T_n{}^2\right)}\right], \tag{29}$$

$$\Gamma_2^{(\omega)}(\mathrm{T}) = \frac{8}{\pi^2}\sum_{n=1}^{\infty}\left[\frac{\frac{\omega\tau_n^2}{\tau_M}\sin^2\left(\frac{\pi n}{2}\right)}{n^2\left(1+\omega^2\tau_n{}^2\right)}\right] = \frac{4}{\pi^2}\sum_{n=1}^{\infty}\left[\frac{\frac{\omega T_n^2}{T_M}\left(1-(-1)^n\right)}{n^2\left(1+\omega^2 T_n{}^2\right)}\right]. \tag{30}$$

Presenting the polarization of the dielectric in the general form (31):

$$\mathrm{P}^{(\omega)}(\mathrm{t}) = \varepsilon_0\left(\hat{\varepsilon}^{(\omega)} - \varepsilon_\infty\right)\cdot \mathrm{E}(\mathrm{t}) \tag{31}$$

according to (26), we write the complex dielectric constant (32):

$$\hat{\varepsilon}^{(\omega)} = \varepsilon_\infty \times \frac{1}{1-\Gamma^{(\omega)}}, \tag{32}$$

Combining (31) and (32) we have (33):

$$\mathrm{P}^{(\omega)}(\mathrm{t}) = \varepsilon_0\varepsilon_\infty\left(\frac{1}{1-\Gamma^{(\omega)}} - 1\right)\cdot \mathrm{E}(\mathrm{t}), \tag{33}$$

Equations (32), (33) are the most generalized at the fundamental frequency of the alternating field and can be further used in detailed studies of the properties and behavior of the spectra of complex dielectric constant in a wide range of field frequencies and temperatures [10]. At a certain level, this issue has already been solved in the study of V.A. Kalytka [6], which describes a scheme for calculating the polarization up to the second odd harmonic multiple of , constructs approximate equations for polarization on an arbitrary frequency harmonic of r multiplicity. However, components of complex dielectric constant were not explicitly calculated in the above work [6], which is the subject of a separate theoretical study based on rather cumbersome mathematical transformations, the result of which is relevant for areas of abnormally high polarization nonlinearities (ultra-low temperatures and strong electric fields) [4; 6; 11].

Now we proceed to the calculation of dielectric constant (32) and polarization (33) in a static uniform electric field. Then, noticing that $\Gamma_2^{(\omega=0)}(\mathrm{T}) = 0$,

$$\Gamma_1^{(\omega=0)}(\mathrm{T}) = \frac{4}{\pi^2}\sum_{n=1}^{\infty}\left[\frac{\frac{T_n}{T_M}\left(1-(-1)^n\right)}{n^2}\right] \tag{34}$$

and, owing to (35):

$$\frac{T_n}{T_M} = \frac{T_{n,D}}{T_{n,D}+T_M}, \tag{35}$$

where:

$$T_{\mathrm{n,D}} = \frac{T_{\mathrm{D}}}{\mathrm{n}^2}, \tag{36}$$

$$T_{\mathrm{D}} = \left(\frac{\mathrm{d}}{\pi a}\right)^2 \left[W^{(0)}\right]^{-1}, \tag{37}$$

when:

$$\frac{T_{\mathrm{n}}}{T_{\mathrm{M}}} = \frac{\frac{T_{\mathrm{D}}}{T_{\mathrm{M}}}}{\mathrm{n}^2 + \frac{T_{\mathrm{D}}}{T_{\mathrm{M}}}}, \tag{38}$$

write:

$$\hat{\varepsilon}^{(\omega=0)} = \varepsilon_{\mathrm{S}}(\mathrm{T}) = \varepsilon_{\infty} \times \frac{1}{1-\Gamma^{(\omega=0)}} = \frac{\varepsilon_{\infty}}{1 - \frac{4T_{\mathrm{D}}}{\pi^2 T_{\mathrm{M}}} \sum_{\mathrm{n}=1}^{\infty} \frac{(1-(-1)^{\mathrm{n}})}{\mathrm{n}^2\left(\mathrm{n}^2 + \frac{T_{\mathrm{D}}}{T_{\mathrm{M}}}\right)}}. \tag{39}$$

On the basis of equality (40):

$$\sum_{\mathrm{n}=1}^{\infty} \frac{(1-(-1)^{\mathrm{n}})}{\mathrm{n}^2\left(\mathrm{n}^2 + \frac{T_{\mathrm{D}}}{T_{\mathrm{M}}}\right)} = \frac{\pi^2}{4} \frac{1}{\frac{T_{\mathrm{D}}}{T_{\mathrm{M}}}} \left(1 - \frac{\mathrm{th}\left(\frac{\pi}{2}\sqrt{\frac{T_{\mathrm{D}}}{T_{\mathrm{M}}}}\right)}{\frac{\pi}{2}\sqrt{\frac{T_{\mathrm{D}}}{T_{\mathrm{M}}}}}\right), \tag{40}$$

from (39) and (33) we obtain static equations:

$$\varepsilon_{\mathrm{S}}(\mathrm{T}) = \varepsilon_{\infty} \mathrm{x}_{\mathrm{D,M}} \cdot \mathrm{cth}\left(\mathrm{x}_{\mathrm{D,M}}\right), \tag{41}$$

$$\mathrm{P_S} = \varepsilon_0 \varepsilon_{\infty} \left(\mathrm{x}_{\mathrm{D,M}} \cdot \mathrm{cth}\left(\mathrm{x}_{\mathrm{D,M}}\right) - 1\right) \cdot \mathrm{E}_0, \tag{42}$$

where:

$$\mathrm{x}_{\mathrm{D,M}} = \frac{\pi}{2}\sqrt{\eta(\mathrm{T})}, \tag{43}$$

$$\eta(\mathrm{T}) = \frac{T_{\mathrm{D}}(\mathrm{T})}{T_{\mathrm{M}}(\mathrm{T})} = \frac{\mathrm{d}^2 n_0 \mathrm{q}^2}{\pi^2 \varepsilon_0 \varepsilon_{\infty} \mathrm{k_B T}} \cdot \frac{W^{(1)}(\mathrm{T})}{W^{(0)}(\mathrm{T})}. \tag{44}$$

From the identity (45):

$$\frac{T_{\mathrm{D}}}{T_{\mathrm{M}}} = \left(\frac{\mathrm{d}}{\mathrm{r_D}}\right)^2 \tag{45}$$

we compute the Debye's shielding radius (46):

$$r_D(T) = d\sqrt{\frac{T_M}{T_D}}, \tag{46}$$

or (47):

$$r_D(T) = \pi\sqrt{\frac{\varepsilon_0\varepsilon_\infty \cdot D_{dif}^{(0)}(T)}{q\,n_0 \cdot \mu_{mob}^{(1)}(T)}}. \tag{47}$$

Then, in the relaxation of the mixed type $(T_M \approx T_D)$ we have (48):

$$r_D(T) = \frac{\pi}{q}\sqrt{\frac{\varepsilon_0\varepsilon_\infty k_B T}{n_0}} \cdot \sqrt{\frac{\exp\left(-\frac{U_0}{k_B T}\right) + \langle D^{(0)}\rangle}{\exp\left(-\frac{U_0}{k_B T}\right) + \langle D^{(1)}\rangle}} \tag{48}$$

In the area of diffusion relaxation, taking $T_D << T_M$, when (49):

$$x_{D,M} = \frac{\pi d}{2r_D} << 1, \tag{49}$$

due to an approximate equation (50):

$$\mathrm{cth}(x_{D,M}) \approx \frac{1}{x_{D,M}} + \frac{x_{D,M}}{3}, \tag{50}$$

from (41), (42) we obtain the approximated equations (51)-(52):

$$\varepsilon_S(T) \approx \varepsilon_\infty\left(1 + \frac{\pi^2 d^2}{12 r_D^2}\right), \tag{51}$$

$$P_S \approx \frac{\varepsilon_0\varepsilon_\infty \pi^2 d^2}{12 r_D^2}. \tag{52}$$

Here, in the numerical evaluation of temperature effects on the Debye's shielding radius $r_D(T)$, the equation (48) is used, in which, at the microscopic level, both thermally activated and tunneling proton transitions are taken into account. At the same time, at the macroscopic level, the type of proton relaxation, due to the large values of the Maxwell relaxation time (owing to small concentrations of protons), is taken as diffusion one. In the case of an infinitesimal ratio of (53):

$$\eta(T) = \frac{T_D(T)}{T_M(T)} << 1, \tag{53}$$

when (54):

$$x_{D,M} = \frac{\pi d}{2 r_D} \to 0, \tag{54}$$

according to the limit (55):

$$\lim_{x \to 0} \left[x \cdot \mathrm{cth}(x) \right] = \lim_{x \to 0} \left[\frac{1}{\frac{\mathrm{th}(x)}{x}} \right] = 1, \tag{55}$$

the dielectric constant tends to high-frequency permeability and the polarization tends to zero. In the area $T_M << T_D$ when (56):

$$\eta(T) = \frac{T_D(T)}{T_M(T)} >> 1, \tag{56}$$

$$x_{D,M} = \frac{\pi d}{2 r_D} >> 1 \tag{57}$$

according to (58):

$$\lim_{x \to \infty} \left[\mathrm{cth}(x) \right] = \lim_{x \to \infty} \left[\frac{1}{\mathrm{th}(x)} \right] = 1 \tag{58}$$

and (59):

$$x \cdot \mathrm{cth}(x) \approx x, \tag{59}$$

we have approximately (60), (61):

$$\varepsilon_S(T) \to \frac{\pi d \varepsilon_\infty}{2 r_D} >> 1, \tag{60}$$

$$P_S \approx \frac{\varepsilon_0 \varepsilon_\infty \pi d}{r_D}, \tag{61}$$

in the case. It is obvious that the Maxwell relaxation caused by high proton concentrations provides abnormally small values of the Debye shielding radius $r_D << d$. At the same time, as in the case of diffusion-relaxation polarization, both the classical and quantum mechanism of proton transfers through the potential barrier is equally taken into account.

Regarding the effect that microscopic mechanism of proton transitions in the potential relief has on the Debye shielding radius, it is easy to see from (48) that in the temperature area (zone) of classical transitions, at temperatures much higher than the critical (62) [10]:

$$T_{cr,mov} = \frac{\hbar \sqrt{2 U_0}}{\pi \delta_0 k_B \sqrt{m}} \tag{62}$$

when (63), (64):

$$\mathrm{T} >> \mathrm{T}_{cr,mov}, \tag{63}$$

$$\frac{\Lambda}{\mathrm{X}} >> 1 \tag{64}$$

and (65):

$$W^{(l)}(\mathrm{T}) \to W^{(l)}_{therm}(\mathrm{T}) = \frac{\nu_0}{2}\exp(-\mathrm{X}), \tag{65}$$

we obtain an equation (66):

$$\mathrm{r_D}(\mathrm{T}) \approx \frac{\pi}{\mathrm{q}}\sqrt{\frac{\varepsilon_0\varepsilon_\infty \mathrm{k_B T}}{n_0}} \tag{66}$$

that is consistent with the representations of the classical statistical theory and, near the temperature of absolute zero $\mathrm{r_D}(\mathrm{T}) \to 0$.

In the temperature area of quantum transitions, at temperatures below the critical ones, when $\mathrm{T} < \mathrm{T}_{cr,mov}$, $\frac{\Lambda}{\mathrm{X}} < 1$ and (67):

$$W^{(l)}(\mathrm{T}) \to W^{(l)}_{tunnel}(\mathrm{T}) = \frac{\nu_0}{2}\left\langle \mathrm{D}^{(l)} \right\rangle, \tag{67}$$

from (68):

$$\mathrm{r_D}(\mathrm{T}) = \frac{\pi}{\mathrm{q}}\sqrt{\frac{\varepsilon_0\varepsilon_\infty \mathrm{k_B T}}{n_0}} \cdot \sqrt{\frac{\left\langle \mathrm{D}^{(0)} \right\rangle}{\left\langle \mathrm{D}^{(1)} \right\rangle}}. \tag{68}$$

In the case of $\mathrm{T} << \mathrm{T}_{cr,mov}$, $\frac{\Lambda}{\mathrm{X}} << 1$, when (69):

$$\left\langle \mathrm{D}^{(l)} \right\rangle \approx \frac{\left(\frac{\Lambda}{\mathrm{X}}\right)^l \exp(-\Lambda)}{1-\frac{\Lambda}{\mathrm{X}}}, \tag{69}$$

we obtain the limit equation (70):

$$\mathrm{r_D}(\mathrm{T}) \approx \frac{\pi}{\mathrm{q}}\sqrt{\frac{\varepsilon_0\varepsilon_\infty \mathrm{k_B T}}{n_0}} \cdot \sqrt{\frac{\mathrm{X}}{\Lambda}} = \frac{\pi}{\mathrm{q}}\sqrt{\frac{\varepsilon_0\varepsilon_\infty \hbar\sqrt{2\mathrm{U}_0}}{n_0 \pi\delta_0\sqrt{\mathrm{m}}}}. \tag{70}$$

The identity (71):

$$\frac{\mathrm{X}}{\Lambda} = \frac{\hbar\sqrt{2\mathrm{U}_0}}{\pi \mathrm{k_B T}\delta_0\sqrt{\mathrm{m}}} \tag{71}$$

was used here. It is easy to see that, taking into account the quantum effects reflected in the expression (72):

$$\left\langle \mathrm{D}^{(l)} \right\rangle \approx \left(\frac{\Lambda}{\mathrm{X}} \right)^{l} \left\langle \mathrm{D}^{(0)} \right\rangle, \tag{72}$$

where

$$\left\langle \mathrm{D}^{(0)} \right\rangle \approx \left(1 - \frac{\Lambda}{\mathrm{X}} \right)^{-1} \times \exp(-\Lambda), \tag{73}$$

in contrast to the classical theory, causes a non-zero value of the Debye shielding radius near the temperature of absolute zero. In this case, assuming $\frac{\Lambda}{\mathrm{X}} \to 0$, $\left\langle \mathrm{D}^{(0)} \right\rangle \to \exp(-\Lambda)$:

$$\left\langle \mathrm{D}^{(1)} \right\rangle \approx \frac{\Lambda}{\mathrm{X}} \left\langle \mathrm{D}^{(0)} \right\rangle, \tag{74}$$

we obtain the same result as above:

$$\mathrm{r_D}(0) \to \frac{\pi}{\mathrm{q}} \sqrt{\frac{\varepsilon_0 \varepsilon_\infty \hbar \sqrt{2\,\mathrm{U}_0}}{n_0 \pi \delta_0 \sqrt{\mathrm{m}}}}. \tag{75}$$

3. Results and discussion

A detailed analysis of the effects of various external factors (variation of the amplitude of the electromotive force (EMF) of the voltage source; alloying; calcination; negative currents) makes it possible to reveal the physical nature of each type of relaxation oscillators (charged structural defects), activated near temperatures corresponding to the experimental maximum density of the TSP $-J_{depol,\exp}(\mathrm{T})$, or dielectric loss tangent $-tg_{\exp}(T)$. Of greatest theoretical and practical interest are the anomalously high nonlinearities exhibited during the polarization of ionic dielectrics in the region of strong electric fields (10-1000 MV/m) [5-7; 11], when, as a result activation of two or more types of relaxation oscillators, against the background of sufficiently high conduction currents, the amplitude of the high-temperature (500-1500 K) current density maximum begins to depend significantly on the intensity of the polarizing field [2; 9].

An important issue is the study of the processes of charge accumulation and relaxation in nanoscale surface plastic deformations (low-temperature electret effect) in the development of fuel cells for hydrogen energy [12-15] in the field of space technology and for electrochemical technology [16-21].

Mathematical modeling of the quantum transfer of protons in systems from thin potential barriers with potential wells containing quantum-well energy levels is relevant in the development of physical principles and schemes of operation of resonant tunneling diodes (RTD) and quantum field effect transistors (FETs) based on proton semiconductors and dielectrics, for microelectronics, radio electronics and quantum electronics [22-28].

Anomalously high transparency of potential barriers of the transverse voltage pattern [29], against the background of sufficiently high frequencies of proton vibrations in transverse potential wells (around 10^{13} c^{-1}) [2-4; 6; 9], make it possible to use the effects of the nanoscale state of the HBC [29] and some other quantum size effects to create ultrafast digital or microwave devices with operating frequencies of more than 1 THz [24; 30-32]. Devices of this type include optical interferometers, interference filters, monochromators, thin-film systems such as resonant tunneling diodes, triodes, etc. [33-36]. The main directions of further studies of the kinetics of low-temperature proton relaxation in crystals with hydrogen bonds (HBC) will be reduced to the construction and development of schemes for analytical solutions of quantum-mechanical kinetic equations, taking into account the effect of proton-phonon interactions on proton-relaxation polarization and conductivity.

At the same time, the methods of the semiclassical kinetic theory of space-charge polarization and depolarization are effective from the point of view of comparative estimates of the effects of various kinds of quantum nonlinearities on the spectra of depolarization currents and on the values of the characteristic parameters of relaxers. It is obvious that the dynamics of interactions in the systems "electron and phonons in a dielectric" and "protons and phonons in a dielectric" [37-39], is not absolutely identical, even if this dielectric is a HBC, due to a number of differences in the conditions of motion of the major charge carriers and in the specifics of their interaction with phonons. The same applies to the mathematical apparatus describing these models [40-58]. In fact, vibrations of a proton, in a certain range of field and temperature parameters (close to breakdown), themselves can generate phonons that affect conduction electrons. In this case, far from breakdown, when the effects of proton relaxation dominate in the dielectric, and electrons are distributed deep in the valence band, the proton already becomes a light mobile particle, and the vibrations of more massive sedentary anions generate phonons for protons and the proton-phonon interaction is activated [59-75]. All these issues can be investigated only in combination with the results of preliminary detailed theoretical studies of the effects of the tunneling motion of protons on the theoretical spectra of the complex permittivity (CDP), polarization and depolarization currents in the HBC, within the framework of the nonlinear quasi-classical kinetic theory, taking into account the transparency of the potential barrier so far only in as a term in the kinetic coefficients of the Fokker-Planck equation, which also determines the scientific significance of the studies carried out in this paper [76-95].

Note that due to the similarity of the diffusion mechanisms of high-temperature proton conductivity in HBC [4; 6; 10] and ionic conductivity in YSZ crystals [96-100], constructed in [4-6] equations of diffusion transfer of protons in an electric field, after some model upgrades, can be used in the mathematical description of the kinetics of transfer of oxygen ions during the polarization of dielectrics such as zirconium dioxide [101-123].

In the study of Yu.M. Annenkov experimentally, in samples of corundum-zirconium ceramics, or CZC (ZrO_2 – Y_2O_3) – n(Al_2O_3), at a frequency of an alternating field of 1 kHz, at the point T = 1250 K, anomalously high dielectric constants $\varepsilon=5\cdot10^6$ were found, due to, according to the authors (Annenkov et al., 2005), structural rearrangement of the oxygen sublattice in the CZC, near the critical temperature (quasi-ferroelectric effect) [124]. The equations of high-temperature (550 – 1500) space-charge polarization of the HBC – nonlinear equations of the Fokker – Planck type [4; 6], can also be applied to the CZC, when describing the mechanism of relaxation motion (transfer) oxygen ions and, accordingly, to the study of the spectra ε (T), near the phase transition temperature, which is important when constructing theoretical methods for predicting the nonlinear electrophysical properties of high-temperature ionic superconductors [125-146].

This class of materials also includes magnesium-doped ceramic crystals of the copper (I) chromite type with hopping hole conductivity [147-149].

Studies of the mechanism of proton relaxation in thermally stimulated depolarization in HBC should be conducted on the basis of the kinetic equation describing the transfer of protons in the field of destructive electret charge accumulated in the dielectric volume for the period of polarization during the time of the order of relaxation time. The value of the polarization relaxation time, as established from the solutions of the nonlinear Fokker-Planck equation, is a discrete value calculated separately for each number n of the relaxation mode. The temperature of the crystal is assumed to be constant and equal to the polarization T_{pol}. In this case, the ratio $\frac{T_{\mathrm{D}}(\mathrm{T}_{pol})}{T_{\mathrm{M}}(\mathrm{T}_{pol})}$ determines the mechanism of formation of the volume-charge distribution in the space between the capacitor plates at the field strength E_{pol} and the type of temperature dependence of the Debye shielding radius of the charge $\mathrm{r}_{\mathrm{D}}(\mathrm{T}_{pol})$.

Thus, the study of relaxation time spectra $T_{\mathrm{n}}(\mathrm{T}_{pol})$ is an important component of the polarization problem and ultimately determines the asymptotics of polarization established in the dielectric during the polarization time t_{pol}. Then, the calculation of the initial polarization of the dielectric, according to the traditional scheme of measuring the thermally stimulated depolarization current, will be conducted at the temperature T_{pol} and intensity of a uniform static polarizing field E_{pol}. In this case, the initial polarization of the experimental sample (76):

$$P_{depol}(0)=\varepsilon_0\varepsilon_\infty\left(\mathrm{x}_{\mathrm{D,M}}\left(\mathrm{T}_{pol}\right)\cdot\mathrm{cth}\left(\mathrm{x}_{\mathrm{D,M}}\left(\mathrm{T}_{pol}\right)\right)-1\right)\cdot\mathrm{E}_{pol}. \tag{76}$$

Here (77):

$$\mathrm{x}_{\mathrm{D,M}}\left(\mathrm{T}_{pol}\right)=\frac{\pi}{2}\sqrt{\eta_{pol}}\ , \tag{77}$$

where (78):

$$\eta_{pol}=\frac{T_{\mathrm{D}}\left(\mathrm{T}_{pol}\right)}{T_{\mathrm{M}}\left(\mathrm{T}_{pol}\right)}=\frac{\mathrm{d}^2 n_0\,\mathrm{q}^2}{\pi^2\varepsilon_0\varepsilon_\infty\mathrm{k}_{\mathrm{B}}\mathrm{T}_{pol}}\cdot\frac{W^{(1)}\left(\mathrm{T}_{pol}\right)}{W^{(0)}\left(\mathrm{T}_{pol}\right)}. \tag{78}$$

The kinetic equation for the process of thermally stimulated depolarization describes the processes of relaxation motion of charge carriers (protons) in the absence of an external electric field $\mathrm{E}(t)=0$, when the linear heating of the crystal (79):

$$\mathrm{T}(t)=\mathrm{T}_0+ct\ , \tag{79}$$

where $c=\mathrm{const}$, T_0- is the initial depolarization temperature, causes the destruction of the polarized state $P_{depol}(0)$, and the current polarization of the dielectric in the process of thermally stimulated depolarization, as a function of time $P_{depol}(t)$, should asymptotically tend to zero $\lim\limits_{t\to\infty}\left(P_{depol}(t)\right)=0$.

The structure of the kinetic equation of proton-relaxation depolarization [5; 6] depends, firstly, on a number of properties of the physical model, under given conditions of the process (laws of temperature change and the intensity of the external electric field in time, the type of electrodes, the effect of strains and mechanical stresses on polarization processes, etc.) and, secondly, on the pre-predicted degree of accuracy of the final analytical solutions.

In this regard, within the phenomenological model of proton conductivity and relaxation in HBC [4-6; 10; 11], two main variants of the choice of this kinetic equation are possible:

1) on the basis of the quasi-classical Fokker-Planck equation for depolarization, with kinetic coefficients (15)-(18) being functions of temperature. In this case, the initial distribution of the bulk charge will be determined by the sum of the distributions of the hetero-charge (associated with the distribution of protons along the dielectric thickness) and homo-charge (associated with the injection of charges from the near-surface area of the electrodes deep into the crystal) [2];

2) on the basis of a general kinetic equation based on the model of a double symmetric potential well perturbed by a uniform electric field $\mathrm{E}(t)$ [9]. In depolarization, the field strength is assumed to be zero. The effect of spatially inhomogeneous homo-charge distribution is not taken into account.

Since in this paper the factor determining the degree of rigor of the mathematical model is, in continuation of the linear kinetic theory [2; 3; 9], the focus on the effects of the interaction of relaxation modes n on the initial polarization (76) inherent in parameter (27), then, to describe the kinetics of depolarization, we choose the second version of the kinetic equation (80) [9]:

$$\frac{\mathrm{d}(\Delta n)}{\mathrm{dt}}+2\Omega(t)\cdot\Delta n=\frac{1}{3}n_0\cdot\Xi(t) \tag{80}$$

Next, we present the average velocity of the probability of proton transitions between potential wells (81):

$$\Omega(t)=\frac{W^{(-)}(t)+W^{(+)}(t)}{2}=\frac{\mathrm{D}_{diff}(t)}{a^2} \tag{81}$$

and the excess velocity of the probability of diffusion transport of protons across the field (82) [11]:

$$\Xi(t) = W^{(-)}(t) - W^{(+)}(t) = \frac{\mathrm{v}_{\mathrm{mob}}(t)}{a}, \tag{82}$$

taking into account (15)-(18), as:

$$\Omega(t) = \frac{\nu_0}{2}\left\{\mathrm{ch}(\zeta(t))\exp(-\mathrm{X}) + \frac{\mathrm{ch}(\eta(t))\exp(-\Lambda) - \mathrm{ch}(\zeta(t))\exp(-\mathrm{X})}{1-\frac{\Lambda}{\mathrm{X}}}\right\}, \tag{83}$$

$$\Xi(t) = \nu_0\left\{\mathrm{sh}(\zeta(t))\cdot\exp(-\mathrm{X}) + \frac{\mathrm{sh}(\eta(t))\cdot\exp(-\Lambda) - \mathrm{sh}(\zeta(t))\cdot\exp(-\mathrm{X})}{1-\frac{\Lambda}{\mathrm{X}}}\right\}. \tag{84}$$

Here (85), (86) [6]:

$$\zeta(t) = \frac{|\Delta \mathrm{U}(t)|}{\mathrm{k_B T}} \approx \frac{\mathrm{qE}(t)a}{2\mathrm{k_B T}}, \tag{85}$$

$$\eta(t) = \Lambda\frac{|\Delta \mathrm{U}(t)|}{\mathrm{U}_0} \approx \frac{\mathrm{qE}(t)\Lambda a}{2\mathrm{k_B T U}_0}. \tag{86}$$

Also we should note the ratio (87):

Obviously, at the critical temperature $\mathrm{T}_{cr,mov}$, the equation (88):

$$\zeta(t) = \eta(t) \tag{88}$$

is performed. In the case of $\mathrm{E}(t) = 0$, assuming $\Omega(t) = W^{(0)}(t)$, $\Xi(t) = 0$, rewriting (80) as (89):

$$\frac{\partial(\Delta n)}{\partial \mathrm{t}} + 2W^{(0)}(t)\cdot\Delta n = 0 \tag{89}$$

and introducing the coordinate averaged polarization (90):

$$P_{depol}(t) = \frac{\mathrm{q}}{\mathrm{d}}\int_0^{\mathrm{d}} \Delta n(\mathrm{x},t)\mathrm{dx}, \tag{90}$$

we formulate a differential equation (91):

$$\frac{\mathrm{d}P_{depol}(t)}{\mathrm{d}t} + 2W^{(0)}(t)\cdot P_{depol}(t) = 0, \tag{91}$$

solved with regard to the initial condition (76). Then (92):

$$P_{depol}(t) = P_{depol}(0)\cdot\exp\left(-2\int_0^t W^{(0)}(t)\right)\mathrm{d}t\cdot \tag{92}$$

Calculation of the current density of the thermally stimulated depolarization (93):

$$J_{depol}(t) = -\frac{\partial P_{depol}(t)}{\partial t}, \tag{93}$$

taking into account (94):

$$c = \frac{\mathrm{dT}(t)}{\mathrm{d}t}, \tag{94}$$

finally gives (95):

$$J_{depol}(\mathrm{T}) = 2P_{depol}(0)\cdot W^{(0)}(\mathrm{T})\cdot \exp\left(-\frac{2}{c}\int_{\mathrm{T}_0}^{\mathrm{T}} W^{(0)}(\mathrm{T})\right)\mathrm{dT}\cdot \tag{95}$$

Equation (95) formally coincides with the Bucci-Riva formula known from thermal depolarization analysis [9], however, the obtained result contains a number of fundamental model differences and additions. First, the kinetic coefficients $W^{(0)}(\mathrm{T})$ are calculated as a function of temperature, taking into account the tunnel transitions of relaxers (protons) through the parabolic potential barrier and, secondly, the initial polarization is quite complex in structure and informative function, reflecting the influence of nonlinear polarization effects (interaction of relaxation modes; nonlinear bulk-charge polarization of mixed type; electret effect, etc.) on the kinetics of depolarization. At the same time, the expression (76) works in a sufficiently wide theoretical range of field parameters (100 kV/m-1000 MV/m) and temperatures (0-1500 K) and allows to reveal at the theoretical level the polarization nonlinearities occurring in the area of ultra-low temperatures (1-10 K) and strong fields (100-1000 MV/m). The phenomenological Bucci-Riva formula, according to the results of numerical calculations, is limited to the experimental range of variation of the macroscopic parameters of the process [150] $\mathrm{E}_{pol} \approx \left(10^5 \div 10^6\right)\ \frac{\mathrm{V}}{\mathrm{m}}$, $\mathrm{T}_{pol} \approx (70 \div 450)\mathrm{K}$. Studies of the integral (96):

$$B(\mathrm{T}) = \int_{\mathrm{T}_0}^{\mathrm{T}} W^{(0)}(\mathrm{T})\mathrm{dT} \tag{96}$$

are conducted by numerical methods [151]. A condition $\mathrm{T}_0 \le \mathrm{T}_{pol}$ is imposed on the initial depolarization temperature. As a rule, when measuring the temperature spectra of TSDC density, experimental values T_0 are taken near the nitrogen temperature (50-70 K), and the polarization temperature is set within $\mathrm{T}_{pol} \approx (300 \div 350)\mathrm{K}$. The rate of linear heating of the crystal $c \approx (0{,}1 \div 1)\frac{\mathrm{K}}{\mathrm{s}}$.

The analytical equation (95) enables to construct theoretical spectra of the density of the thermally stimulated depolarization current $J_{depol,\mathrm{th}}(\mathrm{T})$ and, on this basis, by minimizing the comparison function of the theory with the experiment (97) [7]:

$$\Psi\left(\vec{\zeta}_{\max}\right) = \left[\mathrm{T}_{\max,\mathrm{th}}^{\left[J_{depol,\mathrm{th}}(\mathrm{T};\vec{\zeta})\right]} - T_{\max,\exp}^{\left[J_{depol\exp}(T;\vec{\zeta}_0)\right]}\right]^2 \to \min\ , \tag{97}$$

by programmable computer enumeration of the theoretical values of the characteristic parameters (98):

$$\vec{\zeta} = \{U_{0,th}, \nu_{0,th}, n_{0,th}, a_{th}, \delta_{0,th}\}, \quad (98)$$

in the vicinity (on the set of continuum) temperature of the experimental maximum $T_{max,exp}$, to determine the optimized against the experimental values (99):

$$\vec{\zeta}_0 = \{U_{0,exp}; \nu_{0,exp}; n_{0,exp}; \delta_{0,exp}\}, \quad (99)$$

the theoretical values of these parameters for the temperature $T_{max,th}$, as a vector (100):

$$\vec{\zeta}_{max;\,opt} = \{U_{0,th,max;\,opt}; \nu_{0,th,max;\,opt}; n_{0,th,max;\,opt}; \delta_{0,th,max;\,opt}\} \quad (100)$$

4. Conclusions

Generalized nonlinear expressions for the complex dielectric constant (32) and polarization (33) are formulated, which are performed at the fundamental frequency of the alternating polarizing field. In the case of a stationary homogeneous electric field, the formulas (32), (33) are reduced to the analytical dependences (41), (42), more illustrative and convenient for comparison with the experiment, in comparison with the results of the linear kinetic theory.

From the solution of the general quzasi-classical kinetic equation (91), an expression for the current density of thermally stimulated depolarization (95) is formulated, with regard to the nonlinear polarization effects due to the interaction of relaxation modes of the bulk charge density, which is reflected in (76). The generalized equations that are nonlinear by the field for kinetic coefficients (83), (84) of kinetic equation (80) are formulated. A significant impact of quantum effects on the kinetic coefficients (83), (84) at temperatures much lower than the critical temperature (in the area of nonlinear quantum diffusion polarization) is established.

The proposed scheme of numerical calculation of the characteristic parameters of the relaxers limited to the study of current density of thermally stimulated depolarization (91) by minimization of theory and experiment comparison function (97), by computer enumeration of parameters of relaxers at a set of points of the continuum in the vicinity of the temperature of each mono-relaxational maximum of TSDC density.

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