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## Low-temperature activation of phonons in ferroelectrics

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The paper presents a study of the temperature dependence of the permittivity  $\varepsilon(T)$  of quantum ferroelectrics  $\text{KTaO}_3$  and  $\text{SrTiO}_3$  and the solid solution based on them  $(\text{SrTiO}_3)_{0.85}-(\text{KTaO}_3)_{0.15}$  at temperatures from 5 to 300 K, for the description of which an exponential function with the Boltzmann probability factor and a self-consistent pre-exponential factor is used. It is shown that such an approach in the low-temperature quantum limit is consistent with the Barrett formula, and in the high-temperature limit — with the Curie–Weiss law. Based on this, the activation energies of the processes occurring in three selected temperature ranges (5–30; 30–80; 80–300 K) are determined, where the dependence  $\varepsilon(T)$  has an exponential form. It is assumed that for  $\text{KTaO}_3$  and  $\text{SrTiO}_3$  such activation processes are caused by the temperature excitation of phonons when approaching the Debye temperature. For  $(\text{SrTiO}_3)_{0.85}-(\text{KTaO}_3)_{0.15}$  the deviation of the  $\varepsilon(T)$  dependence at  $T = 30\text{--}80$  K from the exponential one is explained by phonon-phonon interactions of normal modes of crystal vibrations.

**Keywords:** quantum ferroelectric, activation energy, strontium titanate, potassium tantalate, permittivity, phonons.

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### 1. Introduction

Crystals  $\text{KTaO}_3$  and  $\text{SrTiO}_3$  are inorganic compounds with structure of oxygen-octahedral perovskite with common formula  $\text{ABO}_3$ , in which one- (K) or bivalent (Sr) metal is located at vertices of cubic lattice cell, and quinta- (Ta) or tetravalent metal (Ti) is located in center of cube. At that, oxygen O atoms are located in centers of facets.  $\text{KTaO}_3$  and  $\text{SrTiO}_3$  are related to potential (virtual) or quantum [1] ferroelectrics of offset type, where the ferroelectric phase transition is suppressed by quantum fluctuations [2]. At low temperatures  $\text{KTaO}_3$  and  $\text{SrTiO}_3$  demonstrate similarity in proximity to ferroelectric state, and in presence of interaction between soft optical mode and transverse acoustic branch, which leads to softening of the acoustic branch [2]. But,  $\text{KTaO}_3$  has cubic symmetry up to temperature 4 K, where it completely loses stability in relation to soft mode condensation. At same time, in  $\text{SrTiO}_3$  near 105 K there is structural phase transition associated with turn of oxygen octahedrons [2], the phase transition is ferroplastic [3]. The presence of a doping-induced ferroelectric phase has been noted [4]. In description of quantum paraelectricity the theoretical calculations of first principles are often used, they are based on consideration of the electronic subsystem by density functional theory (DFT) and solution of Schrödinger single-particle equation with potential calculated using DFT [5].

The interest to quantum ferroelectrics is also determined by their practical application. So,  $\text{SrTiO}_3$  in form of bulk crystals and thin films is used as substrates to grow oxide layers of high-temperature superconductor due to closeness of their lattice parameters [6]. In heterostructure comprising

epitaxial layers  $\text{KTaO}_3$  and  $\text{SrTiO}_3$ , on facets of interface  $\text{KO}^-/\text{TiO}_2^0$  and  $\text{TaO}^{+2}/\text{SrO}^0$  the sign of two-dimensional hole (2DHG) and electron gas (2DEG), respectively was determined [7]. This opens new prospect in oxide electronics and spintronics, as the physical phenomena, associated with two-dimensional electron gas trapped at boundaries of interface of such heterostructures, include spin polarization, superconductivity, quantum oscillations of magnetoresistance, spin-polarized transport of electrons, permanent photocurrent, Rashba effect, Hall topological effect and Edelstein inverse effect [8].

In suggested article the activation analysis is performed of temperature dependence of dielectric permittivity  $\varepsilon(T)$  at low temperatures (5–300 K) of quantum ferroelectrics  $\text{KTaO}_3$  and  $\text{SrTiO}_3$ , as well as of solid solution based on them  $(\text{SrTiO}_3)_{0.85}-(\text{KTaO}_3)_{0.15}$ . This method was used previously to determine spectra of activation energies of domain and defect structures of classic ferroelectrics  $\text{BaTiO}_3$  and  $\text{Pb}(\text{Zr,Ti})\text{O}_3$  [9,10] based on determined temperature-activation mechanism of temperature dependence of dielectric permittivity  $\varepsilon(T)$  of ferroelectrics [11,12].

Note that the literature analysis of commonly accessible world scientific publications showed absence of data on activation analysis of temperature dependence of the dielectric permittivity  $\varepsilon(T)$  of quantum ferroelectrics  $\text{KTaO}_3$  and  $\text{SrTiO}_3$ , this confirms the novelty of the material in suggested publication.

### 2. Experimental procedure

In article the experimental data are analyzed relating measurements of dielectric permittivity of crystals  $\text{KTaO}_3$

and SrTiO<sub>3</sub>, obtained by authors of article [13], and of the solid solution (SrTiO<sub>3</sub>)<sub>0.85</sub>–(KTaO<sub>3</sub>)<sub>0.15</sub>, obtained by authors of article [14].

Studied sample KTaO<sub>3</sub> represents the crystal grown by seed method, in form of a wafer 10 × 10 × 0.5 mm with epipolished (100) surface (MTI Co.) [13]. The studied samples SrTiO<sub>3</sub> — crystals grown by Verneuil method in form of wafer with size 10 × 10 × 0.5 mm with mirror-finished surface (100) (Furuuchi Chemical) [13]. Gold electrodes were deposited on surface by sputtering. Dielectric permittivity (actual  $\varepsilon'$  and imaginary  $\varepsilon''$  parts) was measured using precision meter LCR (HP 4284A) with applied voltage 0.5 V and frequency 100 kHz. The samples temperature was monitored in temperature range 4–325 K using helium closed-loop refrigerator (Daikin Industries CG308SBR) [13].

Samples (SrTiO<sub>3</sub>)<sub>0.85</sub>–(KTaO<sub>3</sub>)<sub>0.15</sub> were prepared by authors of article [14] as per ceramic technology in form of thin wafer with sputtered gold-platinum electrodes. The complex dielectric permittivity was measured using analyzer 4192 LF Hewlett Packard Impedance Analyzer with amplitude value of metering variable field 100 V/cm with frequency 100 kHz. The samples temperature was monitored in temperature range 4.2–300 K using continuous-flow He-cryostat [14].

By obtained from experiment temperature dependences of the dielectric permittivity  $\varepsilon(T)$  the dependence  $\ln(\varepsilon) - 1/kT$  was plotted, and from sections of this dependence that meet the equation

$$\ln(\varepsilon(T)) = \pm \frac{U}{kT} + \ln(a) \quad (1)$$

Parameters of activation process (activation energy  $U$  and pre-exponential factor  $a$ ) are determined [9].

### 3. Results and discussion

According to ideas relating normal oscillations of atom sublattices of crystal, that are dipole, each such normal oscillation contributes to the dielectric permittivity  $\varepsilon$  of crystal according to expression

$$\varepsilon = \frac{\partial P}{\partial E} = \frac{zN}{m} \frac{1}{\omega_0^2 - \omega^2}, \quad (2)$$

where  $P$  — polarization associated with normal oscillation,  $E$  — amplitude of variable electric field with frequency  $\omega$ ,  $z$  — effective dynamic charge of this oscillation mode (equal to charge of ion for simplest model),  $N$  — number of ions participating in oscillation,  $m$  — reduced mass for this normal oscillation,  $\omega_0$  — frequency of normal oscillation [15].

Note that expression (2) is basic in Ginzburg–Anderson–Cochran (GAC) for ferroelectric transitions of offset type, which occur in the Ginzburg with perovskite structure.

Based on this, the dielectric permittivity can be assumed as macroscopic parameter, representing dynamics

of ferroelectric lattice and determined by phonon spectrum of crystal. At low temperatures the analysis of temperature dependences of dielectric permittivity of the ferroelectric ensures determination of processes of phonons activation at minimum effect of thermal excitations. For this in this paper the dependences  $\varepsilon(T)$  of quantum ferroelectrics KTaO<sub>3</sub>, SrTiO<sub>3</sub> and solid solution based on them (SrTiO<sub>3</sub>)<sub>0.85</sub>–(KTaO<sub>3</sub>)<sub>0.15</sub> are studied in temperature range 5–300 K, obtained by authors of papers [13] and [14], respectively (Figure 1).

To determine the parameters of thermal activation processes of dipole activity manifestation of elements of ferroelectric structure the experimental dependence  $\varepsilon(T)$  (Figure 1) is plotted in coordinates  $\ln(\varepsilon) - 1/kT$  (Figure 2). Sections, where this dependence is close to the linear dependence, are described by the equation (1), where it is converted into exponential equation

$$\varepsilon(T) = a \exp\left(\pm \frac{U}{kT}\right). \quad (3)$$

Equation (3), containing the Boltzmann probability factor, is valid for thermal activation processes and allows determination of the activation energy  $U$  of this process. Sign „±“ considers the decreasing or increasing of the dielectric permittivity during the ferroelectric heating.

Possibility of describing the temperature dependence of dielectric permittivity by equation in form (3) with single activation time is discussed in paper [11], based on this in paper [12] for the ferroelectric based on solid solution Pb(Zr,Ti)O<sub>3</sub> the equation for dependence  $\varepsilon(T)$  was obtained in ferroelectric phase with three activation energies.

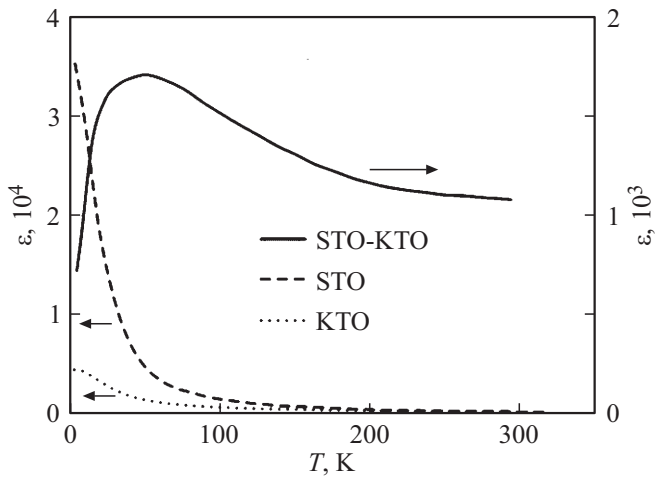
$$\varepsilon(T) = \sum_{i=1}^{i=3} a_1 \exp(a_2 U_i) (a_3 T - a_4)^{-1} \exp\left(-\frac{U_i}{kT}\right). \quad (4)$$

This approach application ensures for the ferroelectrics Pb(Zr,Ti)O<sub>3</sub> and BaTiO<sub>3</sub> determination of the activation processes of interaction of their domain and defect structures at temperatures below Curie temperature and of their activation energy. Exactly small oscillations of the domain walls at the activation energies below 0.1 eV; detachment of the domain walls from securing places on structure defects (mainly oxygen vacancies) at activation energies of 0.1 to 1 eV; the disintegration of the domain structure initiated by migration of the oxygen vacancies at activation energies above 1 eV [10].

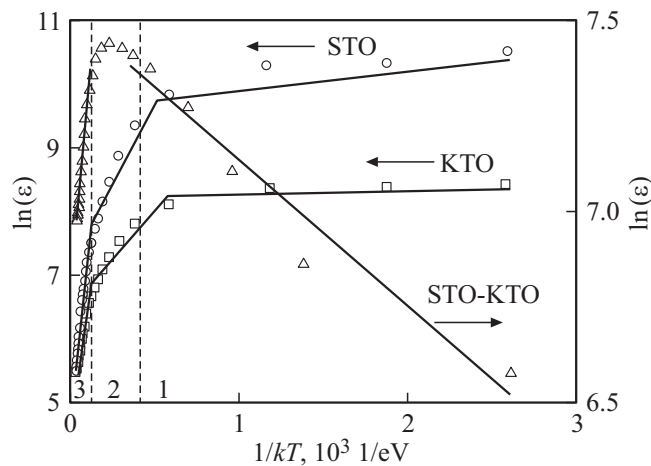
Results of processing the experimental data Figure 1, shown in Figure 2, indicate that for samples KTaO<sub>3</sub>, SrTiO<sub>3</sub> and (SrTiO<sub>3</sub>)<sub>0.85</sub>–(KTaO<sub>3</sub>)<sub>0.15</sub> on dependence  $\ln(\varepsilon) - 1/kT$  we can conditionally select three temperature ranges (1-st — 5–30; 2-nd — 30–80; 3-rd — 80–300 K), where the dependences can be approximated by the linear equation (3). Based on this, values of coefficient  $a$  and activation energy  $U$  are determined (Table 1). Consider that for samples KTaO<sub>3</sub> and SrTiO<sub>3</sub> in three selected sections of temperatures the transition from first section to the third one is accompanied by activation energy increasing. And for

**Table 1.** Values of coefficient  $a$  and activation energy  $U$  of equation (1) at 3 sections of dependence  $\ln(\varepsilon)(1/kT)$  (Figure 2) of studied samples

Sample	1-st section		2-nd section		3-rd section	
	$a$	$U, \text{eV}$	$a$	$U, \text{eV}$	$a$	$U, \text{eV}$
$\text{KTaO}_3$	3615.19	0.0001	632.13	0.0031	167.24	0.0131
$\text{SrTiO}_3$	14249.83	0.0003	1249.13	0.0050	117.89	0.0231
$(\text{SrTiO}_3)_{0.85}-(\text{KTaO}_3)_{0.15}$	1838.12	0.0004	—	—	902.44	0.0045



**Figure 1.** Temperature dependences  $\varepsilon(T)$  for ferroelectrics  $\text{KTaO}_3$  (KTO),  $\text{SrTiO}_3$  (STO) and  $(\text{SrTiO}_3)_{0.85}-(\text{KTaO}_3)_{0.15}$  (STO-KTO) as per data of [13] and [14].



**Figure 2.** Experimental (markers) and approximated (lines) dependences  $\ln(\varepsilon) - 1/kT$  for  $\text{KTaO}_3$  (KTO),  $\text{SrTiO}_3$  (STO) and  $(\text{SrTiO}_3)_{0.85}-(\text{KTaO}_3)_{0.15}$  (STO-KTO) in temperature ranges  $\Delta T$ : 1 — 5–30; 2 — 30–80; 3 — 80–300 K.

sample  $(\text{SrTiO}_3)_{0.85}-(\text{KTaO}_3)_{0.15}$  in the second section we observe maximum of dependence and sign change of activation energy, this further will be associated by us with activation process of phonon-phonon interaction.

Note that as per earlier studies of thermal capacity of ferroelectrics using Debye function for  $\text{KTaO}_3$  and  $\text{SrTiO}_3$  Debye temperatures (311 and 413 K respectively) [16] were determined, temperatures, at which temperature excitation of all frequencies of normal oscillations of crystal lattice. This means that for  $\text{KTaO}_3$  and  $\text{SrTiO}_3$  the monotonous increasing of the activation energy at sections 1–3 Figure 2 with rise of temperature to 300 K can be determined by gradual engagement in the oscillation process of individual normal modes of the crystal lattice of perovskite without significant phonon-phonon interaction between them. In solid solution  $(\text{SrTiO}_3)_{0.85}-(\text{KTaO}_3)_{0.15}$  in second temperature section of dependence  $\ln(\varepsilon)(1/kT)$  Figure 2, presumably, activation of phonon-phonon interaction of normal modes of oscillations inherent to  $\text{KTaO}_3$  and  $\text{SrTiO}_3$  occurs. And this leads to presence of wide maximum (about 100 K) in this section at  $T \approx 43$  K (Figure 2). The presence of this maximum may indicate a phase transition, since the value of the dielectric permittivity in the low-temperature region is noticeably lower than in the high-temperature region [14].

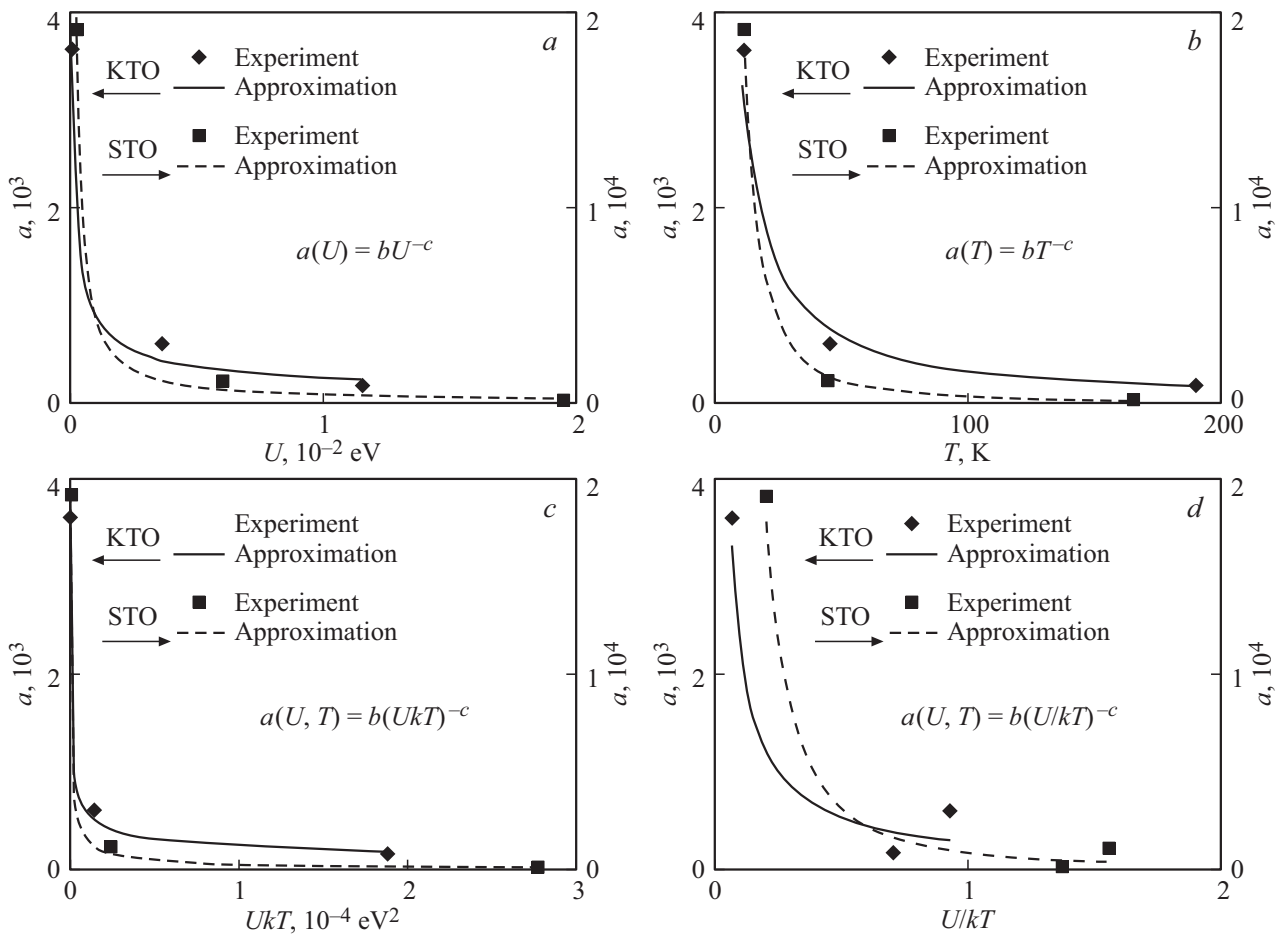
The activation analysis of the temperature dependence of dielectric permittivity of ferroelectrics  $\text{Pb}(\text{Zr,Ti})\text{O}_3$  and  $\text{BaTiO}_3$  permits earlier in paper [9] the determination for three activation processes occurred during heating to Curie temperature of the dependence of coefficient  $a$  of equation (3) on activation energy  $U$  for each of studied samples. This dependence has exponential view

$$a_i(U_i) = a_0 \exp(U_i/U_0), \tag{5}$$

where  $a_0$  and  $U_0$  — coefficients. Such general dependence ensures consideration of the process of approaching to ferroelectric phase transition as series transition between activation processes of dipole elements of structure, self-consistent by dependence (5). Note that self-consistency of the pre-exponential coefficient  $a$  is considered by us as repeatability of dependence (5) in full studied spectrum of activation energies of dipole elements of structure.

At that, unlike said results of paper [9], for quantum ferroelectrics  $\text{KTaO}_3$  and  $\text{SrTiO}_3$ , studied now, the analysis of results in Figure 2 and in Table 1 showed that the pre-exponential coefficient  $a$  can have dependence both on activation energy  $U$  and temperature  $T$ , and on their combination (Figure 3), i.e. can be self-consistency in full measured temperature range

$$a(U) = bU^{-c}, \tag{6}$$



**Figure 3.** Experimental (markers) and approximated (lines) dependences  $a(U)$  (a),  $a(T)$  (b) and  $a(U, T)$  (c,d) for samples  $\text{KTaO}_3$  (KTO) and  $\text{SrTiO}_3$  (STO).

$$a(T) = bT^{-c}, \tag{7}$$

$$a(U, T) = b(UkT)^{-c}, \tag{8}$$

$$a(U, T) = b(U/kT)^{-c}, \tag{9}$$

where  $b$  and  $c$  — coefficients. At that there is no exponential dependence in form (5), which is typical for classic ferroelectrics, this is new result in relation to study [9].

For all obtained in paper approximations of experimental and calculated parameters and coefficients the values of approximation authenticity  $R^2$  were calculated (Table 2).

The exponential dependence (3) for  $\varepsilon(T)$  can be considered as alternative to models of dielectric response of quantum ferroelectrics at low temperatures, which have definite areas of applicability. When studying the quantum ferroelectrics currently there are several approaches: Barrett theory [17] under framework of Slater single-ion model; theory Rechester theory [18] and Vendik theory [19] under framework of theory of soft ferroelectric mode. Authors of these models suggested equations for temperature dependences of the dielectric permittivity and frequency of soft ferroelectric mode.

The Barrett formula [17] is most known, and at the same time having some criticism:

$$\varepsilon(T) = M \left( \frac{1}{2} T_1 \coth\left(\frac{T_1}{2T}\right) - T_0 \right)^{-1}, \tag{10}$$

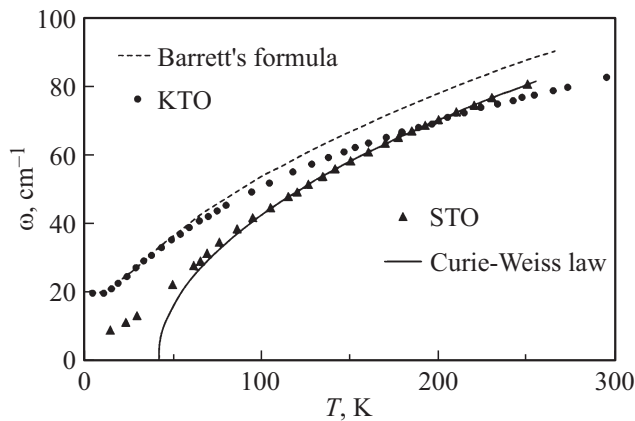
where  $M$  and  $T_0$  — parameters depending on values of electronic polarizability of various atoms of lattice and on geometry of ferroelectric lattice;  $T_1$  — characteristic temperature separating low-temperature region, where quantum effects are important, and Curie–Weiss law is not satisfied, from high-temperature region, where dependence  $\varepsilon(T)$  complies with Curie–Weiss law.

For quantum ferroelectrics the Barrett formula is widely used and amended [20]. Based on formula (10) we obtain also the expression for temperature dependence of soft mode frequency  $\varepsilon(T)$ . So, Figure 4 shows that experimental dependences  $\varepsilon(T)$  for  $\text{KTaO}_3$  and  $\text{SrTiO}_3$  at temperature about 80 K differ from the theoretical curves obtained by Barrett formula (9) and classic Curie–Weiss law, respectively.

In certain temperature ranges for some quantum ferroelectrics (e.g., for  $\text{SrTiO}_3$  at  $T$  from 10 to 75 K [22] or

**Table 2.** Values of approximation authenticity  $R^2$  of studied dependences  $f(x)$  for studied samples

$f(x)$		$R^2$		
		KTaO <sub>3</sub>	SrTiO <sub>3</sub>	(SrTiO <sub>3</sub> ) <sub>0.85</sub> –(KTaO <sub>3</sub> ) <sub>0.15</sub>
$\ln(\varepsilon)(1/kT)$	1	0.8713	0.8529	0.9542
	2	0.9254	0.9243	–
	3	0.9621	0.9199	0.9845
$a(U)$		0.9957	0.9993	–
$a(T)$		0.9957	0.9995	–
$a(UkT)$		0.9988	0.9997	–
$a(U/kT)$		0.9801	0.9972	–



**Figure 4.** Temperature dependences of polar soft modes of samples KTaO<sub>3</sub> (KTO) [21] and SrTiO<sub>3</sub> (STO) [6], approximated by Barrett formula (9) and Curie–Weiss law.

for KTaO<sub>3</sub> below 50 K [23]) the dependence of inverse dielectric permittivity on temperature is non-classical and has view

$$\varepsilon^{-1}(T) = A_0 + \alpha T^2, \quad (11)$$

This is explained by quantum theory of criticality amended by the effects of long-distance dipolar interactions and link of electric polarization field with acoustic phonons [23].

At that, as the model calculation and experiment show, the crossover from low-temperature behavior  $T^2$  to high-temperature classic behavior for KTaO<sub>3</sub> and SrTiO<sub>3</sub> occurs at temperatures 50–100 K [23], this approximately corresponds to the second temperature range selected by us in Figure 2. The temperature dependence of susceptibility near of both classic, and quantum critical points can be determined using self-consistent mean-field theory, where corrections to fluctuations, due to anharmonicity, are determined by fluctuation-dissipation theorem [24].

Note, that in paper [18] during study of quantum effects at second-order phase transition at low temperatures the temperature dependence of energy gap was calculated  $\delta\Delta(T)$  in spectrum of critical optical phonon in the approximation of theory of perturbation, and three temperature ranges were obtained. At low temperatures the dependence

$\delta\Delta^2(T)$  has exponential view ( $\sim \exp(-\Delta_0/T)$ ); at mean temperatures — quadratic dependence ( $\sim T^2$ ); at high temperatures — it is described by classic Curie–Weiss law.

Summary [25] experimentally showed that for SrTiO<sub>3</sub> and KTaO<sub>3</sub> the temperature dependences of dielectric permittivity  $\varepsilon(T)$  and soft mode frequency  $\varepsilon(T)$  have temperature  $T_1$ , above which  $\varepsilon(T)$  meets Curie–Weiss law, and  $\omega^2$  increases linearly with temperature under law  $A(T - T_0)$ . For SrTiO<sub>3</sub>  $T_1 = 100$  K,  $T_0 = 40$  K. For KTaO<sub>3</sub>  $T_1 = 30$  K,  $T_0 = 4$  K.

The suggested in our paper equation (3) for the temperature dependence of the dielectric permittivity is based on classic ideas of statistical physics, and, as it is shown in [26], at temperatures above Curie temperature it can be brought to Curie–Weiss law with Curie–Weiss constant  $C$ :

$$C = \frac{U - kT_0 \ln(a)}{k}, \quad (12)$$

where  $U$  — activation energy,  $T_0$  — Curie–Weiss temperature (close to Curie temperature  $T_C$ ),  $a$  — pre-exponential factor of equation (3),  $k$  — Boltzmann constant.

When discussing the quantum low-temperature limit (when  $T \ll T_1$ ), and considering that hyperbolic cotangent has view

$$\coth(x) = \frac{\exp(2x) + 1}{\exp(2x) - 1}, \quad (13)$$

Barrett formula (10) can be presented as dependence

$$\varepsilon(T) = A(M, T_0, T_1, T) \exp\left(\frac{T_1}{T}\right), \quad (14)$$

where  $M, T_0, T_1$  — ferroelectric parameters. Then Barrett formula can be brought to exponential equation (3).

So, we showed with complete certainty that, like in the case of classic ferroelectrics, for quantum ferroelectrics the dielectric response at low temperatures can be described by the exponential function containing Boltzmann probability factor  $\exp(U/kT)$ . This is due to the fact that the dielectric response of the ferroelectric is mainly determined by the phonon spectra of the crystal comprising normal oscillation modes of the crystal lattice. Deviation from such exponential dependence, apparently, is determined by the phonon-phonon interaction due to structural changes at definite temperatures, like for (SrTiO<sub>3</sub>)<sub>0.85</sub>–(KTaO<sub>3</sub>)<sub>0.15</sub> in temperature range 30–80 K (Figure 2), where according to opinion of authors of paper [14] the phase transition occurs.

## 4. Conclusion

Based on the analysis of temperature dependences of the dielectric permittivity  $\varepsilon(T)$  of quantum ferroelectric of offset type KTaO<sub>3</sub> and SrTiO<sub>3</sub> in temperature range 5–300 K it was determined that this dependence can be conditionally divided into three temperature ranges (1-st — 5–30; 2-nd — 30–80; 3-rd — 80–300 K), with different activation energy for each range. At that, the transition between the activation processes is self-consistent, this is

confirmed by the monotonous rise of activation energy with temperature increasing, and by self-consistent dependence of preexponential factor (on temperature and activation energy). And behavior of dielectric permittivity upon approaching the Debye temperature explains the series activation of phonons associated with normal oscillation modes of the crystal lattice. Excitation of higher high-temperature modes leads to decrease in activity of the dipole mode or its compensation by other modes of normal oscillations. This is confirmed by the decrease in dielectric permittivity with temperature increasing of samples  $\text{KTaO}_3$  and  $\text{SrTiO}_3$  from 5 to 300 K. At same time, maximum of the dielectric permittivity for solid solution  $(\text{SrTiO}_3)_{0.85}-(\text{KTaO}_3)_{0.15}$  in the second temperature range (30–80 K) can be manifestation of the phonon-phonon interaction of dipole active normal oscillation modes of the crystal lattice.

The obtained equation for the temperature dependence of the dielectric permittivity containing the Boltzmann probability factor ensures for the first time determination of the numerical values of activation energy of the processes associated with temperature excitation of phonons. In quantum low-temperature limit this equation is in agreement with known Barrett formula, and in high-temperature limit — with classic Curie–Weiss law for dielectric permittivity.

So, the possibility of theoretical description of dependence on temperature of the dielectric response parameters of both classic, and quantum (low-temperature) ferroelectrics located in the eternal electric field is shown, at that this description is valid under framework of model of thermoactivation processes with wide spectrum of values of activation energy upon both absence and presence of significant phonon-phonon interactions.

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## Conflict of interest

The author declares that she has no conflict of interest.

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