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The Local Order in KTaO_3 Admixture by the Ions of Li^+

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The low temperature behavior of the KTaO_3 type incipient ferroelectric crystals is of constant interest. The quantum fluctuations reduce the transition to the ferroelectric state in these crystals. It is possible that the small amount of the dopant Li^+ can lead, through the elastic interactions, to local glass-like short-range order or even to the relaxor ferroelectric order for $x > 2.6\%$. We presented that the low lithium doped $x = 0.005$ KTaO_3 crystals exhibit the dielectric dispersion of ε' and ε'' , suggesting the glass-like behavior in the low temperature range.

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1. Introduction

The crystal — studied by us — belongs to the group of crystals ABO_3 is similar to the SrTiO_3 and it is called the incipient ferroelectric, which does not soften even at the lowest temperatures [1]. This crystal does not experience the ferroelectric phase transition [2]. Stability of its paraelectric phase results from the projection of the quantum effects of the fluctuation [3–11].

The other interesting feature of this crystal is an influence of the admixtures on its physical properties. For the small concentrations of admixture ions and in the area of the low temperatures, the appearance of the glassy property of the interaction of the short range order is possible. For concentrations $x > 0.022$ the appearance of the polar clusters can be observed, which are characteristic of a relaxor. In this paper an influence of the admixture of Li^+ ions on the properties of the crystal KTaO_3 will be shown. The anomalies observed in the admixed crystal arise from the off-center position of Li^+ . In the small amount of the admixture ($x < 0.022$) the induced electric dipole interacts with each other by the elasticity coupling. This coupling leads to the local electric order [12–19].

Our aim is to show the existence of the areas with the local order in the low temperatures in the KTaO_3 (with a small amount of admixture Li^+ ions) and to make an attempt to their identification depending on the amount of the admixture.

2. Experimental method

The subject of the research were the monocrystals of the KTaO_3 admixed with Li^+ ions. The following crystals of $\text{K}_{1-x}\text{Li}_x\text{TaO}_3$ were tested:

$$x = 0.005 \quad (1.6 \times 3.5 \times 0.41) \text{ mm,}$$

$$x = 0.01 \quad (1.7 \times 3.8 \times 0.72) \text{ mm,}$$

$$x = 0.03 \quad (1.52 \times 4.15 \times 1) \text{ mm.}$$

The properties of the crystals were measured along the crystallographic direction [100]. The silver paste was used for electrodes. Measurements were performed in the range of frequency of the *ac* electric field from 20 kHz to 1 MHz by means of the RLC bridge HP 4275A, in the flow helium cryostat in the temperature range from 3.5 K to 300 K.

3. Results

3.1. The influence of the lithium admixture on the dielectric properties of the KTaO_3

Temperature dependences of the real ϵ' and imaginary ϵ'' parts of the electric permittivity of $\text{K}_{1-x}\text{Li}_x\text{TaO}_3$ ($x = 0.03$, $x = 0.01$, $x = 0.005$) are shown in Fig. 1. The observed dependences show an influence of Li^+ on the dielectric properties of the crystal KTaO_3 . For the concentration $x = 0.03$ the temperature dependence of ϵ' is typical of the relaxors, with characteristic dielectric dispersion [14–17, 19], with the fast increase in the electric permittivity ϵ' value with lowering temperature till getting the value of the real part of the electric permittivity ϵ' . In the low

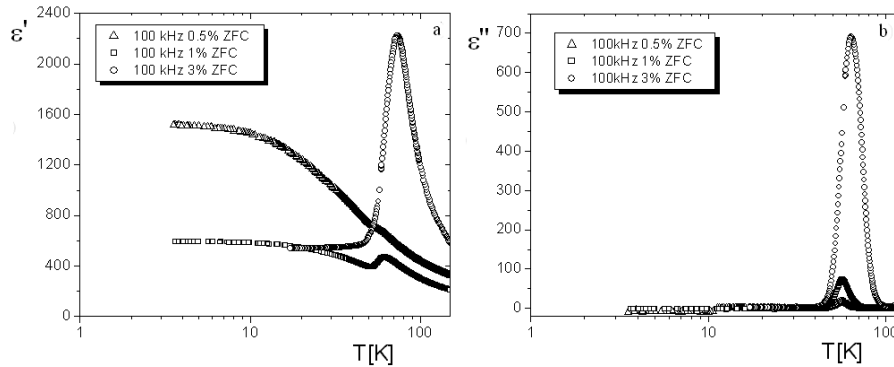


Fig. 1. Temperature dependence of the electric permittivity ϵ' (a) and the imaginary part of the electric permittivity ϵ'' (b) for given concentrations of Li^+ ions at the frequency of *ac* measuring electric field 100 kHz.

temperatures ($T < 40$ K) changes of the electric permittivity ϵ' were not observed for this concentration. For the concentration below the critical value ($x_c < 0.022$), i.e. $x = 0.01$ and $x = 0.005$, the other increase in the electric permittivity ϵ' was observed in this area. For the lower concentration ($x = 0.005$) this increase is much more stronger and the received values ϵ' are bigger and close to those in the pure $KTaO_3$.

3.2. The local order in $K_{1-x}Li_xTaO_3$ crystals

The temperature dependences of the electric permittivity ϵ' and ϵ'' for different frequencies in crystal $KTaO_3$ with admixture Li^+ $x = 0.01$ and $x = 0.005$ are shown in Figs. 2 and 3. For these both concentrations the characteristic dispersion of $\epsilon'(T)$ and $\epsilon''(T)$ are detected in the temperatures range from 40 K to 80 K.

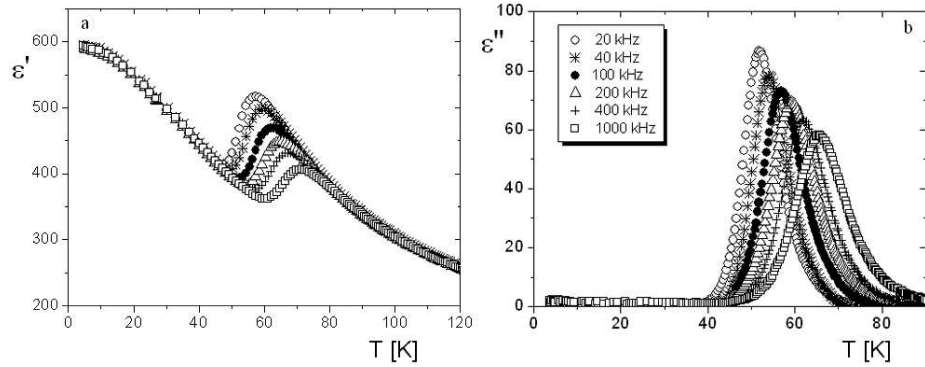


Fig. 2. Dispersion of the electric permittivity ϵ' (a) and the imaginary part of the electric permittivity ϵ'' (b) of $K_{1-x}Li_xTaO_3$ with $x = 0.01$ for given values of the frequency.

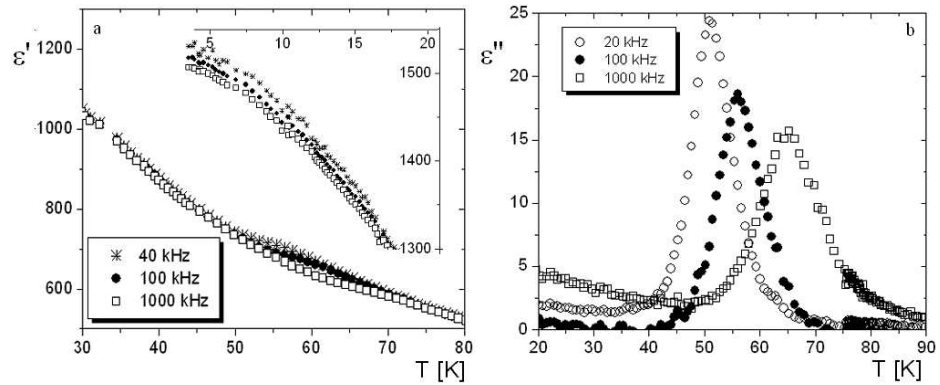


Fig. 3. Dispersion of the electric permittivity ϵ' (a) and the imaginary part of the electric permittivity ϵ'' (b) of $K_{1-x}Li_xTaO_3$ with $x = 0.005$ for given values of the frequency.

An interesting fact is that for the concentration $x = 0.005$ the dielectric dispersion occurs in the area of the lower temperatures. For the electric permittivity $\varepsilon'(T)$ this dispersion begins below the temperature $T = 20$ K (the inset in Fig. 3a). In the temperature dependence of the imaginary part of the electric permittivity ε'' the increase in values ε'' below the temperature $T = 40$ K (Fig. 3b) is detected.

4. Discussion and conclusions

The physical properties of the KTaO_3 crystal can be modified considerably by the collective effects induced by the impurity. These modifications include impurity — induced fluctuations, metastable polarization, phase transition leading to an ordered or dipolar glassy state, etc. The origin of the anomalous behavior in $\text{K}_{1-x}\text{Li}_x\text{TaO}_3$ crystals lies in the fact that the impurities (Li^+ ions) occupy off-center sites and exhibit relaxational motion between equivalent sites with a characteristic time that is temperature dependent. In addition, this off-center occupancy of the impurity has the consequence of polarizing an extended region of host cells surrounding it. The precursor of polar regions significantly enhance the piezoelectric properties by allowing a strong electrostrictive coupling between polarization and strain. The alignment of the polar region provides a sufficient macroscopic polarization for the piezoelectric response to become effective.

The KTaO_3 crystal is a ferroelastic, thus an appearance of *improper glass* phase is possible. The order parameter characteristic of this phase is not the spontaneous polarization, but spontaneous deformation. Therefore, by testing the properties of this crystal, the elastic coupling should be taken into consideration through which the induced dipole moments can interact, even in the small concentration of the admixture. Hence, alike to the tested crystals $\text{KTaO}_3 : \text{Fe}$ with $x = 0.01\%$ and $x = 1\%$ [12, 13], the appearance of a coexistence of polar clusters is possible, in the area of which the short-range order with the paraelectric phase is realized. This phase is called the *improper glass*. Glinchuk theory [19] enables counting the correlation length r_c of interacting dipole moments. In the case of $\text{KTaO}_3 : \text{Li}^+$ this parameter (the size of polar clusters) is $r_0 = 10^{-8}$ m for the concentration $x = 0.01$ and $r_0 = 1.3 \times 10^{-8}$ m for the concentration $x = 0.005$.

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