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Lithium-induced dielectric relaxations in potassium tantalate ceramics

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Abstract

This work reports the effect of lithium doping on the dielectric and polar properties of potassium tantalate. Experimental data were obtained in $K_{1-x}Li_xTaO_3$ ceramics with $x = 0, 0.02, 0.05$ and 0.10 by measuring both the dielectric permittivity from 10^2 to 10^8 Hz, and polarization under an ac electric field driven at 2.5 Hz, for temperatures from 10 to 300 K. The dielectric permittivity exhibits all the relaxations reported for $K_{1-x}Li_xTaO_3$ single crystals. Two dielectric relaxations observed at 40–125 K are ascribed to the individual hopping by 90° and 180° of dipoles created by the off-centre Li ions. Another relaxation observed at 100–200 K is related to 180° -flips of Li pairs for $x = 0.02$ and of polar clusters of interacting Li ions for $x = 0.05$ and 0.10 . In addition to that, an additional relaxation not reported before is presented at 135–235 K for $x = 0.10$ and attributed to 90° -reorientation of Li polar clusters. Both the change from an Arrhenius to a Vogel–Fulcher dependence with increasing lithium content, and the emergence of slim $P(E)$ hysteresis loops around the relaxation temperatures show that the relaxation dynamics in $K_{1-x}Li_xTaO_3$ can be understood if a crossover from a dipolar glass to a relaxor-like behaviour is assumed.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Among perovskites having general formula ABO_3 , there exist three compounds, $SrTiO_3$, $KTaO_3$ (KT) and $CaTiO_3$ [1], which possess polar soft modes but do not exhibit any ferroelectric phase transition down to 0 K. These materials are classified as incipient ferroelectrics and possess a highly polarizable lattice, thus their properties are very sensitive to defects, impurities, elastic and applied electric fields. These factors can easily induce ferroelectricity, intermediate glass- or relaxor-like states, making such systems very attractive from both a fundamental and a technological point of view. In fact, they are attractive candidates for processing microwave devices, nonvolatile memories and photocatalytic water splitters [2–4].

Since KT is very sensitive to impurities, several kinds of substitution ions have been used to study their effect on its properties. In particular, $K_{1-x}Li_xTaO_3$ (KLT) solid solutions with $x < 0.16$ has been intensively studied, wherein lithium ions substitute for K ions on perovskite A sites, having a

coordination number (CN) of 12. The substitution of smaller Li^+ ions ($r_{Li^+} = 1.25 \text{ \AA}$ extrapolated for CN = 12) for larger K^+ ions ($r_{K^+} = 1.64 \text{ \AA}$ for CN = 12) [5] yields a displacement from the centre of A sites by about $\sim 1.26 \text{ \AA}$ along the cubic [001] direction [6]. Displacements of Li ions generate strong local dipole moments (Li dipoles), which couple electrostatically to the KT polar soft mode. The combination of Li-dipoles relaxations, their interactions, and the soft mode leads to complex nature of the dielectric response in KLT [7, 8].

Dielectric measurements on KLT single crystals with $0 < x < 0.02$ reveal only one relaxational peak in $\epsilon(T)$ at low temperature, while for $x > 0.02$, two peaks are observed. These two peaks have been associated with two distinct relaxational processes, termed low- T and high- T relaxations [9–11]. The low- T process with an activation energy of $78 < U < 125$ meV has been associated with isolated Li ions hopping from one (001) position to another [12]. However, the high- T process with an activation energy

of $181 < U < 241$ meV is poorly understood. On the basis of acoustic measurements, it was suggested that this latter process could be due to 180° reorientations of Li ions within dipole pairs [10].

While $K_{1-x}Li_xTaO_3$ single crystals have been intensively studied with considerable success in the last decades, to the best of our knowledge no systematic experimental work has yet been carried out in KLT-based ceramics, so far. This is because polycrystalline $KTaO_3$ is hard to process as a phase pure stoichiometric compound with relatively high density. However, there are several important advantages of ceramics, compared with single crystals: (1) ceramics are less expensive to be processed than single crystals; (2) the doping control in ceramics (including dopant amount and site occupancy) is easier than in single crystals; and (3) ceramics can be tailored by processing in a large variety of shapes, sizes and other properties, making them distinctive candidates for a wider range of applications than single crystals. Currently, they are also highly requested as targets for processing and developing thin films and super-lattices for applications in emergent technologies. Thus, the aim of this work is a systematic study of $K_{1-x}Li_xTaO_3$ by dielectric spectroscopy and polarization inversion, using ceramic samples. The experimental data will be further analysed with comprehensive theoretical models, and the relaxation characteristic parameters obtained thoroughly compared with those earlier reported for single crystals.

2. Experimental procedure

$KTaO_3$, $K_{0.98}Li_{0.02}TaO_3$, $K_{0.95}Li_{0.05}TaO_3$, and $K_{0.90}Li_{0.10}TaO_3$ ceramic samples were prepared by conventional mixed oxide method. Dried grade K_2CO_3 , Li_2CO_3 and Ta_2O_5 reagents were weighed according to the aforementioned compositions with initial (K+Li)/Ta ratio = 1.05 in order to compensate the 5% loss of alkali ions during sintering [13, 14]. After ball milling in alcohol for 5 h using teflon pots and zirconia balls in a planetary mill, the powders were dried, and then calcined at $875^\circ C$ for 8 h. The calcined powders were milled again for 5 h to obtain powders with particle size lower than $5 \mu m$. Pellets of 10 mm in diameter were uniaxially pressed at 100 MPa, covered by powder of the same composition to decrease the loss of potassium, and sintered in closed alumina crucibles at $1300\text{--}1350^\circ C$ for 1 h with a heating and cooling rate of $5^\circ C \text{ min}^{-1}$.

The density of the sintered samples, measured by the Archimedes' method using diethylphthalate as the immersion liquid, is $\sim 88\%$. Room temperature x-ray diffraction analysis did not detect any secondary phase. The grain size was obtained using scanning electron microscopy to be about $6 \mu m$. For the dielectric measurements, sintered samples were polished and gold electrodes were sputtered on both sides.

The real (ϵ') and imaginary (ϵ'') parts of the dielectric permittivity in the frequency range $10^2\text{--}10^8$ Hz as well as the electric polarization versus electric field at 2.5 Hz were measured, using both a Precision LCR Meter (HP 4284A) and a RF Impedance Analyzer (HP 4191A), and a ferroelectric tester (TF analyser 1000 AIXACT) with amplifier, respectively. The

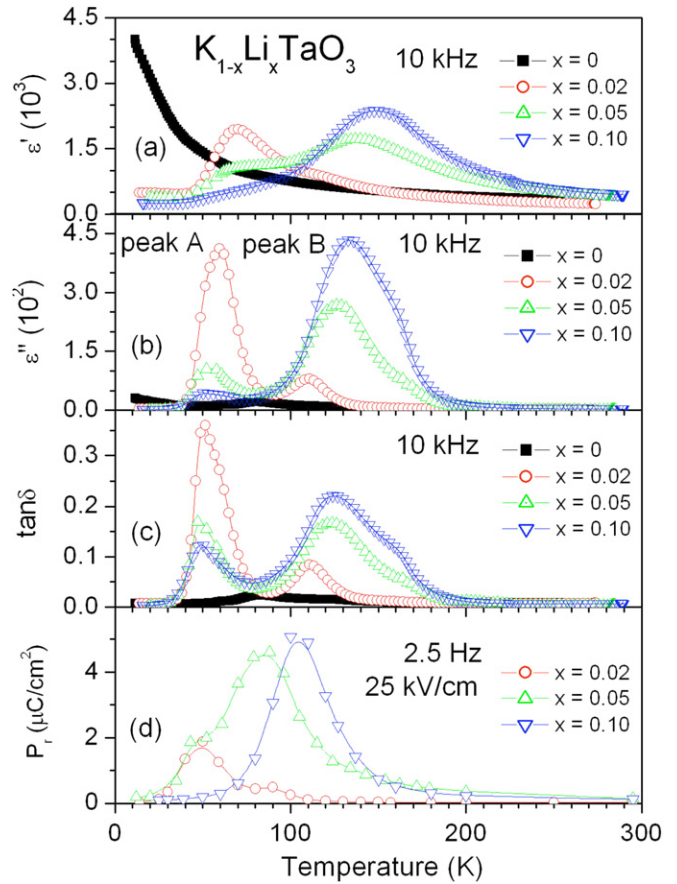


Figure 1. Real ϵ' (a) and imaginary ϵ'' (b) parts of the dielectric permittivity, dissipation factor $\tan \delta$ (c), and remnant polarization P_r (d) of $K_{1-x}Li_xTaO_3$ ceramics with $x = 0, 0.02, 0.05$ and 0.10 as a function of temperature.

temperature could be altered from 10 to 300 K using a He closed cycle cryogenic system (Displex ADP-Cryostat HC-2), equipped with a digital temperature controller (Scientific Instrument Model 9650). The temperature was measured with a silicon diode with a precision of ~ 0.1 K.

3. Results and discussion

3.1. Dielectric anomalies induced by Li doping in $KTaO_3$

The temperature dependence of the real and imaginary parts of the dielectric permittivity ($\epsilon' - j\epsilon''$), as well as of the dissipation factor, $\tan \delta = \epsilon''/\epsilon'$, measured at a frequency of 10 kHz in a heating run on $K_{1-x}Li_xTaO_3$ ($x = 0, 0.02, 0.05$, and 0.10) ceramics, is presented in figures 1(a), (b) and (c), respectively. As seen from figure 1(a), undoped $KTaO_3$ ceramics reveal a classic incipient ferroelectric behaviour, i.e. a continuous increase of ϵ' with temperature decrease similar to that of $KTaO_3$ single crystals. In contrast to undoped $KTaO_3$, $\epsilon'(T)$ of Li-doped KT ceramics reveal a diffuse peak, whose amplitude and position depend on Li content. As shown in figures 1(b) and (c), both $\epsilon''(T)$ and $\tan \delta(T)$ of Li-doped compounds exhibit two peaks: low-temperature peak A about 60 K and high-temperature peak B about 110–135 K at 10 kHz.

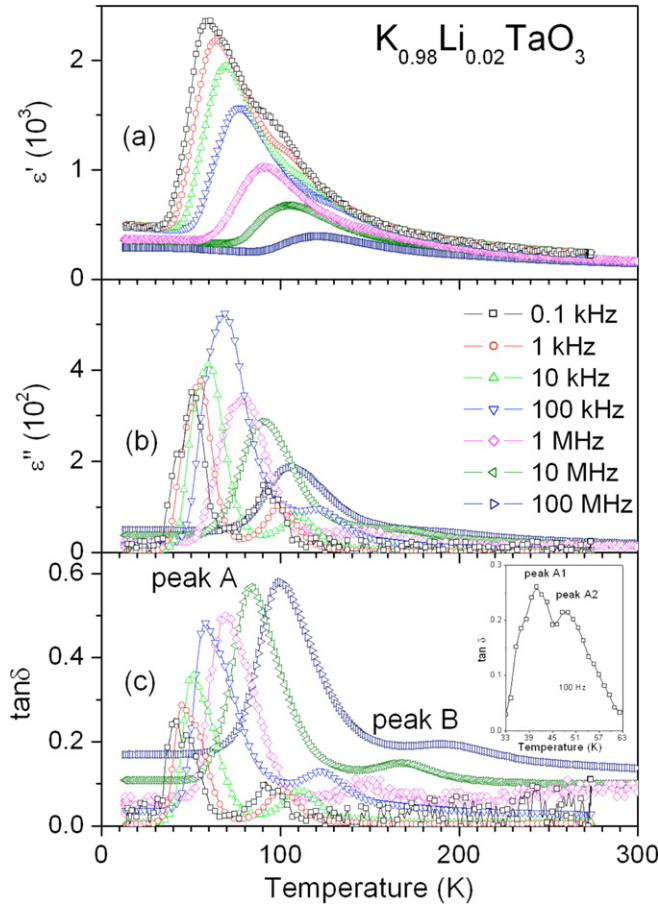


Figure 2. Real ϵ' (a) and imaginary ϵ'' (b) parts of the dielectric permittivity, and dissipation factor $\tan \delta$ (c) of $\text{K}_{0.98}\text{Li}_{0.02}\text{TaO}_3$ ceramics at different frequencies from 10^2 to 10^8 Hz as a function of temperature.

The effect of the frequency (from 100 Hz to 100 MHz) on the dielectric response of $\text{K}_{1-x}\text{Li}_x\text{TaO}_3$ ceramics with $x = 0.02, 0.05$ and 0.10 is shown in figures 2, 3 and 4, respectively. In contrast to undoped KTaO_3 , where no significant frequency dispersion is observed (not shown), $\epsilon'(T)$ and $\epsilon''(T)$ of Li-doped KTaO_3 strongly depend on the frequency. This frequency dependence undoubtedly reveals the relaxation nature of the dielectric permittivity peaks. Peak temperatures increase whereas the real part of the dielectric permittivity decreases with increasing frequency, corresponding in fact to a relaxation-like dispersion. Moreover, the low-frequency data show that for all Li-doped KT under study, peak A consists of two contributions A1 and A2, as shown in the inset of figure 2. Furthermore, for $x = 0.10$, peak B is also shown to include two contributions, labelled as B1 and B2 in the inset of figure 4. Relaxations B1 and B2 were detected for $x = 0.05$ as well, but the relaxation dynamics could be further defined only for the dominant contribution B1.

3.2. Analysis of the dielectric relaxations of $\text{K}_{1-x}\text{Li}_x\text{TiO}_3$ ceramics

The analysis of each dielectric relaxation was carried out assuming a Debye approximation. Accordingly, the set of

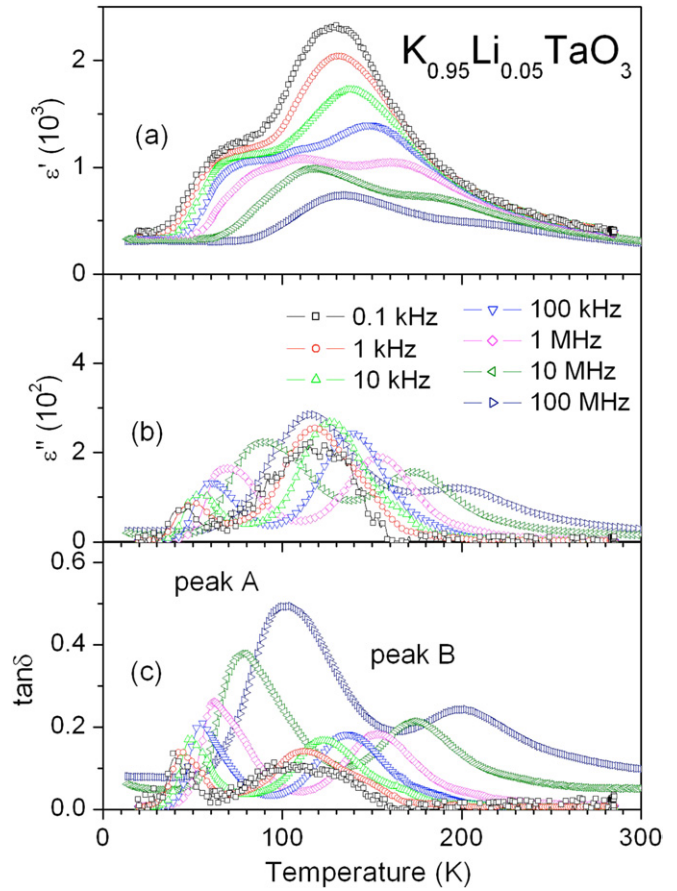


Figure 3. Real ϵ' (a) and imaginary ϵ'' (b) parts of the dielectric permittivity, and dissipation factor $\tan \delta$ (c) of $\text{K}_{0.95}\text{Li}_{0.05}\text{TaO}_3$ ceramics at different frequencies from 10^2 to 10^8 Hz as a function of temperature.

independent dipoles is characterized by a unique relaxation time (τ), which is equal to the inverse of the angular relaxation frequency (ω). All the parameters of the relaxation dynamics were deduced from the temperature dependence of ϵ'' . First, all the relaxations were described by an Arrhenius law:

$$\tau = \tau_0 \exp(U/k_B T), \quad (1)$$

where τ_0 is the relaxation time at infinite temperatures, and U is the activation energy of the dipolar process. In order to determine τ_0 and U a linear fitting to the experimental data plotted as $\ln(\omega^{-1})$ versus $1000/T_{\epsilon''m}$ is carried out in the frequency range 10^2 – 10^8 Hz.

As shown in figure 5, dynamics of both contributions of peak A in all studied KLT ceramics is well described by the Arrhenius law with the relaxation time pre-exponents $\tau_{0A1}, \tau_{0A2} \sim 10^{-13}$ s, and activation energies $U_{A1} \sim 77$ meV and $U_{A2} \sim 96$ meV, as presented in table 1.

Dynamics of peak B for $x = 0.02$ could be analysed in the same way as referred to above (figure 5), wherein $\tau_{0B} \sim 10^{-15}$ s and $U = 212$ meV (see table 2). However, when following the same procedure for peak B1 in $\text{K}_{0.95}\text{Li}_{0.05}\text{TaO}_3$ and peaks B1 and B2 in $\text{K}_{0.90}\text{Li}_{0.10}\text{TaO}_3$ ceramics, a $\tau_0 \sim 10^{-18}$ s was obtained, which is too small to have any physical meaning. Conversely, dynamics of peak B could be achieved

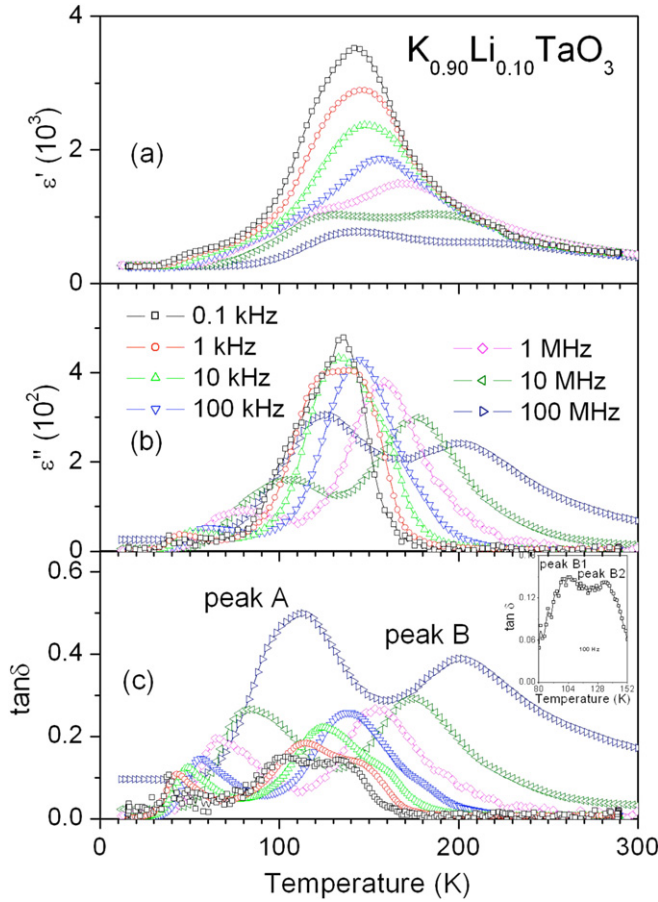


Figure 4. Real ϵ' (a) and imaginary ϵ'' (b) parts of the dielectric permittivity, and dissipation factor $\tan \delta$ (c) of $\text{K}_{0.90}\text{Li}_{0.10}\text{TaO}_3$ ceramics at different frequencies from 10^2 to 10^8 Hz as a function of temperature.

for the temperature dependence of the relaxation time using the Vogel–Fulcher relation:

$$\tau = \tau_0 \exp(U/k_B[T - T_f]), \quad (2)$$

where T_f stands for the freezing temperature of the relaxation process. The fitting procedure yields $\tau_0 \sim 10^{-13}$ s, $U \sim 97$ meV and $T_f \sim 63$ K for peak B1, as presented in table 2. For peak B2 in $\text{K}_{0.90}\text{Li}_{0.10}\text{TaO}_3$ ceramics, $\tau_0 \sim 10^{-14}$ s, $U \sim 81$ meV and $T_f \sim 94$ K were obtained as shown in table 2 as well.

Here we would like to note that the relaxation processes following the Arrhenius law were widely observed in Li-doped KT single crystals by a number of techniques, but usually no more than two relaxations were found [10, 12]. The exception is the early work by Prosandeev *et al* on $\text{K}_{0.957}\text{Li}_{0.043}\text{TaO}_3$ single crystal, where relaxation processes with activation energies of 85, 104 and 241 meV (i.e. similar to 77, 96 and 212 meV obtained in this work) were reported [11]. First relaxation was associated with Fe-related defects, usually detected in KTaO_3 as ‘an unavoidable impurity’ [11]. However, similar parameters obtained by Prosandeev *et al* in weakly doped $\text{K}_{0.994}\text{Li}_{0.006}\text{TaO}_3$ single crystal were associated with the $\pi/2$ -relaxation [15]. Moreover, our analysis of the dielectric response of undoped KTaO_3 ceramics prepared in the

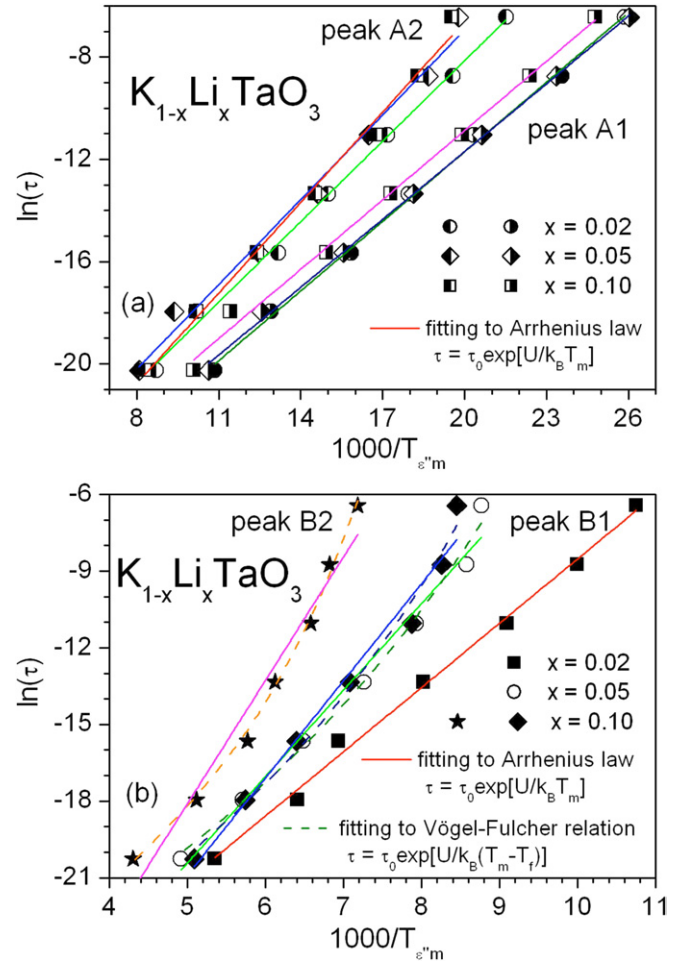


Figure 5. Arrhenius plots $\ln(\tau)$ versus $1000/T_{\epsilon''m}$ for relaxations A (a) and B (b) of $\text{K}_{1-x}\text{Li}_x\text{TaO}_3$ ceramics with fits to the Arrhenius law (solid lines) and the Vogel–Fulcher relation (dash lines).

Table 1. Arrhenius law parameters for the dynamics of the dielectric relaxations A in $\text{K}_{1-x}\text{Li}_x\text{TaO}_3$ ceramics.

Relaxation	x	U (meV)	τ_0 (s)
A1	0.02	78	2.7×10^{-13}
	0.05	78	1.1×10^{-13}
	0.10	76	1.7×10^{-13}
A2	0.02	102	0.8×10^{-13}
	0.05	91	2.2×10^{-13}
	0.10	95	2.4×10^{-13}

same way as Li-doped ceramics does not yield any relaxation process with such parameters, disproving the ‘unavoidable impurity’ mechanism. The second and third relaxations of $\text{K}_{0.957}\text{Li}_{0.043}\text{TaO}_3$, observed in the single crystals, were associated with the off-centre $\pi/2$ -rotations of Li^+ , and the π -rotations of Li^+ pairs, respectively [11].

As referred to above, there are several works on Li-doped KT single crystals that report values of U and τ_0 parameters, obtained by fitting the Arrhenius law to the experimental data. According to the values of those parameters, several mechanisms have been proposed for the observed dielectric relaxations. In this work, we have systematized the earlier reported results, as listed in table 3,

Table 2. Parameters of the Arrhenius law and the Vogel–Fulcher relation for the dynamics of the dielectric relaxations B in $K_{1-x}Li_xTaO_3$ ceramics.

x	Relaxation	Arrhenius law		Vogel–Fulcher relation		
		U (meV)	τ_0 (s)	U (meV)	τ_0 (s)	T_f (K)
0.02	B	212	3.3×10^{-15}			
0.05	B1	292	6.1×10^{-17}	99	6.9×10^{-13}	59
0.10	B1	328	4.7×10^{-18}	96	4.6×10^{-13}	66
0.10	B2	416	4.6×10^{-19}	81	1.6×10^{-12}	94

Table 3. Relaxation mechanisms observed in the $K_{1-x}Li_xTaO_3$ system and their Arrhenius-law parameters.

Proposed mechanism	x	U (meV)	τ_0 (s)	Ref.
Quadrupolar relaxation–reorientation of Li^+ between equivalent sites on orthogonal axes by 90° ($\pi/2$ flipping)	0.006	78	1.4×10^{-13}	[15]
	0.009	86	1.3×10^{-13}	[16]
	0.010	84	4.1×10^{-13}	[17]
	0.016	86	3.4×10^{-13}	[17]
	0.011	86	0.2×10^{-13}	[12]
	0.015	86	6.0×10^{-13}	[10]
	0.035	86	6.0×10^{-13}	[10]
	0.050			
	0.025	86	50×10^{-13}	[9]
	0.033	86	8.0×10^{-13}	[9]
Dipolar relaxation–reorientation of Li^+ between equivalent sites on the same axis by 180° (π flipping)	0.035	86	1.6×10^{-13}	[18]
	0.043	85	0.5×10^{-13}	[11]
	0.011	98	1.0×10^{-13}	[12]
	0.012	125	16×10^{-13}	[19]
	0.016	95	0.6×10^{-13}	[20]
	0.026	95	0.6×10^{-13}	[20]
	0.026	95	0.3×10^{-13}	[21]
	0.025	86	8.0×10^{-13}	[9]
	0.033	112	0.8×10^{-13}	[9]
	0.034			
Reorientation of a pair of coupled dipoles in head-to-tail configurations along the same axis by 180° (π pair flipping)	0.050	99	1.0×10^{-13}	[22]
	0.070			
	0.043	104	0.8×10^{-13}	[11]
	0.025	220	25×10^{-15}	[9]
	0.033	229	10×10^{-15}	[9]
	0.035	233	1.4×10^{-15}	[10]
	0.050	233	1.4×10^{-15}	[10]
	0.043	241	0.9×10^{-15}	[11]
	0.035	207	6.9×10^{-15}	[23]
	0.050	208	5.3×10^{-15}	[23]
0.100	206	8.3×10^{-15}	[23]	
0.160	227	1.8×10^{-15}	[23]	
0.036				
0.049	216		[20]	
0.063				
0.070	181–200		[22]	

to avoid any misinterpretations. Three main groups can be identified in terms of activation energies. The first group, with $U = 78$ – 86 meV obtained for KLT crystals with $x = 0.006$ – 0.050 [9–12, 15–18], is characterized by a quadrupolar relaxation, i.e. reorientation of Li^+ between equivalent sites on orthogonal axes by 90° ($\pi/2$ flipping) [10, 12]. Since these values of activation energy are close to those obtained in this work for the relaxation A1, it can also be associated with individual $\pi/2$ flipping of small lithium ions on the large

potassium sites in KLT ceramics. The second group, with $U = 86$ – 125 meV obtained for KLT crystals with $x = 0.011$ – 0.070 [9, 11, 12, 19–22], is related to dipolar relaxation, i.e. reorientation of Li^+ between equivalent sites on the same axis by 180° (π flipping) [12]. In this range of activation energy also falls the relaxation A2, thus revealing that the corresponding relaxation mechanism is assigned to the π flipping of off-central Li_K^+ in KLT ceramics. The third group, with $U = 181$ – 241 meV obtained for KLT crystals with $x = 0.025$ – 0.160 [9–11, 20, 22, 23], is ascribed to the reorientation of pairs of coupled dipoles in head-to-tail configurations along the same axis by 180° (π pair flipping) [10]. An activation energy inside that range is obtained in this work for KLT with $x = 0.02$, revealing that relaxation B in this composition stems from the π flipping of Li_K^+ pairs.

Regarding the fitting of the Vogel–Fulcher relation to the high- T experimental data in KLT ceramics with $x = 0.05$ and 0.10 , the relaxation parameters obtained evidence the apparent off-centre displacements of small lithium ions on the large potassium sites within polar clusters [7]. Moreover, the similarity of the activation energy for relaxations A2 and B1 as well as A1 and B2 implies the same type of the Li reorientations. The difference is just that Li ion flipping is individual for low- T A relaxations, while it is collective for high- T B relaxations, which then become frozen on cooling down. In contrast to Arrhenius law, there are only few works reporting applicability of the Vogel–Fulcher relation to KLT system. Laguta *et al* obtained $\tau_0 \sim 10^{-14}$ s, $U_{A1} \sim 79$ meV and $T_f \sim 12$ K for $K_{0.97}Li_{0.03}TaO_3$ [17], whereas Yokota *et al* reported parameters $\tau_0 \sim 10^{-12}$ s, $U_{A1} \sim 88$ meV and $T_f \sim 56$ K for $K_{0.93}Li_{0.07}TaO_3$ [24]. The later parameters are close to those obtained in this work for $K_{0.95}Li_{0.05}TaO_3$ and $K_{0.90}Li_{0.10}TaO_3$. Applicability of the Vogel–Fulcher relation is a characteristic feature of relaxors [17].

3.3. Polar behaviour of $K_{1-x}Li_xTaO_3$ ceramics

The emergence of slim S -shaped $P(E)$ hysteresis loops is a characteristic feature of disordered and frustrated systems like dipolar glasses and relaxors [18]. Hence, in order to figure out the actual nature of the dielectric relaxations in $K_{1-x}Li_xTaO_3$ ceramics, $P(E)$ measurements were carried out over the temperature range from 12 to 295 K. Figures 6, 7, and 8 show several characteristic $P(E)$ curves for $x = 0.02$, 0.05 and 0.10 , respectively. Well developed S -shaped hysteresis loops with non-zero remnant polarization P_r suggest the existence of either dipolar glass or relaxor-type states in KLT ceramics at low temperatures, as seen from figures 6 to 8. At room temperature the polarization versus electric field response is linear for all the samples.

Remnant polarization P_r deduced from the loops obtained at electric field of 25 kV cm^{-1} is plotted at the bottom panel of figure 1 as a function of temperature. $P_r(T)$ reveals a distinctive similarity to the behaviour of the dielectric loss, despite the difference in the test signal frequency (10 kHz for dielectric measurements and 2.5 Hz for $P(E)$ measurements). The temperature dependence of P_r is actually characteristic

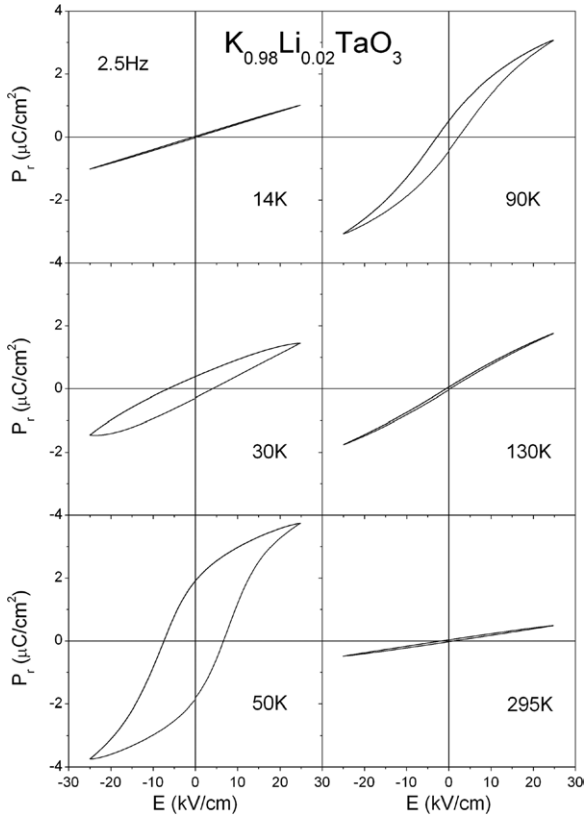


Figure 6. Polarization P versus electric field E of $K_{0.98}Li_{0.02}TaO_3$ ceramics at 2.5 Hz at selected temperatures from 10 to 295 K.

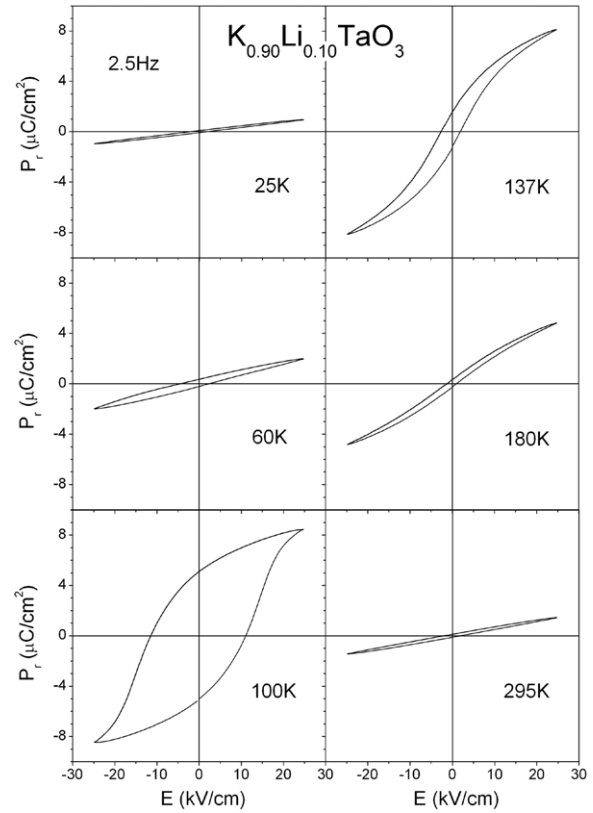


Figure 8. Polarization P versus electric field E of $K_{0.90}Li_{0.10}TaO_3$ ceramics at 2.5 Hz at selected temperatures from 10 to 295 K.

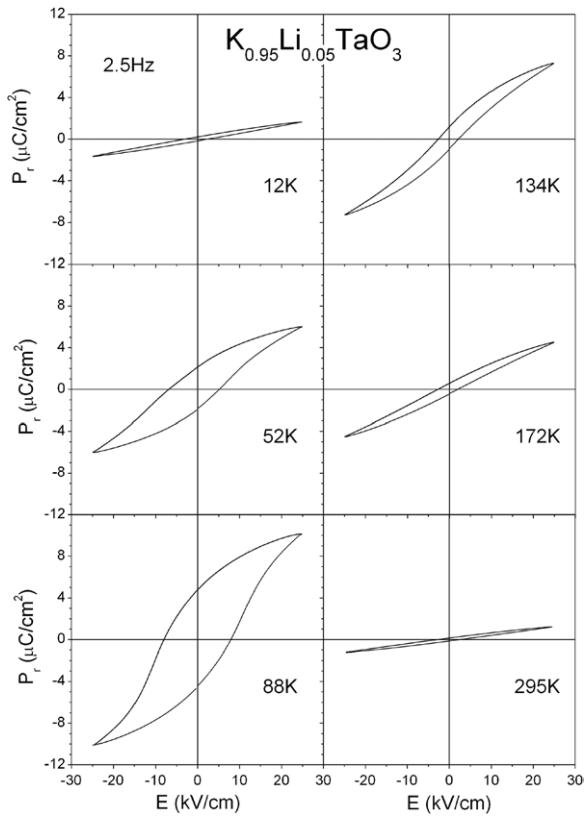


Figure 7. Polarization P versus electric field E of $K_{0.95}Li_{0.05}TaO_3$ ceramics at 2.5 Hz at selected temperatures from 10 to 295 K.

of dipolar glasses and relaxors in unpoled state, revealing peaks around the maximum amplitude temperatures of A and B relaxations, and a smooth decrease of P_r with increasing temperature, in evident contrast to classic ferroelectrics. For KLT single crystals, S-shape hysteresis loops were also reported for $x = 0.035$ [18].

3.4. Crossover from dipolar glass to relaxor-like behaviour

Three main distinct features of dipolar glass or relaxor-type behaviour are observed in $K_{1-x}Li_xTaO_3$ ceramics with $x = 0.02, 0.05$ and 0.10 . Firstly, a rounded peak in the temperature dependence of ϵ' is observed, in contrast to the sharp peaks at the phase transition temperatures characteristic for classic ferroelectrics. Moreover, the ϵ' peak always occurs at a temperature higher than wherein the loss peak is observed (figures 1–4). Secondly, as opposed to classic ferroelectrics both a frequency dispersion of ϵ' in the radio-frequency range, and a rather slow relaxation time are evidenced (figures 2–4). Finally, slim hysteresis loops were observed, as opposed to more squared-shaped loops of classic ferroelectrics (figures 6–8). Furthermore, analysis of the experimental data presented above by the Debye relaxation model, and both Arrhenius and Vogel–Fulcher relations, confirmed the possibility to obtain dipolar glass or relaxor phases in $K_{1-x}Li_xTaO_3$ by just varying the lithium content. For weakly doped $K_{1-x}Li_xTaO_3$ ceramics with $x = 0.02$, only Arrhenius law is valid, thus yielding a dipolar glass behaviour for low lithium content. For moderately doped $K_{1-x}Li_xTaO_3$

ceramics with $x = 0.05$ and 0.10 , though a dipolar glass behaviour is still apparent for A-type relaxations, a compliance with the Vogel–Fulcher relation is distinctly observed for the B-type relaxations. Hence, the dynamics in $K_{1-x}Li_xTaO_3$ can be rather understood in the scope of a crossover from a dipolar glass to a relaxor-like behaviour as the lithium content gradually increases.

4. Conclusions

The dielectric and polar response of $K_{1-x}Li_xTiO_3$ ceramics ($x = 0-0.10$), prepared by conventional method, was analysed in wide frequency and temperature ranges. Dielectric relaxations were induced by Li doping on the background of classic incipient ferroelectric behaviour of undoped $KTaO_3$. For weakly doped $K_{1-x}Li_xTiO_3$ ($x = 0.02$), the relaxation dynamics follows well Arrhenius law with activation energies $U_{A1} = 78$ meV, $U_{A2} = 102$ meV and $U_B = 212$ meV. For moderately Li-doped potassium tantalate ($x = 0.05-0.10$), the low-temperature relaxations also obey Arrhenius law with $U_{A1} = 76-78$ meV and $U_{A2} = 91-95$ meV. In contrast, the high-temperature relaxations follow the Vogel–Fulcher relation with $U_{B1} = 96-99$ meV and $U_{B2} = 81$ meV together with freezing temperatures $T_{FB1} = 59-66$ K and $T_{FB2} = 94$ K. The change from an Arrhenius to a Vogel–Fulcher relation shows that the dynamics in $K_{1-x}Li_xTaO_3$ can be apprehended in the scope of a crossover from a dipolar glass to a relaxor-like behaviour as the lithium content gradually increases. Thus, relaxations A1 and A2 were attributed to quadrupolar ($\pi/2$) and dipolar (π) reorientations of individual Li_K^+ , respectively, whereas relaxation B (B1) is related to the π -reorientation of Li_K^+ pairs (for $x = 0.02$) or clusters (for $x = 0.05$ and 0.10). An additional relaxation B2, detected for the first time for $x = 0.10$, was assigned to $\pi/2$ -reorientation of Li_K^+ clusters. All the relaxations are accompanied by slim S-shape $P(E)$ hysteresis loops. Thus, Li-doped KT ceramics studied in this work may be definitely considered as a reliable representative of $K_{1-x}Li_xTaO_3$ system along with widely reported KLT single crystals.

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