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Title:

## Lithium-induced Dielectric Relaxations in Potassium Tantalate Ceramics

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Abstract

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Keywords:

Quantum paraelectricity, Lithium doped potassium tantalate, Dielectric relaxation properties, Microscopic mechanisms

# 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 a.c. 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  $0.10$ . Besides 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 behavior is assumed.

## I. INTRODUCTION

Among perovskites having general formula  $ABO_3$ , there exist three compounds,  $SrTiO_3$ ,  $KTaO_3$  (KT) and  $CaTiO_3$ ,<sup>1</sup> 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 fundamental and technological point of view. In fact, they are attractive candidates for processing microwave devices, nonvolatile memories and photocatalytic water splitters.<sup>2-4</sup>

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)<sup>5</sup> yields a displacement from the centre of A sites by about  $\sim 1.26 \text{ \AA}$  along the cubic [001] direction.<sup>6</sup> 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.<sup>7,8</sup>

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.<sup>9-11</sup> The low- $T$  process with an activation energy of  $78 < U < 125 \text{ meV}$  has been associated with isolated Li ions hopping from one  $\langle 001 \rangle$  position to another.<sup>12</sup> However, the high- $T$  process with an activation energy of  $181 < U < 241 \text{ meV}$  is poorly understood. On the basis of acoustic measurements, it was suggested that this latter process could be due to  $180^\circ$  reorientations of Li ions within dipole pairs.<sup>10</sup>

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 to 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 the 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 analyzed with comprehensive theoretical models, and the relaxation characteristic parameters obtained thoroughly compared with those earlier reported for single crystals.

## II. EXPERIMENTAL PROCEDURE

KTaO<sub>3</sub>, K<sub>0.98</sub>Li<sub>0.02</sub>TaO<sub>3</sub>, K<sub>0.95</sub>Li<sub>0.05</sub>TaO<sub>3</sub>, and K<sub>0.90</sub>Li<sub>0.10</sub>TiO<sub>3</sub> ceramic samples were prepared by conventional mixed oxide method. Dried grade K<sub>2</sub>CO<sub>3</sub>, Li<sub>2</sub>CO<sub>3</sub> and Ta<sub>2</sub>O<sub>5</sub> 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.<sup>13,14</sup> 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 °C for 8 h. The calcined powders were milled again for 5 h to obtain powders with particle size lower than 5 μm. 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 1300-1350 °C for 1 h with a heating and cooling rate of 5 °C/min.

The density of the sintered samples, measured by the Archimedes' method using diethylphthalat as the immersion liquid, is ~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 μm. For the dielectric measurements, sintered samples were polished and gold electrodes were sputtered on both sides.

The real ( $\epsilon'$ ) and imaginary ( $\epsilon''$ ) parts of the dielectric permittivity in the frequency range of  $10^2$  -  $10^8$  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 analyzer 1000 AIXACT) with amplifier, respectively. The 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.

## III. RESULTS AND DISCUSSION

### A. Dielectric anomalies induced by Li doping in KTaO<sub>3</sub>

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<sub>1-x</sub>Li<sub>x</sub>TaO<sub>3</sub> ( $x = 0, 0.02, 0.05, \text{ and } 0.10$ ) ceramics, is presented in Figures 1a, 1b, and 1c, respectively. As seen from Figure 1a, undoped KTaO<sub>3</sub> ceramics reveal a classic incipient ferroelectric behavior, i.e., a continuous increase of  $\epsilon'$  with temperature decrease similar to that of KTaO<sub>3</sub> single crystals. In contrast to undoped KTaO<sub>3</sub>,  $\epsilon'(T)$  of Li-doped KT ceramics reveal a diffuse peak, which amplitude and position depend on Li content. As shown in Figures 1b and 1c, 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.

The effect of the frequency (from 100 Hz to 100 MHz) on the dielectric response of  $K_{1-x}Li_xTaO_3$  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, labeled 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.

## B. Analysis of the dielectric relaxations of $K_{1-x}Li_xTiO_3$ ceramics

The analysis of each dielectric relaxation was carried out assuming a Debye approximation. Accordingly, the set of independent dipoles is characterized by a unique relaxation time ( $\tau$ ), which is equal to the inverse of the angular relaxation frequency ( $\omega$ ). All the parameters of the relaxation dynamics were deduced from the temperature dependence of  $\epsilon''$ . First, all the relaxations were described by an Arrhenius law:

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

where  $\tau_0$  is the relaxation time at infinite temperatures, and  $U$  is the activation energy of the dipolar process. In order to determine  $\tau_0$  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 of  $10^2 - 10^8$  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 analyzed 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  $K_{0.95}Li_{0.05}TaO_3$  and peaks B1 and B2 in  $K_{0.90}Li_{0.10}TaO_3$  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 by using for the temperature dependence of the relaxation time 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 present in Table 2. For peak B2 in  $K_{0.90}Li_{0.10}TaO_3$  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 notice 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.<sup>10,12</sup> 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 the current work) were reported.<sup>11</sup> First relaxation was associated with Fe-related defects, usually detected in  $\text{KTaO}_3$  as ‘an unavoidable impurity’.<sup>11</sup> 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.<sup>15</sup> Moreover, our analysis of the dielectric response of undoped  $\text{KTaO}_3$  ceramics prepared in the 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  $\text{Li}^+$ , and the  $\pi$ -rotations of  $\text{Li}^+$  pairs, respectively.<sup>11</sup>

As referred to above, there are several works on Li-doped KT single crystals that report values of  $U$  and  $\tau_0$  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, to avoid any misinterpretations. Three main groups can be identified in terms of activation energies. The first group, with  $U = 78\text{-}86$  meV obtained for KLT crystals with  $x = 0.006\text{-}0.050$  (Refs. 9-12, 15-18) is characterized by a quadrupolar relaxation, i.e., reorientation of  $\text{Li}^+$  between equivalent sites on orthogonal axes by  $90^\circ$  ( $\pi/2$  flipping).<sup>10,12</sup> Since these values of activation energy are close to those obtained in the current 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  $U = 86\text{-}125$  meV obtained for KLT crystals with  $x = 0.011\text{-}0.070$  (Refs. 9, 11, 12, 19-22) is related to dipolar relaxation, i.e., reorientation of  $\text{Li}^+$  between equivalent sites on the same axis by  $180^\circ$  ( $\pi$  flipping).<sup>12</sup> In this range of activation energy, also falls the relaxation A2, thus revealing that the corresponding relaxation mechanism is assigned to the  $\pi$  flipping of off-central  $\text{Li}^+_{\text{K}}$  in KLT ceramics. The third group, with  $U = 181\text{-}241$  meV obtained for KLT crystals with  $x = 0.025\text{-}0.160$  (Refs. 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  $180^\circ$  ( $\pi$  pair flipping).<sup>10</sup> An activation energy inside that range is obtained in the current work for KLT with  $x = 0.02$ , revealing that relaxation B in this composition stems from the  $\pi$  flipping of  $\text{Li}^+_{\text{K}}$  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.<sup>7</sup> 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 ions 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,  $U_{AI} \sim 79$  meV, and  $T_f \sim 12$  K for  $K_{0.97}Li_{0.03}TaO_3$ ,<sup>17</sup> whereas Yokota et al. reported parameters  $\tau_0 \sim 10^{-12}$  s,  $U_{AI} \sim 88$  meV, and  $T_f \sim 56$  K for  $K_{0.93}Li_{0.07}TaO_3$ .<sup>24</sup> The later parameters are close to those obtained in the current 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.<sup>17</sup>

### C. Polar behavior 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.<sup>18</sup> So, 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 vs 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 is plotted on the bottom panel of Figure 1 as a function of temperature.  $P_r(T)$  reveals a distinctive similarity to the behavior 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 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  $x = 0.035$ .<sup>18</sup>

### D. Crossover from dipolar glass to relaxor-like behavior

Three main distinct features of dipolar glass or relaxor-type behavior are observed on  $K_{1-x}Li_xTaO_3$  ceramics with  $x = 0.02, 0.05$ , and  $0.10$ . Firstly, a rounded peak in the temperature dependence of  $\varepsilon'$  is observed, in contrast to the sharp peaks at the phase transition temperatures characteristic for classic ferroelectrics. Moreover, the  $\varepsilon'$  peak always occurs at a temperature higher than wherein the loss peak is observed (Figures 1, 2, 3, and 4). Secondly, as opposed to classic ferroelectrics both a frequency dispersion of  $\varepsilon'$  in the radio-frequency range, and a rather slow relaxation time are evidenced (Figures 2, 3, and 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 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 behavior 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 behavior 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 behavior as the lithium content gradually increases.

#### IV. CONCLUSIONS

The dielectric and polar response of  $K_{1-x}Li_xTiO_3$  ceramics ( $x = 0-0.10$ ), prepared by conventional method, was analyzed in wide frequency and temperature ranges. Dielectric relaxations were induced by Li doping on the background of classic incipient ferroelectric behavior 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 obeys Arrhenius law with  $U_{A1} = 76-78$  meV and  $U_{A2} = 91-95$  meV. Contrarily, the high-temperature relaxations follow 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 behavior as the lithium content gradually increases. Thus, relaxations A1 and A2 were attributed to quadrupolar ( $\pi/2$ ) and dipolar ( $\pi$ ) reorientations of individual  $Li^+_K$ , respectively, whereas relaxation B (B1) is related to the  $\pi$ -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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**Table 1.** Arrhenius law parameters for the dynamics of the dielectric relaxations A in  $K_{1-x}Li_xTiO_3$  ceramics.

Relaxation	$x$	$U$ , meV	$\tau_0$ , s
A1	0.02	78	$2.7 \times 10^{-13}$
	0.05	78	$1.1 \times 10^{-13}$
	0.10	76	$1.7 \times 10^{-13}$
A2	0.02	102	$0.8 \times 10^{-13}$
	0.05	91	$2.2 \times 10^{-13}$
	0.10	95	$2.4 \times 10^{-13}$

**Table 2.** Parameters of Arrhenius law and Vogel-Fulcher relation for the dynamics of the dielectric relaxations B in  $K_{1-x}Li_xTiO_3$  ceramics.

$x$	Relaxation	<i>Arrhenius law</i>		<i>Vogel-Fulcher relation</i>		
		$U$ , meV	$\tau_0$ , s	$U$ , meV	$\tau_0$ , s	$T_f$ , K
0.02	B	212	$3.3 \times 10^{-15}$			
0.05	B1	292	$6.1 \times 10^{-17}$	99	$6.9 \times 10^{-13}$	59
0.10	B1	328	$4.7 \times 10^{-18}$	96	$4.6 \times 10^{-13}$	66
0.10	B2	416	$4.6 \times 10^{-19}$	81	$1.6 \times 10^{-12}$	94

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

<b>Proposed mechanism</b>	$x$	$U$ , meV	$\tau_0$ , s	<b>Ref.</b>
Quadrupolar relaxation - reorientation of $Li^+$ between equivalent sites on orthogonal axes by $90^\circ$ ( $\pi/2$ flipping)	0.006	78	$1.4 \times 10^{-13}$	[15]
	0.009	86	$1.3 \times 10^{-13}$	[16]
	0.010	84	$4.1 \times 10^{-13}$	[17]
	0.016	86	$3.4 \times 10^{-13}$	
	0.011	86	$0.2 \times 10^{-13}$	[12]
	0.015	86	$6.0 \times 10^{-13}$	[10]
	0.035			
	0.050			
	0.025	86	$50 \times 10^{-13}$	[9]
	0.033	86	$8.0 \times 10^{-13}$	
0.035	86	$1.6 \times 10^{-13}$	[18]	
0.043	85	$0.5 \times 10^{-13}$	[11]	
Dipolar relaxation - reorientation of $Li^+$ between equivalent sites on the same axis by $180^\circ$ ( $\pi$ flipping)	0.011	98	$1.0 \times 10^{-13}$	[12]
	0.012	125	$16 \times 10^{-13}$	[19]
	0.016	95	$0.6 \times 10^{-13}$	[20]
	0.026	95	$0.6 \times 10^{-13}$	
	0.026	95	$0.3 \times 10^{-13}$	[21]
	0.025	86	$8.0 \times 10^{-13}$	[9]
	0.033	112	$0.8 \times 10^{-13}$	
	0.034	99	$1.0 \times 10^{-13}$	[22]
	0.050			
0.070	104	$0.8 \times 10^{-13}$	[11]	
0.043				
Reorientation of a pair of coupled dipoles in head-to-tail configurations along the same axis by $180^\circ$ ( $\pi$ pair flipping)	0.025	220	$25 \times 10^{-15}$	[9]
	0.033	229	$10 \times 10^{-15}$	
	0.035	233	$1.4 \times 10^{-15}$	[10]
	0.050			
	0.043	241	$0.9 \times 10^{-15}$	[11]
	0.035	207	$6.9 \times 10^{-15}$	[23]
	0.050	208	$5.3 \times 10^{-15}$	
	0.100	206	$8.3 \times 10^{-15}$	
	0.160	227	$1.8 \times 10^{-15}$	
	0.036	216		[20]
	0.049			
0.063				
0.070	181-200		[22]	

## Figure captions

**Figure 1.** Real  $\epsilon'$  (a) and imaginary  $\epsilon''$  (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.

**Figure 2.** Real  $\epsilon'$  (a) and imaginary  $\epsilon''$  (b) parts of the dielectric permittivity, and dissipation factor  $\tan\delta$  (c) of  $K_{0.98}Li_{0.02}TaO_3$  ceramics at different frequencies from  $10^2$  to  $10^8$  Hz as a function of temperature.

**Figure 3.** Real  $\epsilon'$  (a) and imaginary  $\epsilon''$  (b) parts of the dielectric permittivity, and dissipation factor  $\tan\delta$  (c) of  $K_{0.95}Li_{0.05}TaO_3$  ceramics at different frequencies from  $10^2$  to  $10^8$  Hz as a function of temperature.

**Figure 4.** Real  $\epsilon'$  (a) and imaginary  $\epsilon''$  (b) parts of the dielectric permittivity, and dissipation factor  $\tan\delta$  (c) of  $K_{0.90}Li_{0.10}TaO_3$  ceramics at different frequencies from  $10^2$  to  $10^8$  Hz as a function of temperature.

**Figure 5.** Arrhenius plots  $\ln(\tau)$  vs  $1000/T_{\epsilon''m}$  for relaxations A (a) and B (b) of  $K_{1-x}Li_xTaO_3$  ceramics with fits to Arrhenius law (solid lines) and Vogel-Fulcher relation (dash lines).

**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 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.

**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.

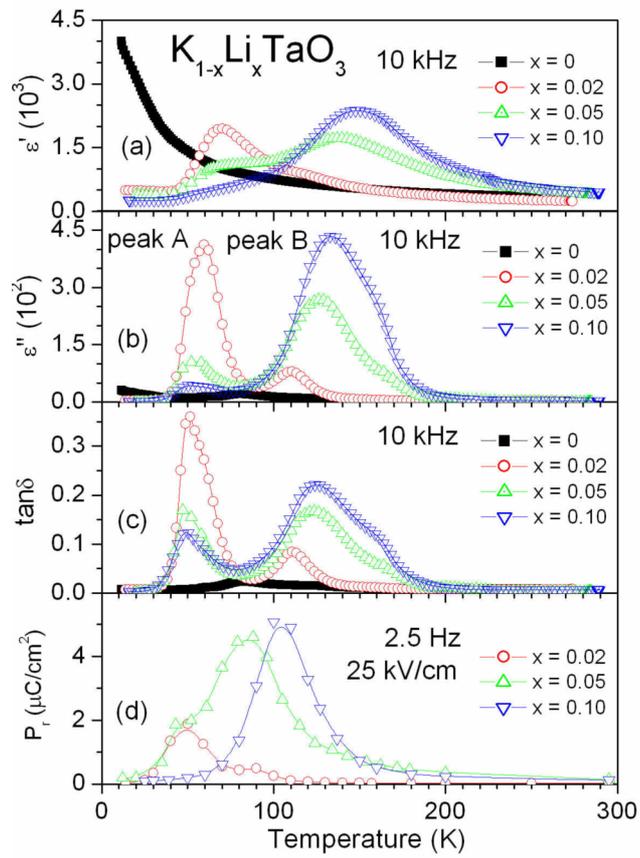


Figure 1

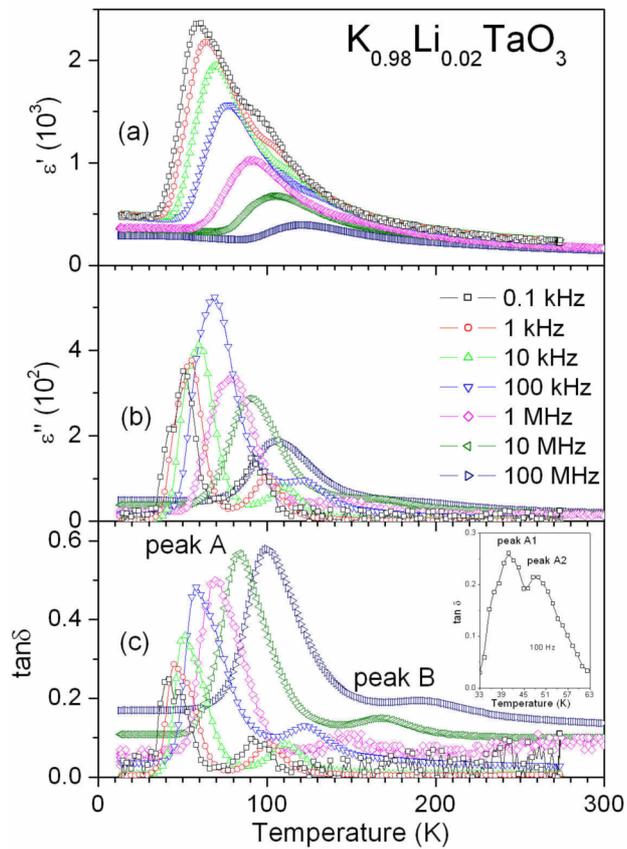


Figure 2

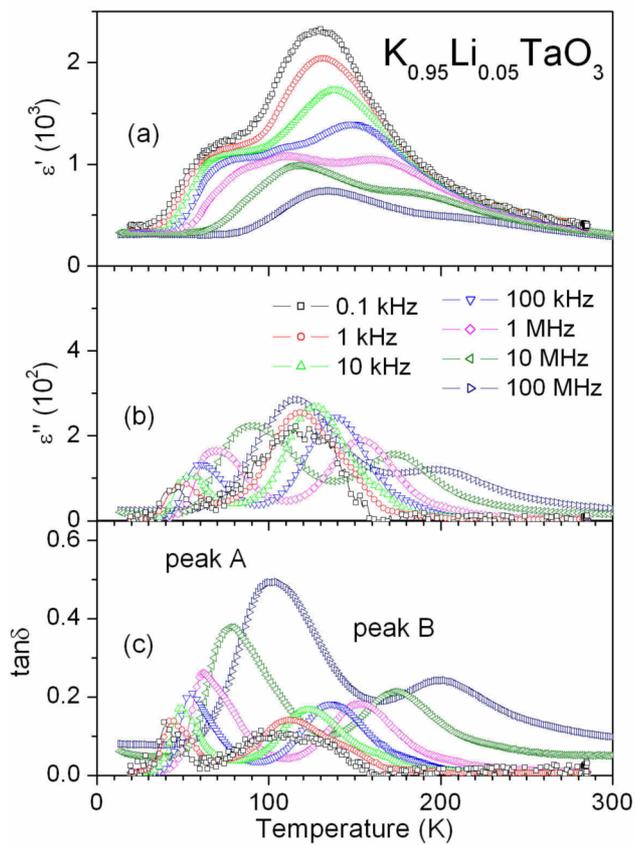


Figure 3

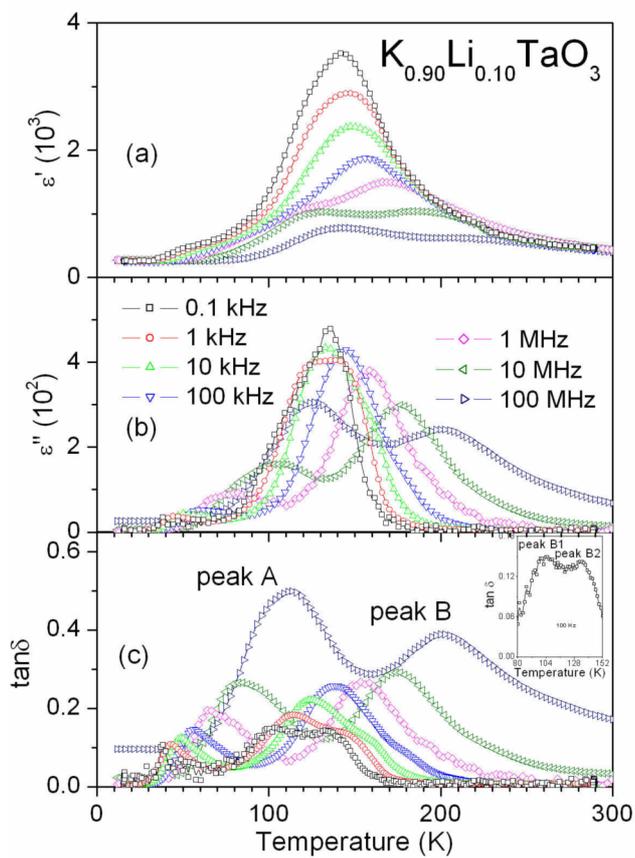


Figure 4

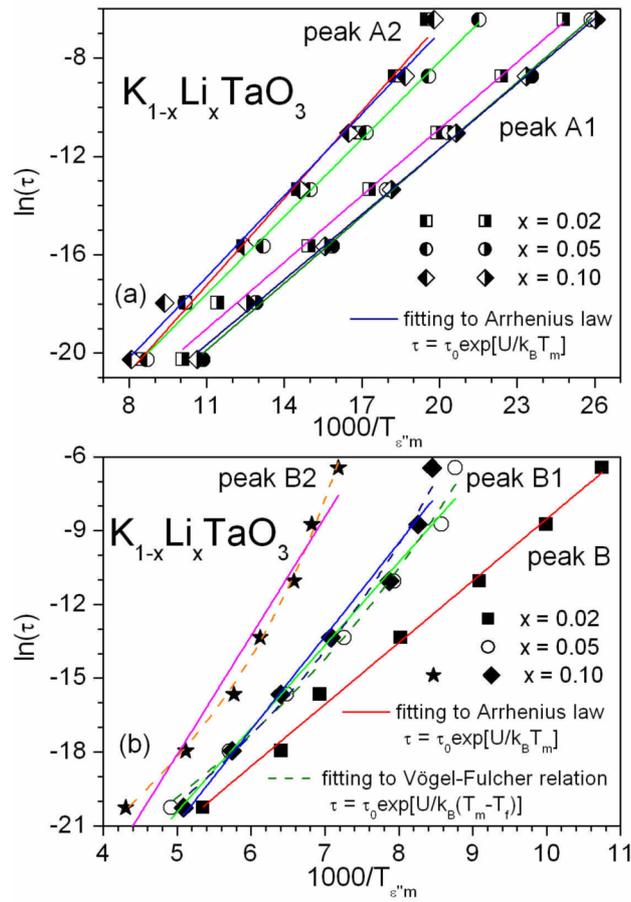


Figure 5

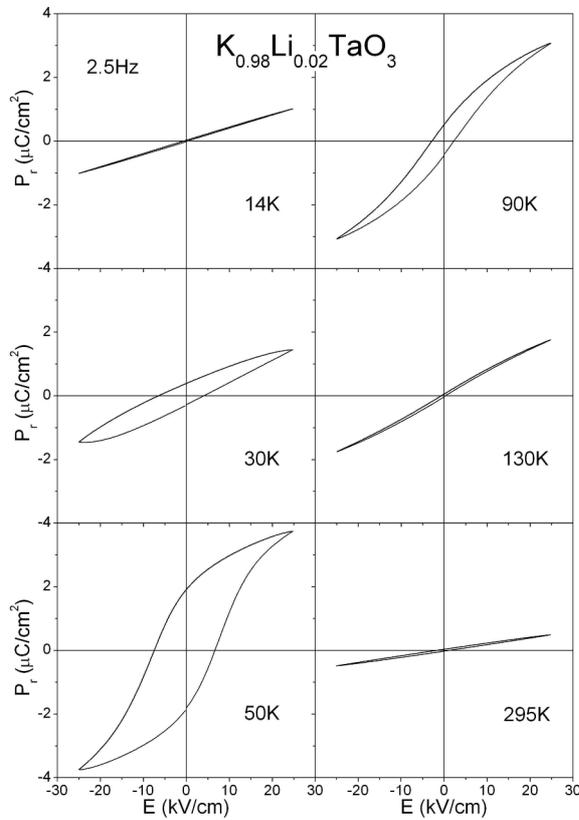


Figure 6

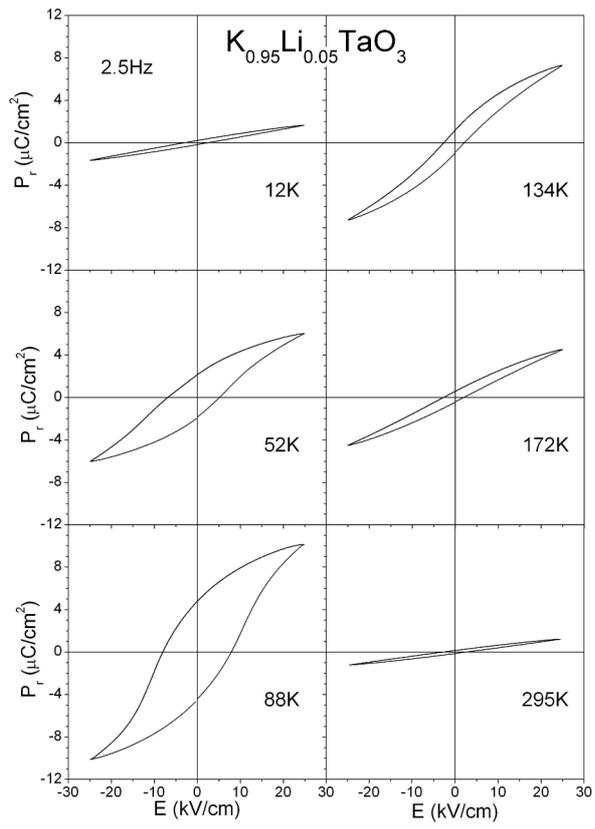


Figure 7

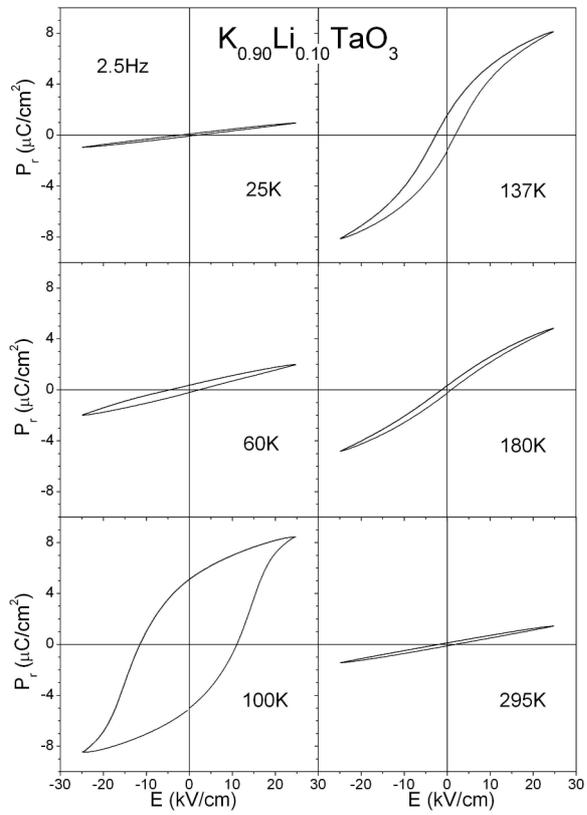


Figure 8