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Characteristics and the nature of the low-frequency dielectric response in moderately concentrated $\text{KTaO}_3\text{:Li}$

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Abstract

The temperature–frequency behaviour of the complex dielectric permittivity of $\text{K}_{0.957}\text{Li}_{0.043}\text{TaO}_3$ (KLT) was studied in detail. Below 250 K a pronounced relaxation-type dielectric dispersion occurs with two wide temperature maxima shifted to lower temperatures increasing in magnitude as the alternating-current monitoring frequency decreases. The results obtained are analysed by using a theory proposed considering the coupling of the TO_1 soft mode (TO standing for transverse optical) to two Li relaxation modes known in moderately concentrated KLT as π - and $(\pi/2)$ -relaxations.

1. Introduction

The attractive properties, dynamics and cooperative dipole ordering effects inherent to the perovskite-type dilute relaxor $\text{K}_{1-x}\text{Li}_x\text{TaO}_3$ (KLT) have already been subjects of extensive study for a long time (see e.g. [1–3] and references therein). Recently [4], we presented detailed studies and a theoretical quantitative analysis of the behaviour of the dielectric permittivity in KLT at a comparatively small Li concentration, $x = 0.006$. The present paper is a natural continuation of [4]. We report detailed measurements and theoretical analysis of the temperature–frequency behaviour of the complex dielectric permittivity for moderately concentrated KLT with $x = 0.042$. So, because the consideration from [4] is applicable for comparatively small Li concentrations in KLT crystals, well below the critical concentration, $x = 0.022$ [5], we extend the theoretical approach of [4] to the case of moderately concentrated KLT ($0.022 < x < 0.06$). In such a region, there is short-range order at low temperatures and long-range ferroelectric order develops for KLT with $x > 0.06$ [5, 6]. We will show that, in accordance with previous observations (e.g. [7–14]), our moderately doped KLT exhibits two

distinct temperature maxima ($(\pi/2)$ - and π -relaxations [11, 12]). We present and consider here a model which couples the two corresponding relaxators to the soft TO lattice mode.

2. Experimental technique

KLT single crystals were grown at the A F Ioffe Physical-Technical Institute by the spontaneous crystallization technique from slowly cooled flux with $x = 0.1$ from the batch described in reference [15]. Based on NMR investigation [16] the Li concentration inside the crystals was usually estimated as $\chi_{in} = 0.35\chi_{melt}$. However, the estimation $\chi_{in} = 0.42\chi_{melt}$ [17] is more reasonable for the Li concentration studied in the present paper. The latter estimate is consistent with the position of the sharp peak in the temperature dependence of the dielectric constant obtained by us in the field-cooling regime (see below).

Selected transparent colourless KLT single crystals were annealed at $T = 400$ K for three hours in the open atmosphere [18]. Examination between crossed polarizers with an optical microscope revealed that the samples prepared were relatively free of inclusions and visible boundaries typical of possible microtwins [19]. Experimental samples were fabricated as thin condensers with Au electrodes evaporated onto the principal faces of $\langle 100 \rangle$ -oriented polished plates of typical dimensions $3 \times 3 \times 0.2$ mm³. Measurements of the dielectric permittivity $\epsilon'(T, f)$ were performed on a 4192 Hewlett-Packard impedance analyser. The samples were suspended in a He-flow cryostat allowing temperature cycling within 5–300 K at a rate of 10–100 mK s⁻¹.

3. Experimental results

In figure 1 the real and imaginary parts of the dielectric permittivity, $\epsilon'(T)$ and $\epsilon''(T)$, measured during the cooling run are presented for $\text{K}_{0.957}\text{Li}_{0.043}\text{TaO}_3$ as a function of temperature at

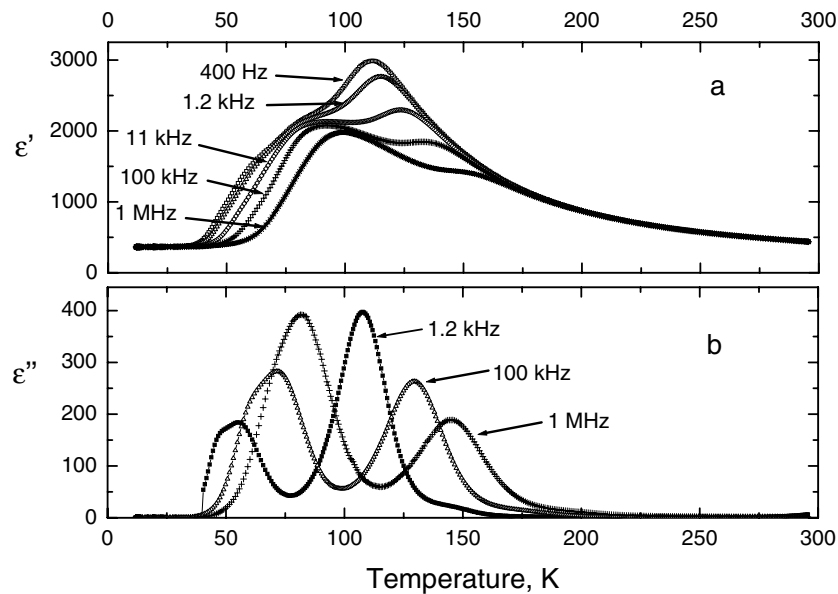


Figure 1. Temperature dependence of the real (a) and imaginary parts (b) of the dielectric permittivity in $\text{K}_{0.957}\text{Li}_{0.043}\text{TaO}_3$ at several selected frequencies.

several selected frequencies. The $\varepsilon'(f, T)$ and $\varepsilon''(f, T)$ dependencies undoubtedly reveal the relaxation nature of the peaks of the dielectric permittivity. Let us consider first the low-temperature, 45–75 K, region.

The low-temperature relaxation obeys well an Arrhenius law [20] with the high-temperature relaxation time $\tau_0 = 2.1 \times 10^{-13}$ s and barrier $U = 1100$ K (compare with the barrier of 950 K for weakly doped KLT [31]). These quantities agree with the well-known characteristics of the $(\pi/2)$ -relaxation [21–26]. It is believed that it is this relaxation that provides the main contribution in dilute KLT (see, e.g., references [1–3, 4, 7, 10]). However, from a detailed analysis of the frequency dependence of the dielectric permittivity at different temperatures, we have found that in the temperature region 45–75 K at least two relaxation processes are operative and the dielectric relaxation is fitted well by the Cole–Cole-type equation [27]

$$\varepsilon(\omega) = \varepsilon_{i\infty} + \frac{\Delta\varepsilon_{i1}}{1 - i(\omega\tau)^{1-\alpha_{i1}}} + \frac{\Delta\varepsilon_{i2}}{1 - i(\omega\tau)^{1-\alpha_{i2}}}. \quad (1)$$

Here $\Delta\varepsilon_{i1,2}$, $\tau_{i1,2}$ and $\alpha_{i1,2}$ are the relaxation depth, mean relaxation time and relaxation distribution Cole–Cole parameters for the first and second relaxations respectively; $i = 1$ corresponds to the temperature interval from 45 to 70 K while $i = 2$ corresponds to the interval from 100 to 140 K. Figure 2 shows the result of the best fitting of this expression to our experimental data at 58 K obtained at $\alpha_{11} = 0.49$ and $\alpha_{12} = 0.3$.

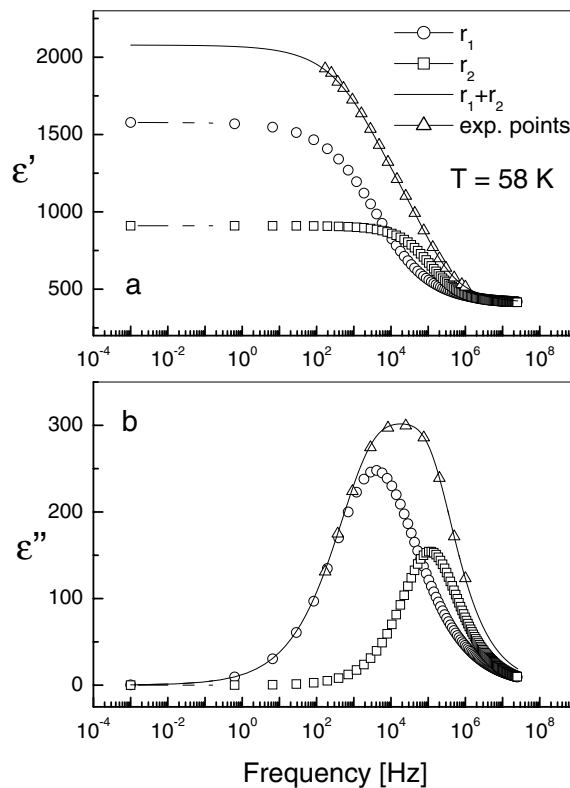


Figure 2. Frequency dependence of the real (a) and imaginary parts (b) of the dielectric permittivity in $\text{K}_{0.957}\text{Li}_{0.043}\text{TaO}_3$ at $T = 58$ K: r_1 and r_2 are the contributions of the first and second relaxators respectively at $\alpha_1 = 0.49$ and $\alpha_2 = 0.3$.

The analysis of the temperature dependencies of the relaxation times, $\tau_{1,2}$, allows us to determine the Arrhenius law relaxation parameters: $\tau_{011} = 8 \times 10^{-14}$ s, $U_{11} = 1200$ K and $\tau_{012} = 5 \times 10^{-14}$ s, $U_{12} = 980$ K. We also found that $\alpha_{11} = 0.5 \pm 0.1$ and $\alpha_{12} = 0.3 \pm 0.1$. We associate the first dominant low-temperature relaxation with the off-centre ($\pi/2$)-rotations of Li^+ . The presence of the second low-temperature relaxation, with activation energy a little smaller than for the off-centre ($\pi/2$)-rotations of Li^+ , was reported earlier in reference [28], where it was associated with Fe-related defects, usually detected in KTaO_3 as ‘an unavoidable impurity’ [29].

Now we turn to the high-temperature relaxation (above 100 K). The high-temperature relaxation obeys well an Arrhenius law with $\tau_{02} = 4 \times 10^{-16}$ s and $U_2 = 2800$ K. We have obtained the result $\alpha_{22} = 0.35 \pm 0.05$. Figure 3 shows the fit at 140 K with the values of the parameters $\alpha_{12} = 0.39$ and $\alpha_{22} = 0.32$. Figure 4 shows the corresponding Cole–Cole diagram clearly evidencing the presence of two relaxation processes. The limitation of the experimental ac frequency makes it very difficult to perform a detailed analysis of both high-temperature relaxation contributions. However, the main dominant relaxation contributions in the high-temperature region can be considered in detail: $\tau_{02} = 8.7 \times 10^{-16}$ s and $U_{22} = 2790$ K, which agree well with the π -relaxation parameters reported in reference [12]. We also performed estimations for the minor high-temperature relaxation in the temperature interval 140–200 K and obtained $\tau_{012} = 9 \times 10^{-15}$ s and $U_{21} \sim 3400$ K. The nature of the latter relaxation is unknown.

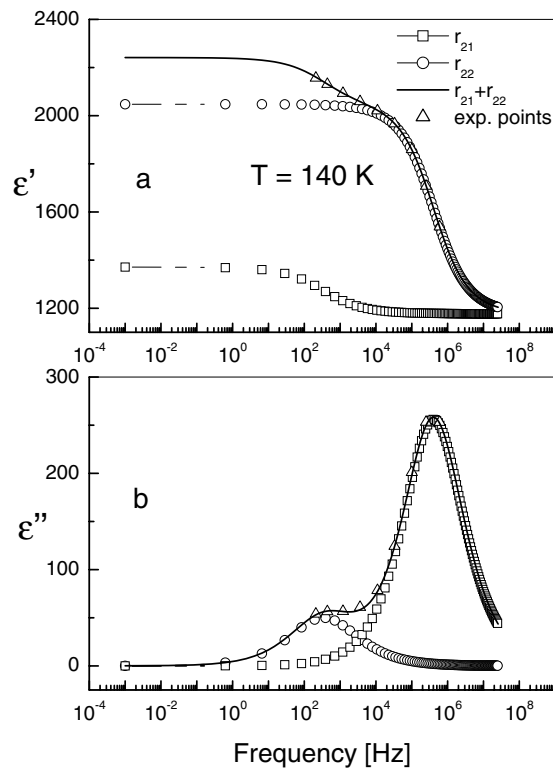


Figure 3. Frequency dependence of the real (a) and imaginary parts (b) of the dielectric permittivity in $\text{K}_{0.957}\text{Li}_{0.043}\text{TaO}_3$ at $T = 140$ K: r_{22} and r_{21} are the contributions of the first and second relaxors respectively obtained on the basis of Cole–Cole equation (2) at $\alpha_{21} = 0.39$ and $\alpha_{22} = 0.32$.

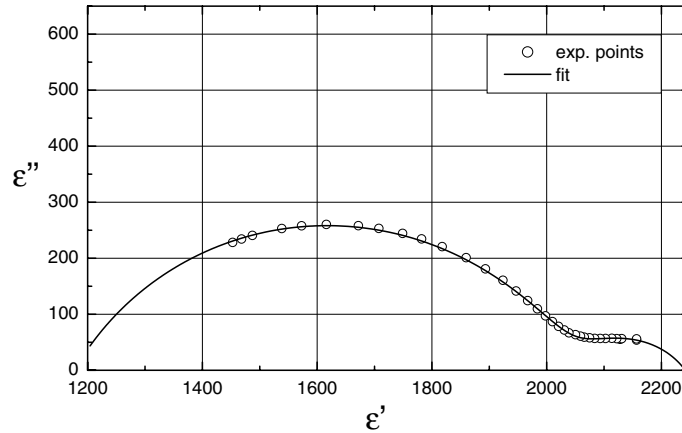


Figure 4. The Cole–Cole diagram for $\text{K}_{0.957}\text{Li}_{0.043}\text{TaO}_3$ at 140 K (experimental data and fit with equation (2)).

The nature of the high-temperature relaxation is a subject of controversy. The non-Debye behaviour and the large activation energy for the high-temperature relaxation process have led researchers to make the assumption that it might be connected with Li impurity clusters [7, 10, 11, 13] and with pairs of lithium dipoles reorienting as a single unit [12, 14, 23, 24].

4. A model

The Li ions substituting for K in KTaO_3 are known to occupy off-centre positions [1] shifted from the centre by a well-known distance (according to theoretical estimations, by 0.6 \AA [25, 32, 33]; experimentally observed displacements are even larger: about $1\text{--}1.2 \text{ \AA}$ [21, 22]). The Li-related dipoles are coupled to the soft mode by the local field [2]. First we consider the effect of this coupling on the Li dipole dynamics. This problem has been explored in many papers (see for example [2, 34–37]). Then we will study the influence of the coupling on the soft mode itself (some preliminary results were given in references [4, 37]). Thereafter we will sum the contributions of the renormalized soft mode and Li-related dipoles to the total dielectric susceptibility and compare the results obtained with experiment.

First we consider two sorts of relaxators, connected with π - and $(\pi/2)$ -reorientations of Li^+ dipole impurities, coupled to the soft mode with polarization P_0 :

$$H = -2E \sum_m \mu_{im} s_{im}^z - \lambda_m P_0 \sum_m s_{im}^z. \quad (2)$$

Here E is the external field, μ_m and λ_m are the dipole moment and coupling constant, respectively, and s_{im}^z are the quasispin operators. The contributions of the $(\pi/2)$ -relaxators ($m = 1$) and π -relaxators ($m = 2$) to the total susceptibility can be expressed as follows [4]:

$$\chi_m = \frac{2\mu_m}{\varepsilon_0} \frac{d\langle s_{im}^z \rangle}{dE} = \frac{4M_1(\omega)}{\varepsilon_0 k_B T} \left(1 + \frac{\lambda_m}{2\mu} \frac{dP_0}{dE} \right) \quad (3)$$

where

$$M_1(\omega) = k_B T \sum \mu_m^2 n_m F_m$$

with

$$F_m = \int \frac{g(x) dx}{4k_B T (1 - i\omega\tau_m(x))}. \quad (4)$$

Here $\tau(x) = \tau_0 \exp[(U + x)/k_B T]$ and $g(x)$ is a Gauss-type distribution function, $g(x) \sim \exp(-x^2/a^2)$; here, a is a parameter of the Gauss distribution.

The host-lattice susceptibility can be derived from the condition of stability of the free energy:

$$\alpha p^2 + \beta p^4 + \gamma p^5 - \sum \lambda_m \langle s_m^z \rangle - E - e = 0 \quad (5)$$

where e is the random field produced by the dipoles. The macroscopic polarization, P_0 , can be found as the average over the random fields:

$$P_0 = \int f(e) p(e) de.$$

Here the distribution function could be taken in the Gauss form:

$$f(e) = (2\pi\delta)^{-1/2} \exp[-(e - E_0 P_0)/2\delta^2]$$

where E_0 and δ are constant [2, 21]. In the paraelectric and dipole glass phases the random fields produce a local polarization while the macroscopic polarization is absent just because of the randomness of the local fields and local polarization. However, it will be shown that the final expression for the dielectric permittivity depends on e through p^2 , as the lattice dielectric permittivity is proportional to $\alpha + 3\beta p^2 + 5\gamma p^4$. This allows one to account for the effect of the random fields by using an average $\langle p^2 \rangle$, which can be estimated as $K/\varepsilon_0^2 \alpha^2$, where K is constant. If one uses a self-consistent approach, one can consider the renormalized susceptibility instead of the susceptibility of the pure lattice.

Taking the derivative with respect to E , one has

$$\chi_0 = \frac{dP_0}{dE} = \int_{-\infty}^{\infty} \left(1 + \lambda_m \frac{\varepsilon_0 \chi_m}{2\mu_m} \right) \frac{f(e)}{A(T, p(e))} de \quad (6)$$

where $A(T, P_0) = \varepsilon_0(\alpha + 3\beta P_0^2 + 5\gamma P_0^4)$. Inserting (3) in (6), one finds

$$\chi_0 = \int \frac{1 + 2M_2/k_B T}{A(T, p(e)) - \varepsilon_0 M_3/k_B T} f(e) de \quad (7)$$

where

$$\begin{aligned} M_2(\omega) &= k_B T \sum \lambda_m \mu_m n_m F_m \\ M_3(\omega) &= k_B T \sum \lambda_m^2 n_m F_m. \end{aligned} \quad (8)$$

The total susceptibility can be now rewritten in the form

$$\chi = \chi_0 + \sum \chi_m = \frac{4M_1}{k_B T} + \int_{-\infty}^{\infty} \frac{D(T, \omega)}{A(T, p(e)) - \varepsilon_0 M_3(\omega)/k_B T} f(e) de \quad (9)$$

where $D(T, \omega) = [1 + 2M_2(\omega)/k_B T]^2$. When fitting this expression to the experimental data, we took into account only the contribution stemming from the $(\pi/2)$ - and π -relaxations.

Figure 5 demonstrates the good agreement of the theoretical fit with the experimental data obtained at 100 kHz (see also the comparison for selected frequencies in figure 6). The dipole moment and concentration were fixed at $0.6 e \text{ \AA}$ [32] and $0.035 (\sim 10^{27} \text{ m}^{-3})$ respectively for both relaxators. A good fit was found with the coupling constant values in the interval from 0.03 to 0.05. These data are lower than those found in reference [4] for smaller Li content. The barriers were at $960 \pm 20 \text{ K}$ and $2850 \pm 50 \text{ K}$ and τ_0 was 10^{-14} s and $8 \times 10^{-16} \text{ s}$ for the first and second relaxators respectively. These data are close to the experimental findings described above. The constant K was found to be $(5.4 \pm 2) \times 10^{-6}$. In order to reproduce well the high-temperature tail of ε' , we used the Barrett formula with the parameters obtained from the fit to this tail: $T_0 = 101 \pm 5 \text{ K}$, $T_s = 113 \pm 8 \text{ K}$ and $C = (8 \pm 2) \times 10^4$. The

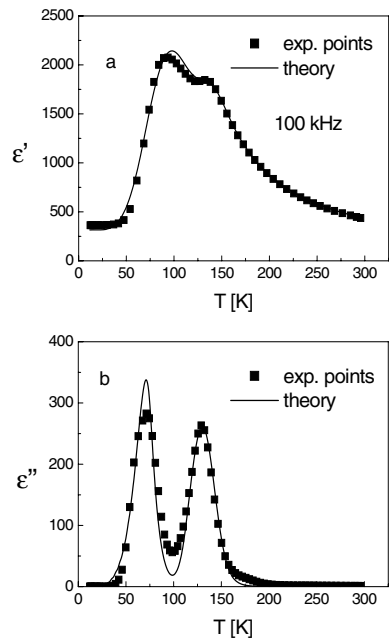


Figure 5. Comparison of the theory and experiment for the $\epsilon'(T)$ (a) and $\epsilon''(T)$ (b) dependencies in $\text{K}_{0.957}\text{Li}_{0.043}\text{TaO}_3$ at 100 kHz (not all experimental points are shown, in order to reveal the theoretical curve clearly).

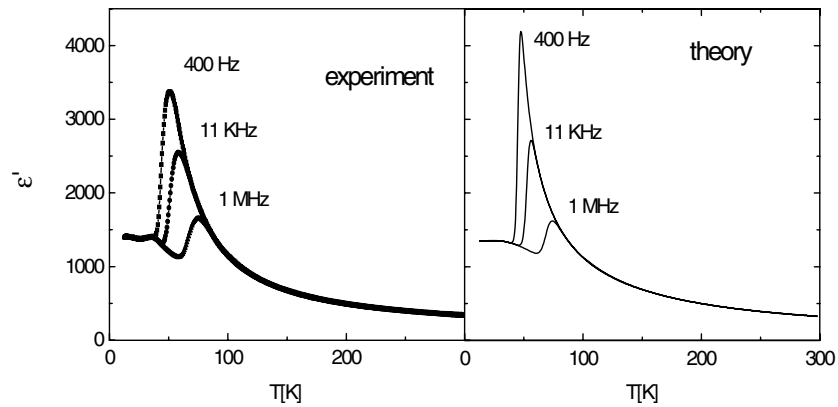


Figure 6. Comparison of theory with experiment for $\epsilon'(T)$ for selected frequencies.

use of the Barrett formula instead of the Curie–Weiss expression can be justified from the fact that the dielectric permittivity does not diverge at the phase transition temperature due to a suppression of the phase transition by random fields. Hence the correlation radius also does not diverge at the phase transition temperature and we assume it to saturate. It is just this saturation that is described by the saturation temperature, T_s , which is governed by the maximal size of the polar region. Thus we assume that the average size of the polar region does not exceed a finite value that corresponds to the formation of a glass state instead of the ferroelectric phase. This assumption is in good accordance with earlier findings of freezing phenomena in KLT [1, 39].

When fitting expression (10) to the experimental data on the frequency dependence of ϵ'' (see figure 7), we obtained the width of the potential barrier distribution function $a = 300$ K (curve 2), but in this case the low frequency part of the permittivity is not reproduced well. We assumed that there is an additional constant (within a frequency interval with a temperature dependent width) contribution, $\delta\epsilon''$, to the dielectric permittivity at low frequencies and obtained a much better fit for $T = 140$ K (curve 1) at $a = 200 \pm 25$ K, $\delta\epsilon'' = 54 \pm 10$ and the frequency width coinciding with the width of the frequency interval studied. The weight of this low-frequency addition was found to decrease with temperature and the frequency width became larger. Figure 8 shows the comparison of the theory with experiment for selected temperatures.

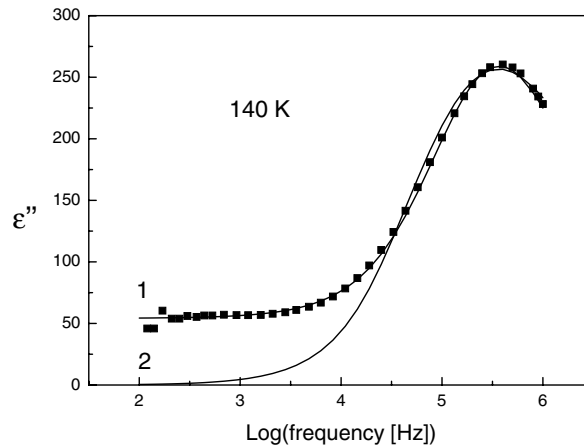


Figure 7. Experimental and theoretical frequency dependencies of $\epsilon''(f)$ at 140 K obtained with the use of a Gauss-type potential barrier distribution and addition of a constant background.

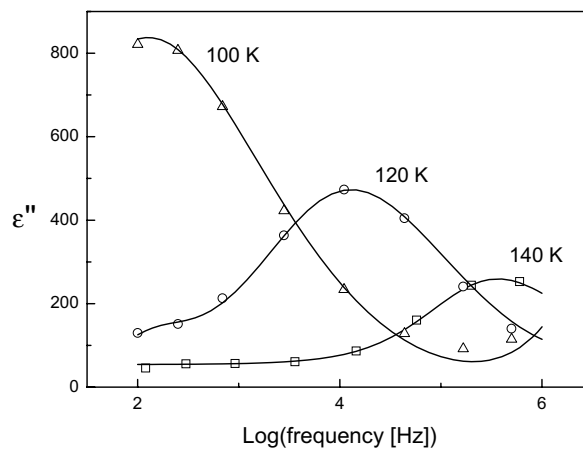


Figure 8. Comparison of the theoretically computed frequency dependencies for selected temperatures with experiment at $a = 200$ K. Only one in ten of the experimental points are shown, in order to reveal the theoretical curve clearly.

The mean potential barrier for the relaxation at 140 K was found at 1950 ± 50 K, but this

value was found also to decrease with temperature. In the linear approximation one has

$$U(T) = U_0 - \zeta k_B T \quad (10)$$

where ζ is thought to be constant. This dependence is well known in the theory of polarons, but for Li impurities in KTL it is observed here for the first time. In our opinion, there is considerable overlap between the theory of the electronic polaron and that of the Li-ionic polaron. Indeed, according to theoretical estimations [25, 32], the Li ion in KLT does not have its own deep potential. Only due to the relaxation of the surrounding ions is the Li ion self-trapped, as a self-trapped electron or hole. One feature of the polaronic behaviour is the temperature dependence of polarization fluctuation characteristics in the polarization cloud around the Li impurity. This dependence leads to (10) for a selected temperature interval. Indeed, if the fluctuation of the potential energy is proportional to temperature, $|\Delta U| = ak_B T$, then the average over fluctuations having different signs gives

$$\langle e^{(U+\Delta U)/k_B T} \rangle = e^{U/k_B T} [e^a + e^{-a}] \cong e^{U/k_B T} e^a = e^{(U+ak_B T)/k_B T}. \quad (11)$$

It is seen that if one averages the potential energy itself, then it is not temperature dependent, but the averaging of the exponential function leads to the final result.

Another reason for the temperature dependence of the potential barrier is the temperature dependence of the frequency of the soft mode. One should remember that the main contribution to the energy of the Li dipole is the polarization energy. This energy decreases with temperature as the dielectric permittivity decreases. This results in a temperature decrease of the potential barrier. Notice that even if the temperature dependence of this contribution is not linear over a wide temperature interval, it can be interpolated using a linear function over the short temperature interval studied. The linear temperature dependence can also stem from the third-order anharmonic interactions leading to the temperature-dependent field, $x\langle xx \rangle \sim Tx$ where χ is the local mode displacement at one of the Li noncentral equilibrium positions.

Figure 9 shows the temperature dependence of the mean potential barrier obtained from fitting equation (9) to the experimental data. It is seen that the linear dependence is followed in the temperature interval studied very well. The same result has been found for the $(\pi/2)$ -relaxation but with an approximately two times lower value of ζ . This sheds light on the nature of the small value of the pre-exponential factor obtained from the Arrhenius plot. Indeed, due to the linear dependence (10), the pre-exponential factor is renormalized:

$$\tau_0^* = e^{-\zeta} \tau_0. \quad (12)$$

The seeming pre-exponential factor, τ_0^* , is a few orders lower than the real pre-exponential factor, τ_0 .

5. Discussion

The π -relaxation process was earlier regarded as being due to Li pairs [12]. There are at least three good arguments in favour of the appearance of Li pair relaxation in moderately doped KLT. The first one is connected with the fact that the potential barrier value obtained experimentally for the second relaxation process is approximately two times larger than the potential barrier value obtained for the first relaxation process [12]. This increase was regarded as being due to the enlarged (total) dipole moment in the Li pair. The second argument was reported in reference [14]. In the authors' opinion, if the π -relaxation were due to the reorientation of single dipoles, one would expect lower activation energy than for the $(\pi/2)$ -relaxation, since the π -relaxation is not accompanied by the reorientation of the strain ellipsoid. The third argument was also given in reference [14]. According to this argument, the second relaxation process should be seen at all Li concentrations.

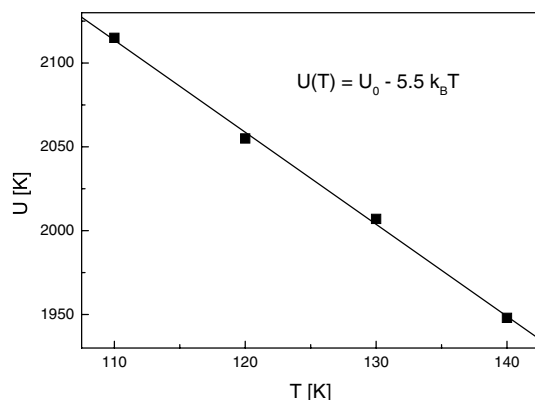


Figure 9. The temperature dependence of the Li potential barrier obtained from the frequency dependence of ϵ'' .

Our computations have shown that, for comparatively small Li concentrations, the second relaxation peak does not appear, in agreement with experiment [4]. Hence this argument cannot be used in favour of the Li-pair idea. The other two arguments should be considered more carefully.

The Li potential relief in KLT has been computed by several theoretical groups [25, 26, 32] and they all found that the values of the potential barriers for the π - and $(\pi/2)$ -relaxations differ appreciably. The first barrier is considerably larger. Hence the second argument is also unreliable.

The most important discrepancy between the Li pair idea and the experimental data stems from the rather low value of the pre-exponential factor in the relaxation time dependence on temperature obtained from the Arrhenius plot. In fact, the value found could even contradict the assumption that this relaxation process is connected with ionic vibrations instead of electronic ones [40, 41]. Fortunately the detailed experimental and theoretical analysis of the frequency dependence of the dielectric permittivity has shown that the nature of this inconsistency lies in the temperature dependence of the potential barrier. We have found the potential barrier to behave linearly with temperature in agreement with earlier findings in the physics of polarons. The mean potential barrier value proves to decrease with temperature and it is just this dependence that led to the seeming decrease of the pre-exponential factor and increase of the mean potential barrier. In fact, on accounting for the frequency dependence of the dielectric susceptibility, the pre-exponential factor and the potential barrier become normal and the problem of the small value of the pre-exponential factor disappears. The final values of the pre-exponential factors for both the relaxations prove to be of the same order, 10^{-13} s. Finally, one can see that the idea of ascribing the π -relaxation to Li-pairs does not contradict the experimental data (after the correction of the pre-exponential factor is made) while it is also not explicitly confirmed by experimental data. Hence, the nature of the π -relaxation remains uncertain up to now and further theoretical and experimental investigations are necessary.

The linear temperature decrease of the Li potential barrier can be easily understood if one takes into account the vibrations of the ions surrounding a Li ion. As the barrier is formed just by the potentials of the polarized surrounding ions, the vibrations of the ions lead to a vibration of the barrier. The larger the vibrations, the larger the decrease of the mean potential barrier. The main reason for this dependence lies in the fact that one should average the exponent instead of averaging the potential barrier itself.

Another finding of the present study is a large Gauss-type distribution of the potential barriers. This finding is in agreement with an earlier study [42]. This distribution could arise, for example, from the disorder of the Li positions and, as a result, from clustering of the Li ions [43]. However, in spite of this clustering effect the single-ion relaxation mainly prevails in the experimental data from the dielectric spectroscopy, although the Li ion proves to be embedded into the random potential of the surrounding Li ions. In our opinion this is a consequence of the fact that the probability of turning of the cluster dipole moment is much less than the probability of turning of the single-Li-ion dipole moment because of the very large potential barrier in the cluster. Such dynamics of Li pairs was considered earlier in the framework of the Glauber model in reference [44]. Nevertheless, when treating the experimental results we saw that the shape of the frequency dependence of the imaginary part of the dielectric permittivity is not symmetrical, and additional relaxators with large potential barriers (or with larger pre-exponential factors) should be added. Perhaps this contribution originates from the Li clusters. This small contribution was found to increase on lowering temperature in good agreement with theory [43].

We did not study here the freezing phenomena, which do exist at low temperatures. Such studies have been performed in many other papers (see for example [1, 39]).

Acknowledgments

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