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Effect of the electric field on the elastic and dielectric properties of a lithium-potassium tantalate crystal

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Abstract. — We have measured in a $K_{1-x}Li_xTaO_3$ crystal with $x = 1.7\%$ the elastic constant with longitudinal acoustic waves of frequency between 30 and 330 MHz and the dielectric constant between 10 Hz and 1 MHz in the temperature range from 4 to 200 K under an electric field applied upon cooling. The main effect of the applied field on the dielectric constant is attributed to space charges. The real part of the elastic constant (the sound velocity) was found to increase when the field was applied perpendicular to the acoustic wave vector and to decrease when it was applied parallel to the wave vector. The effect of the cooling rate was also studied. These results are explained by the occurrence of clusters of oriented dipoles borne by the Li$^+$ ions, favoured by the field.

Introduction.

The study of the disorder generated in KTaO$_3$ crystals by random substitution of K$^+$ ions by smaller Li$^+$ ions enters the general framework of the study of randomly interacting systems. Indeed, because of the different sizes of these ions, the Li$^+$ ions occupy off-centre positions thus generating electric and elastic multipoles. The interaction between the electric dipoles is thought to play a fundamental role in the formation of the low temperature phase, the nature of which is still controversial. The double nature of the multipoles allows the coupling to different probes: the electric dipole located on each substituted site is coupled to the electric field while the elastic quadrupole interacts with the local elastic strain. Moreover, the Li concentration $x$ changes the nature of the low temperature phase: dipolar glass below 2\% and disordered ferroelectric above. All these features, which still need to be carefully checked, make the $K_{1-x}Li_xTaO_3$ (KLT) crystals an attractive system for the study of random interactions in condensed matter [1, 2]. Actually, numerous techniques have already been applied to this system: X-ray diffraction [3], Raman scattering [4], optical birefringence [5, 6], nuclear magnetic resonance [7, 8], dielectric study [5, 9, 10], acoustic study [11-13] etc. However, due perhaps to technical difficulties, the acoustic measurements are rather few.

(*) Associated with the Centre National de la Recherche Scientifique.
In this paper we present the results of high frequency acoustic measurements performed on a KLT crystal of Li concentration equal to 1.7%, a value close to the border which is supposed to separate the two kinds of low temperature phase. In fact, the attention is focused on the role of the applied electric field. Subsidiary dielectric measurements which help with the interpretation of the acoustic data are also given. Finally, some conclusions concerning both the high and the low temperature phases are drawn.

Preliminaries on experiments.

All the present measurements on the complex elastic constant and the complex dielectric constant have been achieved on the same KLT sample. They are part of a systematic study on a series of KLT crystals of various concentrations. In order to keep a coherent labelling inside the series, we refer to this crystal as sample V. Acoustic measurements [12] and dielectric measurements [14] performed on this sample have already been published. In contrast to the present experiments, the former were done without biasing electric field while the latter were focused on the distribution of the relaxation times.

The sample was grown in our laboratory. It is characterized by its transition temperature $T_\gamma$, defined as explained below and found to be equal to 34 K. Then, using the relation $T_\gamma$ (K) = $535 \times (x^{2/3})$ [8], we deduce the lithium concentration $x = 1.7\%$. Another concentration measurement was independently performed by Secondary Ion Mass Spectroscopy (SIMS); using as reference a crystal with 5.7% Li ($T_\gamma = 79$ K) and measuring the ratio of the $^7\text{Li}^+$ -ion emissions, the lithium concentration is found to be $x = 1.6\%$ in our sample V. The agreement is very satisfying.

At room temperature, the KLT crystal has cubic symmetry. All the end-faces of the sample are cut perpendicular to one of the three equivalent fourfold crystallographic axes. Two pairs of opposite faces have received metallic films which serve as electrodes. On one of these electrodes a piezoelectric transducer is bounded, which can generate into the sample longitudinal waves propagating along a [100] axis; therefore the measured elastic constant $C$ coincides above $T_\gamma$ with the elastic constant $C_{\overline{1}}$ of the cubic symmetry class. Between the other pair of electrodes an alternative voltage is applied, for the dielectric measurements; in this case the measured dielectric constant $\varepsilon$ is, for $T$ larger than $T_\gamma$, any $\varepsilon_{\mu\nu}$ component of the isotropic dielectric tensor. Using one pair of electrodes or the other, a static biasing field may be applied during cooling. Then the biasing field may be either parallel or perpendicular to the displacement vector (and the wave vector) for elastic measurements and either parallel or perpendicular to the oscillating electric field vector for dielectric measurements.

The explored temperature range was between 4 K and 200 K. In most of the experiments, the data were recorded when heating the sample, without applied static electric field: they are zero field heating (ZFH) experiments (if a field was applied, this was during the cooling process only).

It is implied that the symbols $C$ and $\varepsilon$ stand for $C(\omega, T, E, r_c, r_h)$ and $\varepsilon(\omega, T, E, r_c, r_h)$ which mean that these quantities are two functions of the frequency $f (\omega = 2 \pi f)$ and the temperature $T$, measured upon heating, with the heating rate $r_h$ and without electric field, after the sample has been cooled in the electric field $E$, with the cooling rate $r_c$.

We present all our data in the form of dimensionless quantities. For the dielectric measurements we use the relative complex dielectric constant $\varepsilon$ while for the elastic measurements we give the reduced change of the complex elastic constant $\delta C/C_0$ which is related to the directly measured quantities by:

$$\text{Re} \left( \delta C/C_0 \right) = \delta C'/C_0 = 2 \frac{\delta V}{V_0}$$

$$\text{Im} \left( \delta C/C_0 \right) = \delta C''/C_0 = \alpha V_0 \ln \frac{10}{(20 \pi f)}$$
where $V_0$ and $C_0$ are respectively the sound velocity and the elastic constant measured at some reference temperature and $\alpha$ is the sound attenuation measured in decibels per length unit. The choice of $C_0$ is in fact of secondary importance since its change results in a simple translation of the velocity curve as a whole. However, in the theoretical part of this paper (see below) it is convenient to choose for $C_0$ the extrapolation of $C$ at $T = 0$.

The acoustic experiments were performed for frequencies $f$ ranging from 30 MHz to 330 MHz. We used an automatic method which was specially conceived for highly accurate sound velocity measurements [15]. The characteristic features observed when the temperature is changed (see Fig. 1, upper part) are:

1) from about 90 K to at least 220 K, a quasi-linear and dispersionless variation of the sound velocity, due to the lattice anharmonicity;

2) a dip in the sound velocity (a slowing down of the acoustic wave), due to a relaxational process, the position and intensity of which are frequency dependent (in the upper part of Fig. 1, it appears near 100 K);

3) an increase of the sound velocity when the temperature increases from the lowest temperatures to about 70 K;

4) a shoulder on this velocity increase, located at about 35 K independently of the frequency, attributed to a transitional effect (see below);

5) an attenuation peak, associated with the sound dip, and also frequency dependent;

6) an attenuation occurring below 70 K.

The frequency range of the dielectric experiments extended from $f = 10$ Hz to $f = 1$ MHz. The main observed features (see the lower part of Fig. 1) are:

1) a positive frequency dependent relaxational step in the real part of the complex dielectric

![Fig. 1. — Upper part: real and imaginary parts of the relative variation of the elastic constant measured at 273 MHz in a KLT sample with 1.7 % Li plotted as a function of temperature. Lower part: real and imaginary parts of the dielectric constant measured at 1 kHz. The two sets of data, performed in the same sample, refer to measurements without applied electric field during cooling and heating.](image-url)
constant (which would correspond to a slowing down of the electromagnetic wave at these frequencies), superimposed on a background which does not depend on frequency;

ii) a relaxational peak in the imaginary part of the complex dielectric constant, corresponding to the step.

Indeed the same process (\(\pi/2\) jump of \(\text{Li}^+\) ions) is responsible for the two relaxational attenuations and dispersions observed in the elastic and dielectric measurements. They appear at different temperatures (100 K and 50 K respectively) because the measurement frequencies are very different (five orders of magnitude). The displacement corresponds to an Arrhenius relaxation with a barrier height equal to 900 K [12].

All these features, in the complex elastic constant as well as in the complex dielectric constant, were observed upon heating without biasing field (ZFH) after a zero field cooling (ZFC). In the following sections the role of the cooling rate and the effects of field cooling (FC) are described.

**Dielectric experiments (data and analysis).**

There is a great difference between the data obtained from experiments with the biasing field \(E\) parallel or perpendicular to the oscillating field. This is shown for \(E = 75\) kV/m in figure 2, where the full lines show the data with \(E = 0\), for reference.

![Dielectric constant plot](image)

**Fig. 2.** — Real (left) and imaginary (right) parts of the dielectric constant measured at 68.5 Hz in a KLT sample with 1.7 % Li plotted as a function of temperature. All the measurements were performed without applied electric field, after cooling either in zero field (full line) or in a 75 kV/m static electric field applied parallel (dashed-doubly dotted line) or perpendicular (dashed-dotted line) to the oscillating electric field.

If the static field applied during FC is parallel to the oscillating field of ZFH, it generates contributions to the real part and the imaginary part of the dielectric constant which are both positive and vary as \(\omega^{-\nu}\). The exponent \(\nu\) and the amplitude of the contributions are temperature dependent: the value of \(\nu\) varies between 0.3 and 0.7 while the amplitudes have maxima near 22 K (dashed-doubly dotted lines). Another contribution of the same type is hardly observed near 50 K.

If the crystal is cooled in a biasing field perpendicular to the oscillating field, in addition to the features observed in the previous case, there are new positive contributions. The first ones are similar to those observed in the parallel case (proportional to \(\omega^{-\nu}\) for both real and
imaginary parts), but here with their maxima near 50 K. The second ones are large, independent of the frequency for the real part and varying as $\omega^{-1}$ for the imaginary part, over our lowest two decades (at higher frequencies, they become negligible); their maxima are also near 22 K, independently of the frequency (dashed-dotted lines). In figure 3, where the logarithmic scales show the frequency behaviour, the two contributions (i.e. the difference between the measurements with and without field) at 22.5 K and 50 K are sketched. The $\omega^{-1}$ contribution corresponds to losses independent of the frequency, that is to say, to a purely ohmic phenomenon.

We first remark that the frequency dependences of our data are in agreement with the Kramers-Kronig relations since they respectively correspond to the two susceptibility forms:

$$
\epsilon(\omega) = A [\cos (\nu \pi/2) - i \sin (\nu \pi/2)](\omega \tau)^{-\nu},
$$

$$
\epsilon(\omega) = A [\pi \delta (\omega \tau) - i \text{p.p.}(1/\omega \tau)],
$$

where $\tau$ is some relaxation time.

Secondly, we notice that in the two quoted experiments the same static electric field was applied along two equivalent perpendicular axes. Therefore, it is expected that the same effect is produced in both geometries. But in one case the remaining fourfold axis is perpendicular to the measuring electrodes, while in the other case it is in their plane. This explains the difference in our two sets of data.

The biasing electric field is known to generate a space charge trapped on the oxygen vacancies [16]. The hopping of the trapped electronic charges leads to a contribution to the complex dielectric constant proportional to $\omega^{-\nu}$ ($0 < \nu < 1$). Such a behaviour has been

![Graph](image-url)

**Fig. 3.** — Log-log plots of the difference between the dielectric constant measured after field cooling and measured after zero field cooling (real part: crosses and imaginary part: open circles) as a function of frequency for a KLT sample with 1.7 % Li for two temperatures. A 75 kV/m static electric field was applied perpendicular to the oscillating electric field during cooling. The dashed lines have a slope equal to $-1$ (top) and $-0.53$ (bottom).
described by Jonscher for many materials [17] and Ngai [18] has explained why it is so commonly encountered. We do observe this behaviour in the two geometries studied.

If the static field is supposed to also generate surface charges which occupy a thin layer at the interface near a biasing electrode, then our result can be understood as follows: if the oscillating field is in the plane of this layer (i.e. is perpendicular to the static field), an ohmic electronic conduction can occur inside the layer since the electrons may have a long path; this is not possible in the other geometry where the electron path is limited by the layer thickness.

Therefore, the main result of our dielectric measurements after a FC process is to show the presence of space charges which persist at temperatures much higher than \( T_t \) as shown by the \( \omega^{-r} \) contribution that extends up to at least 70 K. They strongly perturbed the dielectric measurements, specially under \( T_t \), possibly hiding the effect of the field on the dipoles. They also played a fundamental role in the interpretation of the acoustic experiments above \( T_t \) (see below).

Acoustic experiments (data).

Our experiments, as already said, were nearly all performed upon heating the crystal in zero field. This was because a high voltage applied on the sample could be hazardous for our very sensitive (10\(^{-13}\) W) detection. In order that the temperature of the sample be well equilibrated, our heating rate was low \( (r_h = + 0.25 \, \text{K per mn}) \).

We have first studied the role of the cooling rate in ZFC experiments. This can be seen in figure 4 where the dashed lines are relative to a rapid cooling (RC), with the cooling rate \( r_c = -5 \, \text{K per mn} \) while the full lines correspond to a slow cooling (SC) with \( r_c = -0.25 \, \text{K per mn} \), both with \( E = 0 \). After the SC, the sound velocity has decreased more (by a factor of 2) than after the RC, but the shoulder observed on both near 34 K is much more pronounced in the latter case (upper part of Fig. 4). Similarly, the SC induces a very strong

![Fig. 4. — Relative change of the elastic constant measured at 96 MHz, upon heating in zero field after zero field cooling, in a KLT sample with 1.7% Li as a function of temperature. Top : real part ; bottom : imaginary part. Full line: slow cooling \((-0.25 \, \text{K/mn})\), dashed line: rapid cooling \((-5 \, \text{K/mn})\).](image-url)
attenuation at low temperatures (lower part of Fig. 4). It is very clear on the velocity measurements that the two curves coincide only above 70 K and the quality of the coincidence shows that the discrepancy between them is much larger than our accuracy.

Our heating rate and our SC rate were similar (except for the sign) and, in that case, the velocity and attenuation curves were quasi-identical upon heating and upon cooling, indicating that some thermodynamical equilibrium is practically obtained in these two processes, at any temperature.

The FC processes were achieved with both SC and RC and the biasing field was \( E = 75 \text{ kV/m}. \) Figure 5 shows the role of the electric field and the role of the cooling rate (full line for SC and dashed lines for RC). Obviously, the direction of the field has a strong influence: if the field is applied parallel to the wave vector and the displacement vector (left part), it induces a large decrease of the sound velocity; on the contrary, if it is applied perpendicular to these vectors (right part), the result is an increase. However, the influence on the sound attenuation is rather weak, smaller than in the case \( E = 0 \) (compare Figs. 4 and 5). When the biasing field was \( E = 150 \text{ kV/m}, \) the effect was larger but not twice larger: there is a saturation.

\[
\text{Fig. 5.} \quad \text{Real and imaginary parts of the elastic constant measured at 96 MHz, upon heating in zero field after field cooling, in a KLT sample with 1.7 \% Li as a function of temperature. Static electric field applied parallel (left) or perpendicular (right) to the acoustic wavevector. Full line: slow cooling (– 0.25 K/mn), dashed line: rapid cooling (– 5 K/mn).}
\]

Indeed, the most important result, shown by figure 5, is that the two sound velocity curves relative to the two different cooling rates but the same field \( E \) are different at low temperatures and merge into a unique curve at 34 K. This remarkable temperature is the same for the two field orientations. Therefore, we attribute to this temperature the meaning of a transition temperature \( T_1. \)

The left part of figure 6 presents a sound velocity curve obtained after a ZFC and two curves relative to the two different field orientations. All were recorded after a slow cooling. However, they do not merge at \( T_1. \) Indeed, they coincide only at higher temperatures, near 70 K. This is also true for the sound attenuation, shown in the right part of the same figure.

All these effects do not depend on the frequency in the studied range.
**Acoustic experiments (analysis).**

Our experiments show that the FC process has no effect on the relaxation of the Li\(^+\) ions since the sound velocity and attenuation curves relative to FC and ZFC are identical in the temperature range of the relaxation step and relaxation peak (around 100 K for our frequency range). This is not surprising since the change of the electrostatic energy of an isolated dipole \(\mathbf{E} \cdot \mathbf{p}_{\text{eff}}\) is of the order of 1 K only, while a typical energy of the problem is the barrier height, equal to 1 000 K.

Therefore, only three features observed in our acoustic experiments have to be explained.

1. a) The elastic constant of cubic pure KTaO\(_3\) crystals is known to steadily increase when the temperature is decreased down to 4 K [19]. Here, this behaviour is observed on the sound velocity down to 70 K, but below, an unexpected decrease occurs even upon ZFC. This variation between \(T_i\) and 70 K is the first point to elucidate.

2. b) There is an effect of the electric field on the elastic properties of the KLT crystal, specially on the sound velocity, below \(T_i\). The effect is not weak: for comparison, with \(E = 75 \text{ kV/m}\) the relative velocity change is of the same order as the change induced by the lattice anharmonicity over 200 K. Moreover, it depends on the biasing field orientation. This is the second point to interpretate.

3. c) Between \(T_i\) and 70 K, the sound velocity is not the same after different FC processes. This remanence in the high temperature phase is the third point to explain.

We discuss our experimental results successively according to the following lines:

1. i) the occurrence of packets of oriented dipoles (clusters) which exist even in ZF and high temperature and favorized by the field (this explains the first two points);

2. ii) the presence of space charges, still present above \(T_i\), which generate a residual field (this explains the last point).

However, before a semi-microscopic analysis, we recall the main results of the Devonshire theory [20], which is a macroscopic and phenomenological description of ferroelectrics, because its predictions present some analogy with the observed features (it is expected to apply...
here if the clusters or domains are large enough). In this theory, the four temperatures $T_0 < T_c < T_1 < T_2$ play remarkable roles. They separate the different states listed below:

i) if $T < T_0$, only a ferroelectric phase can exist;

ii) if $T_0 < T < T_c$, a stable ferroelectric phase and a metastable non-polar phase are possible;

iii) if $T_c < T < T_1$, a metastable ferroelectric phase and a stable non-polar phase are possible;

iv) if $T_1 < T < T_2$, the only stable phase is non-polar but an applied field can induce a ferroelectric phase;

v) if $T_2 < T$, the only stable phase is non-polar and even an applied field cannot induce a ferroelectric phase.

Such a system possesses the following properties:

a) in absence of field, it reversibly follows a cycle starting from a temperature higher than $T_1$, going down to a temperature higher than $T_c$ and then coming back to the initial temperature, always staying in a non-polar phase;

b) if it follows the previous cycle with an applied field during the cooling process, it is driven into a ferroelectric phase which persists upon heating, if the field is switched off, up to the temperature $T_1$.

Now, if we assume that $T_1 = 34 \text{ K}$ and $T_c < 4 \text{ K}$ (the lowest temperature in our experiments), our observations are well described. In ZFC down to 4 K, no transition happens: the crystal stays in a non-polar phase. In FC down to 4 K, a stable ferroelectric phase is induced, which becomes metastable when the field is switched off; then upon heating this phase persists up to 34 K where it transforms into a non-polar phase. In this scheme the limit-temperature $T_i$ must be independent of $E$, as we observe in first approximation and must be identified with $T_e$. Indeed some rounding exists, below $T_1$ because of the relaxation of the metastable state towards the stable non-polar state and above $T_1$, due to the residual field associated with the space charges.

In this form, the Devonshire theory is, in principle, valid only for homogeneous media since it neglects fluctuations and disorder. In fact, the KLT system is disordered. This leads to specific properties due, in particular, to static random fields (SRF).

Then we turn to a more microscopic description of the KLT crystal, assuming the presence of clusters which takes into account the fluctuations of polarization and we calculate the effect on the elastic constant. We emphasize that we use the word «cluster» with the simple meaning of packet or small region; in particular, no idea of segregation of lithium atoms is implied.

The off-centre Li$^+$ ions are displaced in one of six equivalent positions along the three local fourfold axes. This fact, associated with the dipolar interactions, gives a trend towards lowering, from cubic to tetragonal, of the symmetry of the crystal and cluster growth. In what follows the superscripts C and T stand for cubic and tetragonal. In zero field there are as many clusters oriented in one direction as in each other. On the contrary, if a field is applied, there are more clusters elongated in the direction of the field than in the perpendicular directions. The elastic constants $C_{33}^T$ and $C_{11}^T$, which govern the propagation of the longitudinal waves along the fourfold axis and the relevant twofold axes respectively, are not equal in a crystal with tetragonal symmetry. If the acoustic wavelength $\lambda (\lambda = 80 \mu \text{m} \text{ at } 100 \text{ MHz})$ is large in comparison with the cluster size, the elastic constant $C$ is approximately the mean value calculated from $C_{11}^T$, $C_{11}^T$, $C_{33}^T$ and the volume fraction occupied by each type of clusters.

Let be $p_i = p_i(T, E, r_e, r_h)$ the volume fraction occupied, at temperature $T$, by clusters with their tetragonal axis along the $i$ axis of the laboratory frame ($i = x, y$ or $z$), after a cooling process in field $E$, with a cooling rate $r_e$ and a heating rate $r_h$. The values that the fraction may
take are limited by the conditions $0 \leq p_i \leq 1$ and $0 \leq p_x + p_y + p_z \leq 1$. If $E = 0$, then $p_x = p_y = p_z$. The $z$-axis is chosen parallel to the acoustic wavevector and therefore the propagation is governed by the mean elastic constant $C$ defined as:

$$C = [1 - (p_x + p_y + p_z)] C_{11}^0 + (p_x + p_y) C_{13}^0 + p_z C_{33}^0.$$  

Due to the lattice anharmonicity, the elastic constant $C_{11}^0$ of cubic pure KTaO$_3$ crystals linearly increases when the temperature is decreased down to 4 K [19]. We assume the same behaviour for the other two elastic constants and moreover we take the same anharmonicity coefficient $b$ for the three cases and we write down, with $i = 1$ or 3 and $X = C$ or $T$:

$$C_{ii}^X(T) = C_{ii}^X(0) - bT.$$  

The contribution due to the relaxation of the Li$^+$ ions is not written since it is noticeable only near 100 K, well above the range of interest for the present discussion. Henceforth, we use reduced quantities, taking as reference $C_0 = C_{11}^0(0)$ and putting

$$K_{11} = C_{11}^0(0)/C_0, \quad K_{33} = C_{33}^0(0)/C_0 \quad \text{and} \quad B = b/C_0,$$

we get:

$$[C(T) - C_0]/C_0 + BT = (p_x + p_y)(K_{11} - 1) + p_z(K_{33} - 1).$$  

The temperature variation is the sum of the linear effect due to anharmonicity and the change due to the three fractions $p_i$.

From our experiments performed in zero field and from others, performed in a field strong enough to exhibit saturation, we put forward a very crude estimate for the reduced elastic constants $K_{ii}$. They are obtained from the difference between the experimental data and the variation, assumed linear in temperature, due to anharmonicity. In the upper part of figure 4, this variation would be represented by a straight line (not drawn but easily imagined) tangent to the curve in the 100-200 K range and cutting the vertical axis near 0.07. This value is valid for all our figures showing $\delta C'/C_0$ since the same reference was chosen. Consequently, the left part of figure 6 shows that the effect of the clusters is slightly positive in one case (dashed-dotted line, corresponding to cooling in a field parallel to the acoustic wavevector) and clearly negative in the other two cases (full line and dashed-dubly dotted line, corresponding to cooling in zero field and in a field perpendicular to the acoustic wavevector, respectively). More precisely, we find:

$$(2/3)(K_{11} - 1) + (1/3)(K_{33} - 1) \approx -0.05,$$

$$(K_{11} - 1) \approx +0.01 \quad \text{and} \quad (K_{33} - 1) \approx -0.15.$$  

Brillouin scattering measurements have been performed [21] on a KLT crystal which exhibits a first order transition at $T = 60$ K. With our convention this corresponds to the concentration $x = 3.8\%$. From their values we can deduce:

$$(K_{11} - 1) \approx +0.04 \quad \text{and} \quad (K_{33} - 1) \approx -0.33.$$  

Considering the difference of concentration, the agreement with our estimate is quite good for the order of magnitude and especially the signs.

In order to complete the model, a calculation of $p_i(T, E, r_c, r_h)$ is needed. It must take into account both the features described by the Devonshire theory and the quadrupolar (probably, dipolar too) fluctuations. This is beyond the scope of the essentially experimental present paper.
However, the model deserves several comments.

i) The observed increase (when \( T \) increases) of the elastic constant above \( T_1 \) was also present in other KLT samples [13] with lithium concentrations equal to 1.5 \%, 3.5 \% (samples I and II of our systematic study) but not clearly for 5 \% (sample III), perhaps because in the last case the transition occurs at too high a temperature (75 K).

ii) These acoustic experiments on sample III support our scenario: upon cooling nothing happens at 75 K whilst upon heating a quasi-discontinuity is observed at this temperature, giving it the meaning of the limit of metastability \( T_1 \). In this case, things are clearer since, because of the higher concentration, a biasing field is not needed to induce the ferroelectric phase.

iii) In ZFC experiments, it is not true that nothing happens at \( T_1 \). Indeed there is a slight shoulder in the sound velocity curve. This may be due to SRF which can locally induce ferroelectric microregions.

iv) In this schema, the weakness of the effect of the electric field on the dielectric properties is attributed to the low anisotropy of the dielectric properties. Indeed, the biasing field in the present experiments is notably smaller than was applied by other authors [22, 23].

v) The role of clusters, with symmetry lower than cubic, present even above \( T_1 \), is reminiscent of the interpretation given by Di Antonio et al. of their Raman scattering data [4] obtained on samples I and II. Because of the symmetry of their probe, they can attribute a polarization to the clusters. In our case, since we use another probe, we only show the quadrupolar nature of the clusters, which is not incompatible with the dipolar nature. We think that they are the same entities in both cases.

vi) Preliminary Raman scattering experiments performed in our sample V by Toulouse [24] show the presence of polarized clusters at temperatures up to 105 K. This clearly confirms our interpretation.

Our dielectric measurements show that a static electric field generates space charges in KLT crystal, still present up to 80 K (see Fig. 5). These charges induce an internal electric field [16] which produces on the elastic constant an effect similar (in the same way) to that given by the external field which has generated the space charges. This explain why the splitting due to FC persists at high temperatures, much above \( T_1 \).

Conclusion.

The acoustic experiments in a KLT crystal with 1.7 \% Li under an applied electric field reported here show the existence of a transition temperature at 34 K, hardly to be seen in zero field. The metastability of the low temperature phase obtained during field cooling is also shown.

The experiments without electric field are in agreement with the occurrence of ordered microregions (clusters) even above the transition which appears very fuzzy (neither first order nor second order). These ordered microregions are obviously present, and probably more developed, below the transition. This result seems to rule out the hypothesis, generally accepted for Li concentrations lower than 2 \%, of a dipolar glass phase.

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[12] Doussneau P., Frénois C., Levelut A., Ziolkiewicz S. and Höchli U. T., *J. Phys. Cond. Matter* **3** (1991) 8369. In this paper the concentration of sample V was given equal to 5%. At that time, the transition at 34 K was not seen in absence of electric field and the SIMS measurements were not performed.