

## Spontaneous and field-assisted transition in $K_{0.984}Li_{0.016}TaO_3$ : The polar pattern by birefringence and second-harmonic generation

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We report birefringence and second-harmonic-generation measurements in  $K_{0.984}Li_{0.016}TaO_3$  at temperatures between 55 and 10 K. On cooling, the local spontaneous polarization grows from  $\approx 1$  to 30 mC/m<sup>2</sup>. We find, however, that its correlation length obtains the value of  $\approx 8$  nm at 55 K and remains unchanged on further cooling. It appears that a "frozen" polar length scale arises from the interaction of the Li dipoles well above the usually defined transition temperature ( $\approx 36$  K).

### I. INTRODUCTION

Lithium-doped potassium tantalate (KTL) has been the object of an extensive experimental and theoretical investigation.<sup>1-3</sup> The Li ions, which substitute for K, can move in six equivalent off-center positions, producing in this way a large dipole moment. Similarly to the case of the alkali halides, the Li dipoles can interact through dipole-dipole forces. However a richer complexity can arise in KTL from the fact that dipoles are hosted in the highly polarizable potassium tantalate matrix which is an incipient ferroelectric by itself. The remanent polarization, the behavior of the susceptibility,<sup>1,4</sup> and NMR data<sup>5</sup> were found to be consistent with a glassy state at low temperature, and the analogy with spin glasses suggested to depict the transition from a paraelectric to a polar state in terms of random dipole interaction.<sup>1,4</sup> Other authors favored the different interpretation of a transition of ferroelectric type;<sup>6</sup> this view was independently supported by calculations of the effective dipole coupling in mean-field approximation.<sup>2</sup> A disordered polar state was proposed to arise from random fields, or, at low doping, from Li dipoles which "freeze" above the transition temperature.<sup>7</sup> But also in this case the question remains whether the Li ions freeze individually or interacting among themselves, and, ultimately, what is the nature of this interaction. While the picture of a glassy state at low temperature has gained consensus for lightly doped KTL, a few experiments have given evidence of the short polar correlation range which is expected in the disordered state.<sup>8-11</sup> At low temperature, no long-range order was evidenced by x-ray structural studies.<sup>8</sup> Raman studies show the polar order to be shorter than in ferroelectrics.<sup>12,13</sup> Absence of Brillouin scattering from microwave pumped phonons indicated the spontaneous polar correlation length  $\xi$  to be smaller than 10 nm,<sup>10</sup> while  $\xi \approx 6$  nm was suggested, always at low temperature, by second-harmonic generation (SHG).<sup>11</sup> To our knowledge, however, no data were reported so far on the polar pattern above the transition temperature (hereafter denoted by  $T_f$ ), and how the pattern evolves on cooling. Tracking the polar correlation length throughout  $T_f$  would give the most direct indications of a criticality in the transition. Furthermore, we feel that the polar features above  $T_f$  can be more directly related in the interaction

among Li dipoles since the displacement of the host matrix is smaller at higher temperatures.

In this paper we address the case of lightly doped KTL. Combined linear birefringence (LB) and SHG measurements show that the spatial correlation of the polarization does not change on cooling below  $\approx 55$  K, which implies that the spontaneous clustering of Li dipoles is already determined above  $T_f$  ( $T_f \approx 36$  K). We derive a constant  $\xi \approx 8$  nm from 55 to 10 K.

### II. THE EXPERIMENT

The sample was grown by the spontaneous nucleation technique. Li concentration was  $x = 0.016$ . At such low doping the quadrupolar order is modest, and one can avoid considering ferroelastic effects in the transition. This simplification is not granted at higher doping; in fact, in  $K_{1-x}Li_xTaO_3$  the degree of spontaneous quadrupolar alignment along one of the cubic axis becomes almost total at  $x > 0.03$ ,<sup>11</sup> and even periodic layers with alternate polarization have been reported for  $x > 0.04$ .<sup>14</sup> The sample was cut into a 5-mm-thick parallelepiped with the sides (parallel to  $x, y, z$ ) oriented along the four-fold crystalline axes. The large thickness was necessary to obtain detectable signals at the higher temperatures. Both the He-Ne beam (employed in LB) and the pump pulse (for SHG measurements) propagated along  $x$ , perpendicularly to the  $y$ - $z$  faces. The sample was cooled from 100 to 10 K (or heated) at the constant rate of 1 K/min. The basic setup is similar to the one of Ref. 15. For SHG, the pump pulse (wavelength 1.06  $\mu$ m, pulse duration 20 ns) was focused, with a long focal length, to obtain a  $\approx 0.5$ -mm spot at the sample. We denote by  $S_{ij}$  the SH energy/pulse collected in a forward  $f/3$  aperture; the subscripts  $i$  and  $j$  ( $i, j = y, z$ ) indicate the polarization direction of the pump and of the SH, respectively. To obtain the magnitude of  $S$ , we calibrated the setup with a quartz platelet (a standard reference for SH generation). For LB, the He-He beam was either collimated to the size of the whole sample for a qualitative inspection, or focused to a waist of 30  $\mu$ m for the quantitative measurements. The spot of the focused He-Ne was kept inside the larger one of the SH pump. LB is observed in KTL when the probing optical field is polarized along the  $[011]$ , or  $[01\bar{1}]$  axis, a fact consistent with the assump-

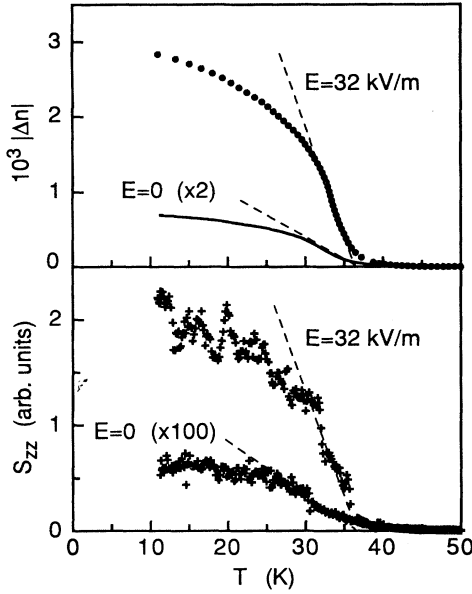


FIG. 1. Temperature dependence of  $|\Delta n|$  (above) and of  $S_{zz}$  (below) recorded in zero-field and field cooling. Broken lines: fit to the linear part with  $|\Delta n| = a(T - T_f)$ .

tion that the local spontaneous polarization  $\mathbf{P}$  is parallel to one of the pseudocubic axis. The quantity measured by LB is  $|\Delta n| = |n_z - n_y|$ , with  $n_z$  and  $n_y$  the average value of the refractive indexes in the probed volume. In the unpoled sample the magnitude of  $|\Delta n|$ , but not its behavior with temperature, changed with the point of measurement. We could identify an area of  $\approx 2$  mm size which appeared rather uniform when viewed through crossed polarizers at  $T \approx 30$  K. Both LB and SHG measurements were performed within this more favorable zone. In this area, we also selected the points where the LB signal maintained full oscillations as  $T$  approached 10 K. Plots of  $|\Delta n(T)|$  and  $S_{zz}(T)$  are reported in Figs. 1 and 2. The ratio  $S_{zz}/S_{yy}$  was found not to depend on

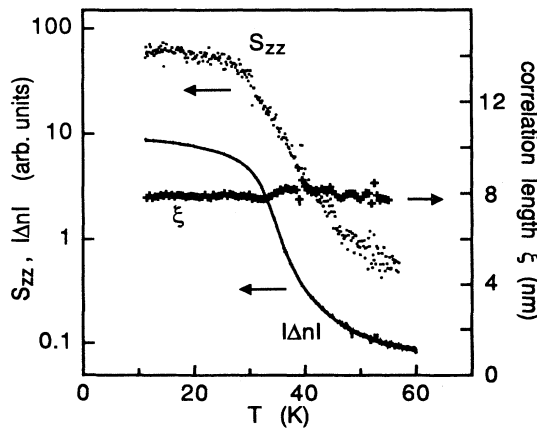


FIG. 2. Plots of  $|\Delta n|$  and  $S_{zz}$  versus  $T$  (logarithmic vertical scale). The correlation length  $\xi$  (vertical scale at the right) is derived from  $|\Delta n|$  and  $S_{zz}$  (see text).

temperature. We also performed measurements in field cooling (FC). The poling field, denoted by  $E$ , was parallel to  $z$ . The field was always applied at 100 K, and the sample was heated to this temperature before cooling at a different  $E$ . Effects of thermal hysteresis were negligible: the same LB and SHG have been found during the heating part of the cycle, when the sample was brought back to 100 K.

### III. RESULTS

#### A. Birefringence

Denoting by brackets the averages on the probed volume, the refractive index anisotropy, which is responsible for LB, is given by

$$\Delta n = C[\langle P_z^2 \rangle - \langle P_y^2 \rangle] = C(f_z - f_y)\langle P^2 \rangle \quad (1)$$

with  $C = 1/2n_0^3(g_{11} - g_{12})$ , and  $\sum_i f_i = 1$ . For pure  $\text{KTaO}_3$ ,  $n_0 = 2.2$ ,  $g_{11} - g_{12} = 0.16 \text{ m}^4/\text{C}^2$ . Assuming that the local values of  $P^2$  do not depend on the direction of  $\mathbf{P}$ , the quantities  $f_i$  coincide with the volume fraction of the three quadrupolar directions ( $\mathbf{P}$  parallel to  $\pm i, i = x, y, z$ ). The temperature dependence of  $\langle P^2 \rangle$  is usually derived from the measured  $|\Delta n(T)|$  by relying on the implicit assumption that  $f_z - f_y$  remains constant throughout the temperature change. As it will be clear below, support to this procedure was here given by the observation that  $S_{zz}/S_{yy}$  did not change with  $T$ . In particular, SHG measurements in zero-field cooling (ZFC) indicated that  $f_y/f_z = 1.6$  in the spot of present measurements (all data here reported have been taken in this spot). Assuming  $f_x = 1/3$ , we then derived  $|f_z - f_y|$  which, substituted together with the measured  $|\Delta n|$  in Eq. (1), allowed to obtain  $P = \langle P^2 \rangle^{1/2}$ .  $P$  increases from  $\approx 1 \text{ mC/m}^2$  at 55 K to  $35 \text{ mC/m}^2$  at 10 K. Some error in  $P$  might result from the assumption  $f_x = 1/3$  and from the fact that, being the He-Ne spot smaller than that of the YAG laser, the exact value of  $f_y/f_z$  involved in LB might have been slightly different from the average one given by SH. We remark that in ZFC one finds a zero average polarization and then  $\langle P_i \rangle = 0$ .

We identify  $T_f$  with the temperature at which the extrapolated linear growth of  $|\Delta n(T)|$  intercepts the abscissa (Fig. 1). As the temperature was decreased below  $T_f$ , it became progressively more difficult to produce a change of SH (or LB) by switching on/off the poling field without returning to high temperature. Indeed  $T_f$  (or  $T_g$ , as the transition temperature is often called) is associated with the stability limit of the remanent polarization.<sup>5</sup> The value we find ( $T_f = 36.5$  K) falls in the range 34–38 K reported in the literature.

The appearance of LB requires  $f_y - f_z \neq 0$ . In lightly doped KTL, the fact that LB occurs also in zero-field cooling can be attributed to built-in stresses which might favor a preferential quadrupolar orientation. Since stresses can couple to the square of  $\mathbf{P}$ , but not to  $\mathbf{P}$ , they are not expected to induce a preferential dipolar orientation.

### B. Second harmonic

In present experiment, the inelastic SH component (hyper-Raman scattering) is expected to be negligible in comparison with the elastic one (hyper-Rayleigh scattering).<sup>16,17</sup> The SH emission can then be accounted for by introducing the nonlinear, second-order optical susceptibility  $\chi^{(2)}$ . The third-order tensor  $\chi^{(2)}$  vanishes in a centrosymmetric material. This is not the case in presence of a local polarization, where one has  $\chi^{(2)}(\mathbf{r}) = \mathbf{b} \cdot \mathbf{P}(\mathbf{r})$ .<sup>18,19</sup> Simplifying to a scalar notation, and assuming a pump beam of wave vector  $\mathbf{k}_\omega$ , the SH field of wave vector  $\mathbf{k}_{2\omega}$  is proportional to  $\int bP(\mathbf{r})e^{i\mathbf{K}\mathbf{r}}d\mathbf{r} = P(\mathbf{K})$ , where the integration is extended to the probed volume and  $\mathbf{K} = \mathbf{k}_{2\omega} - 2\mathbf{k}_\omega$ . Denoting by  $s(\theta)$  the SH intensity radiated at the scattering angle  $\theta$  (the angle between  $\mathbf{k}_{2\omega}$  and  $\mathbf{k}_\omega$ ) one has  $s(\theta) \propto |P(\mathbf{K})|^2$ .

Introducing the correlation function  $g_i(\mathbf{r}) = \langle P_i(0)P_i(\mathbf{r}) \rangle$ , with  $P_i$  the  $i$  component of  $\mathbf{P}$  and its Fourier transform  $G_i(\mathbf{K})$ , the SH signal which is collected within the solid angle  $\Omega$  is given by

$$S_{ij} = Mb_{ijj}L f_j \langle P^2 \rangle \int_{\Omega} G_j(\mathbf{K}) d\Omega, \quad (2)$$

where  $L$  is the sample thickness,  $M$  includes constants and geometrical factors, and the  $b$ 's (reported in Ref. 19 for pure  $\text{KTaO}_3$ ) are related to the Miller coefficients  $\Delta$ . In KTL, the SH intensities detected in the forward  $zz$  and  $yy$  configurations are due to  $P_z$  and  $P_y$ , respectively. The scattering wave vector  $K$  decreases with  $\theta$  and reaches at  $\theta=0$  the minimum value given by  $K_{\min} = k_{2\omega} - 2k_\omega = \pi/l_c$  ( $l_c = 2.8 \mu\text{m}$  in our case). The finite  $K_{\min}$ , which is due to the frequency dispersion of the refractive index, denies a direct probing of the spatial components of  $\mathbf{P}$  of wavelength  $\Lambda = 2\pi/K$  larger than  $l_c$ . For unpoled KTL this restriction is irrelevant since the spontaneous polar correlation length  $\xi$  is shorter than  $l_c$  as it was indicated by the magnitude of the SH and by its isotropic angular distribution.<sup>11,15</sup> Actually isotropic emission implies that  $\xi < \lambda$ , with  $\lambda$  the wavelength of SH. In this case, assuming for both  $P_z$  and  $P_y$  the same correlation function  $g(r) = \exp(-r/\xi)$ , the expression for  $S_{ii}$  ( $i=z,y$ ) simplifies to

$$S_{ii} = MbL f_i \langle P^2 \rangle \xi^3 \Omega, \quad (3)$$

The result is largely independent from the exact  $g(r)$  adopted.<sup>15</sup> From the measured  $S_{zz}$  and  $S_{yy}$  we derived the ratio  $f_z/f_y$  which was used to obtain  $\langle P^2 \rangle$  from LB data.

According to Eqs. (1) and (2), a change of  $G(\mathbf{K})$  with the temperature must manifest itself in a different behavior of  $|\Delta n|$  and  $S_{zz}$  versus  $T$ . Comparing the curves of Fig. 2, one notices that the signals of SH and LB have a similar  $T$  dependence below 55 K, which reveals that the pattern of dipolar order does not change, once established above  $T_f$ . More precisely, making use of Eqs. (1) and (3), one finds that the spontaneous polar correlation length is given by

$$\xi(T) = \text{const} \times [(1 - f_y/f_z) S_{zz}(T) |\Delta n(T)|^{-1}]^{1/3}.$$

The plot  $\xi$  versus  $T$  is reported in Fig. 2. From 55 K

(where we start to detect reliable signals) down to 10 K, the correlation length turns out practically constant with no evidence of criticality. The magnitude  $\xi = 8 \text{ nm}$  was obtained by making use of the calibration of the setup with the reference nonlinear crystal, and by introducing the solid angle of collection and the value of  $b$ . We notice that KTL behaves quite differently to this regard from what we found in  $\text{KTaO}_3:\text{Na}$  and in  $\text{KTaO}_3:\text{Nb}$ . In  $\text{K}_{0.85}\text{Na}_{0.15}\text{TaO}_3$ , for example,  $S_{ii}(T)/|\Delta n(T)|$  increased almost three orders of magnitude and the pattern of the SH emission peaked forward on cooling below  $T_f$ , both findings clearly showing the growth of the polar correlation range.<sup>15</sup>

### C. Measurements with poling field

Plots of  $|\Delta n|$  and  $S_{zz}$  versus  $T$  in FC are shown in Fig. 1. In Figs. 3(a) and 3(b) we reported the dependence of LB and SH on the strength of  $E$  at  $T = 10 \text{ K}$ ; data at  $T \approx T_f$  showed a similar trend. One can notice that  $S_{zz}$  changes significantly with a small applied field, much more than LB. This is consistent with the fact that poling fields of few  $\text{kV/m}$  hardly affect the average quadrupolar orientation, and the change of  $f_z/f_y$  is modest. On the contrary, bringing even a small fraction of dipoles into  $+z$  has a large effect on the correlation length of  $P_z$ .

Interesting is the behavior of LB. Starting from the preferential  $\pm y$  quadrupolar orientation in zero-field cooling (ZFC), and being  $E$  directed along  $z$ ,  $|\Delta n|$  vanished at intermediate poling fields [in Fig. 3(b) one notices that  $|\Delta n| = 0$  at  $E \approx 6 \text{ kV/m}$ ]. From the measured  $|\Delta n|$ ,

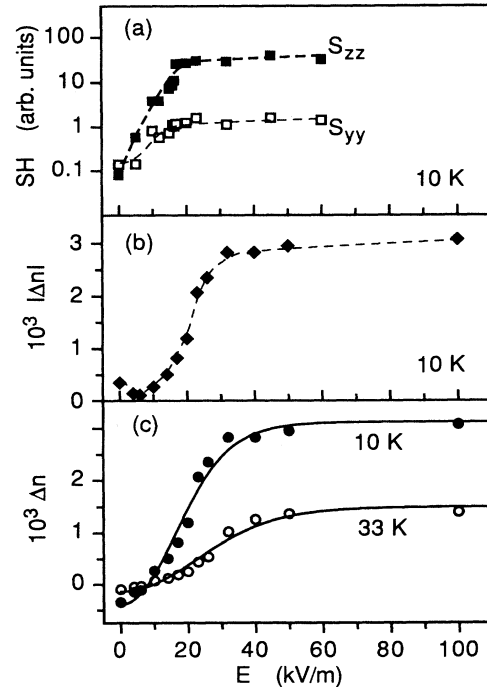


FIG. 3. Plots versus the poling field  $E$  of (a)  $S_{zz}$  and  $S_{yy}$  — note the logarithmic vertical scale; (b)  $|\Delta n|$ ; (c)  $\Delta n$ . The solid lines in (c) are a fit with a Langevin-type function (see text). Temperatures as indicated in the figure.

it is then reasonable to assume that  $\Delta n$  versus  $E$  is given by the plot of Fig. 3(c). The dependence of  $\Delta n$  on the strength of  $E$  appears consistent, at least qualitatively, with the picture that dipoles are arranged in polar clusters of few nanometers size. A crude attempt to relate the changes of  $f_i$  with the magnitude of  $E$  can be done by assuming that a Langevin type of equation applies to polar entities, each reorienting as a whole unit under the action of the poling field. In absence of mutual interaction among units, the probability  $p(i)$  for a unit to be oriented along  $i$ ,  $i = \pm x, \pm y, \pm z$ , is given by  $p(i) \approx Z^{-1} \exp[(-W_i - \mathbf{d}^* \cdot \mathbf{E})/k_B T]$ , where  $Z$  is the partition function,  $W_i$  accounts for the preferential quadrupolar orientation observed at  $E = 0$ , and  $\mathbf{d}^*$  is the effective dipole moment of the unit. The fractions of quadrupolar order can be calculated through  $f_i^* = p(i) + p(i)$ , with  $i = x, y, x$ . From Eq. (1), the predicted dependence of  $\Delta n$  on  $E$  is given by  $\Delta n(E) = D[f_y^*(E) - f_z^*(E)]\langle P^2 \rangle$ . In the model,  $\langle P^2 \rangle$  is independent from  $E$ , and then one has  $\Delta n(E) \propto [f_y^*(E) - f_z^*(E)]$ . Fitting this dependence to the data [solid lines in Fig. 3(c)] gave  $d^* \approx 5 \times 10^{-26}$  cm  $\approx 3000d$ , with  $d$  the bare dipole moment of the off-center Li ion. For the effective  $d_s^*$  of a single Li dipole, Ref. 20 reports  $d_s^* \approx 25d$ , while, from Ref. 2—taking  $\chi = 2000$  for the static susceptibility at  $\approx T_f$  and  $\gamma = 0.1$  for the local-field correction factor—one evaluates  $d_s^* = \chi \gamma d / 3 \approx 70d$ . Whatever the precise value of  $d_s^*$ , the  $d^*$  of the fit is too large to be due to independent Li dipoles and suggests that clusters of  $\approx 40$ – $120$  dipoles reorient as a single unit on poling. With 1.6% doping, one derives an average cluster size of  $\approx 6$  nm, which, considering the crudeness of the model, appears in broad agreement with the spontaneous correlation range derived from SHG.

SH emission changed significantly by applying a poling field:  $S_{zz}$  increased orders of magnitude (Figs. 1 and 3), while the pattern peaked at small  $\theta$ 's with almost all of the SH emitted in a forward cone of  $\approx 0.1$  rad semiaperture (Fig. 4). We notice, however, that SH data in FC (for a thorough investigation see Ref. 14) do not allow to derive unambiguously the polarization pattern and the ratio  $f_z/f_y$  in absence of further information. In fact, the poling field forces a long-range order on the whole crystals, and, writing  $P_z(\mathbf{r}) = \langle P_z \rangle + \delta P_z(\mathbf{r})$ , one has now a large  $\langle P_z \rangle$ , and, very likely, long-wavelength components of  $\delta P_z(\mathbf{r})$  which cannot be assessed by the SH technique due to the limitation on  $K_{\min}$ . In any case, the fact that SH emission is contained in a cone of 0.1 rad dictates that there is a scale length of  $\delta P_z(\mathbf{r})$  comparable or larger than one micron. [Unfortunately, due to stray-light reflections, we could not measure the smaller short-wavelength components of  $\delta P_z(\mathbf{r})$ .] However small,  $\delta P_z(\mathbf{r})$  is not zero; when the crystal was rotated, we also did not observe the Maker fringes which are expected in uniformly polarized media.<sup>19</sup> Permanence of some dipole clusters in the preferential  $\pm y$  quadrupolar direction could be a reason of the spatial fluctuation  $\delta P_z(\mathbf{r})$ . We notice that presence of some disorder under field cooling has been reported by the authors of Ref. 7 from LB and refractive index measurements.

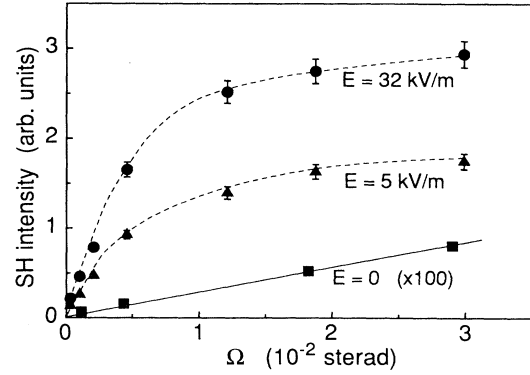


FIG. 4.  $S_{zz}$  at 10 K versus the solid angle of collection at different poling fields. The broken lines are only a guide to the eye. The solid line is a linear fit to the data taken in zero-field cooling.

#### IV. DISCUSSION

By cooling from 55 to 10 K, we observed a large growth of the magnitude of the spontaneous polarization  $P$ , but no changes of its correlation range. The picture we propose is that already at 55 K, and then well above  $T_f$ , the system breaks into patches of  $\approx 10$  nm size which are characterized by an average polarization of  $\approx 1$  mC/m<sup>2</sup>. Responsible for these polar patches are the Li ions which have moved to off-center positions. We remark that inside these clusters some Li ions could still be mobile, and readjustments of single Li dipoles could take place. The growth of  $P$  that is observed on cooling below  $T_f$  [the  $T$  dependence below  $T_f$  even resembles that of ferroelectrics with  $P \propto (T_f - T)^{1/2}$ ] can be attributed to the progressive displacement of the KTaO<sub>3</sub> matrix, which maintains the polar pattern of the Li dipoles above  $T_f$ . We remark that the impurity dipoles ( $d = 1.6 \times 10^{-29}$  cm,  $n_{\text{Li}} = 2.5 \times 10^{20}$  cm<sup>-3</sup>) by themselves can only account for a small fraction of the low-temperature polarization ( $P \approx 30$  mC/m<sup>2</sup>), a fact that calls for the dominant contribution of the matrix.

A similar size of the polar clusters ( $\approx 5$  nm) has been obtained from the linewidth of the TO<sub>1</sub> phonon observed in the same sample through Raman scattering.<sup>13</sup> Also this technique suggested that there are no changes of the size of the clusters in cooling below  $T_f$ ; in fact, while the intensity of the Raman line increased somewhat from above to below  $T_f$ , its linewidth was hardly affected. It is interesting to notice that no true softening of the TO<sub>1</sub> phonon has been observed by Raman scattering in KTL, differently from the case of KTaO<sub>3</sub>:Na, and KTaO<sub>3</sub>:Nb. It appears that the absence of a proper softening mode is associated with a polar correlation length which remains constant.

The picture we propose is consistent with the findings of the early NMR measurements. In fact, the authors of Ref. 5 found that the hopping of the Li ions among the possible sites slows down on cooling from 100 to 50 K. At the latter temperature, the Li ions are frozen in one of the off-center positions for the NMR time scale.

Different is the behavior of Ta ions of the matrix, which they observed to “freeze” off-center well below  $T_f$ . In lightly doped KTL, freezing of part of the Li dipoles above  $T_f$  has been suggested also by Kleemann in order to explain the low-temperature glassy state.<sup>7</sup> On whether the symmetry-breaking defects,<sup>21</sup> in this case the Li ions, freeze individually or interacting among themselves,<sup>22</sup> the correlation range of nanometers that we found definitely supports that they do mutually interact. A cooperative interaction above  $T_f$  appears possible when one considers the size of the distorted matrix around a single Li dipole. A microscopic description of the matrix distortion has been given at zero temperature through lattice-dynamical models.<sup>23</sup> In nominally pure  $\text{KTaO}_3$ , from the Raman line-shape analysis of  $\text{TO}_1$  phonon, Uwe *et al.*, have evidenced that “ferroelectric” regions of distorted lattice form around defects; their radius  $R$  increases  $\approx 3$  times in cooling from 100 to few kelvin, and obtains the value  $R \approx 2a$ , with  $a$  the lattice cell, at 50 K.<sup>24</sup> Similar values were reported in Ref. 16 from a comparison of hyper-Rayleigh and hyper-Raman scattering. In 1.6% KTL the average distance among dopants is  $D \approx 4a$ , and is natural to think that the Li dipoles do interact among themselves.

Present theories, however, are still inadequate to account for the complexity of the interaction. Theoretical approaches predict that the interaction  $-\mathbf{J}_{ij} \cdot \mathbf{d}_i \mathbf{d}_j$  among dipoles leads to a glassy state when  $J$  has a variance larger than its average:  $\text{Var}(J) \gg \langle J \rangle$ ; in the other case,  $\text{Var}(J) \ll \langle J \rangle$ , a ferroelectric long-range order is expected.<sup>1</sup> The difficulty is to establish an expression for  $J$ . Assuming that  $J$  can be approximated with a pure dipole-dipole coupling among randomly distributed impurities, one has  $\langle J \rangle = 0$ . Qualitatively, a glassy state is in agreement with present results. Unfortunately, no quantitative prediction of the magnitude of  $\xi$  are available to compare with our data.

In highly polarizable media, the assumption of a pure dipole-dipole interaction was questioned by Vugmeister and Glinchuk.<sup>2</sup> According to their calculations, due to the lattice distortion around each dipole,  $J$  has also an isotropic component which cooperatively aligns the dipoles and then promotes a long-range ferroelectric order when the ratio  $R/D$  exceeds a threshold. At variance with these last predictions, we find absence of criticality and a short correlation range. In our opinion, however, the discrepancy might be due to the fact that in KTL the Li ions cooperatively interact, but “freeze” in metastable configurations at high temperatures, where  $R$  is still too short for  $J$  to significantly deviate from the pure dipole-dipole form. Present observation that the short-range polar order is already determined well above  $T_f$  is consistent with this possibility. To this end, it is interesting to note that recent calculations<sup>25</sup> have shown that metastable states formed by clusters of closely spaced dipoles can explain the dielectric dispersion above  $T_f$ .

In conclusion, we have shown that the polar state of lightly doped KTL is characterized by microscopic regions where the polarization is oriented along the pseudocubic axes. The preferential quadrupolar orientation appears rather casual, and not related to the mechanism leading to the polar state. The size of the polar correlated patches shows no signs of criticality, the correlation length remaining constant at  $\approx 8$  nm from 55 to 10 K. We suggest that the polar pattern which forms well above  $T_f$  is due to the interaction of the Li dipoles, and that, on cooling, this pattern acts as an imprint for the displacement of the Ta ions of the matrix.

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