

Li AND Nb CODOPING IN DIELECTRIC PROPERTIES AND PHASE TRANSITIONS OF  $\text{KTaO}_3$ : REENTRANT DIPOLAR GLASS AND LONG-RANGE DIPOLE ORDERING.

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The effect of  $\text{Li}^+$  and  $\text{Nb}^{5+}$  impurity off center ions substituting  $\text{K}^+$  and  $\text{Ta}^{5+}$  in the properties of quantum paraelectric  $\text{KTaO}_3$  (KTO) have been extensively studied and the main properties of  $\text{KTO:Li}$  (KLT) and  $\text{KTO:Nb}$  (KTN) have been established already (see e.g. [1-3]). For KLT, these are: a large dipolar  $\langle 100 \rangle$  displacement and slow relaxation of  $\text{Li}^+$ ,  $\text{TO}_1$  mode stiffness, aspiration to clusters and dipolar glass-like states formation and order-disorder-type ferroelectric phase transition (PT). For KTN - faint  $\langle 111 \rangle$  displacements and relaxation of  $\text{Nb}^{5+}$ , polarization enhancement and  $\text{TO}_1$  soft mode dominated displacement-type ferroelectric PT have been found. At the same time KTO doped by both Li and Nb,  $\text{K}_{1-x}\text{Li}_x\text{Ta}_{1-y}\text{Nb}_y\text{TaO}_3$  (KTLN), whose attractive application perspectives were shown recently [4], is studied very scarcely. Practically only in [5] using depolarisation and Raman scattering of light experiments it was found that relaxing symmetry-breaking  $\text{Li}^+$  defects determine the KTLN structure, alter the PT from the soft-mode to order-disorder cubic-tetragonal, increases the transition temperature and decrease the magnitude of the  $\epsilon'(T)_{\text{max}}$ .

We present detailed studies of  $\epsilon'(T, f)$  and  $\tan[\delta(T, f)]$  (10 - 300 K, 100 Hz - 1 MHz) for  $\text{KTLN:Cu}$  with  $x_1 = 0.0014$ ,  $y_1 = 0.024$  (KLTN 0.14/2.4) and  $x_2 = 0.0028$ ,  $y_2 = 0.013$  (KLTN 0.28/1.3). The most surprising properties were found for KLTN 0.14/2.4. Here together with an Arrhenius type relaxation of  $90^\circ$  flips of  $\text{Li}^+$  in the 40–80 K region, with  $\epsilon'(T)_{\text{max}} = 16\,000$  at 47 K (100 Hz), a ferroelectric PT, dominated by a soft  $\text{TO}_1$  mode coupled with a relaxation freezing mode, is found at 39 K. This unusual behavior for such a low concentration of  $\text{Li}^+$  centers is related with a Nb induced  $\text{TO}_1$  mode softness and respective dipolar correlation length  $r_c \sim (v\epsilon')^{1/2}$  increase. However the most striking is that at ~15.5 K another  $\epsilon'(T)$  cusp-shape maximum with  $\epsilon'(T)_{\text{max}} \approx 25\,000$  was found. It is the first observation of the presence of two phase transitions in KLTN and such a sequence of PT in high polarizable perovskites with dipolar off-center impurities. The results are discussed together with the slow dielectric relaxation response and a nonergodic behavior of KLTN 0.14/2.4 observed at low temperatures, Second Harmonic Generation of Nd:YAG laser light experiments and taking into account recent Raman scattering results [6]. The possibility of a low temperature PT into a “reentrant dipolar glass” state together with competition with a long-range dipole ordering in KLTN is suggested.

References:

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