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Short communication

Orientation dependence of dielectric and piezoelectric properties of $(K_{0.95}Li_{0.05})$ $(Ta_{0.40}Nb_{0.60})$ O_3 single crystal

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Abstract

$(K_{0.95}Li_{0.05})$ $(Ta_{0.40}Nb_{0.60})$ O_3 (abbreviated as 0.60: KLTN) single crystal has been grown by the top-seeded melt growth method. Dielectric properties in the *a*- and *c*-direction have been investigated, and the high anisotropy has been found. $\varepsilon_{11}=2130$, while $\varepsilon_{33}=371$ were determined in the unpoled sample. The piezoelectric constants and electromechanical coupling factors have been determined which are $d_{15}=190$ pC/N, $d_{31}=-140$ pC/N, $d_{33}=400$ pC/N, $k_{15}=49.4\%$, $k_{31}=56.2\%$ and $k_{33}=75.0\%$. The crystal orientation dependences of dielectric, piezoelectric and electromechanical coupling properties have been simulated by using the mathematical approach along with phenomenology of the system. These results will be of use for developing high performance lead-free piezoelectric devices.

Keywords: C. Dielectric properties; C. Piezoelectric properties; D. Perovskites

1. Introduction

$KTa_{1-x}Nb_xO_3$ (KTN) solid-solution crystals are well-known electro-optic materials, the crystals structure and properties change with the compositions. Below $x\approx 0.05$, KTN crystals are quantum ferroelectrics, and the ferroelectric transition temperature obeys $T_C=276(x-0.008)^{1/2}$ K [1,2]. When x is between 0.05 and 0.35, KTN crystals have weak diffuse transition characteristics. Above $x\approx$ is betthe compositions exhibit first-order ferroelectric transition and the Curie temperature is $T_C=676x+32$ K [3]. The particular compositions have excellent optical properties, such as linear or second-order electro-optic coefficients. However, the electric properties of KTN crystals were seldom noticed.

For the acoustic applications, the large piezoelectric constants and electromechanical coupling factors are desirable. In the single crystals, electrical properties could be improved by the crystal orientations [4–6]. Recently, we have studied the dielectric, pyroelectric, piezoelectric and ferroelectric properties of Nb-rich Li-doping KTN $(K_{0.95}Li_{0.05}Ta_{1-x}Nb_xO_3)$ crystals [7–10]. Their electric properties were enhanced by Li doping.

In this work, we have modeled the crystal orientation dependence of the dielectric, piezoelectric and electromechanical properties of $K_{0.95}Li_{0.05}Ta_{0.4}Nb_{0.6}O_3$ (0.60: KLTN) single crystal. These results are useful for developing novel piezoelectric devices, and extend the application scope of KTN system.

2. Experimental procedure

The top-seeded melt growth method has been used to grow 0.60: KLTN single crystal. High purity starting materials K_2CO_3, Li_2CO_3, Ta_2O_5 and Nb_2O_5 powders were prepared according to the composition selected in the phase diagram [11,12]. During the growth, the rotation rate of the seed rod was set to 10 rpm, and

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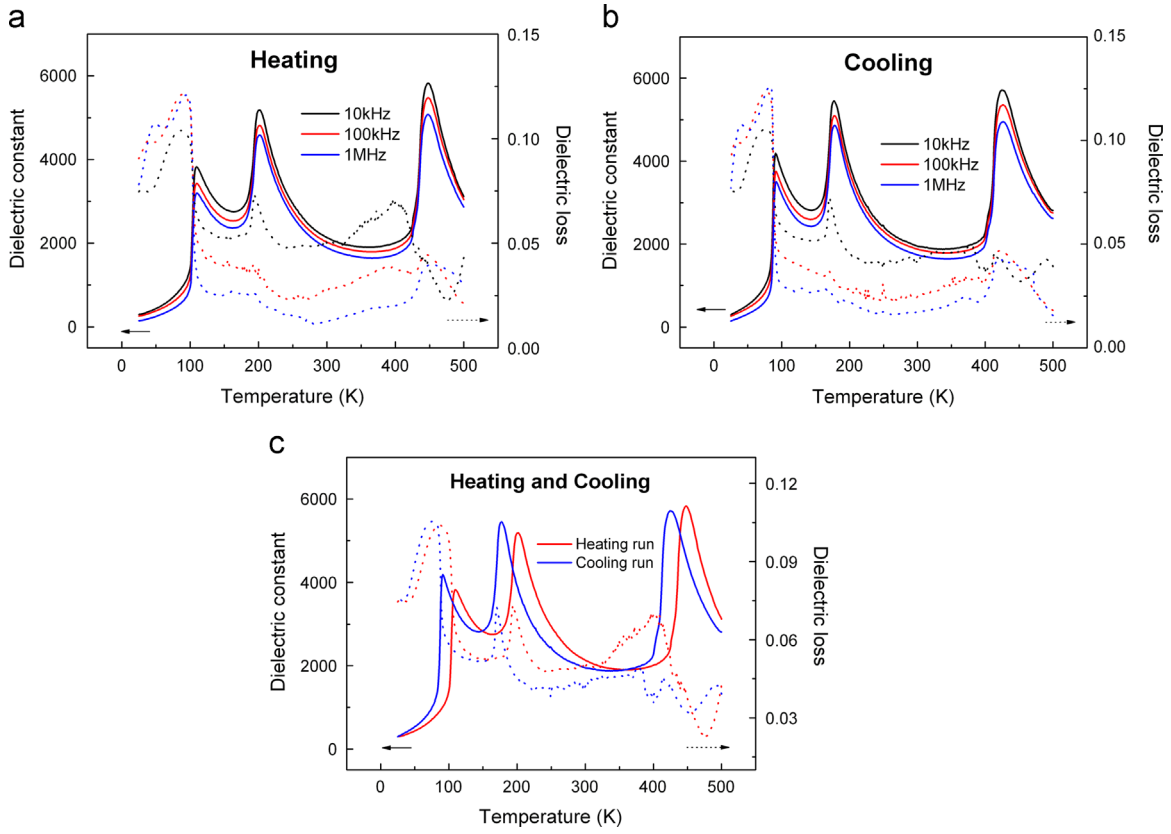

Fig. 1. Dielectric constant and loss of [100] crystallographic direction as a function of temperature at different frequencies: (a) dielectric constant and loss on heating, (b) dielectric constant and loss on cooling, and (c) dielectric constant and loss on heating and cooling runs at 10 kHz.

the pulling speed was 0.5 mm/h. After 6–7 days, high quality single crystals were obtained after entire growth run.

The density of the crystal was around 5.59 g/cm^3. After cutting, polishing, and stress releasing, the samples for measurements were prepared. The dielectric properties were measured as a function of temperature by using an HP4284 LCR meter at different frequencies along the [001] and [100] crystallographic directions. The piezoelectric coefficients were calculated from the resonance and anti-resonance frequencies, which were measured by an HP 4194A impedance/gain phase analyzer.

Table 1
Dielectric constant, loss tangent and phase transition temperatures of [100] and [001] oriented 0.60: KLTN single crystal.

State	T_{R-O} (K)	T_{O-T} (K)	T_C (K)	ε_{rmax}	tan δ_{max}	ε_{rRT}	tan δ_{RT}
[100] Direction							
Heating run	110	200	450	5830	0.10	2130	0.05
Cooling run	90	177	425	5710	0.10	1950	0.04
[001] Direction [4]							
Heating run	88	195	445	2484	0.06	371	0.03
Cooling run	82	188	437	2646	0.07	379	0.03

3. Results and discussion

3.1. Dielectric and piezoelectric measurements

Fig. 1 shows the dielectric constant and loss of [100] crystallographic direction dependence on temperature at different frequencies. The data are collected in the heating and cooling runs and shown in Fig. 1(a) and (b). The results of dielectric constants dependence of temperature reveal the three phase transitions: the transition temperature from rhombohedral to orthorhombic phase, $T_{R-O} \approx 110$ K; the transition temperature from orthorhombic to tetragonal phase, $T_{O-T} \approx 200$ K; and the Curie temperature $T_C \approx$; a K represents transformation from tetragonal to cubic phase, in the heating run at 10 kHz. At low temperature (below 100 K), the values of ε_{11} are very small, while the loss could reach to around 0.10. The anomalies in the dielectric constants at various transition points, e.g. from orthorhombic to tetragonal and finally to cubic phases are evident in the ε vs. T plots. The values of dielectric constant at room temperature are attractive device point of view. The temperature hysteresis between heating and cooling runs of $\Delta T_{R-O} \approx 20$ K, $\Delta T_{O-T} \approx 23$ K, and $\Delta T_C \approx 25$k, clearly suggest the first order phase transition behavior. Table 1 lists the summary of the results for T_{R-O}, T_{O-T}, T_C; the corresponding maximum and room temperature values of the dielectric constants and loss (ε_{rmax}, tan δ_{max}) at 10 kHz.

Fig. 2 shows the dielectric constant and loss along the [100] and [001] crystallographic directions. Comparing the phase transition points of these two crystallographic directions, some drift at the phase transition peaks can also be noticed. In both heating and cooling runs, the values of ε_{11} are more than 5 times of the ε_{33}. The typical value of ε_{11} at room temperature is ~2130, and the maximum in whole temperature range is about 5800 (at the T_C). For