Dielectric properties of $K(Ta_{0,51}bN_{0,49})O_3$ Single Crystal

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***Abstract* — Dielectric properties of $K(Ta_{0,51}Nb_{0,49})O_3$ [KTN] single crystals with [100], [001] and [011] crystallographic orientation were studied. Temperature dependencies of dielectric permittivity of monodomain and polydomain crystals of these orientations are compared.**

Keywords - component; dielectric permittivity; single crystal; phase transition; domain structure

I. INTRODUCTION

The enhancement of properties of lead free ferroelectrics is a subject of a strong current interest [1]. One of the most intriguing approaches to obtain such enhancement involves the so called 'domain engineering' where a high density of domain wall structure is created under different mechanical and electrical conditions [2-3]. For these purposes, crystals are used in orientations which do not coincide with the polar direction. The present study was motivated by a desire to study dielectric properties of $K(Ta_{0,51}Nb_{0,49})O_3$ or KTN single crystal with [011] orientation which is suitable for engineered domain configuration and to compare its properties with those in the directions of different crystallographic axes <100>. KTN is a solid solution of $KNbO_3$ and $KTaO_3$ constituting a mixed-crystal series exhibiting the classical phase transition sequence [4]: cubic — tetragonal — orthorhombic — rombohedral phases. Also it is one of the perovskite-type ferroelectric oxides that have tetragonal phase at room temperature. High-quality KTN crystals have very large electro-optic effects at room temperature [4]. The phase transition temperatures can be modified by changing the concentration of $KTaO_3$ in the solid solution, facilitating the selection of required temperature region. To the best of the authors' knowledge KTN dielectric properties have not been reported in details.

In present work polydomain and monodomain crystals were studied. Monodomain samples were prepared by poling below phase transition temperature.

II. EXPEERIMENTAL PROCEDURE

KTN crystals were grown by top seeded solution growth. Samples were cut and polished into 4,5×4,5×0,5 plates, with the main side perpendicular to [011], [001] and [100] that is equivalent [010] directions of the pseudocubic lattice. [100] oriented samples were prepared from [001] oriented monodomain crystal plate by cutting perpendicular to the direction [100]. Than [100] oriented samples were shaped and polished into 4,5×0,5×0.2 lamellas. The main sides of all samples were then fully sputter coated with Pt 100 nm in thickness to form a capacitor. Polarizing microscope was also used for observation of samples' domain structure and verifying the quality of their monodomain state.

For monodomain crystals samples were poled with electric field 5 kV/cm below temperature of phase transition (tetragonal-cubic) but in its vicinity.

The dielectric response was measured as a function of temperature, frequency and amplitude of the ac electrical signal using a computer controlled setup consisting of a temperature chamber Delta 9023 with multimeter HP 34401A for temperature measurements and a precision LCR-meter HP 4284A.

At room temperature the domain structure of the polydomain crystals consisted of 180-degree and 90-degree domains corresponding to tetragonal phase (fig. 1).

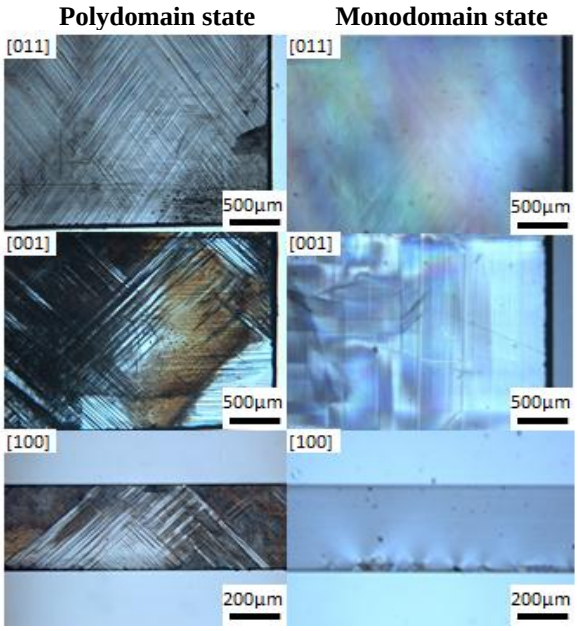

Fig. 1. Domain structure of [011], [001] and [100] oriented samples.

III. Results And Discussion

Dielectric properties of every type of samples were examined in the cubic tetragonal orthorhombic and rhombohedral phases in temperature range between -100°C and 180°C on heating and on cooling. Temperature rate was kept 0,5 K/min. Dielectric permittivity was measured at ac voltage 1 V for 4 different frequencies: 100 Hz, 1 kHz, 10 kHz and 100 kHz. For these frequencies no dispersion was observed. Temperatures of phase transitions for both orientations of crystal were the same. From rhombohedral to orthorhombic phase, phase transition temperature was $T_{r\text{-}o}$ = -69°C with temperature hysteresis 8 K. From orthorhombic to tetragonal phase, phase transition temperature was $T_{o\text{-}t}$ = 11°C with temperature hysteresis 12 K. From tetragonal to cubic phase it was $T_{t\text{-}c}$ = 92°C with temperature hysteresis 4 K. The temperature dependencies of dielectric permittivity during heating and during cooling for samples oriented along ferroelectric axes [001] are presented on fig. 2.

The temperature dependency of dielectric permittivity and dielectric loss tangent was measured for both polydomain and monodomain crystals. The most stable values were observed in temperature region between − 60°C and 180°C (fig.3,4)

It was found that for polydomain state in [011], [001] and [100] (that is equivalent [010]) directions at room temperature the values of effective dielectric permittivity were around 3400, 2100 and 3500 correspondingly.

Monodomain state was induced after poling below phase transition temperature between tetragonal and cubic phases. To prevent destroying of monodomain state during heating/cooling samples were cooled to 5°C – temperature that is above but in the immediate vicinity of phase transition temperature between orthorhombic and tetragonal phases. After that the temperature dependency of dielectric permittivity was measured during heating until 80°C. Dielectric permittivity for [011] in monodomain state was 10% lower than for polydomain state because of the absence of domain structure influence. For [001] oriented samples the value of dielectric permittivity at room temperature for monodomain state was around 350 that is 10 times lower than the value for polydomain state. For [100] oriented samples the value of dielectric permittivity at room temperature for monodomain state was around 6000, that is more than two times higher value than the value for polydomain state.

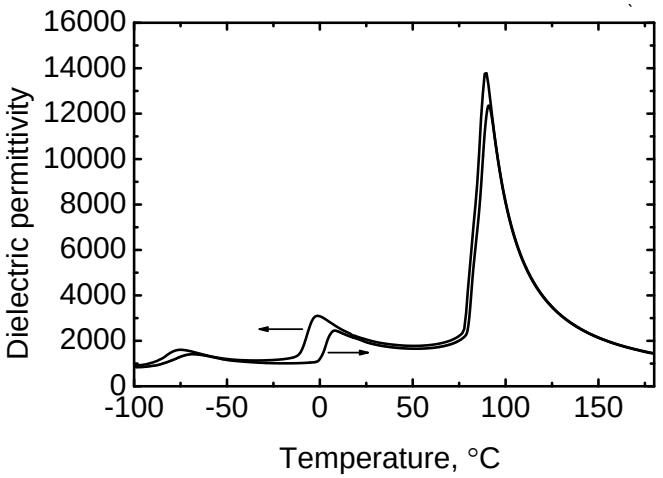

Fig. 2. Temperature dependence of dielectric permittivity measured in ferroelectric direction [001].

The calculation of dielectric permittivity for monodomain state showed that obtained values related to the equation

$$\varepsilon^{[100]} = \varepsilon^{[010]} = 2\varepsilon^{[011]} - \varepsilon^{[001]}.$$

This relation follows from the rotation of the third rank tensor for tetragonal phase. The calculated values of ε^{100} are presented on fig. 5. Difference between measured and calculated values is lower than 5 %. This result shows that the quality of induced monodomain state by poling was high and there is no domain contribution to measured values of dielectric permittivity.

IV. Conclusion

Dielectric properties of KTN single crystals of [001], [100] and [011] orientations were studied. Strong influence of poling on dielectric permittivity of [001] oriented crystal was found, with ten times reduction in dielectric permittivity upon poling. At the same time for [100] oriented monodomain crystal dielectric permittivity was more than two times higher than the

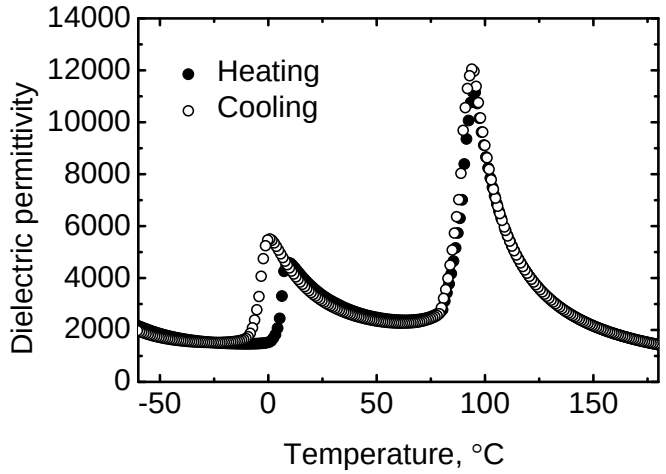

Fig. 3. Temperature dependence of dielectric permittivity measured in [011] direction (polydomain state).

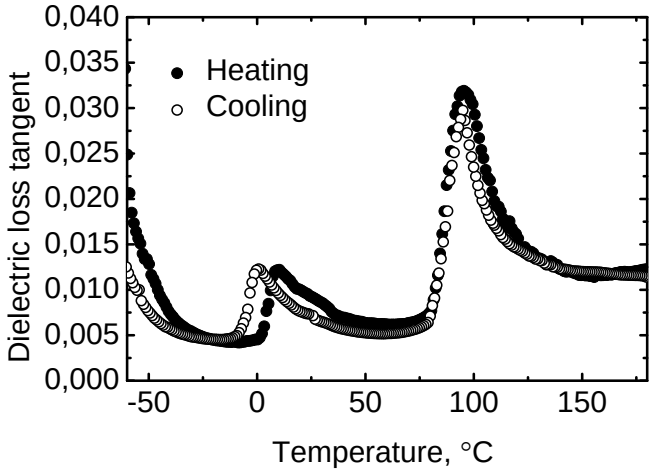

Fig. 4. Temperature dependence of dielectric loss tangent measured in [011] direction (polydomain state).

value for polydomain state. Measured temperatures of phase transitions and high values of dielectric permittivity in [011] direction make this material perspective for usage in tetragonal phase for domain engineering purposes in temperature range from 10 to 80 °C.

V. Acknowledgements

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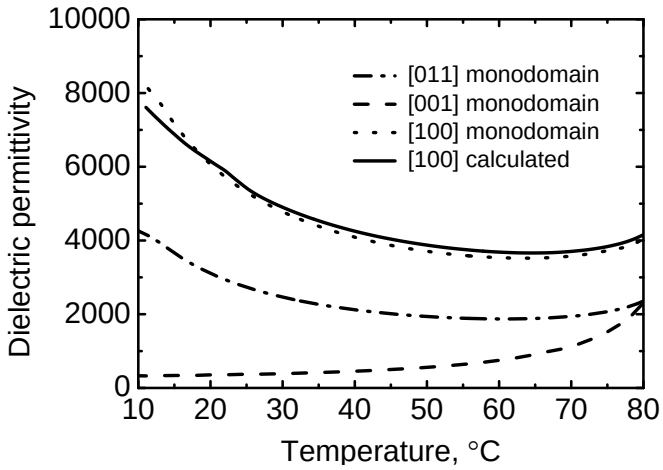

Fig. 5. Temperature dependence of dielectric permittivity measured in [100], [001] and in [011] directions for monodomain state during heating.

VI. References

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