Dielectric Properties of a $K_3Li_{1.88}Nb_{5.12}O_{15.24}$ Crystal from 90 to 300 K

V. G. Gurtovoi, A. U. Sheleg, and S. A. Guretskii
Joint Institute of Solid-State and Semiconductor Physics, Belarussian Academy of Sciences,
ul. Brovki 19, Minsk, 220072 Belarus

e-mail: hurtavy@ifttp.bas-net.by
Received May 25.2006

Abstract—The electrical conductivity, dielectric permittivity, and loss tangent of a $K_3Li_{1.88}Nb_{5.12}O_{15.24}$ crystal have been measured at temperatures from 90 to 300 K and frequencies of 0.1, 1, 10, and $10^3$ kHz. The results demonstrate that the dielectric permittivity of the crystal increases with increasing temperature and drops with increasing frequency. The plots of $\tan \delta$ versus temperature show maxima characteristic of semiconductors. The conductivity of the crystal increases by several orders of magnitude with increasing frequency. The conductivity and dielectric properties of the $K_3Li_{1.88}Nb_{5.12}O_{15.24}$ crystal are shown to be anisotropic.

DOI: 10.1134/S0020168507050184

INTRODUCTION

$K_3Li_{2-x}Nb_{5+x}O_{15+2x}$ (KLN) crystals are potentially attractive materials for piezoelectric, nonlinear optical, and electron-optical components owing to their broad transmission window, high optical damage threshold, and good nonlinear optical and electro-optic properties. Below their ferroelectric transition temperature, KLN crystals have a tetragonal structure with room-temperature lattice parameters $a = 12.5764 \pm 0.0002$ Å and $c = 4.0149 \pm 0.0001$ Å [1].

Note that stoichiometric KLN crystals are unstable. Stable KLN crystals can only be grown if the melt contains Nb in excess of the stoichiometric content. Varying the melt composition, Li et al. [2] grew KLN crystals with lattice parameters in the ranges $a = 12.49$–12.60 Å and $c = 4.01$–4.06 Å.

The dielectric properties of KLN were investigated in [3, 4]. Kim and Lee [3] studied the effect of chemical composition on the dielectric permittivity of KLN near its phase transition. Kang and Yoon [4] measured the dielectric permittivity of KLN in one direction at temperatures from 273 to 820 K and frequencies of 10, 50 and 100 kHz. They, however, did not specify the exact chemical composition of their crystals.

In this paper, we report the electrical conductivity and dielectric properties of a $K_3Li_{1.88}Nb_{5.12}O_{15.24}$ crystal in two different crystallographic directions at temperatures from 90 to 300 K and frequencies of 0.1, 1, 10, and $10^3$ kHz. The lithium content of the crystal was determined by measuring its $90^\circ$ phase matching wavelength at room temperature.

EXPERIMENTAL

The electrical conductivity $\sigma$, dielectric permittivity $\varepsilon$, and loss tangent $\tan \delta$ of the KLN crystal were measured with an E7-12 digital multimeter at a frequency of 1 MHz and with an E7-14 multimeter at frequencies of 0.1, 1, and 10 kHz. Temperature-dependent $\sigma$, $\varepsilon$, and $\tan \delta$ data were obtained during quasi-steady-state continuous heating at a rate of about 0.5 K/min. The sample was mounted in a purpose-designed holder, which was then immersed in liquid-nitrogen vapor.

The samples, in the form of single-crystal KLN plates about 1 to 2 mm in thickness, with (001) or (110) surfaces, were cut from Czochralski-grown single-crystal boules. The cut surface was oriented parallel to the (001) plane to within 5–8' using x-ray diffraction. The (110) surface was a natural growth surface.

The sample temperature was monitored with a Chromel–Copel thermocouple, whose junction was situated on the sample surface. The temperature was controlled by a temperature controller using a heater embedded in the sample holder. Electrical contacts were made with silver paste.

RESULTS AND DISCUSSION

Figure 1 shows the temperature dependences of $\varepsilon$ and $\tan \delta$ in the [110] direction for the KLN single crystal at temperatures from 90 to 300 K and different frequencies. As seen in Fig. 1a, the dielectric permittivity of KLN varies insignificantly with temperature in the range 90–200 K, at a level $\varepsilon_1 = 270$ at frequencies from $10^2$ to $10^4$ Hz and $\varepsilon_2 = 240$ at $10^6$ Hz. In the temperature range ~200–270 K, the dielectric permittivity
The temperature dependences of $\tan\delta$ (Fig. 1b) show maxima typical of semiconductors. The maxima are located in the temperature range where the dielectric permittivity rises steeply. At low temperatures (from liquid-nitrogen temperature to 150 K), $\tan\delta$ is close to zero, independent of frequency. As the temperature is raised from 150 to 250 K, $\tan\delta$ increases sharply; in the temperature range 250–280 K, $\tan\delta$ drops. Above 280 K, $\tan\delta$ increases again owing to conduction losses. As seen in Fig. 1b, the maxima in $\tan\delta(T)$ shift to higher temperatures with increasing frequency. This is associated with high-frequency relaxation processes, as is the reduction in $\varepsilon$ with increasing frequency.

Figure 2 shows the $\varepsilon(T)$ and $\tan\delta(T)$ curves for the KLN crystal in the [001] direction at temperatures from 100 to 300 K and different frequencies. The curves for the [001] direction are seen to be similar in shape to those for [110], but the sharp rise in $\varepsilon$ and $\tan\delta$ occurs at higher temperatures. The dielectric permittivity begins to grow at $T \sim 250$ K; near 300 K, $\varepsilon$ reaches...