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Single crystal growth of $KNbO_3$ and application to surface acoustic wave devices

Kazuhiko Yamanouchi[a,*], Yasuo Wagatsuma[b], Hiroyuki Odagawa[b], Yasuo Cho[b]

[a]*Department of Electronics, Tohoku Institute of Technology, 35-1 Yagiyama Kasumicho, Taihaku-ku, Sendai, 982-8577, Japan*
[b]*Research Institute of Electrical Communication, Tohoku University, Sendai, Japan*

Abstract

Potassium niobate ($KNbO_3$) has large piezoelectric constants. The surface acoustic wave (SAW) substrates with high couplings are very important for wide-band SAW filters and SAW devices. Therefore, the large size $KNbO_3$ single crystals are required for SAW device applications. In this paper, the techniques of a large size of $KNbO_3$ using the top seeded solution growth techniques (TSSG) with large size of crucible of 60 mmϕ platinum are described. The results show that $50\times50\times15$ mm^3 single crystals are obtained. Also, the poling techniques of $KNbO_3$ crystal are investigated by using the nonlinear scanning dielectric microscope. The propagation characteristics of SAW and piezoelectric leaky surface waves in $KNbO_3$ single crystal were investigated theoretically. The results show that the electro-mechanical coupling coefficient of the surface wave propagating along the *X*-axis of the rotated *Y*-cut plane is extremely large: $K^2=0.53$, compared to K^2 of 0.055 for $LiNbO_3$. The experimental results of the above $KNbO_3$ substrates agree with the theoretical ones. Also the $KNbO_3$ substrates showed experimentally the zero temperature coefficients of frequency (TCF) around 20 °C.

Keywords: Crystal growth; $KNbO_3$ single crystal; Surface acoustic wave (SAW); Super large K2; Wide band SAW filter

1. Growth of $KNbO_3$ crystals

Potassium niobate ($KNbO_3$) has been receiving much attention due to its excellent properties, such as a large electro-optic coefficient, a high nonlinear optical coefficient and excellent photo-refractive characteristics. Furthermore, $KNbO_3$ has large piezoelectric constants.

Top seeded solution growth (TSSG) techniques of single domain material have been applied for a $<110>$ seed and growth of relatively large size (100 g; thickness 20 mm) pure $KNbO_3$ crystals. However, larger size $KNbO_3$ single crystals for SAW applications are required.

$KNbO_3$ is an incongruently melting oxide and crystals have to be grown from a K_2O rich, non-stoichiometric high temperature solution. Fig. 1 shows the phase diagram of K_2O–Nb_2O_5 components. The single crystals are grown from the $KNbO_3$ + liquid phase.

Top-seeded solution growth procedure has been applied for large size of $KNbO_3$ growth. To increase the crystal size, we have set up new equipment housing a cylindrical Pt crucible for nearly 180 ml (diameter = 6 cm) of liquid phase. The main growth parameters are as follows: raw materials → K_2CO_3: Nb_2O_5 = 1.0: 1.05, seed orientation → [010], seed rotation → 10 rpm, crucible → Pt (60 mm diameter, 70 mm height), pulling speed → 0.2 mm/h.

The rate of temperature decrease during the growth and cooling process is very important, because the existence of two phase transitions while cooling to room temperature, as well as the incongruent melting behavior, are most likely limiting constraints to obtain specimens as large as possible for $LiNbO_3$. The temperature characteristics during the growth and cooling process are shown in Fig. 2. Fig. 3 shows the grown $KNbO_3$, crystal showing the size $50\times50\times15$ mm^3.

Also, the poling techniques of $KNbO_3$ crystal are investigated by using the nonlinear scanning dielectric microscope and the poling direction is determined and poling is performed under the applied electric field of 2 kV/cm in oil. The single domain crystals are obtained.

2. Propagation characteristics of SAW on $KNbO_3$

Campbell and Jones[1] discussed optimum crystal cuts and propagation directions for the effective coupling of $LiNbO_3$, which is based on the computation of the difference between a velocity of the surface wave in a free surface and that in a metallized surface. The investigation

* Corresponding author. Tel./fax: +81-22-229-1205.
E-mail address: yamasw@tohtech.ac.jp (K. Yamanouchi).

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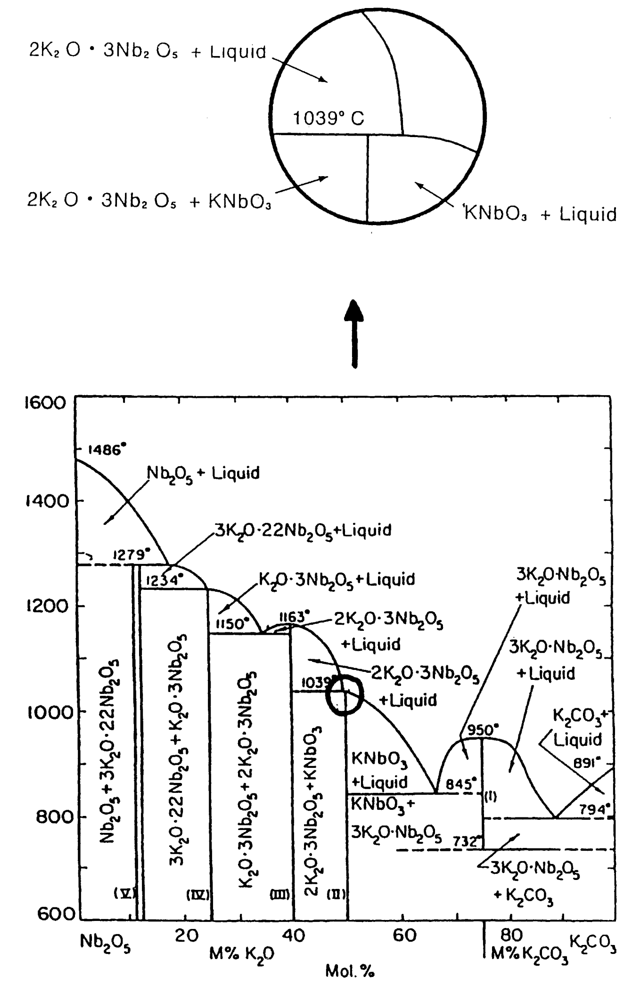

Fig. 1. Phase diagram of K_2O–Nb_2O_5.

of a piezoelectric leaky surface wave has been analyzed by Yamanouchi and Shibayama.[2]

Here we report the computation and experimental results for the surface waves and piezoelectric leaky surface waves in $KNbO_3$. The values of the elastic and piezoelectric constants of $KNbO_3$ were taken from the work of Zgonik et al.[3] We investigated the waves propagating along the *X*-axis of the rotated *Y*-cut plane and other cuts in $KNbO_3$.

The results show that the electro-mechanical coupling coefficient of the surface wave propagating along the *X*-axis of the rotated *Y*-cut plane is very large, $K^2=0.53$, compared to K^2 of 0.055 for $LiNbO_3$. Also, the $KNbO_3$ substrates show the zero temperature coefficients of frequency (TCF) around 20 °C.

2.1. Theoretical analysis and experimental results of $KNbO_3$ single crystal substrate

$KNbO_3$ has large piezoelectric constants. Therefore, surface waves with very large electro-mechanical coupling coefficients are expected.

In this section, we derive fundamental theoretical equations for leaky surface waves and elastic surface waves and present calculated results.

The coordinate system employed is shown in Fig. 4. In this figure, X_1 is taken in the direction for propagation, X_3 in the direction transverse to the propagation, and the elastic surface waves decay exponentially in the X_2 direction inside the piezoelectric infinite plate. The displacement u_i ($i=1$, 2, 3) and electric potential ϕ are given as follows:

$$u_i=\sum_{n=1}^{4}A^{(n)}\alpha_i^{(n)}e^{\Omega(n)\kappa X_3+j\kappa(lX_1-vt)} \tag{1}$$

$$\phi=\sum_{n=1}^{4}A^{(n)}\alpha_4^{(n)}e^{\Omega(n)\kappa X_3+j\kappa(lX_1-vt)} \tag{2}$$

where $l=1+\ j\sigma$(σ is decay constant/wavelength).

For the normal surface waves, $\Omega^{(n)}$ become in decay constants in the direction of the depth, but for the leaky surface waves, one of the $\Omega^{(n)}$ represents the growing constant in the direction of the depth.

The equation relating $\Omega^{(n)}$ and ν is found by substituting Eqs. (1) and (2) into the equation of motion, the piezoelectric equations and Maxwell's equations, giving

$$F\left(\Omega^{(n)},\nu\right)=0. \tag{3}$$

The boundary conditions for the elastic quantities are

$$T_{3j}|_{X_3=0}=0 \tag{4}$$

and the boundary condition for the electric quantities,

$$\phi|_{X_3=0}=0\ \text{for metallized surface,} \tag{5a}$$

$$D_3|_{X_3=0}=-\varepsilon_0\frac{\partial\phi^e}{\partial X_3}|_{X_3=0}\ \text{for free surface,} \tag{5b}$$

where ϕ^e is the electric potential of the free space ($X_2>0$) and D_2 the electric displacement in the X_2-direction.

The boundary condition equation is given from Eqs. (4) and (5):

$$G\left(\Omega^{(n)},\nu\right)=0. \tag{6}$$

Successive values of the phase velocity ν are tried until the function $G\left(\Omega^{(n)},\nu\right)$ is made equal to zero within some pre-determined accuracy, the $\Omega^{(n)}$ being determined from Eq. (3). Fig. 5 shows the electrode pattern

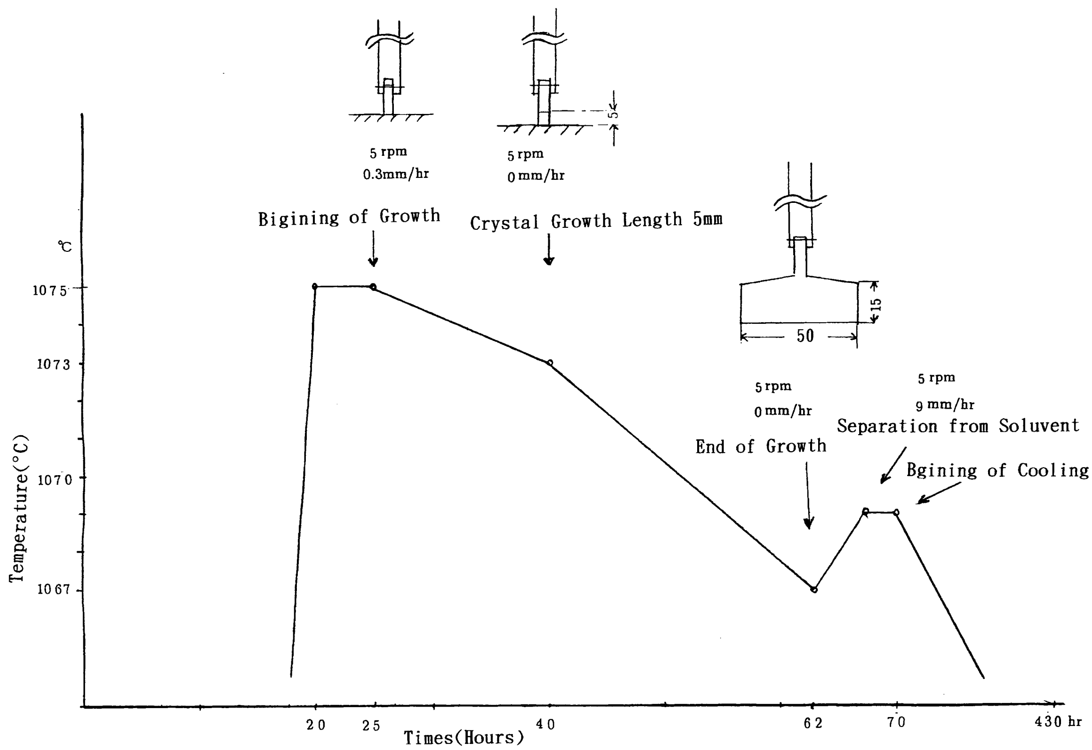

Fig. 2. Temperature cycle from crystal growth to room temperature at cooling process.

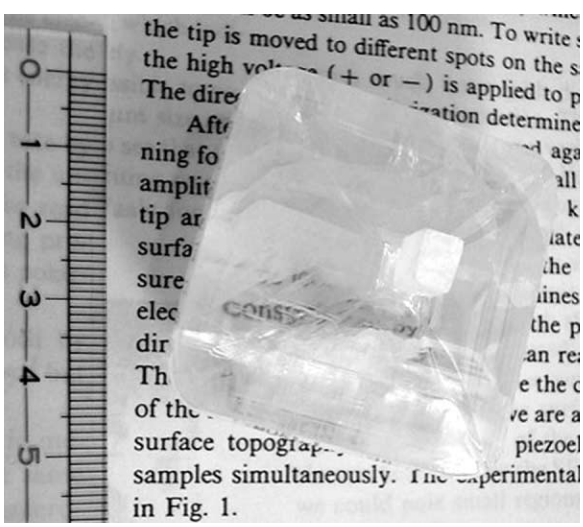

Fig. 3. Grown $KNbO_3$ single crystal with showing size (50×50×15 mm^2).

for measurements of SAW velocities. The velocities of open and short surface, v_f and v_m are given by

$$v_f = (L_2 - L_1)/(t_{BC} - t_{AB}) \quad (7)$$

$$v_m = L_3/\left(L_3/v_f + t_{CD} - t_{BC}\right) \quad (8)$$

where t_{AB}, t_{BC} and t_{CD} are delay times as shown in Fig. 5. The experimental wavelength is 20 μm and Al thickness is 0.4 μm.

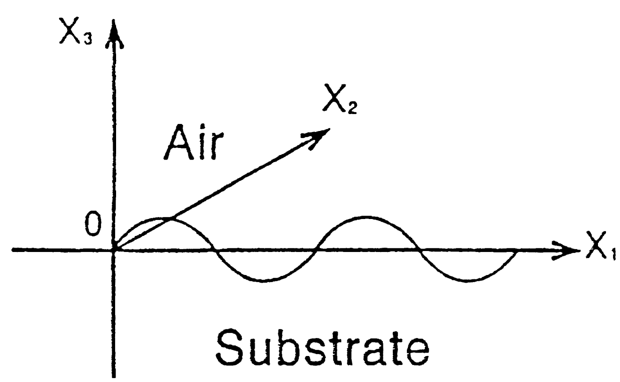

Fig. 4. Coordinate system used for calculation.

Fig. 6 shows curves of velocities versus rotating angle for a longitudinal wave, two transverse waves and surface acoustic wave. In this figure the solid line indicates the velocities of the surface wave in the free surface adjoining the vacuum and the dotted line the velocities in the metallized surface.

If the former is denoted by v_f and the latter by v_m, the effective electro-mechanical coupling coefficient can be expressed as $K^2 = 2\,(v_f - v_m)/V_m$. Fig. 7 shows the K^2 of a SAW. For the surface acoustic wave the K^2 has a maximum value at about 0° and a minimum at 90°. The maximum value of K^2 is about 0.53. This value is approximately 10 times larger than the value of 0.055 for $LiNbO_3$. The experimental results obtained by measuring v_f and v_m are plotted in Figs. 6 and 7 (○

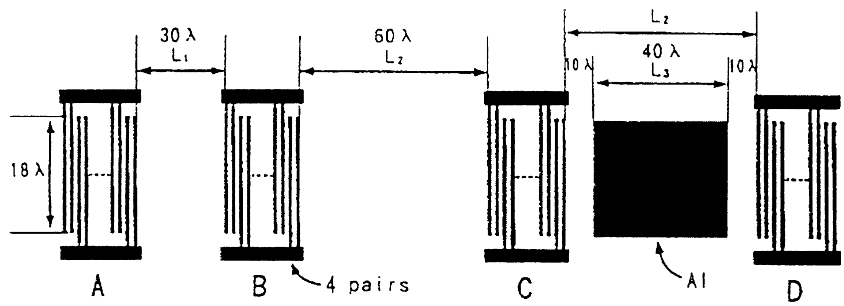

Fig. 5. Electrode pattern for measuring of SAW velocity.

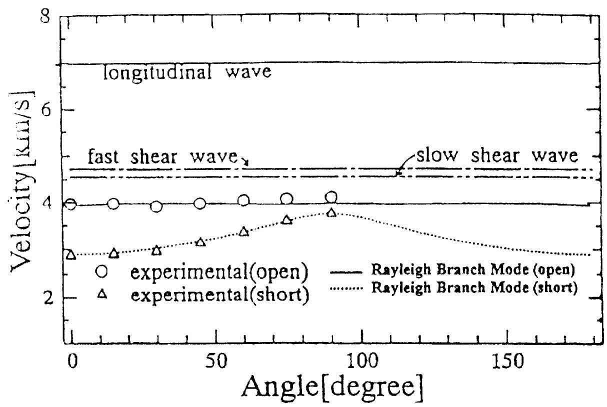

Fig. 6. Velocities of bulk and surface waves for rotated *Y*-cut, *X*-propagating $KNbO_3$ versus rotation angle.

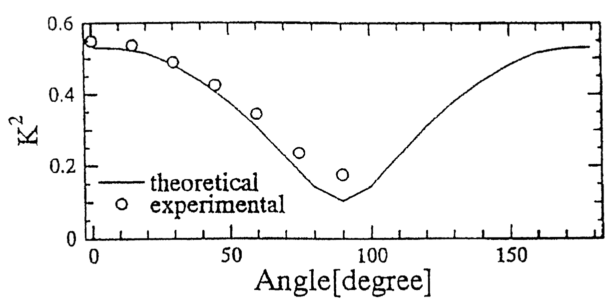

Fig. 7. Electromechanical coupling K^2 versus angle rotated *Y*-cut, *X*-propagating $KNbO_3$.

and Δ). The experimental and theoretical values are in good agreement.

Fig. 8 shows curves of velocities on the *Y*-cut plate versus the propagation angle for longitudinal wave, two transverse waves and surface acoustic wave. In this figure the solid line indicates the velocities of the surface wave in the free surface adjoining the vacuum and the dotted line indicates the velocities in the metallized surface. The leaky surface waves are observed near the velocity of 4900 m/s in calculation.

The frequency response on 45° *Y–X* $KNbO_3$ for conventional interdigital transducers with pair number (*N*) of 4 and aperture of 18 λ (λ: the wavelength of SAW) is

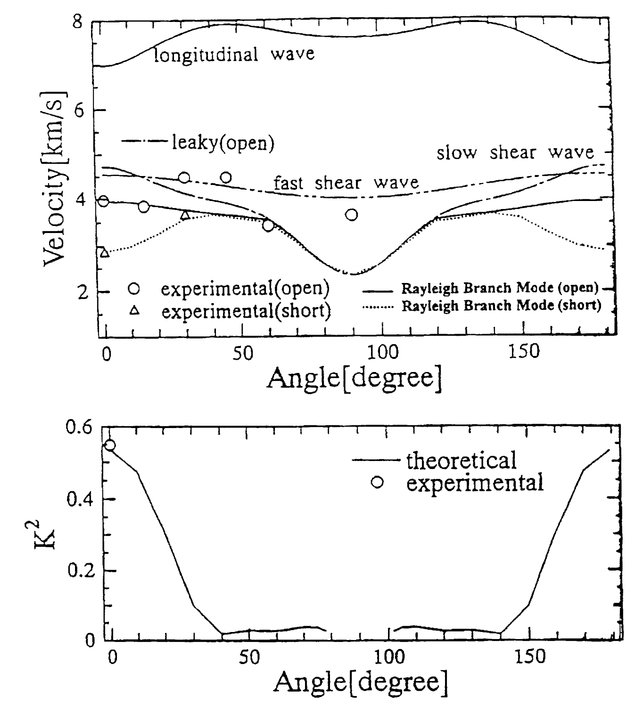

Fig. 8. Propagation characteristics on *Y*-cut $KNbO_3$ plate.

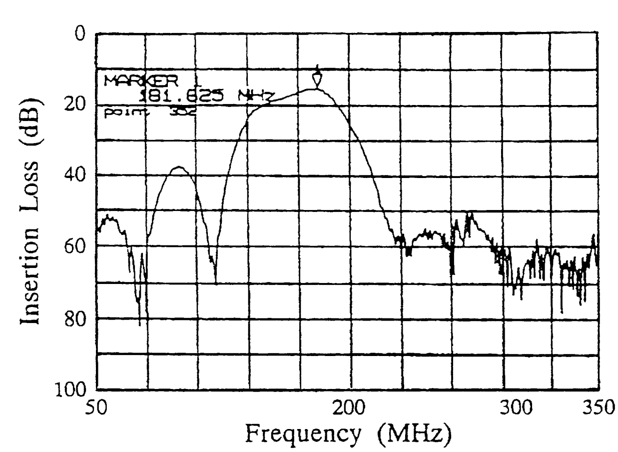

Fig. 9. Frequency response of two IDTs with $N=4$ finger pairs and aperture of 18 λ on 45° *Y–X* $KNbO_3$.

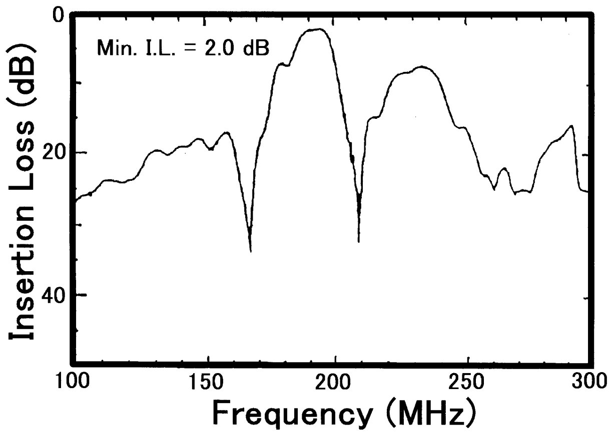

Fig. 10. Frequency response of FEUDT with $N=8$ finger pairs and aperture of 18 λ on 60° Y–X $KNbO_3$.

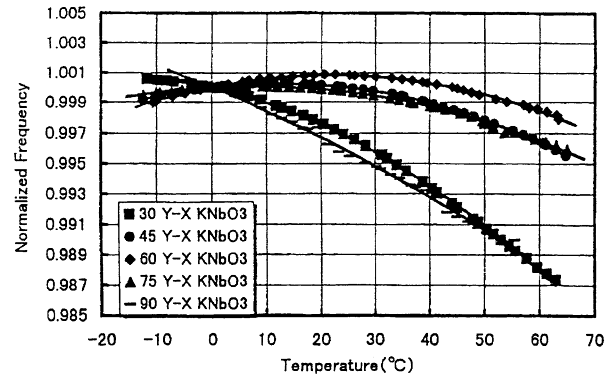

Fig. 11. Frequency versus temperature behavior of selected $KNbO_3$ substrates.

shown in Fig. 9. The radiation conductance is about 5 ms, corresponding to a K^2 value of 0.4.

The frequency response on 60° Y–X $KNbO_3$ for the floating electrode type unidirectional transducer (FEUDT) with pair number of 8 and 18 λ is shown in Fig. 10.

The insertion loss of 2.0 dB with the directivity of 10 dB under the matching circuit is obtained. The band width is about 20%.

2.2. Temperature properties

The temperature coefficient of frequency (TCF) is given by the following equation

$$\mathrm{TCF} = -\frac{\mathrm{d}\tau}{\mathrm{d}T} = \frac{\mathrm{d}f}{\mathrm{d}T} = -\frac{1}{l}\cdot\frac{\mathrm{d}l}{\mathrm{d}T} + \frac{1}{v}\cdot\frac{\mathrm{d}v}{\mathrm{d}T} \tag{9}$$

where τ is the delay time; T is the temperature, l is the propagation distance and f is the frequency.

Fig. 11 shows the experimental results of temperature characteristics. TCF response shows the parabolic characteristics. The 45° Y–X and 60° Y–X $KNbO_3$ substrates have the zero TCF around 20 °C, as shown in Fig. 11.

3. Conclusion

We studied the growth of $KNbO_3$ and the propagation characteristics of SAW in $KNbO_3$ single crystals, theoretically and experimentally. We obtained super-high coupling of SAW with a K^2 value of 0.53. We are now investigating very wide-band and low-loss filters using unidirectional IDTs and GHz-range SAW devices.

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