Characterization of LiNbO3 crystals by line-focus-beam acoustic microscopy

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Characterization of $LiNbO_3$ crystals by line-focus-beam acoustic microscopy

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Line-focus-beam (LFB) acoustic microscopy is applied to quantitative characterization of piezoelectric $LiNbO_3$ crystals to demonstrate the usefulness of this new analytical technique. Experimental relations between chemical composition ratios of Li/Nb and leaky surface acoustic wave (LSAW) velocities for 128°*YX* $LiNbO_3$ wafers are determined. LSAW velocity measurements are carried out for commercial wafers obtained from a series of crystal growths. Small changes of 0.092% are detected due to the compositional variation. It is estimated that the "effective" congruent composition in the production line is 48.440 Li_2O mol% with the density of 4647.4 kg/m^3.

Piezoelectric single crystals of $LiNbO_3$ and $LiTaO_3$ have been widely used as substrates for surface acoustic wave (SAW) devices such as filters and resonators. Their characteristics relate directly to the elastic properties of wafers of their materials, especially the SAW velocity. It is therefore most important to use homogeneous wafers of the same quality in SAW properties for mass production of SAW devices. Studies have been conducted on crystal growth conditions and wafer fabrication processes in order to obtain high quality wafers.[1-4] Until recently, only the method of interdigital electrodes formed on wafers has been available to evaluate the elastic properties.

However, line-focus-beam (LFB) acoustic microscopy has recently become available[5] to provide a new technology applicable to quantitative material characterization in scientific and industrial fields.[6] Characterization is made by determining the propagation characteristics, viz., velocity and attenuation of leaky surface acoustic waves (LSAWs) on the boundary between a sample and the water coupling liquid, through the $V(z)$ curve analysis.[5] This method is most useful for this application since direct evaluation of the elastic properties along the desired wave propagation direction can be made nondestructively in small areas at chosen positions with higher measurement accuracy.

Although there are many kinds of related scientific and industrial problems to be resolved and inspected by this method, it is of fundamental importance to discuss the elastic variations not only within a single wafer, but also among wafers and crystal lots, which are associated with chemical composition.

In this letter we describe the application of a LFB acoustic microscope system to investigate the elastic properties of 127.86° rotated *Y*-cut *X*-propagating (128°*YX*) $LiNbO_3$ wafers, with special concern for the compositional variations occurring in the industrial production process. The most recent system,[7] operating at a frequency of 225 MHz with a relative velocity measurement accuracy better than ±0.005% at a point and ±0.02% over a scanning area of 75 mm×75 mm, was used for the measurements of the LSAW velocities.

It is first necessary to determine experimentally the relations between chemical composition ratios of Li/Nb and LSAW velocities. Four kinds of 128°*Y*-cut $LiNbO_3$ crystals of 2-in.-diam grown with the different compositions of the starting materials of 47, 48, 49, and 50 Li_2O mol%, which were studied previously,[4] were used for the measurements. Two wafers were taken from the top (*T*) and bottom (*B*) of each boule. The LSAW velocities were measured for eight samples, each being 0.5 mm thick. The velocities measured at the center of each wafer are chosen for discussion. The densities of the samples were also determined, which must be considered as average values for the wafers because compositional variations occur in a wafer along radial and thickness directions. Figure 1 shows the measured LSAW velocities, as a function of the wave propagation direction, together with the calculated values using the physical constants reported by Warner *et al.*[8] Clearly different characteristics were obtained due to the different compositions. The elastic properties in the *X*-axis (0°) direction are of greatest interest in the 128°*Y*-cut $LiNbO_3$ wafers employed for SAW devices in Fig. 1.[9] Figure 2 shows the experimental results of the relations among the LSAW velocities, densities, and chemical compositions for 128°*YX* $LiNbO_3$ wafers. In the figure, it is also shown that the wafer compositions[4] evaluated by the measurements of the Curie temperatures are different from those of the starting materials and depend on the wafer position. It is very clear that both the LSAW velocity and density strongly depend on the composition. As the Li_2O content increases, the LSAW velocity increases linearly with the rate of 37.16 m/s/mol% around the congruent composition and the density decreases with the rate of 7.25 kg/m^3/mol%. The sensitivity of the measurement system to the compositional change is 0.02691 mol%/m/s. The resolution is better than 0.005 mol%, corresponding to 0.2 °C in the Curie temperature. It is seen from Fig. 2 that the congruent composition to produce high quality $LiNbO_3$ wafers with homogeneous elastic properties in a crystal ingot is around 48.45 Li_2O mol%, with a density of about 4647 kg/m^3.

In order to study the quality of 128°*YX* $LiNbO_3$ wafers, a series of four crystals of about 110 mm length for 3 in. wafers were pulled from the melt, recharged by the starting material of the same composition in the same cru-

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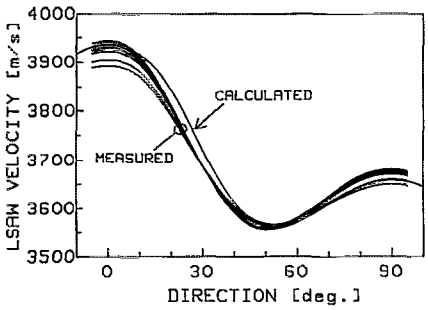

FIG. 1. LSAW velocities for 128°*Y*-cut $LiNbO_3$ wafers with different compositions.

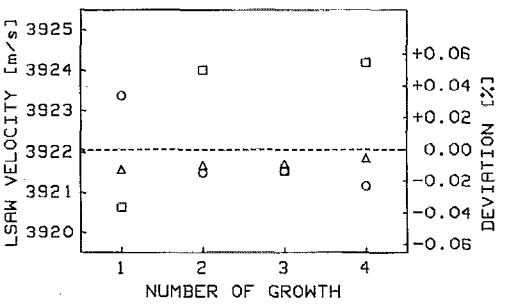

FIG. 3. Variations of LSAW velocities measured for 128°*YX* $LiNbO_3$ wafers taken from a series of four crystals in the production line. Wafer positions at 5, 55, and 105 mm are indicated with ○, □, and △, respectively.

cible of the crystal growth system under the same conditions for growth, according to the normal schedule of the production line. Such multiple growths are part of the normal process from the point of view of reducing the production cost. Three wafers 0.5 mm thick were obtained at the wafer positions of 5, 55, and 105 mm from the crystal head of each boule. Measurements were made at the center of 12 wafers. The results are shown in Fig. 3, where it is seen that slight variations are found. It can be considered that the variations were caused by subtle changes of chemical and thermal conditions in the melt during the pulling processes. The average value of the LSAW velocity is 3922.07 m/s, and the maximum difference is 3.60 m/s corresponding to a maximum deviation of 0.092%. Referring to Fig. 2, the chemical composition is estimated to be 48.440 Li_2O mol% with variation of 0.097 mol%. The average density measured for all the wafers is 4647.4 kg/m^3 and the maximum difference is 1.6 kg/m^3. The value is reasonable for the congruent composition as predicted in Fig. 2, but it cannot always explain the variations of LSAW velocities measured locally at the center. From the above investigation, it has been shown that the "effective" congruent composition is 48.440 Li_2O mol% with a density of 4647.4 kg/m^3 in the production line. The composition is very close to that of 48.45 Li_2O mol% reported by O'Bryan *et al.*[3] A similar investigation has been performed for *X*-112°*Y* (*X*-cut 112.2° rotated *Y*-propagating) $LiTaO_3$ wafers obtained from a series of seven crystals. The maximum change of LSAW velocities is 0.031%, which is smaller than that for the 128°*YX* $LiNbO_3$ wafers. It seems that this means different characteristics in elastic quality between $LiNbO_3$ and $LiTaO_3$ crystals. Detailed discussions will be reported elsewhere.[10]

The usefulness of the LFB system has been successfully demonstrated showing that chemical composition changes, which result in local density changes, can be quantitatively determined as elastic property changes through LSAW velocities.

The present system, with the distinctive features described above, is extremely useful not only for selection of wafers and control of the production line, but also for research and development studies of new materials for SAW devices. The application is easily extended to development and evaluation of MgO-doped $LiNbO_3$ crystals[11,12] for integrated optoelectronic devices, because elastic quality of the crystals related to chemical composition, including the MgO dopants, is primary to the optical quality.

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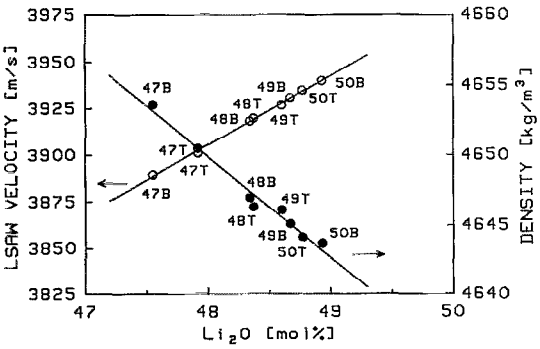

FIG. 2. Experimental relations among LSAW velocities, composition ratios of Li/Nb, and densities of 128°*YX* $LiNbO_3$ wafers.

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