Crystal Growth of Lithium Niobate and Potassium Lithium Niobate by μ-PD Method

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The recent developments of nonlinear optical materials are important steps toward applications in the field of blue and green second harmonic generation (SHG). Usually, single crystals used in optoelectronics are grown to large sizes because of the cost of material. However, high-quality alternative crystals have not yet been produced by conventional growth techniques. In contrast, in micro single crystals which are just device size with diameters below 1 mm, high-quality crystals with low defect density and composition homogeneity can be expected to produce by development a new growth process. Among these, $LiNbO_3$ (LN) and $K_3Li_{2-x}Nb_{5+x}O_{15+2x}$ (KLN) single crystals have considerable interest in the miniaturization and high efficiency of the devices because of the extended interaction length and high optical intensity. In this paper the growth of micro single crystals are described by the micro pulling down (μ-PD) method, even though KLN is difficult to grow due to the formation of cracks and the composition change by the conventional crystal growth methods. Further growth conditions, distribution coefficients and SHG properties are described and discussed, as compared with those of bulk crystals grown by the Czochralski (CZ) method.

There are several methods for the growth of micro single crystals in "fiber" form. Among these, the μ-PD method which is represented in Fig. 1, involves growing single crystals through a micro-nozzle by pulling in the downward direction, has been developed with several advantages : 1) Crystals of device size (not more than 1 mm in diameter) without degradation in quality or cracks can be grown. 2) Melt convection is suppressed by the use of a narrow-distance nozzle by the capillary effect. This allows the growth from incongruent melt because of the effective segregation coefficient near unit.

Using the μ-PD method the raw materials are melted within a crucible with a micro-nozzle at the bottom. Subsequently, the molten material is passed through the micro-nozzle to form a micro single crystal (rod shape). Then, crack-free LN micro single crystals have been grown with uniform diameters in the 60 μm to 800 μm range, independent of melt composition (48.6, 50 and 58 mol%Li_2O). Voids, subgrain boundaries and dislocations (dislocation density is nearly zero) were not found in crystals having diameters of 500 μ m or smaller. The homogeneous axial distribution of the chemical composition reflects an effective distribution coefficient of unity.

Also, crack-free and homogeneous KLN micro single crystals with a cross section not more than 1 mm in diameter and a length of 150 mm have been successfully grown by the μ-PD method, with high growth rates. Good control of the micro crystal diameter has been observed using a micro-nozzle and an afterheater. Especially, no fractures normal to the c-axis have been observed after growth, unlike the cases of crystals grown by the top-seeded solution growth (TSSG) and CZ methods. KLN was capable of frequency doubling from 790 nm to 920 nm by SHG at room temperature suitable for non-critical phase matching (NCPM), and considered on the interchange between incongruent melt composition and SHG properties.

In Fig. 2, correlation between the composition of crystals and the wavelength of SHG applications from ultra-violet to green region is found. Furthermore, a near homogeneous distribution of chemical composition has been found independent on the melt compositions at the μ -PD crystals because of the suppression of melt convection by the use of a narrow-distance nozzle by the capillary effect and by the very stable and linear temperature distribution in the growth region. These results provide the dependence of the most appropriate composition for SHG applications from ultra-violet to green region and the developments of future crystal growth technologies for a new material having improved properties with composition homogeneity from incongruent melt compositions.

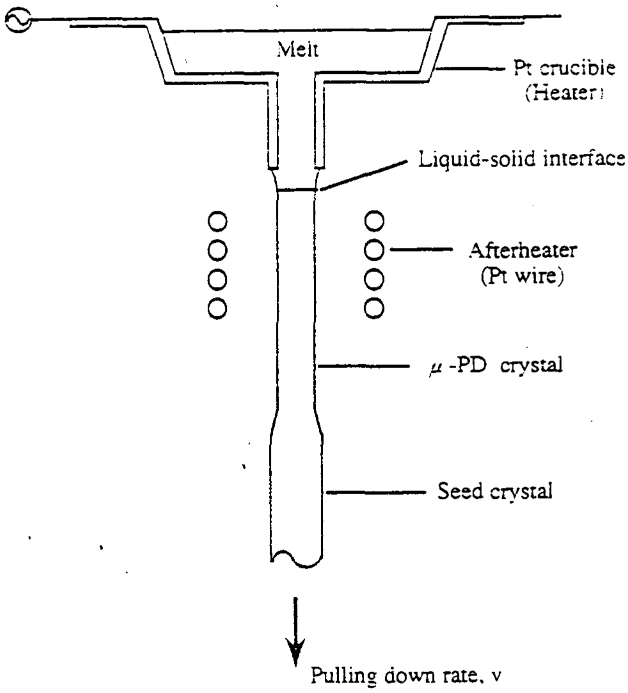

Fig. 1. Scheme of the μ-PD method.

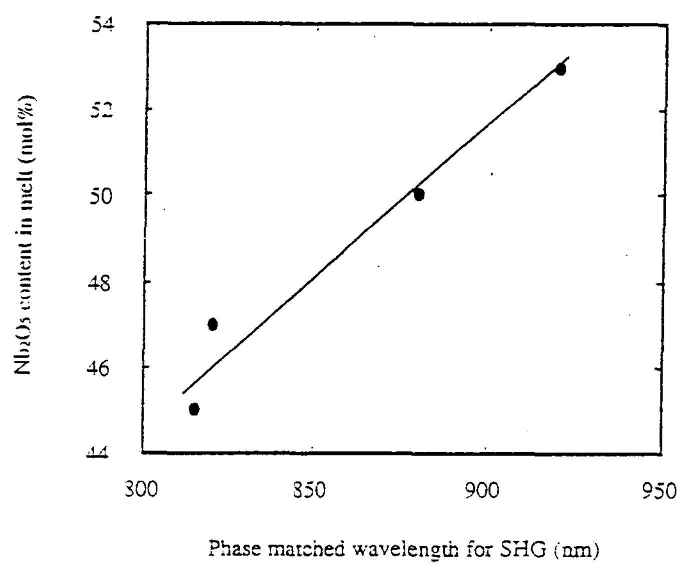

Fig. 2. Correlation between melt composition and NCPM wavelength for KLN crystals.