

## Compositional uniformity in the MgO-doped LiNbO<sub>3</sub> crystal that is concurrently congruent and stoichiometric

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LiNbO<sub>3</sub> (LN) crystals grown under the external electric field by  $\mu$ -PD method have been studied. Doping a certain amount of MgO enables LN to be stoichiometric and congruent simultaneously (denoted as cs-MgO:LN, Li<sub>2</sub>O:Nb<sub>2</sub>O<sub>5</sub>:MgO = 45.3:50:4.7mol%)[1]. It is considered that no segregation of any ionic species takes place during crystal growth of cs-MgO:LN when an appropriate current is imposed, and thus makes it possible to obtain a high-quality LN crystal.

### Background

LiNbO<sub>3</sub> (LN) crystals have received much attention due to their excellent nonlinear properties, and have been applied to a great varieties of devices. The segregation of ionic species is influenced by the interface electric field during the growth of LN crystal. This study was conducted to understand the effect of an interface electric field on the compositions of LN single crystal including cs-MgO:LN via the  $\mu$ -PD method in order to obtain a high-quality LN crystal.

### Experimental

The current injection on the composition of LiNbO<sub>3</sub> was conducted by using the  $\mu$ -PD method. In order to study the segregation behavior of ionic species at the interface, specimens were quenched very rapidly during growth to freeze the solute distribution in the melt. Therefore, we could observe Mg distributions both in the solid and liquid near the interface via EPMA. The Curie temperature and melting temperature of these LN crystals were measured by DSC.

### Results

In the binary system of Li<sub>2</sub>O-Nb<sub>2</sub>O<sub>5</sub>, the effect of an external electric field on the growth of LN single crystal was shown in Fig. 1. In the ternary system of Li<sub>2</sub>O-Nb<sub>2</sub>O<sub>5</sub>-MgO, the compositional variation of cs-MgO:LN crystal by external current injection was determined by Curie temperature. It was found that the current injection to the growth of cs-MgO:LN did not influence the bulk compositional in the grown crystal (Fig. 2), that is,  $k_{eff}=1$ . However, the compositions at the interface varied in association with the interface electric field. Quenched samples that maintain the distribution of solute during crystal growth were analyzed by EPMA (Fig. 3), therefore, the effect of electric field on the interface of solid-liquid could be directly observed. It was found that the intrinsic electric field or the forward current lead to the decrease of Mg concentration in the liquid at the interface,  $C_{L(i)}^{Mg}$ , while the reverse current increased  $C_{L(i)}^{Mg}$ . There exists a certain value of imposed current that was able to counterbalance the intrinsic electric field so that no segregation occurred at the interface.

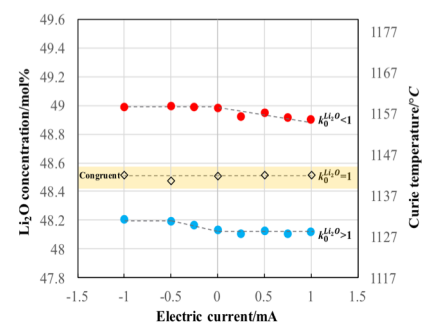


Fig. 1 Electric-current dependent concentration of Li<sub>2</sub>O in LiNbO<sub>3</sub> crystals.

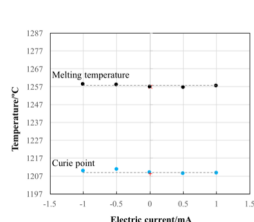


Fig. 2  $T_m$  and  $T_c$  of cs-MgO:LN.

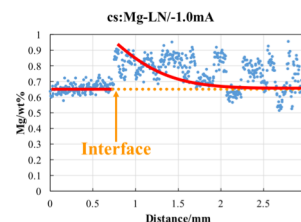
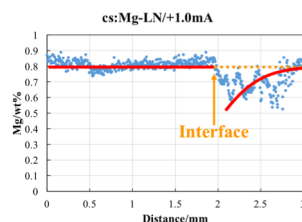


Fig. 3 MgO distribution under (a) +1.0mA and (b) -1.0mA external current.

[1] H. Kimura, S. Uda, Journal of Crystal Growth, 2009, 311(16): 4094-4101.