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Investigation of bulk laser damage threshold of lithium niobate single crystals by Q-switched pulse laser

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Bulk laser damage threshold of 2-in.-diam. MgO-doped $LiNbO_3$ crystals grown using the Czochralski technique has been measured using a Q-switched Nd^{3+}:$LiYF_4$ laser. The threshold showed dependence on the crystal quality. The highest bulk laser damage threshold (14 J/cm^2 at 1.053 μm wavelength with 1-ns pulse width) was obtained in the $LiNbO_3$ crystal doped with 1 mol % of MgO. The lowering of bulk laser damage threshold was observed in the crystals doped with MgO at more than 3 mol %. These crystals contained the local aggregations of MgO which increased the scattering centers and decreased the transparency.

Lithium niobate single crystals have been used in various optical devices such as linear electro-optic devices or nonlinear optical devices, because of the excellent electro-optic effects, nonlinear properties, and low loss waveguide capabilities. However, it has been said that the low optical damage resistance and low bulk damage threshold of $LiNbO_3$ severely limited its application to nonlinear optics in the visible region.

Zhong *et al.*[1] and D. A. Bryan *et al.*[2] have reported optical damage resistance is greatly improved in the bulk crystals of $LiNbO_3$ doped with more than 4.5 mol % MgO. In our previous paper,[3] we also reported that no optical damage is observed in $LiNbO_3$ with 1–5 mol % MgO dopant irradiated by the Ar laser of an incident power of 44 W/cm^2 and an exposure time of up to 10 min. These improvements in optical damage will contribute to the realization of various optical devices using with lasers of short wavelength.

On the other hand, $LiNbO_3$ crystals which can withstand irradiation of energy density over 10 J/cm^2 with 1 ns pulse width at 1.053 μm laser wavelength, are necessary for such applications as frequency doubling and Q-switching of pulse lasers. Nevertheless, it has been thought that the bulk laser damage threshold of $LiNbO_3$ is not enough to be utilized for second harmonic generators of high power lasers. J. L. Nightingale *et al.*[4] have reported that the surface damage threshold of MgO-doped $LiNbO_3$ was measured to be 610 MW/cm^2 at 1.064 μm and 340 MW/cm^2 at 0.532 μm.

In the present paper, we discuss the crystal-growth conditions and the damage resistant properties of MgO-doped $LiNbO_3$ against Q-switched pulse. We measured bulk damage threshold, optical transmission spectra and Mg distribution in the crystal and discussed the relation between the bulk damage threshold and the optical quality of the crystals. We showed $LiNbO_3$ single crystals intrinsically have high damage threshold and indicated its usefulness as a nonlinear optical material.

MgO-doped $LiNbO_3$ single crystals with a diameter of 2 in. were grown using the conventional Czochralski technique. The pulling directions were chosen along the Z- and Y-axes of $LiNbO_3$. Three kinds of concentration levels of MgO, 1, 3, and 5 mol % were added to the noncongruent $LiNbO_3$ melt. High-purity Nb_2O_5, Li_2CO_3 and MgO powder (99.99%) were used as the raw materials. The pulling rate was experimentally varied from 1 to 3 mm/h, the rotation rate from 8 to 20 rpm, and the temperature gradient above the melt surface was also varied from 20 to 50 °C/cm by adjusting the position of Pt crucible. As-grown crystals were poled above the Curie temperature by the conventional field cooling method. The surface of $LiNbO_3$ crystals were finished by mechanochemical polishing with colloidal silica.

Measurement of the bulk laser damage threshold was performed using a Q-switched Nd^{3+}:$LiYF_4$ (YLF) laser (1.053 μm:ω) in a transverse and longitudinal single mode.[5] The pulse widths of the fundamental waves were 1 ns for the short pulse and 25 ns for the long pulse. The second-harmonic waves (0.526 μm:2ω) were also used and the pulse widths were 0.6 and 20 ns. The pulse was focused into Z-cut $LiNbO_3$ samples by a lens of 3-cm focal length. The occurrence of damage was observed by the naked eye. A He-Ne laser was used to identify small damage spots. The focal point was moved shot by shot. Fused silica was used as a reference of standard. The incident laser power was determined by monitoring with a combination of a biplanar phototube and an oscilloscope, which was calibrated by a calorimeter at each wavelength and pulse width. The bulk laser damage threshold is defined as the laser peak power at which the damage happened. Optical transmission spectra was investigated with a spectrometer. Magnesium concentrations in the crystals were determined

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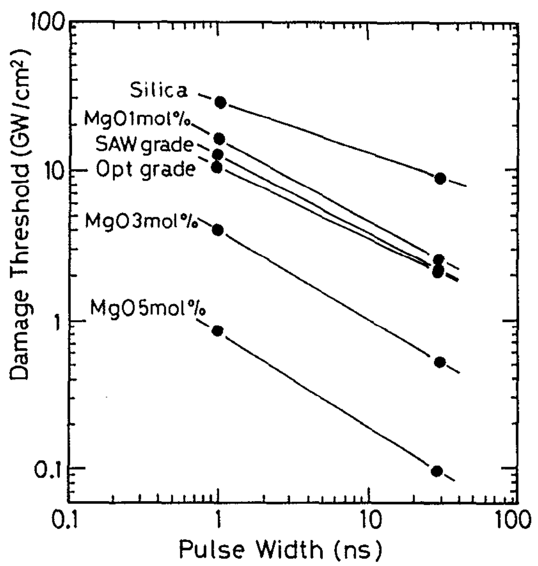

FIG. 1. Dependence of bulk laser damage threshold of $LiNbO_3$ on pulse widths. The laser wavelength was 1.053 μm(ω). Closed circles show the data measured in this experiment.

by electron probe microanalysis (EPMA) using pure metal Mg as a standard.

The results of the bulk laser damage threshold are shown in Figs. 1 and 2. The experimental error is less than at most $\pm 10\%$ due to an ambiguity in calibration of the calorimeter. We investigated six types of samples: optical grade (containing $Fe < 1$ ppm, no subgrain boundary), SAW grade ($Fe > 1$ ppm, some subgrain boundaries), MgO 1, 3, 5 mol % doped crystals ($Fe < 1$ ppm), and fused silica. Every crystal shows higher damage threshold than the values reported before.[4] SAW grade crystals, optical grade crystals and MgO 1 mol % doped crystals have high damage threshold (> 10 J/cm^2 at 1.053 μm, 1 ns). However the crystals doped with 3 and 5 mol % MgO have a lower damage threshold than other crystals. There is a tendency for the damage threshold to decrease as the amount of MgO dopant becomes greater. This damage threshold dependence on the MgO doping level is contrary to that of optical damage resistance due to the photorefractive effect as reported before.[3]

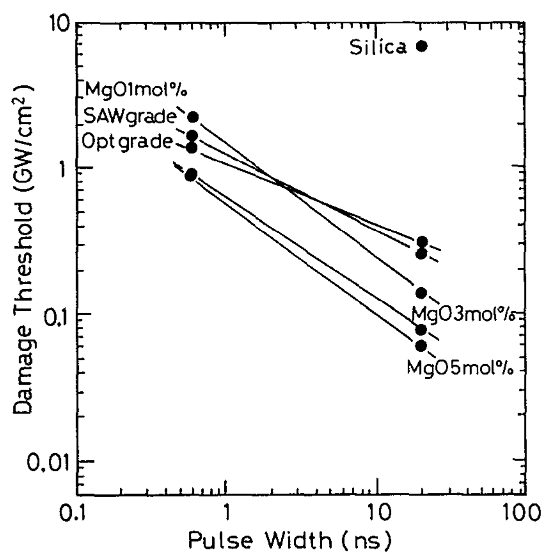

FIG. 2. The dependence of bulk laser damage threshold of $LiNbO_3$ on pulse widths. The laser wavelength was 0.526 μm(2ω). Closed circles show the data measured in this experiment.

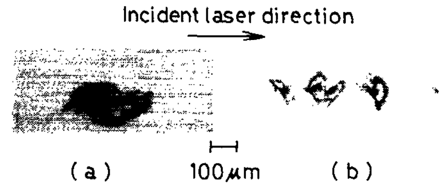

FIG. 3. Typical damages of the crystals; (a) optical grade crystal which have high bulk laser damage threshold; (b) MgO 3 mol % doped crystal which have low damage threshold. The laser wavelength was 1.053 μm and the pulse width was 25 ns.

The damage threshold was lowered with the increasing of the pulse width at both ω and 2ω. This may be due to the fact that the thermal effect is more emphasized with the irradiation of long pulse laser than of short pulse laser. Comparing Figs. 1 and 2 the damage threshold was lowered as the wavelength of the incident laser was shortened. Therefore, the thermal effect is one of the important mechanisms for laser damage in the time interval investigated here.

Figure 3 shows typical damage traces observed in the crystals irradiated by the laser with wavelength of 1.053 μm and pulse width of 25 ns. These kinds of damages were considered to be induced by a dielectric break down, and classified into two categories according to the shape of damages. The first type was observed in crystals with high damage threshold [Fig. 3(a)]. Only one damage, with size of about 200 μm, occurred in the crystal along the incident laser direction, and the damage pattern was similar to other materials such as silica or KDP.[5] The other type of damage was observed in crystals which had low damage threshold [3 and 5 mol % MgO doped crystals, Fig. 3(b)]. The damage occurred at several spots along the direction of the incident laser. The extent of the damage sites were about 50–100 μm. This suggest that in the latter case, some crystalline defects existed, which made the threshold lower. Optical transmission spectra are shown in Fig. 4. The transmission were not calibrated for reflection loss. The difference of reflection losses between samples were neglected, because the dependence of the refractive indices on MgO concentration and wavelength are negligibly small (1×10^{-2}).[3] Comparing the transparency of MgO-doped crystals with that of optical grade crystal, no decrease in transparency was observed in the 1 mol % MgO doped crystal. However, MgO doping of more than 3 mol % caused a decrease of transparency. Much doping of MgO also shifted the fundamental absorption edges toward the

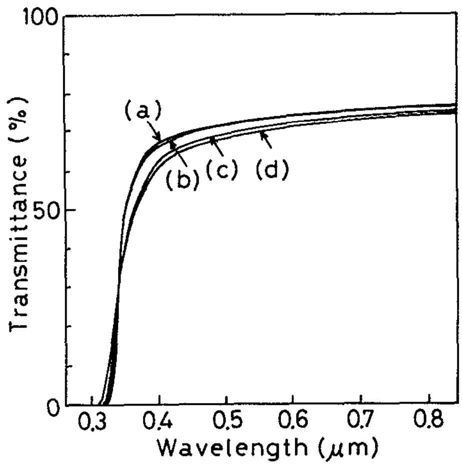

FIG. 4. Transmission spectra of $LiNbO_3$; (a) optical grade crystal; (b) MgO 1 mol % doped crystal; (c) MgO 3 mol % doped crystal; (d) MgO 5 mol % doped crystal. Transmission is not calibrated for reflection loss.

shorter-wavelength side. This shift is consistent with the previous report.[6]

Figures 5(a) and (b) show the Mg distribution in crystals doped with 1 and 5 mol % MgO, respectively. The crystal doped with 1 mol% MgO showed the uniform distribution of MgO along the pulling direction within experimental error [Fig. 5(a)]. On the other hand, local aggregations of Mg were frequently detected in the crystals doped with MgO more than 3 mol % as shown in Fig. 5(b). This aggregation of Mg caused the gathering of subgrain boundaries and imperfection of poling; meaning weak and vague traces of the domain were observed in this crystal.[3] These inhomogeneity increased the scattering centers which decreased the transparency of the crystal.

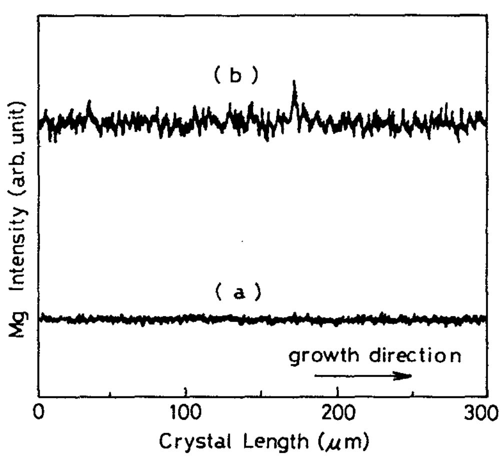

FIG. 5. Mg distribution in MgO doped crystal measured by electron probe microanalysis; (a) MgO 1 mol % doped crystal; (b) MgO 5 mol % doped crystal.

There was a strong correlation between the damage threshold and the transparency. The damage threshold tended to decrease with the decrease of the transparency. We think that the threshold of about 14 J/cm^2 is the intrinsic damage threshold of $LiNbO_3$ crystals, and that the scattering centers in the crystal lead to the lowering of bulk laser damage threshold.

In summary, this work showed that $LiNbO_3$ crystal intrinsically have high damage threshold. This work also showed that there is not any similar dependence on crystal quality between the bulk damage threshold and the optical damage resistance. We conclude that the main reason for the low bulk laser damage threshold of $LiNbO_3$ is the scattering centers such as local aggregations of Mg or vacancy clusters which are generated in crystals during crystal growth. It has been said that doping MgO of more than 4.5 mol % was effective in improving optical damage resistance. However, high MgO doping caused the problems of reducing the damage threshold and optical homogeneity. We have to improve the quality of crystals doped with MgO more than 3 mol % by developing the condition and technique of crystal growth, so that the new properties of the high MgO doped $LiNbO_3$ single crystals permit new flexibility in the design of various optical devices.

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[1] G.-G. Zhong, J. Jian, and Z. K. Wu, in *Proceedings of the 11th International Quantum Electronics Conference,* IEEE. Cat. No. 80, CH1561-0, June 631 (1980).

[2] D. A. Bryan, R. Gerson, and H. E. Tomaschke, Appl. Phys. Lett. **44**, 847 (1984).

[3] Y. Furukawa, M. Sato, F. Nitanda, and K. Ito, J. Cryst. Growth **99**, 832 (1990).

[4] J. L. Nightingale, W. J. Silva, G. E. Reade, and A. Rybicki, SPIE Laser Nonlinear Optical Materials **681**, 20 (1986).

[5] A. Yokatani, T. Sasaki, K. Yoshida and S. Nakai, Appl. Phys. Lett. 55, 2692 (1989).

[6] K. Polgar, L. Kovacs, I. Foldvari, and I. Cravero, Solid State Commun. **59**, 375 (1986).