The impact of MgO-doped near-stoichiometric lithium niobate crystals on the THz wave output characteristics

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Abstract: The control experimental study on the THz wave parametric oscillator (TPO) output characteristics based on the congruent LiNbO₃ crystal (CLN) and stoichiometric MgO-doped lithium niobate (SLN) crystal is performed. As a nonlinear medium, in the aspect of the THz wave output, experiments show that the congruent LiNbO₃ crystal is more stable than the SLN crystal. Compared with the CLN crystal, SLN showed significant photorefractive effect, which adversely the stability of the THz wave output. Experiments indicated that different molar concentration of MgO doped, can significantly change the photorefractive properties of SLN crystal. The results showed that with the increase of MgO doping concentration, the photorefractive of SLN gradually become weaker, and THz wave output stability has the significantly increase. The output stability of mol 5.0% MgO droped SLN crystal, has not significantly different with the CLN crystal, while the peak THz energy output increased 28%.

1. Introduction
In the middle of the last century, Y R.Shen[1], B.C.Johnson and J.Nishizawa[2] have proposed the theory that sub-millimeter electromagnetic wave can be generated by nonlinear optical crystal. In 1996, the Ito lab of Japan first reported the research work about THz wave parametric oscillator based on nonlinear LiNbO₃ crystal[3]. This THz wave source has the characteristics of frequency tunable, narrow line width (high monochromatic), high spatial and temporal coherence, high energy output, and operation at room temperature. The threshold of TPO is closely related with the parameter of resonant cavity itself. In addition, experiments show that the characteristics of the crystal material had made a great impact on the operation threshold of oscillator. With the rapid development of LiNbO₃ research, choosing the appropriate crystal to reduce the TPO-threshold and to optimize the output characteristics is not only necessary, but also has the exploration significance in the research.

LiNbO₃ crystal has the important feature and application prospects, primarily due to its elements doping[4,5]. In recent years[6], compared with congruent LiNbO₃ crystal (CLN), stoichiometric MgO-doped lithium niobate (SLN) crystal is more attracting attention, many researchers have achieved the significant progress in this area. The LiNbO₃ crystal is the typical non-stoichiometric ratio crystal, the LiNbO₃ crystal which is widely used is the same ingredient allocated proportion ([Li]/[Li]=48.3~48.6/51.7~51.4) at present. The LiNbO₃ crystal (CLN) grewed in the same ingredient allocated proportion has obviously superiority in the growth technology, but it bing the large number...
of intrinsic defects at the same time, mainly due to the antisite defect of (NbLi) and vacancies of (VLi). The intrinsic defects in CLN reduced the many properties of crystal. The growth intrinsic defects of stoichiometric ([Nb]/[Li]=1:1) LiNbO₃ crystal (SLN) was reduced obviously. Compared with the CLN crystal, the results show that the SLN crystal performance has shows considerable improvement, only in terms of nonlinear coefficient and electro-optic coefficient enhanced about 30%. The nonlinear coefficient of CLN is 13.8 pm / V, the SLN is 17.5 pm / V. The photo-damage resistance of CLN is 100kW/cm², and the SLN photo-damage resistance is 104kW/cm².

From the application view of TPO, we hope that crystal non-linear coefficient has a large nonlinear coefficient and the strong ability of anti-photorefractive. The big nonlinear optics coefficient is not only advantageous in reduces the oscillation threshold of TPO, but also can enhances the THz wave energy output; meanwhile, the photorefractive resistance ability is benefical to the stability of THz wave radiation. In this paper, 5% mol MgO doped SLN crystal was used as a nonlinear optical medium of TPO. The proposal of this experiment is to enhance the THz wave output by using the large nonlinear coefficient of SLN, and reduced the threshold of TPO. Simultaneously, the photorefractive effect is expected to reduce due to the MgO doped.

2. The impact of SLN and CLN on the output stability of the TPO

Only from the level of crystal growth and crystal structure can make a fundamental interpretation for the physical performance differences among CLN and SLN, and different molar concentration of MgO doped SLN crystal. This paper showed the qualitative analysis by the contrast experiment. In the contrast experiment used four types LiNbO₃ crystal: (1) the congruent LiNbO₃ crystal (CLN); (2) near-stoichiometric 0.89% mol MgO: SLN crystal; (3) 1.7% mol MgO: SLN; (4) 5.0% mol MgO: SLN crystal. The crystal geometry of four types LiNbO₃ is same, all for the 65mm × 10mm × 10mm. Experimenting[] in the exact same experimental condition, all types crystal as the nonlinear medium of a TPO with the 154mm cavity length, respectively, the THz wave colleted in a vacuum calorimeter, and then the calorimeter signal inputed oscilloscope[] to record the electrical signals amplitude corresponding to THz wave.

Figure 1 is the THz wave output with time of 0.89% mol MgO: SLN in 27 minutes. Labview control platform automatically recorded the output voltage in every 10 seconds. You can see, the signal amplitude of THz wave first reached a peak in four minutes, then gradually attenuated to a certain value. In the initial stage THz wave rised because of the YAG laser device was turned on, there is a gradual process of laser energy rised and stabilized, THz wave signal also tended to increase with the laser intensity increased; When the laser output has stabilized, the output of the THz wave

![Figure 1](image1.png)
[Figure 1. Output curve of 0.89mol%MgO:SLN in 27 minutes](image1.png)

![Figure 2](image2.png)
[Figure 2. Output curve of 0.89mol%MgO:SLN in 47 minutes](image2.png)
reflected attenuation. This showed that physical property of the 0.89% mol MgO: SLN is not stable, will change in the light. Figure 2 is the output curve of 0.89% mol MgO: SLN in 47 minutes in the case of laser output stabilized. The curve also reflected the instability characteristics of SLN, and with continuing of the illumination time, THz wave output even attenuated to zero. Figure 3 is the output curve of 1.7% mol MgO: SLN crystal when the laser energy output stabilized. The curve showed the THz wave output still attenuated when the pump incidence, then tended to stabilize at a lower average level. Compared with 0.89% mol MgO: SLN, the crystal output stability has improved. Figure 4 is the output curve of 5.0% mol MgO: SLN crystal under the same conditions. Compared with the above three types crystal, the THz wave output of crystal stabilized at a average level, reflected the stability of the physical properties of crystal.

Figure 5 is the output curve of the congruent LiNbO$_3$ crystal (CLN) under the same conditions. Compared with the SLN, output of CLN is stable, reflected the stability of its physical properties better than near-stoichiometric SLN crystal. The contrast experiments showed the instability of the THz wave output was due to photorefractive effect of LiNbO$_3$ crystal.

3. Comparison of the oscillation threshold and energy output of CLN and 5% mol MgO: SLN

For THz wave parametric oscillator, the adverse impact of the photorefractive are (1) phase mismatch caused by photorefractive effect would affect the efficiency of THz wave radiation; (2) the loss of light in the crystal become very serious; (3) photo-damage caused by photorefractive would seriously influences the performance of TPO.

![Figure 3. Output curve of 1.7mol% MgO:SLN in 72 minutes](image3)

![Figure 4. Output curve of 5.0mol% MgO:SLN in 72 minutes.](image4)

![Figure 5. Output curve of CLN in 72 minutes](image5)
Figure 6 is the threshold comparison of the SCN-TPO with that of the CLN-TPO at 160mm cavity length. To ensure the comparability in experimental test, the frequency of output THZ wave both are 1.2 Thz, and the crystal length are 65mm.

From the curve, the pump laser threshold of SCN-TPO approximately is 23mJ/pulse, and MgO:SLN approximately is 18mJ/pulse. That is to say: 5%mol MgO:SLN crystal used in the THz wave parametric oscillator make the TPO threshold reduced 21.7%. Just like front introduces, the cause is that 5%mol MgO:SLN has the big nonlinear optics coefficient, and dopes the appropriate mole fraction MgO, which suppressed the photorefractive effect. The vibration threshold value reduces means: at the same level condition of the incident pump luminous energy invariable, the output THz wave in all frequency tuning range is to increase. Figure 7 is under the same pump energy is 20mJ/pulse, the comparison of THz output of 65mm 5% mol MgO: SLN and CLN crystal with equal length. The peak energy of SLN is 90pJ/pulse, while CLN is 65pJ/pulse, the peak energy of SLN has increased 28%.

4. CONCLUSIONS
As a nonlinear medium which generated THz wave on the output stability of the device, the CLN crystal of congruent ratio growth is more stable than the near-stoichiometric SLN crystal.

The experiment results indicated that compared with the CLN crystal, SLN showed significantly photorefractive effect; But can significantly change the photorefractive properties of SLN crystal by different molar concentration of MgO-doped. With the increase of MgO doping concentration, the photorefractive of SLN gradually becomes weaker the output stability has the increase. Experiments pointed out that the THz wave output stability of mol 5.0% MgO:SLN crystal, has not significantly different with the CLN. In the contrast experiment of TPO with the 160mm cavity length and 65mm crystal length, the pump laser threshold of the 5% mol MgO: SLN crystal decreased by 21.7% than the CLN crystal, while the peak energy output increased 28%.

This paper only analysed some differences on the THz-wave output of the MgO: SLN and CLN crystal used in THz wave parametric oscillator, relevant experimental phenomenon can be used as a reference.

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**Acknowledgments**
Authors wishing to acknowledge the financial support from the National Natural Science Foundation of China (Grant No. 60971015, 60771053), Natural Science Foundation of Shaanxi Province (Grant No:8JK402), Science Foundation of Xi’an University of Technology (Grant No. 108-210703) and Well-educated Staff Science and Technology Start-up Foundation of Xi’an University of Technology (Grant No. 108-210801). The authors especially thanks the Ito-lab of RIKEN for their support.