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Efficient generation of red light by frequency doubling in a periodically-poled nearly-stoichiometric $LiTaO_3$ crystal

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An efficient generation of red light in a periodically-poled nearly-stoichiometric $LiTaO_3$ (PPSLT) by extracavity single-pass frequency doubling of a diode-pumped, Q-switched Nd:YVO4 laser at 1342 nm was realized. An average power of 1.4 W of the 671 nm red light is obtained at the fundamental power of ~2.8 W with the conversion efficiency of 50%. The high conversion efficiency and steady output of red light indicate that the thick PPSLT is a competitive candidate for frequency conversion in order to construct a compact all-solid-state red laser. © *2004 American Institute of Physics*. [DOI: 10.1063/1.1772525]

Optical superlattices based on quasi-phase matching (QPM) theory, such as periodically-poled $LiTaO_3$ (PPLT),[1,2] periodically-poled $LiNbO_3$ (PPLN)[3–5] and periodically-poled $KTiOPO_4$ (PPKTP),[6,7] are promising materials for second-harmonic-generation (SHG) of visible light from an IR laser. As for the congruent $LiTaO_3$ (CLT) and $LiNbO_3$, they have large nonlinear coefficients and wide transparent ranges[8,9] and can get large size of single crystals. But the high coercive fields make them difficulty to fabricate several-millimeter-thick periodically-poled samples that are suitable for high-power operation. Now, most of the poled samples are of the thickness to be limited to 0.5–1.0 mm. For KTP, although it has low coercive field, it is difficult to grow large crystals that are long enough for efficient frequency conversion.

Recently, techniques such as the double crucible Czochralski method[10,11] and the vapor transport equilibration method[12] have been developed to grow stoichiometric $LiTaO_3$ (SLT) crystal. In a SLT crystal, the nonstoichiometric defect is significantly reduced, which leads to a reduction of the coercive field by one order and an increase of the optical damage threshold by two to three orders.[13] In this letter, we report the periodic poling of a nearly SLT crystal with the thickness of 1.2 mm and the characteristics of QPM-SHG performance with the PPSLT crystal pumped by a Q-switched $Nd:YVO_4$ laser.

In this work, the nearly SLT crystal is grown by use of a top-seeded solution growth technique from a stoichiometric melt with the addition of 11 mol % of K_2O as flux. As mentioned in Ref. 14, the position of the ultraviolet (UV) absorption edge is a very sensitive indicator of the composition of $LiNbO_3$ and $LiTaO_3$ crystals. The measurement showed that the UV absorption edge of our sample was located at 263 nm. We estimate from the result that the [Li]/[Ta] in the crystal is about 49.6/50.4, therefore close to stoichiometric. The coercive field of the crystal is ~2 kV/mm at room temperature, which is one tenth that of CLT.

According to the QPM theory, the effective nonlinear coefficient of a periodic superlattice is $d_{\mathrm{eff}}=2d_{33}/\pi m \times \sin(\pi m D)$, where m is the QPM order and D is the duty cycle. In order to get the maximum of d_{eff}, we choose m to=1 and $D=0.5$. The QPM condition in a collinear interaction is

$$k_s - 2k_f - \frac{2\pi}{\Lambda} = 0, \tag{1}$$

where k_f and k_s are the wave vectors of the fundamental and the second harmonic, respectively, Λ is the period, and we have $k_{(f,s)}=2\pi n_{(f,s)}/\lambda_{(f,s)}$, where n_f and n_s are the refractive indices of the fundamental and the SH, respectively, and both of them are the function of wavelength and temperature. For this study, the wavelength of fundamental is 1342 nm that is a strong emission line of the $^4F_{3/2}-^4I_{13/2}$ transition of Nd^{3+} ion in YVO_4 crystal. From Eq. (1), we preset the period of the PPSLT $\Lambda=14.61$ μm. The phase matching temperature is around 110 °C for a CLT crystal with this period in terms of Sellmeier equation in Ref. 15. The Sellmeier equation of SLT has been given by the authors in Ref. 16. Expectably, the actual phase-matching temperature for the present crystal will be different from the preset value due to the critical dependence of refractive indices on the ration of [Li]/[Ta] in the crystal.

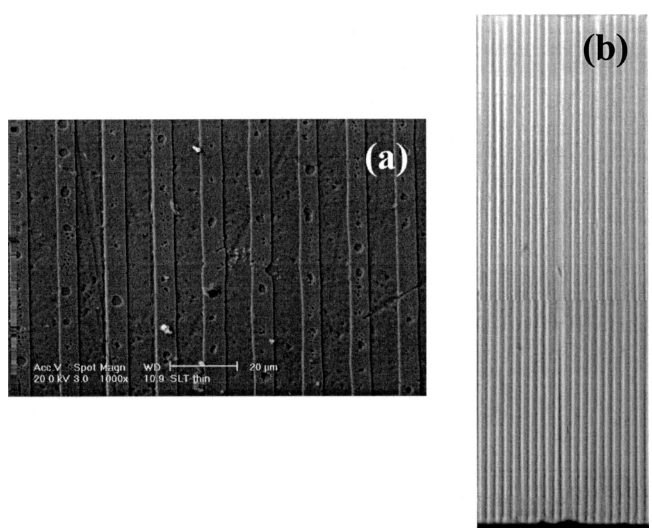

FIG. 1. (a) Scanning electron microscope micrograph of etched domain-inverted patterns on the +C surface. (b) Cross-sectional view of Y face of the PPSLT.

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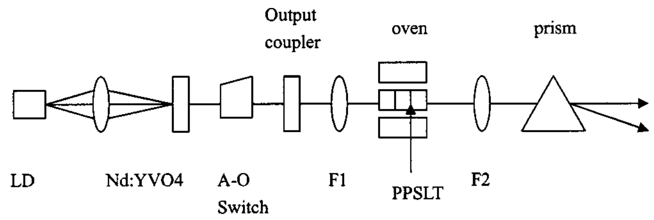

FIG. 2. The schematic experiment setup. Nd:YVO4 crystal: The front end face with film HR@1342 nm and AR@809 nm is a reflective mirror of the cavity, the output end face is coated with film AR@1342 nm to suppress the loss; the output coupler of the cavity with the film T=8%@1342 nm.

The PPSLT sample with 20 mm in length and 1.2 mm in thickness was fabricated using the conventional electrical poling technique.[17] Figures 1(a) and 1(b) are the micrographs of +C and Y face of the PPSLT. We can see from Fig. 1(a) that the inverted domain distribution is uniform on the +C surface and the duty cycle is close to 50%. Figure 1(b) is the cross-sectional view of the Y face, which indicates that the inverted domains penetrate through the whole thickness of 1.2 mm and domain boundary is smooth. This is favorable for frequency conversion application.

The schematic experiment setup is shown in Fig. 2. The fundamental sources was a laser-diode-pumped, Q-switched, 1342 nm $Nd:YVO_4$ laser. The gain crystal was 3 mm $\times$3 mm$\times$5 mm in size. The cavity was composed of two mirrors, one of them being the gain crystal itself coated on its front surface, the other an output coupler. The properties of the films coated on these two mirrors are shown in Fig. 2. In this laser system, an acousto-optical Q-switch was laid into the cavity, which generated pulses with a duration of ~40 ns at a repetition rate of 20 kHz. The focus length of the lens $F1$ was 25 mm, and a waist spot about 50 μm in diameter inside the sample was estimated. The actual maximum fundamental average power incident into the sample was ~2.8 W and the corresponding peak power was 3.3 kW. The two end faces of the sample were polished but no antireflection coating was used. Taking into account the Fresnel reflection of about 13% on the front surface of the PPSLT, the peak intensity at the waist was about 170 MW/cm^2 under the power level. A heater was used to heat the sample to the phase-matching temperature with an accuracy of 0.1 °C. The output red light and the fundamental beam were separated with a prism, and were detected with a power meter, respectively.

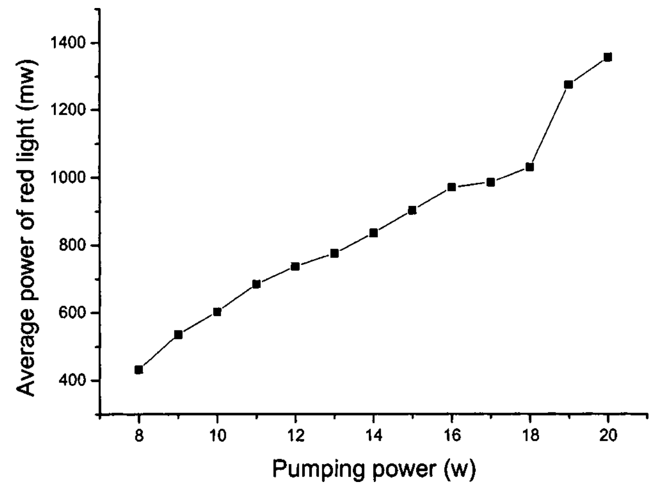

FIG. 4. Average power of red light vs pumping power at the temperature of 191.9 °C.

Figure 3 shows the temperature tuning curve of red light and the normalized SHG efficiency versus the sample temperature. The measured phase-matching temperature was 191.9 °C with the full width at half maximum of ~3 °C. The measured temperature bandwidth is very close to that of the theoretical one, which indicates that the sample phase matches over the whole 20 mm length, and that the refractive index inside of the crystal is uniform in the wavelength range. An output power of 1.4 W was obtained at the maximum fundamental power of 2.8 W with conversion efficiency up to 50%. Meantime the pumping power was about 20 W, which corresponds to a light–light conversion efficiency of 7%. By raising the pump power or increasing the effective length of the sample, we are sure to increase the output power of red light. The average power of red light versus pumping power at the temperature of 191.9 °C is shown in Fig. 4.

Figure 5 displays the facula of the output red light that exhibits a circularly shaped beam with a Gaussian profile, which was scarcely seen using the previously thinner PPLT or PPLN samples. Figure 6 shows the stability of the output beam in 0.5 h during the experiment period and it exhibited only a fluctuation of ~1.0%, which indicates that the nearly SLT crystal had a homogeneous optical quality and no observably optical damage took place at the temperature and the power level. Actually, during the whole experiment pro-

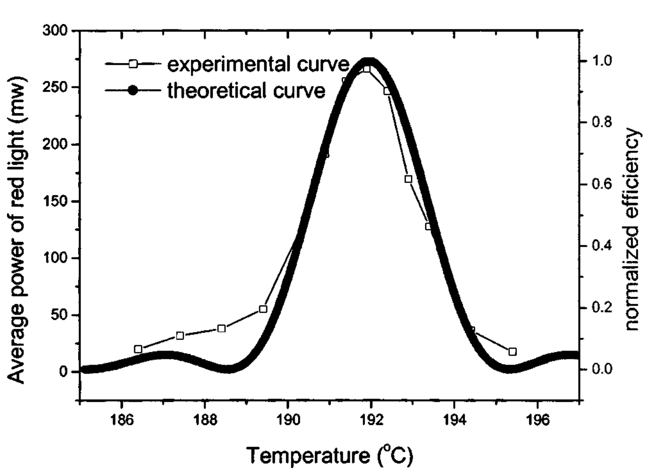

FIG. 3. Measured temperature tuning curve for red light and the dependence of the normalized SHG efficiency on the bulk temperature.

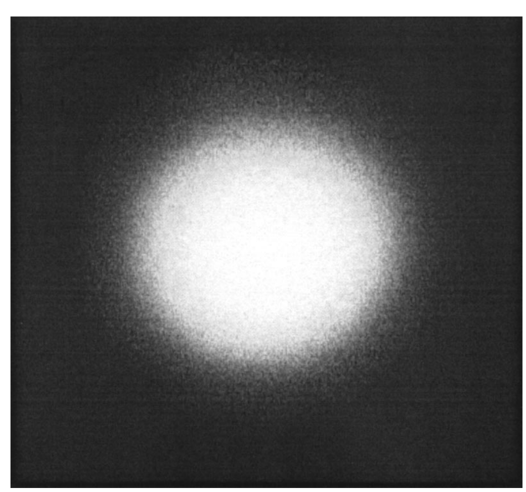

FIG. 5. The facula of the output red light beam.

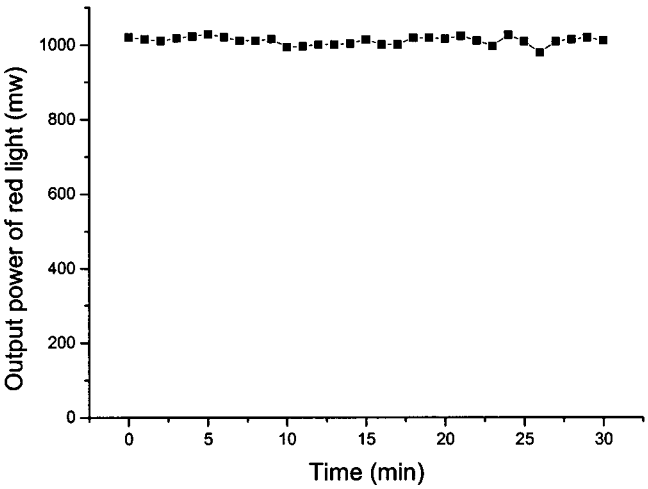

FIG. 6. Stability of the output power of red light.

cess that persisted for several days, the output power kept stable. And according to our observation, that the main factor causes the fluctuation is the temperature of heater who has a fluctuation of ~0.1 °C. The output power is sensitive to the temperature variation due to the narrow temperature bandwidth of the PPSLT sample.

In summary, a 1.2-mm-thick PPSLT was fabricated for the objective of the frequency doubling of 1342 nm laser. The sample exhibits a good poling uniformity, smooth domain boundary and high optical damage threshold. Under a high focus intensity of 170 Mw/cm^2 at the waist, the PPSLT sample performs well, no optical damage was observed and the output was stable during the whole experiment process. The high conversion efficiency of output red light indicates a large efficient nonlinear coefficient of SLT crystal. Also, because of using the thicker sample, the beam shape was better than that of a thin sample. To get an accurate design of nonlinear optical devices, however, educing the Sellmeier equation for the crystals with different stoichiometric ratio is a necessary work in future.

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