Appl. Phys. B 67, 29–32 (1998)

Applied Physics B
Lasers
and Optics

Spectral properties and quasi-phase-matched second-harmonic generation in a new active medium: optical superlattice Nd:MgO:$LiNbO_3$

Y.Q. Lu[1], J.J. Zheng[1], Y.L. Lu[1], N.B. Ming[2]

[1] National Laboratory of Solid State Microstructures, Nanjing University, Nanjing 210093, P.R. China
(Fax: +86-25/3300-535, E-mail: yqlu@nju.edu.cn)
[2] CCAST (World Laboratory), P.O. 8730, Beijing 100080, and National Laboratory of Solid State Microstructures, Nanjing 210093, P.R. China

Received: 22 August 1997/Revised version: 10 January 1998

Abstract. A new multifunctional active medium – periodically poled optical superlattice Nd:MgO:$LiNbO_3$ crystal – was grown in our Laboratory. The absorption and fluorescence spectra were measured and show some differences from the results of common Nd:MgO:$LiNbO_3$ with monodomain structure. Third-order quasi-phase-matched violet second-harmonic generation in the sample was demonstrated to characterize the quality of the crystal. The properties indicate that the periodically poled optical superlattice Nd:MgO:$LiNbO_3$ has great potential for constructing a blue or green light source through self-frequency-doubling operation using the maximum nonlinear optical coefficient d_{33} of $LiNbO_3$ and the high-gain π polarization at the same time.

PACS: 42.55; 42.65; 42.70

Nd:MgO:$LiNbO_3$ combines the excellent laser properties of Nd^{3+} with the electro-optical and nonlinear optical properties of $LiNbO_3$, which permits the design of multifunctional laser devices. MgO doping was found to greatly reduce the photorefractive damage [1]. In 1986, T.Y. Fan et al. first reported the laser oscillations in Nd:MgO:$LiNbO_3$ and demonstrated that it is suitable for self-Q-switching and self-frequency-doubling (SFD) [2]. Later, the diode-pumped laser oscillation [3, 4] and cw SFD laser operation [5] in Nd:MgO:$LiNbO_3$ were successfully demonstrated.

Nd:MgO:$LiNbO_3$ crystal with monodomain structure has several shortcomings. Its largest nonlinear coefficient d_{33} can not be used and it can not be phase matched when the fundamental wavelength is shorter than 1 μm because of insufficient birefringence, hence it is impossible to frequency double the 0.94 μm $^4F_{3/2} - {}^4I_{9/2}$ transition of Nd^{3+} for blue light generation. Moreover, in standard Nd:MgO:$LiNbO_3$, the low-gain σ polarization (E⊥C) has to be used for achieving 90° phase matching [2]. Though the insertion of a quarter-wave plate in the oscillation cavity can force the laser change to σ polarization when the SFD laser is operated at the high-gain π (E//C) polarization, the cavity losses are increased consequently [5]. To overcome these shortcomings, a technique was proposed to use the periodically poled optical superlattice Nd:MgO:$LiNbO_3$ crystal. The $(e^{\omega} + e^{\omega} \rightarrow e^{2\omega})$ quasi-phase-matching (QPM) can easily utilize the high-gain π polarization and the maximum d_{33} at the same time.

The idea of QPM was proposed by Bloembergen in 1962 [6]. It predicts that the nonlinear optical effects can be enhanced in both birefringent or non-birefringent crystals with periodic modulation of nonlinear susceptibilities, i.e. in optical superlattice crystals. In $LiNbO_3$ with monodomain structure, the largest nonlinear coefficient d_{33} is not phase matchable. Since d_{33} is about 7.5 times larger than the ordinarily used d_{31}, we may expect a QPM enhancement factor of $(d_{33}/d_{31})^2(2/\pi)^2 \approx 23$ in an optical superlattice $LiNbO_3$ with the modulation period of $2L_c$ in the process of second-harmonic generation (SHG), where L_c is the coherence length. In addition, this enhancement can be achieved at an arbitrary temperature over the crystal's entire transparency range [7–12]. Thus if there is an optical superlattice Nd:MgO:$LiNbO_3$ crystal, the efficient blue or green light generation by SFD of the $^4F_{3/2} - {}^4I_{9/2}$ transition or the $^4F_{3/2} - {}^4I_{11/2}$ transition line will both be able to be achieved. This is impossible in ordinary monodomain Nd:MgO:$LiNbO_3$.

In this work, we will report the growth of optical superlattice Nd:MgO:$LiNbO_3$ crystals with proper modulation period. The absorption and fluorescence spectra were recorded. The 3rd-order QPM SHG in an optical superlattice Nd:MgO:$LiNbO_3$ sample with the modulation period of 7.5 μm was measured to characterize the quality of the sample.

1 Crystal growth

The optical superlattice Nd:MgO:$LiNbO_3$ crystals were grown along the *a* axis by the Czochralski technique in a carefully designed asymmetric temperature field as we described early [7, 9–11]. The starting materials were prepared by doping 4.5 mol% MgO and 0.2 wt% Nd_2O_3 in a congruent melt of $LiNbO_3$ (Li_2CO_3/Nb_2O_5=48.6/51.4) with the chemical purity of 99.99%. The resulting crystals were clear, transparent, and slightly blue in color. After being *b*-face cut and polished, the crystals were etched in 1HF:2HNO_3 for 10 min

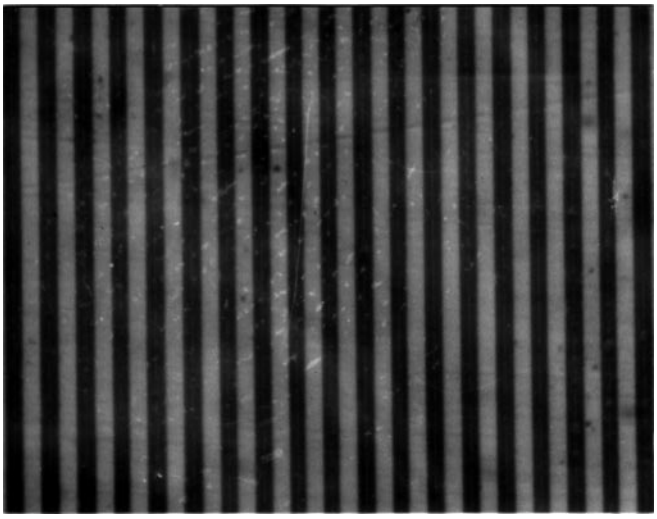

Fig. 1. Optical micrograph showing periodic ferroelectric domain structure of *b* face in an optical superlattice Nd:MgO:$LiNbO_3$ crystal

and examined in an optical microscope to reveal the ferroelectric domain structures. Figure 1 shows the optical micrograph of a sample. The positive and negative domains are almost equal and the domain structure shows good periodicity. The sample has the dimensions $2.5 \times 3 \times 3$ (a × b × c) mm^3 with the modulation period of 7.5 μm and period fluctuation of below 4%. In the experiments below, it was chosen to make the spectral properties and SHG measurement. The light transmitting direction is along the *a* axis.

2 Spectral properties

Figure 2 shows the π and σ polarized absorption spectra at 300 K of the optical superlattice Nd:MgO:$LiNbO_3$ sample in the spectral domain 550 nm–850 nm. The electronic energy levels reached from the ground state $^4I_{9/2}$ can be identified from the peak position of the recorded absorption spectrum and are listed in Table 1. The data of π and σ polarization show some difference, but both have three main absorption bands in the spectra: one is near 600 nm, the other two are at about 750 nm and 810 nm. In the 810 nm region, three absorption peaks can be observed in the spectrum of σ polarization. They are at 808 nm, 813 nm, and 824 nm. The maximum one is at 808 nm. In the π-polarization case, four linked peaks exist and form an intensive absorption band with the central wavelength of 814 nm. The height is larger than the three peaks of σ polarization. It is very beneficial for the easily available 810-nm diode laser pumping. Comparing these data with that of common Nd:MgO:$LiNbO_3$ with monodomain structure, some differences in the peak position and relative intensities are appreciated. For example, the 753 nm peak of π polarization is very much higher than the π-polarization 758 nm peak, whereas in the case of common Nd:MgO:$LiNbO_3$, the heights of the two peaks are generally equal [13]. Perhaps this is caused by the existence of periodic domain structure, but the physical origin is not clear.

The fluorescence spectra were measured by using a tunable cw Ti:sapphire laser excitation at 814 nm. A polarizer and a monochromator were used to analyze the spectra and a detector received the emitted light from the slit of the monochromator. Figure 3 shows the fluorescence spectra near 1.09 μm, corresponding to the transition of $^4F_{3/2} - {}^4I_{11/2}$. In this figure, the spectral peaks at 1080, 1085, 1093, and 1108 nm exist in both π and σ polarization conditions. In the case of σ polarization, the intensity of the 1093 nm spectral

Table 1. Electronic energy levels and related peak positions in the polarized absorption spectra of Nd:MgO:$LiNbO_3$

	State	$^4G_{5/2}$, $^2G_{7/2}$	$^4F_{7/2}$, $^4S_{3/2}$	$^4F_{5/2}$, $^2H_{9/2}$
π polarization	Peak position /nm	582, 590, 598	744, 753, 758, 768	814
σ polarisation	Peak position /nm	589, 599, 611	744, 751, 756, 765	808, 813, 824

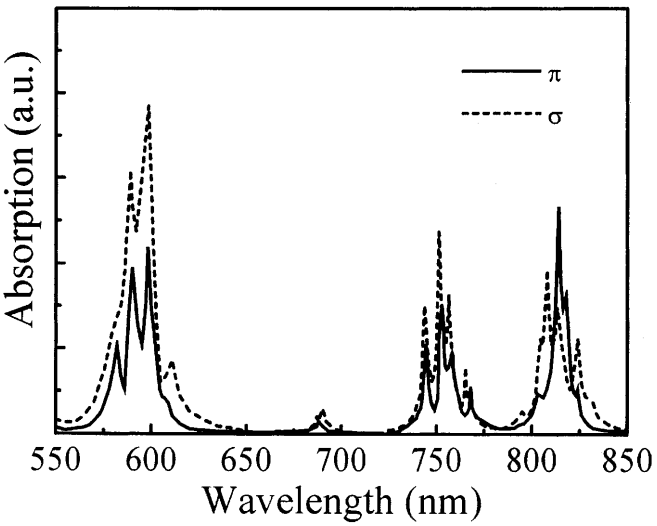

Fig. 2. Polarized absorption spectra of an optical superlattice Nd:MgO:$LiNbO_3$ crystal. The *solid line* corresponds to π polarization and the *dashed line* to σ polarization

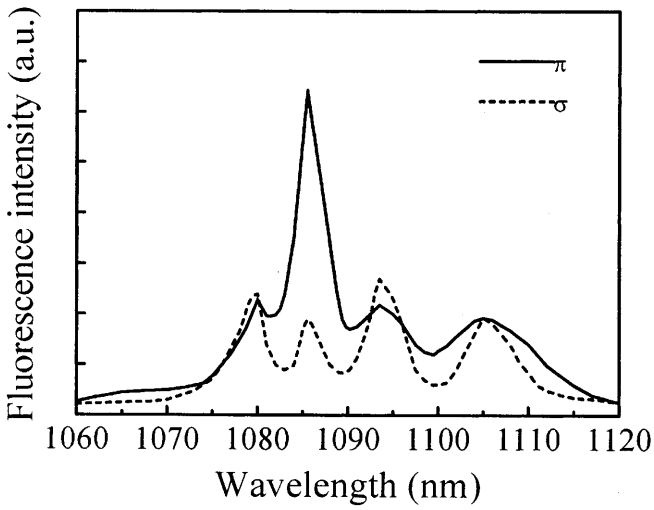

Fig. 3. Polarized fluorescence spectra of an optical superlattice Nd:MgO:$LiNbO_3$ crystal near 1.09 μm. The *solid line* corresponds to π polarization and the *dashed line* to σ polarization

line is the maximum. Whereas in the π polarization case, the 1085 nm line is the most intensive one and it is also much higher than the 1093 nm line of σ polarization. It shows that the π-polarization excitation is really high gain. These results are generally similar to previous published data of common Nd:MgO:$LiNbO_3$, except that the peak position and peak intensity show a few differences.

3 Quasi-phase-matched second-harmonic generation

Using a tunable dye laser (Model ND6000, Continuum Co.), the third-order QPM SHG in the same periodically poled Nd:MgO:$LiNbO_3$ sample was measured for characterizing the nonlinear optical properties. The dye laser is pumped by a Nd:YAG laser with a pulsewidth of 8 ns and a repetition rate of 10 Hz. The fundamental beam was focused by a $f = 30$ cm lens on the sample with the light spot area of about 1 mm^2. The beam polarization is along the c axis, just corresponding to the high-gain π polarization. Figure 4 shows the SHG energy dependence on the fundamental wavelength. A peak is obtained for the fundamental wavelength of about 790 nm. The full width at half maximum (FWHM) is about 2 nm. However, the calculated FWHM of a sample with perfect periodic domain structure with the same thickness and the same modulation period is only 0.2 nm. This great difference should be due to the irregular fluctuation of modulation period. The focused fundamental beam could also make the FWHM wider. On the other hand, the second-harmonic light is near the ultraviolet absorption edge, the Nd^{3+} also has slight absorption of fundamental light, and they both may affect the shape of our SHG tuning curve in Fig. 4. According to the QPM theory [14] and the Sellmeier equation coefficient [15] of Nd:MgO:$LiNbO_3$, the 1st-order QPM optimum fundamental light of this sample is at 1088 nm, just nearing the 1085 nm maximum spectral peak. These results shows that the sample is suitable for the generation of SFD of the 1085 nm line of π polarization.

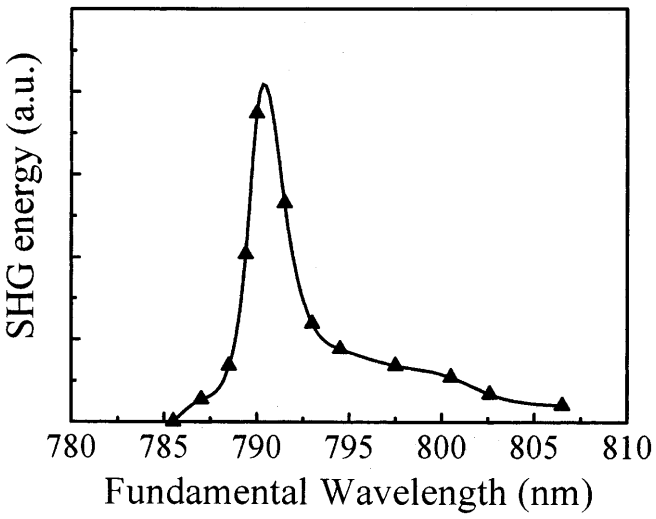

Fig. 4. SHG energy against fundamental wavelength generated through 3rd-order QPM in an optical superlattice Nd:MgO:$LiNbO_3$ sample with a 7.5 μm modulation period

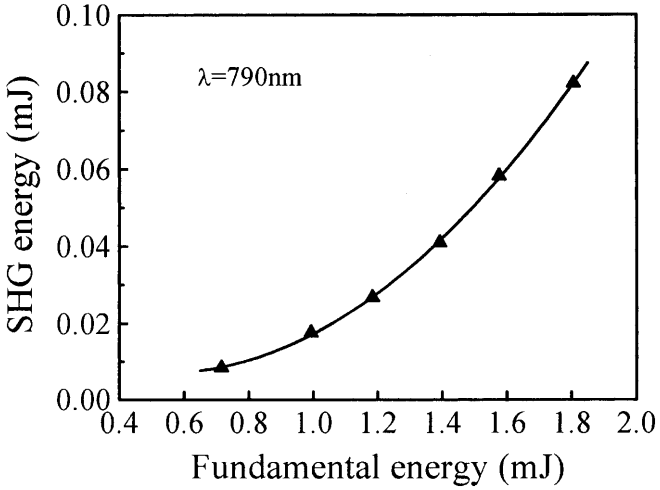

Fig. 5. Measured SHG pulse energy as a function of the single pulse energy of the fundamental light

Figure 5 shows the harmonic energy dependence on the fundamental energy at the optimum wavelength of 790 nm. The SHG energy is nearly proportional to the square of the fundamental energy. This square relationship indicates that no photorefractive damage takes place. When the fundamental energy is 1.8 mJ, a 395 nm violet light with a single-pulse energy of 0.08 mJ is obtained. The measured conversion efficiency is 4.4%. Comparing with the theoretical efficiency of 45.17%, which is calculated according to the incident power and the spot size in the crystal under the plane-wave model and the nondepletion approximation [16], the efficiency is not high. We think that this is mainly due to the period fluctuation, the uncoated light-transmitting facet and the crystal absorption of the fundamental and second-harmonic light. Furthermore, the nondepletion approximation will also make the calculated efficiency higher than the experimental result even if the influence of the period fluctuation is considered.

4 Conclusions

In summary, the periodically poled Nd:MgO:$LiNbO_3$ crystal was successfully fabricated in our laboratory. The absorption and fluorescence spectra show some differences from common Nd:MgO:$LiNbO_3$ with monodomain structure. SHG measurement indicates that this material is suitable for SFD operation using the high-gain π polarization at room temperature. In addition, the blue and green SFD operation were both achievable in optical superlattice Nd:MgO:$LiNbO_3$ which is impossible in common Nd:MgO:$LiNbO_3$. Further works in our laboratory are intended to improve the crystal quality by optimizing the crystal growth technics or using the electric-field poling method and to construct a prototype device by diode-laser pumping.

References

1. G. Zhong, J. Jian, Z. Wu: Proceedings of the 11th International Quantum Electrics Conference, IEEE Catalog No. 80CH 1561-0 (Institute of Electronics Engineers, New York 1980) p. 631
2. T.Y. Fan, A. Cordova-Plaza, M.J.F. Digonnet, R.L. Byer, H.J. Shaw: J. Opt. Soc. Am. B **3**, 140 (1986)

3. A. Cordova-Plaza, T.Y. Fan, M.J.F. Digonnet, R.L. Byer, H.J. Shaw: Opt. Lett. **13**, 209 (1988)
4. N. Mackinnon, C.J. Norrie, B.D. Sinclair: J. Opt. Soc. Am. B **11**, 519 (1994)
5. R. Li, J. Wang, X. Liang, C. Xie, K. Peng: Chin. Phys. Lett. **10**, 231 (1993)
6. A. Armstrong, N. Bloembergen, J. Ducuing, P.S. Pershan: Phys. Rev. **127**, 1918 (1962)
7. D. Feng, N.B. Ming, J.F. Hong, Y.S. Yang, J.S. Zhu, Z. Yang, Y.N. Wang: Appl. Phys. Lett. **37**, 607 (1980)
8. G.A. Magel, M.M. Fejer, R.L. Byer: Appl. Phys. Lett. **56**, 108 (1990)
9. Y.L. Lu, L. Mao, S.D. Cheng, N.B. Ming, Y.T. Lu: Appl. Phys. Lett. **59**, 516 (1991)
10. Y.Q. Lu, Y.L. Lu, G.P. Luo, X.F. Cheng, C.C. Xue, N.B. Ming: Electron. Lett. **32**, 336 (1996)
11. Y.Q. Lu, Y.L. Lu, C.C. Xue, J.J. Zheng, X.F. Chen, G.P. Luo, N.B. Ming, B.H. Feng, X.L. Zhang: Appl. Phys. Lett. **69**, 3155 (1996)
12. W.K. Burns, W. McElhanon, L. Goldberg: IEEE Photo. Tech. Lett. **6**, 252 (1994)
13. H. Loro, M. Voda, F. Jaque, J. Garcia Sole, J.E. Munoz Santiuste: J. Appl. Phys. **77**, 5929 (1995)
14. M.M. Fejer, G.A. Magel, D.H. Jundt, R.L. Byer: IEEE J. Quantum Electron. **QE-28**, 2631 (1990)
15. G. Xu, M. Gong, Y. Guo, J. Chen, B. Li, G. Zhai, K. Han: Chin. J. Lasers **18**, 324 (1991) (in Chinese)
16. A. Yariv, P. Yeh: *Optical Waves in Crystals* (Wiley, New York 1984) pp. 519–528