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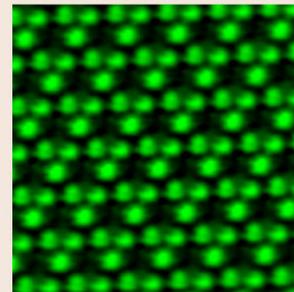
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Femtosecond violet light generation by quasi-phase-matched frequency doubling in optical superlattice LiNbO₃

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We report the 390 and 385 nm violet light generation by frequency doubling of a tunable 90 fs, 82 MHz Ti:sapphire laser in two optical superlattice LiNbO₃ samples through third-order quasiphase matching (QPM). With the average incident infrared light power of 770 mW, 9.7 and 2.9 mW output second-harmonic lights were obtained without photorefractive damage. The QPM wavelength acceptance bandwidth measurement indicates that violet light generation with higher efficiency is possible. © 1996 American Institute of Physics. [S0003-6951(96)04347-1]

With the development of the mode-locked femtosecond Ti:sapphire laser,¹ the generation of ultrashort pulses with high repetition rate has become possible in the near-infrared wavelength range.² However, for many applications, for example, experiments on molecules and wide-gap semiconductors, pulses at shorter wavelengths are required. Second-harmonic generation (SHG) in nonlinear optical crystals including LiNbO₃ and BBO represents a powerful technique to convert infrared pulses to the ultraviolet-blue spectral range. Up to date, a variety of frequency conversion schemes has been proposed and efficient blue lights generated. Among them, there are the directly frequency doubling,³ intracavity,⁴ and extracavity⁵ SHG. However, all these schemes are based on the conventional phase-matching (PM) technique. To the best of our knowledge, the quasiphase-matching (QPM) technique⁶ which is achieved in optical superlattice crystals (i.e., crystals with periodic nonlinear optical coefficient modulation) has not been applied in such an ultrashort pulse frequency conversion area. As the QPM technique allows frequency doubling at room temperature over the crystal's entire transparency range using the largest nonlinear coefficient, the utility of a single material can be extended. For example, in LiNbO₃, due to the insufficiency of birefringence, PM condition cannot be satisfied when the fundamental wavelength is shorter than 1 μm , thus, the generation of blue or violet light is impossible; The maximum nonlinear coefficient d_{33} , which is about 7.5 times larger than the commonly used d_{31} cannot be used. Whereas in optical superlattice LiNbO₃ short wavelength light generation used d_{33} is possible, we may expect a QPM enhancement factor of $(d_{33}/d_{31})^2(2/\pi)^2 \approx 23$ in the harmonic generation process.⁷ In our previous works, we have demonstrated the efficient blue or violet light generation by directly frequency doubling the CW diode laser or picosecond pulsed

optical parametric oscillator.⁸⁻¹¹ For femtosecond short-wavelength light generation, the high peak power intensity of femtosecond infrared pulses must make the process of QPM SHG more efficient. On the other hand, the femtosecond pulses broadening arisen from the group-velocity mismatch (GVM) is a severe problem in the harmonic generation process, which requires a nonlinear crystal with high nonlinearity and shorter interaction length. Obviously, the QPM technique allows obtaining the same harmonic generation efficiency in a much thinner crystal, so the pulse broadening effect could be weakened. Thus, the optical superlattice crystal has great potentials in ultrashort harmonic generation. Furthermore, the possible higher photorefractive damage threshold caused by the periodic domain structure¹² is also beneficial for SHG of the high-frequency femtosecond Ti:sapphire amplifier.

In this letter we report the efficient femtosecond violet light SHG in two optical superlattice LiNbO₃ samples through third-order QPM by using a tunable femtosecond Ti:sapphire laser with high repetition rate. The QPM acceptance bandwidth was measured with a pulsed dye laser to analyze the experimental results.

An argon-pumped 90 fs, 82 MHz Ti:sapphire laser (Tsunami: Spectra-Physics) with a linewidth of about 8.0 nm was used as the fundamental light source. The diameter of the output infrared beam was 3 mm. After propagating through two convex lenses with their focusing lengths of 10 and 5 mm, the beam diameter was compressed to 1.5 mm and then normally injected into the sample. An infrared-cut filter was used to block the fundamental light and then let the passed harmonic light directly enter the detector.

Two optical superlattice LiNbO₃ samples grown by the Czochralski method as described earlier¹³ were selected for the experiments. Their average modulation periods are 7.2 μm (sample 1) and 6.9 μm (sample 2) with the period fluctuation of 4.0% and 3.5%, respectively. Their thicknesses are

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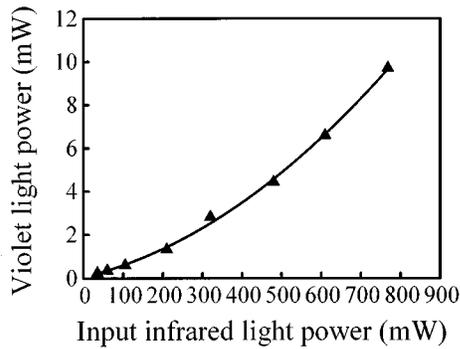


FIG. 1. The output power of the 390 nm violet light as a function of the fundamental light power in sample 1.

2.8 and 0.8 mm. Each sample has a $3 \times 3 \text{ mm}^2$ ($b \times c$) light-transmitting face with the fundamental beam propagated in the uncoated a face. The polarization direction of the fundamental wave is along the c axis, just for satisfying the QPM condition.

After tuning the output wavelength of the Ti:sapphire laser, the optimum QPM wavelength of sample 1 was found to be 780 nm. Figure 1 shows the dependence of the output power of the 390 nm violet light on the input infrared light power with consideration of the violet light loss of the filter. Along with the increasing of the input power, the violet light power increases as a square relationship. When the infrared light power went up to 770 mW, 9.7 mW violet light generation was obtained without photorefractive damage in the crystal. The SHG conversion efficiency was 1.3%.

Figure 2 shows the experimental results in sample 2. The optimum fundamental light is at 770 nm and the 385 nm violet light was obtained. With the 770 mW incident infrared light, 2.9 mW violet light was generated, corresponding to the conversion efficiency of 0.4%. Although the thickness of this sample is only 0.8 mm and the QPM theory predicts that the conversion efficiency is proportional to the square of the sample thickness, from our experimental results,

$$(\eta/L^2)_{\text{sample 2}} : (\eta/L^2)_{\text{sample 1}} = 3.7:1 > 1,$$

where η is the conversion efficiency and L is the sample thickness. Furthermore, the thinner sample thickness of sample 2 also made the GVM smaller, so sample 2 is more suitable for the femtosecond light SHG. However, the reasons why the theory prediction has so large a deviation are

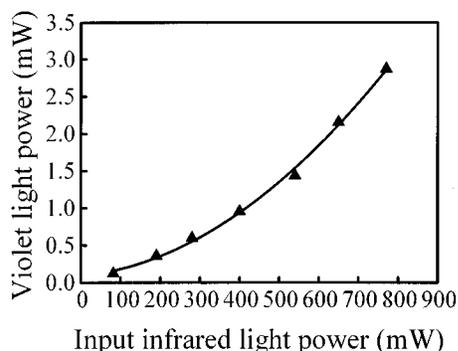


FIG. 2. The output power of the 385 nm violet light as a function of the input infrared light power of 770 nm infrared light in sample 2.

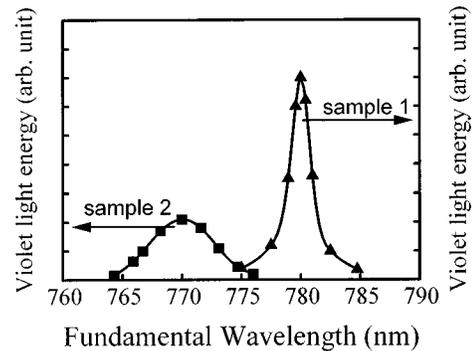


FIG. 3. Second-harmonic light energy as a function of the fundamental wavelength in sample 1 and sample 2 measured by a pulsed dye laser.

complicated. We think that the better periodicity of sample 2 might be an influential factor, but the main reason must have something to do with the different wavelength acceptance bandwidth of the samples.

A tunable dye laser (model ND6000: Continuum Co.) with a pulsewidth of 8 ns and linewidth of 0.2 cm^{-1} was used as the fundamental light source for measuring the acceptance bandwidth. The experimental setup was similar to the femtosecond frequency doubling situation. Figure 3 shows the relationship between the energy of the generated violet pulse and the fundamental wavelength. The measured fundamental wavelengths with the highest conversion efficiency are also 780 nm (sample 1) and 770 nm (sample 2), which are the same as the results of the femtosecond SHG. From the figure, the wavelength acceptance bandwidth [full width at half-maximum (FWHM) of the wavelength tuning curve] of sample 1 is about 2.0 nm, which is greatly larger than the theoretical prediction of less than 1 nm according to Eq. (30) in Ref. 7. As the equation is based on the ideal QPM condition (only consider the perfect periodic structure), the period fluctuation must be the major reason to the bandwidth expansion. Furthermore, the thickness of sample 1 is 3.5 times as much as that of sample 2 and their QPM wavelengths are nearly the same, the acceptance bandwidth of sample 2 should be also nearly 3.5 times as much as that of sample 1 according to the equation,⁷ but the measured acceptance bandwidth of sample 2 was 6.4 nm, which is not in good accordance with the theoretical prediction. In fact, since the fundamental wavelength, the sample thickness, and the period fluctuation are all able to influence the wavelength acceptance bandwidth and even the detailed functional spatial form of the periodic fluctuation does something with the acceptance bandwidth, obtaining the acceptance bandwidth of a sample numerically is very difficult. However, whether in the case of sample 1 or sample 2, the measured acceptance bandwidth is smaller than the linewidth of the Ti:sapphire laser, thus, only a part of the fundamental light could be used for frequency doubling and a lot of fundamental light power was wasted. So the linewidth of the fundamental light and the acceptance bandwidth are two important factors for our consideration in the femtosecond frequency conversion process. Only if the linewidth is smaller than the sample's acceptance bandwidth can higher conversion efficiency be obtained. As we know, the femtosecond pulse usually has a wider linewidth, so it is beneficial for increasing the wave-

length acceptance bandwidth of a sample. It is just the wider acceptance bandwidth of sample 2 that makes the $(\eta/L^2)_{\text{sample 2}}$ larger. To date, there have been some attempts to increase the QPM acceptance bandwidth, for example, using optimized domain grating¹⁴ or the variable-spaced phase reversal technique.¹⁵ Comparing to the unchangeable and the narrow acceptance bandwidth in the conventional PM SHG process, the wide QPM bandwidth and its tunability seem very attractive. We hope that future acceptance bandwidth controlling techniques will make the QPM femtosecond SHG more efficient.

In summary, we first demonstrated the femtosecond pulsed violet light SHG in optical superlattice LiNbO₃ crystals through third-order QPM. 9.7 mW 390 nm and 2.9 mW 385 nm lights were obtained. The conversion efficiencies were 1.3% and 0.4%, respectively. The acceptance bandwidth measurement indicated that SHG with higher conversion efficiency is possible.

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