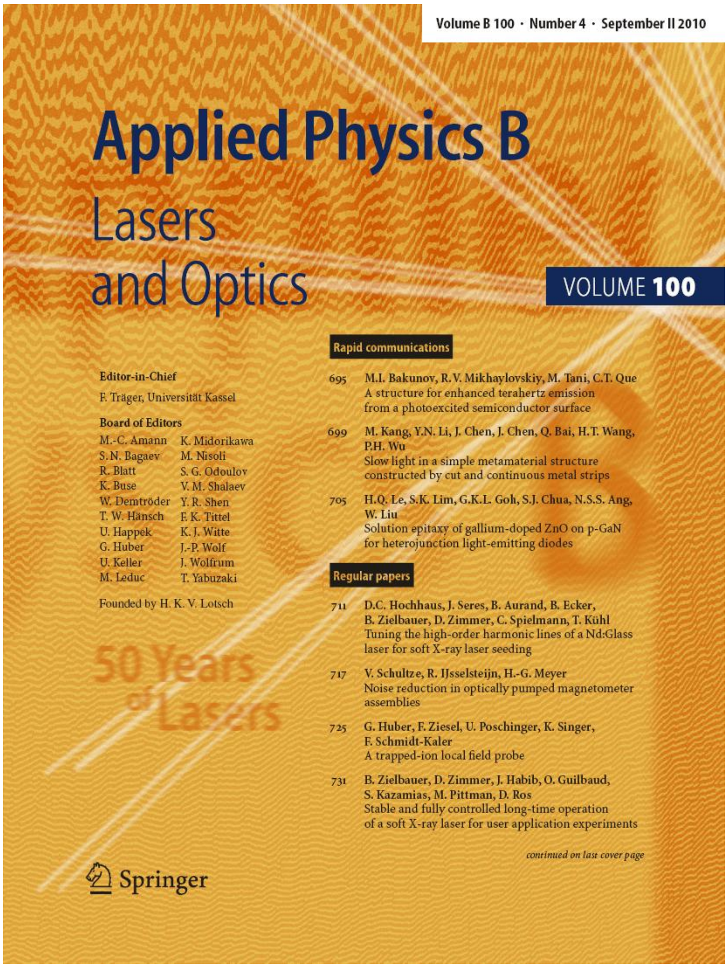
ISSN 0946-2171, Volume 100, Number 4



Volume B 100 · Number 4 · September II 2010

Applied Physics B

Lasers and Optics

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Appl Phys B (2010) 100: 755–758
DOI 10.1007/s00340-010-4047-1

Applied Physics B
Lasers and Optics

Non-collinear efficient continuous optical frequency doubling in periodically poled lithium niobate

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Received: 9 December 2009 / Revised version: 9 March 2010 / Published online: 6 May 2010

Abstract Non-collinear continuous frequency doubling with relatively high conversion efficiency is demonstrated in periodically-poled lithium niobate. It is shown that about 80% of the total second harmonic energy can be collected to one output channel by tuning incident angle. The fractional phase-matching condition is introduced to interpret the phenomenon. These results make it possible for direct power applications of continuous frequency doubling over the broad spectral range from the visible to the infrared without temperature or angle tuning.

1 Introduction

Second-harmonic generation (SHG) has been widely used in modern physics research. Efficient SHG is generally achieved through the crystal birefringence [1] or the quasi-phase-matching (QPM) technique [2], in which processes the wave vectors of fundamental frequency (FF) and second harmonic (SH) beams are collinear. Since 2000, 2-D artificially created non-linear optical superlattices [3–6] have been widely reported, which make great progress with multidirectional and multi-QPM SHG processes [7]. However, QPM technique always requires strict phase-matching (PM) conditions, resulting in its disadvantages of narrow non-linear-response bandwidth and sensitivity to the period of the QPM grating and temperature. The complex fabrication techniques and the expensive cost also prevent wide applications of 2-D non-linear optical superlattice. As a potential substitution, a new type of random material, for example the SBN crystal [8–13], has been intensively studied in the last decade for its new characteristics of broadband spectrum response [9, 11] and linear growth of SH intensity with the crystal length [14, 15]. The most fascinating feature of the SBN crystal is the relaxation of the phase-matching (PM) condition resulting from the randomly-distributed needle-like antiparallel domains. Nevertheless, such structure causes the SH beams to radiate in a cone with low conversion efficiency, decreasing its directivity and power applications. Till now, only signal analysis has been reported in SBN crystals [11]. It seems to be "impossible for power applications using this phenomenon" [8]. Generally speaking, efficient, PM-condition-relaxed, broadband SHG solutions have not been reported yet.

In this paper, we report an efficient SHG process, with both advantages of 2-D non-linear optical superlattices and random materials, in a simply structured 1-D non-linear grating, in which process a multidirectional, PM-condition-relaxed, continuous optical frequency doubling occurs. Systematically investigation shows that a new SHG mechanism should be introduced in our experiment. Relatively high conversion efficiency is obtained and single output beam with most of the SH energy is proved by easily tilting the incident beam. These results will open novel possibilities for efficient, PM-condition-relaxed, broadband SHG in a simply structured conversion crystal without temperature or angle tuning. They also provide a method to double femtosecond pulses efficiently.

2 Experimental setup

Figure 1 is the schematic of our experiment and the SH patterns on the screen. A z-cut $20 \times 5 \times 0.5$ mm^3 PPLN sample with a period of 30 μm and a duty ratio 1:1 is used as

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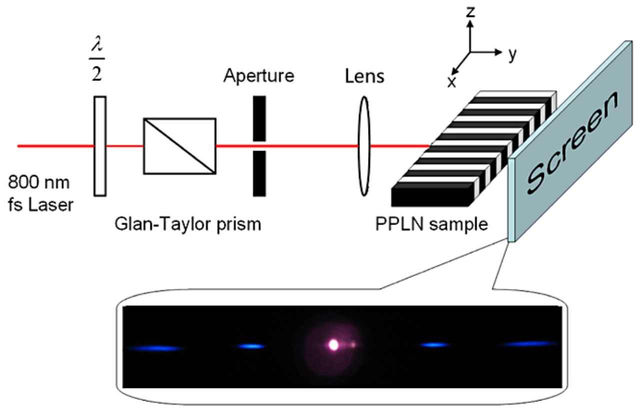

Fig. 1 Experimental setup and the SH patterns on the screen

the conversion crystal. The light source is a Ti: Sapphire oscillator producing about 100 fs pulses at an 84 MHz repetition rate at wavelength 800 nm and full width half maximum (FWHM) pulse energies of about 3.5 nJ which provides FF with bandwidth over 30 nm. A half-wave plate and a Glan–Taylor prism are employed to adjust the FF power and its polarization. The FF beam is directed along the domain wall and loosely focused by a 100 mm focal length lens, resulting in a focal spot of about 0.1 mm. A 0.1 mm wide slit is used to measure the spectrum and the intensity of the SH at different angles with respect to the y-axis in the $x-y$ plane.

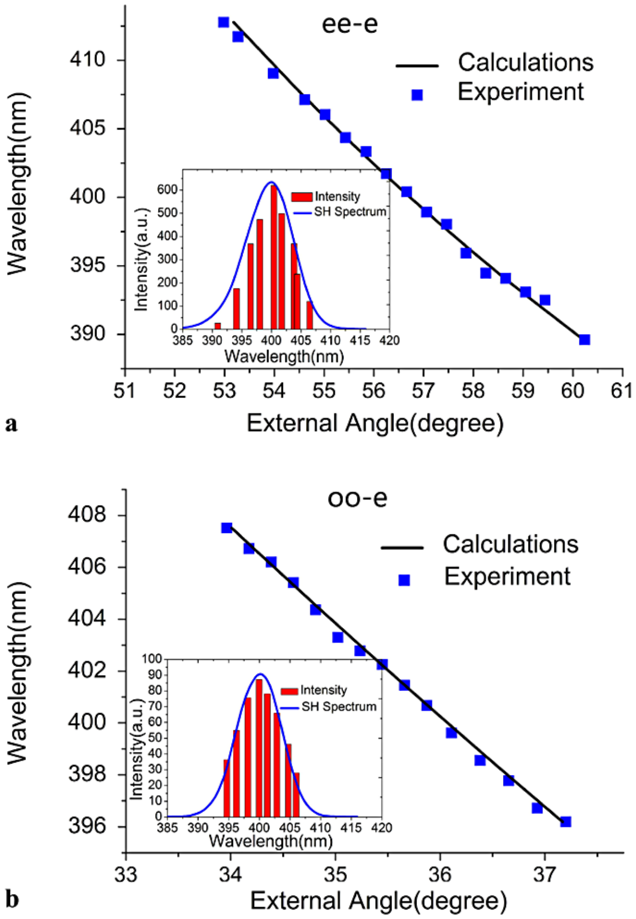

Fig. 2 The dependence of the SH wavelengths on the diffracting angles in (**a**) ee-e process and (**b**) oo-e process. *The insets* are the corresponding SH spectra and the intensity distributions with wavelengths in each process

3 Results and discussion

See the SH patterns in Fig. 1, the central red spot is the transmitting FF beam. Two pairs of SH beams are generated. The outside pair is generated by the e-polarized component of FF through d_{33} and the inner pair is generated by the o-polarized component of FF through d_{31}. Both pairs of the SH beams are e-polarized. They have good beam qualities after a relatively long interacting distance of 5 mm in the PPLN, compared with the short 0.5 mm interacting distance in earlier studies [16, 17]. It can be seen from the screen that the SH beams broaden themselves in the $x-y$ plane while remaining their initial dimensions in the $y-z$ plane. Such a broadening mechanism has not been mentioned in previous studies. In order to study this broadening mechanism, precise angular measurements of spectrum and intensity were made with the 0.1 mm slit. The situations of e-polarized FF and o-polarized FF are identified as ee-e and oo-e processes respectively. The results are shown in Fig. 2. Solid lines are theoretical calculations which will be discussed later. It is obvious that different components of the SH beams diffract at different angles. The larger the SH component's wavelength is, the smaller the diffracting angle is. The inserts in Fig. 2 are the intensity distributions of some SH components compared with the total SH spectrum profiles. Each frequency component of the FF beam has been doubled efficiently.

Similar to that in SBN crystal, broadband SHG is achieved in our experiment, but the mechanism seems quite different. In SBN crystals, randomly sized and distributed domains provide various reciprocal vectors [8], which compensate for the phase mismatch of all components of the FF. In our PPLN sample, the period is fixed and only a unique reciprocal vector is provided. As shown in Fig. 3(a), $\mathbf{G}_1$ is the reciprocal vector provided by the PPLN. This fixed reciprocal vector $\mathbf{G}_1$ can only fulfill the PM condition for a special component $\mathbf{k}_1$ of the FF in the SHG process. If another component $\mathbf{k}_1'$ of the FF can be doubled efficiently, another reciprocal vector $\mathbf{G}_2$ must be required, which cannot be provided by the same PPLN. So a fulfilled PM condition is not suitable for the broadband SHG process in our experiment. New explanations should be suggested. Two types of fractional PM conditions are appropriate here, the Cherenkov phase-matching condition resulting from the non-linearity existing in domain walls [16] and the non-

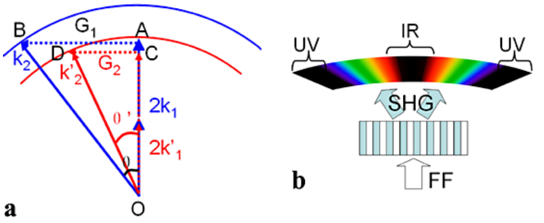

Fig. 3 (**a**) Non-integer order phase-matching condition in which the reciprocal vectors G1 and G2 do not need to be integer order; and (**b**) schematic of a pair of symmetric "Rainbow" generated from FF beam with ultra-wide spectrum

integer order PM condition only in non-linear optics [17]. Both of them indicate a longitudinal PM (LPM) condition that only the FF and SH vectors are required to fulfill the vector relation in the triangle in Fig. 3(a). By the mechanism of this LPM condition, all the frequency components of the FF will be doubled efficiently. The SH diffracting angle is determined by $\cos\theta = 2k_1/k_2$, a direct derivation from LPM condition. The calculation results are the solid lines in Fig. 2, in good agreement with the experimental results. If a FF beam has ultra-wide spectral width, the SH beams of different wavelengths will diffract continuously at different angles and form a pair of beautiful "Rainbows" as shown in Fig. 3(b). If the FF beam is monochromatic, the SH beam should be well collimated, which could be considered as good SH source. Thus continuous frequency doubling by the LPM condition is demonstrated in PPLN. The converting spectral range is determined by the transparency window of lithium niobate, from visible to infrared. Temperature has little influence on this SHG process in PPLN, because the LPM condition is always satisfied.

The conversion efficiency is important because it determines whether this phenomenon in our experiment has potential power applications. For most of non-collinear SHG processes, the conversion efficiencies are rather low, always less than 10^{-3}, which is impossible for power applications. In our experiment, ee-e and oo-e processes are measured separately and the results are shown in Fig. 4. There are two channels for the SH beams. When the FF is e-polarized and the injecting power is 225 mW, we get the SH conversion efficiency of about 0.65% in one channel. The total SH conversion efficiency should be 1.3%, which is relatively high for such non-collinear SHG processes. When the FF is o-polarized, the total conversion efficiency is lower, of about 0.6%. This is because d_{33} is larger than d_{31} in lithium niobate crystals. The inset in Fig. 4 gives the quadratic relationship between the SH and FF powers. From Fig. 4, we also find that the SH conversion efficiency increases with the FF power linearly. It should be noted that because the half-wave plate and the Glan–Taylor prism cause pulse energy loss when we rotate the FF polarization, the SH beams are only generated when the FF power is larger than 150 mW instead of 0 mW. This relatively high conversion efficiency, which we think is the highest reported so far, will open possibilities for direct power applications of SHG in such configurations. Through this continuous frequency doubling, femtosecond pulses can be doubled efficiently and the SH pulse can be reconstructed by the use of optical gratings.

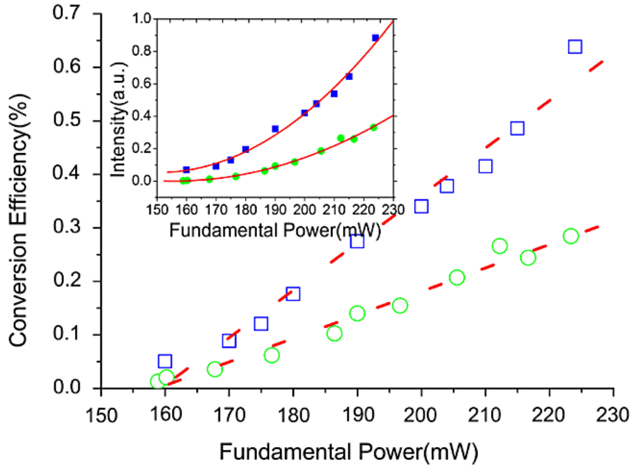

Fig. 4 SH conversion efficiency as a function of the FF power which is fitted linearly. *The inset* is the SH intensity changing with the FF power which is fitted to be quadratic. *The blue dots* are experimental data in the ee-e process and *the green dots* in the oo-e process

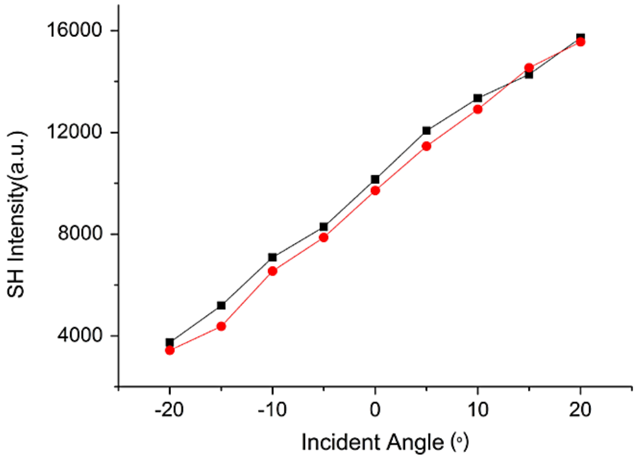

Fig. 5 Intensity of one SH beam of the pair changes with the incident angle with respect to the y-axis

Although the total SH energy is divided into two channels, we find in the experiment that by tuning the incident angle in x–y plane, we can collect most of the SH energy into one channel. This phenomenon has not been reported in any previous researches. Experimental results of ee-e process are shown in Fig. 5. When the incident angle is tuned from −20° to 20°, the SH energy in one channel increases linearly with the angle. In this tuning process, up to 80% of the total SH energy is collected in this channel, which can maximize the utilization of the total SH energy. Moreover, according to either the Cherenkov phase-matching theory or the non-integer order phase-matching theory, we predict an increase of conversion efficiency when the period of the PPLN decreases. Poling technology is rather mature today that $\chi^{(2)}$ non-linear gratings with sub-micron period could be produced [18], which would significantly increase the conversion efficiency.

4 Conclusion

In conclusion, we have demonstrated efficient continuous frequency doubling in simply structured 1-D PPLN with the converting range from visible to infrared. Fractional PM condition is discussed as a different mechanism from that of the SHG in 2-D non-linear superlattices and SBN crystals. A relatively high conversion efficiency is observed in our experiment, and most of the SH energy can be collected in one output channel by tuning the incident angle. We believe these results will pave the way for research on multidirectional, PM-condition-relaxed, broadband SHG and open novel possibilities for direct power applications of efficient continuous frequency conversion in a single simply structured conversion crystal without temperature or angle tuning.

Acknowledgements This research was supported by the National Natural Science Foundation of China (No. 10734080 and No. 10574092); the National Basic Research Program “973” of China (No. 2006CB806000), and the Shanghai Leading Academic Discipline Project (B201).

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