Cascaded third-harmonic-generation converter based on a single ADP crystal*

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Based on a specially designed optical structure, an efficient cascaded third-harmonic-generation (THG) output of a 1064-nm, pico-seconds pulse laser is successively realized by using an $NH_4H_2PO_4$ (ADP) crystal that acts as the second-harmonic-generation component and sum-frequency-generation component. The maximum THG output is 1.61 mJ, and the highest conversion efficiency from 1064 nm to 355 nm reaches 35%, which are obviously superior to the results obtained using a KDP crystal under the same circumstance. The further phase-matching analysis indicates that this THG configuration of ADP crystal can be applied to various fundamental wavelengths in a range of 1 μm–1.1 μm. Compared with the previously reported KDP THG converter, which is based on a similar principle, the present ADP THG converter is favorable for large-energy, high-efficiency operation because of the larger effective nonlinear optical coefficient $d_{\rm eff}$ and higher laser damage threshold.

Keywords: third-harmonic-generation, ADP crystal, polarization, ultraviolet laser

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1. Introduction

Nowadays, with the help of nonlinear optical (NLO) materials, harmonic generation of near infrared laser at 1-μm waveband has been an important method to obtain ultraviolet (UV) coherent light. This method includes several different routes, i.e., direct third-harmonic-generation (THG) based on third-order nonlinearity,[1,2] quasi-phase-matching (QPM) from a quasi-periodic optical superlattice (QPOS),[3–7] and cascaded THG based on birefringent phase-matching (BPM).[8–13] For the former two schemes, the UV light beam can be obtained simply by using a single NLO medium. However, the third-order nonlinear coefficients of NLO materials are too small, which restricts the practicability of direct THG, and the QPOS technique is plagued by its complicated preparation processes, high price, and low damage threshold. Therefore, currently, the most popular and efficient approach to obtaining a solid-state UV laser source, i.e., the BPM cascaded THG, is still the third route. It includes two steps: a phase-matching (PM) second-harmonic-generation (SHG) process, followed by a PM sum-frequency-generation (SFG) process. Correspondingly, two independent NLO materials are required. For some special NLO crystals and special wavelengths, the PM directions of SHG and SFG are identical or very adjacent. Even in these occasional cases, two blocks of NLO crystals are still essential because the SHG (2ω) polarization generated by the SHG process is usually different from the 2ω polarization required by the SFG process.

Recently, we reported a novel single crystal BPM cascaded THG converter, which has been successfully applied to KDP crystal.[14] This is a kind of design based on polarization optics.[15–22] For 1064 nm, 40 ps, 10-Hz fundamental laser pulses, the maximum THG output was 1.13 mJ, and the highest overall conversion efficiency was 30.7%.

As a traditional NLO crystal, ammonium dihydrogen phosphate ($NH_4H_2PO_4$ or ADP) possesses a similar structure, linear and nonlinear optical properties to KDP crystal, which can be used as 2ω, 3ω, and 4ω harmonic generators of 1-μm lasers.[23–28] Compared with KDP crystal, ADP has larger effective NLO coefficient $d_{\rm eff}$ ($\sim$ 1.1 times that of KDP) and higher laser damage threshold ($\sim$ 2 times that of KDP). In addition, ADP is more suitable for rapid growth with high optical quality than KDP.[29] In this work, by utilizing our single crystal BPM cascaded THG technique, efficient THG conversion of 1064 nm is demonstrated in ADP crystal. The maximum UV output and the highest THG conversion efficiency are 1.61 mJ and 35%, respectively, which are obviously su-

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perior to the results of KDP crystal.[14] Such an outstanding performance can be attributed to the larger NLO coefficient d_{eff} and the ability to resist the laser damage to ADP crystal, and exhibits a promising application foreground.

2. Design principle

Figure 1 shows the work principle of the single crystal BPM cascaded THG converter. A quarter-wave plate (QWP) of 2ω wavelength is used to adjust the polarization of the SHG wave, and a round-trip optical path is used to realize the SHG and SFG processes in one bulk NLO crystal, successively. In this design, the linear polarization of the fundamental wave, the optical axis of the QWP and the bisection plane of the o-light and the e-light of the NLO crystal are adjusted to be parallel to each other. In this way, the QWP does not rotate the fundamental wave (ω) polarization in the whole process, whereas after the forward and backward passing through the QWP twice, the linear polarization of the 2ω wave generated by type-II SHG will be rotated 90° and change e-light into o-light, because the total phase delay is π, or $\lambda/2$. In the process of the backward propagation, the o-polarized 2ω wave interacts with the e-polarized component of the residual ω wave in the NLO crystal (type-II SFG, $o_{2\omega} + e_{\omega} \rightarrow e_{3\omega}$). As a result, the 3ω wave can be obtained from the output end. This configuration retains the advantages of the traditional BPM cascaded THG, like high optical conversion efficiency and high damage threshold, and brings some new merits, such as adjustable working wavelength and reduced production cost due to saving an NLO crystal, at the same time. In fact, such a design can be applied to other cascaded frequency up-conversion processes, or other NLO crystals, such as DKDP, DADP, and BIBO, as long as the relevant cascaded frequency up-conversion processes occur at similar crystal orientations.

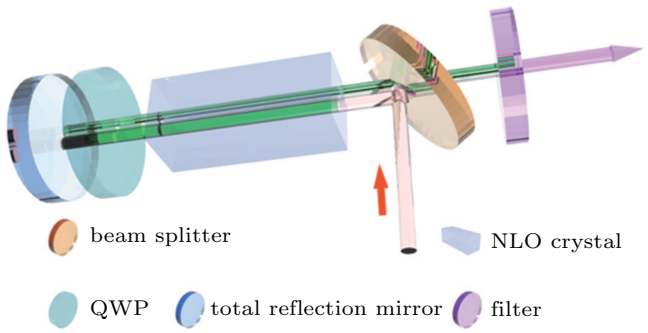


Fig. 1. (color online) Schematic diagram of the single crystal BPM cascaded THG converter.

3. Theoretical analysis

Based on the refractive index dispersion equation of the ADP crystal,[30] the PM angles of type-II SHG and type-II THG are calculated for different fundamental wavelengths at 1-μm waveband (1000 nm–1100 nm). The corresponding tuning curves of PM angles theta (θ) are shown in Fig. 2, and some representative data are listed in Table 1. It should be noted that to make d_{eff} optimized, the PM angle phi (ϕ) of type-II NLO frequency conversion processes are always kept at 0° in ADP crystal. From Fig. 2 and Table 1, it can be found that in this waveband the PM directions of type-II SHG and type-II THG are very close and about at ($\theta = 60°$, $\phi = 0°$). The largest θ angle deviation is only 3.5°, which appears at 1100 nm. These calculation results indicate that for the THG of various wavelengths at 1 μm∼ 1.1 μm waveband, ADP is available for the single crystal BPM-cascaded THG structure mentioned above.

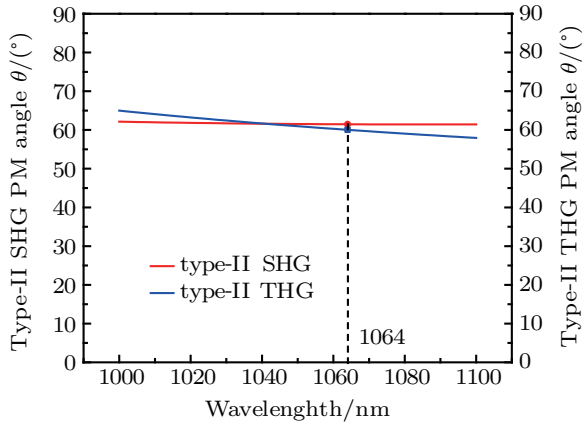


Fig. 2. (color online) Tuning curves of type-II SHG and type-II THG PM angles theta (θ) versus wavelength in a range of 1000 nm–1100 nm in ADP crystal.

For the common Nd:YAG, Nd:YVO_4 laser (1064 nm), the type-II SHG and type-II THG PM angles are (61.5°, 0°) and (60.0°, 0°) respectively, corresponding to a deviation of 1.5°, which is similar to the value of KDP crystal (0.8°). At the same time, ADP possesses larger d_{eff} and higher laser damage threshold than previously reported KDP crystal, as shown in Table 2.

Table 1. Representative data in Fig. 2.

Fundamental wavelength	1000 nm		1030 nm		1053 nm		1064 nm		1080 nm		1100 nm	
NLO process	SHG	THG	SHG	THG	SHG	THG	SHG	THG	SHG	THG	SHG	THG
$\theta/(°)$	62.1	65.0	61.7	62.4	61.5	60.8	61.5	60.0	61.4	59.1	61.4	57.9

Table 2. Comparison between SHG and SFG parameters for THG output of 1064 nm in ADP and KDP crystals.

Crystal	Phase-matching type	Phase-matching scheme	Phase-matching angles[30]	Effective NLO coefficient d_{eff} (pm/v)[31]	Laser damage threshold
ADP	type-II SHG	$o_\omega + e_\omega \rightarrow e_{2\omega}$	$\theta = 61.5^\circ$, $\phi = 0^\circ$	0.39	1.6 GW/ cm^2
	type-II SFG	$o_{2\omega} + e_\omega \rightarrow e_{3\omega}$	$\theta = 60.0^\circ$, $\phi = 0^\circ$	0.40	(355 nm, 7 ns)[26]
KDP	type-II SHG	$o_\omega + e_\omega \rightarrow e_{2\omega}$	$\theta = 59.1^\circ$, $\phi = 0^\circ$	0.35	0.7 GW/ cm^2
	type-II SFG	$o_{2\omega} + e_\omega \rightarrow e_{3\omega}$	$\theta = 58.3^\circ$, $\phi = 0^\circ$	0.35	(355 nm, 7 ns)[32]

4. Experiment and results

The ADP crystal used for the laser experiment was grown by the point-seed rapid growth method from aqueous solution. The experimental sample was processed along (61.5°, 0°), i.e. its type-II SHG direction of 1064 nm, with dimensions of 20 mm×20 mm×9 mm. Its transmission end faces were optically polished but uncoated.

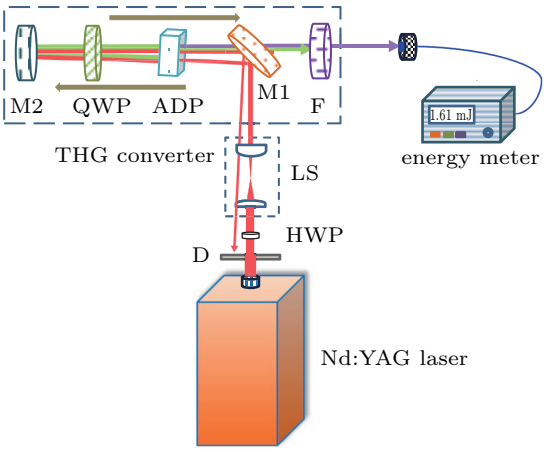


Fig. 3. (color online) Experimental setup of the single ADP crystal BPM cascaded THG converter.

The experimental setup is shown in Fig. 3, and the ambient temperature was 25 °C. The fundamental light source was a mode-locked Nd:YAG laser operating at 1064 nm, with an original beam diameter of 8 mm, a pulse width of 40 ps, and a repetition rate of 10 Hz. After a diaphragm (D), the diameter of fundamental light was reduced to 4 mm, accompanied with improving beam quality. A 1064-nm half-wave plate (HWP) was used to adjust the linear polarization of the fundamental laser to be parallel to the bisection plane of the *o*-light and the *e*-light of the ADP crystal. In this manner, the type-II SHG process could be realized. Meanwhile, the polarization requirements of this converter were satisfied. A lens system (LS) that was composed of two plane-convex lens ($f = 100$ mm and 50 mm, respectively) was used to compress the fundamental beam diameter to a half to elevate the energy density. Then the fundamental beam entered into the THG converter that consisted of a beam splitter M_1 (45° HR@1064 nm & 45° HT@355 & 532 nm), an ADP crystal, a 532-nm mica QWP, a total reflection mirror M_2 (HR@1064 & 532 nm) and a filter (F) (HT@355nm & HR@532 nm). The beam splitter M_1 was placed at ∼ 45° inclination angle with respect to the central main light path, and the optical axis of the QWP was adjusted to be parallel to the polarization of the fundamental wave,and the bisection plane of the *o*-axis and the *e*-axis of the ADP crystal. The distance between the ADP crystal and the M_2 mirror was about 15 mm. In this experiment, the M_1 was highly transmitted for the 2ω and 3ω waves ($T > 99\%$ at 532 nm and 355 nm) and highly reflected for the fundamental wave ($R > 99\%$ at 1064 nm), which are basically opposite to the M_1 used in Ref. [14]. The residual fundamental laser after the SFG process will be reflected by M_1 rather than transmit through it. In this way, the propagation direction of the residual fundamental laser can be easily controlled by M_1, and make the backward optical path of the reflective fundamental laser separated from the forward optical path of the incident fundamental laser, which can better avoid being interfered by the returned beam to the fundamental light source.

During the experiments, the inclined angles of the ADP crystal and M_2 were finely tuned to reach a maximum THG output. Because the PM angles of type-II SHG and type-II THG have a discrepancy of 1.5°, i.e., an exterior angle discrepancy of ∼ 2.3°, the forward and the backward optical paths were not overlapped as shown in Fig. 3. The residual fundamental laser after the SFG process was reflected by M_1, but not along the original path. In the transmitted light beams of M_1, the 3ω laser was selected out by the filter F from the unconverted 2ω laser and detected by an energy meter.

The THG output and overall conversion efficiency from 1064 nm to 355 nm are demonstrated in Fig. 4. The inset displayed a THG facula. Considering the fundamental Fresnel losses of the ADP crystal, a maximum 355-nm output of 1.61 mJ was obtained at a fundamental energy of 4.64 mJ. The highest 3ω conversion efficiency was 35%, which was obviously better than the result of KDP crystal (30.7%) under similar experimental conditions.[14] From Fig. 4, it can be seen that the THG conversion efficiency tended to be saturated when the fundamental energy was larger than 2.5 mJ. In the whole process of the experiment, the THG output was stable because the forward and the backward beams were not overlapped in the crystal and would not interfere with each other.

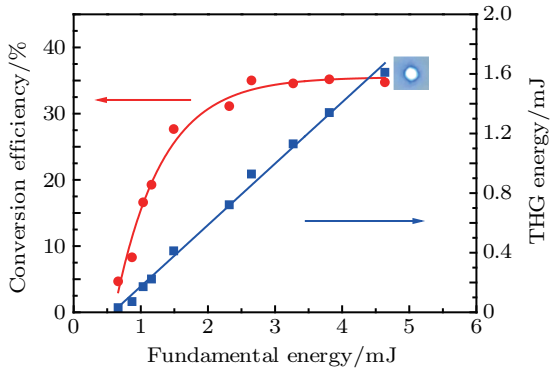


Fig. 4. (color online) THG output energy and conversion efficiency as functions of the fundamental energy. Inset: a THG facula.

5. Conclusion and perspectives

In this paper, a kind of single ADP crystal BPM cascaded THG converter was reported, which can efficiently associate type-II SHG with type-II SFG. The maximum THG conversion efficiency from 1064 nm to 355 nm reached 35%, which was obviously better than the previously reported result of KDP crystal. As an excellent NLO medium for the single crystal BPM cascaded THG converter, ADP has larger d_{eff} and higher laser damage threshold than KDP. At the same time, it is more suitable for the rapid growth technique. All of these characteristics are favorable for the future practical applications of this component.

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