Non-critical phase-matching conditions for fourth harmonic generation of DKDP crystal

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Abstract: The non-critical phase matching (PM) conditions of deuterated dihydrogen phosphate (DKDP) crystal are investigated in this paper for fourth harmonic generation (FHG) of Nd:YAG crystal and Nd:glass lasers, which include exterior angle deviation, deuterium content and PM temperature. DKDP crystals of different deuterium content were grown from aqueous solutions and cut for type-I non-critical phase-matching with the direction at 90° to the crystal Z axis (θ = 90°) and at 45° to the crystal X axis (Φ = 45°). The samples are measured for FHG at 1064 nm and 1053 nm, respectively. The deuterium content of DKDP crystal that supports non-critical phase-matching for FHG experiments is confirmed by measuring the relationship of deuterium content and phase-matching angle. Additionally, the non-critical phase matching temperature is also examined.

OCIS codes: (190.4400) Nonlinear optics, materials; (190.2620) Harmonic generation and mixing; (140.3515) Lasers, frequency doubled.

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1. Introduction

KDP/DKDP crystals are indispensable nonlinear optical materials that perform laser frequency conversion in inertial confinement fusion (ICF) project, owning to their excellent properties including high transmittance from infrared to ultraviolet regions, moderate

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nonlinear-optical coefficient, high laser induced damage threshold, good optical homogeneity and ability of growing into large size of meter order [1–4]. In 1982, researchers in Lawrence Livermore National Laboratory of United States reported the fourth harmonic generation of KDP crystal [5]. For the same frequency conversion procedure, DKDP has two advantages compared with KDP. One is that DKDP crystal can effectively reduce stimulated Raman scattering [6,7] that may result in laser damage, the other is that DKDP crystal can achieve 90 degree non-critical phased-matching (NCPM) such that it is easier to obtain high conversion efficiency. As the refractive index of DKDP crystal can vary with the deuterium content [8] or temperature, the PM angle for FHG of Nd:YAG laser (1064 nm) and Nd:glass laser (1053 nm) can be adjusted accordingly. When the PM angle is 90° to the crystal Z axis (θ = 90°), i.e. the angular NCPM is realized, the output power and the conversion efficiency can be substantially improved, as shown by Steven T. Y. et al. recently [9]. With a 70% deuterated DKDP crystal, they obtained 1.9 J NCPM FHG output for 1053 nm laser, and the conversion efficiency reached 79%.

In this paper, we studied the NCPM FHG conditions of DKDP crystal for the first time by using Nd:YAG laser (1064 nm) and Nd:glass laser (1053 nm) as the fundamental waves, which include PM angle vs. deuterium content, and PM angle vs. temperature. We hope that this present work can provide more references for FHG application of DKDP crystal.

2. Experiment setup

2.1 Crystal growth

The DKDP crystals were grown from different aqueous solutions with the deuterium content of 70%, 80%, 90%, 93% and 98% by using the point-seed rapid growth method. The deuterium content of DKDP crystals are 62%, 74%, 86%, 90% and 97%, respectively. The relationship between the deuterium content of solution (D_s) and the deuterium content of crystal (D_c) is as below [10]

$$D_c = 0.68\,D_s \times e^{0.00382\,D_s} \qquad (1)$$

Crystallization was performed in temperature ranged from 48°C to 41°C. The velocity of temperature-reduction is maintained at 0.1°C/day. The crystal rotated in the mode of “forward-stop-backward” with a speed about 77 rpm [11]. A typical crystal photo is shown in Fig. 1, which reveals no visible macroscopic defects.

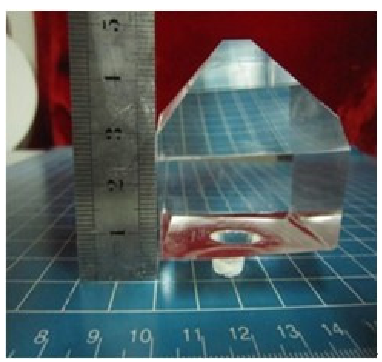

Fig. 1. DKDP crystal grown by point-seed rapid growth method with the deuterium content of 86%.

2.2 Preparation of samples

All of DKDP crystals have similar size and the samples for FHG experiment are cut for type I non-critical phase-matching with the direction at 90° to the crystal Z axis (θ = 90°) and at 45° to the crystal X axis (Φ = 45°), as shown in Fig. 2 and Fig. 3. The thickness of all the samples is 10~12 mm.

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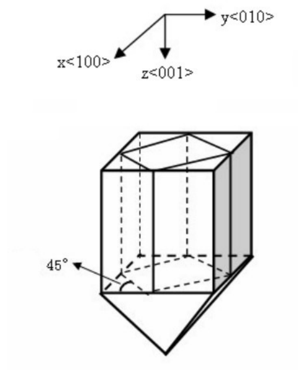

Fig. 2. Cutting schematic diagram of DKDP crystal.

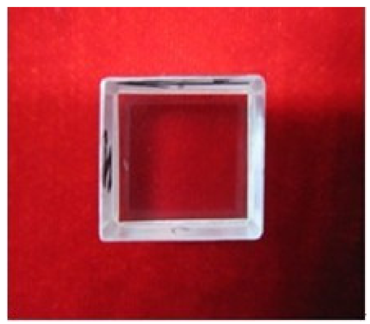

Fig. 3. DKDP crystal sample after processing.

2.3 FHG experiment

The Experimental setup for measuring FHG of DKDP crystal is shown in Fig. 4. The testing fundamental wavelength used for measuring exterior angle deviation is 1064 nm and 1053nm with pulse width of ~10 ns, and the repetition rate of 1 Hz. A KTP crystal is used for second harmonic generation (SHG), and a filter is used to block the residual fundamental light. The DKDP sample is mounted on an adjusting frame that can rotate around the direction of $\theta = 90°$ and keep $\Phi = 45°$ at the same time, in this way the exterior angle between actual PM direction and $\theta = 90°$ direction can be determined.

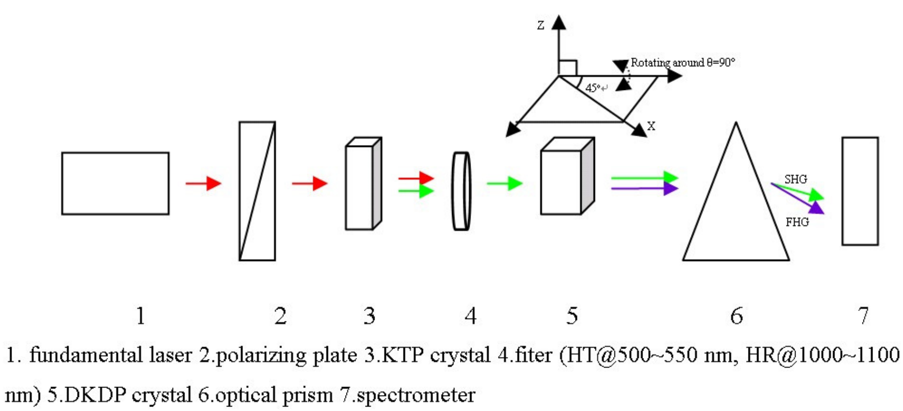

Fig. 4. Experimental setup for measuring FHG of DKDP crystal.

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As shown in the inset of Fig. 4, the KTP crystal is cut for type-II (oee) and the DKDP crystal is cut for type-I (ooe). The DKDP sample shown in Fig. 4 was placed in a cooper cube whose temperature was controlled with an accuracy of ± 0.1°C. The cooper cube was kept tightly sealed (one side is covered with optical glass and the other side is covered with quartz glass) to prevent the DKDP sample from circulation of air and temperature diffusivity.

3. Results and discussion

Figure 5 shows the FHG spectrogram at 1053 nm. The experimental results of PM angle at 26 °C are listed in Table 1. When the fundamental wavelength is 1064 nm, all DKDP samples of different deuterium contents can achieve NCPM by adjusting the spatial angle. However, when the fundamental wavelength is 1053 nm, the DKDP crystals with high deuterium content (86%, 90%, 97%) cannot achieve NCPM. The reason is that for these three crystals the extraordinary light refractive index of 263 nm becomes greater than the ordinary light refractive index of 526 nm as the deuterium content of DKDP crystal increase, so in these samples phase matching for FHG cannot be reached.

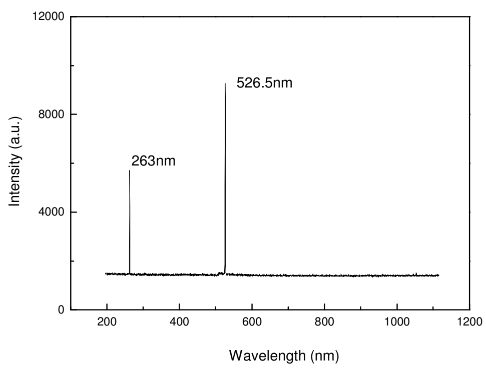

Fig. 5. Spectrogram of fourth harmonic generation of 1053nm.

Table 1. Exterior deviation angle between FHG PM direction and 90° NCPM direction of DKDP crystal with different deuterium contents (T = 26°C)

Deuterium content /%	62	74	86	90	97	Remark
Exterior angle deviation /°	10	8.7	7	4.7	3.7	$\lambda_{fundamental}$ = 1064 nm
	8.7	3	—	—	—	$\lambda_{fundamental}$ = 1053 nm

Furthermore, we examined the NCPM temperature for the samples with high deuterium content (≥74%) at 1064 nm and 1053 nm, respectively. The results are shown in Table 2. The changing of exterior deviation angle with crystal temperature for the 74% DKDP crystal at the fundamental wavelength of 1064 nm is shown in Table 3.

Table 2. The relationship between deuterium content of DKDP crystal and FHG NCPM temperature

Deuterium content /%	74	86	90	97	Remark
FHG NCPM temperature /°C	87	73.7	63.1	47.6	$\lambda_{fundamental}$ = 1064 nm
	28.2	20	—	—	$\lambda_{fundamental}$ = 1053 nm

Table 3. The relationship between exterior deviation angle and crystal temperature for the 74% DKDP sample

Temperature /°C	26	34.4	43.3	47.5	56.2	65.3	81.5	83.4	87	Remark
Exterior deviation angle /°	8.7	8	7	6.7	5.7	5	2.3	1.3	0	$\lambda_{fundamental}$ = 1064 nm

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From Tables 1, 2 and 3, following information can be obtained:

(1) With the increase of deuterium content, exterior deviation angle and NCPM temperature decrease.

(2) When the fundamental wavelength is 1064 nm, all of DKDP crystals cannot realize NCPM FHG at a room temperature around 25°C. Even for the sample with the highest deuterium content (97%) there is still an exterior deviation angle of 3.7°. To compensate this small angle, the crystal temperature has to be elevated to 47.6°C to reach NCPM.

(3) When the fundamental wavelength is 1053 nm, NCPM FHG can be realized in DKDP crystal by adjusting the deuterium content at room temperature. In a temperature range of 20~30°C, the deuterium contents of 74~86% would be the optimum. When the deuterium content is higher, the NCPM temperature would be lower than 20°C.

In a previous work [9], NCPM FHG of Nd:YLF laser was achieved at a temperature of 18.5 ± 0.1°C, using a DKDP crystal with 70% deuteration level. In present work, as seen in Table 2, NCPM at a similar crystal temperature (20°C) was achieved by an 86% deuterated crystal. We think the discrepancy might came from the uncertainty of the deuteration level determined by the method of reference [10]. For DKDP crystals grown by traditional method (period of 4 months), the accuracy of the methods is declared to be ± 0.2% D [10]. The samples we used in this paper were grown by point-seed rapid method (period of 7 days), so it is possible that the rapid growth conditions here lead to an obvious smaller k_{eff} than that calculated from Eq. (1), in this situation the deuteration level used in this paper might has an uncertainty of ~15%. Nevertheless, we speculate that our nominal 86% DKDP crystal can be equal to the 70% DKDP crystal of S. T. Yang et al’s [7,10]. The NCPM temperatures for type-I FHG are similar, and it illustrates that the refractive index are similar. Besides, the Raman shifts are also adjacent, which is measured to be ~888 cm^{-1} for our 86% DKDP crystal, and ~887 cm^{-1} for their 70% DKDP crystal [7]. This fact reflects that these two crystals have similar structure.

4. Conclusion

We investigated type-I NCPM FHG conditions for DKDP crystal, including deuterium content and crystal temperature. When the fundamental wave is 1064 nm (Nd:YAG laser), NCPM cannot be realized at room temperature for all deuterium contents. Nevertheless, for most DKDP sample. NCPM is still possible by elevating crystal temperature. Our research has shown that higher Deuterium content correspond to lower NCPM temperature. For the 97% DKDP sample, its NCPM temperature is 47.6°C. When the fundamental wave is 1053 nm (Nd:glass laser), NCPM can be realized at room temperature for DKDP crystals with proper deuterium content. At a temperature of 20°C the optimum Deuterium content is 86%, while at a temperature of 28°C the optimum Deuterium content is 74%.

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