Homogeneity of large rapidly grown ADP crystals

Liu Hong, Wang ZongQi, Hu Chen, Teng Bing, Huang JinXin and Ji ShaoHua
College of Physics, Qingdao University, Qingdao 266071, People’s Republic of China
1 Author to whom any correspondence should be addressed.
E-mail: hchhide@163.com, 5108tb@163.com(Teng Bing) and jeshhua@163.com

Abstract

DKDP crystal is a type of well-known excellent nonlinear optical material that can be used as 2ω, 3ω, and even 4ω harmonic converters. Compared to DKDP crystal, ADP has a larger nonlinear optical coefficient and higher laser damage threshold, so a larger energy and higher conversion efficiency can be expected from ADP crystals. In this paper, a high-quality ADP crystal (118 × 137 × 160 mm) was grown through point-seed rapid growth method within one month. The corresponding optical properties and homogeneity for noncritical phase-matching fourth harmonic generation were investigated. For the ADP sample with cross-section of 110 mm, the obtained homogeneity noncritical phase-matching temperature is 0.13 °C, which is superior to a DKDP crystal with the same growth parameters. Moreover, UV transmittance property of various thicknesses for ADP crystals was also explored. The transmittance at 263 nm is 88% and decreased 0.36% per millimeter. This result is much lower than the 0.63% for the DKDP crystal in the same thickness range. These results indicate ADP crystals are very promising candidates for noncritical phase-matching fourth harmonic generation.

1. Introduction

Ammonium dihydrogen phosphate (NH₄H₂PO₄, i.e., ADP) crystals and potassium dihydrogen phosphate (KH₂PO₄, i.e., KDP) crystals are well-known multifunctional crystal representatives of the phosphate family and widely used in frequency conversion for coherent radiation of high-power lasers such as second harmonic generation (SHG), third harmonic generation (THG), and fourth harmonic generation (FHG) [1–7]. Nowadays, noncritical phase-matching (NCPM) fourth harmonic generation (FHG) of a 1053 nm Nd:glass laser and a 1064 nm Nd:YAG crystal laser were most concentrated in deuterated potassium dihydrogen phosphate (DKDP) crystals, which attracted lots of attention on the drivers in inertial confinement fusion (ICF) facilities [8–12]. Both DKDP and ADP crystals can be grown from aqueous solution by point-seed rapid growth method. In order to obtain large size and enhance the utilization rate of the crystals, ADP crystals were processed in the Type-I NCPM direction, which is 90° relative to the z-axis (Θ = 90°) and 45° relative to the x-axis (Φ = 45°) [13, 14]. Because of the H/D exchange in DKDP crystals, the refractive index can be continuously adjusted via deuterium segregation, results in inhomogeneity and deuterium segregation difference in the DKDP crystals [15–17]. Compared to DKDP crystals, ADP has larger nonlinear optical (NLO) coefficient, higher laser damage threshold, shorter UV transmission cut-off, faster growth speed, and much lower production cost [18–21]. However, previous research indicates ADP crystals have a narrower temperature-acceptance and larger transverse stimulated Raman scattering (TSRS) gain than DKDP crystals [22–26]. This produces a limitation to its application in NCPM FHG.

In this work, the spatial homogeneity of rapidly grown ADP crystals was investigated through UV transmission and NCPM temperature exploration. We also discussed the NCPM FHG application. The results indicate ADP crystals are superior to DKDP crystals, and they will be a powerful competitor to the commonly used DKDP and KDP crystals. The study results also provide some insight into understanding the homogeneity of crystal materials.
2. Crystal growth and sample preparation

DKDP crystals and ADP crystals can be grown using either a traditional growth method or point-seed rapid growth method [27, 28]. Compared to the traditional growth method, the point-seed rapid growth method has faster growth rate due to the large supersaturation, resulting a better utilization. However, the crystal quality is very sensitive to the crystal growth parameters. The crystal quality mainly depends on the supersaturation and the temperature in the crystal growth process. For both ADP and DKDP crystals, all the crystal growth parameters are controlled to ensure a fair comparison. In this experiment, the supersaturation value is set to a very sensitive to the crystal growth parameters. The crystal quality mainly depends on the supersaturation and the temperature in the crystal growth process. For both ADP and DKDP crystals, all the crystal growth parameters are controlled to ensure a fair comparison. In this experiment, the supersaturation value is set to a value of 2.5% via crystal-growth software to prevent the formation of crystal defects. Crystallization was performed in the temperature range of 325–316 K, and the growth rate of the crystal was approximately 5.3 mm·d⁻¹. The crystal was rotated in a forward-stop-backward mode at a speed of 77 r·min⁻¹. ADP crystal was processed in the Type-I NCPM direction, which is 90° relative to the z-axis (Θ = 90°) and 45° relative to the x-axis (Φ = 45°). As shown in figure 1(a) high-quality ADP crystal with a size of 118 × 137 × 160 mm, was chosen and grown in one month using the point-seed rapid growth method. The growth period was reduced by 85% compared to the traditional growth method. The ADP crystal was cut into 5 samples (figure 1(b)). The distance between the samples is 50 mm, and the cutting zone was 110 × 110 mm above. In this study, we chose a 66% DKDP crystal grown at a supersaturation value of 2.5% in the study because of its ready availability in our laboratory, and its samples have been already performed on FHG experiment [17].

3. FHG temperature experiment

Angular acceptance and temperature acceptance are critical parameters for frequency multiplication materials for NCPM FHG. Previous studies based on the NCPM FHG in ADP crystals could give estimation of external angular bandwidth of ADP is 50.5 mrad·cm⁻¹/², which corresponds to an angular phase-mismatch sensitivity, ∂κ/Δ2Θ, of 4.36 × 10⁻³ mrad²·cm⁻¹ [29]. This value is superior to those in a rapid growth DKDP crystal. However, the temperature bandwidth for the ADP crystal was measured to be 0.6 °C·cm, and the temperature phase-mismatch sensitivity (∂κ/∂T) was 9.3 °C⁻¹·cm⁻¹. These values represent an obstacle for the application in NCPM FHG. In previous research, we found homogeneity of the crystal plays a key role for the stability and efficiency of the NCPM FHG output energy [30]. This is very important to achieve large crystals in NCPM FHG. For example, compared to the ADP crystal, DKDP crystals have advantages with respect to temperature bandwidth and phase-mismatch sensitivity compared with the ADP crystal, but deuterium doping decreases homogeneity and yields 5.4% deuterium difference [24, 25], which causes phase-mismatch. In this work, studies on the homogeneity of DKDP and ADP crystals grown using the rapid growth method for similar crystal growth parameters are conducted.

As shown in figure 2, temperature tuning was performed on both ADP and DKDP crystal samples with the same thickness (5 mm). The NCPM temperature (TNCPM) of DKDP (66% deuterated level) crystal is near 32.5 °C, and the full-width at half-maximum (FWHM) is 4.01 °C, which yields a temperature bandwidth of 2.0 °C·cm and the temperature phase-mismatch sensitivity (∂Δκ/∂T) is 2.78 °C⁻¹·cm⁻¹. These results are in good agreement with those of S. Yang at the Livermore laboratory [6]. By adjusting the crystal temperature at 33.7 °C, NCPM could be achieved in the ADP crystal. Compared to DKDP crystals, the FWHM of the temperature...
tuning curve was 0.84 °C in ADP crystal, which is much narrower than DKDP crystal. This corresponds to the temperature bandwidth of 0.42 °C·cm and the temperature phase-mismatch sensitivity (∂Δκ/∂T) of 1.35 °C⁻¹·cm⁻¹. With the development of the ICF project, homogeneity of crystal, governing stability and efficiency of the NCPM FHG become more and more important. To obtain an efficient and stable NCPM FHG, the inhomogeneity of the T_{NCPM} difference in a large-size crystal should be less than the value that corresponds to the temperature bandwidth at 95% of maximum output power.

As shown in figure 2, the value for the temperature bandwidth at 95% of maximum output power is 1.14 °C for DKDP crystals, which is much larger than the 0.25 °C for ADP crystals. Unfortunately, due to superior temperature bandwidth, the optical quality of the DKDP crystals applied in NCPM FHG suffers from the inhomogeneity of the spatial deuterium distribution. Based on our earlier research, a NCPM temperature deviation of 1.14 °C indicates a 1.87% deuterated-level difference [25]. However, grown at a supersaturation level of 2.5% using a point-seed rapid growth method, the DKDP crystals used 5.3% deuterium deviation (~3.23 °C NCPM temperature deviation), which is much larger than the allowable value according to theory (1.87%), limits the DKDP crystals applications in NCPM FHG due to the low conversion efficiency and decreasing output power. Compared to DKDP crystals, ADP crystals show a larger effective NLO coefficient, a laser induced threshold and superior homogeneity [26].

At a grown supersaturation of 2.5%, ADP crystal exhibits faster growing rate and larger size than DKDP crystal (110 × 110 mm) compare to DKDP crystal (80 × 80 mm).

4. UV transmission

In addition to effective nonlinear optical coefficients and laser-induced thresholds, UV transmittance is another crucial parameter for crystal materials used for NCPM FHG. In this experiment, the five ADP crystal samples cut from (110) were characterized using UV transmission, processed to obtain the same thickness, 5 mm, after the NCPM experiment (figure 1(b)). The transmittance spectra of the ADP and DKDP crystals are recorded with a (HITACHI, U-3500) ranging from 200 nm to 1100 nm.

One can see from figure 4(a), all samples had excellent UV transmission, and according to previous research, the UV transmission of rapid growth ADP crystals is superior to that of rapid growth DKDP crystals, which results in higher FHG output power. As shown in figure 1(b), sample 1 was located at the bottom close to the support shelf, where it is subjected to more dislocation than in other regions. This leads to decreasing UV
transmittance, and the UV transmittance ranging from 200 nm to 400 nm is 80%, much lower than other samples. The results indicate that the FHG sample should be chosen selectively in addition to a rich dislocation area to obtain a high optical quality. As shown in the inset in figure 4(a), these five samples have a transmittance of 77.5%, 90.0%, 90.6%, 88.9%, and 91.1% at 263 nm, respectively. These results are much better than the rapid growth DKDP samples and indicate that the rapidly grown ADP crystals have a much lower level of absorption loss than the rapidly grown DKDP crystals. In other words, the ADP crystal is more favorable for a better NCPM FHG output.

As introduced in the NCPM temperature part, the increasing crystal thickness not only leads to a decrease of temperature acceptance, but also reduces UV transmittance. Therefore, we proposed a comparable study of absorbance at 263 nm as a function of crystal thickness. We cut another six samples of ADP crystal from (110). The thicknesses of the ADP samples were 5.00 mm, 6.60 mm, 8.00 mm, 8.94 mm, 9.94 mm and 10.70 mm, and the thicknesses of the DKDP samples were 4.36 mm, 5.24 mm, 6.22 mm, 7.46 mm, 8.34 mm, 9.26 mm and 10.28 mm, respectively. The UV transmittance is monitored with two laser energy meters (4ω-1) and (4ω-2). In order to prevent spatial inhomogeneous influence on the UV transmittance, all the samples are cut from the same location in the ADP and DKDP crystals, close to the location of sample 2.

As shown in figure 4(b), UV transmission at 263 nm for the ADP and DKDP crystal samples with different thicknesses were conducted. The results indicate linear absorption exists both in ADP and DKDP crystals, and the UV transmission decreases with increasing of crystal thickness. Linear absorption based on the UV transmittance and crystal thickness was calculated. We found ADP crystal has a value of 0.36% per millimeter in thickness, which is better than 0.63% of the DKDP crystal. And in the same thickness, ADP crystal has a better
UV transmittance (3% above) than DKDP crystal. As a consequence, this can give a higher FHG output and conversion efficiency.

5. Conclusion

In this work, a large ADP crystal (118 × 137 × 160 mm) was grown by point-seed rapid growth method. The ADP crystal has a superior homogeneity, i.e., 0.13 °C NCPM temperature distribution deviation, which is much less than DKDP (0.25 °C). The result indicates the rapidly grown ADP crystal is more suitable for NCPM FHG in the ICF project than the rapid grown DKDP crystal, whose NCPM temperature distribution deviation is 3.23 °C larger than 1.14 °C (in theory). With the crystal growth parameters changed, including supersaturation and temperature, the DKDP crystal could suffer from lower homogeneity while the ADP crystals are less influenced. Compared to the DKDP crystal, the ADP crystal has a higher transmittance and a lower linear absorption at 263 nm (0.36%·mm⁻¹). More detailed study of homogeneity in series of rapidly grown DKDP crystals still under investigation and will be reported soon.

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ORCID iDs

Liu Hong https://orcid.org/0000-0002-5508-7524
Teng Bing https://orcid.org/0000-0002-0298-1434

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