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# Noncollinear third-harmonic generation with large angular acceptance by noncritical phase matching in KDP crystal

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**The scheme of prefocusing to focus sum frequency generation (SFG) without a lens is proposed and experimentally verified in this study. Noncollinear type-I noncritical phase-matching SFG to generate a third-harmonic wave with large angular acceptance is presented. The principle of broad angular acceptance and the advantages of this PM configuration are also described in detail. External angular bandwidth of 7.33° for noncollinear SFG was measured in a 2 mm long $KH_2PO_4$ (KDP) crystal, which is in reasonably good agreement with the theoretical calculation. The mechanism of broadband SFG and the prefocusing scheme make it possible for the realization of convergent third-harmonic generation without involving a lens, which provides a promising way to avoid damage to optical components during the focusing of high-energy UV light in high-power laser facilities.** 

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At present, the key factor for the maximum load capacity in the operation of a large solid laser facility is the damage threshold of optical elements [1]. For example, the National Ignition Facility in the USA has an Nd:glass laser delivering pulses of energy up to 1.8 MJ carried by 192 beams and peak power of 500 TW of ultraviolet light with wavelength of 355 nm for performing ignition target experiments [2–4]. Such high-energy UV beams are accompanied by obvious nonlinear effects and, thus, might damage the lenses when focusing and limit the capability of a high-power laser facility [5].

Since the late 1970s, scholars have been studying and measuring the damage thresholds of optical materials, which can be roughly divided into three major aspects: avalanche ionization, multiphoton absorption, and inclusion-induced damage [6]. Of course, a damage mechanism is not confined to what is mentioned above; other patterns such as ultrasound-mediated destruction and small-scale self-focusing also might play a part [7]. Actual damage of optical materials might be the result of the interaction of a variety of damage mechanisms in proportion. Recently, lots of experiments and theoretical researches on laser-induced damage of K9 glass also have been reported. Short wavelength laser, particularly the ultraviolet wavelength, has greater damage power than a longer wavelength laser to fused silica, as it induces not only thermal damage caused by the defect and impurity of the medium but also avalanche-ionization breakdown [8]. Besides, focusing position of crystal and laser beam diameter take effect in the damage threshold as well [9].

In order to solve the existing problems as noted, in this Letter, we propose a new way (prefocusing) to focus $3\omega$ light without a lens in a high-energy level. Figure 1 shows the comparison of the existing scheme [Fig. 1(a)] for the focusing of $3\omega$ light, which might damage the lens at high energy and the revised scheme [Fig. 1(b)]. By prefocusing the fundamental and second-harmonic waves before $KH_2PO_4$ (KDP)/$K(D_xH_{1-x})_2PO_4$ (DKDP) crystals, $3\omega$ light could focus automatically without a lens, which requires the crystal to have broad phase-matching (PM) angular acceptance. Nonlinear crystals, on the other hand, have a much higher damage threshold [1,7].

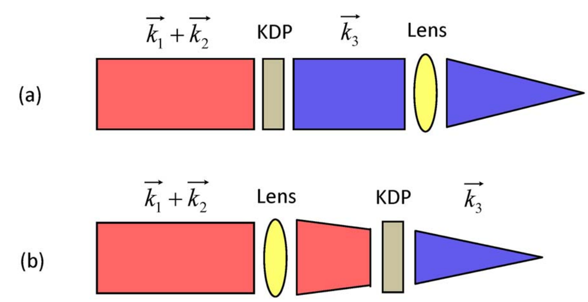


**Fig. 1.** Comparison of existing scheme (a) for focusing $3\omega$ light and revised scheme, (b) for automatically focused $3\omega$ light.

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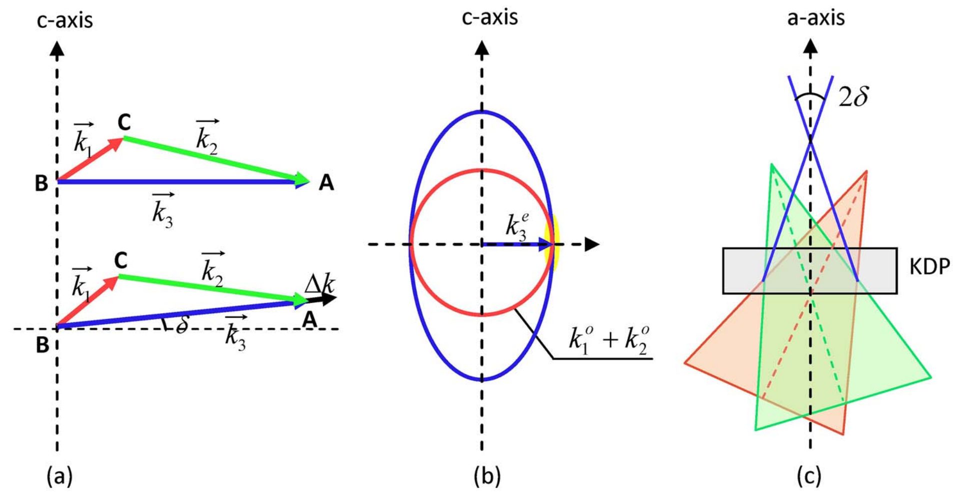


**Fig. 2.** (a) Principle of noncollinear broad angular acceptance. (b) Noncollinear type-I NCPM for broadband SFG. (c) Principle of automatically focused SFG.

However, the crystalline properties of KDP and its analogs [10] limit them to shape a curved surface for simultaneous frequency conversion and beam focusing. At the moment, fabrication of a KDP lens is technically unrealizable.

In collinear PM configurations, noncritical PM (NCPM) has more advantages over critical PM, including a small PM angular sensitivity, a large effective nonlinear optical coefficient, high utilization of the as-grown crystal, and no beam walk-off [11]. However, NCPM along the $\theta = 90°$ direction of different crystals and different PM types corresponds to different specific wavelengths [12]. For current inertial confinement fusion facilities, the fundamental wavelength is 1053 nm; unfortunately, sum frequency generation (SFG) Type-I NCPM wavelength for KDP crystal is about 738 nm. Here, we propose a noncollinear type-I PM configuration setting SFG ($\rightarrow k_3$) at $\theta = 90°$ direction with large PM angular bandwidth and automatic focusing to overcome the existing problem of optical damage when focusing high-energy UV laser beams, as shown in Fig. 2.

In Fig. 2(a), the fundamental wave ($\rightarrow k_1$), second-harmonic wave ($\rightarrow k_2$), and SFG ($\rightarrow k_3$) satisfy the noncollinear wave-vector-matching relationship $\rightarrow k_1 + \rightarrow k_2 = \rightarrow k_3$, where $\rightarrow k_3$ is set at the $\theta = 90°$ direction. Assuming $\rightarrow k_1$ and $\rightarrow k_2$ deviate from their PM directions $\delta$ degrees, then, the new SFG $\rightarrow k_3'$ is supposed to be along with the minimum phase-mismatching direction. For type-I PM ($o + o \rightarrow e$), the new magnitude of $\rightarrow k_1'$ and $\rightarrow k_2'$ remain the same, and $\rightarrow k_3'$ would diverge from 90° direction $\delta$ degrees as well, and the magnitude of it would enlarge slightly, as the KDP crystal is a negative uniaxial crystal. In this condition, wave-vector-mismatching $\Delta k$ could be seen as the variation between the refractive index at $\theta = 90°$ direction and the refractive index at other directions in an SFG refractive index ellipsoid. Therefore, a noncollinear type-I PM configuration is expected to have a broad angular acceptance range when $\rightarrow k_3$ is set at the $\theta = 90°$ direction [Fig. 2(b)]. The principle of automatically focused $3\omega$ light is shown in Fig. 2(c). Apart from the direction of $\theta = 90°$, SFG exists in other directions on account of small PM angular sensitivity. SFG deviated from 90° direction $\delta$ degrees at the border of the laser spot on KDP crystal is the result of deviation of $\delta$ degrees for fundamental and second-harmonic waves. Besides, SFG focal length is related to the focal length of fundamental and second-harmonic waves. The relationship between the deviation of angle $\delta$ and the conversion efficiency $\eta$ could be written as follows:

$$\eta \propto \sin c^2\left(\frac{\Delta k \cdot L}{2}\right), \tag{1}$$

$$k_3 = |\rightarrow k_3| = n_{3\omega}^e \cdot \frac{3\omega}{c}, \tag{2}$$

$$k_3' = |\rightarrow k_3'| = \frac{n_{3\omega}^o n_{3\omega}^e}{[(n_{3\omega}^o)^2 \sin^2(90° - \delta) + (n_{3\omega}^e)^2 \cos^2(90° - \delta)]^{1/2}} \cdot \frac{3\omega}{c}, \tag{3}$$

$$\Delta k = k_3' - k_3, \tag{4}$$

where $n_{3\omega}^o$, $n_{3\omega}^e$ are the refractive indexes of ordinary and extraordinary third-harmonic waves, respectively, $L$, $3\omega$, and $c$ are the crystal length, the frequency of third-harmonic wave, and light velocity, respectively. The angular acceptance here is defined as full width at half maximum (FWHM) of the conversion efficiency.

The experimental setup for noncollinear SFG is shown in Fig. 3. The fundamental wavelength is 1064 nm, with a repetition rate of 20 Hz, a pulse width of 4 ns, and horizontal polarization. The second-harmonic wavelength is 532 nm, with 3 ns pulse width and vertical polarization. The intensities of $1\omega$ and $2\omega$ are 17.4 MW/cm$^2$ and 2.5 MW/cm$^2$, respectively. Two beams of lasers meet a certain angular relationship considering $\rightarrow k_1 + \rightarrow k_2 = \rightarrow k_3$ inside the KDP crystal, as well as the refractive effect of the interface between the crystal and the air, and go through the lens, respectively, for prefocusing. The focal lengths of lenses 6 and 7 are 150 and 175 mm, respectively. As is shown in Fig. 2(a), the internal angles among the three beams of lasers are $A = 6.63°$, $B = 13.51°$, $C = 159.86°$, and the corresponding external angles are $A' = 10.05°$ and $B' = 20.44°$, which means the experimental angle between the fundamental wave and the second-harmonic wave is 30.49°. The half-wave plate and polarizer set in the 1064 nm optical path was used to convert the horizontal polarization to vertical polarization and ensure the incident light is ordinary for a KDP crystal. The KDP sample that we used was 10 mm × 10 mm × 2 mm in size and (90°, 45°) in the cutting direction. The sample was mounted on an adjustable frame, which could rotate around the $\theta = 90°$ angle, and was placed at the off-focused intersection of the fundamental wave and the second-harmonic wave. The generated SFG laser (355 nm) from the KDP crystal was measured with an energy calorimeter. The spatial overlapping of the fundamental wave and second-harmonic wave is longer than the crystal. The inset of Fig. 3 shows the photos of an SFG spot in different positions. The spot sizes were getting smaller and then larger again as we moved forward the screen, and the focal points of the fundamental wave, second-harmonic wave, and SFG were almost in the same focal plane, which exhibits obvious autofocusing phenomenon and confirms our prediction in experiments.

By slowly adjusting the KDP optical axis deviated from $\theta = 90°$, we could measure the angular acceptance of the PM with an energy calorimeter for the third-harmonic output. The result is shown in Fig. 4. At a room temperature of 20°C, the FWHM of the angle acceptance of the noncollinear type-I NCPM for a 2 mm long KDP crystal was measured to be 7.33°, which was equivalent to the inner angular bandwidth of 4.88° based on the refractive index of KDP crystal. The experimental data agrees reasonably well with theoretical calculation for KDP crystal, which verifies our prediction and gives a basis to the scheme of prefocusing. The inset of Fig. 4 shows the relationship between the crystal length and internal PM angular acceptance for noncollinear type-I NCPM. As we can see, decreasing the KDP crystal length properly could achieve larger PM angular acceptance. The insertion point plots the experimental inner angular bandwidth versus 2 mm long KDP crystal. Crystal thickness is determined by the trade-off between conversion efficiency and angle tolerance. Thinner crystal could achieve larger phase-matching angular acceptance but have lower conversion efficiency.

Figure 5 shows the relationship between the third-harmonic output $I_3$ and the product of the fundamental harmonic and the second-harmonic energy $I_1 \times I_2$. The fitting line is a straight line through the zero point, which matches the theoretical prediction. The slope here is relevant to the wavelengths of the incident light, refractive indexes of the crystal, and effective nonlinear optical coefficient. Based on the trend of the fitting line, we could obtain higher third-harmonic output as we increase energy of the fundamental and second-harmonic waves. However, the relationship in which $I_3$ is proportional to $I_1 \times I_2$ is only valid for the nondepleted conversion case, and it breaks down as intensities are increased to go in the saturated regime of the conversion.

In the experiment, we have only taken the KDP crystal as an example; other crystals also could be feasible for prefocusing and the configuration of noncollinear type-I NCPM for SFG. Compared with a regular lens focusing [Fig. 1(a)], the upconversion efficiency in this configuration is slightly lower due to the overlapping, but the angular acceptance is larger; the tolerance for $M^2$ value is supposed to be larger as well. Besides, this kind of PM configuration has no requirement or limitation for fundamental wavelengths, which means we do not have to regulate temperature, doping ratio in crystal [13,14], or other ways to satisfy the specific fundamental wavelength for small PM angular sensitivity such as collinear SFG NCPM. Moreover, even though the experimental angle between the fundamental and the second-harmonic waves is not exactly 30.49°, the KDP crystal could still generate a third-harmonic wave as the incident fundamental wave, and

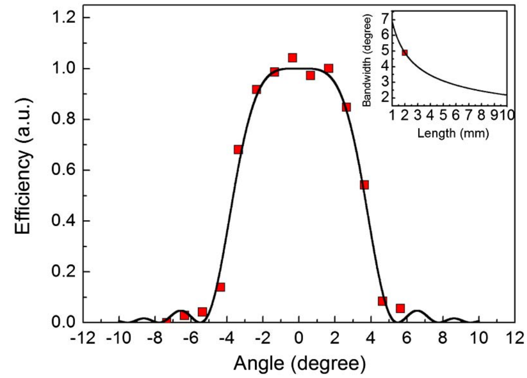


**Fig. 4.** External angular acceptance for noncollinear type-I NCPM in a 2 mm long KDP crystal. Relationship between internal angular acceptance and crystal length is plotted in the inset.

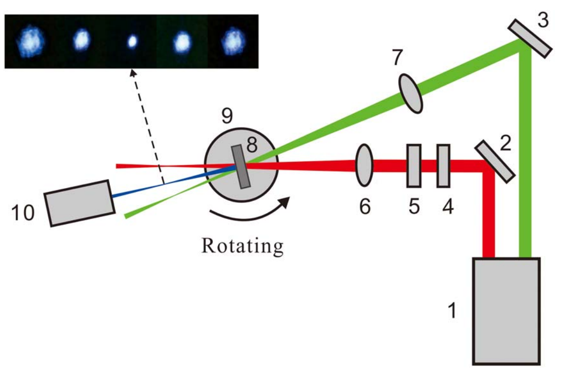


**Fig. 3.** Experimental setup. 1, Nd:YAG nanosecond laser; 2, 3, mirrors; 4, half-wave plate; 5, polarizer; 6, 7, lenses; 8, KDP crystal; 9, adjustable frame; 10, energy calorimeter.

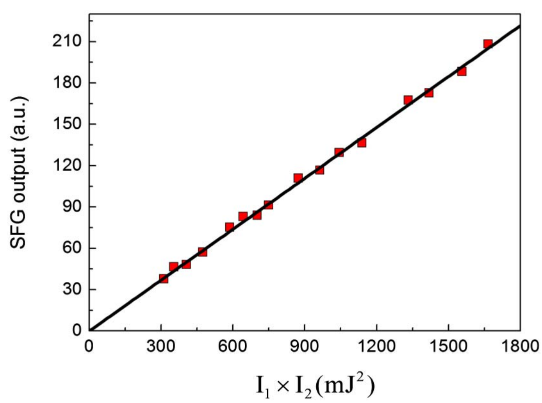


**Fig. 5.** Measured SFG energy versus the product of the fundamental energy and the second-harmonic energy.

the second-harmonic wave has a range of incident angles after being focused but has smaller PM angular bandwidth, which could be compensated by decreasing the crystal length, as we previously discussed. The scheme we propose has such distinct advantages that make it a promising candidate for application in ultraviolet focusing of ultrahigh-energy laser beams such as in laser ignition.

In summary, we have proposed the scheme of prefocusing to focus an SFG without a lens, that is, using lenses to focus the incident fundamental and the second-harmonic waves before KDP crystal for SFG. The new PM configuration for broad-band THG we propose is to ensure the scheme of prefocusing is possible and feasible. By noncollinear type-I NCPM, we could achieve broad PM angular bandwidth such as traditional collinear NCPM configuration.

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