

Conical second harmonic generation in KDP crystal assisted by optical elastic scattering

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Abstract: We observed the generation of phase-matching conical second harmonic generation (SHG) inside KDP crystal under anomalous dispersion condition, which is attributed to complete phase-matching assisted by fundamental wave (FW) and scattering wave. The double-ring pattern of the conical SHG implies that elastic scattering can stimulate two different polarization states of scattering wave. Furthermore, SH ring of KDP displays inhomogeneous intensity around the ring, which has relevance to effective nonlinear coefficient, and it can be used to investigate crystal symmetry.

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1. Introduction

In nonlinear optics, since Bloembergen et al. first proposed the theory of phase-matching in 1962 [1], numerous experiments have successively demonstrated that SHG could be realized through different approaches depending on the principle of phase-matching between interacting waves, such as birefringence phase-matching or quasi-phase-matching (QPM) [2, 3]. Besides traditional collinear and non-collinear frequency conversion [4], phase-matching conical process have also been observed in nonlinear optical materials. For example, second to fifth conical harmonic waves were simultaneously generated in a single two-dimensional (2D) nonlinear photonic crystal [5], which was proposed for applications in crystal symmetry studies, high resolution optical microscopy, or even photon entanglement [6–9]. In addition, phase-matching conical SHG process has also been discovered in various circumstances, such as conical SHG generated by a Bessel beam in a periodically poled KTP crystal [10], conical SHG obtained in LiTaO₃ crystal [11], conical SHG observed in SBN crystal, which is excited by intensive laser light propagated along the crystallographic *c* axis [12], etc..

Either in bulk medium or photonic micro-structure, FW could induce elastic scattering waves along with various spatially directions which is ascribed to internal defects of crystals. In these experimental schemes about conical SHG generation, one of the FWs is provided by elastic scattering, such as in LiNbO₃, a bulk medium, scattering SH ring is derived from the correlation between FW and scattering wave under anomalous-like dispersion condition [13]. Experimentally, scattering SH ring is also demonstrated in one-dimension (1D) of quasi-periodical [14] and 2D periodical [15, 16] optical crystals, for instance, in 1D PPLN, conical SHG has already been discovered [17]. On account of poly-directional scattering waves inside of a crystal, FW vector is able to find a particular direction of scattering wave vector which helps assisting conical SHG. Scattering-assisted conical SHG in LiNbO₃, a 3m symmetry crystal, manifests itself as a uniform second harmonic (SH) ring [13, 17]. In view of conical SHG process, different effective nonlinear coefficients influence the intensity distribution of SH ring. Hence, the intensity distribution of the SH ring can in return reveal the crystal structure.

In this work, we observed scattering-assisted phase-matching conical SHG inside KDP, a $\bar{4}2m$ symmetry crystal. It is confirmed that the elastic scattering stimulates different polarization states of scattering wave to match different types of phase-matching with the FW. Furthermore, each SH ring displays inhomogeneous intensity, which illustrates the structure of the $\bar{4}2m$ symmetry crystal.

2. Anomalous-like-dispersion region

On account of elastic scattering process, the scattered FW could accessibly find a particular scattering wave vector to satisfy a non-collinear phase-matching requirement. Phase-matching condition for SHG can be written as

$$\vec{k}_1 + \vec{k}_1' \geq \vec{k}_2. \quad (1)$$

Where $\vec{k}_1, \vec{k}_1', \vec{k}_2$ are wave vectors of the fundamental, scattering and SH waves, respectively. Equation (1) directly implies that the refractive index of FW must be larger than that of SH wave, which can be achieved in anomalous dispersion region. Previous study has already demonstrated that anomalous-dispersion-like environment [18] can be obtained in a birefringent crystal. Negative uniaxial crystal, such as LiNbO₃, LiTaO₃ and KTP, can mimic anomalous dispersion. In this case, Eq. (1) can be satisfied and SHG could be generated with the assistance of optical elastic scattering. Spatially, such scattering-assisted SHG is visualized as conical SHG.

Scattering-assisted conical SHG in KDP crystal requires anomalous dispersion condition likewise. In Fig. 1, \vec{k} represents for wave vector, k_0 is the wavenumber in the air, n represents for refractive index, subscripts 1, 2 represent for FW and SH wave, subscripts o, e represent for ordinary polarized (o-polarized) and extraordinary polarized (e-polarized) state, respectively (parameters for scattering wave are denoted with a superscript).

To observe scattering-assisted conical SHG in KDP crystal, anomalous-like dispersion should be satisfied. As it is shown in Fig. 1, for oo-e SHG phase-matching, the fundamental wavelength is longer than 0.518 μm [Fig. 1(a)], and for oe-e, only if the fundamental wavelength should be longer than 0.730 μm .

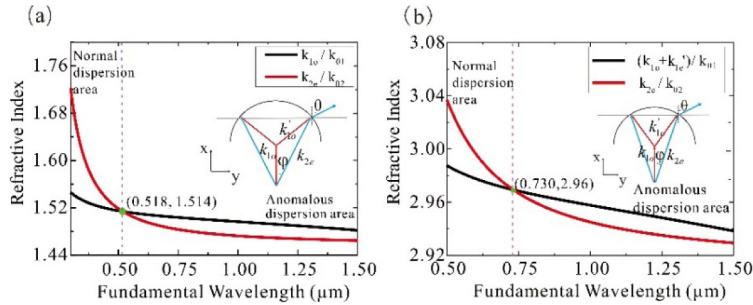


Fig. 1. (a) Dispersion of KDP crystal. The inset shows oo-e phase-matching diagram of conical SHG under anomalous dispersion condition in 2D. (b) Refractive indices relationships for oe-e phase-matching conical SHG. The inset shows oe-e phase-matching diagram of conical SHG under anomalous dispersion condition in 2D.

3. Experiment and discussion

In the experiment, the KDP crystal was Z-cut with a dimension of $5 \times 10 \times 10 \text{ mm}^3$ ($x \times y \times z$). The laser source was an optical parametric amplifier (TOPAS, Coherent Inc.) pumped by a Ti: Sapphire femtosecond system with a pulse width of 80 fs, and a repetition rate of 1 kHz. The input power was about 3 mW. The laser beam was loosely focused by a 100-mm focal lens. The sample was laid on a rotation stage near the focusing position to avoid super-continuum effect, as shown in Fig. 2(a). The incidental FW propagated along the x-axis of crystal, with a beam waist of 50 μm . For the sake of creating anomalous-dispersion-like condition for both oo-e and oe-e process, the input wavelength was tuned from 800 nm to 1400 nm. The input FW was o-polarized, and the output was projected on a screen, 6.2 cm away from the crystal. A Glan-Taylor prism was used to determine the polarization state of emitting SH rings on the screen.

Experimentally, maintaining o-polarized FW, the outgoing conical SHG projected on the screen was observed as two concentric rings with different radii [(Fig. 2(b)], which corresponded to oo-e and oe-e phase-matching, respectively. Each conical SHG has a different half-cone angle due to material dispersion.

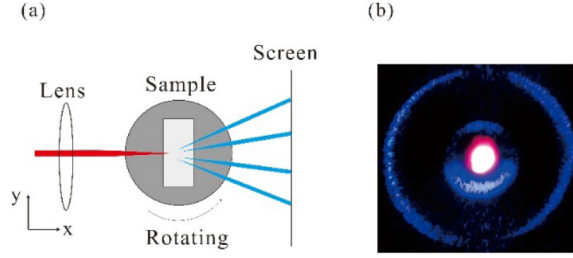


Fig. 2. (a) Schematic of the experimental set up. (b) Images for a typical concentric emitting SH ring pair when incident FW is o-polarized (800 nm).

With regard to oo-e phase-matching conical SHG, the external half-cone angle θ satisfies:

$$\theta = \arcsin \left\{ n_{2e} \sin \left[\arccos \left(\frac{n_{2e}}{n_{1o}} \right) \right] \right\}. \quad (2)$$

The experimental measurement fits well with the theory prediction evaluated by Eq. (2) as shown in Fig. 3(a), which certifies that the outer ring is assuredly complies with oo-e phase-matching SHG. Analogy to oo-e phase-matching, we can directly obtain the relationship for oe-e phase-matching

$$\theta = \arcsin \left\{ n_{2e} \sin \left[\arccos \left(\frac{4n_{2e}^2 + n_{1o}^2 - n_{1e}^2}{4n_{2e}n_{1o}} \right) \right] \right\}. \quad (3)$$

This curve can be checked in Fig. 3(a), which is also in accordance with the experimental data.

Compared with these two phase-matching mechanisms at the same wavelength, the half-cone angle of oe-e is always smaller than that of oo-e. Consequently, inner SH ring is based on the oe-e phase-matching mechanism, the outer one abides by the oo-e phase-matching mechanism as shown in Fig. 2(b). Since the FW is o-polarized, it can be infer that polarization state is also altered by elastic scattering process [19].

When the FW was e-polarized, we also obtained one SH ring on screen. Generally, ee-e phase-matching conical SHG does not exist in KDP crystal, it can be explained that the conical SHG is referred to eo-e phase-matching. The o-polarization of FW wave is from the scattering of incident e-polarization FW wave. Under this condition, the external half-cone angle can be expressed as

$$\theta = \arcsin \left\{ n_{2e} \sin \left[\arccos \left(\frac{4n_{2e}^2 + n_{1e}^2 - n_{1o}^2}{4n_{2e}n_{1e}} \right) \right] \right\}. \quad (4)$$

The relationship is shown in Fig. 3(b), which agrees well with experimental value.

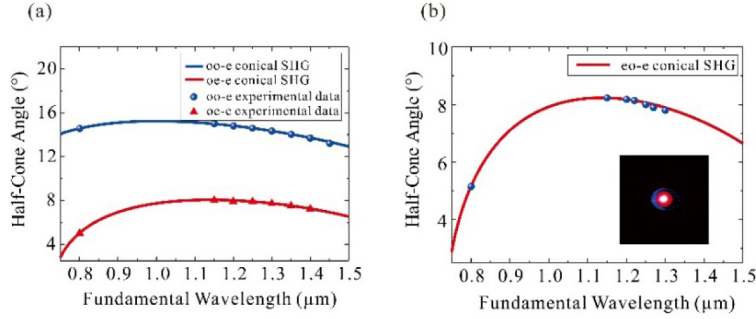


Fig. 3. (a) The half-cone angles of two phase-matching mechanisms. (b) eo-e phase-matching half-cone angle as a function of fundamental wavelength. The inset displays eo-e phase-matching SH ring.

As we can see experiment phenomena in KDP crystal [Fig. 2(b)], the concentric SH rings on the screen display inhomogeneous intensity respectively. The SH rings exhibit the same intensity distribution even we changed the fundamental wavelengths. To gain insight into this, we can calculate the intensity of SHG, which can be expressed as [20]

$$I_2 \propto \frac{8\pi^2 L^2 |I_1| |I_1'|}{n_1 n_1' n_2 c \epsilon_0} d_{eff}^2, \quad (5)$$

where I_1, I_1', I_2 are light intensities of the fundamental, scattering and SH waves, respectively. d_{eff} is the effective nonlinear coefficient, L is the crystal length, c is light velocity in vacuum, ϵ_0 is vacuum permittivity. The elastic scattering intensity of FW is uniform with symmetry around its propagation direction, so that the SH rings intensity distribution would be related to the influence of effective nonlinear coefficient. For instance, previous researches about LiNbO₃ indicates that SH ring of LiNbO₃ displays a uniform intensity distribution [10, 16], which is the result of that the effective nonlinear coefficient of LiNbO₃ is a constant in such symmetry. However, in KDP crystal, effective nonlinear coefficient d_{eff} is affected by the angle of scattering wave and it can be calculated as

$$d_{eff} = \vec{a}_2(2\omega) \cdot \vec{d} : \vec{a}_1(\omega) \vec{a}_1'(\omega), \quad (6)$$

where, $\vec{a}_1(\omega), \vec{a}_2(2\omega)$ are unit vectors of FW, and SH wave, respectively, and \vec{d} is the nonlinear optical coefficient tensor.

Concentric SH rings of KDP crystal both display inhomogeneous intensity on different spots, which can be analyzed by the two phase-matching mechanisms separately. We set θ as the angle between wave vector and z-axis of crystal, φ as the angle between wave vector projection in x-y plane and x-axis of crystal, and γ as the azimuthal angle of any point on the SH rings. Unlike LiNbO₃ whose nonlinear effective coefficient is only related to θ , the effective nonlinear coefficient of KDP associates with θ and γ .

Therefore, for oo-e phase-matching conical SHG, the intensity of spots on the outer SH ring can be derived by the square of effective nonlinear coefficient as

$$d_{eff}^2 = (d_{36} \sin \theta_{2\omega} \sin \varphi)^2, \quad (7)$$

where $d_{36} = 4.61 \times 10^{-12}$ m/V is the second-order nonlinear optical susceptibility of KDP crystal. The curve for Eq. (7) is showed in Fig. 4(a), which reveals that high intensity spots

on the ring must be laid on the same direction as the FW polarization direction, and the relatively darker points are laid on the direction paralleled to the z-axis of the crystal. As shown in Fig. 4(c), the experimental phenomenon fits the simulation results well.

Contrarily, for oe-e SH ring, spots on vertical direction are brighter with larger effective nonlinear coefficient. The square of effective nonlinear coefficient is calculated as ($d_{14} = 5.03 \times 10^{-12} \text{ m/V}$):

$$d_{\text{eff}}^2 = (d_{14} \sin \theta_{2\omega} \cos \theta_{\omega} \cos \varphi + d_{36} \sin \theta_{2\omega} \cos \theta_{\omega} \cos \varphi)^2. \quad (8)$$

As in Fig. 4(b), the curve illustrates that highest intensity dots on the SH ring are laid on the same direction as the z-axis of crystal, and the relatively darker points are along the FW polarization direction [Fig. 4(d), Fig. 2(b) inner SH ring].

Although the effective nonlinear coefficient of oe-e is larger than that of oo-e, the e-polarized scattering light intensity is weaker than that of o-polarized light. Taking this into consideration, integrally, the outer oo-e phase-matching SH ring has the similar intensity with the inner ring. In addition, with a wide range of anomalous dispersion region, KDP can realize conical SHG closed to deep ultraviolet region.

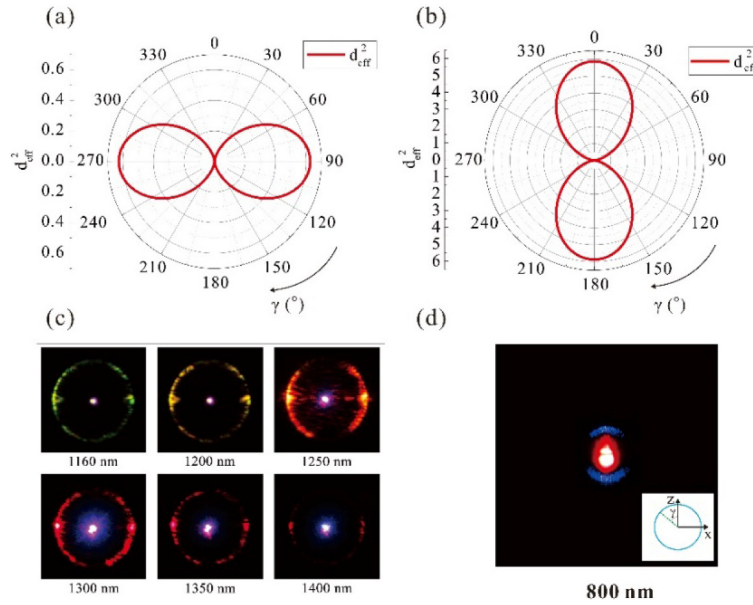


Fig. 4. (a) oo-e phase-matching conical SHG, effective nonlinear coefficient of different spots on SH ring. (b) oe-e phase-matching conical SHG, effective nonlinear coefficient of different spots on SH ring. (c) Images for oo-e SH ring individually with various wavelengths. (d) Images for oe-e phase-matching SH ring with the fundamental wavelength at 800nm.

Because the parameters of SHG rings, such as amount, angle and intensity distribution, is related to symmetry of the nonlinear optical crystal, as for KDP, it is described by Eq. (7), and (8), it provide an effective way to check the symmetry of unknown crystal.

4. Conclusion

In summary, the observation of phase-matching conical SHG by optical elastic scattering in bulk anomalous-dispersion-like medium can be attributed to the assistance between FW and scattering wave. We further experimentally expound that optical elastic scattering process can contribute to alternative polarization states of scattering wave. Furthermore, we revealed the intensity distribution of KDP emission SH ring, which can be consequently ascribed to

altered effective nonlinear coefficient. By analyzing effective nonlinear coefficient, higher intensity spots on emitting SH ring has higher value of effective nonlinear coefficient, which can potentially be applied to detect symmetry structure of nonlinear optical crystals.

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