Interactive length of fundamental wave and second harmonic generated on the surface of anomalous dispersion medium

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In this Letter, a new method is presented to calculate the interactive length between the fundamental wave and second harmonic generation (SHG) for the configuration of total internal reflection on the inner surface of a nonlinear crystal. Three independent experiments are designed to measure the bandwidths of this second harmonic wave. The theoretical expression of the intensity of SHG is obtained through a nonlinear coupled wave equation. The interactive length of this phase-matched SHG can be calculated mathematically by utilizing the measured bandwidths and the intensity equation. There is no existing method to obtain the interactive length either from theoretical calculations or by experimental measurement. This method can be applied to estimate the extremely short interactive volume in nonlinear processes.

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Nonlinear frequency conversion processes in optics have been extensively studied since the invention of light amplification by stimulated emission of radiation[1–9]. The efficiencies of nonlinear parametric processes are determined by the extent of phase matching. Because of the effect of dispersion, it is difficult to naturally attain the condition of phase matching in interacting waves. In previous research, phase matching can be achieved by various artificial techniques, such as tuning temperature and angle in birefringent crystals, and applying periodic-poled nonlinear crystals with a change of the sign of second-order nonlinear susceptibility[9–13]. The periodic change of second-order nonlinear susceptibility can provide an additional reciprocal wave vector to fulfill the phase-matching condition in nonlinear parametric processes. New types of phase matching, like nonlinear Cherenkov radiation, have also been researched extensively[14–17]. Nonlinear parametric processes that are generated on the boundary of a bulk medium have been researched deeply in previous work[15–20]. Such nonlinear parametric processes have novel phase-matching types like scattering-assisted conical second harmonic generation (SHG) and reflective-light-assisted sum frequency. The sharp second-order susceptibility modulation from 1 to 0 can give rise to the enhancement of the conversion efficiency. The phase-matching mechanism of nonlinear parametric processes generated from complete phase matching of incident light and reflected light on the boundary of bulk crystals was investigated deeply in our previous research[17,18]. The diagrams of this phase-matching condition are shown in Fig. 1(a). When the wavelength of the fundamental wave exceeds 1030 nm, the phase velocity of the corresponding polarized wave exceeds the ordinary fundamental wave in birefringent crystals, as shown in Fig. 2(b). Under this anomalous-like dispersion environment, the wave vectors of the fundamental wave and SHG form a complete triangle on the boundary of the bulk crystal, as shown in Fig. 1(b). From the deduced nonlinear coupling wave equation, we can

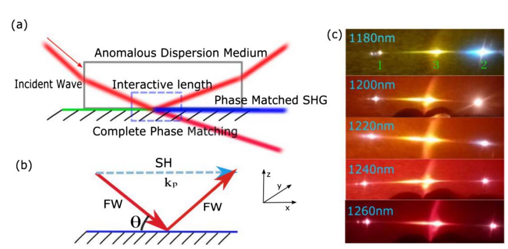

Fig. 1. (a) Diagrams of SHG generated by incident and reflected fundamental waves. (b) The triangle phase-matching type of the SHG spot. (c) The photographs of SHG with different fundamental wavelengths (1180 nm, 1200 nm, 1220 nm, 1240 nm, and 1260 nm).

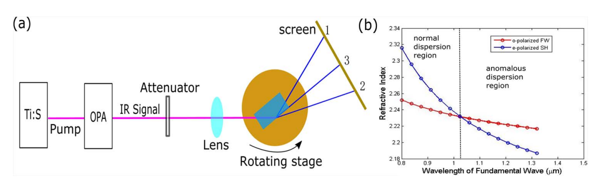

Fig. 2. (a) Experiment setup. (b) Comparison of the refractive index of an extraordinary-polarized second harmonic beam and that of an ordinary-polarized fundamental beam in a lithium niobate crystal.

conclude that the intensity of SHG I_2 is related to Δk. The scalar equation of this phase-mismatched factor can be demonstrated as $\Delta k = 4\pi[n_{2e}(T) - n_{1o}(T)\cdot\cos\theta]/\lambda_1$. Incident angle, operating temperature, and pump wavelength are three parameters directly related to the intensity of the noncollinear SHG. By varying incident angle, operating temperature, and pumping wavelength to a small extent, several phase-mismatched noncollinear second harmonics (SHs) can occur under this anomalous-like dispersion condition. Their generation mechanisms are attributed to the large acceptance of SHG from complete phase matching of incident light and reflected light on the boundary of the nonlinear bulk medium[17]. The conversion efficiency of the phase-matching noncollinear SHG is up to 15.74%, with an extremely short interactive volume for one single total reflection on the surface of the nonlinear crystal. When one of these three parameters is changed in one precise phase-matching situation, the precise balance is broken. Although two methods that are based on beam-pulse overlap and conversion efficiency can be used to investigate the interactive length, these methods both require specific information for both the beam width of the fundamental wave (FW) and second-order nonlinear susceptibility. These requirements are obstacles for investigations when this information is unknown. In this Letter, a nonlinear coupled wave equation of this type of SHG is deduced. The relationship of the intensity of SHG with the interactive length can be obtained from the coupled wave equation. Through three independent experiments, three parameters of bandwidth are measured, respectively. After measuring the bandwidths and deducing the intensity equation, one method is proposed to calculate the interactive length of this SHG through relative equations without knowing the beam width of the FW and the second-order nonlinear susceptibility of a nonlinear crystal.

In the following, the coupled wave equation for a triangle phase-matching type of SHG generated by incident light and reflected light on the boundary of a nonlinear crystal will be solved. The fundamental wave illuminates the inner boundary of the bulk crystal with an internal angle of θ. The fundamental wave is a Gaussian beam and the modulation along the z axis is neglected for simplicity; the dispersion function of the amplitude turns out to be e^{-x^2/a^2}, where a is the beam width. The relevant time-independent parts of the amplitude of the incident and reflected fundamental waves are

$$E_1 = A_1 e^{-z^2/a^2} e^{i(k_1\cos\theta\cdot x + k_1\sin\theta\cdot z)}, \tag{1}$$

$$E_1' = A_1 e^{-z^2/a^2} e^{i(k_1\cos\theta\cdot x - k_1\sin\theta\cdot z)}. \tag{2}$$

The expression of the SH wave is defined as

$$E_2 = A_2(x,z)e^{ik_2\cdot x}. \tag{3}$$

In the above equations, $A_1 e^{-z^2/a^2}$ and A_2 represent the amplitude of the fundamental wave and the second harmonic wave, respectively. k_1 and k_2 are the wave vectors of the FW and SHG, respectively. The dispersion function of the amplitude of the FW turns out to be e^{-z^2/a^2}, where a is the beam width. The nonlinear coupled wave equations are deduced from the Maxwell equations. Because of the undepleted pump approximation, we can consider the amplitude of the fundamental wave as unchanged. Under these assumptions, the nonlinear coupled wave equation of the amplitude of the SH wave turns out to be[20]

$$\frac{\partial}{\partial x}A_1(x,z) = 0, \tag{4}$$

$$\left(\frac{i}{2k_2}\frac{\partial^2}{\partial z^2} + \frac{\partial}{\partial x}\right)A_2(x,z) = -i\frac{k_2}{2n_{2\omega}^2}h(z)\chi^{(2)}\left(A_1 e^{-\frac{z^2}{a^2}}\right)^2 \cdot e^{i(k_2 - 2k_1\cos\theta)\cdot x}, \tag{5}$$

where $n_{2\omega}$ denotes the refractive index of the SH wave, $\Delta k = k_2 - 2k_1\cos\theta$ is the phase-mismatched factor, ω is the angular frequency of the FW, and $h(z)$ denotes the distribution function of $\chi^{(2)}$. It represents the structure function of the nonlinear crystal. In this case, $h(z)$ has the following form:

$$h(z) = \begin{Bmatrix} 1, \\ 0 \end{Bmatrix} \begin{matrix} z \geq 0 \\ z < 0 \end{matrix}. \tag{6}$$

To solve Eq. (5), we change the amplitude from the space function $A_2(x,z)$ into the Fourier spectrum:

$$A_2(x,k_z) = \int A_2(x,z)\cdot e^{ik_z z}\,\mathrm{d}z. \tag{7}$$

We substitute Eq. (7) into the nonlinear coupled wave equation, and seek two partial differential equations for z. Then Eq. (5) takes the form

$$\left(\frac{\partial}{\partial x} - i\frac{k_z^2}{2k_2}\right)A_2(x,k_z) = -i\frac{k_2}{2n_{2\omega}^2}\chi^{(2)}(A_1)^2 e^{i\Delta kx} \cdot \int e^{-2z^2/a^2}\cdot h(z)\cdot e^{ik_z z}\,\mathrm{d}z. \tag{8}$$

where $\Delta k = k_2 - 2\cos\theta\cdot k_1$ is the phase mismatched factor. We apply the Fourier-transform technique on both sides of Eq. (8), and then solve the corresponding inhomogeneous linear equation. The x integration limit is from 0 to x, where x is the interactive length. The amplitude of the second harmonic wave can be expressed as

$$A_2(k_z,x) = i\frac{k_2}{2n_{2\omega}^2}\chi^{(2)}\cdot A_1^2\cdot\frac{e^{i\left(\Delta k - \frac{k_z^2}{2k_2}\right)x} - 1}{i\left(\Delta k - \frac{k_z^2}{2k_2}\right)}\cdot e^{i\frac{k_z^2}{2k_2}x} \cdot \int e^{-2z^2/a^2}\cdot h(z)\cdot e^{ik_z z}\,\mathrm{d}z. \tag{9}$$

The intensity of the SH $I_2(x, k_z) = |A_2(x, k_z)|^2$ can then be expressed as

$$I_2(k_z,x)=\left[\frac{k_2}{2n_{2\omega}^2}\chi^{(2)}\right]^2 I_1^2\cdot x^2\cdot \mathrm{sinc}^2\left[\left(\Delta k-\frac{k_z^2}{2k_2}\right)\frac{x}{2}\right]|H(k_z)|^2. \tag{10}$$

As we analyzed in our previous research[17], k_z is the transverse wave vector of the generated SH. The symbol sinc represents a function of $\mathrm{sinc}(x) = \frac{\sin(\pi x)}{\pi x}$. The situation of $k_z = 0$ corresponds to this phase-matching SHG. The intensity of this central SHG can then be expressed as

$$I_2(k_z, x) = \left[\frac{k_2}{2n_{2\omega}^2}\chi^{(2)}\right]^2 I_1^2\cdot x^2\cdot \mathrm{sinc}^2\left|\Delta k\cdot\frac{x}{2}\right||H(k_z)|^2, \tag{11}$$

where x is actually the interactive length, $\Delta k = k_2 - 2k_1\cos\theta$ is the phase-mismatched factor, and $H(k_z) = H(0) = \int e^{-2z^2/a^2}\cdot h(z)\cdot e^{ik_z z}\,\mathrm{d}z = 2\sqrt{\frac{\pi}{8}}a$.

Under anomalous-dispersion-like conditions, the requirement for phase matching of noncollinear SHG generated by incident light and reflected light on the boundary of a bulk crystal is analyzed to be $k_2 = 2k_1\cos\theta$[17]. A z-cut 5% (mole fraction) MgO:$LiNbO_3$ crystal of 3 mm × 10 mm × 2 mm size is utilized as the experimental crystal. The experiment setup is shown in Fig. 2(a). The two symmetrical SHG spots, 1 and 2, are collinear SHG of the incident and reflective wave, respectively. The SHG marked as 3 is the phase-matching noncollinear SHG generated by incident light and reflected light on the boundary of the bulk crystal. The intensity of the SHG marked as 3 can be significantly enhanced when the phase-matching requirement is satisfied. Equation (10) shows that both the radiation angle and the intensity of this noncollinear SHG generated on the boundary are mainly determined by the term $\mathrm{sinc}^2(\Delta k\cdot x/2)$. The conversion efficiency is measured as up to 15.74% for this surface complete phase-matching SHG. $\Delta k = k_2 - 2k_1\cos\theta$ is the phase-mismatched factor. When k_2 equals $2k_1\cos\theta$, Δk equals 0 under this condition, and $\mathrm{sinc}^2(\Delta k\cdot x/2)$ equals 1. The intensity of SHG is the maximum in this condition. This noncollinear SHG is under the phase-matching condition. The experimental setup is demonstrated in Fig. 2(a). There are three parameters determining the scalar quantity of the factor Δk. The scalar equation of the phase-mismatched factor of this SHG generated on the boundary can be expressed as $\Delta k = 4\pi[n_{2e}(T) - n_{1o}(T)\cdot\cos\theta]/\lambda_1$. There are three parameters that are directly related to the phase-mismatched factor. These parameters are internal incident angle, temperature of crystal, and wavelength of FW. The intensity of the SHG I_2 is proportional to $d_{\mathrm{eff}}^2 I_1^2\cdot x^2\cdot\mathrm{sinc}^2(\Delta k\cdot x/2)$ as demonstrated in the deduced energy equation of SHG in Eq. (11). When one of these parameters in one precise phase-matching situation is changed, the precise balance is broken. The intensity of SHG decreases from the maximum. The interactive length x is unchanged under the slowly varying envelope approximation. Therefore, the phase-mismatched factor Δk exclusively decides the intensity of SHG. By utilizing three independent experiments that measure the bandwidth of these three parameters, three statistical data can be obtained to calculate the interactive length through the corresponding equations. Through mathematical calculations, when $\Delta k\cdot x/2$ equals 1.3916, $\mathrm{sinc}^2(\Delta k\cdot x/2)$ goes to 1/2. This situation corresponds to half of the maximum energy of the SHG. By measuring the bandwidths of angle, temperature, and wavelength through three independent experiments, these measured bandwidths can be substituted into the corresponding equations to calculate the interactive length as follows:

$$\Delta k = 4\pi[n_{2e}(T) - n_{1o}(T)\cdot\cos\theta]/\lambda_1, \tag{12}$$

$$\Delta k\cdot x/2 = 1.3916. \tag{13}$$

The incident angle, temperature, and wavelength are the three direct parameters that determine the phase-mismatched factor of the noncollinear SHG on the boundary of the nonlinear crystal. In order to estimate the interactive length through mathematical calculation, we need to obtain the bandwidth of these three parameters first. For the investigation of the bandwidth of angle, the bandwidths of angles that correspond to different fundamental wavelengths are measured. The inner incident angle θ is related to the scalar quantity of the phase-mismatched factor Δk, as shown in Eq. (12). Ordinary-polarized fundamental waves with wavelength of 1064, 1200, 1400, and 1600 nm are, respectively, used to illuminate the inner boundary of the nonlinear crystal. The temperature of the experiment is set at 29°C. A femtosecond laser pulse is produced by an optical parametric amplifier (TOPAS, USF-UV2) (repetition frequency 2 kHz and pulse duration 50 fs). The temperature and fundamental wavelength are kept unchanged once the condition is confirmed to make sure the angle is the only changing parameter. A receptor is used to measure the energy of SHG. When the nonlinear crystal is rotated to change the incident angle, the maximum and half of the maximum energy of the SHG can be measured. By measuring the corresponding angles the bandwidths of the angles can be obtained. The external angle bandwidths are measured to be 0.49°, 0.54°, 0.53°, and 0.50°. These measured bandwidths correspond, respectively, to fundamental waves with wavelengths of 1200, 1240, 1280, and 1300 nm. Experimental results are shown in Figs. 3(a)–3(d). Then these measured bandwidths are transformed into inner incident angles through Snell's equation and substituted into Eqs. (12) and (13). The interactive lengths are calculated to be 0.4012, 0.4220, 0.4210, and 0.443 mm, respectively, through these equations.

Temperature is the second parameter that directly determines the refractive coefficients of the fundamental wave and second harmonic wave $n_{1o}(T)$ and $n_{2e}(T)$. In order to have a relatively precise temperature bandwidth,

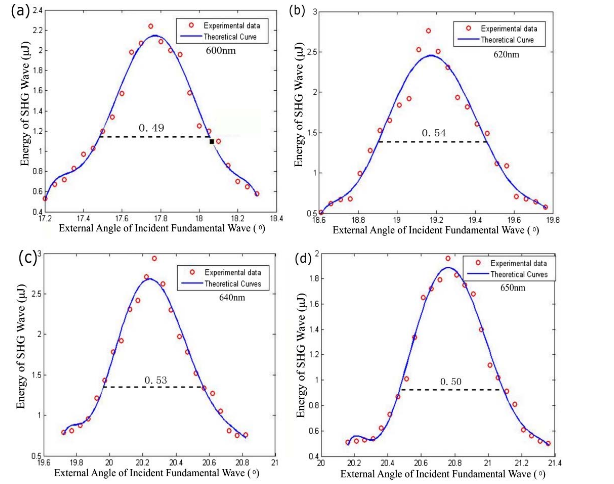

Fig. 3. (a) The measured energy of SHG with the relationship of angles (fundamental wavelength of 1200 nm) and the energy changing process. (b) The measured energy of SHG with the relationship of angles (fundamental wavelength of 1240 nm) and energy changing process. (c) The measured energy of SHG with the relationship of angles (fundamental wavelength of 1280 nm) and energy changing process. (d) The measured energy of SHG with the relationship of angles (fundamental wavelength of 1300 nm) and energy changing process.

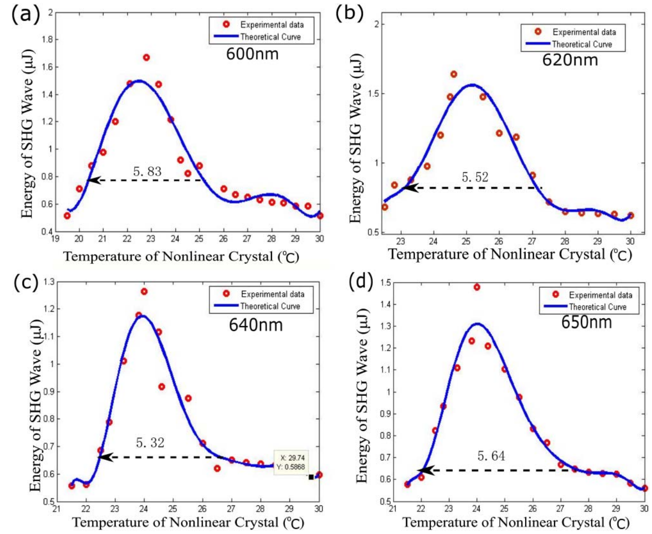

Fig. 4. (a) The measured energy of SHG with the relationship of temperature (fundamental wavelength of 1200 nm) and the energy changing process. (b) The measured energy of SHG with the relationship of temperature (fundamental wavelength of 1240 nm) and the energy changing process. (c) The measured energy of SHG with the relationship of temperature (fundamental wavelength of 1280 nm) and the changing process of energy. (d) The measured energy of SHG with the relationship of temperature (fundamental wavelength of 1300 nm) and the energy changing process.

we measure the bandwidth of different fundamental waves with wavelengths of 1200, 1240, 1280, and 1300 nm. An optical parametric amplifier (TOPAS, USF-UV2) is used to produce femtosecond laser pulses (repetition frequency 2 kHz and pulse duration 50 fs). When the temperature of the nonlinear crystal is changed, the refractive coefficients of the fundamental wave and second harmonic wave will change subsequently due to their dependence on temperature. Equation (12) shows that the refractive coefficients of the fundamental wave and second harmonic wave are directly related to the scalar quantity of the phase-mismatched Δk. The results show that changing the temperature of a nonlinear bulk crystal can disturb the balance of the phase-matching condition because it causes the scalar quantity of Δk to deviate from 0. The incident angle and the fundamental wavelength are kept unchanged in order to make sure the temperature is the only changing parameter. When the temperature of the nonlinear crystal is changed, the maximum and half of the maximum energy of the SHG can be measured. By measuring the corresponding temperatures, the bandwidths of the temperature can be obtained. The temperature bandwidths of the crystal measured are 5.83°C, 5.52°C, 5.32°C, and 5.64°C, respectively, as shown in Figs. 4(a)–4(d). The measured bandwidths are substituted into Eqs. (12) and (13). Then the interactive lengths are calculated to be 0.4639, 0.4821, 0.4780, and 0.5092 mm, respectively.

Wavelength is the third parameter that directly determines the scalar quantity of this phase-mismatched factor Δk. The optical parametric amplifier femtosecond laser system (50 fs pulses at a repetition rate of 1 kHz) is used to provide the fundamental wave[17]. The fundamental wave has a 75 nm bandwidth. One precise incident angle corresponds to only one fundamental wavelength. The external angles are fixed from 14° to 20°, respectively. These external angles correspond to different fundamental wavelengths (centric wavelengths from 1112 nm, 1122 nm, 1150 nm, 1164 nm, 1184 nm, 1210 nm, to 1246 nm). In order to make sure the wavelength is the only changing parameter, the incident angle and temperature are kept unchanged. From the obtained spectral components of SHG, the bandwidth of SHG with a wavelength of 575 nm is measured to be approximately 3 nm[17]. Considering the quadratic relationship of the intensity between SHG and the fundamental wave, the calculated wavelength bandwidth of the fundamental wave is 6 nm. The measured bandwidth is substituted into Eqs. (12) and (13). The interactive length is calculated to be about 0.4142 mm. The interactive lengths of this phase-matching SHG independently calculated through three bandwidth parameters are very close. The calculated interactive lengths that are deduced from angle, temperature, and wavelength bandwidths have some differences. The difference mainly comes from imprecise experimental measurements because the data of the three different bandwidths measured by the experiment equipment are not so accurate. Some work still has to be done to improve the accuracy of the equipment for these bandwidth experiments. The maximum is 0.5092 mm, the minimum is 0.4012 mm, and the average is 0.4463 mm. The interactive length between the incident light and reflected light is not that short for two main reasons. The first reason is that the incident light is a Gaussian beam. The incident light does not illustrate the boundary of the crystal with the shortest beam width for avoiding destroying the nonlinear crystal, so it has longer beam width. The second reason is that the incident light illuminates the boundary with an angle θ. This oblique incident lengthens the interactive length.

In summary, we study theoretically the coupled wave equation of SHG generated by a Gaussian beam on the boundary of quadratic bulk nonlinear media. By using the Fourier-transform technique, the coupled wave equation of this SHG is solved. Through three independently designed experiments that focus on the bandwidth of three parameters, which determine the phase matching geometry of this SHG, statistical data are obtained. Then by utilizing the measured bandwidths and the coupled wave equation through mathematical calculations, the interactive length of this phase-matching SHG is calculated. This method will provide useful information to estimate an extremely short interactive volume. This method can also be used to provide specific information for both unknown beam widths of FW and nonlinear second-order susceptibility of nonlinear crystals by corresponding equations.

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