Simulation of the thermo-optic coupling effect in mid-infrared second harmonic generation of ZnGeP₂ crystal

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The thermo-optic coupling process of second harmonic generation was numerically simulated in ZnGeP₂ crystals pumped by a pulsed CO₂ laser at the wavelength of 9.6 μm, under the strong and weak cooling conditions. The conversion efficiencies, temperature distributions were calculated during the evolution of the thermo-optic coupling. The results showed that the thermo-optic coupling was weak in the strong cooling condition, which nearly did not disturb the conversion processes and temperature distribution, while in the weak cooling case, the temperature distribution showed a great influence on the conversion efficiency and light intensity. Finally, it was found that compensation of the phase mismatch induced by the thermal effect can well recover the conversion efficiency.

Keywords: *second harmonic generation; thermo-optic coupling; ZnGeP₂ crystal*

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1. Introduction

The thermo-optic coupling effect of optical frequency conversion frequently occurs in a nonlinear crystal, disturbing the conversion process to some extent. Commonly, the light absorbed in a related nonlinear crystal turns to heat which enhances the temperature inside. This local temperature rising causes a relative change of refractive indices, destructs the phase-matching (PM) condition and lowers the conversion efficiency [1–3]. In a common frequency conversion process, such as second harmonic generation (SHG), the thermo-optic coupling effect is generally unable to be avoided without cooling, especially in the infrared range. In early 70s, Okada and Ieiri discussed the relation between the phase mismatch and the absorption assuming a Gaussian heating distribution [2]. Dreger and McIver, modeled a new wave equation

set of SHG including the thermal effect and considered the influence of temperature on the conversion efficiency [4]. Seidel and Mann developed a numerical model including the thermo-optic coupling by an iteration method [5], in which they assumed a simple case, where a cylindrical symmetry exists in the transverse dimensions. Huang et al. proposed a simple model to calculate the thermal effect in BBO crystal, assuming an infinite transverse boundary [6]. At the same time, Sabaeian et al. presented a similar model to treat the temperature evolution in KTP crystal for type II SHG, with the first and the second transverse boundary conditions [7]. We have developed a model to consider the thermo-optic coupling effect accounting for the thermal anisotropy, and applied it to the simulation of the temperature distribution in GaSe [8]. An accurate evaluation and analysis of the thermo-optic coupling effect provides an assistance to reduce such a thermal effect through an ambient temperature control with a frequency converter, as it was already

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done before in some special cases [9–11]. ZnGeP₂ crystal is a powerful crystal for mid-infrared light generation, due to its large nonlinear coefficient, high damage threshold and high thermal conductivity [12–17]. In this paper, we will apply the former model [8] to ZnGeP₂.

2. Thermo-optic coupling equations

Fig. 1 shows the beam incident into a uniaxial nonlinear crystal, whose coordinate origin is at the incident surface, optical axis is in the x-z plane, θ is PM angle, ρ the walk-off angle, L the crystal length, the extraordinary and ordinary beams are polarized in x and y directions, respectively. So the x axis points to the beam lateral walk-off direction.

The coupled wave equations for type I SHG (where $\Delta_{\perp} = \partial^2/\partial x^2 + \partial^2/\partial y^2$) are [8]:

$$\begin{aligned} \frac{\partial A_1}{\partial z} + \frac{1}{2ik_1}\Delta_{\perp}A_1 + \rho_1\frac{\partial A_1}{\partial x} + \frac{\alpha_1}{2}A_1 - i\frac{\omega}{c}\frac{\partial n_1}{\partial T}TA_1 \\ = \frac{i\omega}{cn_1}d_{eff}A_1^*A_2 \exp[i(k_2 - 2k_1)z] \end{aligned} \quad (1)$$

$$\begin{aligned} \frac{\partial A_2}{\partial z} + \frac{1}{2ik_2}\Delta_{\perp}A_2 + \rho_2\frac{\partial A_2}{\partial x} + \frac{\alpha_2}{2}A_2 \\ - i\frac{2\omega}{c}\frac{\partial n_2}{\partial T}TA_2 = \frac{i\omega}{cn_2}d_{eff}A_1^2 \exp[i(2k_1 - k_2)z] \end{aligned} \quad (2)$$

where $A_{1,2}$ and $k_{1,2}$ are the electric field amplitudes and wave vectors, respectively, while $\rho_{1,2}$, $\alpha_{1,2}$, $n_{1,2}$, d_{eff} , $\partial n_{1,2}/\partial T$, T are, respectively, the fundamental and harmonic beam walk-off angles, absorption coefficients, refractive indices, effective nonlinear coefficient, the derivative of refractive index on temperature and the temperature difference between inside and outside the crystal. The coupled wave equations can be solved by the typical split-step method or the Fourier transformation method [18]. The heat transfer equation can be

described as [8]:

$$\begin{cases} \frac{\partial T}{\partial t} - \left(a_X^2 \frac{\partial^2 T}{\partial X^2} + a_Y^2 \frac{\partial^2 T}{\partial Y^2} + a_Z^2 \frac{\partial^2 T}{\partial Z^2} \right) = F'(x, y, z, t) \\ \left(T - H \frac{\partial T}{\partial z} \right) \Big|_{z=0} = 0, \left(T + H \frac{\partial T}{\partial z} \right) \Big|_{z=L} = 0 \\ T|_{t=0} = \phi(x, y, z) \end{cases} \quad (3a)$$

with

$$\begin{cases} T|_{x=0} = 0, T|_{x=W_x} = 0 \\ T|_{y=0} = 0, T|_{y=W_y} = 0 \end{cases} \quad (3b)$$

or

$$\begin{cases} \partial T / \partial x|_{x=0} = 0, \partial T / \partial x|_{x=W_x} = 0 \\ \partial T / \partial y|_{y=0} = 0, \partial T / \partial y|_{y=W_y} = 0 \end{cases} \quad (3c)$$

where $F'(x, y, z, t) = G(x, y, z, t)/(C\rho_c)$, G , the heat flux density produced at (x, y, z) , $a_j^2 = K_j/(C\rho_c)$ with the crystal heat capacity C , ρ_c , the mass density of the crystal, K_j ($j = X, Y, Z$) is the principal thermal conductivity, W_x and W_y are the crystal widths in x and y directions, respectively, $H = K_3/h$ and h is the air heat transfer coefficient, K_3 is the thermal conductivity in direction z. The heat transfer equation 3a can be rewritten as:

$$\begin{aligned} \frac{\partial T}{\partial t} - \left(a_1^2 \frac{\partial^2 T}{\partial x^2} + a_2^2 \frac{\partial^2 T}{\partial y^2} + a_3^2 \frac{\partial^2 T}{\partial z^2} \right) \\ = F(x, y, z, t)F(x, y, z, t) = F'(x, y, z, t) + \delta a^2 \frac{\partial^2 T}{\partial x \partial z} \end{aligned} \quad (4)$$

The solution of equation 4 can be derived through a Fourier transform (equations 10, 11 in [8]).

Thus, a numerical solution at an arbitrary instant can be obtained by solving simultaneously the equation within a certain time interval until it forms a stable field distribution. The temperature distribution is sampled in the x-y-z space region as a lattice of $64 \times 64 \times 40$ lattices within $[-4w_0, 4w_0] \times [-4w_0, 4w_0] \times [0, L]$, where w_0 is the beam waist radius. The Fourier transformation method is used to solve equations 3 which are also discretized as 64×64 lattices in the x-y plane within $[-4w_0, 4w_0] \times [-4w_0, 4w_0]$. At the same time, the 4-order Runge-Kutta algorithm is utilized to ensure a certain precision, where the single-step error is controlled to be less than 10^{-4} .

Fig. 1. Schematic diagram of a focused beam incident into a uniaxial nonlinear crystal, where a rotation (θ) relation between the laboratory coordinate system (x, y, z) and the crystallographic directions (X, Y, Z) is shown.

3. Numerical simulation

In the following part, we explore the SHG (ee-o) coupling process in the ZnGeP_2 crystal pumped by a pulsed CO_2 laser at $9.6 \mu\text{m}$, with a Gaussian TEM_{00} mode.

For concision, the laser pulse is simplified as a square wave in time. The average pumping intensity in the cross section of the crystal, is $I(x, y) = I_0 \exp[-2(x^2 + y^2)/w^2]$, where I_0 is the power density at the focus, which is $I_0 = 2E_p/(\pi \cdot \Delta\tau \cdot w_0^2)$, E_p is the pulse energy, $\Delta\tau = 30 \text{ ns}$ is the pulse width with the repetition rate of 10 Hz . The average power intensity is taken as 100 mW/cm^2 , which is close to the damage threshold of ZnGeP_2 . At this level of pumping, a thermal lens effect will not occur. Since the temperature varies slowly comparing with light pulses, it is reasonable to assume that the heat source is independent of the pumping pulse shape. The absorption coefficients of the fundamental and harmonic beams are, respectively, taken as 0.2 cm^{-1} and 0.1 cm^{-1} (this is a middle absorption case), and the thermal conductivity is shown as $K_{\perp} = 35 \text{ W/(m}\cdot\text{K)}$, $K_{\parallel} = 36 \text{ W/(m}\cdot\text{K)}$ with the heat capacity $400 \text{ J/(kg}\cdot\text{K)}$ [19]. The refractive index and the temperature dependence come from the data in [19]. It gives the PM angle: $\theta = 1.17 \text{ rad}$ with the walk-off angle: $\rho_1 = 0.0093 \text{ rad}$. Considering different convections [20], we take the heat transfer coefficient as $10.0 \text{ W/(m}^2\cdot\text{K)}$ on a weak cooling condition but $200 \text{ W/(m}^2\cdot\text{K)}$ for a strong

cooling. Here, we introduce a strong cooling condition (equation 3b) in which a circulating water device dictates cooling, with the consideration of the side boundaries, and a weak cooling condition on which the side boundaries are almost adiabatic (equation 3c). It should be noted that the incident and exit surfaces are not cooled in the strong cooling case.

Under the strong and weak cooling conditions, the thermo-optic couplings in the SHG process have been simulated with 5 mm beam waist radius and 10 mm crystal length. Here, we use scaled coordinates $\xi = x/w_0, \eta = y/w_0, \sigma = z/L$ and the longitudinal axis is at the line $\xi, \eta = 4$. In the first case (strong cooling), the temperature varying throughout 15 s is shown in Fig. 2, sampled with the interval $\Delta t = 0.5 \text{ s}$. The evolutions of fundamental and harmonic beams are plotted in Fig. 3. The temperature distribution demonstrates weak gradient in transverse direction due to the large thermal conductivity as expected, while a rather flat distribution is found in the longitudinal direction which can be attributed to the uniform spreading of heat source in this direction. The time of temperature rising is about 4 s and a total stabilization should be achieved at a time over 10 s according to the variation of the central average temperature illustrated in the inset in Fig. 2b (the temperature is averaged over the z coordinate at the longitudinal axis). In the second case (weak cooling), relative simulation results are illustrated in Fig. 4 and 5 with the interval $\Delta t = 50 \text{ s}$ (within 100 s , a small time interval 5 s has been chosen, but there was no obvious difference in the final temperature distribution). The maximal temperature is almost 34 times of that calculated in the first case, as more severe impact has been exerted on SHG. Boundary temperature continuously increases with time restricted to the adiabatic condition. Since more heat energy is stored in the crystal, the stabilization time is greatly prolonged (nearly 230 times). However, the final distribution of temperature, subtracting at 40°C background, is similar to that of strong cooling.

In the first case, the SHG is hardly affected by the temperature (Fig. 3) and the stabilization of temperature finally brings stable coupling of the

Fig. 2. Temperature distribution in the (x, z) plane (a), transient temperature distribution along the x direction at $\eta = 4, \sigma = 1$ (b), along the longitudinal axis (c) under the strong cooling condition within/during 15 s. The inset in (b) illustrates the evolution of the central average temperature.

Fig. 3. The variations of fundamental and harmonic pulse energies inside the crystal (a), the evolution of fundamental (b) and harmonic (c) intensity profiles in the x direction ($\eta = 4, \sigma = 1$) within 15 seconds, under the strong cooling condition.

beams. The quality of the output harmonic beam almost keeps unchanged, which can be checked in Fig. 3a. This phenomenon does not occur in the second case which is shown in Fig. 5. Within 50 s, the conversion efficiency is much lowered by the

quick temperature rising. After 200 s, the SHG is totally ruined by the quick thermal effect. The temperature bandwidth in this case is about 48 °C, which means the temperature acceptance is about 24 °C (half of the bandwidth). Thus, the efficiency reduction is inevitable even in the weak cooling condition.

Simulation for different beam waists and pumping powers have been carried out as well as for a comparison. The temperature distribution and harmonic evolution are similar to those presented above. Their results only differ in the magnitudes of minimal and maximal temperatures and induced conversion reduction. Since the PM angle is small (0.0093 rad), the optical axis is nearly along the z coordinate. Then, the transverse thermal conductivity is dominated by $K_{X,Y}$. The thermal anisotropy is negligible. Taking the central average temperature as a simple reference, the scaled phase mismatching constant at time t can be written as:

$$\Delta k_a(t) = (2\tilde{\gamma}_1 - \tilde{\gamma}_2)\bar{T}(t)/\bar{f} \quad (5)$$

where $\bar{T}$ represents the central average temperature. Equation 5 represents the maximal phase mismatching constant induced by temperature variation. Then, the compensated phase mismatching constant should be $\Delta k = -\beta\Delta k_a$, where β is a positive number less than 1.0. In a compensated coupling process, the temperature increase is higher due to a larger absorption of the harmonic beam, so the phase mismatch should be compensated constantly to keep a maximal output. Since the temperature distribution is rather uniform inside the crystal, the phase compensation can have a quite good control on the efficiency restoration (the difference is less than 5 %).

4. Conclusions

In this paper, we have applied the thermo-optic coupling model for type I SHG to the popular infrared crystal: ZnGeP₂, under the strong and weak cooling conditions. We simulated some extracavity SHG processes of pulsed CO₂ laser at the wavelength of 9.6 μm. Relative demonstrations of the evolution of the temperature and beams were

Fig. 4. Temperature distribution in the (x, z) plane (a), transient temperature distribution along the x direction at $z = 4$, $\sigma = 1$ (b), along the longitudinal axis (c) under the weak cooling condition in the range of 3500 s. The inset in (c) illustrates the evolution of the central average temperature.

Fig. 5. The variations of fundamental and harmonic pulse energies inside the crystal (a), the evolution of fundamental (b) and harmonic (c) intensity profiles in the x direction ($\eta = 4, \sigma = 1$), within 3500 s, under the weak cooling condition.

analyzed. The numerical results infer that the presence of thermal effects can lead to a reduction of conversion efficiency as usual. Since ZnGeP_2 has a large thermal conductivity, the temperature gradient is relatively small inside the crystal, which implies that a strong cooling will have a good effect

in practice. The simulation implies that the phase compensation for the thermal effect in ZnGeP_2 crystal is still a good method to recover efficiency.

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