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Theoretical Analysis of Second Harmonic Generation Considering Laser Absorption with Repetitive Irradiation of Focused Beam

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In the frequency conversion, phase matching is essential to obtain the most effective efficiency. However the crystal temperature deviation causes undesirable conversion because the phase matching condition depends on the temperature. Frequency conversion efficiency is higher as the input intensity is higher. Thus the focused beam is usually used as input in order to obtain the higher efficiency, but the focusing also yields high temperature-rise and large temperature gradient near the focal spot; conversion efficiency decrease in the case that the degree of focusing is too much because of its large effect of thermal phase mismatching. Most of conventional theoretical-studies exclude effects of laser absorption and treated the problem as steady state although practical frequency conversion is non steady state problem with repetitive irradiation. We analyzed theoretically second harmonic generation considering laser absorption with repetitive irradiation of focused beam, and discussed the contribution of both the focused beam divergence and temperature distribution induced by repetitive irradiation to conversion efficiency. Based on these analysis results, we estimate the several thermal controls that utilize the internal temperature predicted. As a result we obtained larger enhancement in the control considering the internal crystal temperature.

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1. Introduction

Frequency conversion using nonlinear optical crystals is indispensable technique for laser precision microfabrications especially in application of solid state lasers. In the harmonic generation, phase matching is essential to obtain the most effective conversion efficiency [1]. However the phase matching condition depends on temperature. Thus a temperature change of crystal due to laser absorption causes thermal phase mismatching and yields undesirable conversion such as either decline or fluctuation of the efficiency and distortion of beam profile [2-4]. Therefore frequency conversion requires a thermal control, but it is unavoidable that temperature distribution is formed in the crystal even under a strict thermal control. This is the thermal problem on frequency conversions. On the other hand frequency conversion efficiency is higher as the input intensity is higher. Thus a focused beam is usually applied as input fundamental in order to obtain the higher efficiency. However the focusing also yields high temperature-rise and large temperature gradient near the focal spot; too tight focusing decreases conversion efficiency due to its large effect of thermal phase mismatching.

Elucidation of internal temperature distribution should be important in order to realize the more effective and stable conversion. An experimental measurement of internal temperature is very difficult especially in frequency conversions with focused beam. However most of conventional theoretical studies excluded the input fundamental depletion and effects of laser absorption [1, 5-7]. There are several studies considering laser absorption, but they are based on one pulse irradiation [8] or steady state problem on the assumption of low repetition rate [9]. Practical frequency conversion is non steady state problem with repetitive irradiation. We are not aware of any studies dealing with the thermal problem as non steady state.

In this paper we analyzed theoretically second harmonic generation (SHG) considering the influence of laser absorption with repetitive irradiation of focused beam. We constructed coupling model composed of SHG including laser absorption in the nonlinear optical crystal and the crystal temperature variation by heat conduction equation. Nd:YAG laser is supposed for input fundamental and $KTiOPO_4$ (KTP) crystal for nonlinear optical crystal. Contribution of both the focused beam divergence and temperature distribution induced by repetitive irradiation to conversion efficiency is investigated. Based on these analysis results, we estimate the several thermal controls that utilize the internal temperature predicted.

2. Analysis method

2.1 Phase matching condition

Phase matching condition on SHG is achieved when refractive index of crystal for fundamental is equal to that of second harmonic. Refractive index depends on a wavelength of light λ, a temperature of medium T and incident angle of light to medium θ. Each refractive index is expressed as, $n_{\omega}(\lambda_{\omega}, T, \theta)$, $n_{2\omega}(\lambda_{2\omega}, T, \theta)$ where subscripts ω, 2ω correspond to fundamental and second harmonic.

Phase matching is usually achieved through selecting the phase matching temperature T_{pm} before seeking the phase matching angle θ_{pm} that satisfies the equation; $n_{\omega}(T_{\mathrm{pm}}, \theta_{\mathrm{pm}}) = n_{2\omega}(T_{\mathrm{pm}}, \theta_{\mathrm{pm}})$.

2.2 Focused SHG including laser absorption

Two axisymmetric coordinate systems are applied to the nonlinear optical crystal in order to use a finite difference model as shown in **Fig. 1**. One is used for focused beam propagation (r, z) and the other is used for heat conduction (R, z). The coordinates system for focused beam whose beam waist is r_{F}, focal length is f_{F} and Rayleigh length is z_{R} is given by the variation of $1/e^2$ radius $r_e(z)$ as a hyperbolic function [10].

$$(z - f_{\mathrm{F}})^2 / z_{\mathrm{R}}{}^2 - r_e{}^2(z) / r_{\mathrm{F}}{}^2 = -1 \tag{1}$$

Using beam quality factor M^2 (M-squared) and beam divergence θ_{F}, we can express r_{F} and z_{R} as

$$r_{\mathrm{F}} = 2\mathrm{M}^2 / k \tan\theta_{\mathrm{F}}\ ,\ z_{\mathrm{R}} = 2\mathrm{M}^2 / k \tan^2\theta_{\mathrm{F}} \tag{2}$$

where k is wave number.

Input fundamental intensity $I\,(r, 0)$ is supposed that the spatial distribution is Gaussian.

$$I(r,0) = \frac{2E_{\mathrm{p}}}{\pi r_e(0)^2 \tau_{\mathrm{p}}} \exp\left[-2\left(r/r_e(0)\right)^2\right] \tag{3}$$

where E_{p} is pulse energy, τ_{p} is pulse duration (FWHM).

When the grids of focused beam coordinates are enough fine, the following governing equation of one-dimensional SHG including laser absorption can be applied to each lattice [8].

$$\begin{cases} \dfrac{dA_{\omega}(z)}{dz} = -i\eta_{\omega}\omega d_{\mathrm{eff}} A_{\omega}^{*}(z) A_{2\omega}(z) \exp\left[-\alpha_{2\omega} z\right] \exp\left[-i\Delta k z\right] \\ \dfrac{dA_{2\omega}(z)}{dz} = -i\eta_{2\omega}\omega d_{\mathrm{eff}} A_{\omega}^{2}(z) \exp\left[\left(\alpha_{2\omega} - 2\alpha_{\omega}\right) z\right] \exp\left[i\Delta k z\right] \end{cases} \tag{4}$$

where i is imaginary unit, ω is angular frequency of fundamental, d_{eff} is nonlinear coefficient, A is complex amplitudes, η is impedance of medium, 2α is absorption coefficient. The parameter Δk indicating the degree of phase matching, and is given by

$$\Delta k = 2\omega\left(n_{2\omega} - n_{\omega}\right)/c_0 \tag{5}$$

where c_0 is light velocity. The relationship between complex amplitude A and its intensity I is given by

$$I = \left|A^2\right| / 2\eta \tag{6}$$

Eq. (4) can be solved numerically for the lattice (i, k) by using a discrete intensity at z_{k-1}; $I_{\omega,\,i,\,k-1}$, $I_{2\omega,\,i,\,k-1}$ as the initial value. This gives the intensities at z_k which are treated as plane waves; $I'_{\omega,\,i,\,k}$, $I'_{2\omega,\,i,\,k}$, and internal heat generated in the lattice (i, k); $w_{i,k}$, as the decay of total intensities.

$$w_{i,k} = \left[\left(I'_{\omega,\,i,\,k} + I'_{2\omega,\,i,\,k}\right) - \left(I_{\omega,\,i,\,k} + I_{2\omega,\,i,\,k}\right)\right] / \Delta z_k \tag{7}$$

Then the modified intensities; $I_{\omega,\,i,\,k}$, $I_{2\omega,\,i,\,k}$, are calculated considering the focusing effect. These intensities are used as the initial value for the next lattice (i, k+1). For the first lattice (i, 1), discrete input intensities derived from Eq.(4) is used as the initial value. The above procedure is repeated along crystal length z in each i component, consequentially the distribution of intensities; $I_{\omega,\,i,\,k}$, $I_{2\omega,\,i,\,k}$, and internal heat; $w_{i,k}$, in the focused beam coordinates are derived as a result of one-pulse irradiation.

Conversion efficiency is evaluated by the ratio of second harmonic energy at the length z; $E_{2\omega}(z)$, to input fundamental energy; $E_{\omega}\,(0)$ (= E_{p}). $E_2\,(z)$ is obtained by integration of $I_{2\omega}\,(r, z)$ in the radial and time direction.

2.3 Repetitive irradiation

Applying the coordinate transformation to internal heat for focused beam coordinates; $w_{i,k}$, gives internal heat for heat conduction coordinates; $w_{j,\,k}$. Temperature distribution in the crystal after the pulse irradiation is described by the two-dimensional non-steady heat conduction equation with internal heat $w_{j,\,k}$. Because there is no laser absorption between the pulse and the next pulse, the heat conduction equation is solved without internal heat. When the next pulse is irradiated, the refractive indices n_{ω} and $n_{2\omega}$ are calculated using the temperature given by just before this pulse. The indices change at this time due to their temperature dependence, Eq. (4) is solved for the next pulse with this state.

This procedure composed of SHG including laser absorption and the crystal temperature variation is repeated with repetition rate f_{p} during irradiation time.

2.4 Analysis conditions

We supposed Nd:YAG (wavelength is 1064 nm) for the input fundamental and KTP ($\mathrm{KTiOPO_4}$) for the nonlinear optical crystal. Physical properties of KTP are tabulated in **Table 1** [1, 11, 12]. The analysis conditions without the beam divergence are fixed as **Table 2**. This is assumed that phase matching condition is satisfied at the beginning of irradiation. The beam divergence is varied from 3 to 10 deg as shown in **Fig. 2**. Heat transfer to the ambient whose temperature is room temperature 293 K is adopted as boundary condition. The heat transfer coefficient is supposed to be 10 W/m^{2}K in static air.

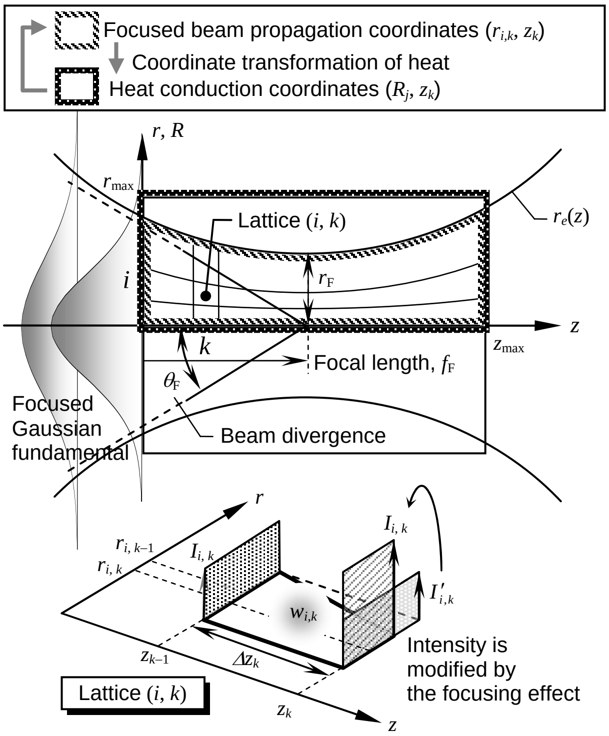


Fig. 1 Analysis model

Table 1 Physical properties of KTP

Absorption coefficient	
$2\alpha_1$ (fundamental, 1064 nm)	0.5 [m^{-1}]
$2\alpha_2$ (second harmonic, 562 nm)	4.0 [m^{-1}]
Density ρ	$2.945\text{x}10^3$ [kg/m^3]
Specific heat c (at 293 K)	$6.85\text{x}10^2$ [J/kgK]
Thermal conductivity	
K_r (r direction)	3.064 [W/mK]
K_z (z direction)	2.173 [W/mK]
Effective nonlinearity d_{eff}	$2.15\text{x}10^{-23}$ [F/V]

Table 2 Analysis condition

Crystal: KTP, Laser: Nd:YAG	
Crystal radius $r_{\max}$	2 [mm]
Crystal length $z_{\max}$	5 [mm]
Focal length f_F	2.5 [mm]
Pulse Energy E_p	5 [mJ]
Pulse duration τ_p	50 [ns]
Beam quality factor M^2	10
Beam divergence θ_F	3 ~ 10 [deg]
Repetition rate f_p	2 [kHz]
Phase matching temperature T_{pm}	293 [K]
Initial temperature T_0	293 [K]
Phase matching angle θ_{pm}	24.6134 [deg]

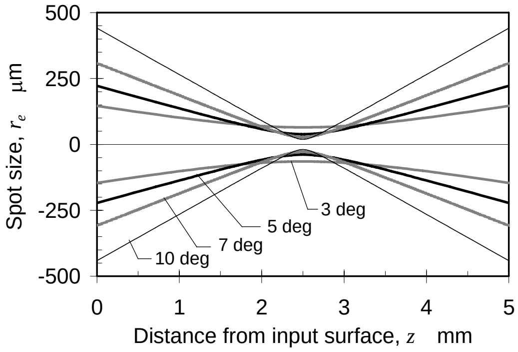


Fig. 2 The variation of spot size with the crystal length for the various beam divergences

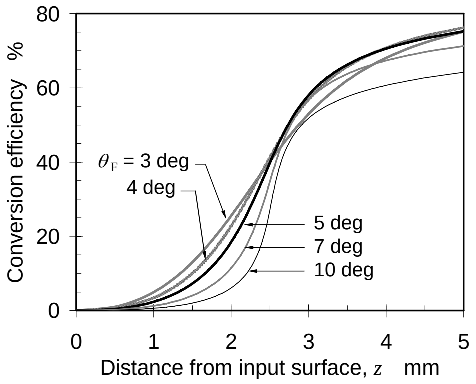


Fig. 3 The dependence of the beam divergence on conversion efficiency distribution along the crystal length obtained by one pulse irradiation with phase matching condition

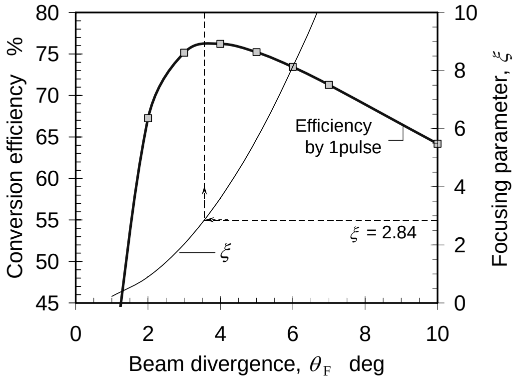


Fig. 4 The dependence of beam divergence on the output conversion efficiency obtained by one pulse irradiation with phase matching condition and on focusing parameter ξ; the ratio of crystal length to the confocal length

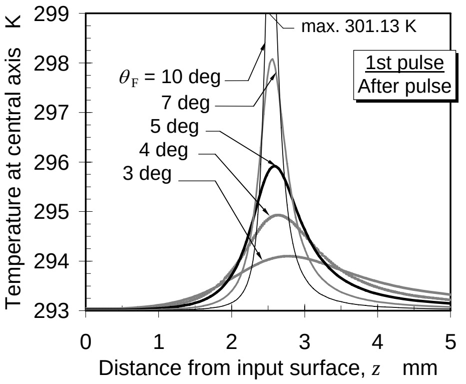
3. Results and discussions

3.1 One pulse irradiation without thermal phase mismatching condition

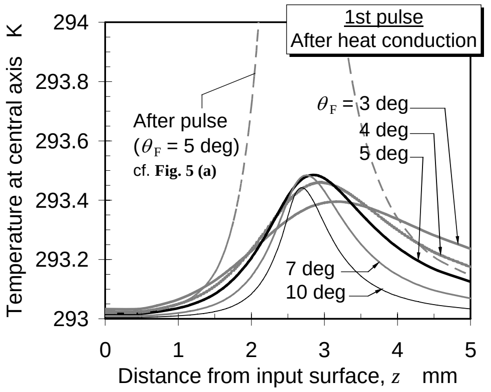
This section describes a SHG including laser absorption by one focusing pulse irradiation without thermal phase mismatching. **Figure 3** shows the dependence of the beam divergence on conversion efficiency distribution along the crystal length. Conversion efficiency is generally higher as the input intensity is higher in harmonic generations. Larger beam divergence leads its intensity to be higher at the focal point, but lower around the input and output side of the crystal. Thus the rise of efficiency is slow around the input side, is very quick around the focal point, and is slow again around the output side. In the case of smaller beam divergence, efficiency rises slowly through the crystal compared with larger one. As a result, output efficiencies (at which z = 5 mm; crystal length $z_{\max}$) are obtained as shown in **Fig. 4**. It shows that the highest efficiency is observed at about 3.5 deg beam divergence. It has been already reported by Boyd et al. that the optimum efficiency is obtained at where the focusing parameter; the ratio of crystal length to the confocal length, is 2.84 [8]. Figure 4 also contains this focusing parameter, and our result shows good agreement. However this is a result under the ideal condition that is a steady state without thermal effect.

3.2 Temperature distribution formed by one pulse irradiation

A pulse irradiation as described in the previous section induces temperature rise due to laser absorption. **Figure 5 (a)** shows temperature distribution along the central axis of the crystal after one pulse irradiation. Temperature rise is a little higher at the output side than at the input side because the absorption coefficient of second harmonic is about eight times as high as that of fundamental in KTP [1]. In addition, higher temperature at focal point is observed as the beam divergence is larger. Temperature distribution which affects frequency conversion directly is not after the pulse irradiation but before the next pulse irradiation. As a result of heat conduction during the interval between the 1st and 2nd pulses (500 μs under 2 kHz repetition rate), temperature distribution is diffused as shown in **Fig. 5 (b)**. The difference of temperature at focal spot between beam divergences almost disappears by the heat conduction.



(a) After the pulse irradiation



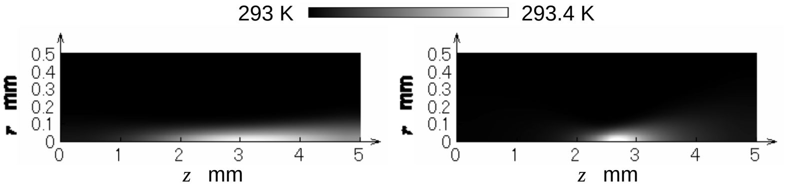
(b) After the heat conduction

Fig. 5 Temperature distribution along the central axis of the crystal after 1 pulse irradiation and before the 2nd pulse irradiation (after the heat conduction during the interval between the 1st and 2nd pulses)

Figure 6 shows temperature distribution near the central axis of crystal (r < 0.5 mm) corresponding to Fig. 5 (b). It is found that a relatively gradual temperature gradient is induced along the direction of crystal length z in the case of larger beam divergence. When the 2nd pulse is irradiated, second harmonic is generated with thermal phase mismatching caused by temperature distribution as like Fig. 6.

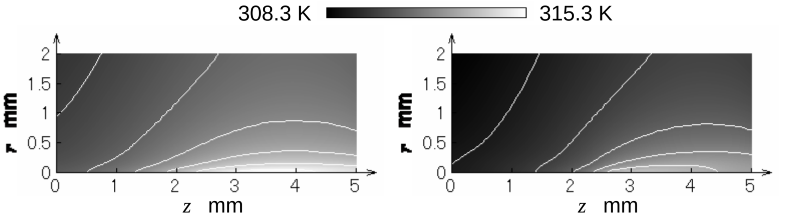
3.3 Time variation of the output conversion efficiency and temperature of the crystal with repetitive irradiation

Temperature distribution is affected by both the heat accumulation and the heat conduction during repetitive irradiation. **Figure 7** shows temperature distribution in the crystal at irradiation time 2 min. It is the time before the 240,000th pulse irradiation because the repetition rate is 2 kHz. The difference of the temperature gradient appeared in Fig. 6 become a little smaller in Fig. 7 by the heat conduction. The overall temperature of the crystal is higher compared with Fig. 6 by the heat accumulation. The average temperature is 311.08 K in Fig. 7 (a) and 309.78 K in Fig. 7 (b). In addition, the output conversion efficiency with the state of each temperature distribution is 47.16 % and 56.60 %, respectively. Plotting these average temperatures and the efficiencies as a function of the irradiation



(a) θ_F = 3 deg **(b)** θ_F = 10 deg

Fig. 6 Temperature distribution near the central axis of crystal (r < 0.5 mm) before the 2nd pulse irradiation



(a) θ_F = 3 deg
(Average temp. is 311.08 K,
output efficiency is 47.16 %)

(b) θ_F = 10 deg
(Average temp. is 309.78 K,
output efficiency is 56.50 %)

Fig. 7 Temperature distribution in the crystal at irradiation time 2 min (before the 240,000th pulse irradiation). Contours are every 1 K.

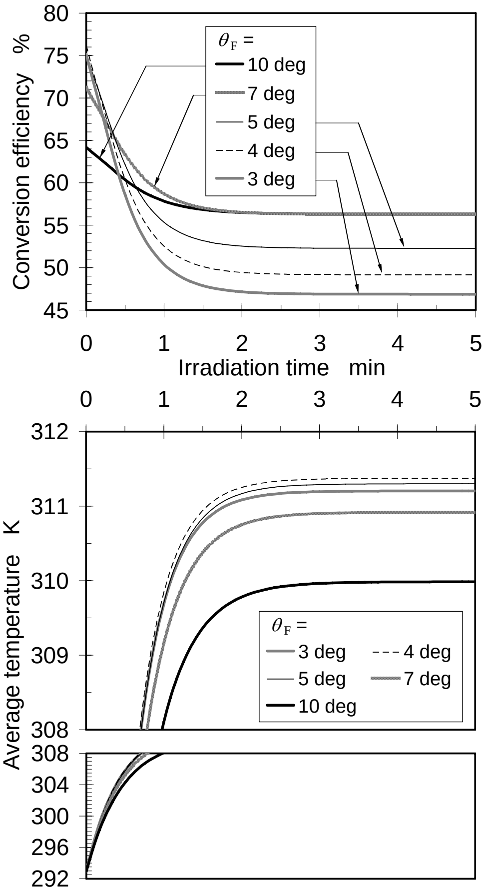


Fig. 8 Time variation of output conversion efficiency average temperature of crystal with irradiation time

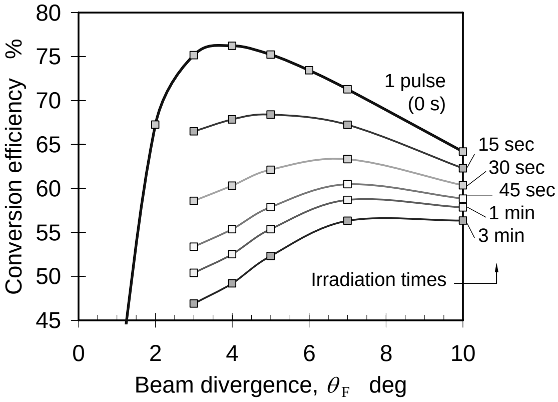


Fig. 9 The dependence of beam divergence on the output conversion efficiency obtained at each irradiation time

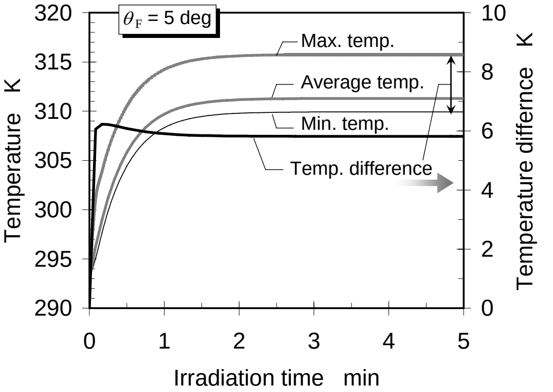


Fig. 10 Time variation of the maximum, minimum, average temperature of the crystal (left ordinate) and the difference between maximum and minimum temperature (right ordinate) under the beam divergence θ_F = 5 deg

time gives **Fig. 8**. Both the average temperature and the efficiency become almost constant after the irradiation time 2 min under the present conditions. The highest efficiency is obtained by 7 deg and 10 deg beam divergence. Adding this time variation of efficiencies to Fig. 4; the beam divergence dependence on efficiency, yields **Fig. 9**. The result shows that the optimum beam divergence changes with irradiation time. It is concluded that an analysis based on a steady state is not enough for the investigation of practical frequency conversions such as under the repetitive irradiation.

3.4 Evaluation of a thermal control using the calculated internal temperature distribution

This section describes a thermal control using the calculated temperatures in the previous section. Adjusting the phase matching temperature T_{pm} to the temperature at enough irradiation (e.g., 5 min) may be effective for a high and stable harmonic generation. Temperature is obvious only at a surface of crystal in terms of a possible measurement experimentally. However temperature in the crystal is higher than that at surface; the maximum temperature tends to appear near the focal spot and the minimum does at a crystal surface such as shown in Fig. 7. **Figure 10** shows time variation of the average temperature (already shown in Fig.8) and of the maximum, minimum temperature in the crystal under the beam divergence θ_F = 5 deg. The difference between maximum and minimum temperature is also plotted. When the temperatures are stable, the maximum temperature is 315.7 K and the minimum temperature is 310 K. Therefore we estimated the two control conditions as followings: phase matching temperature is adjusted to maximum temperature 315.7 K, and minimum temperature 310 K. Expression of conversion efficiency obtained by the former control is expressed as $\eta_{T\max}$, the latter $\eta_{T\min}$ and no controlled η_0 (already shown in Fig.8). The result of them is **Fig. 11**. Both the two control conditions achieve higher and stable conversion compared with no controlled condition though their efficiencies at the begging of irradiation are lower. The control considering the internal crystal temperature (T_{pm} = 315.7 K) has a larger enhancement by about 5 % than the surface temperature (T_{pm} = 310 K).

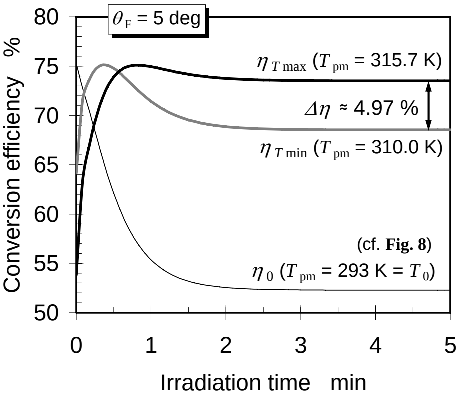


Fig. 11 Time variation of output conversion efficiencies of the various thermal control conditions under the beam divergence θ_F = 5 deg. η_0: The phase matching temperature is the initial temperature of crystal T_0, without control (= 293 K). $\eta_{T\max}$ and $\eta_{T\min}$: The phase matching temperature is maximum and minimum temperature of crystal after enough irradiation (= 315.7 K and 310 K).

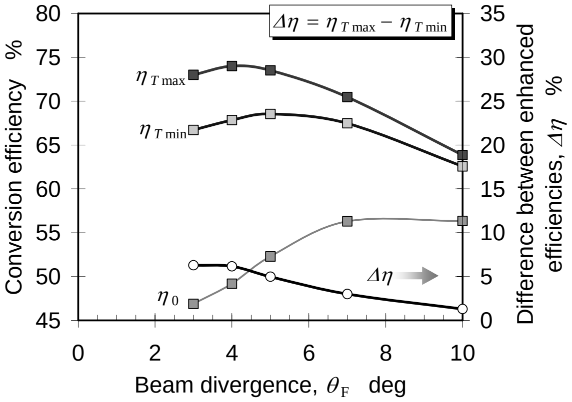


Fig. 12 The dependence of beam divergence on the difference between enhanced output conversion efficiencies after enough irradiation

Similar controls are applied to the other conditions of beam divergence. The control considering the internal crystal temperature $\eta_{T\max}$ shows higher improvement in all conditions as **Fig. 12**. The difference between enhanced efficiencies $\Delta\eta$ becomes higher as smaller beam divergence. The reason is considered that an influence where the temperature rises is larger as the beam divergence is smaller.

Conclusions

We analyzed theoretically second harmonic generation (SHG) considering the influence of laser absorption with repetitive irradiation of focused beam as non steady state problem. We constructed coupling model composed of SHG including laser absorption in the nonlinear optical crystal and the crystal temperature variation by heat conduction equation. KTP crystal is supposed for nonlinear optical crystal. Focused Nd:YAG laser is supposed for input fundamental wave and its beam divergence is varied from 3 to 10 deg. Contribution of both the focused beam divergence and temperature distribution induced by repetitive irradiation to conversion efficiency is investigated.

The optimum beam divergence using the focusing parameter was found to depend on the irradiation time. Therefore an analysis based on a steady state is not enough for the investigation of practical frequency conversions.

In addition, we estimate the several thermal controls based on the above analysis results. The larger enhancement of the conversion efficiency was theoretically obtained by using the calculated internal crystal temperature which is unable to be measured experimentally, compared with the surface temperature. The improvement has a tendency to appear in smaller beam divergence. Such a control is expected to be more effective for frequency conversion with high power lasers.

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