Evaluation of Second Order Nonlinearity in Periodically Poled KTiOPO₄ Crystal Using Boyd and Kleinman Theory

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Focusing lens

Fundamental wave (860 nm)

Second harmonic wave (430 nm)

Abstract—The effective nonlinearity E_{NL} of periodically poled KTiOPO₄ crystal is precisely measured with SHG experiment at various focusing conditions. The result shows the optimum focusing condition which maximizes the E_{NL} at focusing parameter ξ at around 2.8 and is consistent with Boyd and Kleinman theory. The analysis based on the theory shows the nonlinear optical coefficient d_{33} of 17.5 [pm/V].

I. INTRODUCTION

The second order nonlinearity in nonlinear optical crystal is very important effect for many applications. One of the important utilization of the second order nonlinear effect is efficient generation of second harmonic wave. There are many examples of previous research about second harmonic generation (SHG). Another important utilization is optical parametric process in order to generate squeezed states of light. Squeezed light is an important resource for quantum optics and quantum information processing with continuous variables [1]. In both cases, high nonlinearity is important factor.

A periodically poled KTiOPO₄ (PPKTP) is one of the promising nonlinear optical crystal because of its high nonlinearity. In a recent work of SHG, Targat, *et al.* achieved 75% of conversion efficiency at 461 nm by using a PPKTP crystal in an external resonant cavity with a bow-tie configuration [2]. In recent experiments for generating highly squeezed light with subthreshold optical parametric oscillator (OPO), Suzuki, *et al.* -7.2 ± 0.14 dB [3] and Takeno, *et al.* achieved -9.01 ± 0.14 dB [4] of squeezing at 860 nm by using a PPKTP crystal, respectively. In both cases, efficient SHG and squeezing experiments, high nonlinearity is very important factor.

A typical indication resulting from second order nonlinear effect is the effective nonlinearity E_{NL} (W⁻¹) which is defined by

$$E_{NL} = \frac{P_{2\omega}}{P_{\omega}^2} \tag{1}$$

where P_{ω} and $P_{2\omega}$ are the power of fundamental and second harmonic wave respectively. It is shown that the conversion efficiency with an external cavity configuration depends largely on the E_{NL} [5]. In case of squeezing

Fig. 1. Schematic diagram of experimental setup for second harmonic generation.

PPKTP

experiment by using an OPO, an oscillation threshold is one of the important parameter and related with the E_{NL} [6].

The purpose of this work is to measure the E_{NL} of PPKTP crystal at various focusing conditions and find out the optimum focusing. The experimental results are analyzed by utilizing famous Boyd and Kleinman (BK) theory which gives precise description of SHG with focused Gaussian light beams [7]. And the second order nonlinear optical coefficient d_{33} is also derived from theoretical calculations.

II. EXPERIMENTAL SETUP AND CONDITION

A schematic of experimental configuration is shown in Fig. 1. A continuous-wave Ti:Sapphire laser at 860 nm is used as a fundamental wave. Spatial transverse mode of the laser beam is Gaussian mode. The 860 nm beam is focused with a plano-convex lens (productions of Thorlabs Inc.) and introduced into a PPKTP crystal (production of Raicol Crystals Ltd.) with 10 mm of length l and $1*1 \text{ mm}^2$ of cross section. The position of the beam waist is aligned to be located at almost center of the crystal. Both crystal surfaces are highly anti-reflective coated at 860 nm and 430 nm. The crystal assembly is attached with a Peltier device and temperature controlled at 40°C for achieving phase matching condition. Second harmonic wave generated at the PPKTP crystal is separated by a harmonic separator which highly reflects 430 nm beam and transmits unconverted 860 nm beam. The power of second harmonic wave is detected by optical sensor (Advantest Q82017A).

Harmonic

separator

Optical

power meter

Focusing length	Beam waist size w_0	Focusing parameter ξ	E_{NL}
(mm)	(µm)		(W^{-1})
50.6	6.5	17.36	0.0137
75.7	9.8	7.77	0.0256
101.0	13.0	4.38	0.0331
126.4	16.3	2.81	0.0348
151.7	19.5	1.95	0.0351
176.8	22.7	1.44	0.0322
202.1	25.9	1.11	0.0267
252.7	32.3	0.71	0.0195
303.1	38.7	0.50	0.0141
404.2	51.5	0.28	0.0081
505.4	64.1	0.18	0.0056

TABLE I SUMMARY OF EXPERIMENTAL CONDITIONS

The SHG experiment is conducted with various focusing conditions which are realized by changing focusing length of lenses. Table. I summarizes experimental conditions of focusing length of lenses, beam waist size w_0 in radius, focusing parameter ξ described later in Eq.(2), and experimental results of E_{NL} at each conditions. The accurate focusing length of lenses at 860 nm is calculated by using radius of curvature of lenses and refractive index of lens material (=1.510 :BK7 glass). Gaussian beam waist size w_0 at the center of the crystal is calculated by analysis of ABCD matrix which consists of optical elements with an incident beam size of 2.1 mm. The focusing parameter ξ is defined in the BK-theory as

$$\xi = l/b \tag{2}$$

where l is crystal length and b is confocal parameter of the Gaussian beam. The b is calculated from beam waist size w_0 and wave vector of fundamental wave k_{ω} as

$$b = w_0^2 k_\omega \tag{3}$$

where k_{ω} is expressed as

$$k_{\omega} = \frac{2\pi}{\lambda_{\omega}} n_{\omega} \tag{4}$$

with the refractive index of nonlinear optical crystal n_{ω} (=1.840) at fundamental wavelength λ_{ω} (=860 nm). It is obvious that the confocal length defined by Eq.(3) is twice the Rayleigh length of Gaussian beam.

III. EXPERIMENTAL RESULTS AND THEORETICAL ANALYSIS

Fig. 2 shows one of the typical SHG measurement which is obtained at focusing condition of $w_0 = 16.3$ μ m, ($\xi = 2.81$). The horizontal axis is fundamental wave power P_{ω} observed before the PPKTP crystal. The SHG power $P_{2\omega}$ increases as parabolic curve with the P_{ω} . And the conversion efficiency η which is calculated from a relation of $P_{2\omega}/P_{\omega}$ increases in direct proportion to the P_{ω} . These property agrees well with a consequence from conventional nonlinear optics. The η can be described in association with E_{NL} as following





Fig. 2. Experimental result of single pass frequency doubling. Horizontal axis is input power of fundamental wave. Circles and squares indicate the power of second harmonic wave at 430 nm and the conversion efficiency respectively. Solid lines are calculation results with Eq. (5)and (6), respectively.

In the same way, the $P_{2\omega}$ is expressed as

$$P_{2\omega} = E_{NL} P_{\omega}^2. \tag{6}$$

Solid lines in Fig. 2 are calculation results with Eq. (5) and (6) respectively with 0.0348 (W⁻¹) of E_{NL} . By this means, the E_{NL} at various focusing condition is evaluated from the coefficient of fitting curve.

Similar SHG experiments are repeated at various focusing conditions summarized in Table. I. Fig. 3 shows summary of experimental results of the E_{NL} versus (i) beam waist size w_0 and (ii) focusing parameter ξ , respectively. It is noteworthy that there is optimum focusing condition which mazimizes the E_{NL} . In order to verify these property, experimental results are analyzed with the BK-theory. Solid lines in Fig. 3 (i) and (ii) represent calculation results based on the BK-theory. Theoretical description of the E_{NL} in MKS unit is given by

$$E_{NL} = K l k_{\omega} h_m(\xi) \tag{7}$$

where constant parameter K is

$$K = \frac{2\omega^2 d_{eff}^2}{\epsilon_0 n_\omega^2 n_{2\omega} c^3 \pi}.$$
(8)



Fig. 3. Summary of experimental results under various focusing condition. (i) and (ii) represent E_{NL} dependence on beam waist size w_0 and focusing parameter ξ , respectively. Circles indicate experimental results and solid lines are calculation results from Boyd and Kleinman theory based on Eq.(7).

The $h_m(\xi)$ in Eq. (7) is the optimized BK-factor which is explained in detail in appendix. In Eq.(8) the ϵ_0 is vacuum permittivity (= 8.85×10^{-12} [F/m]), the c is speed of light (= $3 * 10^8$ [m/s]), and the $n_{2\omega}$ (=1.938) is refractive index at second harmonic wave length of 430 nm. The effective nonlinear optical coefficient d_{eff} in Eq. (7) is calculated from nonlinear optical coefficient d_{33} of KPT crystal with following relation $d_{eff} = (2/\pi)d_{33}$. Solid lines in Fig. 3 are calculation results with nonlinear optical coefficient d_{33} of 17.5 [pm/V] and shows good agreement with experimental results. In the BK-theory it is reported that the $h_m(\xi)$ has has maximum value of 1.068 at $\xi = 2.84$. Experimental results shows the optimum E_{NL} is achieved at around 2.8 of ξ . These results agree well with the consequence from the BKtheory. Moreover, the value of d_{33} is very close to previously reported value of 16.5 [pm/V] by Shoji, et al. in KTP crystal at 852 nm [8].

IV. CONCLUSION

In conclusion, the effective nonlinearity E_{NL} of PP-KTP crystal is precisely measured with SHG experiment at various focusing conditions. The result shows the optimum E_{NL} is achieved with focusing parameter ξ at around 2.8 and agree well with the BK-theory. The nonlinear optical coefficient d_{33} is evaluated as 17.5 [pm/V] based on the BK-theory and very closed to previously reported value by Shoji, *et al.* [8].

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APPENDIX

The expression of the optimum Boyd and Kleinman factor $h_m(\xi)$ is derived theoretically in appendix. In original theory the BK-factor h is given as a function of all the parameters σ , β , κ , ξ , and μ which can be optimized. In experiments of this work, the double refraction parameter β is zero, since both fundamental and second harmonic are ordinary waves and have same polarization in a quasi phase matching configuration. Therefore, there is no walk-off between both waves in a crystal. The absorption parameter κ is assumed to be zero, since the power of both fundamental and second harmonic is low level in this experiment. The focusing position parameter μ is also expected to be zero. The focusing position is aligned at almost center of the crystal while experiments. So, the h is expected to be a function of the only σ and ξ as below

$$h(\sigma,\xi) = \frac{1}{4\xi} \iint_{-\xi}^{\xi} \frac{\exp\left[i\sigma\left(\tau - \tau'\right)\right]}{\left(1 + i\tau\right)\left(1 - i\tau'\right)} d\tau d\tau'.$$
(9)

The Eq.(9) can be easily translated to following equation

$$h(\sigma,\xi) = \frac{1}{4\xi} \left(\int_{-\xi}^{\xi} \frac{\cos\sigma\tau + \tau\sin\sigma\tau}{1 + \tau^2} d\tau \right)^2.$$
(10)

The phase mismatching parameter σ is expressed as

$$\sigma = \frac{1}{2}b\Delta k \tag{11}$$



Fig. 4. Integrand in Fq.(10) as a function of τ at various σ .



Fig. 5. Summary of calculation results of the function h at various σ . The envelope of various h curves corresponds to the optimum BK-factor $h_m(\xi)$ which yields the maximum E_{NL} at certain value of ξ .

where mismatching in wave vectors Δk is given as

$$\Delta k = 2k_{\omega} - k_{2\omega}.\tag{12}$$

The integrand in Eq.(10) as a function of τ is shown in Fig. 4 at various phase mismatching parameters σ . It is noteworthy that the value of function increases at the



Fig. 6. Calculation result of optimum phase mismatching parameter σ_m which yields the optimum Boyd and Kleinman factor $h_m(\xi)$.

lower τ and sinusoidally varies at the higher range of τ by increasing the σ . So there is possibility that the BK-factor h improves by optimizing the σ . Fig. 5 shows calculation results of the h as a function of ξ at various σ . The optimized Boyd and Kleinman factor $h_m(\xi)$ corresponds to the envelope curve in Fig. 5 and yields the maximum E_{NL} at certain value of ξ . This envelope is identical to the curve with the condition of B=0 in Fig.2 in ref. [7].

The optimum σ (= σ_m) which attains the optimum $h_m(\xi)$ is shown in Fig. 6. Experimentally the optimum σ_m is achieved by fine tuning of the phase matching temperature or adjustment of the crystal orientation in order to maximize the SHG power. The mechanism that the *h* improves with increasing the phase mismatching parameter σ from zero to one is not clear for the author yet.