Journal of the Optical Society of Korea, Vol. 2, No. 2, September 1998, pp. 54-57

Birefringence measurements of 1mol%Mg:LiNbO$_3$ with Noncollinear-phase-matching cone

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(Received June 26, 1998)

A noncollinear-phase-matching cone of second harmonic generation(SHG) was observed in a LiNbO$_3$ crystal doped with 1 mol% MgO. Birefringence refractive indices can be accurately evaluated by analysing the temperature phase matching characteristic for SHG combined with the measurement of the half cone angle. The electro-optic coefficient can also be determined from the observed change of the half cone angle when a DC electric field is applied along the optic axis.

I. INTRODUCTION

Giordmaine reported the first observation of a second harmonic conical ring with Potassium dihydrogen Phosphate (KDP) in 1961 [1]. This effect was explained as the phase matched interaction for second harmonic generation between the on-axis field of the incident pump beam and the off-axis field scattered from internal or surface scattering centers. Bates developed the quantitative analysis to describe noncollinear-phase-matching in negative uniaxial crystals and verified the Giordmaines hypothesis experimentally [2]. He calculated the temperature dependence of birefringence and the phase matching temperature from the temperature dependence of the half cone angle. It was also found that only one cone is produced in negative uniaxial crystals and two cones are produced in biaxial crystals[3] and this effect was observed even in organic crystals[4] and photorefractive materials(polymer[5], crystals[6-8]).

LiNbO$_3$ (Lithium niobate: LN) is an attractive nonlinear optical crystal that has been used for second harmonic generation (SHG) as well as optical integrated circuits. The birefringence parameters such as birefringence refractive indices and the electro-optic (EO) coefficient are important for applications of LN. However, the measurement of refractive indices requires the fabrication of accurate prisms[9-12] and the EO coefficients can be measured by using either the crossed polarizer method[13-16] or the phase-matching shift method[17-19] for which the experimental set-up is relatively complicated.

In the present work, we demonstrate a simple method to measure all the important parameters of birefringence by analysing the noncollinear-phase-matching cone and temperature-tuning characteristics of SHG. Some birefringence measurements in LN doped with 1mole% MgO are reported for the first time.

II. THEORY

When an incident light wave propagates in a medium, it can be scattered by a surface or internal scattering center. When the vector phase matching condition of second harmonic generation (SHG) between the incident wave and the scattered wave is satisfied, the second harmonic will be generated. Type I (o + o → e) phase matching is possible in LN because LN is a negative uniaxial crystal. Due to the momentum conservation in this nonlinear optical interaction, the angle phase matching condition can be written as

$$\mathbf{k_1} + \mathbf{k_2} = \mathbf{k_3} \tag{1}$$

where $\mathbf{k_1}$ and $\mathbf{k_2}$ are the wave vectors for the fundamental and scattered waves of frequency ω, and $\mathbf{k_3}$ is the wave vector of the second harmonic wave of frequency 2ω [2]. Consider the case where the fundamental beam of frequency ω propagates through the crystal normal to both the c-axis(optic axis) and the y-axis as shown in Fig. 1. The vector Eq.(1) can be decomposed into x- and y- components in the plane normal to the optic axis, i.e.,

$$k_3\ cos\theta_2 = k_1 + k_2\ cos\theta_1\ , \tag{2}$$

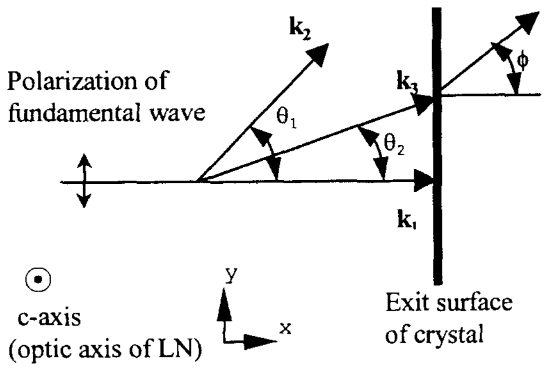


FIG. 1. Schematic diagram for noncollinear SHG in 1%Mg:LiNbO3.

$$k_3\ sin\theta_2 = k_2\ sin\theta_1\ . \tag{3}$$

From Eqs. (2) and (3), we obtain

$$cos\theta_2 = n_e(2\omega)/n_o(\omega), \tag{4}$$

where $n_e(2\omega)$ is the extraordinary index of refraction at the second harmonic frequency and $n_o(\omega)$ is the ordinary index of refraction at the fundamental frequency. By combining Eq.(4) with Snell's law, one can show that the half cone angle (ϕ) outside the crystal is given by the following expression, i.e.,

$$sin^2\phi = [\frac{n_e(2\omega)}{n_o(\omega)}]^2\ [n_o(\omega)^2 - n_e(2\omega)^2]. \tag{5}$$

Provided that $\Delta n = n_o(\omega) - n_e(2\omega) \ll 1$, Eq.(5) can be approximated as

$$sin^2\phi = 2n_o(\omega)\Delta n. \tag{6}$$

This result indicates that one can determine n_o and n_e if one knows Δn and ϕ, which can be directly measured from noncollinear-phase-matching cone.

The temperature dependence of birefringence can be investigated from the temperature phase matching characteristic of SHG [19]. In the case of negligible pump depletion, the intensity of SHG, $I_2(2\omega)$, with the crystal length L can be written as[20]

$$I_2(2\omega) \propto I(\omega)^2 L^2 \left[\frac{sin(\Delta kL/2)}{(\Delta kL/2)}\right]^2, \tag{7}$$

where $I(\omega)$ is the intensity of incident light with the frequency ω and $\Delta kL/2$ is phase change parameter. Here, Δk can be written as

$$\Delta k = \frac{\omega}{c}\Delta n = \frac{\omega}{c}(T - T_{pm})\frac{\partial \Delta n}{\partial T}, \tag{8}$$

where T_{pm} and $\frac{\partial \Delta n}{\partial T}$ are the noncritical phase matching temperature and the thermo-birefringence coefficient for SHG. Eq(8) indicates that Δn can be evaluated from the temperature phase matching characteristic of SHG.

Furthermore, when the DC electric field, E_o is applied parallel to the c-axis of the LN crystal, an additional birefringence change of refractive index, Δn^E, will be introduced as a linear function of E_o, i.e.,

$$\Delta n^E = n_o^E - n_e^E = r_{eff}E_o, \tag{9}$$

where r_{eff}=$(r_{13}n_o^3 - r_{33}n_e^3)/2$, and r_{13} and r_{33} are electro-optic coefficients. As a result, the half cone angle will be changed when E_o is applied. By combining Eqs. (6) and (9), we have

$$sin^2\phi_E - sin^2\phi = 2n_o(\omega)r_{eff}E_o, \tag{10}$$

where ϕ_E is the half cone angle outside crystal with the DC electric field applied parallel to the c-axis.

III. EXPERIMENT AND RESULTS

The MgO-doped LiNbO3 single crystals were grown along the c-axis by the Czochralski technique from melt in a Pt crucible. The starting materials were high purity Li_2CO_3(5N), Nb_2O_5(5N), and MgO(4N). The mixed powder was of congruent composition with Li/Nb = 48.6/51.4 and contained 1 mol% MgO. The grown crystals were annealed at 1200°C for 10 hours to remove thermal strain. The crystal samples were poled by applying DC 5 V/cm at 1200°C [21]. The dimension of the crystal was 5.2×4×6.8(c-axis) mm^3.

For SHG experiments, the laser source used in our experiments is a commercial 10 Hz Q-switched Nd:YAG laser (Lumonics, Model HY-750) which has a multitransverse spatial mode intensity profile whose diameter is approximately 8mm. The temporal profile of the laser was observed to be near Gaussian with a pulse duration of 24 ns (FWHM). For SHG experiments, we used a nearly Airy beam. In order to obtain an Airy profile for the input beam, we passed the output beam of the same laser through the 1-mm-diameter circular aperture, followed by free-space propagation of 1.5 m. The beam divergence of our Airy beam was ~ 1.8 mrad. The incident fundamental beam propagates along the direction normal to the optic axis and it is polarized as an ordinary ray to satisfy the type I phase matching condition for SHG as shown in Fig. 1.

The intensity of SHG was measured as a function of temperature and the experimental results are shown as open circles in Fig. 2. The intensity of the fundamental beam was set to be low so that intensity depletion of the fundamental beam would be negligible. The solid curve is the best fit to the corresponding data by using Eq.(7). The consistent agreement between the solid curve and experimental data implies that the optical uniformity of the sample crystal is good. T_{pm} was found to be 39.0°C. $\partial\Delta n/\partial T$ can be evaluated to

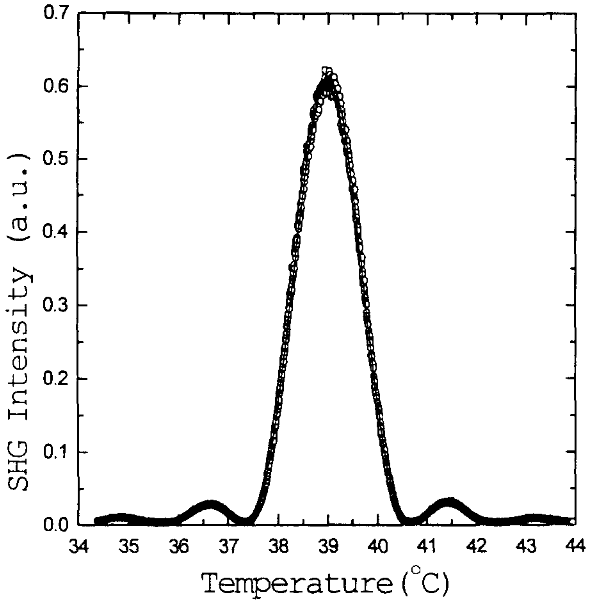


FIG. 2. Temperature phase matching curve of noncritical SHG in 1%Mg:LiNbO$_3$; experimental data(open circle) and the best fit curve(solid line).

be $(5.9\pm 0.1)\times 10^{-5}$ $^o\mathrm{C}^{-1}$ from this temperature phase matching curve by using Eqs.(7) and (8).

We observed the noncollinear phase matching ring at room temperature when the propagation direction of the fundamental beam was normal to the c-axis. The result is shown in Fig. 3(a). The observed ring is an ellipse whose diameter along the c-axis is shorter than that along the y-axis. However, the symmetry of the ring is broken when the propagation direction of the fundamental beam is not normal to the c-axis as shown in Fig. 3(b). It was confirmed that the optic axis in a negative uniaxial crystal can be determined easily from the shape of the ring. Obscure spots and rings in Figs. 3 and 4 were due to multiple reflections from the two surfaces of the crystal. The half cone angle of noncollinear phase matching was measured to be $3.76^o\pm.05$ ^{o} at room temperature(25^{o}C). In this experiment, such an accurate measurement of the half cone angle was obtained because it was possible to measure the radius of rings within an error of one part per thousand. Here, each radius was determined by measuring the distance between the center of the ring and the center of the ring-band. Combining our experimental results with Eqs.(6) and (8), the refractive indices of the fundamental ordinary wave and the second harmonic extraordinary wave are evaluated to be $n_o(\omega) = 2.277\pm.002$ and $n_e(2\omega) = 2.276\pm.002$ at room temperature.

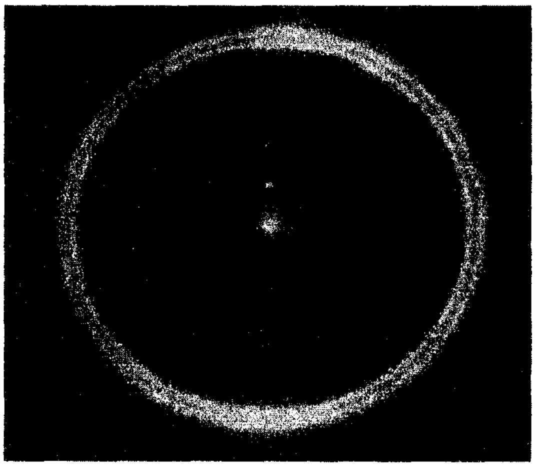
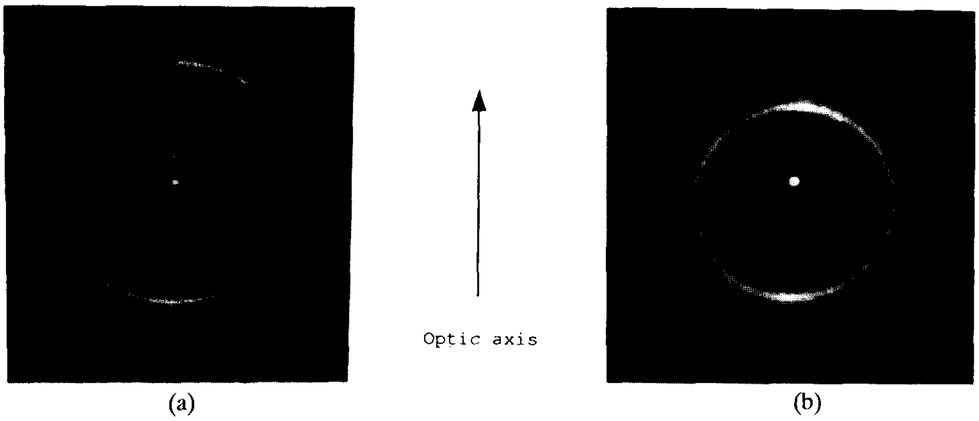


FIG. 4. Experimental photograph of the change of half cone angle with the DC electric field of 5.36 kV/cm(inner ring; parallel to the optic axis) and -5.36kV/cm(outer ring; anti-parallel to the optic axis).

When DC electric fields of 5.36kV/cm and -5.36kV/cm were applied parallel and anti-parallel to the c-axis at room temperature, the half cone angles were changed into 3.63^o and 3.90^o as shown in Fig. 4. The double rings in this picture were taken by



(a) (b)

FIG. 3. Experimental photographs of noncolinear phase matching ring when the propagation direction of the fundamental beam is (a) normal to the optic axis and (b) not normal to the optic axis(~ 80^o off from the c-axis).

using a technique of double exposure. The duration of each exposure was 30 seconds. Combining these results with Eq.(9) and (10), the electro-optic coefficient, r_{eff}, can be evaluated to be -(1.21±.02)$\times 10^{-8}$ cm/V. This value is consistent with the corresponding value ((-1.11±.07)$\times 10^{-8}$ cm/V) that was measured previously with the phase matching temperature shift method [19].

IV. CONCLUSIONS

Temperature dependence and the noncollinear-phase-matching cone of second harmonic generation were investigated in a $LiNbO_3$ crystal doped with 1 mol% MgO. We succeeded in measuring all the important birefringence parameters such as $n_o(\omega) = 2.277$, $n_e(2\omega) = 2.276$, $\partial \Delta n/\partial T = 5.9\times 10^{-5}$ $^oC^{-1}$, and the electro-optic coefficient (1.21±0.02)$\times 10^{-8}$ cm/V. Compared to any other methods, our method has an advantage of easy operation for measuring birefringence parameters, though it is restricted to negative uniaxial crystals.

ACKNOWLEDGMENTS

The present studies were supported in part by the Basic Research Institute Program, Ministry of Education, 1997, Project No. BSRI-97-2415 and also by Research Programs of KIST.

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