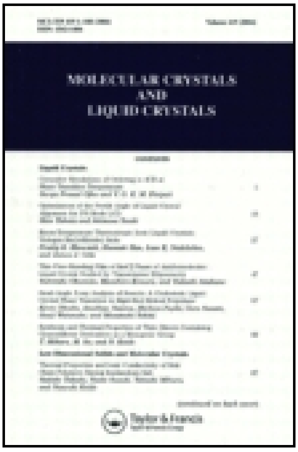

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Blue and Green Second Harmonic Generation in Poled Polymer on the Inorganic Crystal Substrate

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BLUE AND GREEN SECOND HARMONIC GENERATION IN POLED POLYMER ON THE INORGANIC CRYSTAL SUBSTRATE

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Abstracts Optical second harmonic generation in the form of Cerenkov radiation is investigated using KH_2PO_4 (KDP) substrate and poled polymer thin film waveguide. The copolymer of MMA and DR1-substituted methacrylate molecules were oriented by corona poling precess after spin coating on the KDP substrate. The conversion efficiency strongly depends on the waveguide geometry. The high conversion effficency is obtained by theoritical analysis when the nonlinear optical coefficient direction of a substrate is opposite to that of a waveguiding layer. In experiment, we haveconfirmed that the SH power by Cerenkov type phase matching using KDP crystal as a substrate is larger than that of using pyrex as a substrate.

INTRODUCTION

Recently the field of nonlinear optics is growing rapidly and nonlinear optical phenomena have been concentrated on applications in optical switch[1], electrooptics[2] and frequency conversion[3-6]. In these effects frequency conversion, especially second harmonic generation, has been intensively studied for converting near infrared wavelength into the visible wavelength. SHG devices have been reported in the form of guided mode-to-mode phase matching, Cerenkov type phase matching, and quasi-phase matching. In Cerenkov radiation mode, the phase matching condition between the fundamental guided mode and the second harmonic radiation mode can be automatically satisfied by adjusting waveguide parameters and it is possible to achieve high second harmonic generation although the second harmonic wave is absorbed in the waveguiding layer. To achieve high efficiency SHG, the overlapping of the cross sectional field distributions between the fundamental wave and the second harmonic wave is one of the important parameters.

In this paper, poled copolymer [poly (MMA-co-DR1MA)] was used as a waveguiding layer on nonlinear substrate and linear substrate. The optimum poled polymer thickness was investigated and the Cerenkov SHG power was compared.

SAMPLE FABRICATION

Copolymer of methylmethacrylate (MMA) and Disperse Red1 (DR1)-substituted methacrylate (DR1 10wt.%) was spin coated on the nonlinear substrate (KDP crystal) and the linear substrate (pyrex). The KDP crystal was cut in the size, $20 \times 24 \times 1.5mm^3$, and its face planes were polished for spin coating of the polymer. The sample was prebaked at 80℃ to remove the residual solvent and moisture and was poled by corona discharge at DC 5kV, 110℃ for 15 minutes. In the process of corona poling, raising temperature and cooling (0.4℃/min) were carried out very carefully to avoid the damage of the KDP crystal substrate. The absorption specturm of the poled polymer film before poling is shown in Fig. 1 as a solid line together with the absorption curve (dashed line) after poling. The absorption peak of poled polymer film exhibited red shift accompanying decrease of the optical density perpendicular to the film after poling as shown in Fig. 1. The nonlinear optical coefficient of the poled polymer film was estimated as d_{33}=13.6 pm/V by Maker fringe method using Q-switched Nd-YAG laser (1064nm). To estimate d_{33} of the poled olymer the second harmonic power from d_{11} of the quartz crystal was used as a reference. The refractive indecies of the poled polymer measured by m - line method and ellipsometry are 1.524, 1.527, 1.582 and 1.494 at

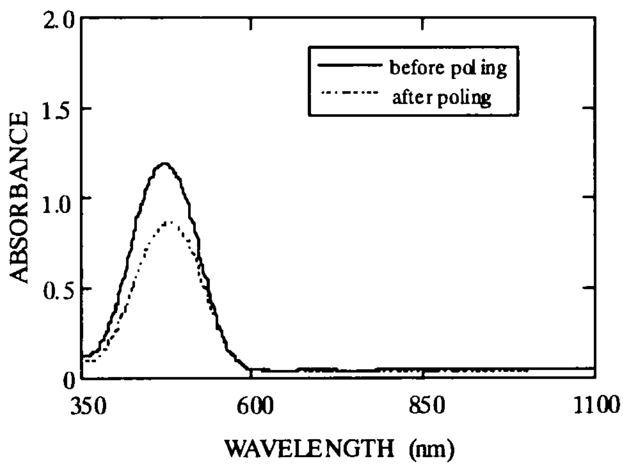


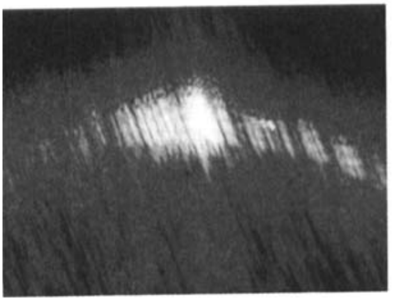
FIGURE 1. The absorption spectrum of poly(MMA-co-DR1MA).

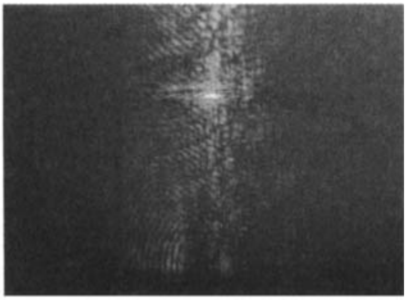
wavelength 1064nm, 880nm, 532nm and 440nm, respectively.

EXPERIMENT AND RESULTS

First, we perfomed Cerenkov type SHG experiment using the sample with nonlinear waveguiding layer-nonlinear substrate. Since two nonlinear coefficients, d_{33} of the poled polymer and d_{14} of KDP crystal substrate, were used to generate Cerenkov type second harmonic generation it is necessary to use TM fundamental wave. Fig. 2(a) is the photograph of the green Cerenkov type second harmonic output generated from Q-switched Nd-YAG laser (1064nm) by prism coupling. Cerenkov type SHG is advantageous to this case the second harmonic wave is absorbed in the waveguiding layer. The polymer film thickness on the KDP crystal was estimated as 1.25μm from the incidence angle of the fundamental beam. We fabricated the another film thickness 1.6μm and measured the second harmonic power using Q-switched Nd-YAG laser (1064nm) as a fundamental beam. Fig. 3 shows the second harmonic power at film thickness 1.25μm, 1.6μm. The circle is for the second harmonic power at 1.25μm and the triangle is for the second harmonic power at 1.6μm. The film thickness was controlled with ease by spin speed. The second harmonic power at 1.6μm film thickness was 50 times as low as that of 1.25μm. The experimental results are good agreement with the theoritical results as shown in Fig. 4. Varing the film thickness, the second harmonic power is lower than that of the optimum thickness because the overlap integral becomes small. This results from the difference of propagation constant on the film thickness. Fig. 4 is the results calculated by eq. (1).

$$P^{2\omega} = \frac{\sqrt{2}\pi\omega^2 L\left(n_{eff}^{\omega}\right)^4 \left(P^{\omega}\right)^2 S^2}{c^3 \varepsilon_0 \left(W_{eff}^{\omega}\right)^2 D n_s^{2\omega} \sin\alpha} \tag{1}$$





(a) (b)

FIGURE 2. The photograph of the Cerenkove type second harmonic wave on the screen. See Color Plate I.

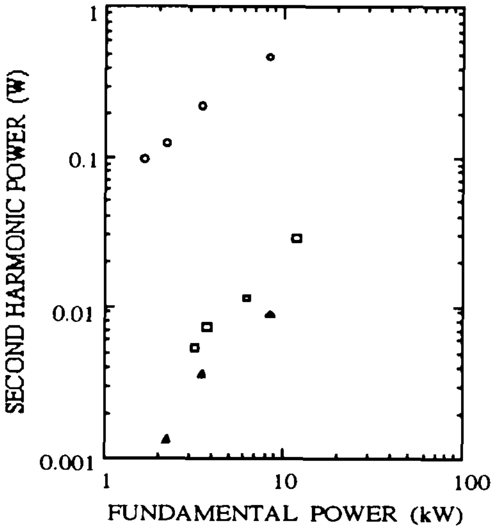


FIGURE 3. The SH power versus fundamental power. The circle is for nonlinear waveguiding layer-nonlinear substrate at film thickness 1.25μm and the triangle is for nonlinear waveguiding layer-nonlinear substrate at film thickness 1.6μm. The square is for nonlinear waveguiding layer-linear substrate at film thickness 0.8μm.

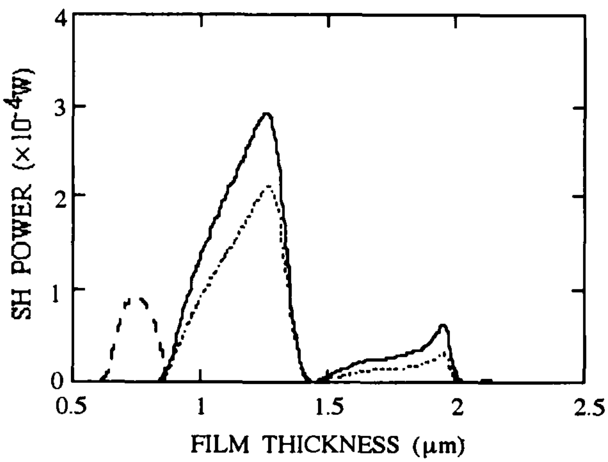


FIGURE 4. The SH power versus filmthickness in nonlinear waveguiding layer-nonlinear substrate (solid and dashed line) and nonlinear waveguiding layer-linear substrate (long dashed line) structures.

n_{eff}^{ω} in eq. (1) is effective refractive index of the fundamental guided mode $n_s^{2\omega}$ are refractive indecies of the substrate at the second harmonic wavelength. P^{ω} is fundamental power. D is fundamental beam width and W_{eff}^{ω} is effective guide thickness for the fundamental guided mode. α is Cerenkov angle defined by

$$n_{eff}^{\omega} = n_s^{2\omega} \cos\alpha \tag{2}$$

S which plays an important role in second harmonic power is overlap integral between the fundamental and second harmonic magnetic field distributions as follows;

$$S = \int_{-\infty}^{-t} d_s \left[H_y^{\omega}\right]^2 H_y^{2\omega} dx + \int_{-t}^{0} d_g \left[H_y^{\omega}\right]^2 H_y^{2\omega} dx \tag{3}$$

where H_y^{ω} is mode function of TM guided mode at the fundamental wave and $H_y^{2\omega}$ is that of TM radiation mode at the second harmonic wave. d_i is the nonlinear optical coefficient (g:waveguiding layer and s:substrate). L is interaction length. We investigated the effect of nonlinear optical coefficients in waveguiding layer and substrate. The second harmonic power of the sample with the opposite signs in nonlinear waveguiding layer and nonlinear substrate (solid line in Fig. 4) is larger than that of the sample with the same signs in nonlinear waveguiding layer and nonlinear substrate (dashed line in Fig. 4). This means that the overlap integral is reduced by interference effect[7] in waveguiding layer-substrate interface when the waveguiding layer and the substrate have the same sign nonlinear coefficients. Fig. 2(b) is the photograph of the Cerenkov type second harmonic output generated from mode locked Ti-Sapphire laser (880nm).

Second, we carried out the Cerenkov type SHG experiment using the sample with nonlinear waveguiding layer-linear substrate and measured the second harmonic power using Q-switched Nd-YAG laser (1064nm) as shown in Fig. 3 (the square). Comparing the second harmonic power, we have known that the second harmonic of the sample with nonlinear waveguiding layer-linear substrate is smaller than that of the sample with nonlinear waveguiding layer-nonlinear substrate because the overlap integral of the sample with nonlinear waveguiding layer-linear substrate is affected only in the waveguiding layer region from eq. (3). This experiment is good agreement with the analysis as shown in Fig. 4.

DISCUSSION

From Fig. 1 the absorption is very small near 370nm. It is possible to generate guided second harmonic wave from fundamental guided mode using 740nm as a fundamental beam. The mode dispersion of nonlinear waveguiding layer and linear substrate is shown in Fig. 5. The refractive index of the substrate at the fundamental wavelength is higher than that of the substrate at the second harmonic wavelength because of anomalous dispersion. The phase matching between fundamental first mode and second harmonic first mode is possible. The phase matching filmthickness is 1.124mm and the second harmonic power is larger than that of Cerenkov phase matching type.

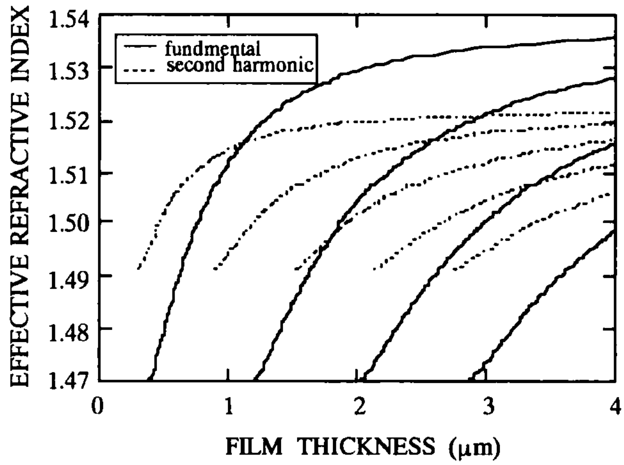


FIGURE 5. The mode dispersion curve of nonlinear waveguiding layer-linear substrate structure. The fundmental wavelength is 740nm and second harmonic wavelength is 370nm.

CONCLUSIONS

The Cerenkov type second harmonic generation of a Q-switched Nd-YAG laser (1064nm) and mode locked Ti-Sapphire laser (880nm) was observed in the waveguide structure which consists of poled poly(MMA-co-DRIMA) and KDP crystal even if the poled polymer flim has absorption at the second harmonic wavelength. We confirmed that the second harmonic power of nonlinear waveguiding layer-nonlinear substrate structure was much larger than that of nonlinear waveguiding layer-linear substrate structure

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