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A NOVEL PHASE MATCHING TECHNIQUE FOR A POLED POLYMER WAVEGUIDE.

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Abstract An electric field induced dynamic phase-matching which can improve the thickness tolerance of waveguide SHG device is first demonstrated using organic nonlinear optical (NLO) materials with low glass transition temperature (Tg). The advantage of this technique is the possibility to control an effective refractive index of NLO waveguide at room temperature. As a result of refractive index variation induced by an external electric field, the effective phase-matching dimension can be controlled from 1.272 μm to 1.260 μm. The phase-matching condition i.e. conversion efficiency of second harmonic light can be easily tuned by changing a poling electric field. The obtained net conversion efficiency of low Tg NLO slab waveguide was found to be 1.5 %. No decay of conversion efficiency for low Tg NLO waveguide is observed in the presence of an electric field.

INTRODUCTION

Second-order nonlinear optical (NLO) materials have recently attracted a great deal of attention for waveguide electro-optic (EO) modulation and frequency doubling applications.[1-4] The interest in NLO materials is a result of the ease in designing and processing the materials.[5] A noncentrosymmetric structure can be introduced in the medium by electric field poling and the polymers can be fabricated into good optical quality devices.[6-7] However, the electrical poling induced second-order NLO properties are not temporary stable at ambient temperature.[8] The general approach for achieving stable second-order NLO properties is to incorporate the NLO chromophores in a polymer which has a high glass transition temperature (Tg).[9] The relaxation of the poled molecules is considerably restricted at temperatures much lower than Tg.

Several approaches have been developed to provide temporal stability in NLO polymers using thermal crosslinking[10] photocrosslinking and spontaneously crystallization process to form a noncentrosymmetric structure.[11] We recently reported a novel approach to obtain a stable second harmonic generation (SHG) using a low Tg material.[12] This approach is based on the fact the molecular orientation of low Tg materials can be controlled by changing the applied electric field at room temperature. Therefore stable SHG properties can be expected in the presence of an applied electric field. Furthermore, the phase-matching properties of low Tg materials can be controlled easily due to the large refractive index variation induced by electrical poling.

Organic SHG devices in optical waveguides have not been developed extensively because of the difficulty in achieving phase-matching. It was proposed for the phase-matching to utilize the difference between the mode dispersion of the fundamental and the second harmonic waves in thin film waveguides.[13] The phase-matching in the guided mode is satisfied by controlling or adjusting the waveguide thickness.[14] However, it is not easy to prepare the appropriate thickness waveguides by which the phase-matching condition is satisfied. To avoid this difficulty, the use of artificial periodic structure[15] Cerenkov radiation[16] and noncollinear light path geometry have already been demonstrated.[17]

In the present paper, we report an electric field assisted dynamic phase-matching in a guided mode, using a low Tg NLO material, which can control the effective phase-matching thickness by an applied electric field.[18]

EXPERIMENTAL

Preparation of Optical Waveguide

A low Tg NLO materials used in this experiment are shown in Figure 1. 4-nitroaniline (PNA) is coupled to an ethylene glycol chain (PEG) to form a ω,ω'-di(p-nitroaniline)-ethylene glycol. The detail of the synthetic scheme of the soft-gel material abbreviated as PEG-PNA was described in reference.[12] The sample was purified using an electric-dialysis to eliminate the effect of ion species during the poling procedure. The sample was added between two indium tin oxide (ITO) glass plates which were covered by a fused silica buffer layer as shown in Figure 2. The thickness of the buffer layer is 0.95 μm. The taper gradient of PEG-PNA is 0.05 μm/mm. The refractive indices were accurately measured using the TaFD21 prism coupler according to m-line method.[19] TM-polarized Q-switched Nd:YAG laser (1064 nm), its harmonic beam and He-Ne laser was

used for the light source and out couple angle was measured as a function of poling voltage.

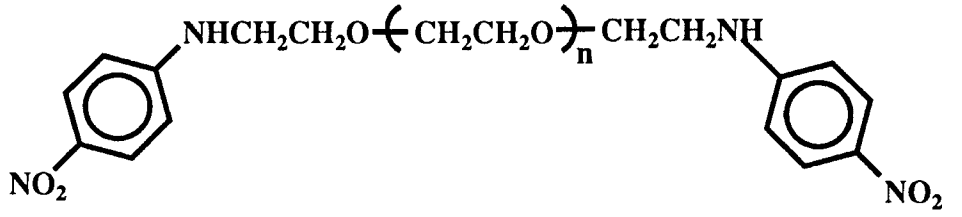

Figure 1. Chemical structure of PEG-PNA. (n=20)

Determination of Nonlinear Optical Coefficient

The second-order nonlinear optical coefficient d33 of the PEG-PNA was measured by Maker fringe technique. The Second harmonic (SH) signal was obtained by illuminating a Q-switched Nd:YAG laser and compared with that of y-cut quartz. The SH power $P^{2\omega}$ in a uniaxial poled materials generated by p-polarized fundamental power P^{ω} is given by

$$P^{2\omega}(\theta) = (512\pi^3 / A) d_{33}^2 p^2(\theta) F(\theta) \tag{1}$$

$$F(\theta) = t_i^4 t_\omega^4 T_{2\omega} t_0^2 l_\omega^2 \left[\sin \psi(\theta) / \left(n_\omega^2 - n_{2\omega}^2\right)\right]^2 \tag{2}$$

where A is the area of the laser beam spot. t_0 and t_i are the Fresnel factors for substrates. t_ω and $T_{2\omega}$ are fundamental and second harmonic Fresnel factors respectively and $\psi(\theta)$ is the angular dependence of the SH power.[20]

$$\psi(\theta) = \left(\frac{2\pi l}{\lambda}\right)\left(n_\omega \cos\theta_\omega - n_{2\omega} \cos\theta_{2\omega}\right) \tag{3}$$

The n_ω and $n_{2\omega}$ the refractive indices of the polymer waveguide at the fundamental and SH wavelength. θ_ω, $\theta_{2\omega}$ are the internal angles of the fundamental and SH wavelength. The projection factor $p(\theta)$ is given by

$$p(\theta) = \left(\sin^2\theta_\omega + \cos^2\theta_\omega / 3\right)\sin\theta_{2\omega} + 2\sin\theta_\omega \cos\theta_\omega \cos\theta_{2\omega} / 3 \tag{4}$$

Using eq. (1-4), the second-order nonlinear optical coefficient d33 of PEG-PNA films was determined to be 1-3 pm/V.

Phase-matched SHG Experiments

The Nd:YAG laser was TM polarized in order to use d33 of PEG-PNA efficiently. The fundamental beam was focused by 40 times magnificated microscope objective lens and coupled into the waveguide from the edge of the substrate as shown in Figure 2.

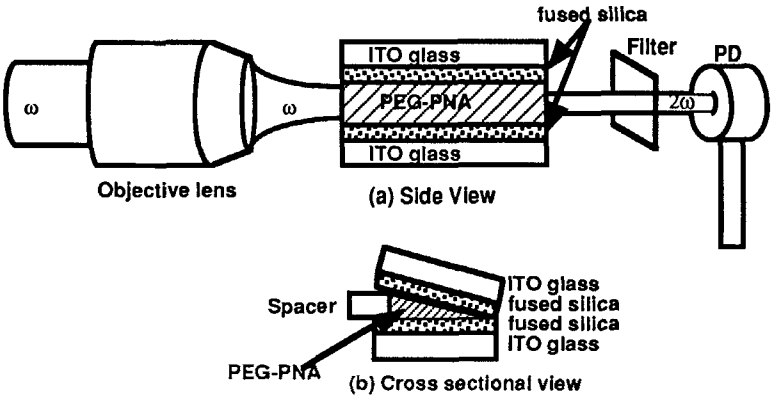

Figure 2. Experimental setup for measurement of phase-matched SHG.

The generated phase-matched SH beam was detected by a pin-photodiode behind the YAG-cut filter and interference filter. The phase-matching point is adjusted by both transitional shift of the waveguide in the taper direction and change of an applied electric field.

RESULTS AND DISCUSSIONS

Table 1 shows the experimentally obtained refractive indices of PEG-PNA and calculated phase-matching thickness as a function of poling voltage. The film thickness of unpoled sample corresponds to the hypothesis phase-matching thickness because of its SHG inactivity. The phase-matched conversion between the fundamental 0 order mode and SH 2nd order mode was selected. The refractive indices for TM mode increas with an increase of the poling electric field, whereas the phase-matching thickness decrease. The largest linear polarizability α_{xx} of PEG-PNA aligns along the poling electric field, which induce the large refractive index change by poling.

Table 1. Measured refractive indices and calculated phase-matching thickness of PEG-PNA waveguide.

Poling Voltage (V/μm)	n(ω)	n(2ω)	Phase-matching thickness (μm)
0	1.500	1.558	1.2719
15	1.501	1.559	1.2682
30	1.502	1.560	1.2645
45	1.503	1.561	1.2609

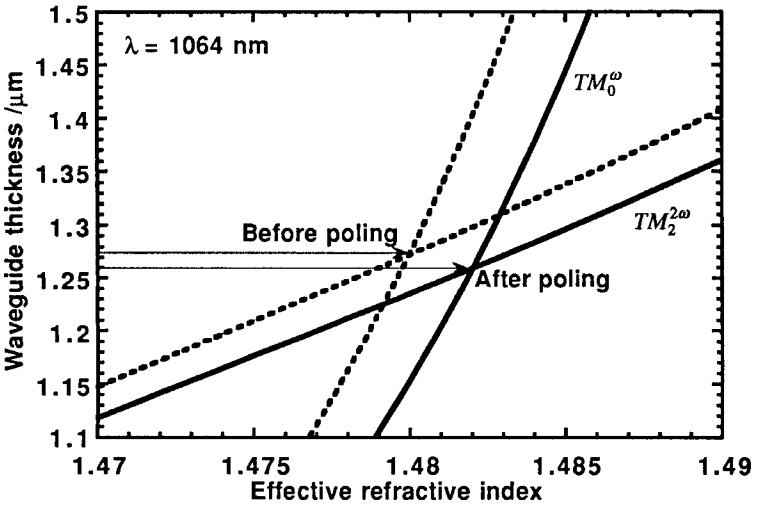

Figure 3. Mode dispersion curves of PEG-PNA waveguide.
(Solid line; After poling, Dotted line; Before poling)

The dispersion of refractive indices of poled material is larger than that of unpoled material, which is due to the red-shift of absorption maxima induced by poling. The red-shift of absorption maximum results from the strong dipole-dipole interaction of NLO chromophore. In order to relax the phase-matching condition, the increase of dispersion of refractive indices after poling plays a very important role. Wide range of effective phase-matching condition cannot be expected without red-shift of cutoff wavelength induced by poling. It is clear that the effective phase-matching thickness can be controlled

from 1.272 μm - 1.260 μm using electric field as shown in Figure 3. This effective phase-matching thickness variation induced by poling is a very helpful for achieving a phase-matching of taperless waveguide, because the full width of half maximum (FWHM) of conventional 3 layer slab waveguide is very small. The tuning range of PEG-PNA waveguide dimension for phase-matching is at least one order of magnitude larger compared with FWHM of 3 layer waveguides.[21] Phase-matching between TM_0^{ω} and $TM_1^{2\omega}$ is also available but this configuration cannot increase the conversion efficiency due to the low overlap integral.[13] The propagation length of the waveguide is 2 mm.

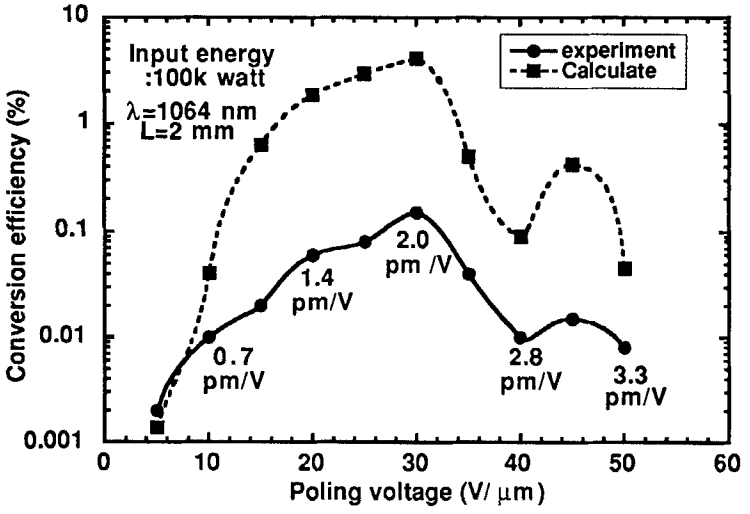

Figure 4. Conversion efficiency of PEG-PNA waveguide as a function of poling voltage.

Figure 4. shows conversion efficiency of the PEG-PNA waveguide as a function of the poling electric field at room temperature. The maximum conversion efficiency can be obtained with a 1.265 μm waveguide thickness, when a 30 V/μm electric field is applied. The nonlinear optical coefficient of the 30 V/μm poled sample is 2.0 pm/V, whereas the conversion efficiency of 3.3 pm/V sample poled at 50 V/μm decrease than that of the 2.0 pm/V sample due to the phase-mismatch. These experimental results suggest that the electric field assisted dynamic phase-matching can control the phase-matching condition by an applied electrical field. In order to obtain an efficient conversion efficiency, the channel type waveguide is required because of the high optical power density. Electrical field assisted dynamic phase-matching enables us to fabricate the channel waveguide

easily with the ionic exchange technique because the effective phase-matching thickness can be controlled. The experimental conversion efficiency of PEG-PNA waveguide is smaller than that of the theoretical calculated value because of the following reasons.

1) The incident laser beam not only excites the TM_0^{ω} mode but also the TM_1^{ω} mode, which lowers the power density of the fundamental beam.
2) The roughness of the fused silica buffer layer protect to satisfy the phase-matching condition.
3) The relatively small phase-matching area compared with laser beam diameter due to the tapered structure reduce the conversion efficiency.
4) Several experimental parameters such as beam diameter in the waveguide were uncertain.

The experimental SH tune curve in Figure 4 seems to be wider than that of the theoretical calculation, which is mainly due to the tapered structure of waveguide. To increase the tuning range of phase-matching thickness, the large dispersion of refractive indices induced by poling is necessary. This large dispersion of refractive indices can be expected by synthesizing the medium with high concentration of PNA. SHG experiments for low Tg NLO materials with high concentration of the NLO dye are underway and will be published elsewhere. Another key parameter is the refractive indices of the buffer layer. Choosing the appropriate substrate which has a slightly smaller refractive index than that of NLO material, the intersection point for phase-matching relatively shifts to the right side in Figure 3. The temporal SHG stability of PEG-PNA waveguide was examined. No decay of conversion efficiency is observed in the presence of electric field.

CONCLUSION

Electric field assisted phase-matching was first demonstrated at room temperature using low Tg NLO materials. This phase-matching technique completely eliminates the relaxation problem of the NLO chromophore and enables the control of the phase-matching condition. This procedure will be a promising approach for the practical application in SHG devices.

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