Frequency Doubling in Poled Polymers using Anomalous Dispersion Phase-matching

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ABSTRACT

We report on a second harmonic generation in a poled polymer waveguide using anomalous dispersion phase-matching. Blue light ($\lambda = 407$ nm) was produced by phase-matching the lowest order fundamental and harmonic modes over a distance of 32 μm. The experimental conversion efficiency was $\eta = 1.2\times10^{-4}$, in agreement with theory. Additionally, we discuss a method of enhancing the conversion efficiency for second harmonic generation using anomalous dispersion phase-matching to optimize Čerenkov second harmonic generation. Our modeling shows that a combination of phase-matching techniques creates larger conversion efficiencies and reduces critical fabrication requirements of the individual phase-matching techniques.

1. INTRODUCTION

Frequency doubling of near-IR semiconductor lasers has attracted much attention as a potential route to short wavelength laser sources. Recent advances in organic material design and inorganic material processing have provided candidate materials that are promising for coherent short wavelength device applications. Among the potential applications are high density optical data storage, laser printing, spectroscopy, and display technology. Polymeric materials are interesting for these applications because they have large nonlinearities, low losses, and are easily cast into waveguide structures (including multilayers). In addition they are inexpensive, easily prepared, and compatible with a variety of substrate materials. Recently, polymeric LB (Langmuir-Blodgett) films have been optimized for second harmonic generation and have produced conversion efficiencies approaching those of inorganic materials. [1] This level of performance is stimulating new research efforts.

Efficient frequency doubling of lower power laser light requires large nonlinearities and fundamental intensities, as well as phase-matching over an appreciable propagation lengths. The degree to which these parameters can be optimized is dependent on the material and may also depend on other parameters being optimized. The equation describing conversion efficiency for second harmonic generation is

$$\eta_{shg} = d_{NL}^2 L^2 I_\omega S_{211}^2 sinc^2(\Delta kL/2) \tag{1}$$

where d_{NL} is the nonlinear optical coefficient, L is the propagation length, I_ω is the fundamental intensity, S_{211} is the overlap between fundamental and harmonic fields, and Δk is the wave-vector phase-matching.

Low power lasers can achieve intensities necessary for nonlinear optical interactions by spatially confining light to a waveguide as in integrated optics. Since light in a waveguide is confined to a small cross-sectional area and propagates over long lengths without diffraction, more efficient frequency conversion can

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take place. The advantage due to increased energy density in waveguided devices over bulk devices can be as high as the ratio of the length of the waveguide to the wavelength of light, which can, in principle, be 10^4. [2] The quantized properties of waveguide modes allows for additional phase-matching schemes, but may add practical difficulties due to critical design parameters.

An important requirement for efficient second harmonic generation is phase-matching, which refers to the degree of coherence between the nonlinear polarization and the generated harmonic radiation. Phase-matching in waveguides has been achieved using modal dispersion phase-matching (MDPM), quasi-phase-matching (QPM), colliding pulse or counter propagating phase-matching (CPPM), and Čerenkov phase-matching(CPM). [3–6] Unfortunately, these phase-matching techniques contain a parameter or a processing variable that must be critically controlled to permit phase-matching over long distances. MDPM matches modes of different order by adjusting waveguide thickness and hence the effective index so that modes of different order travel at the same velocity. Waveguide thickness variations of 0.1% limit the phase-matched interaction length to a few millimeters. In addition, conversion efficiencies in MDPM are reduced because of small overlap between the fundamental and harmonic fields. QPM requires periodic spatial modulation of the nonlinearity and has a narrow acceptance band width. Deviations from periodicity can severely limit conversion efficiency. In CPPM, mixing of counter propagating guided waves produces harmonic light normal to the direction of fundamental propagation and is therefore difficult to collect. In CPM the lowest order mode cutoff occurs at a wavelength between the fundamental and the second harmonic. The phase-matching condition is noncritically obtained for certain wave-vectors in the radiated mode. As a result, the harmonic light propagates in the cladding where optical losses are minimal. A number of studies have been carried out, both theoretical [7]- [15] and experimental to optimize conditions for Čerenkov second harmonic generation. Experiments have been carried out for inorganic crystals [16,17], crystal cored fibers [18,19], polymers [20]- [22], and Langmuir-Blodgett films [23,24].

2. ANOMALOUS DISPERSION PHASE-MATCHED SECOND HARMONIC GENERATION

To achieve ADPM (anomalous dispersion phase-matched) second harmonic generation, a nonlinear optical chromophore that absorbs between the fundamental and harmonic wavelengths is placed in a host polymer that exhibits normal dispersion. At a particular concentration, the normal dispersion of the host material is canceled by the anomalous dispersion of the chromophore, yielding equal refractive indices at the fundamental and harmonic wavelengths. In addition, the guest-host system must be processed to eliminate inversion symmetry (poled) and transparent at both the fundamental and harmonic wavelengths. In this technique, increasing the nonlinear optical moiety concentration increases the refractive index at the fundamental wavelength (normal dispersion) while it decreases the index at the harmonic wavelength (anomalous dispersion). ADPM has many advantages over conventional phase-matching techniques. First, ADPM chromophores can have large nonlinear optical susceptibilities because their first excited electronic state is in the visible region of the spectrum. The correlation between optical nonlinearity and the inverse excited state energy is well documented and translates into an order of magnitude increase in the nonlinear optical susceptibility for ADPM dyes. [25,26] Second, the nonlinear optical susceptibility is naturally resonantly enhanced because both the fundamental and harmonic wavelengths are located near the absorption maximum. Third, ADPM allows conversion between lowest order fundamental and harmonic modes, resulting in a large overlap integral. ADPM second harmonic generation has been demonstrated in solution measurements (EFISH) electric field induced second harmonic generation. [27] Residual absorption in the uv transparency window was the main limitation for EFISH using ADPM.

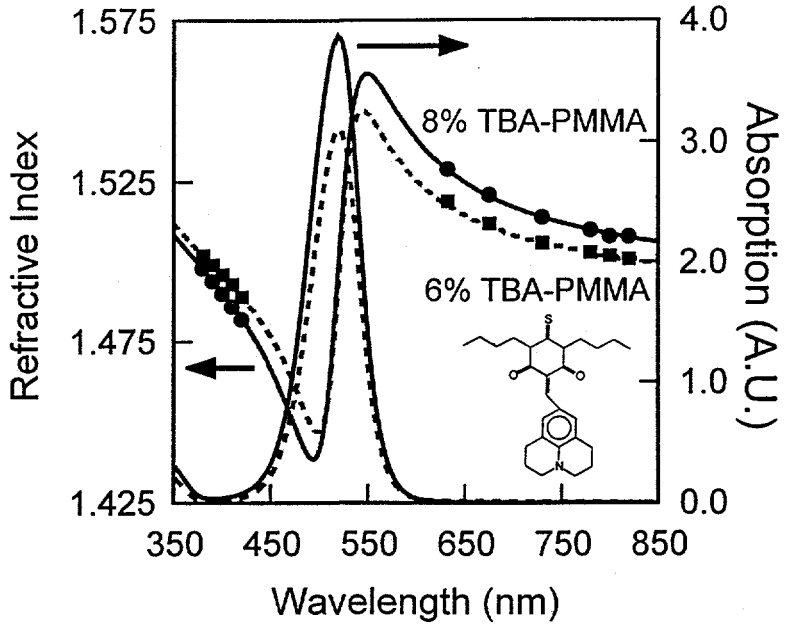

FIG. 1. Data points represent refractive index measurements of 6% and 8% TBA-PMMA doped films. Solid lines show fits to a two term Sellmeier equation using the absorption maximums and oscillator strengths determined from absorption measurements. Also shown are the absorption spectra of 6% and 8% TBA-PMMA films.

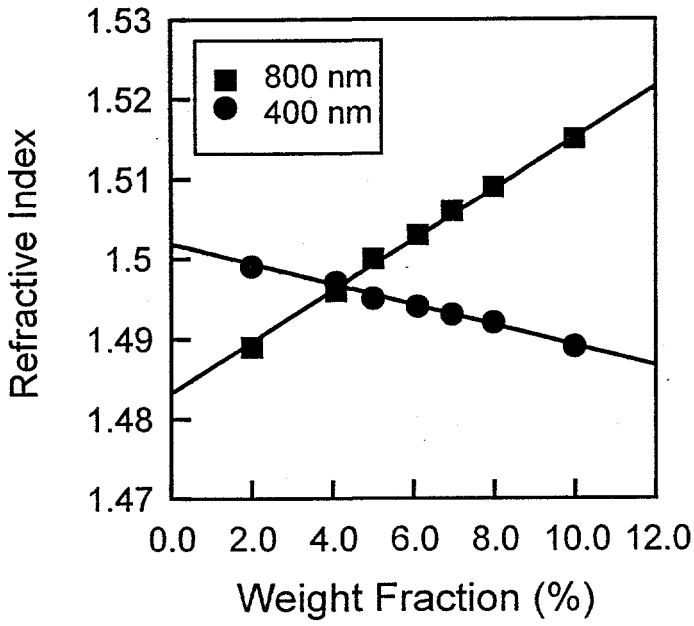

FIG. 2. Bulk refractive indices measured at 800 nm and 400nm as a function of chromophore weight fraction for thiobarbituric acid doped PMMA.

The nonlinear optical guest-host system was made by mixing PMMA with the nonlinear optical moiety TBA, shown in Figure (1). The synthesis and characterization of this chromophore have been described elsewhere. [28] This dye had an ADPM figure of merit of 185 (ratio of molar absorptivity at the absorption peak to the molar absorptivity at the uv transparency window). Although the ADPM figure of merit for TBA is large compared to other conventional noncentrosymmetric nonlinear optical moieties, the residual absorption at the ADPM concentration limited the coherence length ($l_c < 1.0$ mm). Waveguides were prepared using a film floating technique. [29] To produce noncentrosymmetric ordering necessary for second order nonlinearities, free-floating films were corona poled on ITO glass. Typical poling fields were 1.0 MV/cm as measured by an electrostatic voltmeter. The nonlinear optical susceptibility component perpendicular to the film, d_{33},was measured at a wavelength of $\lambda = 1.367$ μm using a rotational Maker fringe (RMF) experiment. [30] The susceptibility at $\lambda = 815$ nm was calculated using the two-level model to correct the $\lambda = 1.367$ μm data for dispersion. [31]

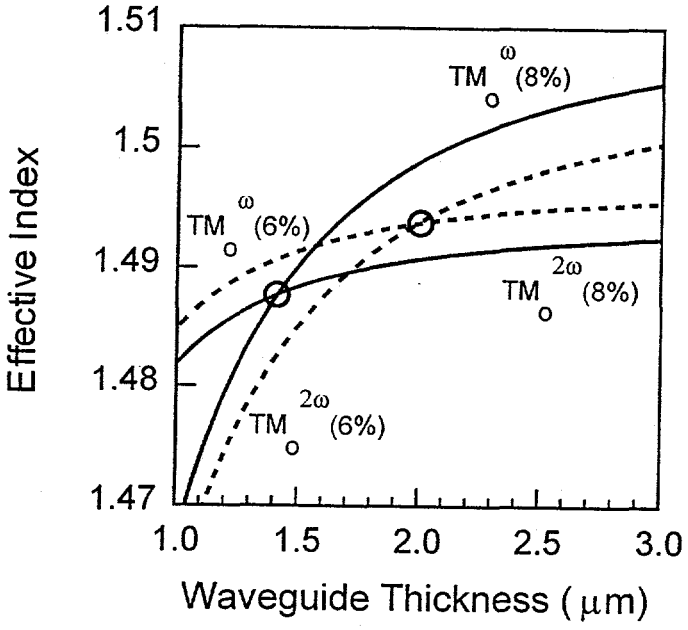

FIG. 3. The effective index for $TM_o^{2\omega}$ and $TM_o^{2\omega}$ modes is plotted for 6%(dotted) and 8%(solid) films using refractive indices measured after electric-field-poling.

Refractive indices were measured with TM polarized light using the prism coupling technique. The refractive index data was fit to a two term Sellmeir equation using the absorption maximum wavelength and oscillator strength obtained from UV-visible absorption measurements. Figure (1) shows the measured refractive indices (data points) and resulting fits for 6% and 8% TBA-PMMA by weight fraction. Also shown in Figure (1) are the respective absorption spectra. The bulk refractive index of a thiobarbituric acid derivative (TBA) doped PMMA (poly-methyl-methacrylate) is shown in Figure (2) as a function of moiety concentration at the fundamental and harmonic wavelengths. Care must be taken when measuring the refractive indices of films prepared using the floating technique. Incomplete removal of NLO film from the water soluble polymer causes critical errors in the refractive index measurements and destroys the delicate phase-matching condition. In the case of waveguided second harmonic generation, it is not the bulk refractive indices that must be phase matched, but rather the effective mode indices. Figure (3) shows the effective index as a function of waveguide thickness for lowest order fundamental and harmonic TM modes using refractive indices measured after poling. The intersection points in Figure (3), marked by

open circles, indicate phase-matching at film thicknesses of 2.0 μm and 1.4 μm for the 6% and 8% films, respectively. As Figure (3) shows, the film thickness can be used to adjust the phase-matching condition.

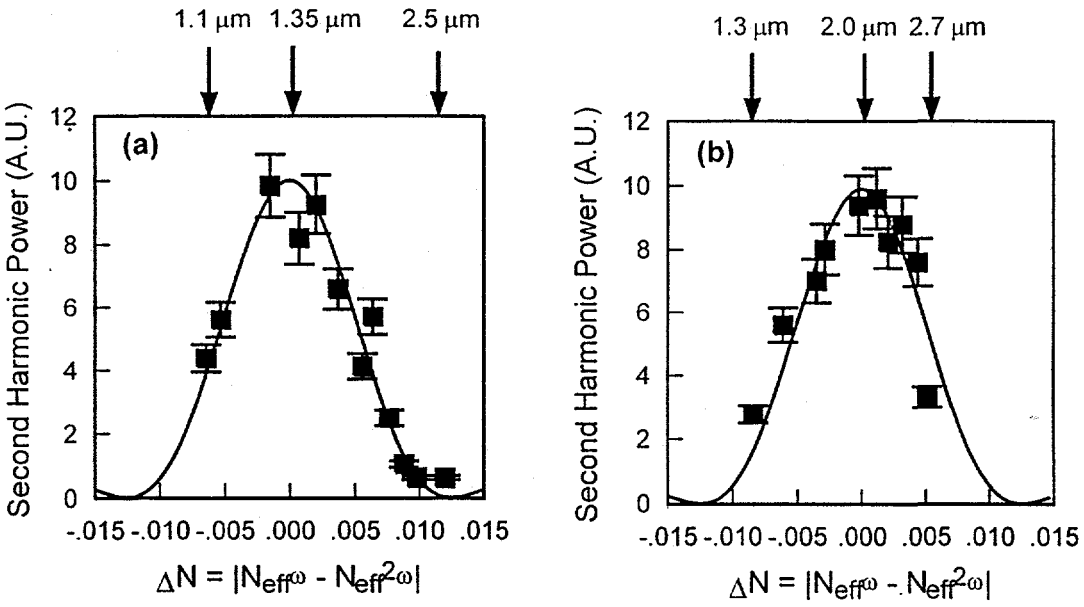

FIG. 4. Experimentally measured second harmonic intensity is plotted for films of various thickness(data points). The maximum amount of second harmonic was obtained for thicknesses of 2.0 μm and 1.4 μm for 6% and 8% TBA-PMMA films, 4(a) and 4(b) respectively. Solid line is a fit to $sinc^2(x)$ using the amplitude and propagation length as adjustable parameters.

Pulsed light at 815 nm from a frequency tripled Nd:YAG pumped optical parametric oscillator entered the experiment through a bandpass filter centered at the fundamental wavelength. [32] The beam was split into the sample and reference arms of the experiment. The reference beam passed through a quartz wedge that was rotated to a Maker Fringe maximum and served to eliminate shot-to-shot fluctuations. The sample beam passed through a focusing lens and was in-coupled and out-coupled through a high refractive index equilateral prism at the synchronous angle. The out-coupled light passed through appropriate filters and the harmonic was detected with a photo-multiplier tube. The measurement was repeated on samples of different thickness. The thicknesses of the films were measured independently with a Dek-Tak profilometer to an accuracy of $\pm 2.0\%$. The relative amount of second harmonic power is plotted vs phase mismatch calculated from independent thickness measurements in Figures 4(a) and 4(b). The data points in Figures 4(a) and 4(b) could only be measured over a limited thickness range. Thicker films permit conversion processes between higher order modes and films thinner than 1.0 μm were difficult to prepare using the floating technique. It was necessary to correct the integrated second harmonic signal for thickness and poling field variations. The solid line in Figures 4(a) and 4(b) represents a fit to the $sinc^2(x)$ function using the amplitude and the propagation length as adjustable parameters. A good fit was obtained with the $sinc^2(x)$ function using a propagation length of 32 μm. Overall Figures 4(a) and 4(b) show an order of magnitude increase in second harmonic power for 8%-TBA films and a factor of five increase for 6%-TBA films at the phase-matching thicknesses calculated in Figure (3). Agreement of the location of the phase-matching thickness with those predicted in Figure (3) is strong evidence of anomalous dispersion phase-matching. Additional experiments confirmed the quadratic dependence on fundamental power. We observed a decay in second the harmonic power that decayed over time with the rate and degree of decay

increasing with generated harmonic power. This suggests that photodegradation of the dye is occurring, probably due to two-photon absorption. We are currently investigating improved materials. Figure (5) shows the decay of second harmonic generation as a function of time for two incident fundamental powers.

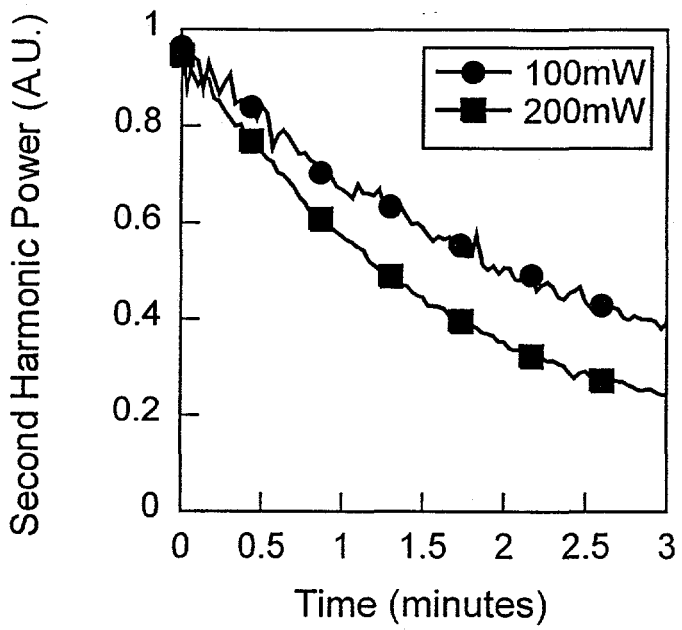

FIG. 5. Decay of second harmonic generation as a function of time and incident power.

A mode-locked Ti:Sapphire (80 MHz repetition rate) was used to measure the conversion efficiency. Mode-locked Gaussian pulses at 815 nm were 15 ps in duration and produced an average power of 400 mW. The conversion efficiency was measured for a 6%-TBA film. The experimental conversion efficiency was defined as the ratio of harmonic power before out-coupling to the fundamental power coupled into the waveguide. Defining P_ω as the fundamental pump power incident on the prism coupler and $P_{2\omega}$ as the harmonic power measured after the prism, the conversion efficiency is

$$\eta_{exp} = \frac{P_{2\omega}}{\kappa^2 P_\omega}, \tag{2}$$

where we have assumed equal in-coupling and out-coupling coefficients κ. A coupling efficiency of $\kappa = 0.27$ was estimated from mode reflectivity scans. An average fundamental power of $P_\omega = 133$ mW was focused onto the prism base with a 7.5 cm focal length lens and produced 1.2 μW of harmonic power. The resulting experimental conversion efficiency was $\eta_{exp} = 1.2 \times 10^{-4}$. The theoretical conversion efficiency was calculated from experimental parameters using coupled mode theory that includes absorption at the harmonic wavelength. The effect of absorption in this case is negligible because the propagation length is much shorter than the $1/e$ absorption length. The calculated conversion efficiency was $\eta_{theory} = 1.7 \times 10^{-4}$, in good agreement with experimental results considering uncertainties in various parameters.

3. ČERENKOV PHASE-MATCHING

In order to overcome the difficulties presented by both absorption and criticality, we propose a second harmonic generation scheme based on Čerenkov phase-matching with anomalous dispersion to achieve zero transverse dispersion. In Čerenkov phase-matching the second harmonic light propagates as a radiation mode, while the fundamental is guided. By guiding the harmonic light in the cladding, the effect of absorption in the core of the waveguide is minimized, as we will demonstrate below. The criticality of fabrication is also eased in this scheme.

Čerenkov second harmonic generation occurs when the following conditions are satisfied. First, the fundamental must be guided, which requires

$$n_c^\omega < n_g^\omega \tag{3}$$

where n_c is the bulk index of refraction of the cladding at the fundamental wavelength, and n_g is the bulk index of refraction of the waveguide. Second, the index of the cladding at the harmonic wavelength must be greater than the core

$$N^\omega < n_c^{2\omega} \tag{4}$$

where N^ω is the effective guided-wave index for the lowest order mode at the fundamental frequency and $n_c^{2\omega}$ is the bulk index of refraction of the cladding at the harmonic wavelength. Under these conditions, harmonic light traveling in the core will travel faster than harmonic light propagating in the cladding as a radiation mode. Phase fronts exiting at the Čerenkov angle, θ_c, will be in phase and interfere constructively as light propagates down the waveguide. The Čerenkov angle is then defined as

$$\theta_c = \cos^{-1}\left(\frac{N^\omega}{n_c^{2\omega}}\right). \tag{5}$$

An illustration of the Čerenkov phase-matching process is shown in Figure (6) for one-sided Čerenkov second harmonic generation. Harmonic light generated in the waveguide is represented by rays in the waveguide. Ray $\overline{AB}$ which represents second harmonic light radiated into the cladding is noncritically and automatically phase-matched with the ray leaving at point C at the wavefront $\overline{BC}$ when both rays leave the waveguide at an angle θ_c. The length of $\overline{BC}$ measures the degree of transverse phase-matching.

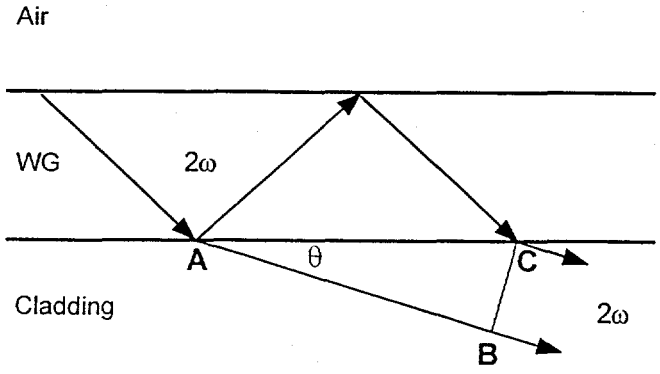

FIG. 6. Illustration of one-sided Čerenkov phase-matching.

Hashizume and co-workers have recently analyzed Čerenkov generation theoretically for a variety of waveguide configurations, including slab waveguides. [13] The optimum conditions for frequency conversion can be determined by examining the case of TM_{ω}^{0} to $TM_{2\omega}^{0}$ conversion using a d_{22} (2 direction perpendicular to the film) second harmonic coefficient for a symmetric waveguide with a nonlinear guiding layer. In this case the authors show that the second harmonic efficiency, η is given by,

$$\eta = \frac{P^{2\omega}}{P^{\omega}} = \frac{\mu_0 \omega^2 L}{cWD_{eff}^2} P^{\omega} \frac{d_{22}^2}{(N^{\omega})^2 n_2^{2\omega}} \frac{J_1 J_2 S^2}{\sin\theta_c} \tag{6}$$

where μ_0 is the permeability of free space, d_{22} the second harmonic coefficient, θ_c the Čerenkov angle, ω the fundamental frequency, c the speed of light, W the waveguide width, D_{eff} the effective waveguide thickness which relates to the dimensions of the confined light, L the propagation length, and P^{ω} the incident power. Note that the conversion efficiency increases as L rather than L^2 as in other phase-matching schemes. In addition, we define

$$J_1 = \frac{1}{1 + \frac{(\kappa_{2\omega}^2 - \gamma_{2\omega}^2)^2}{4\kappa_{2\omega}^2 \gamma_{2\omega}^2} \sin^2 \kappa_{2\omega} D}, \tag{7}$$

$$J_2 = 1 - \frac{\kappa_{2\omega}^2 - \gamma_{2\omega}^2}{\kappa_{2\omega}^2} \sin^2 \frac{\kappa_{2\omega}}{2} D \tag{8}$$

and

$$S = \frac{\sin(\kappa_{2\omega} + 2\kappa_{\omega})D/2}{(\kappa_{2\omega} + 2\kappa_{\omega})/2} + \frac{\sin(\kappa_{2\omega} - 2\kappa_{\omega})D/2}{(\kappa_{2\omega} - 2\kappa_{\omega})/2} + 2\frac{\sin(\kappa_{2\omega} D/2)}{(\kappa_{2\omega}/2)} \tag{9}$$

where

$$\kappa_{\omega} = \sqrt{k_{\omega}^2 (n_g^{\omega})^2 - \beta_{\omega}^2} \tag{10}$$

and

$$\gamma_{\omega} = \sqrt{\beta_{\omega}^2 - k_{\omega}^2 (n_c^{\omega})^2} \tag{11}$$

with β_{ω} the guided mode propagation constant, k_{ω} the wave vector magnitude, D the waveguide thickness, and κ and γ, the extinction coefficients. Upon examination of Equation (6), three criteria for optimizing the efficiency were found: [13]

1. The quantity S is optimized when transverse phase matching is attained, i.e. $n_g^{\omega} = n_g^{2\omega}$.
2. The quantity J_1 is maximized when $n_c^{2\omega}$ is slightly larger than $n_g^{2\omega}$ causing stronger radiation of the second harmonic into the cladding.
3. Strong confinement of the fundamental or small D_{eff} which corresponds to large $n_g^{\omega} - n_c^{\omega}$.

We have modeled the effect of these three factors from a similar theory of Onda and Ito [12] which also takes into account the effect of absorption in the guiding layer on the efficiency of second harmonic generation. In this modeling, we have used the following reasonable parameters in calculating the second harmonic efficiency:

Waveguide length	$L = 1.0$ cm
Waveguide width	$W = 2\mu$m
Fundamental optical power	$P^{\omega} = 100$ mW
Effective nonlinearity	$d_{eff} = 10$ pm/V
Fundamental wavelength	$\lambda = 800$ nm
Fundamental guide index	$n_g^{\omega} = 1.502$
Harmonic guide index	$n_g^{2\omega} = 1.500$
Fundamental clad index	$n_c^{\omega} = 1.483$
Harmonic clad index	$n_c^{2\omega} = 1.505$

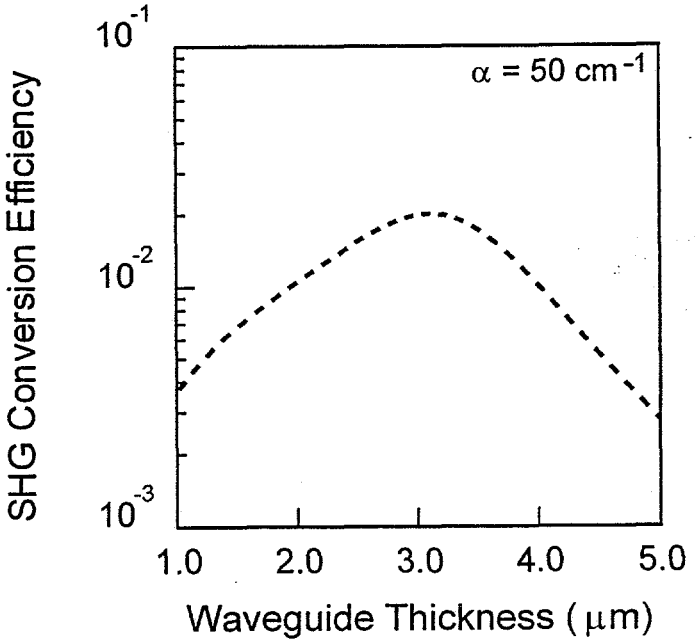

FIG. 7. The dependence of the second harmonic efficiency on the waveguide thickness with the parameters given below.

The conversion efficiency was determined by the program as a function of thickness. An example is shown in Figure (7). This figure represents the optimum efficiency possible with the parameters above. A conversion efficiency of 2% is found at a thickness of about 3.25 μm. It is important to note from Figure (7) that the thickness dependence of the efficiency is not critical; in fact, the conversion efficiency is little affected over a range of up to 0.5 μm. This peak efficiency was obtained by optimizing the refractive indices under the three criteria above using starting values for the refractive indices as listed above. The results are unaffected by the absorption on an order on magnitude basis as shown in Figure (8). By employing anomalous dispersion to obtain transverse phase-matching to $|n_g^{\omega} - n_g^{2\omega}| \sim 0.005$, Čerenkov second harmonic radiation between 100-1000 times that obtained in the literature without transverse phase-matching is expected. The turnover

in the efficiency is due to the absorption length becoming comparable or shorter than the coherence length. It is seen that reasonably small absorption at the second harmonic frequency in the waveguide does not have a major effect on the efficiency since the second harmonic is guided in the cladding. However, some effort is required to minimize absorption, since if the absorption length becomes much smaller than the coherence length, the conversion will be limited.

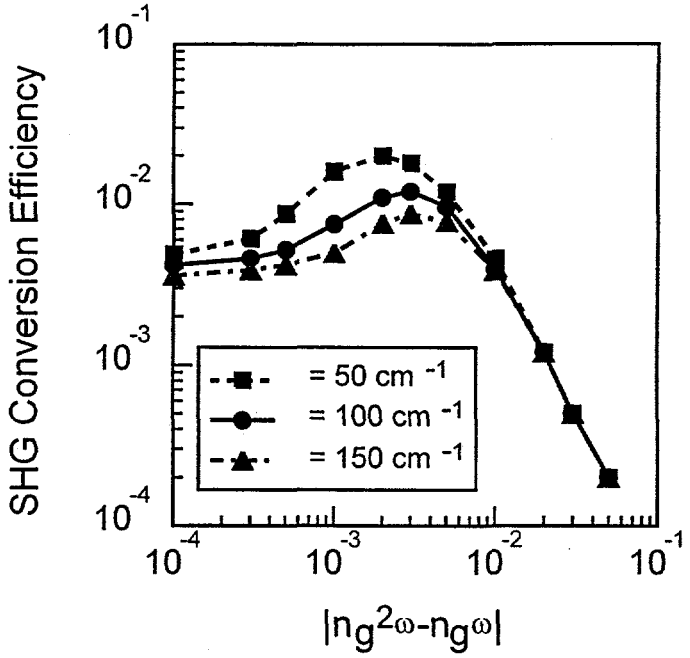

FIG. 8. Čerenkov phase-matched second harmonic efficiency with absorption coefficient as a parameter.

4. SUMMARY

In summary, anomalous dispersion phase-matched second harmonic generation was demonstrated in a poled polymer waveguide. The maximum conversion efficiency was $\eta_{exp} = 1.2 \times 10^{-4}$. Larger conversion efficiencies are can be obtained by confining light to a channel and using host polymers that have greater dispersion, allowing higher concentrations of chromophore to produce phase-matching. Low conversion efficiencies are mostly attributed to the short propagation length of 32 μm. Calculations including absorption at the harmonic predict conversion efficiencies close to 1% for a 2.0 mm propagation length in a 5.0 μm wide channel waveguide.

We have discussed the use of polymeric waveguides in integrated optics devices for frequency conversion. For guided-wave second harmonic devices we have found, through simulations that the efficiency of Čerenkov phase-matched second harmonic generation can be enhanced greatly through the use of anomalous dispersion. The Čerenkov process also overcomes the drawbacks of anomalous dispersion phase-matching, namely, the critical fabrication required, and the effects of residual loss, and possibly two-photon absorption. We have found that a 100-fold increase over current designs can be attained through anomalous dispersion phase-matching. Experiments are in progress to demonstrate the calculations in this paper.

5. ACKNOWLEDGEMENTS

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