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Anomalous Dispersion Enhanced Čerenkov Phase-Matching

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ABSTRACT

We report on a scheme for phase-matching second harmonic generation in polymer waveguides based on the use of anomalous dispersion to optimize Čerenkov phase matching. We have used the theoretical results of Hashizume et al. and Onda and Ito to design an optimum structure for phase-matched conversion. We have found that the use of anomalous dispersion in the design results in a 100-fold enhancement in the calculated conversion efficiency. This technique also overcomes the limitation of anomalous dispersion phase-matching which results from absorption at the second harmonic. Experiments are in progress to demonstrate these results.

I. INTRODUCTION

Waveguide structures are often preferred for nonlinear optical and electro-optic interactions since the waveguide structure provides for highly intense optical radiation to propagate over long distances. This provides for the monotonic build-up of converted light in parametric or frequency conversion interactions, and the accumulation of phase-shifts in optical Kerr and Pockels devices. Among the important potential applications are frequency upconversion for optical communication and data storage. In frequency conversion, the generation of ultraviolet and blue laser sources would find applications in data storage since the data density depends on diffraction limited focusing which in turn is inversely proportional to the wavelength squared. Thus, a low cost short wavelength laser technology is needed for optical storage. In addition, the generation of blue-green and green lasers may be useful for underwater communication. Low cost visible lasers in red, blue, and green would find application in display devices. These devices require materials with large optical nonlinearities and viable phase-matching techniques to maximize conversion. We describe here a scheme using anomalous dispersion to enhance second harmonic generation through optimization of the coherence properties of the interaction.

For frequency conversion devices, the phase-matching property and optical nonlinearity largely determine the device efficiency.^{1,2} Phase-matching refers to the degree of coherence between the nonlinear optical polarization and the generated light. The coherence is generally limited by the fact that the nonlinear polarization and the light radiated by the polarization have different spatial frequencies due to the dispersion of the refractive index. A characteristic length describing the degree of coherence (coherence length) is defined by

$$l_c = \frac{\lambda}{4(n^{2\omega} - n^\omega)}$$

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where n^ω is the refractive index at frequency ω and λ the fundamental wavelength. When not phase-matched, these different spatial frequencies interfere over the propagation length, thus limiting the nonlinear optical conversion. Phase-matching occurs when the two indices of refraction are equal leading to an infinite coherence length. In the phase-matched condition, the dispersion is canceled by one of several means as described below. In this case, the converted intensity grows monotonically with length of propagation.

In integrated optics, the light propagates within a waveguide. Since light in a waveguide is confined to a very small cross-sectional area and propagates over long lengths, more efficient frequency conversion can 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 .³ The quantized properties of waveguide modes allows for additional phase-matching schemes, but may add practical difficulties due to critical design parameters. In the case of waveguides, it is not the bulk refractive indices that must be phase matched, but rather the mode indices. Because guided waves must satisfy the mode conditions, the two polarization eigenmodes propagate with different refractive indices even in an isotropic material, and the dispersion in the effective mode index of refraction is greater than that of the bulk index of refraction.

Efficient operation also requires low-loss waveguides where the guided-wave energy is highly confined over considerable distance in the nonlinear optical waveguide so that a large fraction of energy is converted. In addition, the fundamental and second-harmonic waves need to be confined in such a way that much of their energy interferes constructively. All the modes not only exhibit confinement in the central guiding region, but also in the cladding region as an evanescent field. The fraction of energy confined in each depends on the thickness of the guiding region, the relative bulk indices in the regions, the wavelength, and the order of the mode. For nonlinear waveguides, the efficiency is optimized with the lowest order mode since it yields the most energy in the waveguide. For a nonlinear cladding, a higher order mode is preferred since it will yield more energy in the cladding. In most schemes, both the fundamental and second harmonic waves are confined to the waveguide, and the efficiency is related to the overlap integral between the fundamental and harmonic guided modes.

Phase-matching has been achieved in waveguides using a number of techniques including the use of birefringence, modal dispersion, and periodic structures. Another method of phase-matching that has received considerable attention consists of having the second harmonic light not confined in the waveguide.^{8,9} This technique is known as Čerenkov phase-matching since the radiation from the waveguide exits in a cone in analogy to Čerenkov radiation from charged particles. In this case, the lowest order mode cutoff occurs at some wavelength between the fundamental and the second harmonic. The phase-matching condition is attained in a noncritical way for certain wave-vectors in the radiated mode. A number of studies have been carried out, both theoretical⁹⁻¹⁷ and experimental. Experiments have been carried out for inorganic crystals^{9,18,19}, crystal cored fibers^{20,21}, polymers²²⁻²⁴, and Langmuir-Blodgett films^{25,26}. All experimental studies to date are in regimes far from optimum conditions delineated in recent theoretical studies. The scheme proposed here utilizes anomalous dispersion to realize the optimum conditions for the Čerenkov process.

II. ANOMALOUS DISPERSION PHASE-MATCHING

We have been investigating anomalous dispersion as a method of phase-matching.^{2,27,28} In this scheme, a nonlinear optical moiety is chosen so that it absorbs at frequencies between the fundamental and the second harmonic, but minimally at either frequency. The refractive index of the moiety will exhibit

anomalous frequency dispersion (lower refractive index at higher frequency), so that when incorporated with a normally dispersive polymer a concentration will be found where the positive dispersion of the polymer cancels the negative dispersion of the nonlinear optical moiety yielding the phase-matched condition.

The anomalous dispersion scheme is made possible by recent investigations of electric-field-poled polymeric materials as second-order nonlinear optical materials.^{2,29} In these materials, an organic molecule which possesses a large optical nonlinearity is incorporated in a passive glassy host, usually a polymer. The material is given second-order nonlinear optical properties through an electric-field poling process. In this process, the molecules are aligned by an electric field when the host is in a state of enhanced molecular mobility. The alignment is frozen-in by returning to a state of low molecular mobility. The glass-rubber phase transition in certain polymers provides one possible method for controlling the molecular mobility. These polymeric materials have many attractive processing features such as formation into thin film waveguides on semiconductor substrates.

The ability to adjust the concentration of moieties allows for the anomalous dispersion scheme. This scheme has a number of advantages; foremost is the ability to use molecules that have their first electronic excited state in the visible region of the spectrum. A strong inverse correlation between optical nonlinearity and the energy of the first excited state has been found.²⁸ As seen in Figure 1, anomalous dispersion dyes offer increases in the useful second order hyperpolarizability, β , of about an order of magnitude leading to a two order of magnitude increase in efficiency. A second advantage is the ability to phase-match any component of the nonlinear optical tensor since it is an intrinsic material phase-matched condition, independent of geometry.

Fig. 1 Plot of $\mu\beta$ product vs. useful second harmonic wavelength. The lower line is a least squares fit to the data, the upper (dashed) line is a guide to the eye.

We demonstrated this scheme, in principle, in a liquid solution of Foron Brilliant Blue (FBB) in

acetonitrile.²⁷ Second harmonic generation of 0.954 μm wavelength light was measured as a function of concentration of FBB. The absorption at the second harmonic wavelength was finite, but relatively small. The data in Figure 2 shows a 50 fold increase in signal at the phase-matched concentration of around 0.045 M. Chemical synthetic studies were carried out to identify a nonlinear optical dye which is more appropriate for anomalous dispersion phase matching.³⁰ A barbituric acid dye (BA) was found to possess a sharp absorption in the wavelength range between 400 and 800 nm. The dye structure of BA (molecule X) along with other barbituric acid derivatives are shown in Figure 3, and the absorption spectra in Figure 4. The spectrum of BA indicates low loss at both 400 and 800 nm wavelength. We were able to identify the zero dispersion (400 and 800 nm) concentration of a guided mode in a waveguide composed of PMMA with BA dissolved in, as shown in Figure 5.²

Fig. 2 Second harmonic intensity versus concentration for solutions of FBB in acetonitrile near the anomalous dispersion phase-matched condition.

The anomalous dispersion scheme for waveguided second harmonic generation does suffer from two important drawbacks. First, it contains a critical parameter to achieve full phase-matching. To attain phase-matching, the indices of refraction must be equal within at least a part in 10^4 at 400 and 800 nm. This requires the ability to control concentration and/or thickness to that precision. In both cases, the fabrication would be critical to those parameters. A second important drawback is the residual absorption in the molecule at 400 nm. The absorption, though low, limits the propagation length to less than a millimeter. The measured absorption of BA at the phase-matched concentration at 400 nm is about $\alpha = 20\text{cm}^{-1}$. At 800 nm the absorption is low enough to propagate over several centimeters.

Fig. 3 Molecular structure of barbituric acid dyes. (X=BA)

Fig. 4 Absorption spectrum of barbituric acid dyes in acetonitrile.

III. 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 which is optimized by 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.

Fig. 5 Refractive index difference for waveguides of BA in PMMA as a function of concentration. The index difference is measured at 800 and 400 nm.

If n_c is the index of refraction of the cladding of the waveguide, and n_g is the index of refraction of the waveguide, then conditions for Čerenkov phase-matching include,

$$n_c^\omega < n_g^\omega \quad (2)$$

in order to confine the fundamental frequency and

$$N^\omega < n_c^{2\omega} \quad (3)$$

where N^ω is the effective guided-wave index for the lowest order mode at the fundamental frequency. This second condition is necessary for the radiation of the second-harmonic from the guide into the cladding. This necessity is made clear when the boundary conditions are solved, and the conservation of transverse momentum leads to the phase-matched condition,

$$n_c^{2\omega} \cos \theta = N^\omega. \quad (4)$$

Thus the phase-matching condition is noncritically and automatically attained when second-harmonic light radiates into the cladding at an angle from the propagation direction given by,

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

If the cladding thicknesses are properly chosen the harmonic light can enter the cladding and propagate as a guided mode.

In a recent study, Hashizume and co-workers have analyzed Čerenkov generation theoretically for a variety of waveguide configurations, including slab waveguides.¹⁵ 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,

$$\eta = \frac{P_{2\omega}}{P_\omega} = \frac{\mu_0\omega^2 L}{cW D_{eff}^2} P_\omega \frac{d_{22}^2}{(N\omega)^2 n_{2\omega}^2} \frac{J_j J_i S^2}{\sin \theta} \quad (6)$$

where μ_0 is the permeability of free space, d_{22} the second harmonic coefficient, θ 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 intensity increases as L rather than L^2 as in other phase-matching schemes. In addition,

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

$$J_i = 1 - \frac{\kappa_{2\omega}^2 - \gamma_{2\omega}^2}{\kappa_{2\omega}^2} \sin^2 \frac{\kappa_{2\omega}}{2} D \quad (8)$$

and

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

where

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

and

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

with β_ω the guided mode vector magnitude, 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:¹⁵

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_i 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 Ito¹⁴ 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	$L = 5$ cm
Waveguide width	$W = 2\mu\text{m}$
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. 6 The dependence of the second harmonic efficiency on the waveguide thickness at optimum conditions (see Figure (7) below).

The conversion efficiency was determined by the program as a function of thickness. An example is shown in Figure (6). This figure represents the optimum efficiency possible with the parameters above. The high conversion efficiency of 16% is found at a thickness of about $3.75 \mu\text{m}$. It is important to note from Figure (6) that the thickness dependence of the efficiency is not critical; in fact, the conversion efficiency is little affected over a range of up to $0.5 \mu\text{m}$. 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. Details of the optimization for the three criteria are shown in Figure (7). Here it is seen that the transverse phase-matching condition ($n_g^\omega - n_g^{2\omega}$) is most important. By employing anomalous dispersion to obtain 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. In addition, Figure (7) indicates that conversion does not critically depend on $n_g^\omega - n_g^{2\omega}$, and absorption does not affect the order of magnitude of conversion. 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. The fundamental confinement ($n_g^\omega - n_c^\omega$) also has a significant effect on the conversion efficiency. However, most guides would be designed in the range of $n_g^\omega - n_c^\omega \sim 0.005 - 0.1$ due to material constraints. In this region, the confinement has less of an effect. The strength of harmonic radiation ($n_c^{2\omega} - n_g^{2\omega}$) is seen to have little effect on the conversion efficiency, however; the small index difference between core and cladding will cause additional waveguide losses because of the absorption of the harmonic evanescent fields in the core. This effect can be minimized by using thicker cladding layers or increasing the index difference between core and cladding. The efficiencies we obtain will have small corrections due to pump depletion and scattering losses.

IV. SUMMARY

To this time, conversion efficiency by the Čerenkov method has been limited by the inability to employ materials and structures which optimize the process. We have shown that the Čerenkov process can be optimized by rational design using anomalous dispersion and polymeric materials resulting in extremely large conversion efficiencies without critical steps. This optimization occurs near the intrinsic phase-matched condition, but is not critically dependent.

We have reviewed the use of polymeric waveguides in the design of nonlinear 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. We have found that a 100-fold increase over current designs can be attained through anomalous dispersion phase-matching.

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FIG. 7. Optimization of Čerenkov phase-matched second harmonic efficiency with absorption coefficient as a parameter. a) Transverse phase-matching (using anomalous dispersion), $n_g^\omega - n_g^{2\omega}$. b) Fundamental confinement, $n_g^\omega - n_c^\omega$. c) Harmonic confinement, $n_c^{2\omega} - n_g^{2\omega}$.

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