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Phase-matched second harmonic generation over 7 mm is demonstrated using the modal dispersion of a composite 4-dimethylamino-4'-nitrostilbene/polyetherimide channel waveguide. For a 1-mm-long sample a record figure of merit for polymers of $\eta = P_{2\omega}/(P_{\omega}L^2) = 14\%/W\text{ cm}^2$ was measured at 1550 nm. This is comparable to the value reported for LiNbO₃ at that wavelength. The efficiency in longer samples is limited by fundamental and harmonic waveguide losses. © 1996 American Institute of Physics. [S0003-6951(96)01253-3]

Second harmonic generation (SHG) has been a powerful tool for accessing shorter wavelengths in the blue and UV region. Several inorganic crystals have been employed successfully for that purpose. Nonlinear optical polymers, on the other hand, have mostly a strong absorption in this region, making them less attractive for these applications. However, with the realization that second order nonlinear effects can also be used for cascading phenomena such as all-optical switching, spatial solitary waves, etc. and for wavelength conversion, a new field of potential applications is emerging.^{1,2} Cascading, for example, requires phase-matched efficient SHG, low propagation loss at the fundamental and harmonic wavelength, and operation at one of the telecommunication wavelengths (1300 or 1550 nm) is desirable. So far, these conditions have been implemented only in ferroelectric crystals, like KTP³ and LiNbO₃,⁴ but poled polymers with comparable or even higher nonlinear $d^{(2)}$ -coefficients could be a cheaper alternative to these traditional SHG materials. So far, the efficiencies reported for poled polymers using quasi-phase matching (QPM),⁵⁻⁸ modal dispersion phase matching (MDPM),⁹⁻¹³ or anomalous dispersion phase matching¹⁴ do not compete with those of ferroelectric crystals. In this letter we report on phase-matched SHG in a composite channel waveguide made of 4-dimethylamino-4'-nitrostilbene (DANS) polymer and polyetherimide. Our SHG figure of merit $\eta = P_{2\omega}/(P_{\omega}L)^2 = 14\%/W\text{ cm}^2$ is competitive with those of the best inorganic crystals.¹⁵ Phase matching was maintained for up to 7 mm, demonstrating that multilayer spin coating can provide the thickness tolerances necessary for useful propagation distances.

Efficient MDPM involves the matching of two waveguide modes in terms of both their phase velocities (phase matching) and their optical fields (mode matching). In our case, phase matching is achieved between the TM₀₀^ω and TM₁₀^{2ω} modes using the modal dispersion of the waveguide. For a perfectly symmetric DANS waveguide, this combination diminishes the figure of merit η , because the positive and negative contributions cancel each other in the overlap integral

$$\left| \int \int d^{(2)}(x,y) E_{\omega}^2(x,y) E_{2\omega}^*(x,y) dx dy \right| \propto \sqrt{\eta} \quad (1)$$

due to the sign change of the harmonic field $E_{2\omega}^*(x,y)$. To optimize the efficiency, our guiding region consists of a linear polyetherimide layer and a nonlinear DANS-polymer layer. Figure 1 shows the vertical modal field distribution of the fundamental and harmonic fields. Contributions to the overlap integral come only from the DANS layer ($d^{(2)} \neq 0$), where both fields are positive. This geometry allows a significant improvement of the overlap integral over our previously reported results.¹³

Unlike QPM, MDPM does not require a periodic structure in propagation direction. This simplifies the fabrication of low loss waveguides and is an advantage over QPM, where fabricating these nonlinear phase-match gratings in poled polymers is technologically challenging.⁷ Another issue is the sensitivity of the phase-matching condition to fluctuations of waveguide parameters p (e.g., thickness, refractive index, temperature, etc.). To assess this problem, the wave vector mismatch

$$\Delta\beta = \frac{4\pi}{\lambda} (N_{2\omega} - N_{\omega}) - \frac{2\pi}{\Lambda} \quad (2)$$

FIG. 1. Calculated vertical field distribution of the TM₀₀^ω and TM₁₀^{2ω} modes of the composite waveguide. The thicknesses of the guiding layers are $h_{\text{DANS}} = 0.53\text{ }\mu\text{m}$ (DANS polymer) and $h_{\text{PEI}} = 0.60\text{ }\mu\text{m}$ (polyetherimide).

FIG. 2. Sample construction of the composite poled polymer channel waveguide.

(λ —fundamental wavelength, $N_{\omega,2\omega}$ —fundamental and harmonic effective index, Λ —QPM grating period, last term present only for QPM) can be expanded into a Taylor series¹⁶

$$\Delta\beta(p) = \left. \frac{\partial\Delta\beta}{\partial p} \right|_{p_0} \delta p + \frac{1}{2} \left. \frac{\partial^2\Delta\beta}{\partial p^2} \right|_{p_0} (\delta p)^2 + \dots, \quad (3)$$

where $\Delta\beta(p_0)=0$ denotes the phase-matching point. QPM has the advantage that noncritical phase matching ($\partial\Delta\beta/\partial p|_{p_0}=0$) can be designed for one of these waveguide parameters p by an appropriate choice of the grating period.¹⁷ This extra degree of freedom is not present in MDPM, making precise control of the waveguide parameters a necessity. For a propagation length of 1 cm, the acceptable thickness tolerances δh for each of the two guiding layers are only a few nm. Besides high efficiencies, our research has therefore focused on the question of whether spin coating technology can provide the accuracy necessary to maintain phase matching over useful distances.

The samples were fabricated by sequential multilayer spin coating onto a silicon wafer. Special care was taken to avoid dissolving away a previously deposited polymer layer during spin coating. The guiding region consists of a nonlinear DANS-polymer layer and a linear polyetherimide layer, sandwiched between two 3- μm -thick layers made of PC polymer, which is a crosslinkable buffer polymer provided by AKZO. Poling was performed by applying a nominal electric field of 180 V/ μm between the top aluminum electrode and the silicon substrate near the glass temperature of the DANS polymer (142 °C). With the field still applied the samples were then cooled down to room temperature, freezing the orientation of the DANS chromophores and giving rise to a macroscopic nonlinearity ($d^{(2)}$ coefficient). Subsequently the samples were diced into pieces of different lengths (1.0 to 7.2 mm) for end-fire coupling. Horizontal light confinement is provided by photobleaching of the DANS chromophore through a mask, creating channel waveguides similar to the strip-loaded geometry (Fig. 2).

The linear and nonlinear characterization of the samples was performed with a NaCl:OH⁻ color center laser, synchronously pumped by a Nd:YAG laser. It provides 6 to 8 ps pulses tunable from 1500 to 1650 nm. First the throughput for samples of different lengths ($L=1.0, 2.0, 3.0$, and 7.2 mm) was carefully measured. In each sample the throughput was averaged for a number of identical waveguides. From these measurements an incoupling efficiency of 40% and a

fundamental waveguide loss $\alpha_\omega=7\text{ dB/cm}$ was deduced. With the same procedure the waveguide loss $\alpha_{2\omega}$ at the SHG wavelength was determined using a Ti:sapphire laser. The value is approximately 20 dB/cm and decreases towards the infrared. We attribute this high loss coefficient to the absorption tail from the broad charge transfer peak of the DANS chromophore. This interpretation is consistent with fits to the absorption in the DANS side-chain polymer.¹⁸ This high SHG absorption creates another problem, since it causes bleaching of the chromophore, which was observed at SHG average intensities above 100 W/cm².

An important device performance parameter is the figure of merit (FOM), or normalized efficiency, which is defined as $\eta = P_{2\omega}/(P_\omega L)^2$. Occasionally the FOM is calculated based on the fundamental and harmonic output, however we prefer to give the internal FOM, which is based on the guided fundamental peak power P_ω at the waveguide entrance and the guided harmonic peak power $P_{2\omega}$ at the end. The highest FOM $\eta=14\%/W\text{ cm}^2$ was measured in a 1-mm-long waveguide. To our knowledge this is the highest non-resonant value reported for poled polymers at this wavelength. For longer samples this number drops due to the presence of waveguide loss. The dependence of the FOM on the sample length L agrees fairly well with the theoretically expected formula

$$\eta = 2 \exp[-(\alpha_\omega + \alpha_{2\omega}/2)L] \cdot \frac{\cosh(\Delta\alpha L) - \cos(\Delta\beta L)}{(\Delta\alpha L)^2 + (\Delta\beta L)^2} \cdot \eta_0 \quad (4)$$

that takes into account the measured waveguide losses ($\alpha_\omega = 7\text{ dB/cm}$, $\alpha_{2\omega} = 20\text{ dB/cm}$). Here η_0 denotes the FOM of the lossless case and $\Delta\alpha = \alpha_{2\omega}/2 - \alpha_\omega$. When this behavior is found it implies that the efficiency of the longer samples is limited by these losses, rather than fluctuations and/or a drift with distance of the phase-matching condition.

To clarify this question, the spectral dependence of the efficiency was investigated by tuning the fundamental wave-

FIG. 3. SHG tuning curve for a 3-mm-long waveguide. The inset shows a tuning curve of a 7.2-mm-long sample in the same wavelength range (1495–1520 nm).

FIG. 4. SHG acceptance bandwidth (FWHM) vs sample length L (measured data and theoretical fit).

length while recording the SHG power. Figure 3 shows two examples of such SHG tuning curves for a 3- and a 7.2-mm-long sample. In the presence of waveguide loss the FOM is described by Eq. (4). But for $\Delta\alpha L = (\alpha_{2\omega}/2 - \alpha_{\omega})L < 2$ the $\Delta\beta L$ -dependence function can be approximated by the usual $\text{sinc}^2(\Delta\beta L/2)$ function of the lossless case.¹⁹ Since $\Delta\alpha L \leq 0.5$ for our waveguides, the SHG acceptance bandwidth is therefore nearly unaffected by the waveguide loss. This enables the investigation of the phase-matching behavior even for propagation lengths longer than the individual loss lengths $L_{\omega,2\omega} = 1/\alpha_{\omega,2\omega}$. The full width at half maximum (FWHM) of the bandwidths has been measured for samples of different lengths, as shown in Fig. 4. The curve displaying the theoretically expected $1/L$ -dependence of the bandwidth is in very good agreement with the measured data. This supports our conclusion that phase matching is maintained for up to 7 mm.

In conclusion, we have demonstrated an SHG figure of merit $\eta = 14\%/W \text{ cm}^2$ for a 1-mm-long waveguide, comparable to the $27\%/W \text{ cm}^2$ reported in LiNbO_3 .¹⁵ Phase matching was maintained for up to 7 mm with reduced efficiencies.

This demonstrates the usefulness of multilayer spin coating technology for phase-matched SHG applications. The waveguide losses still have to be reduced to an acceptable level, which is feasible with other existing nonlinear polymers.

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