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Second-harmonic generation in reactive-ion etched *N'* ethyl *N*-ethanol-4-(nitrophenylazo)phenylamino polymer waveguides at telecommunication wavelengths

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Second-harmonic generation using modal dispersion phase matching has been demonstrated in reactive-ion etched waveguides based on the organic nonlinear polymer poly (methyl methacrylate)-Disperse Red 1 [*N'* ethyl *N*-ethanol-4-(nitrophenylazo)phenylamino]. The measured propagation losses were 6 dB/cm at the fundamental and 10 dB/cm at the second-harmonic wavelength, dramatically less than we obtained previously for the harmonic in photobleached waveguides. A figure of merit of $\eta=0.1\%/W$ for second-harmonic generation was obtained with a modest nonlinearity of 3 pm/V at 1607 nm in a 2 mm long waveguide. © 1999 American Institute of Physics. [S0021-8979(99)06318-5]

I. INTRODUCTION

Second-order nonlinear optical polymer materials have been studied because of their applications in high speed electro-optic modulators,¹ second-harmonic generation (SHG),² $\chi^2:\chi^2$ -cascading,³ etc. In particular, cascading of second-order processes has been shown to give rise to large nonlinear phase shifts, thus giving attractive possibilities for all-optical signal processing if efficient harmonic generation can be achieved.⁴

Efficient SHG requires phase matching. Phase matching in optical waveguides can be achieved using various configurations, e.g., birefringence phase matching,⁵ modal dispersion phase matching (MDPM),⁶ quasiphasematching,⁷ and Cerenkov-type phase matching.⁸ These phase-matching schemes have been demonstrated in optical waveguides of the well-known inorganic nonlinear optical crystals with relatively large nonlinear optical coefficients, such as LiNbO₃,⁹ KNbO₃,⁵ LiTaO₃,¹⁰ and KTiOPO₄.¹¹ A number of techniques have recently led to promising SHG in polymers.^{12–15} The Disperse Red 1 (DR1) results were very promising in terms of normalized figures of merit, but the absolute conversion efficiency was limited by the very large losses at the second harmonic, of the order of 100 dB/cm.¹⁴ Here, we report on using reactive ion etching to fabricate low loss DR1 channel waveguides and report on the first modal dispersion phase matching experiments in these waveguides. The waveguides were fabricated by spin coating and oxygen (O₂) plasma etching.¹⁶

II. DR1 POLYMER WAVEGUIDES

In polymers, the macroscopic nonlinearity is determined by the molecular hyperpolarizability of the nonlinear optical

chromophores, their number density within the polymer, and their degree of orientational order. This noncentrosymmetric order is usually achieved by alignment of the chromophore dipoles in a high static electric field that is applied after heating the polymer close to its glass transition temperature T_g . The multiple layers needed to make the waveguides are fabricated by the spinning of successive polymer layers. Hence, it is straightforward to make the high index guiding films from combinations of second-harmonic active and inactive films. This facilitates MDPM which has been successfully demonstrated in 4-diethylamino-4'-nitrostilbene (DANS) and DR1 waveguides.^{13,14} MDPM requires tight control of the thicknesses of the layers in the waveguide structure. Because the second-harmonic wave is carried in a higher spatial order mode, a special waveguide structure needs to be designed in order to avoid cancellation of the overlap integral given by

$$\left| \int \int dx dy d(x,y) e_{\omega}^2(x,y) e_{2\omega}^*(x,y) \right|^2,$$

where d represents the nonlinear-optical coefficient and e_{ω} , and $e_{2\omega}$ denote the fundamental and second-harmonic fields, respectively. Figure 1 shows the basic planar structure used in this work. A Disperse Red 1-poly (methyl methacrylate) side-chain polymer with 24% dye concentration by weight and a glass transition temperature of 131 °C is used as the nonlinear material. The core of the waveguide consists of a nonlinear DR1 and a linear polycarbonate (PC) layer, sandwiched between two lower index poly (methyl methacrylate) (PMMA) cladding layers. The field distributions of both the fundamental TM_{00}^{ω} and the second-harmonic $TM_{01}^{2\omega}$ are shown in Fig. 1(a). The bottom cladding layer was deposited with a thickness of more than 3 μm to avoid optical tunneling losses into the Si substrate. The nonlinearity, schematically shown in Fig. 1(b), is “turned off” in the PC layer when the second-harmonic field changes sign. By this means a nonvanishing overlap integral is obtained.

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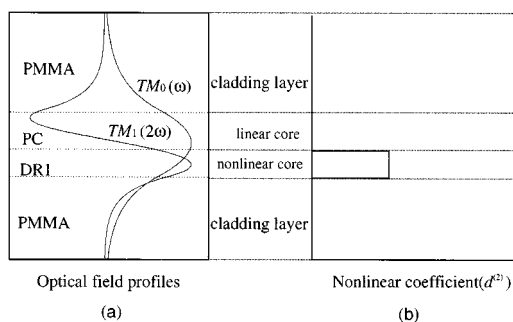


FIG. 1. (a) Basic waveguide structure for MDPM SHG and optical field distributions for the $TM_{00} \rightarrow TM_{01}^{2\omega}$ interaction. (b) Schematic representation of the variation of the optical nonlinearity across the height dimension of the waveguide.

In order to design a phase-matched structure, the refractive indices of all materials were determined at several wavelengths between 633 and 1550 nm using the m -line technique with an accuracy of 10^{-3} . The slab waveguide structure was prepared by multilayer spin coating onto a silicon substrate. The layer thickness was controlled by adjusting both the polymer's concentration in the solution and the rotation (spin) speed. In order to remove the solvent in the thin film polymers, the sample was baked for several hours after every spin-coating step.

For efficient SHG, it is essential to provide optical confinement in two dimensions, i.e., to create a channel waveguide. Such structures can be formed by for example plasma etching or photobleaching. Photobleaching with UV light from a Hg lamp has often been used to fabricate channel waveguides based on nonlinear polymers. However, photobleaching is unsuitable for polymers with a low photosensitivity that would require long bleaching times and high UV intensities. For DR1 very long bleaching times of more than 24 h with intensities of 20 mW/cm^2 would be needed due to the low spectral content of the "UV" lamp we used in the appropriate wavelength range below 400 nm.¹⁷ In this article, channel waveguides based on DR1 were fabricated by O_2 plasma etching with precise control of the etching. The waveguide structure is depicted in Fig. 2. A channel pattern was defined on the cross-linkable PC layer by a standard photolithographic process. Subsequently, the PC polymer layer was etched to a depth of $0.4 \mu\text{m}$ [Fig. 2(a)] to form a single mode ridge waveguide at the fundamental wavelength. A PMMA polymer layer was then spin-coated as a top-cladding layer. A gold electrode was deposited for poling using thermal evaporation. Poling was done using the contact-poling method¹⁸ at a temperature of 127.4°C near the glass transition temperature of DR1 ($T_g = 131^\circ\text{C}$) for about 1 h with a dc bias voltage of 650 V applied across the four polymers layers of $7.4 \mu\text{m}$ total thickness as shown in Fig. 2(b). Note that no effort was taken to optimize the field across the nonlinear polymer layer by optimizing the relative T_g 's of the different layers. With the field still applied the sample was cooled down to room temperature, fixing the orientation of nonlinear chromophores resulting in a macroscopic nonlinearity [$d^{(2)}$ coefficient]. The nominal poling field was approximately $90 \text{ V}/\mu\text{m}$, averaged across the full

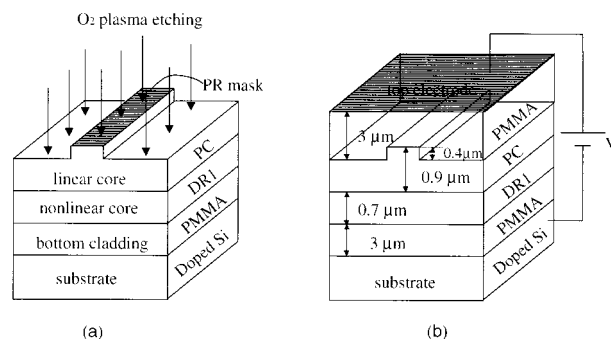


FIG. 2. (a) The ridge waveguide structure fabricated by using O_2 plasma etching with a photoresist mask. (b) Contact electrode poling configuration for the nonlinear DR1 side-chain polymer.

structure. Finally, the waveguides were cleaved into pieces of different lengths.

III. WAVEGUIDE CHARACTERIZATION

In the past, the second-harmonic conversion efficiency of polymer waveguides was often limited by high losses at the fundamental and second-harmonic waves.¹⁴ In particular, high absorption losses at the second-harmonic wavelength of $0.78 \mu\text{m}$ of $\approx 100 \text{ dB/cm}$ were shown to cause a considerable reduction in the conversion efficiency. To estimate the absorption loss, planar waveguides of DR1 of $1.5 \mu\text{m}$ thickness were fabricated on silicon oxide and their propagation loss was measured. This result is shown in Fig. 3(a). The measured loss was less than 3 dB/cm in the spectral range 0.78 – $1.6 \mu\text{m}$. The loss in the DR1 polymer due to absorption at the second-harmonic wavelength of $0.78 \mu\text{m}$ was estimated to be more than one order of magnitude smaller than that reported for the DANS side-chain polymer that has given the highest reported SHG conversion efficiency in polymer waveguides to date. The propagation losses of the channel waveguide used for SHG measured by the cut-back method are shown in Fig. 3(b). The second-harmonic loss was measured by using a laser diode at $0.78 \mu\text{m}$ and a Ti:sapphire laser at $0.83 \mu\text{m}$. The measured propagation loss in the channel waveguide was below 6 dB/cm for the fundamental and 10 dB/cm for the second-harmonic, respectively.

For the second-harmonic measurements, a NaCl:OH^- color center laser (Burleigh FCL 120) synchronously pumped by a mode-locked Nd:YAG laser with a repetition rate of 76 MHz (Coherent Antares) was used. The system produces 7 – 9 ps pulses with a tunability from 1500 to 1650 nm . The laser beam was polarized such that a TM mode was excited in the waveguide since the DR1 chromophores in the sample were aligned perpendicular to the substrate. The beam was end-fire coupled into the channel waveguide using a $\times 40$ objective lens. The power was measured using Ge and Si detectors for the fundamental and second harmonic waves, respectively.

The measured second-harmonic power as a function of wavelength is shown in Fig. 4. The peak normalized conversion efficiency, defined by $\eta = P_{2\omega}/P_{\omega}^2$, was $0.1\%/W$ in the 2 mm long waveguide (which corresponds to $2.5\%/W\text{cm}^2$).

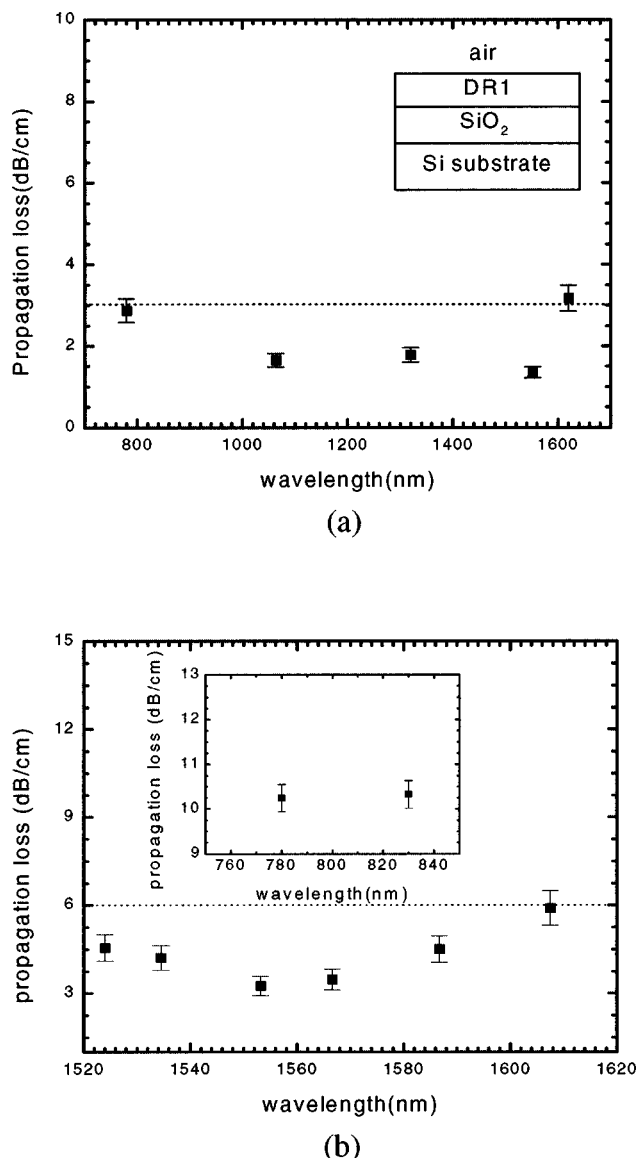


FIG. 3. (a) Measured planar waveguide loss of nonlinear DR1 side chain polymer at near infrared wavelengths. The planar waveguide structure is shown in the inset. (b) The measured channel waveguide loss of the SHG device for fundamental and second-harmonic wavelengths (inset).

The phase-matching wavelength was 1607 nm. Based on the measured losses and conversion for the 2 mm sample, we calculate from

$$P_{2\omega} = 2\eta_0 P_{\omega}^2 e^{-(\alpha_{\omega} + \alpha_{2\omega}/2)L} \frac{\cosh(\Delta\alpha L) - \cos(\Delta\beta L)}{(\Delta\alpha)^2 + (\Delta\beta)^2},$$

the variation in the guided wave power with sample length,¹⁴ shown in Fig. 5. Here $\Delta\alpha = 0.5\alpha_{2\omega} - \alpha_{\omega}$. The peak in the conversion efficiency will occur for a sample length of about 8 mm, which is far from the sample length used. Hence, the estimated peak conversion efficiency for an optimum sample would be 0.3%/W, the best obtained to date for polymers.

It can be seen from Fig. 4 that the measured tuning curve is distorted as compared to the theoretical curve, indicating an effective phase-matching length shorter than the sample length. From the measured conversion efficiency we estimate a nonlinear coefficient of approximately 3 pm/V, based on a

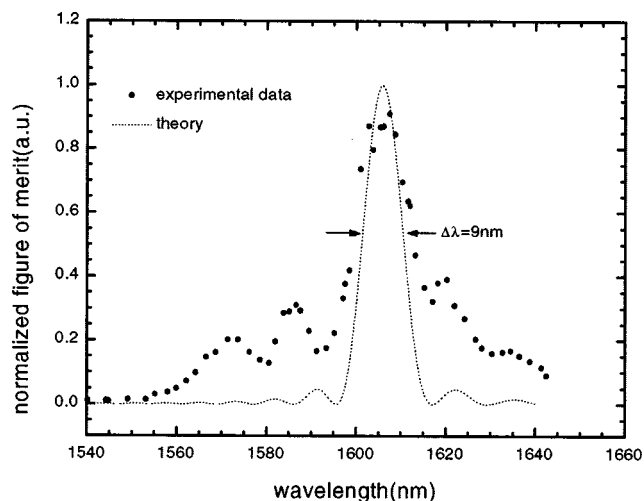


FIG. 4. SHG spectrum of a 2 mm long waveguide as a function of wavelength. The phase matched resonance peak is at 1607 nm. The dotted line shows the calculated spectrum.

2 mm phase-matched sample length. Also, the phase-matching wavelength for the TM_{00}^{ω} and $TM_{01}^{2\omega}$ interaction was shifted by about 50 nm relative to the expected value. We attribute these distortions to the different poling field strengths in the ridge and lateral surrounding regions, respectively, which may result in variations of the refractive indices. In addition, partial material mixing at the interfaces between the polymer layers may have occurred giving rise to further index nonuniformities.

The measured conversion efficiency is comparable to the highest reported previously for DR1 side-chain polymer waveguides.¹⁴ In that case, the effective nonlinearity was at least larger by a factor of two due to the reversal of the nonlinearity in the appropriate part of the waveguide necessary to remove destructive interference from the overlap integral. Note that $\eta \propto d_{\text{eff}}^2$. It is also comparable to the one

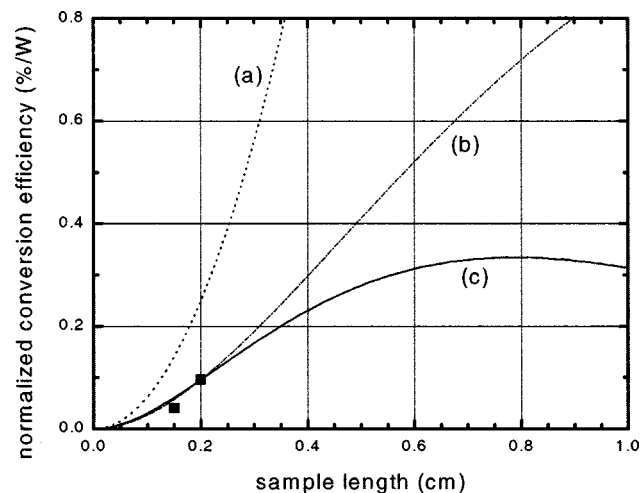


FIG. 5. Calculated variation in the SHG conversion efficiency in %/W vs phase-matched channel waveguide length for the experimental parameters. The squares indicates the measured value. The lines represent the calculation for (a) a lossless waveguide, (b) a waveguide with $\alpha_{\omega} = 3$ dB/cm and $\alpha_{2\omega} = 5$ dB/cm, and (c) $\alpha_{\omega} = 6$ dB/cm and $\alpha_{2\omega} = 10$ dB/cm, with a nonlinear coefficient of 3 pm/V.

reported for DANS.¹³ However, compared to DANS, DR1 is advantageous because of its much lower absorption at the second-harmonic wavelength. The existing structure offers a potential improvement by one order of magnitude over previous results when samples phase matched over 8 mm can be made. By further optimization of the waveguide design and poling procedure, we expect to increase the second-harmonic figure of merit by at least another order of magnitude.

In conclusion, we have demonstrated design, fabrication, and characterization of a low loss DR1 waveguide for SHG using MDPM at a wavelength of 1.6 μm . O₂ plasma etching was used to fabricate polymer channel waveguides with low propagation loss at both wavelengths, offering an alternative to UV photobleaching. By further development of this fabrication technique, we ultimately envision the use of such waveguides for $\chi^2:\chi^2$ cascading based all-optical devices operating at telecommunication wavelengths.

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