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We have investigated three different quasi-phase-matching approaches to second-harmonic generation (SHG) in DANS (4-dimethylamino-4′-nitrostilbene) poled polymer channel waveguides at 1.5 μm. Periodic photobleaching and periodically poled electrodes deposited directly on the film produced unacceptably high propagation losses. However, periodic electrodes on the substrate gave low losses and useful SHG. © *1996 American Institute of Physics.* [S0003-6951(96)04009-8]

The field of second-order nonlinearities in waveguides has found a significant application in cw second-harmonic generation (SHG), which enabled the fabrication of compact, blue light sources. More recently, it has gained new attention due to the possibility of using cascaded $\chi^{(2)}$ effects for all-optical switching, spatial solitons, etc.[1] Because cascading applications usually require pulsed sources, frequently at communications wavelengths, it is desirable to both work at specific wavelengths and minimize the refractive index dispersion with wavelength. Of the different phase-matching techniques demonstrated to date, one of the most successful has been quasi-phase-matching (QPM) essentially because it allows any frequency to be doubled.[2] It involves a periodic modulation of the refractive index or the nonlinear coefficient $d^{(2)}$ $(2\omega; \omega, \omega)$ such that the harmonic fields generated in different parts along the waveguide interfere constructively at the output. QPM also allows the phase matching of (a) the diagonal $d^{(2)}$ tensor elements which are typically the largest and (b) the lowest order mode which typically leads to the highest waveguide overlap integrals (and therefore efficiency), by choosing the appropriate periodicity for the nonlinear grating. These conditions have been demonstrated primarily in QPM inorganic crystal waveguides to date.[3]

Another material class which can exhibit very large second-order nonlinearities is poled polymers. They could ultimately have certain advantages over ferroelectric materials due to ease of processing, low costs, etc. Periodic poling,[4–6] photobleaching,[7] and laser ablation[8] have already been used to create a periodic modulation of the $d^{(2)}$ coefficient in polymers. These studies were all performed on different materials and with different processing conditions. As a result, to date no direct comparison between any of these approaches has been carried out. In this letter, we report QPM-SHG implemented by periodic photobleaching and by periodic poling with different electrode locations and compare their relative problems and merits in the side-chain DANS (4-dimethylamino-4′-nitrostilbene) polymer.

The samples were fabricated by multilayer spin coating onto silicon or fused silica substrates. In order to spatially separate the guided mode fields from the absorbing electrodes, the guiding DANS layer was sandwiched between two buffer layers (PC polymer, provided by AKZO). All three polymeric layers were 2.1 μm thick and the aluminum electrodes used for poling had a thickness of about 0.05 μm. For the photobleaching study, a planar electrode was first deposited on the glass substrate, the multilayers were then spun on and a second planar electrode was deposited onto the buffer film surface. After poling, the top electrode was removed by chemical etching. A 14 μm period grating was photobleached into the slab waveguide by illumination from above with an argon ion laser through a mask. The grating periodicity was determined from $\Lambda = 2\pi/|\Delta\beta|$, $\Delta\beta = 2\beta_1 - \beta_2$, where β_1 and β_2 are the fundamental and harmonic propagation wave vectors of the waveguide. To create digital electrodes with this periodicity, one of the aluminum layers was photolithographically patterned. Two sets of periodically poled samples were fabricated, the first on a silicon substrate (which acted as the bottom electrode) with the patterned electrode on top of the multilayer stack, and the second with the patterned electrode beneath the polymer stack (directly on the fused silica substrate, see Fig. 1). All of the samples were poled with an electric field of 100 V/μm for 30 min at 137 °C (near the glass transition temperature of 142 °C) and subsequently cooled down to room temperature. A d_{33} of 25 pm/V has been measured at a fundamental wavelength of 1.5

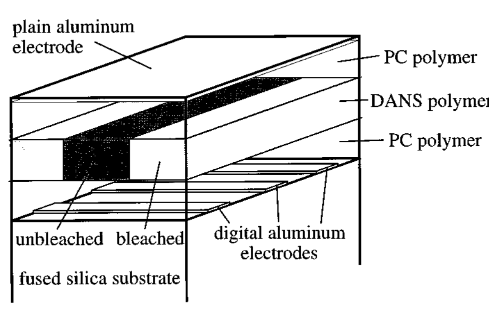

FIG. 1. Sample geometry with periodic electrode ($\Lambda = 14$ μm) on the substrate.

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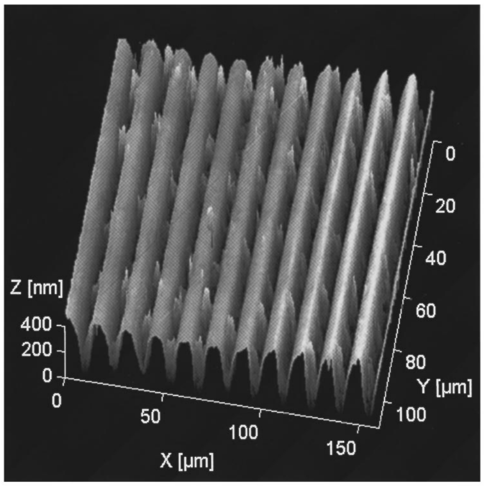

FIG. 2. Three-dimensional surface plot of the periodically poled sample with digital electrode on the multilayer stack surface. Maximum deformation amplitude is larger than 300 nm.

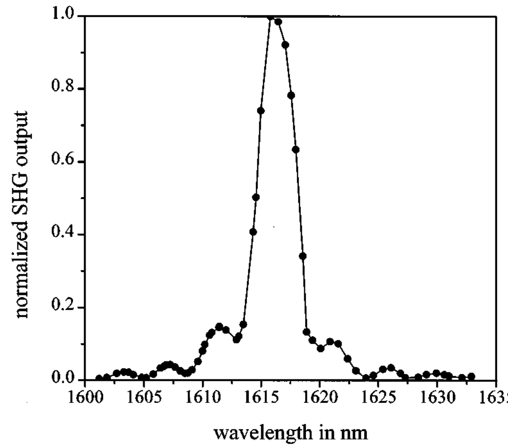

FIG. 3. Normalized second-harmonic output vs detuning. The acceptance bandwidth corresponds to a phase-matching length of 0.2 cm.

μm using the Maker fringe method for a poling field of 90 V/μm across planar (no periodicity) electrodes. The measurement was performed versus the reference of quartz (0.4 pm/V). Channel waveguides with widths from 1 to 5 μm were then photobleached by exposure to blue/UV light through a mask for 2 h. Finally, the samples were diced into pieces of several lengths for endfire coupling.

The waveguides were first characterized by evaluating their throughputs and deducing from such measurements the linear losses at the fundamental wavelength. Very large losses were measured for the photobleached grating sample. At 1.5 μm, the measured loss was 25 dB/cm, more than an order of magnitude larger than the absorptive loss in DANS. The origin was quickly identified as coherent scattering of the fundamental into the substrate via the resulting strong index modulation induced by the photobleaching. That is, the regions of different refractive index act like a phased array antenna in converting the guided wave into radiation fields. This was confirmed by beam propagation analysis through a periodic index structure with an index change of 0.04 between alternate waveguide regions. In addition, the photobleaching process changes the thickness of the polymer film. In order to measure these thickness changes, the samples were studied with a phase-shift interferometer, as reported in detail elsewhere.[9] The surface (not shown) exhibited a periodic deformation of about 80 nm or 1.3% of the total stack thickness.

The propagation losses for the two periodically poled geometries were radically different. For periodic electrodes on the substrate, waveguide losses were estimated to be a maximum of 5 dB/cm. However, a surprisingly large 40 dB/cm was measured for waveguides in which the periodic electrodes were deposited onto the multilayer stack.

In order to determine the origin of this excessive loss, the samples were also studied using phase shift interferometry. It was found that the polymer under the electrodes was squeezed when a strong field was applied, leading to a periodic thickness perturbation which scattered the light both coherently and incoherently out of the waveguide. Figure 2 shows a three-dimensional map of the thickness variations, where the imbedded regions correspond to the digital electrode grating. The maximum deformation amplitude was found to be more than 300 nm (or 5% of the polymer stack) in the poled waveguide device.

Systematic investigations with different grating periods exhibited an increase in the deformation amplitude for finer electrode gratings. For example, the deformation for a grating period of 18 μm was about 10%–20% smaller than that of a 14 μm grating. These results can be explained by the electrostatic forces during the poling and the resulting viscous flow of the polymer in the rubbery state. For large grating periods, a similar flow of the polymer is, as observed, only possible near the edges of the electrodes. It is these periodic deformations which led to the large propagation losses.

For samples with a plain electrode on top, the deformation of the polymer waveguide is, therefore, minimized. The surface (not shown) exhibits only a slight periodic deformation of about 20 nm or 0.3% of the stack thickness. Furthermore, it was observed that the regions that had been exposed to the UV lamp of the mask aligner during the photobleaching were reduced in thickness, causing the channels to stand out like small ridges. The effect is still small (about 0.5% of the stack thickness), and does not seem to influence the device performance in our case.

The SGH measurements were performed with a synchronously pumped NaCl:OH$^-$ color center laser, which produces 6–9 ps pulses with tunability from 1.50 to 1.65 μm. By tuning the wavelength, the second-harmonic power was investigated versus the detuning $\Delta\beta L_{\mathrm{SAMPLE}}$. For the periodically photobleached sample, the full width of 7 nm indicated an effective phase-matching length of $L_{\mathrm{PM}}=0.6$ mm $<L_{\mathrm{SAMPLE}}=2$ mm, in good agreement with the measured loss coefficient. Even for such small values of L_{PM}, $P(2\omega)/P^2(\omega)=5\times10^{-4}$%/W was measured. For the periodically poled sample with electrodes on the upper surface, the measured conversion efficiency was even less because of the larger propagation losses.

The best results were obtained with periodic electrodes deposited on the substrate surface. As shown in Fig. 3, the expected $\mathrm{sinc}^2(\Delta\beta L_{\mathrm{PM}})$ dependency is observed. From the phase-matching curve and the waveguide parameters, the length L_{PM} over which phase matching is maintained was determined to be 0.2 cm, which is very close to the sample length of $L=0.25$ cm. The figure of merit for this sample is

$$\eta=\frac{P_{2\omega}}{P_{\omega}^{2}L^{2}}=0.05\%/\text{W-cm}^{2}.$$

Although this efficiency, to the best of our knowledge, is the highest reported for a polymeric channel waveguide, it is still very modest compared to that expected from the high off-resonant d_{33} coefficient of 25 pm/V. As discussed previously by Khanarian and co-workers, the spatial modulation of d_{33}, i.e., $\Delta d_{33}(z)$ is small with respect to its average value in this poling geometry.[10] In order to utilize the high nonlinearity of polymers, a better geometry, or poling procedure and/or material has to be found that allows complete modulation of the nonlinearity.

In conclusion, we have investigated three QPM-SHG geometries for fundamental beams at 1.5 μm. It was found that QPM under certain conditions led to large scattering losses. For example, the large radiative losses found for the index modulation which accompanies photobleaching made this approach unattractive. Periodic film deformation via poling electrodes on the free film surface also led to large radiative losses and proved unsuitable. These deleterious effects were minimized by changing the waveguide design, i.e., using periodic poling electrodes on the substrate surface, leading to a figure of merit $\eta=0.05\%/\text{W-cm}^2$ for SHG with a phase-matching length of 0.2 cm. A large improvement of the SHG conversion efficiency is still to be expected for an improved poling geometry that allows a better modulation depth of the d_{33} coefficient.

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