1-1-1993

2Nd-Harmonic Generation By Counter Propagating Beams In 4-Dimethylamino-4'-Nitrostilbene Side-Chain Polymer Channel Wave-Guides

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Cite as: Appl. Phys. Lett. **63**, 3405 (1993); https://doi.org/10.1063/1.110155
Submitted: 21 June 1993 . Accepted: 05 October 1993 . Published Online: 04 June 1998

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Second harmonic generation by counter propagating beams in 4-dimethylamino-4′-nitrostilbene side-chain polymer channel waveguides

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(Received 21 June 1993; accepted for publication 5 October 1993)

We demonstrate surface emitted second harmonic generation due to mixing of counter propagating waves in in-plane poled, low loss, channel waveguides of 4-dimethylamino-4′-nitrostilbene (DANS) side-chain polymers. Single film DANS side-chain polymer waveguides yield high conversion efficiency devices.

The mixing of counter propagating guided waves to generate second harmonic (SHG) normal to the surface was initially demonstrated in Ti in-diffused $LiNbO_3$ waveguides over a decade ago.[1] Several applications were also demonstrated or proposed, including picosecond signal processing,[2] picosecond optical transient digitizers,[3] and spectrometers.[4] The device potential of this phenomenon for ultrafast signal processing, wavelength division demultiplexing, etc., has attracted attention recently.[5] In the last few years, several works in GaAs based waveguides were reported,[5–10] and the conversion efficiency was improved dramatically by using a form of quasi-phase matching in multilayered structures. On the other hand, organic polymers have also been shown to have great potential for second-order nonlinear optical applications because they have large nonresonant nonlinearities and ease of fabrication. In this letter we report the first observation of this effect in polymer waveguides made from a single film of 4-dimethylamino-4′-nitrostilbene (DANS) side-chain polymer. The measured nonlinear cross-section coefficient is comparable to that for single film GaAs waveguides, in good agreement with our calculations.

DANS side-chain polymers are one of the most highly developed poled polymers for applications to electro-optical switching devices. Their relatively high glass transition temperature provides long-term stability of the poled state at room temperature and large poling field strengths provide nonlinear coefficients d as high as 200 pm/V at a resonant wavelength ($\lambda = 1.064$ μm).[11] Furthermore, structures with lateral confinement can be made by photobleaching[12] which allowed us to fabricate low loss channel waveguides.[13]

The device efficiency for counter propagating mixing SHG is given by the nonlinear cross section A^{NL} which is defined as[1,14]

$$I(2\omega) = A^{NL} I_+(\omega) I_-(\omega), \tag{1}$$

where $I_\pm(\omega)$ is the line intensity in W/m of the guided waves at the fundamental frequency, i.e., the power per unit width of the guided wave wave front. Also, $I(2\omega)$ is the intensity of the second harmonic light generated normal to the waveguide, in W/m^2. Analysis of the field generated by the nonlinear polarization shows that the nonlinear cross section is strongly dependent on the superposition integral $S(\infty)$ between the guided mode profile of the fundamental beams and the generated second harmonic field:

$$A^{NL} = \epsilon_0 \omega k_0(2\omega) n_c(2\omega) |C|^4 |S(\infty)|^2, \tag{2}$$

with

$$S(\infty) = \int_{-\infty}^{\infty} \frac{d_{22}(x') f_y^2(x')}{n_f(2\omega)} \times \exp[ik_0(2\omega) n_f(2\omega) x'] dx', \tag{3}$$

where $k_0(2\omega) = 2\omega/c$, and $n_c(2\omega)$ is the refractive index of the cover and $n_f(2\omega)$ is the complex refractive index of the film. Furthermore, $f_y(x')$ is the guided mode transverse distribution and C is the normalization constant of the guided mode for the fundamental beam.[4] Obviously, the properties of the nonlinear cross section are directly related to the properties of the superposition integral which needs to be optimized.

We first examine numerically the trade-offs in $S(\infty)$ for the DANS waveguide parameters. The magnitude of the overlap integral oscillates with increasing film thickness t. Since the guided mode profile $f_y(x')$ is broadened with increasing thickness and the refractive index $n_f(2\omega)$ includes absorption, the envelope of this oscillation decreases with increasing thickness when normalized to input power. The important tendencies are clearly shown in Fig. 1 based on typical DANS parameters [$n_f(\omega) = 1.63$, $n_f(2\omega) = 1.8$]. Because the peak of $f_y(x')$ moves towards the cover-film interface when $n_c(\omega)$ is large, approaching the value of $n_f(\omega)$, the influence of $n_c(\omega)$ on A^{NL} [Fig. 1(a)] indirectly demonstrates the influence of the guided mode profile $f_y(x')$. The three curves correspond to the first three smallest film thicknesses for which the oscillating A^{NL} is maximum. The first and largest peak value is obtained for $n_c(\omega) = 1.49$. The corresponding $n_f(2\omega)$ dependence is evaluated with an air cladding and a fused

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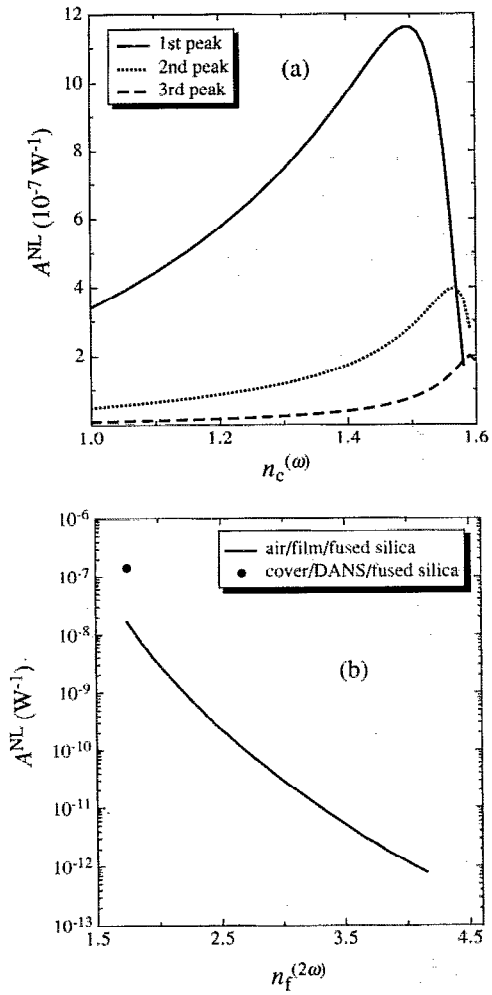

FIG. 1. Variation of A^{NL} with refractive index; (a) The dependence on the refractive index of the cladding at $\lambda=1064$ nm with the DANS polymer film and the fused silica substrate. (b) The dependence on the refractive index of the film at $\lambda=532$ nm.

silica substrate [Fig. 1(b)]. The nonlinear cross section A^{NL} at the film thickness of 1 μm is estimated from the envelope of the oscillating decay for comparison. Since the oscillating period is inversely proportional to $n_f(2\omega)$, the integral within each period dramatically decreases for large refractive indices of the film. In this experiment, the cover polymer which has $n_c(\omega)=1.55$ was used and Fig. 1(b) indicates large potential of this device.

A schematic of the cross section of the fabricated DANS polymer waveguide is shown in Fig. 2. DANS side-chain polymer films were made by spin coating onto fused silica substrates which have two coupling gratings, one at each end, and poling electrodes on the glass surface. The period of the grating was 0.5 μm and the spacing of the poling electrodes was 20 μm. The film thickness of 0.83 $\pm$0.04 μm was determined from calibrated data on the spinning rate and the solid concentration in the spinning solution. A 4-μm-wide channel waveguide with parabolic tapered mode couplers on both sides[13] was constructed by photobleaching used to reduce the film index outside the channel region. In this process the film is exposed through a mask to collimated light from a Hg-arc lamp ($\lambda=436$

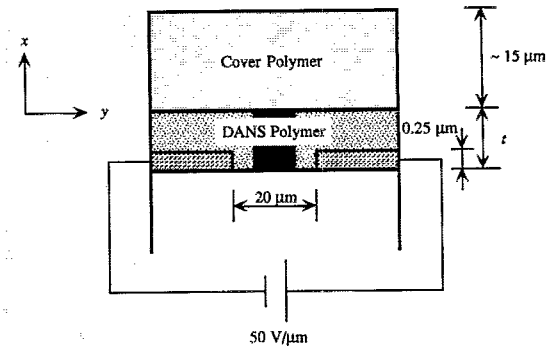

FIG. 2. Schematic cross section of the waveguide.

nm) for 4 h. The effective device length was 1 cm. A 15-μm-thick cover layer was spin coated on top of the DANS polymer in order to prevent discharge through the air during poling and to create a better guided mode profile for efficient surface emitting SHG. Since a large lateral component (in-plane, along the y axis in Fig. 2) of the second-order nonlinearity is required for efficient conversion, plane parallel poling was performed at just below the glass transition temperature of $T_g=142$ °C. Although the applied electric field strength was only 50 V/μm for the sample investigated for SHG, the highest field which can be reached was determined to be as large as 200 V/μm. The nonlinear coefficient d_{22} was measured at 1.064 μm (on resonance) after poling with 50 V/μm and a value of 26 pm/V was determined. This value is lower by a factor of 2 compared to the value derived from the measurement of the electro-optical coefficient for poling with the same value of field normal to film.[11] This is reasonable because the field lines in our geometry are curved, resulting in field being used to align molecules normal as well as parallel to the surface.

The surface emitted SHG power was implemented by coupling 100-ps Q-switched mode locked pulses at 1064 nm into both sides of the waveguide. A pulse was split into two identical halves and the overlap of these two pulses in the waveguide was optimized by changing the optical path length of one arm. The coupling efficiency of the grating couplers was determined by measuring the transmission change and the total coupling efficiency into the channel waveguide was estimated as 10% for each of the two couplers, including the tapered mode transition loss.[13] The SHG power was detected in the direction normal to the surfaces by a photomultiplier tube through a lens. Since the generated light has a large divergence angle along the y axis, the power collected by the lens was only 30% of the total generated power, including reflection losses. The measured SHG power was 17 mW for a 32 W input power which yields 0.64 GW/cm^2 of peak intensity in the channel waveguide. The nonlinear cross section was evaluated as 8.5×10^{-9} W^{-1} by assuming a Gaussian shape for the pulses in the pulse train. This value is larger than that for a single layer GaAs film and the generated second harmonic light was easily seen by eye under room light.[5]

The thickness dependence of the magnitude of the non-

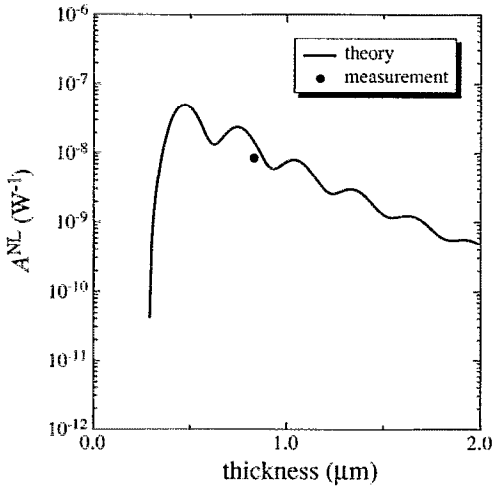

FIG. 3. Thickness dependence of the nonlinear cross section for DANS side-chain polymer waveguides with a cladding refractive index of 1.55 and a fused silica substrate.

linear cross section was calculated for the DANS side-chain polymer and is shown in Fig. 3. The measured value of the nonlinear coefficient d_{22} for the 50 V/μm poling field was used, and absorption and multiple reflection were included in the calculation. The calculated value 1.45×10^{-8} W^{-1} at a thickness of 0.83 μm agrees with the measured value. The maximum efficiency would be obtained at a film thickness of 0.47 μm with $A^{NL}=5.0\times10^{-8}$ W^{-1}. A simple extrapolation to our largest poling voltages to data of 200 V/μm gives 8×10^{-7} W^{-1}, the comparable to that of a multiple layer GaAs film.[5] Such an efficient single layer device should make it possible to operate over a large frequency range, and to demonstrate new applications such as spectrometers, etc. In principle, extending the device to multiple layer films with alternate films of zero activity should lead to even larger efficiencies.

In summary, counter propagating mixing SHG was demonstrated with polymer channel waveguides for the first time. High efficiency was observed in the poled DANS side-chain polymer. The reasonable agreement observed between the experimental values and theory supports the promise for even higher efficiencies with optimized devices.

The authors acknowledge the support of the Air Force Office of Scientific Research (AFOSR-91-0339). We also thank J. Ross for preparing the coupling gratings and the poling electrodes.

[1] R. Normandin and G. I. Stegeman, Opt. Lett. **4**, 58 (1979).
[2] R. Normandin and G. I. Stegeman, Appl. Phys. Lett. **36**, 253 (1980).
[3] R. Normandin and G. I. Stegeman, Appl. Phys. Lett. **40**, 759 (1982).
[4] G. I. Stegeman, J. J. Burke, and C. T. Seaton, in *Optical Engineering: Integrated Optical Circuits and Components*, edited by L. D. Hutcheson (Dekker, New York, 1987), Vol. 13.
[5] R. Normandin, S. Letourneau, F. Chatenoud, and R. L. Williams, J. Quantum. Electron **QE-27**, 1520 (1991); H. Dai, S. Janz, R. Normandin, R. L. Williams, and M. Dion, Electron. Lett. **28**, 1540 (1992).
[6] D. Vakhshoori and S. Wang, Appl. Phys. Lett. **53**, 347 (1988).
[7] D. Vakhshoori, J. Walker, S. Dijaili, S. Wang, and J. S. Smith, Appl. Phys. Lett. **55**, 1164 (1989).
[8] R. Normandin, R. L. Williams, and F. Chatenoud, Electron. Lett. **26**, 2088 (1990).
[9] D. Vakhshoori, R. J. Fischer, M. Hong, D. L. Sivco, G. J. Zydzik, G. N. S. Chu, and A. Y. Cho, Appl. Phys. Lett. **59**, 896 (1991).
[10] R. Normandin, H. Dai, S. Janz, A. Delage, J. Brown, and F. Chatenoud, Appl. Phys. Lett. **62**, 118 (1993).
[11] W. H. G. Horsthuis, G. R. Möhlmann, and H. W. Mertens, *Digest of the Conference on Nonlinear Guided-Wave Phenomena* (OSA, Cambridge, England, 1991), paper MD1, pp. 70–73.
[12] M. D. J. Diemeer, F. M. M. Suyten, E. S. Trommel, A. McDonach, J. M. Copeland, L. W. Jenneskens, and W. H. G. Horsthuis, Electron. Lett. **26**, 379 (1990).
[13] A. Otomo, S. M.-Neher, G. I. Stegeman, W. H. G. Horsthuis, and G. R. Möhlmann, Electron. Lett. **29**, 129 (1993).
[14] P. J. Vella, R. Normandin, and G. I. Stegeman, Appl. Phys. Lett. **38**, 759 (1981).