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3Rd-Order Nonlinearity Of 4-Dialkylamino-4'Nitro-Stilbene Wave-Guides At 1319 Nm

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The intensity dependent optical properties of 4-dialkylamino-4′nitro-stilbene polymer channel waveguides were measured at 1319 nm with a pulse modulated Mach–Zehnder interferometer to be $n_2=0.8\times10^{-13}$ cm^2/W and $\beta_2<0.08$ cm/GW. This material is promising for all-optical switching at 1319 nm because it satisfies both the W and T figures of merit.

DANS (4-dialkylamino-4′nitro-stilbene) moieties in the form of side chain polymers have proven very attractive for nonlinear optics.[1,2] For example, the third order nonlinearities are of interest for all-optical switching.[2,3] It has already been shown that the nonlinear coefficient n_2 (defined by $n=n_0+n_2I$) is comparable to that of soluble polydiacetylenes like poly-4BCMU.[2,4] The key question is whether the two switching figures of merit (FOM) $W>1$ and $T<1$ are satisfied at a communications relevant wavelength such as 1319 nm.[3] These FOM, defined in detail later, quantify whether a 2π nonlinear phase shift is achievable over one linear (W) or nonlinear (T) absorption length. It has in fact proven difficult to find materials which do satisfy these FOM. Here we report measurements of DANS nonlinearities which show that this material is promising for all-optical phenomena at 1319 nm.

The DANS molecule was first developed for its high second order nonlinearity. When poled in channel waveguides, efficient electro-optical switching devices have already been demonstrated at 1319 nm.[1] The channel waveguides were fabricated by photobleaching, i.e., by supplying photons of sufficient energy to induce *cis-trans* isomerization which leads to reduced nonlinear activity and refractive index in the bleached region.[5] The desired guiding region of high index (channel) is blocked from the photobleaching light. Another method of supplying energy to the *cis-trans* isomerization process is via the simultaneous absorption of two or more photons. If there is a significant two photon coefficient at the operating wavelength, usually 1319 nm, then the device operating characteristics will degrade in time. Here, we report an upper limit to the two photon absorption coefficient at 1319 nm.

Our experiments were performed on 1.65 cm long single mode channel waveguides formed by photobleaching spin-coated DANS. The polymer, composed of 35% side chain DANS and 65% of backbone polymer, was spin-coated onto oxidized silicon substrates to form a 1.8 μm thick film. Channel waveguides were formed by exposing the film to UV light through a mask in which only the channel region is blocked. The UV bleached part of DANS film has $n=1.567$ while the unbleached part of DANS has a refractive index 1.601. These values were established by calibration measurements on thin films. The estimated depth (based on a series of calibrations of films of different thickness) to which the DANS is bleached is 0.7 μm after 6 h of UV light exposure. These material parameters, along with the mask width, were used in computer program for calculating waveguide modes. We used the Fourier analysis method for solving the scalar wave equation, subject to the usual boundary conditions.[6] This method shows that the 3 μm width and 1.8 μm thick DANS film supports only a single mode at the wavelength 1319 nm. The calculated effective area, defined by

$$A_{\text{eff}}=\frac{[\int\int|E(x,y)|^2dx\,dy]^2}{\int\int|E(x,y)|^4dx\,dy}, \tag{1}$$

where $E(x,y)$ is the field distribution, is 8.0 μm^2 with an effective waveguide index $N_{\text{eff}}=1.570$.

The nonlinear refractive index was measured by a pulse modulated Mach–Zehnder interferometer as shown in Fig. 1. The prototype version of this interferometer has been described in detail elsewhere.[4] A mode-locked Nd:YAG laser operating at 1319 nm provided 90 ps full-

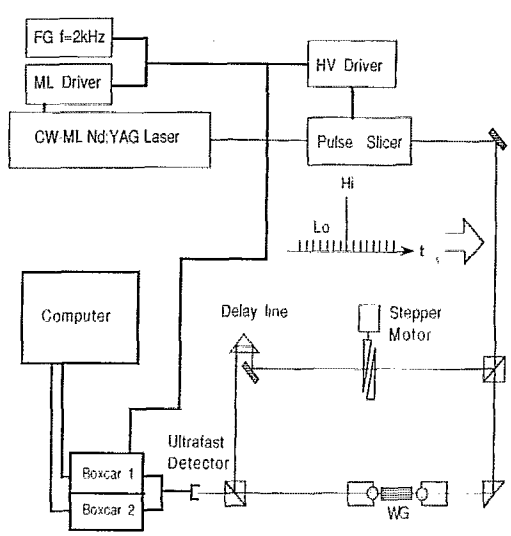

FIG. 1. Experimental arrangement of pulse modulated Mach–Zehnder interferometer for direct measurement of n_2.

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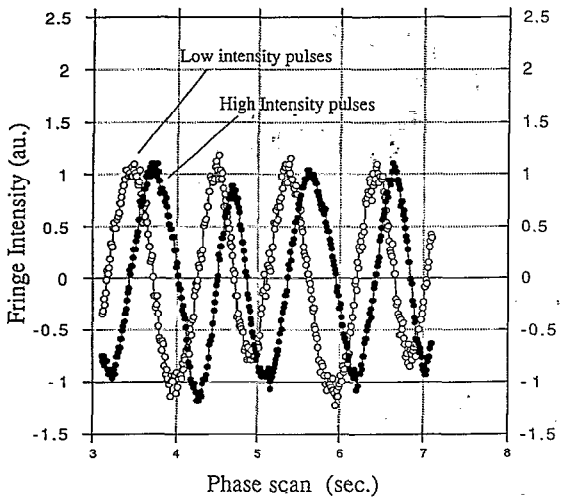

FIG. 2. Measured fringe scan for both high and low intensity pulses. The measured phase difference is 0.5π rad.

width at half-maximum (FWHM) pulses at a 76 MHz repetition rate. An electro-optic pulse slicer which was synchronized to the mode locker was used to select one high intensity pulse every 0.5 ms and to attenuate the rest of the mode-locked pulses. The DANS waveguide sample was put in one arm of the interferometer. We put a delay line in the reference arm to make the path length of the two arms equal. Because of the sample nonlinearity, the high intensity pulses transmitted through the waveguide propagate through a different optical path length. This will give a phase shifted fringe pattern for the high intensity pulses compared to the low intensity pulses. Two matched detectors and boxcar gated averagers were used to sample these two fringe patterns which were scanned at 1 Hz by pushing a wedged plate into the beam path of a reference arm. We were able to scan more than 100 fringes without any detectable fringe amplitude change. It should be noted that this interferometer becomes an all optical Mach–Zehnder switching device by itself if the two beams from both arms are well aligned with each other. We tested it without a sample and obtained more than 95% modulation by pushing the wedge plate into the beam path. In order to avoid any slow thermal refractive index changes, the 3 ns gate of the boxcar used for the low intensity pulses was opened to catch the interference pattern for the low intensity pulse immediately preceding the selected high intensity pulse.

We took two sets of fringe scan data, shifted by $\Delta\phi^{NL}$, as shown at Fig. 2 for the high and low intensity pulses. Converting these sine wave sets into a data set of $\Delta\phi^{NL}$ is not trivial because there are phase and amplitude fluctuations in the data due to laser noise and vibrations of optical components in the interferometer. As discussed in Ref. 4, to maximize the information we did Hilbert transformations instead of Fourier transformations on each fringe pattern.[4] Then, the phases were compared point by point between the high and low intensity data sets. The standard deviation of the 300 phase differences which constituted a data set were used to estimate the measurement error, less than $\pi/50$ for most cases.

The nonlinear phase change experienced by the waveguide modes is a function of the optical beam intensity which varies over the waveguide cross section and pulse duration. The spatial averaging over the waveguide field profile can be taken into account by using the effective area so that $\Delta n_{\text{peak}} = n_2 P(0)/A_{\text{eff}}$, where $P(0)$ is the peak input power just inside the waveguide. The measured phase shift $\Delta\phi^{NL}$ is already averaged for a given temporal pulse shape when we take data with a gated averager which has much wider gate width than the optical pulse width. We modeled in detail the temporal pulse shape dependence of the interferometer output to simulate the relationship between the measured phase shift and the nonlinear phase shift at the peak intensity of a Gaussian pulse. This modeling shows that $\Delta\phi^{NL}_{\text{avg}} = 0.753\Delta\phi^{NL}_{\text{peak}}$ within 2% error for phase shifts up to several π in $\Delta\phi^{NL}_{\text{peak}}$.

The linear loss due to scattering along the waveguide was measured to be 1.7 dB/cm (0.4 cm^{-1}) with an IR camera and frame grabber. The nonlinear phase shift accumulated over a length L with loss α present is given by

$$\Delta\phi^{NL}_{\text{peak}} = \frac{2\pi}{\lambda_{\text{vac}}} \Delta n_{\text{peak}} L_{\text{eff}}, \quad L_{\text{eff}} = \int_0^L e^{-\alpha z}\, dz.$$

Here, L_{eff} is 1.21 cm. The maximum peak power difference of 39.1 W between the high and low intensity pulses gave a 0.5π phase shift, shown in Fig. 2. This gives $\Delta\phi^{NL}_{\text{peak}} = 0.66\pi$, which in turn gives $\Delta n_{\text{peak}} = 3.6 \times 10^{-5}$. Using $P(0) = 0.5\epsilon_0 N_{\text{eff}} c \int\int |E(x,y)|^2 dx\, dy$ with $P(0) = 39 \pm 5$ W gives $n_2 = (0.8 \pm 0.2) \times 10^{-13}$ cm^2/W for the polymer with 35% DANS doping. The main source of the uncertainty comes from estimating the intensity inside the waveguide.

The nonlinear absorption coefficient β_2 was measured with small modifications to the experimental setup shown in Fig. 1. The interferometer reference arm is replaced by a reference channel for monitoring the input intensity. Since some hysteresis in transmission was observed when the intensity of the cw mode-locked (cw-ML) pulse train was changed with a half-wave plate and a polarizer, we used the pulse slicer again to avoid coupling efficiency changes due to the thermal effects. One high intensity pulse was selected and modulated for every 152 000 low intensity pulses so that the average power through the waveguide was changed very little even though the intensity of the high intensity pulses were changed from the minimum to the maximum. In the presence of linear loss, $I(L) = I(0)/[1 + \beta_2 I(0)\ L_{\text{eff}}]$.[4] When there is no linear absorption the relation between the input intensity and the output intensity is given by $I(L) = I(0)/[1 + \beta_2 L I(0)]$, where $L = 1.65$ cm in our case. We could not observe any change in the throughput with cw-ML laser operation which gave a maximum intensity of 0.28 GW/cm^2 inside the waveguide. Next we used QS-ML (Q-switched mode-locked) operation which gives about a factor of 80 more peak power. With a pulse slicer we selected one of the peak pulses at the center of each QS-ML pulse train. When the intensity of these selected pulses was increased, we destroyed several waveguides at intensities > 1.1 GW/cm^2, about one-tenth

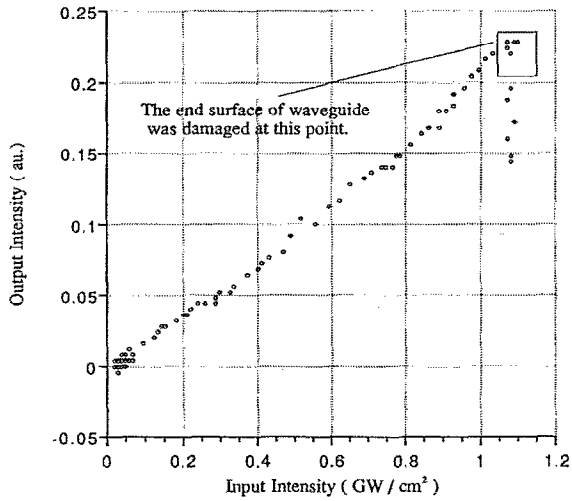

FIG. 3. Measured nonlinear transmission change with QS-ML operation of the Nd:YAG laser.

of the maximum intensity we could in principle get inside of our waveguide. Typical data are shown in Fig. 3. However, no nonlinear absorption was observed up to the damage threshold intensity. This result gives the upper bound of $\beta_2 < 0.08$ cm/GW. Note that our results also give an upper limit to the three photon absorption coefficient $\beta_3 < 0.34$ cm^3/GW^2 (in $\alpha = \beta_3 I^2$). We observed no degradation of the sample due to GW/cm^2 pulses over the duration of the experiments (~ 10 h). The damage observed originated at the cleaved input facets which could probably be improved by better preparation techniques. Our previous experience is that the damage threshold for DANS films at normal incidence is in excess of 5 GW/cm^2.

We can now evaluate the two nonlinear figures of merit W and T. In order to have reasonable throughput as well as switching, we need $W = \Delta n/\alpha\lambda > 1$.[7] For the experimental parameters at the input surface damage threshold intensity, $W > 1.6$. However, with better input facets so that the material and not end-face damage threshold can be assumed, W increases by an order of magnitude. Even without linear loss, two photon absorption can reduce the device throughput and spoil switching. We need the $T = 2\beta_2\lambda/n_2$ parameter to be smaller than one.[8] We found $0.26 > T$. Therefore, both FOM are easily satisfied, making this material a prime candidate for all-optical switching. An intensity of 3.5 GW/cm^2 and a peak power of 140 W would be required to switch a nonlinear directional coupler, comparable to the best results recently implemented in AlGaAs waveguides below half their band gap.[9]

In summary, we have measured the third order nonlinearities of the DANS side-chain polymer at 1319 nm. The figures of merit obtained for all-optical switching are very promising. Finally, an upper bound of 0.08 cm/GW was found for the two photon absorption coefficient.

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