1-1-1994

Assessment Of Single-Crystal P-Toluene Sulfonate As An All-Optical Switching Material At 1.3-Mu-M

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Cite as: Appl. Phys. Lett. **65**, 1742 (1994); https://doi.org/10.1063/1.112902
Submitted: 10 May 1994 . Accepted: 27 July 1994 . Published Online: 04 June 1998

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(Received 10 May 1994; accepted for publication 27 July 1994)

We evaluate the potential of single crystal PTS (*p*-toluene sulfonate) for all-optical applications at the 1.32 μm communications wavelength by measuring the electronic and thermo-optic nonlinearities, and the multiphoton absorption with picosecond pulses. With appropriate heat sinking, duty cycles greater than 25% should be possible without significant cumulative thermal effects for 1 W operating powers. © *1994 American Institute of Physics.*

Materials must satisfy a series of figures of merit to be useful for ultrafast all-optical switching and related applications.[1] First, they must exhibit a large, nonresonant, optical Kerr-like response ($n_2 > 10^{-12}$ cm^2/W) for the intensity-dependent refractive index in order to operate at watt peak powers in centimeter long devices. Second, a nonlinear phase shift of at least π should be possible over $1/e$ attenuation distances for reasonable device throughput. This requirement is embodied in two figures of merit $W = n_2 I_{SW}/\alpha_1 \lambda > 1$ and $T = \alpha_2 \lambda / n_2 < 1$ (for 2π phase shift), where α_1 and α_2 are the one and two photon absorption coefficients respectively, and I_{SW} is the intensity required for a 2π phase shift. Finally, absorption leads to index changes via the thermo-optic effect which accumulate over many pulses and compete with the all-optical index changes. Assuming that N pulses, Δt in width, are required for a cumulative thermal index change equal to 10% of the electronic index change, then the maximum allowed duty cycle is $D = 10N\ \Delta t/\tau$% for any pulse width where τ is the thermal relaxation time of the device structure.[2] We have recently shown that the one-dimensional conjugated polymer PTS, poly[2,4-hexdiyne-1,6-diol-bis-*p*-toluene-sulfonate], shows a record high, nonresonant, nonlinear response of 2×10^{-3} cm^2/GW at 1.6 μm and satisfies the T parameter as well.[3] Here we show that PTS in the other important telecommunication window around 1.3 μm should satisfy all of the above stated requirements.

PTS has already been grown in an integrated waveguide format with low losses in the near infrared.[4] Here we concentrate on predevice linear and nonlinear measurements in a bulk single crystal approximately 210 μm thick. Linear transmission was limited by Fresnel losses at the surfaces of the crystal, and exceeded 80% for radiation polarized along the conjugation axis. Nonlinear losses were measured with a versatile parametric laser system with the Z-scan technique. An example of our experimental results for both the open and closed aperture Z-scan is shown in Fig. 1 for input pulses at 1.30 μm. As a result of the fits, also shown in Fig. 1, a two-photon absorption coefficient of 20 ± 4 cm/GW (α_2) is obtained. In addition, a positive intensity dependent refractive index n_2 of $3.2 \pm 0.3 \times 10^{-3}$ cm^2/GW is measured from the closed aperture data. The latter result was reconfirmed with a hybrid Mach–Zehnder interferometer, which has been developed to measure nonlinearly induced phase shifts as small as $\pi/100$.[5] In this case, just as previously observed for the measurements at 1.6 μm, a weak linear dependence (Fig. 2) of both nonlinear coefficients on input intensity was observed, suggesting the presence of higher order effects at input peak intensities used which ranged between 0.5 and 7 GW/cm^2, i.e., $\Delta n = n_2 I + n_3 I^2$ and $\Delta\alpha = \alpha_2 I + \alpha_3 I^2$. From the measured variations versus intensity we deduced the next higher order loss coefficient and intensity dependent refractive index to be respectively $\alpha_3 = 5.2 \pm 0.3$ cm^3/GW^2 and $n_3 = -1.5\times10^{-4}$ cm^4/GW^2. Similar effects have been reported at other wavelengths previously in polydiacetylenes.[6,7] These higher order effects are negligible for typical device switching intensities ($\simeq \lambda/n_2 L \simeq 20$ MW/cm^2 for $L = 2$ cm and a 2π phase shift). We directly evaluate a value of $T = 0.75$. Based on linear loss measurements reported by Thakur *et al.* and our experimental estimate of 10 GW/cm^2 for the damage threshold intensity, we estimate that $W > 100$.[4] Therefore both loss-related figures of merit are acceptible for PTS at this wavelength.

An important issue which is frequently overlooked in assessing materials is the possibility that cumulative (over many pulses), large, optically induced index changes can exceed the instantaneous, Kerr-effect index changes at high data rates. This effect is quantified by the duty cycle defined previously. For example, in the case of semiconductor waveguides, Gabriel *et al.* demonstrated that both free carrier absorption and thermo-optic phase drifts can play an important role.[2] Here we employed a nonlinear Mach–Zehnder interferometer developed to measure both the instantaneous (single pulse) refractive nonlinearity and the accumulated thermal phase shift generated by a 76 MHz train of 100 picosecond pulses. Figure 3 shows the phase shift induced by

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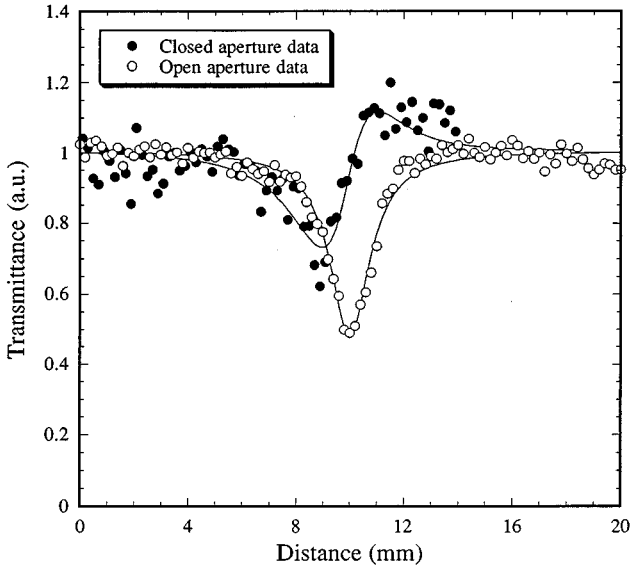

FIG. 1. Typical Z-scan data at 1.3 μm. The open circles show the open aperture Z-scan showing the presence of two-photon absorption with pulses of 5.66 GW/cm^2. The filled circles show the result of a closed aperture Z-scan at 0.8 GW/cm^2.

2000 pulses of varying intensity. The induced phase shift varies quadratically with input intensity, implying a two photon absorption origin for the thermal phase shift. At the highest intensity used, 170 MW/cm^2, the two photon induced absorption is 3 cm^{-1}, much larger than the typical linear loss

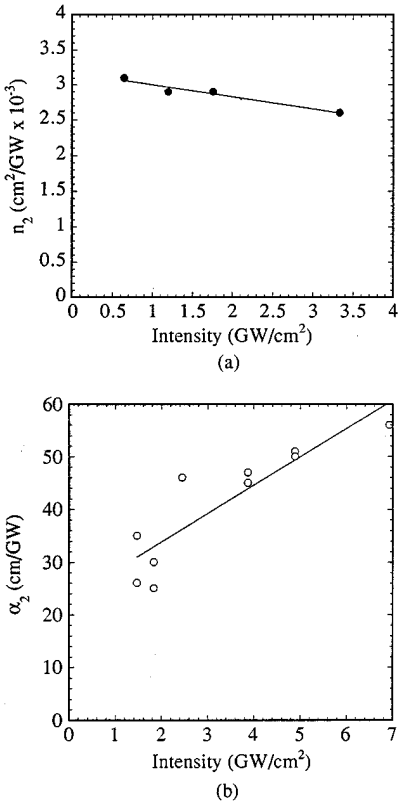

FIG. 2. Variation in the effective n_2 (a), and α_2 (b), with peak input intensity.

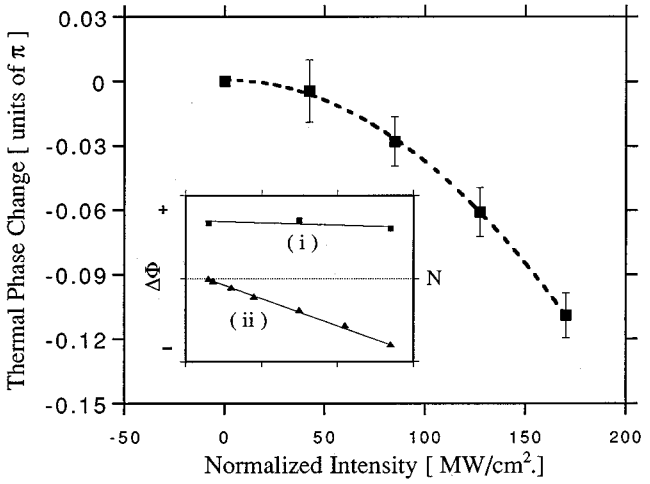

FIG. 3. Accumulated thermal nonlinear phase change for a PTS sample for 2000, 100 ps pulses as a function of the input intensity. Shown in the inset is a schematic of the nonlinear phase shift with number of pulses for both the electronic and thermal nonlinearities.

of 0.2–0.3 cm^{-1}. We therefore conclude that two-photon absorption is the dominant loss mechanism for $I>15$ MW/cm^2.

The maximum allowed duty cycle is estimated from the interferometrically measured phase shifts. A single pulse measurement of the electronic nonlinearity with 170 MW/cm^2 input intensity showed a positive phase shift of $\pi/5$ in our 210 μm platelet. (This corresponds to $n_2=4\times10^{-12}$ cm^2/W, in good agreement with the Z-scan measurement obtained at intensities one order of magnitude larger.) Because the phase shift is linear in intensity, this would correspond to a phase shift of $\pi/5$ [20/170]$=\pi/42.5$ at the typical switching intensity of 20 MW/cm^2 for a 2-cm-long device. It required 3000, 170 MW/cm^2, pulses to shift the fringes by-$\pi/5$ via the thermal process. Because this is a two photon activated process, $N=2.55\times10^4$ pulses will be required to produce a phase shift of $\pi/42.5$ at a 20 MW/cm^2 switching intensity. Furthermore, with a well heat-sunk environment to dissipate the heat, for example, a sapphire substrate for a waveguide, a thermal decay time of 1 μs is a conservative upper limit. This implies a 25% duty cycle, i.e., switching of pulses separated by three pulse widths is feasible at the stated intensity levels with minimum cross talk due to thermal effects.

In summary, our measurements show that single crystal PTS satisfies all of the important figures of merit for ultrafast all-optical devices at 1.3 μm. its nonlinearity is orders of magnitude larger than that of other materials used in the communications spectral windows which also satisfy the same figures of merit, for example, silica and chalcogenide glasses in fiber form, or AlGaAs.[7–9] For example, a 2-cm-long PTS device with a 2 μm guiding core would need a source of approximately 1 W peak (0.25 W average, 25% duty cycle) power. Such sources should become available soon at 1.3 μm, making nonlinear devices viable and compatible with current state of the art fiber technology. Finally, we note that the loss figures of merit are also very good at 1.60 μm, allowing even larger duty cycles at that wavelength.[3]

The research at CREOL was supported by AFOSR (91-0339).

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