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Measurement of the complex nonlinear refractive index of single crystal *p*-toluene sulfonate at 1064 nm

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Z-scan at 1064 nm was used with single, 35 ps pulses to measure the nonlinear refraction and absorption in single crystal PTS (*p*-toluene sulfonate). Detailed analysis of the *Z*-scan data based on $\Delta n = n_2 I + n_3 I^2$ and $\Delta \alpha = \alpha_2 I + \alpha_3 I^2$ yielded $n_2 = 5(\pm 1) \times 10^{-12}$ cm²/W, $\alpha_2 = 100(\pm 20)$ cm/GW, $n_3 = -5(\pm 1) \times 10^{-21}$ cm⁴/W² and $\alpha_3 = -5(\pm 1)$ cm³/GW.² The resulting two photon figure of merit *T* for PTS is marginal for high throughput, all-optical waveguide switching at 1064 nm.

Since the first measurements of large nonlinear optical coefficients in π -electron conjugated polymers, such materials have been studied for all-optical applications.¹⁻⁶ Soluble polydiacetylenes such as poly-4BCMUs are attractive because low loss (1 db/cm) integrated optical devices can be fabricated by standard coating techniques.⁷ However, the nonlinear refractive index coefficients n_2 reported in the transparency region ($n_2 \sim 10^{-13}$ cm²/W) are only a few hundred times those of fused silica, and two-photon induced losses, specifically at 1064 nm, are large.^{5,6} (Here n_2 is defined by $n = n_0 + n_2 I$ where n is the refractive index and I the intensity.) However, for single crystal PTS (*p*-toluene sulfonate), a large range of very large values of n_2 have been reported at 1064 nm, specifically 3×10^{-11} cm²/W,⁸ 5.5×10^{-12} cm²/W,⁹ and 1.1×10^{-12} cm²/W.¹⁰ Although technically difficult, the fabrication of channel waveguides has been demonstrated.⁸ Such large coefficients, if accompanied by low linear and nonlinear losses, could make this material useful for all optical applications. Although the linear loss has been demonstrated to be small, a range of two-photon absorption coefficients α_2 (50–120 cm/GW) has also been reported.¹⁰⁻¹² As a result, the two-photon figure of merit, $T = 2\alpha_2 \lambda / n_2$, which needs to be less than unity for useful applications varies from 0.3 to 22. In this letter we report single-pulse, picosecond, *Z*-scan measurements on single crystal PTS which allow us to sort out the large uncertainty in the reported n_2 , α_2 , and *T* values, and hence to assess the potential of PTS for switching devices.

For the *Z*-scan measurement, we used a mode locked and *Q*-switched Nd:YAG laser operating at a 10 Hz with 35 ps pulses.¹³ The laser output was first spatially filtered in air through a 25- μ m-diam pinhole and then collimated, with special care taken to avoid introducing additional aberrations to the phase front. A 10 cm focal length lens focused 0.2 to 2 μ J of the 1064 nm radiation into a 20 μ m spot on a single crystal PTS sample approximately 200- μ m-thick as measured by a digital micrometer. The optical damage threshold was estimated experimentally to be >15 GW/cm² for our

conditions. By comparing the linear transmission of two perpendicular polarizations we concluded that most of the transmission losses were due to absorption along the polymer backbone, and not due to scattering.

Typical *Z*-scan results are shown in Figs. 1 and 2 for incident light polarization along the conjugation axis. (No nonlinear effects were observed with orthogonally polarized light.) For the initial analysis, we assumed $\Delta n = n_2 I$ and $\Delta \alpha = \alpha_2 I$ only. The open aperture *Z*-scan was used to deduce the two-photon absorption coefficient as well as the laser beam waist and the position of the focus. These parameters were then used to fit the closed aperture *Z*-scan results. Because of the large transmission changes due to both two-photon absorption and large nonlinear phase shifts, a full Fresnel diffraction calculation had to be performed and the methodology was verified by calibration measurements on

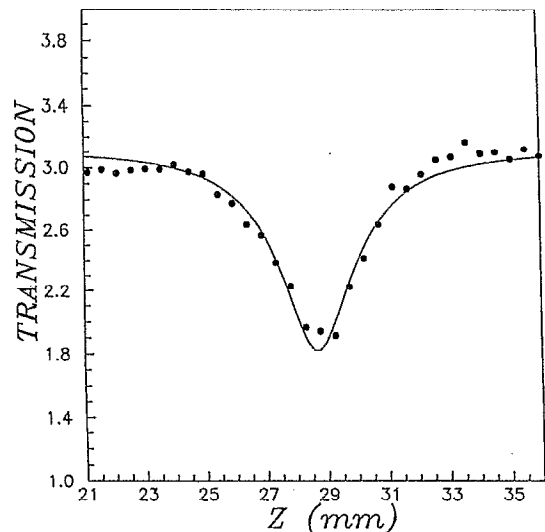


FIG. 1. Nonlinear transmission as a function of sample position for the open aperture case.

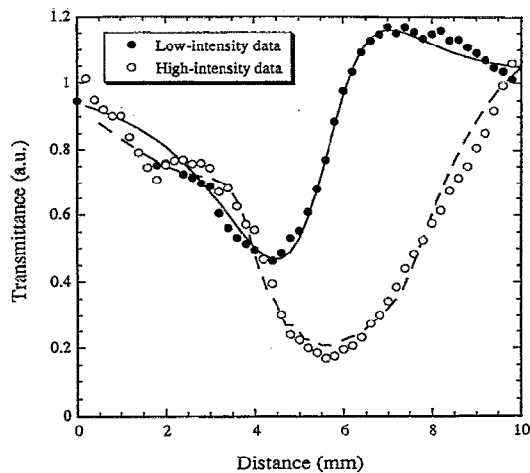


FIG. 2. Nonlinear transmission for an aperture $S=0.5$. The filled circles indicate low-intensity data (focusing, $I=0.9 \text{ GW/cm}^2$), and the open circles indicate high-intensity data (defocusing, $I=9 \text{ GW/cm}^2$). Note the shifts from the center position of the two minima as well as the sharp slope changing sign. In both cases the curves show an effect dominated by TPA.

CS_2 for which values of n_2 are known, giving comparable results to those of van Stryland within $\pm 30\%$.¹³ Experimental data was assumed satisfactory when low intensity ($< 50 \text{ MW/cm}^2$) scans gave no observable change on the transmission for both the open and closed aperture cases. The experimental procedure, including intensity scans, was reproduced three times on two different samples.

Our initial analysis indicates a variation in n_2 with incident intensity as shown in Fig. 3. n_2 appears to vary linearly with increasing intensity from 0 to 9 GW/cm^2 , implying that the material response at this wavelength and pulse width can be better described by $\Delta n = n_2(0)I + n_3(0)I^2$. Furthermore, the two-photon absorption coefficient, α_2 , also displayed a linear dependence on intensity as shown in Fig. 4,

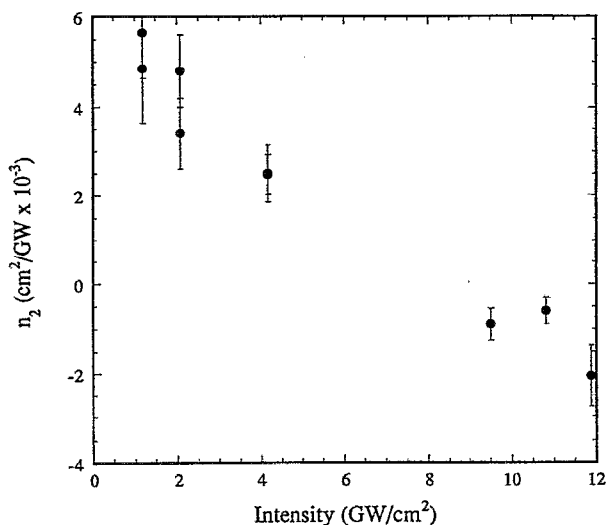


FIG. 3. Variation of the effective nonlinear refractive index coefficient n_2 as a function of the intensity, as measured with 35 ps pulses at 1064 nm from a 10 Hz Nd:YAG laser. The analysis used assumed $\Delta n = n_2 I$.

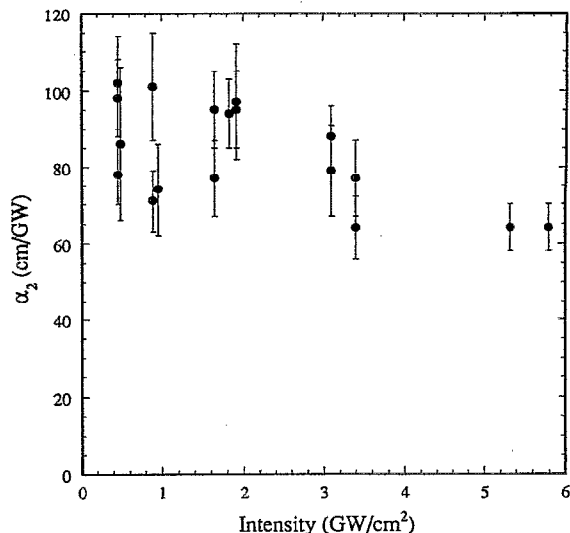


FIG. 4. Variation of the two-photon absorption coefficient α_2 as a function of the intensity, as measured with 35 ps pulses at 1064 nm from a 10 Hz Nd:YAG laser. The analysis used assumed $\Delta\alpha = \alpha_2 I$.

implying that including higher order effects via $\alpha(I) = \alpha_2(0)I + \alpha_3(0)I^2$ would also better describe the nonlinear absorption. As a result of the presence of higher-order intensity dependencies in the nonlinear absorption and refraction, calculations involving strictly n_2 and α_2 tend to break down at moderate to high intensities. Consequently, performing a linear fit to the results versus intensity is not a valid means of analysis, and the results serve only as the impetus to including the higher-order terms in the original fitting routines.

An iterative series of Fresnel diffraction calculations were performed on all of the data including the $n_3 I^2$ and $\alpha_3 I^2$ terms in the analysis. Examples of the resulting fits at low and high intensities are shown in Fig. 2. This revised calculation gave $n_2(0) = 5(\pm 1) \times 10^{-3} \text{ cm}^2/\text{GW}$, $n_3(0) = -5(\pm 1) \times 10^{-3} \text{ cm}^4/\text{GW}^2$, $\alpha_2(0) = 100(\pm 20) \text{ cm/GW}$, and $\alpha_3(0) = -5(\pm 1) \text{ cm}^3/\text{GW}^2$. Note that the effect of the higher order nonlinearity is sufficiently strong to reverse the sign of the nonlinear index change at a cw intensity of 1 GW/cm^2 .

Our low intensity value for $n_2(0)$ of $5 \times 10^{-12} \text{ cm}^2/\text{W}$ compares well with Ref. 9 ($n_2 = 5.5 \times 10^{-12} \text{ cm}^2/\text{W}$) obtained by using a modified Sagnac ring interferometer in which any slow thermal effects are avoided by design. Thermal effects are minimized in our case by the low repetition rate and the short pulses employed. Our linear extrapolation to $I=0$ gives $\alpha_2(0) = 100 \text{ cm/GW}$, which corresponds well with the value reported in Ref. 12 of 120 cm/GW . The uncertainty in the measured value of α_2 is probably due to thickness variations in PTS known to occur because of its laminar (stratified) structure. Repeated measurements performed on the same or a nearby spot were repeatable to $\pm 15\%$, verifying the technique's reproducibility.

Similar unusual behavior at 1064 nm has been reported before in the red form of poly-4BCMU in solution using degenerate four wave mixing.¹⁴ In that case, increasing the

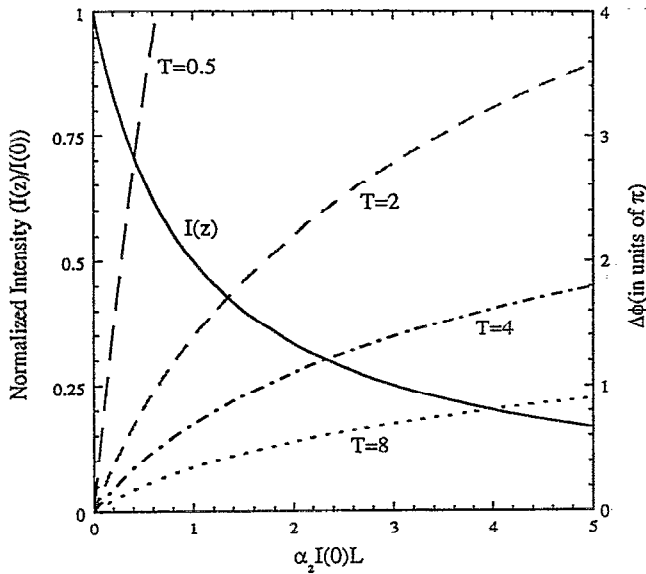


FIG. 5. Variation in the nonlinear phase shift $\Delta\phi_{NL}$ and the transmission vs the parameter $\alpha_2 I(0)L$ for representative T .

intensity led to an increase in $n_2(I)$ from a negative to a positive value, and to an increase in the two-photon absorption coefficient. Some of the features were interpreted in terms of a Stark shift and saturation of the two-photon level which occurs higher in energy than the energy of the two photons being absorbed. In our case, the two-photon absorption is occurring with a total two-photon energy between two two-photon active states (located at 2.05 and 2.7 eV), implementing those two two-photon states in Charra's and Nunzi's model could explain qualitatively the effects described here.¹⁵ The interpretation in Ref. 14 also included contributions from excited species in a three level model. Clearly the origin of these intensity dependencies merits further investigation.

There are two figures of merit used to assess the suitability of a nonlinear material for all-optical switching devices.¹⁶ If linear absorption dominates the loss, $W = \Delta n / \alpha \lambda > 1$ is needed. Here $W > 1$ for $I > 22 \text{ MW/cm}^2$, based on the attenuation coefficients reported in Ref. 11. When two-photon absorption (α_2) dominates the loss, $T = 2\alpha_2 \lambda / |n_2|$ should be less than unity. For PTS, $T > 3$ at 1064 nm. Shown in Fig. 5 are the two-photon absorption trade offs between the accu-

mulated nonlinear phase shift $\Delta\phi^{NL} = 4\pi \{ \ln[1 + \alpha_2 I(0)L] \} / T$ and the relative throughput $1/[1 + \alpha_2 I(0)L]$. Clearly for $T > 1$, large nonlinear phase shifts come at the expense of the throughput. Furthermore, it has previously been shown that with increasing T , the switching power increases and the switching becomes incomplete in a nonlinear directional coupler.¹⁷ Therefore, we conclude that PTS is marginal for all-optical applications at 1064 nm.

In conclusion, we have measured the complex nonlinear refractive index of PTS at 1064 nm. Both the nonlinear refractive index n_2 and the two-photon absorption coefficient α_2 were found to vary linearly with intensity, which could partially explain the large range of previously reported values. Unfortunately, at this wavelength, two-photon absorption prevents PTS from being a useful material for all-optical switching. We speculate that α_2 decreases at longer wavelengths making $T < 1$.

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