

# Large phase shifts in $\text{As}_2\text{S}_3$ waveguides for all-optical processing devices

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A large phase shift of  $4.7\pi$  at  $1.53\ \mu\text{m}$  has been observed from a low-loss ( $0.2\ \text{dB/cm}$ ), small-core  $\text{As}_2\text{S}_3$  waveguide fabricated by dry etching. The strength of the nonlinear response was limited by photosensitivity and photocrystallization of the  $\text{As}_2\text{S}_3$  films at  $1.53\ \mu\text{m}$ , far below the material bandgap. © 2005 Optical Society of America

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Self-phase modulation (SPM) can be used for all-optical processing in high-speed optical communication systems, allowing the creation of all-optical switches, supercontinuum generators, and all-optical regenerators.<sup>1–3</sup> However, the low optical nonlinearity of silica glass means that a significant nonlinear response at low pulse power can be obtained only after propagation through hundreds of meters of silica fiber. For many such applications chip-scale devices that integrate optical components to yield the desired functionality are preferable. To obtain a strong nonlinear response in integrated structures where the path length may have been reduced to only a few tens of centimeters requires a large reduction in the mode field area from the  $100\ \mu\text{m}^2$  of standard single-mode fibers to  $\approx 1\ \mu\text{m}^2$ , accompanied by a large increase in the third-order nonlinearity of the waveguide material.

Chalcogenide glasses are known to possess high third-order optical nonlinearity (up to 27,000 times that of silica has been reported<sup>4–6</sup>) and generally good nonlinear figures of merit. Although the photosensitivity is employed to directly write channel waveguide structures into thin chalcogenide films,<sup>7</sup> unfortunately, in most cases the photoinduced refractive-index changes are not stable, and alternative approaches are required for waveguide fabrication.

In this context we recently reported high-refractive-index-contrast  $\text{As}_2\text{S}_3$  rib waveguides with losses of  $\sim 0.25\ \text{dB/cm}$  at  $1.55\ \mu\text{m}$  fabricated by standard photolithography and dry etching.<sup>8</sup> Furthermore Ponnampalam *et al.*<sup>9</sup> reported the production of rib waveguides by selective chemical etching of photodarkened  $\text{As}_2\text{S}_3$  glass and obtained losses also down to  $0.25\ \text{dB/cm}$ . These values are sufficiently low to allow integrated devices containing waveguides several tens of centimeters long to be fabricated. In addition, the high refractive index contrast of the chalcogenide waveguide supports a small mode, thus enhancing the optical field intensity for fixed optical power. This can also support low-loss bends with small bend radii, making it possible to coil the waveguides to the extend interaction length to hundreds of centimeters while occupying only a small area on a chip. Thus chalcogenide waveguides should allow all-optical processing at low peak power in compact structures.

In this paper, we report the measurement of large nonlinear phase shifts due to strong self-phase modulation in waveguides based on our latest fabrication methods that produce small-core rib  $\text{As}_2\text{S}_3$  waveguides by using inductively coupled plasma (ICP) etching. A phase shift of up to  $4.7\pi$  was observed in a 6 cm long waveguide with a  $1530\ \text{nm}$  pulse source. Under the extreme conditions of these experiments degradation of the waveguides was observed, as has been reported by other researchers.<sup>10</sup> The threshold for degradation limits the maximum intensity that can be used with  $\text{As}_2\text{S}_3$ , and that has some implications for future device design.

$\text{As}_2\text{S}_3$  films about  $2.5\ \mu\text{m}$  thick were deposited by ultrafast pulsed laser deposition (PLD) onto oxidized silicon wafers. The  $\text{As}_2\text{S}_3$  rib waveguide shown in Fig. 1 was etched by using ICP as described in Ref. 8. The optimum conditions leading to a smooth and vertical sidewall profile were similar to those reported in Ref. 8, except that a much lower rf power ( $300\ \text{W}$ ) and slightly higher bias voltage ( $170\ \text{V}$ ) were used because the ICP produces a much higher etch rate in  $\text{As}_2\text{S}_3$  than does the helicon etcher that was used for the majority of the work reported in Ref. 8. A photoresist mask rather than a metal mask could be used with ICP etching because a reasonable  $\text{As}_2\text{S}_3$  etch selectivity (3:1) to photoresist was obtained, and only a relatively shallow etch depth ( $\approx 1.5\ \mu\text{m}$ ) was required in these high-refractive-index-contrast waveguides. The benefits of the photoresist mask are significantly simplified processing and an improvement in waveguide quality. The sidewall roughness was significantly lower compared with the results presented in Ref. 8 and was estimated to be  $< 20\ \text{nm}$

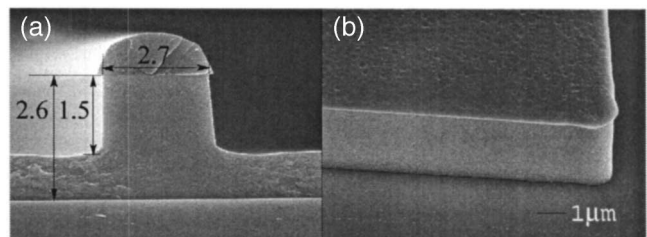


Fig. 1. (a)  $\text{As}_2\text{S}_3$  waveguide for SPM measurement (micrometers). (b)  $\text{As}_2\text{S}_3$  pattern, showing a smooth sidewall.

from scanning electron microscopy images such as those shown in Fig. 1(b).

The waveguide dimension for SPM measurements is shown in Fig. 1(a). A high-numerical-aperture (NA) fiber was used to launch light into the laterally tapered  $\text{As}_2\text{S}_3$  waveguide with a polysiloxane cladding. The effective fundamental modal area  $A_{\text{eff}}$  of the waveguide was calculated to be  $\sim 4.2 \mu\text{m}^2$  by using commercial Olympios software, resulting in the mode mismatch loss of 1.2 dB. The total insertion loss of the 6 cm long waveguide was 5 dB, corresponding to an estimated propagation loss of  $\approx 0.2 \text{ dB/cm}$ . The value is somewhat lower than that we have previously reported<sup>8</sup> and is consistent with the lower sidewall roughness obtained using ICP etching.

Figure 2(a) illustrates the experimental apparatus used for self-phase modulation in the  $\text{As}_2\text{S}_3$  waveguide. The laser source consists of a single-pass optical parametric amplifier based on a 10 mm long periodically poled MgO-doped lithium niobate (MgO:PPLN) crystal. The optical parametric amplifier was seeded with a narrowband tunable diode laser at a wavelength of 1530 nm and pumped at 1064 nm with 13 ps pulses from a long-cavity diode-pumped passively mode-locked Nd:YVO<sub>4</sub> laser.<sup>11</sup> The laser source specially designed for this experiment can provide an average power of  $\approx 20 \text{ mW}$  at maximum, which corresponds to a pulse peak power of 1300 W.

The 1530 nm pulse spectrum is shown in Fig. 2(b) and corresponds to an approximately Gaussian pulse with duration of 7 ps. However, in this spectrum there is some asymmetry, which was also observed in the spectrum of the 1064 nm pump pulses, most likely indicating asymmetry in the pulse envelope, which would not be detected using our autocorrelator. Note that in all these measurements the cw background has been subtracted from the measured spectrum.

The high power output of a 95/5 fiber coupler was spliced into a 5 mm long high-NA fiber and used to launch 1530 nm pulsed laser light into the waveguide, while the low output was used for power monitoring. The input power was varied by adjusting the coupling from the optical parametric amplifier source into the coupler. The lengths of input-output fibers were kept as short as possible to ensure that they did not introduce any additional phase shift at high power.

For the spectral broadening measurement, only the TE mode was excited in the waveguides. Figure 3(a)

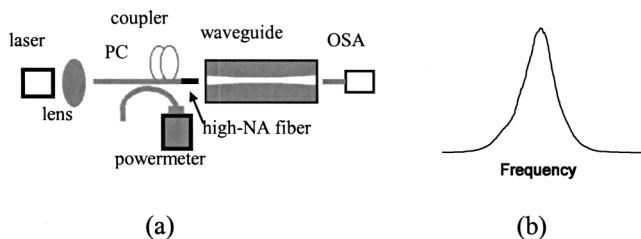


Fig. 2. (a) Experimental setup used for measuring SPM in the  $\text{As}_2\text{S}_3$  waveguide. (b) Laser source spectrum, showing some asymmetry.

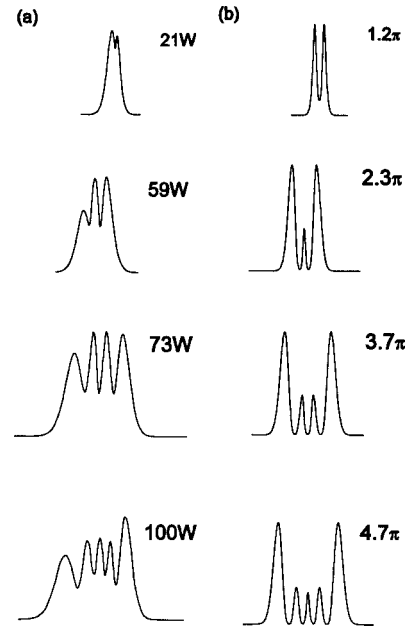


Fig. 3. (a) Experimental spectral broadening, corresponding to different peak powers in the waveguide. (b) Simulated spectra for increased peak power.

shows several spectra corresponding to different peak intensities. These spectra are compared with numerical simulations based on the nonlinear Schrödinger equation, as shown in Fig. 3(b). The calculation from the dispersion of the refractive index of  $\text{As}_2\text{S}_3$  films<sup>12</sup> and literature reports<sup>7</sup> indicate that line dispersion  $\beta_2$  of  $\text{As}_2\text{S}_3$  films at 1530 nm is about  $4 \times 10^{-3}$  to  $6 \times 10^{-3} \text{ ps}^2/\text{cm}$  and can be neglected. Therefore only the spectral broadening produced by SPM was considered in these simulations.

It can be seen that there is some asymmetry in the measured spectra that can be qualitatively understood to arise from asymmetry in the pulse shape.<sup>13</sup> Since the number of peaks in the spectra depends on the maximum phase shift rather than the shape of the pulse, counting the peak is a better measure of maximum phase shift than measuring the frequency width.<sup>13</sup> Through comparison of the spectral broadening between our measurements and the simulations, it can be seen that the largest phase shift from self-phase modulation was  $4.7\pi$  for a peak power of 100 W in the waveguide. A phase shift as large as  $4.7\pi$  is sufficient for many applications, exceeding that required for all-optical switching and close to that required for an all-optical regenerator. To fit the experimental results we used the value  $n_2 = 2.93 \times 10^{-14} \text{ cm}^2/\text{W}$  for the nonlinear refractive index of  $\text{As}_2\text{S}_3$ , which is consistent with our previous measurements for bulk glass.<sup>8</sup> Values for the nonlinearity for bulk  $\text{As}_2\text{S}_3$  glasses reported by other researchers range from  $2.2 \times 10^{-14}$  to  $6.6 \times 10^{-14} \text{ cm}^2/\text{W}$ ,<sup>5,14</sup> while thermally evaporated  $\text{As}_2\text{S}_3$  films have been reported to have values between  $0.8 \times 10^{-14}$  and  $6.9 \times 10^{-14} \text{ cm}^2/\text{W}$ ,<sup>7,14</sup> the higher values corresponding to photodarkened films.

Although in principle a larger phase shift or even spontaneous Raman scattering should be obtained if the power were increased to an even higher value,

when this was attempted, the spectral broadening was observed to decrease. Since the average incident intensity was much less than the thermal damage threshold, linear absorption and two-photon absorption should be negligible under these conditions. Therefore we investigated this phenomenon further.

Observation of the output beams with an infrared camera showed that as the input power was increased the output first increased proportionally until a critical point was reached, when the output beam power started to fluctuate rapidly. If the input power was further increased, the output power dropped almost immediately, indicating that the waveguides had been damaged. This appears to be related to the unexpected photosensitivity of  $\text{As}_2\text{S}_3$  waveguides at  $1.5\ \mu\text{m}$ , which was first reported by Hô *et al.*<sup>10</sup> At high peak intensity the refractive index first increases, causing the modes to redistribute. With a further increase in intensity laser-induced crystallization occurs,<sup>15</sup> resulting in a rapid increase in waveguide loss. The critical intensity representing the threshold for photodegradation was found to be  $\approx 2\ \text{GW}/\text{cm}^2$ . This threshold corresponds to an average power of only  $\approx 1.4\ \text{mW}$  propagating in the waveguide, suggesting that the photocrystallization is not a thermal process.<sup>16</sup>

To illustrate the effect of high intensity on the average refractive index in the waveguides, Fig. 4 compares the output images of an  $\text{As}_2\text{S}_3$  waveguide directional coupler before and after injection of a beam at an intensity of  $2.6\ \text{GW}/\text{cm}^2$ . The coupling ratio has changed from  $\approx 70:30$  to  $\approx 55:45$  owing to the change of refractive index caused by photodarkening by the high-intensity pulses. In these couplers the interaction length (5 cm) was much longer than the coupling length ( $\approx 1.2\ \text{cm}$ ), making it difficult to calculate the exact change of refractive index caused by photodarkening. However, an approximate value could be estimated because a similar change in coupling purely due to the Kerr response required intensities of  $2\text{--}2.5\ \text{GW}/\text{cm}^2$  and corresponded to a change in material index of  $5 \times 10^{-5}$  to  $8 \times 10^{-5}$ .

Raman spectra have indicated that our PLD  $\text{As}_2\text{S}_3$  films contain  $\text{As}_4\text{S}_4$  structures similar to that of thermally evaporated ones.<sup>10</sup> Such a composition has a different bond structure and has been observed to be unstable even at room temperature, especially for films that contain defects. The higher photodegradation threshold compared with that obtained in Ref.

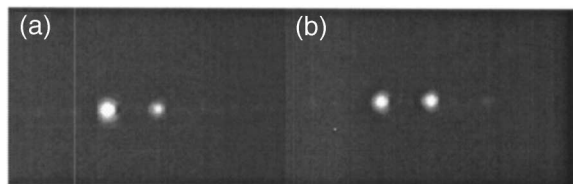


Fig. 4. Output beam images of a directional  $\text{As}_2\text{S}_3$  coupler (a) before and (b) after a high-peak-intensity exposure in the waveguide.

10 may imply that the PLD  $\text{As}_2\text{S}_3$  films contain fewer  $\text{As}_4\text{S}_4$  clusters.

In conclusion, we have reported a large SPM-induced phase shift of  $4.7\pi$  at  $1.5\ \mu\text{m}$  from the  $\text{As}_2\text{S}_3$  waveguides first fabricated by dry etching. Any further increase of the phase shift was prevented by photosensitivity and photocrystallization of  $\text{As}_2\text{S}_3$  films irradiated at intensities above a few gigawatts per square centimeter by the  $1.53\ \mu\text{m}$  beam. Clearly this places a significant limitation for practical devices, and in particular larger phase shifts will not be achieved by simply decreasing the mode volume to increase the intensity in the waveguides because of this photodegradation, although the approach is useful in reducing the operating power of the nonlinear device. Hence, to achieve large phase shifts, longer path lengths will inevitably need to be employed, but the peak intensity must remain well below  $1\ \text{GW}/\text{cm}^2$ , at least for  $\text{As}_2\text{S}_3$ . Thus typical device length in  $\text{As}_2\text{S}_3$  waveguides will need to lie in the  $10\text{--}50\ \text{cm}$  range, which requires the production of structures with propagation loss below  $0.1\ \text{dB}/\text{cm}$ . We believe our results indicate that this is achievable with our current technology.

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