Resonant cavity-enhanced photosensitivity in As$_2$S$_3$ chalcogenide glass at 1550 nm telecommunication wavelength
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We report the first (to our knowledge) experimental observation of resonant cavity-enhanced photosensitivity in As$_2$S$_3$ chalcogenide glass film at 1550 nm telecommunication wavelength. The measured photosensitivity threshold is $<0.1$ GW/cm$^2$, and a photoinduced refractive index increase as large as 0.016 is observed. The photosensitive process is athermal; further, we confirm the absence of two-photon absorption in As$_2$S$_3$, suggesting that defect absorption accounts for the energy transfer from photons to glass network. Besides its potential application for reconfigurable photonics circuit, such photosensitivity is also an important design consideration for nonlinear optical devices using chalcogenide glasses.

Chalcogenide glasses (ChGs), namely, the amorphous compounds containing sulfur, selenium, and/or tellurium, have emerged as a promising material for IR photonics. It has been well documented that these glasses exhibit photosensitivity to near-bandgap illumination [1–3], a property that has been exploited for device fabrication [4–6]. On the other front, the performance characteristics of photonic devices strongly depend on the refractive indices of their constituent materials, thus the recent surge of interest in using chalcogenide glasses as a Kerr medium for ultrafast optical-switching calls for an in-depth study of glass photosensitivity to light at telecommunication wavelengths, given the high optical power density involved in the nonlinear processes. Data on the photosensitivity of sulfide glasses at telecommunication wavelengths, however, are scarce to date largely due to the much weaker interaction between glass and photons with energy well below its bandgap and the lack of sensitive characterization techniques [7].

Here we report the first (to our knowledge) experimental observation of resonant cavity-enhanced photosensitivity of As$_2$S$_3$ at 1550 nm telecommunication wavelength, using our recently developed resonant cavity refractometry technique [8]. In resonant cavity refractometry, an optical resonant cavity is leveraged as a sensitive spectroscopic tool to detect refractive index and/or optical absorption change in the material comprising of the cavity and thereby provide information on glass structural evolution kinetics that are otherwise difficult to obtain using conventional metrology techniques.

In this study, the optical resonators used are 30 μm radius microdisks coupled to a bus waveguide using a pulley configuration and patterned in thermally evaporated As$_2$S$_3$ (n = 2.4) glass film. Details of the device fabrication are described elsewhere [9–11]. The bus waveguides are 800 nm wide. Both microdisks and bus waveguides are 450 nm in height. A layer of SU8 polymer cladding (n = 1.57) is coated on top of the devices to prevent surface oxidation.

Prior to measurement, the resonators are aged at room temperature until the resonant wavelength is stabilized. Table 1 lists the linear optical parameters of the microdisk resonators near 1550 nm wavelength, measured at low input-power levels to avoid nonlinear effects. In the microdisk, optical power is enhanced by more than 15 dB at resonance with respect to the bus waveguide. Notably, the resonators exhibit a negative thermo-optic (TO) coefficient as a result of the SU8 polymer over cladding, from which we infer an As$_2$S$_3$ material TO coefficient of $\sim4 \times 10^{-5}$ K$^{-1}$, in good agreement with a previous report [12].

In the first set of nonlinear optical measurements, TM polarization light from a tunable laser is amplified using an erbium-doped fiber amplifier (EDFA) and then end coupled into a bus waveguide through an optical fiber. The output light from the bus waveguide is fiber coupled, and its power is recorded using an optical spectrum analyzer (OSA). The unoptimized coupling loss per facet is $\sim(4.5 \pm 1)$ dB. Figure 1 plots the transmission spectra of the microdisks measured at different input power levels; the arrows indicate the temporal sequence of the measurements; i.e., the measurement starts with a low input power (linear regime), and then the coupled power into the bus waveguide is increased up to 18.6 dBm and finally decreased back to the linear regime. From the figure it is clear that the resonant peak is distorted toward a shark-fin-like shape at high input power, suggesting a cavity instability phenomenon. Our calculations indicate that the optical power inside the resonator is still 1 order of magnitude smaller than...
that required to trigger Kerr instability. Moreover, the resonant peak is slanted toward a shorter wavelength, which cannot be explained by the photosensitivity effect that we will discuss in the next paragraph. Therefore we attribute the peak shape distortion to a TO cavity instability, which results from heat deposited during a high-power measurement and the subsequent resonant wavelength change. Similar cavity instability phenomena have been reported in As$_2$Se$_3$ microspheres recently [13]. The power threshold for triggering such instability is 2 orders of magnitude higher in our case owing to the improved heat conduction from the on-chip microdisk to silicon substrate heat sink. Using the device TO coefficient from Table 1, the temperature increase of the microdisk is calculated to be $7 \, ^\circ\text{C}$ even at 18.6 dBm input power, and thus no thermal-induced material degradation is expected. For nonlinear optical applications, such an increased instability threshold is critical, as it allows higher power for enhanced nonlinear functionalities. In addition, the partial TO coefficient compensation between Chg (with a positive TO coefficient) and polymer cladding (with a negative TO coefficient) suggests possible further increase of the instability threshold by employing an athermal design.

In addition to the transient TO response, the permanent resonant wavelength redshift shown in Fig. 1 after the high-power test indicates a nontransient refractive index change in As$_2$S$_3$. To verify the origin of such index modification, a series of pump–probe measurements is performed using a setup schematically illustrated in Fig. 2. In the test, light from a pump laser amplified by an EDFA is used to induce index change in the As$_2$S$_3$ microdisk, and the corresponding resonance shift is measured independently using a probe laser. Figure 3 shows the transmission spectra evolution when the pump light is (a) off and (b) on microdisk resonance. Compared with Fig. 1, in Fig. 3 no peak distortion occurs, because in this case the peak shift originates from a “cross-induced” phenomenon. Apparently, a resonant peak redshift is observed when the pump light is actively locked to the microdisk resonant wavelength; in contrast, when the pump light is off-resonance, no resonant peak shift is observed at even the maximum pump power. This result suggests that the glass index change stems from light coupled into the microdisk at resonance and hence resonantly enhanced photon–matter interaction. From Fig. 3, we can also estimate the threshold power density for such cavity-enhanced photosensitivity to be $0.1 \, \text{GW/cm}^2$, based on the onset of peak shift at a resonant pumping power of 9.6 dBm. This threshold figure is 1 order of magnitude smaller compared with a previously reported value [7], probably because the resonant cavity refractometry method we use enables the sensitive detection of small refractive index variation. We measure refractive index change as large as $1.6 \times 10^{-2}$ due to cavity-enhanced photosensitivity and no obvious saturation behavior has been observed yet.

From a material perspective, we seek to understand the origin of such photosensitivity. Since heating effect during our tests is minimal ($<7 \, ^\circ\text{C}$), we can exclude heat-induced structural relaxation (“annealing”). In addition, since the photon energy at 1550 nm wavelength is much smaller than the optical bandgap of As$_2$S$_3$, other mechanisms that possibly account for the photosensitivity necessarily involve energy transfer to glass network via subgap absorption, including defect (midgap localized states) and nonlinear absorption with the latter mainly referring to two-photon absorption (TPA). To clarify the energy-transfer mechanism, we notice that optical resonance extinction ratio critically depends on optical loss, a phenomenon we can leverage to accurately measurement nonlinear absorption in As$_2$S$_3$ microdisks. Details regarding the data processing

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**Table 1. Linear Optical Parameters of the As$_2$S$_3$ Microdisk Resonators Used in This Study**

<table>
<thead>
<tr>
<th>Cavity Q factor</th>
<th>FSR</th>
<th>Optical Power Enhancement</th>
<th>Equivalent Loss</th>
<th>Thermo-optic Coefficient</th>
</tr>
</thead>
<tbody>
<tr>
<td>$60,000 \pm 10%$</td>
<td>626 GHz</td>
<td>($15.2 \pm 0.5$) dB</td>
<td>2.4 dB/cm</td>
<td>$-5 \times 10^{-6} , \text{K}^{-1}$</td>
</tr>
</tbody>
</table>

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**Fig. 1.** (Color online) Transmission spectra of an As$_2$S$_3$ microdisk resonator near a resonant peak measured at different power levels; the arrows indicate the temporal sequence of measurements. The resonant peak redshift after high-power measurement manifests the photosensitivity effect, and the peak shape distortion at high power is a consequence of thermo-optic cavity instability.

**Fig. 2.** (Color online) Schematic illustration of the pump–probe measurement setup used to characterize cavity-enhanced photosensitivity in As$_2$S$_3$ glass.
Fig. 3. (Color online) Transmission spectra of As$_2$S$_3$ microdisks measured using a low-power probe beam in a pump-probe setting (a) when the pump light is off-resonance; (b) when the pump light is locked with microdisk resonant wavelength, cavity-enhanced photosensitivity leads to the nontransient redshift of resonance, and the spectra with different pump power are offset vertically for clarity.

methodology can be found elsewhere [14]. Figure 4 gives the measured nonlinear absorption component (on top of existing linear absorption) as a function of circulating power inside the resonator. In the presence of TPA, nonlinear absorption is supposed to be linearly proportional to the circulating power; the lack of such linearity in Fig. 4 suggests the absence of TPA within the accuracy of our cavity-enhanced absorption spectroscopy technique. The technique gives an upper limit of TPA coefficient in As$_2$S$_3$ film to be $1.5 \times 10^{-13}$ m/W. The upper limit of nonlinear absorption determined from our measurement (0.008 dB/cm) is almost 2 orders of magnitude smaller compared with linear absorption, and therefore we conclude that subgap defect absorption is responsible for the energy transfer from 1550 nm wavelength photons to the glass network in the photosensitive process. Since defect density and energy distribution in glasses are highly processing-dependent, we expect purposive control of the photosensitivity threshold via optimizing film deposition and treatment processes.

In summary, we report the first experimental observation and characterization of cavity-enhanced photosensitivity in ChGs at 1550 nm telecommunication wavelength. The measured photosensitivity threshold is $<0.1\text{GW/cm}^2$. We further leverage cavity-enhanced absorption spectroscopy for measuring nonlinear absorption in thin films, to confirm the absence of TPA in As$_2$S$_3$, which suggests that the energy transfer from 1550 nm photons to the glass network occurs via defect absorption. Such photosensitivity is an important design consideration for chalcogenide glass nonlinear optical devices, given the high optical power density involved. In addition, this discovery demonstrates a highly selective technique for in situ tuning of ChG resonators and thus paves the path toward reconfigurable photonic circuits.

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References