

# Third order cascaded Raman wavelength shifting in chalcogenide fibers and determination of Raman gain coefficient

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**Abstract:** Cascaded Raman wavelength shifting up to three orders from 1553 nm to 1867 nm is demonstrated in  $\text{As}_2\text{S}_3$ -chalcogenide fibers. Due to a long zero dispersion wavelength for the sulfide fiber ( $>4.5 \mu\text{m}$ ), pumping the fiber at 1553 nm results in generation of cascaded Stokes orders based on stimulated Raman scattering. Using the threshold power for the Raman orders, we estimate the Raman gain coefficient for the  $\text{As}_2\text{S}_3$  fibers to be  $\sim 5.7 \times 10^{-12} \text{ m/W}$  at 1550 nm. Observation of higher Raman orders is limited by damage to the fiber at input intensities  $>1 \text{ GW/cm}^2$ .

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OCIS codes: (190.5650) Raman effect; (190.4370) Nonlinear optics, fibers.

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## References

1. F. Smektala, C. Quemard, L. Leneindre, J. Lucas, A. Barthélémy, C. De Angelis, "Chalcogenide glasses with large non-linear refractive indices," *J. Non-Cryst. Solids* **239**, 139-142 (1998).
  2. T. Cardinal, K. A. Richardson, H. Shim, A. Schulte, R. Beatty, K. Le Foulgoc, C. Meneghini, J. F. Viens and A. Villeneuve, "Non-linear optical properties of chalcogenide glasses in the system As-S-Se," *J. Non-Cryst. Solids* **256 & 257**, 353-360 (1999).
  3. J. M. Harbold, F. Ö. Ilday, F. W. Wise, J. S. Sanghera, V. Q. Nguyen, L. B. Shaw, I. D. Aggarwal, "Highly nonlinear As-S-Se glasses for all-optical switching," *Opt. Lett.* **27**, 119-121 (2002).
  4. M. Asobe, T. Kanamori, K. Naganuma, H. Itoh, T. Kaino, "Third-order nonlinear spectroscopy in  $\text{As}_2\text{S}_3$  chalcogenide glass fibers," *J. Appl. Phys.* **77**, 5518-23 (1995).
  5. J. S. Sanghera, I. D. Aggarwal, "Active and passive chalcogenide glass optical fibers for IR applications: a review," *J. Non-Cryst. Solids* **256 & 257**, 6-16 (1999).
  6. D.-P. Wei, T. V. Galstian, I. V. Smolnikov, V. G. Plotnichenko, A. Zohrabyan, "Spectral broadening of femtosecond pulses in a single-mode As-S glass fiber," *Opt. Express* **13**, 2439-2443 (2005).
  7. S. D. Jackson, G. Anzueto-Sánchez, "Chalcogenide glass Raman fiber laser," *Appl. Phys. Lett.* **88**, 221106 (2006).
  8. Govind P. Agrawal, *Nonlinear Fiber Optics* (Academic Press, 1989), Chap. 8.
  9. A. K. Abeeluck, C. Headley, "Continuous-wave pumping in the anomalous- and normal-dispersion regimes of nonlinear fibers for supercontinuum generation," *Opt. Lett.* **30**, 61-63 (2005).
  10. M. E. Lines, "Ultralow-Loss Glasses," *Annu. Rev. Mater. Sci.* **16**, 113-135 (1986).
  11. A. A. Kikineshi, I. Rosola, "Material dispersion in  $(\text{As}_2\text{S}_3)_x(\text{AsI}_3)_{1-x}$  glasses," *Proc. SPIE* **3890**, 502-506, 1999.
  12. Stuart D. Jackson and Paul H. Muir, "Theory and numerical simulation of nth-order cascaded Raman fiber lasers," *J. Opt. Soc. Am. B* **18**, 1297-1306 (2001).
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## 1. Introduction

We report for the first time to our knowledge, observation of up to three orders of cascaded Raman wavelength shifting (CRWS) in highly non-linear  $\text{As}_2\text{S}_3$ -(sulfide) fibers. We shift light by  $\sim 300$  nm from the pump wavelength at 1553 nm using three Stokes orders. Also, based on the threshold powers for the three Raman orders, we estimate the Raman gain coefficient ( $g_R$ ) for the sulfide fibers to be  $\sim 5.7 \times 10^{-12}$  m/W. Previously reported values for the nonlinear parameters of the sulfide glass vary widely from  $\sim 35\times$  [1] to about  $73\times$  [2], to  $\sim 220\times$  [3] that of fused silica. The  $g_R$  for our sulfide fibers is  $\sim 89$  times that of fused silica at 1550 nm and our results are in agreement with the value reported for sulfide fibers in Ref. [4]. Beyond three orders, damage occurs in the fibers, corresponding to a damage threshold of  $\sim 1$  GW/cm<sup>2</sup> [5].

For mid IR light generation, all-fiber based sources are attractive as they provide for operation at room-temperature and avoid moving parts. Chalcogenide fibers are good candidates for all-fiber based sources due to their low loss in the IR and high nonlinearity coefficients, up to  $\sim 900$  times compared to fused silica fibers [1-3]. Nonlinear phenomenon occurring in the chalcogenides has therefore attracted attention recently. Strong spectral broadening of femtosecond pulses based on self-phase modulation (SPM) in sulfide fibers has been reported by Wei et al. [6]. Recently, a Raman fiber laser in  $\text{As}_2\text{Se}_3$  chalcogenide fibers operating at  $\sim 2100$  nm with a  $\text{Tm}^{3+}$  pump has also been reported [7]. Using nanosecond pump pulses, we are able to observe CRWS up to three orders in the sulfide fibers.

For nanosecond pulses, CRWS is the dominant nonlinear effect observed at telecom wavelengths in sulfide fibers. Due to the long zero dispersion wavelength (ZDW) of the fibers ( $>4.5$   $\mu\text{m}$ ), pumping at 1550 nm implies operation in the strongly normal dispersion regime. Therefore, modulation instability (MI) does not phase-match and pulse breakup leading to super continuum (SC) generation is not observed. However, beyond a Raman threshold, the pump is frequency-shifted to generate a Stokes wave through stimulated Raman scattering (SRS) [8]. Higher-order Stokes lines also appear at high pump powers when the Stokes power becomes large enough to pump the next-order Stokes line. The cascaded effect occurs until the pump eventually shifts into the anomalous dispersion regime and generates MI seeded SC [9]. Hence, in the case of sulfide fibers, a large ZDW suggests several cascaded Stokes orders and wavelength shifting up to  $\sim 4.6$   $\mu\text{m}$ . However, owing to a low damage threshold for the sulfide fibers, CRWS beyond three orders is not observed.

## 2. Fiber specifications and experimental set-up

We use two different sulfide fibers in our experiments. For both the fibers, the cladding glass ( $\text{As}_{40}\text{S}_{60}$ ) was made by melting appropriate mixtures of purified As and S in an evacuated silica ampoules. Thermally compatible higher index version core glasses were made similarly by doping with Se or Sb, resulting in an  $\text{As}_{40}\text{Se}_2\text{S}_{58}$  or  $\text{As}_{38}\text{Sb}_2\text{S}_{60}$  core for fiber 1 and fiber 2, respectively. The core glasses were fabricated as 1 cm thick cylindrical rods, and drawn at  $400^\circ\text{C}$  to 1 mm diameter canes. The cladding glass was made as 3.5 cm boules that were subsequently extruded through a vitreous C die at  $330^\circ\text{C}$  into 14 mm outer diameter (OD), 1 mm inner diameter (ID) tubing. The core canes were sleeved with the cladding material to make rod-in-tube preforms that were subsequently redrawn at  $400^\circ\text{C}$  into acrylate coated fibers with different core diameters. Sulfide fiber 1 has a core diameter of  $\sim 6.5$   $\mu\text{m}$ , whereas fiber 2 has a core diameter of  $\sim 5.4$   $\mu\text{m}$ . The refractive index of the two glasses at 1549 nm was measured using a Metricon prism coupler. Both the fibers have a cladding with refractive index of 2.438. For fiber 1, the core has a refractive index of 2.448 and a numerical aperture (NA) of 0.22 whereas; fiber 2 has a core refractive index of 2.456 and a NA of 0.3.

Figure 1(a) illustrates the loss measured in 2.03 m of fiber 1 and 7.6 m of fiber 2. The absorption peaks beyond 2.7  $\mu\text{m}$  for both fibers, are due to impurities such as  $\text{H}_2\text{S}$  or from oxides of the original cationic elements or water molecules [10]. The linear fiber loss at 1.55  $\mu\text{m}$  wavelength is  $\sim 0.75$  dB/m for fiber 1 and  $\sim 1.1$  dB/m for fiber 2. In the case of sulfide fibers, residual modes have been found to exist in the cladding [6]. To eliminate these modes, gallium can be used on the bare ends of the fiber. Presence of metal coating prevents

excitation of unwanted cladding modes at the input end. Based on such measurement, the loss in the core is observed to be  $\sim 1$  dB/m for fiber 1 and  $\sim 1.25$  dB/m for fiber 2.

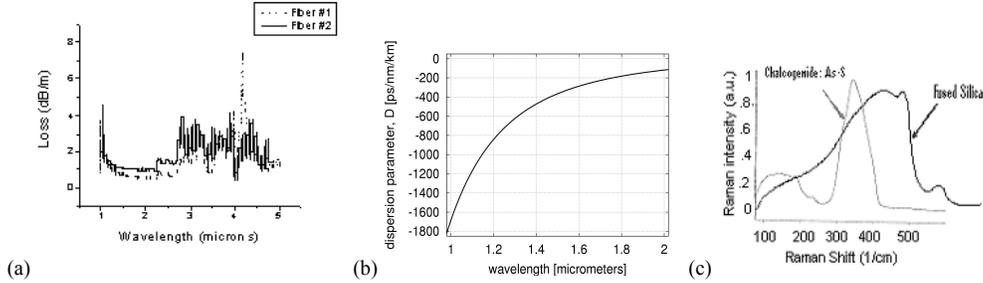


Fig. 1 – (a) Loss curves in the 1-5  $\mu\text{m}$  region for the two fibers. (b) Dispersion curve for both fibers. (c) Comparison of Raman spectra measured in bulk- $\text{As}_2\text{S}_3$  material using  $90^\circ$  scattering geometry and fused silica (Ref. [8]).

The two fibers have similar dispersion characteristics. Based on curve fitting of the refractive index, we obtain the following approximation to the fiber refractive index in the 1000-2000 nm range:

$$n^2 = a_0 + \frac{a_1^2}{\lambda^2} + \frac{a_2^4}{\lambda^4} - \frac{\lambda^2}{a_3^2}, \quad (1)$$

where,  $a_0 = 5.85$ ,  $a_1 = 482.38$  nm,  $a_2 = 491.41$  nm,  $a_3 = 10387.35$  nm. Figure 1(b) illustrates the dispersion curve for the two fibers in the 1000-2000 nm range. Both fibers have a ZDW of  $\sim 4.6$   $\mu\text{m}$ , which compares to a similar value of 4.81  $\mu\text{m}$  for sulfide glass as quoted in [11]. Raman spectrum measured for a polished  $\text{As}_2\text{S}_3$ -glass surface using a 752.5 nm source in a  $90^\circ$  scattering geometry is depicted in Fig. 1(c). The Raman spectrum for fused silica has been superimposed to provide a comparison in the line shape. The spectrum shows a distinct peak at a Stokes shift of  $\Delta k = 345$   $\text{cm}^{-1}$  for sulfide fibers as compared to 440  $\text{cm}^{-1}$  for the case of fused silica. The sulfide fiber also has a narrower Raman gain linewidth ( $\sim 90$   $\text{cm}^{-1}$ ) compared to fused silica ( $\sim 250$   $\text{cm}^{-1}$ ). The peak value for  $g_R$  for sulfide fibers is expected to be larger than that of fused silica owing to the higher nonlinearity, and is estimated later in the paper based on our experimental results.

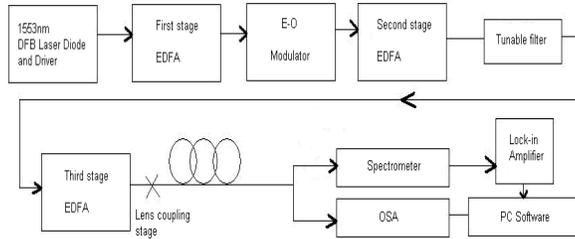


Fig. 2(a) Block diagram of experimental set-up

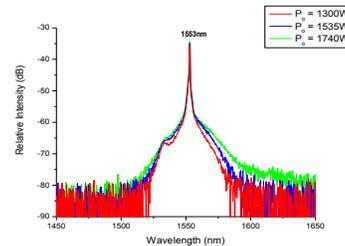


Fig. 2(b) Spectrum from EDFA

A block diagram of the experimental setup is illustrated in Fig. 2(a). A distributed feedback (DFB) laser diode provides the 1553 nm seed light with 2-ns pulse width at a 5 kHz repetition rate. An electro-optic (E-O) modulator synchronized to the pulse generator and a tunable filter are used to suppress the amplified spontaneous emission. The light is then boosted up through three stages of erbium-doped fiber amplifier (EDFA) and peak powers of up to  $\sim 1.74$  kW can be obtained. Figure 2(b) shows the spectrum of the laser with a peak at 1553 nm. The spectrum is clean and without significant nonlinear effects. Lenses are used to

couple light into the sulfide fibers and an efficiency of  $\sim 70\%$  is achieved. The short wavelength spectrum (700-1770 nm) at the output is measured by an optical spectrum analyzer (OSA) while the long wavelength spectrum is measured by dry nitrogen purged, grating spectrometer followed by a long-wavelength enhanced type InGaAs detector. A lock-in amplifier and PC setup is used to acquire data from the detector.

### 3. Results

We observe CRWS up to three orders in the sulfide fiber 1. Figure 3(a) shows the spectrum from 12 m of fiber 1 at a peak incident power of 350 W. The pump is shifted out to 1867 nm using three cascaded Raman orders corresponding to a wavelength shift of  $\sim 314$  nm. The first Raman order is observed at 1641 nm for incident peak powers above 100 W, while the second order is observed at 1743 nm for peak incident powers in excess of 200 W. The third Raman order appears at an input peak power of 350 W at 1867 nm. Figure 3(b) illustrates the power evolution of the first two cascaded Raman orders in fiber 1. The figure shows that as the pump power is increased above 200 W up to 235 W, the second order increases significantly.

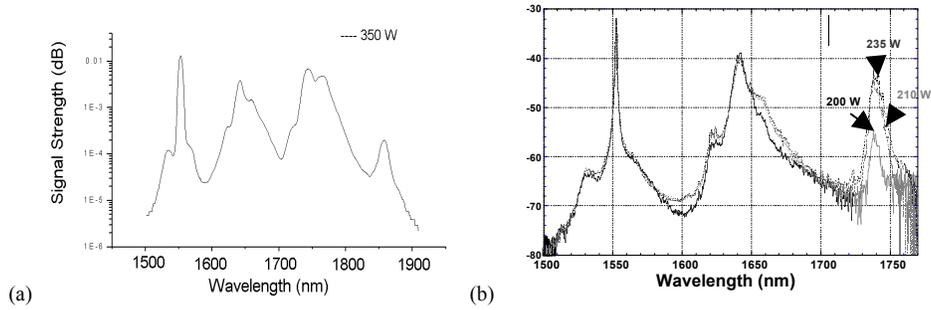


Fig. 3- (a) Generation of three orders of cascaded Raman in 12 m fiber 1 at 350 W peak incident power. (b) Evolution of first and second orders with power in fiber 1.

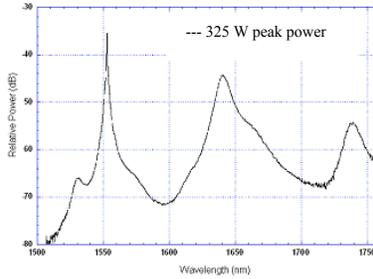


Fig. 4- Generation of two orders in 20 m fiber 2 at an input peak power of 325 W

We also observed cascaded shifting up to two orders in 20 m of fiber 2 at an input peak power of 325 W (Fig. 4). The higher power required for the generation of the second order in fiber 2 as compared to fiber 1 is attributed to the higher loss in the fiber as is evident from Fig. 1(a) and presence of possible scattering centers. In both the fibers, the first and second Raman orders occur at similar wavelengths. The wavelength of 1641 nm for the first and 1743 nm for the second Raman orders, correspond to a Stokes' shift of  $\Delta k = 345 \text{ cm}^{-1}$  and agrees with the value quoted in [4]. In the output spectrum we observe that the higher-order Raman peaks are asymmetric compared to the pump. Such asymmetry is due to the fact that shorter wavelength components pump the longer wavelength side. Also, the broadening of the higher orders is attributed to the line width of the Raman gain spectrum as depicted in Fig. 1(c).

In both the sulfide fibers, shifting beyond three orders is not observed as damage to the input end occurs consistently at peak incident powers above 350 W. For an effective core

diameter of  $\sim 33 \mu\text{m}^2$ , peak power in excess of 350 W correspond to an intensity of  $\sim 1 \text{ GW}/\text{cm}^2$  at the fiber end and corresponds to the damage threshold for the fiber reported in [5].

The threshold power of 100 W for the first Raman order in fiber 1 is used to estimate the  $g_R$  for the sulfide fiber. The relation between total gain,  $G_A$  and the  $g_R$  is given by, [8]:

$$G_A = \exp\left(\frac{g_R P_0 L_{\text{eff}}}{A_{\text{eff}}}\right), \quad (2)$$

where  $P_0$  is the peak pump power and  $A_{\text{eff}}$  is the effective area.  $L_{\text{eff}}$  is the effective length for an attenuation  $\alpha$ , which is given by

$$L_{\text{eff}} = \frac{1}{\alpha} (1 - e^{-\alpha L}) \quad (3)$$

The fiber 1 has a length of 12 m and a loss of  $\sim 1 \text{ dB}/\text{m}$  in the core as determined by using mode-stripping gallium at the ends of the fiber. The coupling efficiency into the core is  $\sim 70\%$  with a  $\pm 10\%$  variation. The Fresnel loss at the fiber end is  $\sim 17\%$  due to the refractive index mismatch between the sulfide fiber and air. The power in the first Raman order is assumed to correspond to a 90 dB Raman gain, at which the background noise grows to the pump level. Assuming random polarization of the pump in the fiber, we estimate a  $g_R$  of  $\sim 5.7 \times 10^{-12} \text{ m}/\text{W}$  for these fibers, which is  $\sim 89$  times larger than that of fused silica at  $1.55 \mu\text{m}$ .

Simulations are also performed to confirm the experimentally determined value for  $g_R$ . Using the model described in [12], evolution of the higher Raman orders are obtained by solving the system of non-linear boundary value problems for ordinary differential equations (BVODE) describing the slowly varying field approximation. In addition to the boundary conditions at the input end of the fiber, we also take into consideration various experimental constraints for fiber 1. Specifically, we take into account a 17% Fresnel reflection loss at the fiber output end and a coupling loss of 30%. For a linear loss of  $1 \text{ dB}/\text{m}$  in the fiber core, the  $L_{\text{eff}}$  of 12 m of fiber 1 is 4.1 m. The simulations are performed assuming a  $g_R$  90 times that of fused silica for the fiber medium for a cw pump signal at 1550 nm.

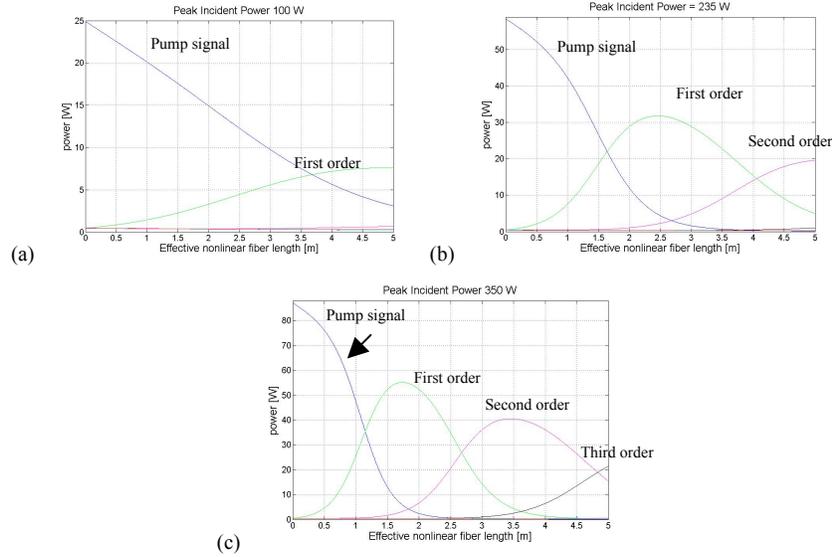


Fig. 5 (a)-(c): Theoretical calculation of pump and higher order evolution.

Simulation results for 5 m of  $L_{\text{eff}}$  are depicted in Figs. 5 (a)-(c). The plots illustrate the evolution of the first, second and third orders respectively against  $L_{\text{eff}}$  of the fiber. The incident power on the fiber end is specified to be the same as that in experiments. For a  $g_R$ , 90 times that of fused silica, plot 5(a) shows significant generation of the first Raman order in  $\sim 4$

m effective length of the fiber at an incident power of 100 W. Also, at input power of 200 W, simulations suggest appearance of the second Stokes order in about 4.5 m effective length of the fiber (Fig. 5(b)). Similarly, in Fig. 5(c), the third order starts to appear in about ~4 m of effective fiber length for a peak input power of 350 W, as observed experimentally. Thus, simulations for a fiber with  $g_R$  90 times that of fused silica correlate to the experimental results observed in the sulfide fiber 1. Notice that unlike in the experimental results, the simulations indicate that the pump gets completely depleted as light is shifted to higher orders, hence suggesting lower threshold powers for each order compared to those observed in the experiments. Such discrepancy arises due to the assumption of continuous wave pump in the simulations as against a quasi-cw pump (2 ns pulses) in the experimental set-up. Also, the fraction of the pump that leaks into the cladding while propagating along the length of the fiber does not contribute significantly to the Raman gain and remains at the pump wavelength.

#### 4. Discussion

The long ZDW wavelength of the sulfide fibers is responsible for the observation of SRS. In our experiments, by pumping with 2 ns pulses in the normal dispersion regime, MI phase matching is prevented and SC generation does not occur. Also, by using nanosecond pulses SC generation through SPM broadening is avoided [6]. Whether CRWS or SC is predominant in the fiber depends on the wavelength of the pump or the shifted pump with respect to the ZDW of the fiber [9]. There is some uncertainty in the deduced value for  $g_R$  in the sulfide fibers. One uncertainty in our experimental analysis arises from the nature of the loss occurring in the fiber core. In sulfide fibers, inevitable leakage of light into the cladding has been observed in our set-up and also in the past, [6]. Also in the case of fiber 2, we speculate the presence of scattering points along the length of the fiber. As a result, the loss inside the fiber may not be linear, and the  $L_{eff}$  of the fiber may be shorter than the estimated value of ~4.1 m. However, the estimated value of ~89 times that of fused silica for  $g_R$  in sulfide fibers, corresponds to similar values of ~73x, [2] and ~69x, [4] that of fused silica.

Consistent damage was observed at the input end of both fibers above 350 W of incident power, corresponding to an intensity of ~1 GW/cm<sup>2</sup>. Hence, although we expect several orders of CRWS extending into the mid-IR, generation of higher orders in the sulfide fibers is limited by the optical damage threshold for the fiber material. As a result, generation of light in the mid-IR using CRWS in sulfide fibers would require starting out with a Tm<sup>3+</sup>-doped cladding pumped fiber laser at 2.2  $\mu$ m. A wavelength shift corresponding to three Stokes orders implies light generation up to ~2.85  $\mu$ m for the sulfide fiber within the damage threshold limits. In order to achieve higher Stokes orders in these fibers, there is a need to understand the mechanism of the damage observed in these fibers. Techniques such as cooling the fiber, or encapsulating the fiber ends with heat sink material could potentially alleviate the low damage threshold for these fibers.

In summary, CRWS up to three Stokes orders is observed in sulfide fibers. Owing to the long zero dispersion wavelength of the fibers (~4.6  $\mu$ m), strong normal dispersion is observed in the telecommunication window. As a result, SRS is observed to be the dominant nonlinear effect for 2 ns pump pulses, resulting in cascaded orders. Wavelength shifting in excess of 300 nm using three orders is achieved from 1553 nm to 1867 nm. We also deduce a value for the  $g_R$  of the sulfide fiber based on the threshold power for the first order. A value of ~5.7x10<sup>-12</sup> m/W is obtained which is ~89 times that of fused silica at 1550 nm. Our estimation is also verified through numerical simulations. However, we observed only up to three cascaded Raman orders in 12 m length of the fiber due to the low damage threshold of the sulfide fibers, which is found to be ~1 GW/cm<sup>2</sup>. As a result, in spite of a high Raman gain coefficient, CRWS-based mid-IR light sources near 5  $\mu$ m would require pumping from an optical parametric amplifier at ~3.3  $\mu$ m in sulfide fibers.

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