

Article

Exact Solution of the Raman Response Function of Chalcogenide Fiber and Its Influence on the Mid-Infrared Supercontinuum

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Abstract: We fabricated a core-cladding Ge–Sb–Se glass fiber with a Ge_{12.5}Sb₁₅Se_{72.5} core and Ge₁₅Sb₁₀Se₇₅ cladding, achieved a supercontinuum spectrum spanning from 2 μm to 9 μm by pumping the Ge–Sb–Se fiber with a core diameter of 11 μm using a femtosecond laser pump at 3.8 μm, and numerically simulated the supercontinuum generation using the generalized nonlinear Schrödinger equation. In particular, we investigate the effect of the different Raman response functions that were calculated using the traditional single Lorentzian model and a multiple vibrational mode model on the evolution of the supercontinuum by comparing the supercontinua obtained from simulation and experimental results. We demonstrate that the Raman response function generated by the multiple vibrational mode model captures the actual response behavior of the material, and the supercontinuum generated using this model has more accuracy. To the best of our knowledge, this is the first reported study on supercontinuum generation in Ge–Sb–Se fiber utilizing a Raman response function calculated using the multiple vibrational mode model. This significant advancement enables more accurate simulation of supercontinuum generation in fibers with a multi-peaked structured Raman gain spectrum and holds great potential for optimizing the performance of various mid-infrared supercontinuum sources.

Keywords: chalcogenide; Raman response function; mid-infrared; supercontinuum

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1. Introduction

Supercontinuum (SC) generation is a nonlinear phenomenon characterized by dramatic spectral broadening of intense light pulses passing through a nonlinear material due to the combined effects of chromatic dispersion and various nonlinear processes [1]. SC light sources possess numerous advantages, including a large bandwidth, brightness, high coherence, and a multi-octave-spanning spectrum. Consequently, they have achieved remarkable success in scientific research and find applications in various fields, such as national defense security [2], biomedicine [3], fluorescence imaging [4], and spectroscopy [5].

The mid-infrared (MIR) SC has drawn much attention in recent years because it covers two important atmospheric windows at 3 μm to 5 μm and 8 μm to 12 μm where most molecules display fundamental vibrational absorption, leaving distinctive spectral fingerprints [6]. MIR fiber laser source host materials primarily include fluoride, heavy metal oxides, and chalcogenide (CHG) glasses. For example, Eslami et al. [7] reported octave-spanning SC generation from 1.2 μm to 2.5 μm with 600 mW average power in a short length of a multimode fluoride fiber with a 100 μm core diameter due to the main contribution of higher-order modes. Le et al. [8] achieved all-normal dispersion (ANDi) of a lead-bismuth-gallate glass solid-core photonic crystal fiber (PCF) and SC generation

in the wavelength range of 0.9 μm to 2.5 μm pumped using low peak power. Ghosh et al. [9] reported the design and fabrication of an ultra-high birefringence up to 10^{-2} polarization-maintaining $\text{As}_{38}\text{Se}_{62}$ tapered PCF with 3 rings of air holes in the cladding and experimentally generated MIR SC from 3.1 μm to 6.02 μm . Recently, the use of non-glass fibers in MIR SC generation has attracted much interest. Adamu et al. [10] achieved a 4.3-octave-wide spectrum from 0.2 μm to 4 μm in a gas-filled hollow-core antiresonant fiber that consists of a hollow core surrounded by seven non-touching silica capillaries with a wall thickness of ~ 0.64 μm , forming a core with a diameter of ~ 44 μm . Van et al. [11] demonstrated that it was possible to optimize the dispersion characteristics of the CHG fibers for SC generation by filling the selected liquids into cladding air holes and studied the tapered fibers for broad SC generation. SC generation from selective filling of MOFs has been experimentally demonstrated using water [12], carbon tetrachloride [13], and toluene [14]. It can improve the nonlinearity and optimize the dispersion characteristics of fibers generated using this method, which provides an alternative approach to the study of broadband SC generation.

CHG glasses have numerous desirable and promising optical properties, such as a high value of nonlinear refractive index, high damage threshold, low phonon energy, and excellent crystalline quality, making them excellent candidates for MIR SC generation [15]. Arsenic-containing glasses are the most used host materials in CHG fibers, and numerous studies have been conducted on improving the spectral width, coherence, and engineering of optical fiber waveguides. For example, Cheng et al. [16] experimentally demonstrated SC spanning from ~ 2.0 μm to 15.1 μm using a 3 cm long CHG step-index fiber (SIF) by pumping it at 9.8 μm with a peak power of ~ 2.89 MW. Islam et al. [17] reported a three-hole suspended-core fiber (SCF) that used AsSe_2 as background material. The proposed SCF obtained a zero dispersion wavelength (ZDW) at 2.55 μm , and a broadband SC extending from 1 μm to 14 μm was achieved by pumping 10 kW peak power and 50 fs duration laser pulses at 2.56 μm . Li et al. [18] reported 1.5 μm to 8.3 μm broadband SC generation with high coherence property from 15 cm long $\text{As}_2\text{S}_3/\text{As}_{38}\text{Se}_{62}$ tapered optical fiber (TOF) pumped at 3.75 μm by controlling the waist core diameter using the homemade tapering platform to realize the ANDi characteristic of fibers. Compared with conventional As-S and As-Se glasses, Ge-Sb-Se glasses exhibit enhanced thermal and mechanical durability, a higher Raman gain coefficient, increased optical nonlinearity, an ultrafast optical response, a superior infrared transmission range, and excellent infrared transmittance [19]. Furthermore, the substitution of highly toxic As with Sb in Ge-Sb-Se glasses contributes to their reduced toxicity and improved environmental friendliness. Ge-Sb-Se glasses have gained significant attention in recent years. Ou et al. [20] reported a high nonlinear $\text{Ge}_{15}\text{Sb}_{25}\text{Se}_{60}/\text{Ge}_{15}\text{Sb}_{20}\text{Se}_{65}$ SIF with a high damage threshold of 3674 GW/cm², and the SC from 1.8 μm to 14.0 μm was obtained by optical parametric amplification (OPA). Medjouri et al. [21] designed an all-solid PCF with ANDi profile using two compatible CHGs, namely $\text{Ge}_{15}\text{Sb}_{15}\text{Se}_{70}$ and $\text{Ge}_{20}\text{Se}_{80}$, which were used as background material and for solid rods, respectively, by pumping 50 fs duration laser pulses with a total energy of 900 pJ at 3 μm , and a broadband SC with ~ 5 dB bandwidth spanning from 1.6 μm to 7.0 μm was generated in 1 cm long PCF.

The generation of SC in fibers involves the interaction between numerous linear and nonlinear effects and is a very complex physical process, resulting from the interplay of a number of effects, such as self-phase modulation (SPM), cross-phase modulation (XPM), four wave mixing (FWM), stimulated Raman scattering (SRS), and soliton fission [22–25]. It is worth noting that SRS is an important nonlinear process in SC generation. When the pump laser intensity is sufficient, the high frequency components act as a pump source for lower frequencies, which is the main cause of significant broadening of the SC in the long wavelength region [26]. SRS is closely related to the Raman gain coefficient and the Raman response function. Therefore, an accurate Raman response function is of great significance for accurately simulating SC generation. The Raman gain spectrum of fused silica is commonly represented by a single Lorentzian function, and the corresponding

Raman response function becomes a simple damped harmonic oscillation [27]. The Raman gain spectra and Raman response functions of As_2S_3 and As_2Se_3 show a similar form to that of fused silica [28]. However, the Raman gain spectrum of CHG glasses, such as Ge–As–Se, Ge–Sb–Se, and Ga–Sb–S, have a multi-peaked structure, and the Raman response function calculated using the conventional single Lorentzian model differs significantly from the actual situation, leading to a substantial impact on the accurate simulation of SC and Raman laser output. Unfortunately, some of the literature has overlooked this issue. For example, Vays et al. [29] report broadband SC generation through a flat dispersion profile of the $\text{Ge}_{11.5}\text{As}_{24}\text{Se}_{64.5}$ PCF, and the Raman response function of $\text{Ge}_{11.5}\text{As}_{24}\text{Se}_{64.5}$ material is directly represented by Lorentzian spectral profile. This not only reduces the accuracy of the Raman response function but also affects the subsequent SC generation. Furthermore, certain studies misapplies the Raman response function of As_2S_3 and As_2Se_3 . For instance, Medjouri et al. [30] directly used the Raman response function of As_2Se_3 as a replacement for $\text{GaSb}_{32}\text{S}_{60}$ material in their study of broadband coherent SC in ANDi fiber.

Currently, there are only a few reports on the Raman response function of CHG materials with a multi-peaked structure Raman gain spectrum, and the use of a traditional single Lorentzian model can significantly impact the accuracy of the Raman response function. In this study, Ge–Sb–Se fiber was experimentally prepared and Raman spectra, refractive index, and other material parameters were obtained. According to the multiple vibrational mode model that was proposed by Dawn based on previous spectroscopic data and an earlier proposed model [31], we establish a multi-vibration model in Ge–Sb–Se fiber for the first time to solve the Raman response function. The Raman gain spectrum obtained by performing a Fourier inverse transform of the Raman response function, which is calculated using this model, exhibits remarkable agreement with the Raman gain spectrum derived directly from the Raman spectra. This robust correspondence serves as compelling evidence for the effectiveness of our proposed model. The Raman response function generated by the multiple vibrational mode model and single Lorentzian model are brought into the generalized nonlinear Schrödinger equation (GNLSE) to simulate SC generation, and the results showed that the SC calculated using the Raman response function obtained from the multi-vibration model exhibited a better fit with the experimental results.

This multi-vibration model can also be applied to other materials with a multi-peaked structure Raman gain spectrum. It provides a more accurate solution for the Raman response function based on material characteristics and serves as an important basis for more precise simulation and optimization of SC and Raman laser output.

2. Experimental

The ChG SIF used in this work had a core made of $\text{Ge}_{12.5}\text{Sb}_{15}\text{Se}_{72.5}$ glass and a cladding made of $\text{Ge}_{15}\text{Sb}_{10}\text{Se}_{75}$ glass; both had sufficient thermal compatibility so that the components could be drawn down in the same fiber. A vacuumed melt quenching technique was used to synthesize the glass samples with high-purity elemental raw materials of Ge, Sb, Se (99.999%), similar to that previously reported [32]. SC generation was measured further based on the Ge–Sb–Se fiber. The experimental set-up is depicted in Figure 1. A tunable MIR OPA system (~150 fs full-width at half maximum (FWHM), 1 kHz repetition rate, and 2–10 μm tunable wavelength) was used as an exciting source. A pair of polarizers was used to control the output power of the pump, and the beam from the OPA was coupled to the fiber via a calcium fluoride lens with a focal length of a 75 mm. The output SC from the fiber was collected into the input slit of a monochromator, and the spectra were measured by an infrared spectrometer (Infrared systems, FPAS, Winter Park, FL, USA) with a liquid nitrogen-cooled HgCdTe (MCT) detector (1–16 μm wavelength range). Furthermore, the refractive indices were measured using an infrared spectroscopy ellipsometer (J.A. Woollam IR-Vase II, Lincoln, NE, USA). The Raman spectra of the samples were obtained in the range of 25–400 cm^{-1} using a Raman

spectroscopy (Renishaw InVia, Renishaw, Gloucestershire, UK). All these measurements were carried out at room temperature.

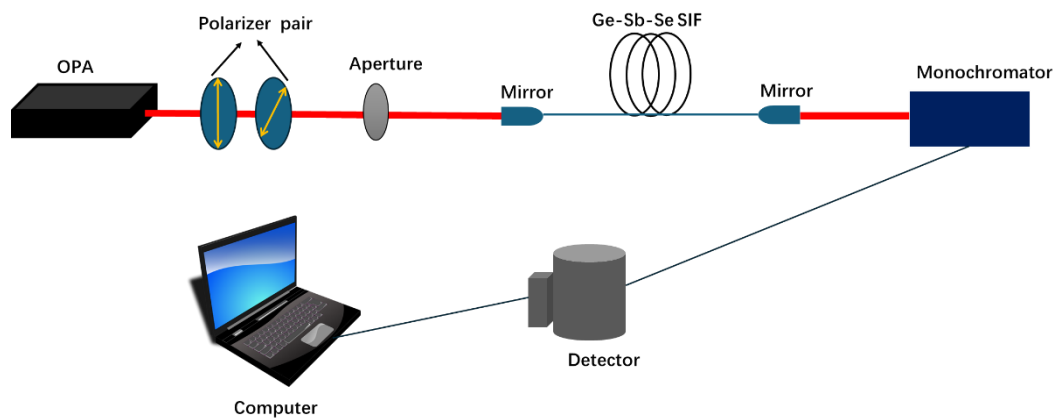


Figure 1. Experiment setup for SC generation in the Ge–Sb–Se SIF.

3. Theoretical Model

3.1. Numerical Simulation

The transmission of light waves in an optical fiber can be expressed by Maxwell's equations. In general, a slowly varying envelope is used to approximate the value of the rapidly varying component in an electric field for further simplification. By introducing a delay coordinate system T for moving the group velocity of the pulse, the optical envelope is expressed using the GNLSE as follows [33]:

$$\frac{\partial A(z, T)}{\partial z} = -\frac{\alpha}{2} A(z, T) + \sum_{k \geq 2} \frac{i^{k+1}}{k!} \beta_k \frac{\partial^k A(z, T)}{\partial T^k} + i\gamma \left(1 + \frac{i}{\omega_0} \frac{\partial}{\partial T} \right) \left[A(z, T) \int_{-\infty}^{\infty} R(T') |A(z, T - T')|^2 dT' \right] \quad (1)$$

where $A(z, T)$ is the optical field envelope, and α denotes the loss coefficient. The entire right-hand second term is a wavelength dispersion effect that includes the high-order dispersion. The last term includes the nonlinear effects, and $R(t)$ is the nonlinear response function that consists of two parts $f_R h_R$ and $(1 - f_R) \delta(t)$, which indicate instantaneous electronic and delayed Raman contributions, respectively. Here, h_R is the Raman response function, and f_R represents the Raman fractional contribution to the overall nonlinear response. In addition, $\delta(t)$ is the Dirac delta function.

3.2. The Raman Response Function Solved Using the Multiple Vibrational Mode Model

Raman-induced changes in the spectrum and temporal envelope of an ultrashort pulse are governed by the time-domain Raman response function of the material. Previous researchers have made significant contributions to the development of models for the Raman response function. In 1989, Blow et al. [34] reported the Raman response function expression of fused silica by approximating the Raman gain coefficient spectrum as single Lorentzian function, wherein a simple damped harmonic oscillation can be expressed by Equation (2).

$$h_R(t) = \frac{\tau_1^2 + \tau_2^2}{\tau_1 \tau_2^2} \exp(-t/\tau_2) \sin(t/\tau_1) \quad (2)$$

Here, $\tau_1 = 12.2$ fs, and $\tau_2 = 32$ fs. This model uses three parameters to provide the correct location and peak value of the dominant peak in the Raman gain spectrum. Because of its

simplicity, it has a great fit with the experimental result within 100 fs, and this simple model has been widely adopted by other researchers [35–37]. However, the Raman response function in Equation (2) only considers the main peak of the Raman gain spectrum of fused silica and ignores the influence of other small Raman gain peaks; it does not provide a correct quantitative description of Raman-induced phenomena and leads to difficulty in comparing theoretical and experimental results. To address this limitation, Dawn et al. [20] proposed a multiple vibrational mode model. The Raman gain spectrum of the materials were regarded as having a multi-peaked structure, and multiple Gaussian functions were used to reproduce the Raman gain spectrum. The model was adapted an intermediate-broadening model using Lorentzian and Gaussian convolutions, as shown in Equation (3).

$$h_R(t) = \sum_{i=1}^n \frac{A_i}{\omega_{v,i}} \exp(-\gamma_i t) \exp(-\Gamma_i^2 t^2 / 4) \sin(\omega_{v,i} t) \quad (3)$$

Here, A_i is the amplitude of the i th vibrational mode, and $\omega_{v,i}$, γ_i , and Γ_i are related to the Gaussian component position, Lorentzian FWHM, and Gaussian FWHM, respectively. Figure 2 demonstrates the comparison of Raman response functions obtained from the single Lorentzian model [38] and the multiple vibrational mode model [31] as well as using the experimental results provided in Ref. [39] for fused silica. It is evident that the Raman response function generated using the single Lorentzian model gradually tends to 0 after 150 fs, whereas the Raman response function generated using the multiple vibrational mode model consistently matches the experimental results.

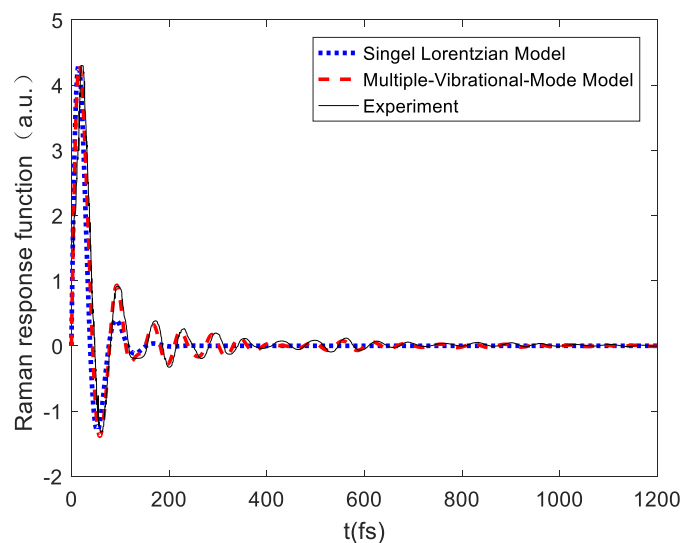


Figure 2. The comparison of Raman response functions of fused silica. The blue curve is the single Lorentzian model [38], the red curve is the multiple-vibrational-mode model [31], and the black curve is the experimental result [39].

4. Results and Discussion

4.1. The Raman Response Function of Ge–Sb–Se Glass

According to previous work of the researchers in this paper, when fused silica is used as a reference standard, the Raman gain spectrum of Ge–Sb–Se or other glasses can be calculated by comparing spontaneous Raman spectra measured under the same experimental conditions, as shown in Equation (4) [40].

$$\frac{g_{RG\beta-sample}^{k,\gamma}(\omega_p - \Omega_{\beta-sample}^k)}{g_{RG\beta-sio2}^{k,\gamma}(\omega_p - \Omega_{\beta-sio2}^k)} = \frac{K_{GR-sample}^{k,\gamma} K_{sR-sio2}^{k,\gamma}}{K_{sR-sample}^{k,\gamma} K_{GR-sio2}^{k,\gamma}} \cdot \frac{(\omega_p - \Omega_{\beta-sio2}^k)^3 n_{sio2}(\omega_p) n_{sample}(\omega_p - \Omega_{\beta-sample}^k)}{(\omega_p - \Omega_{\beta-sample}^k)^3 n_{sample}(\omega_p) n_{sio2}(\omega_p - \Omega_{\beta-sio2}^k)} \cdot \frac{[1 - R(\omega_{p-sio2})][1 - R(\omega_p - \Omega_{\beta-sio2}^k)]}{[1 - R(\omega_{p-sample})][1 - R(\omega_p - \Omega_{\beta-sample}^k)]} \cdot \frac{I_{\beta-sample}^{k,\gamma}(\omega_p - \Omega_{\beta-sample}^k)}{I_{\beta-sio2}^{k,\gamma}(\omega_p - \Omega_{\beta-sio2}^k)} \quad (4)$$

where $\Omega_{\beta}^k \approx \Omega = \omega_p - \omega_s$ is the frequency shift of the Raman peak from the laser frequency ω_p , and $R(\omega) = \frac{[n(\omega) - 1]^2}{[n(\omega) + 1]^2}$ is the reflectance coefficient at normal incidence.

The parameters $K_{sR}^{k,\gamma}$ and $K_{GR}^{k,\gamma}$ are constants that contain all the phonon and electromagnetic constant parameters, and the difference between them is $K_{sR}^{k,\gamma}$, which contains the Bose–Einstein correction factor $F_{BE}(v, T) = 1 + [\exp(hv/kT) - 1]^{-1}$ (T , h and k refer to temperature, Planck constant, and Boltzmann constant, respectively) that accounts for the thermal statistical fluctuation for each individual mode with an energy kT . From Equation (4), it is evident that the ratio of the Raman gain coefficient of other materials to that of fused silica can be determined by comparing the ratio of their respective refractive indices, reflectivity coefficients, Raman spectral intensities, and Raman frequency shifts. Generally, it is widely acknowledged in the field that the Raman gain coefficient of fused silica remains constant when the pump wavelength is held constant. If we employ fused silica as the reference standard, the Raman gain coefficients of Ge–Sb–Se or other glass materials can be determined through a comparative analysis of spontaneous Raman spectra acquired under identical experimental conditions. By leveraging this approach, we not only ensure precise calculations but also establish a robust framework for assessing and contrasting the Raman gain coefficients of various glass compositions.

The Raman gain spectrum of $\text{Ge}_{12.5}\text{Sb}_{15}\text{Se}_{72.5}$ glass obtained using this method is shown in Figure 3. Here, the dashed line represents the spectrum modeled using seven Gaussian functions, and each Gaussian peak corresponds to a vibrational mode of a unit composing the glass structure. Table 1 provides the parameters used in the multiple vibrational mode model, and the ratio of the Gaussian linewidth to the Lorentzian linewidth is 3.0 in this work.

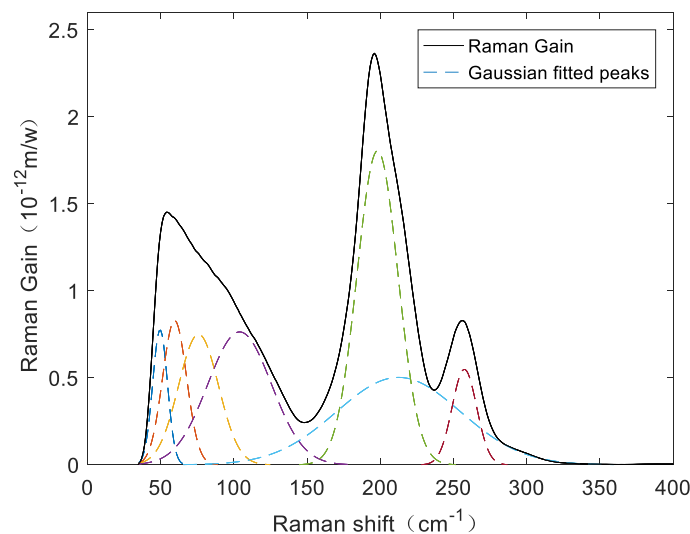
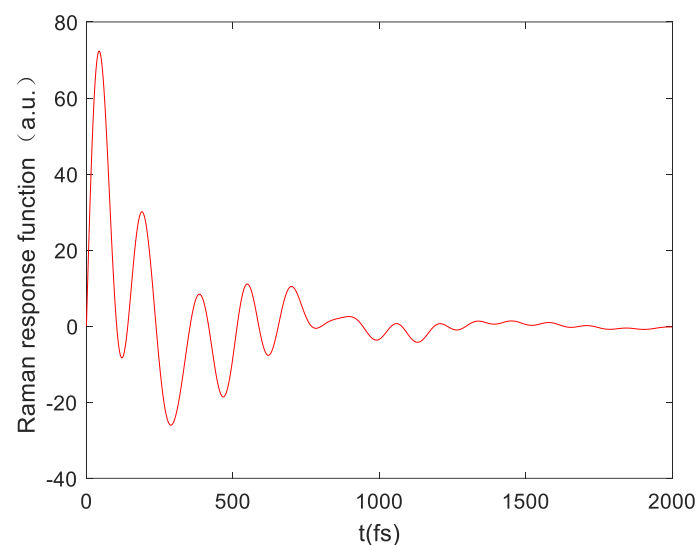


Figure 3. The Raman gain spectrum of $\text{Ge}_{12.5}\text{Sb}_{15}\text{Se}_{72.5}$ glass calculated using Raman spectra (solid line) and seven different Gaussian functions (dashed line).

Table 1. Values of the parameters used in the multiple-vibrational-mode model.

Mode Number <i>i</i>	Component Position (cm ⁻¹)	Peak intensity $A_i = A_i'/\omega_{v,i}$ (10 ⁻¹² m/w/THz)	Gaussian FWHM (cm ⁻¹)	Lorentzian FWHM (cm ⁻¹)
1	49.63	4.14	10.07	3.36
2	59.55	7.20	16.35	5.54
3	75.90	10.86	27.30	9.10
4	103.96	17.61	43.48	14.49
5	198.10	26.79	27.99	9.33
6	212.44	22.58	84.78	28.26
7	257.42	4.87	16.74	5.58

Based on Equation (3) and data in Table 1, the Raman response function of Ge_{12.5}Sb₁₅Se_{72.5} glass is depicted in Figure 4. It is noteworthy that the Raman response function of Ge_{12.5}Sb₁₅Se_{72.5} glass is characterized by a sum of multiple damped harmonic oscillations, in contrast to the simple damped harmonic oscillation observed in the single Lorentzian structure Raman gain materials. In addition, the presence of multiple peaks in the Raman gain spectrum suggests the contribution of multiple vibrational modes to the Raman gain, thereby supporting the superiority of the multiple vibrational mode model in accurately representing the actual response behavior of Ge_{12.5}Sb₁₅Se_{72.5} glass.

**Figure 4.** The Raman response function of Ge_{12.5}Sb₁₅Se_{72.5} glass calculated using the multiple-vibrational-mode model.

The Raman gain spectrum is proportional to the imaginary part of the transfer function as reported by Agrawal [41], which is given as follows.

$$g_R(\omega) = \sum_{i=1}^N \frac{A_i}{2} \int_0^{\infty} \left\{ \cos[(\omega_{v,i} - \omega)t] - \cos[(\omega_{v,i} + \omega)t] \right\} \exp(-\gamma_i t) \exp(-\Gamma_i^2 t^2 / 4) dt \quad (5)$$

There are no reports on experimental or accurate theoretical simulations about the Raman response function of Ge_{12.5}Sb₁₅Se_{72.5} glass. Therefore, to verify the accuracy of the Raman response function solved using the multiple vibrational mode model, we inversely deduce the Raman gain spectrum using Equation (5) and compare it with the experimentally measured data obtained from Equation (4), and the result is shown in Figure 5.

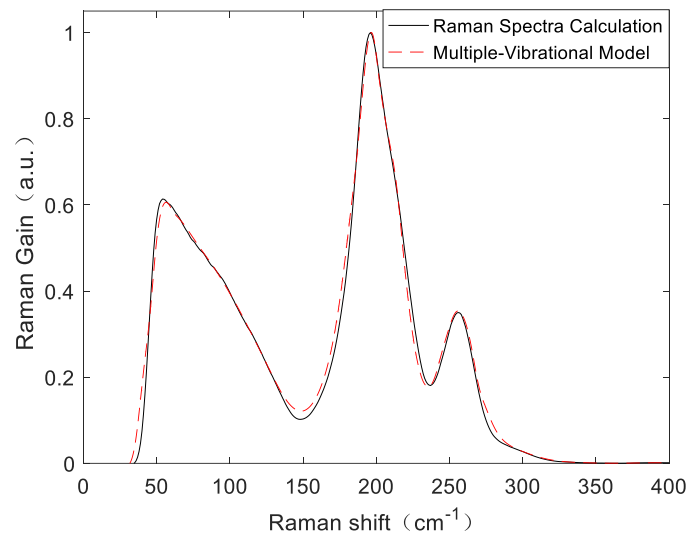


Figure 5. The Raman gain spectrum obtained from the multiple-vibrational-mode model (red) and the Raman spectra (black).

Figure 5 demonstrates that the curve profiles generated using the multiple vibrational mode model and the Raman spectra are consistent with only a small difference observed around 150 cm⁻¹. Hence, we conclude that the multiple vibrational mode model is plausible. The Raman fraction can be obtained from the Raman response function using Equation (6) [42].

$$g_R(\omega) = \frac{2\omega_0}{c} n_2 f_R \text{Im}[\tilde{h}_R(\omega)] \quad (6)$$

Here, the transfer function $\tilde{h}_R(\omega)$ is the Fourier transform of the Raman response function, ω_0 is the center frequency, c is the speed of light in vacuum, and n_2 is the nonlinear refractive index of the Ge_{12.5}Sb₁₅Se_{72.5} glass. The Raman fraction f_R of the SIF fiber is obtained as 0.4615.

4.2. Other Parameters Needed in SC Simulation

In order to further validate the influence of an accurate Raman response function on the precise simulation of supercontinuum, this study compares the SC obtained using two different methods, namely, the single Lorentzian model and the multiple vibrational mode model, with experimentally measured results. Consequently, an experimental setup was prepared to generate SC using a Ge–Sb–Se SIF with a 11 μm core diameter and a fixed core-cladding ratio of 11/305. The parameters required for SC simulation, such as refractive index, dispersion, effective model area and nonlinear coefficient, were calculated using Equation (1). Additionally, the fiber's loss coefficient was determined using a cut-back method, the loss spectrum of the Ge–Sb–Se SIF as shown in Figure 6. As noted in the figure, the Ge–Sb–Se SIF shows a lower loss of 1.62 dB/m at 6 μm. The absorption peaks centered at 2.86 and 4.5 μm, corresponding to O–H and Se–H bonds, respectively, are still present evidently due to pollution by oxygen or water molecules; therefore, we selected 3.8 μm as the pumping wavelength for our experiments in this study.

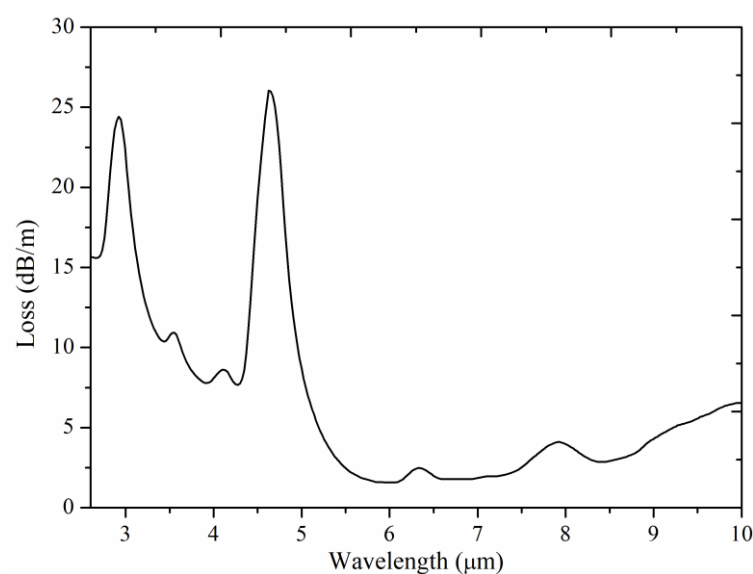


Figure 6. The loss spectrum of the Ge–Sb–Se SIF measured using the cut-back method.

The wavelength-dependent linear refractive indices of the CHG glasses used for the design of the proposed fiber are expressed using the Sellmeier dispersive formula as follows [43]:

$$n^2 = 1 + \frac{B_1 \lambda^2}{\lambda^2 - C_1} + \frac{B_2 \lambda^2}{\lambda^2 - C_2} \quad (7)$$

where B_1 and B_2 are dimensionless coefficients, and C_1 and C_2 are related to the glass resonance wavelengths [44]. Table 2 provides the different values of the Sellmeier coefficients, and Figure 7 depicts the variation of the linear refractive index with wavelengths for both $\text{Ge}_{12.5}\text{Sb}_{15}\text{Se}_{72.5}$ and $\text{Ge}_{15}\text{Sb}_{10}\text{Se}_{75}$ ChG glasses.

Table 2. Sellmeier coefficients for $\text{Ge}_{12.5}\text{Sb}_{15}\text{Se}_{72.5}$ and $\text{Ge}_{15}\text{Sb}_{10}\text{Se}_{75}$ CHG glasses.

$\text{Ge}_{12.5}\text{Sb}_{15}\text{Se}_{72.5}$		$\text{Ge}_{15}\text{Sb}_{10}\text{Se}_{75}$	
$B_{1,2}$	$C_{1,2}$	$B_{1,2}$	$C_{1,2}$
45.2485	36530.3824	5.6454	0.0530
6.1319	0.0276	1.5523	2287.1588

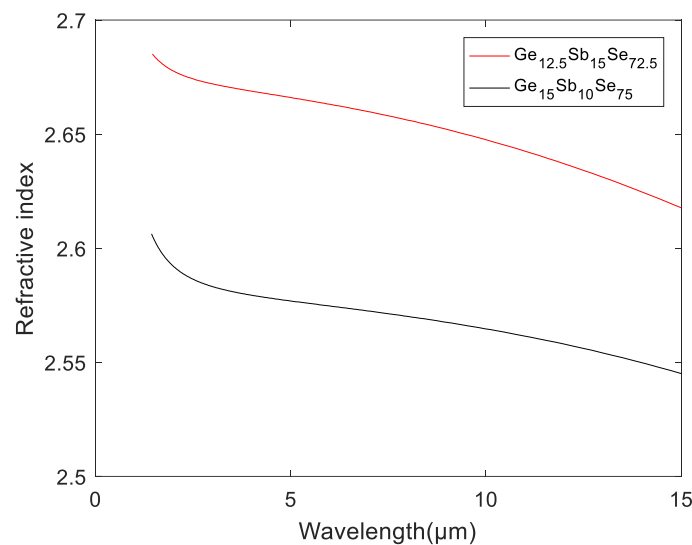


Figure 7. Variation in the $\text{Ge}_{12.5}\text{Sb}_{15}\text{Se}_{72.5}$ and $\text{Ge}_{15}\text{Sb}_{10}\text{Se}_{75}$ CHG glasses refractive indices with wavelengths.

During the SC generation process, the impact of dispersion outweighs the impact of loss. The higher-order dispersion obtained by the propagation constant at any frequency relative to the pulse central frequency can be expanded as the Taylor series expansion as noted in Equation (8) [45].

$$\beta(\omega) = \beta_0 + \beta_1(\omega - \omega_0) + \frac{1}{2!}\beta_2(\omega - \omega_0)^2 + \frac{1}{3!}\beta_3(\omega - \omega_0)^3 + \dots \quad (8)$$

Here $\beta_k = \left(\frac{d^k \beta(\omega)}{d\omega^k} \right)_{\omega=\omega_0}$, and the variable k takes values up to 10 in this study. The

β_2 in Equation (8) corresponds to the group velocity dispersion (GVD). It determines the extent to which the different spectral components of the ultrashort pulse propagate at different phase velocities in the SIF, resulting in these different frequency components having arrived at the output at different times. This feature is recognized as the main factor contributing to spectral broadening. To quantitatively represent GVD, the dispersion coefficient D is commonly incorporated into the calculation process, as illustrated in Equation (9) [46].

$$D(\lambda) = -\frac{\lambda}{c} \frac{d^2 \text{Re}(n_{\text{eff}})}{d\lambda^2} = -\frac{2\pi c}{\lambda^2} \beta_2 \quad (9)$$

Here, $\text{Re}(n_{\text{eff}})$ represents the real part of the effective refractive index, and c is the speed of light. In order to accurately calculate dispersion, it is essential to determine the effective refractive index of the fiber. As the effective refractive index is related to refractive index of the fiber's core and cladding as well as the waveguide structure of the fiber, we simulated our design to calculate the refractive index of the core and cladding of Ge–Sb–Se SIF in Figure 7. The dispersion curve of Equation (9) is shown in Figure 8.

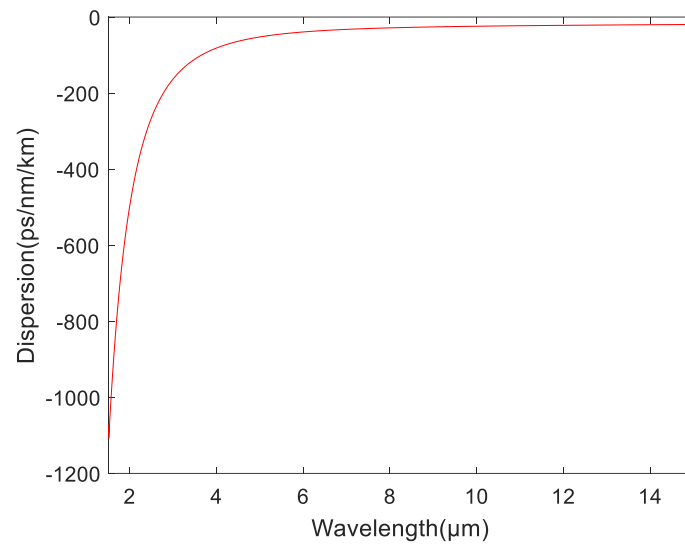


Figure 8. Variation of the dispersion profile with wavelengths for Ge-Sb-Se SIF.

The investigation of nonlinear effects in fibers is a highly intricate task, as it necessitates a comprehensive analysis of the contributions of each effect to the process of spectral broadening. This complexity is further compounded by the interactions among various effects. The nonlinear coefficient is typically denoted as demonstrated in Equation (10) [47]:

$$\gamma(\lambda) = \frac{2\pi n_2}{\lambda A_{\text{eff}}} \quad (10)$$

where A_{eff} denotes the effective mode area of the optical propagating into the fiber, and n_2 is the nonlinear refractive index. The A_{eff} is shown in Figure 9; it demonstrates an increasing trend as the wavelength increases. Specifically, when the wavelength extends from 3 μm to 9 μm , there is a corresponding increase in the effective mode field area from 52.7 μm^2 to 81.1 μm^2 . The nonlinear coefficient used in the simulation of this study is 0.875/W/m at a pump wavelength of 3.8 μm .

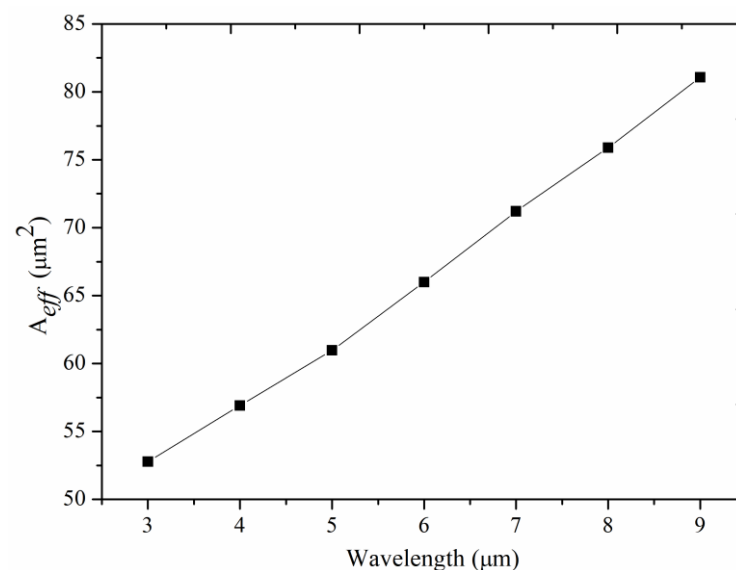


Figure 9. Variation in the effective mode area with wavelengths for Ge-Sb-Se SIF.

4.3. Comparison of SC Calculated Using Different Methods with Experimental Results

Figure 10 presents a comparative analysis between the SC generated by laser pulses at $3.8\ \mu\text{m}$ with a peak power of 500 kW and a duration of 150 fs and the corresponding experimental results obtained under identical conditions. In Figure 10a, the Raman response function utilized is generated using the multiple vibrational mode model, whereas in Figure 10b, a simple damped harmonic oscillation based on the single Lorentzian model reported in Ref. [21] is employed. The observed disparity between the SC generated using the Raman response function is based on the simple damped harmonic oscillation and the experimental result and is particularly noted in the longer wavelength range. Conversely, the SC generated using the Raman response function derived from the multiple vibrational mode model exhibits a remarkable agreement with the experimental results across the entire range of wavelengths. Figure 10c,d shows the spectral evolution of the simulation in Figure 10a,b, respectively. This outcome highlights the significant impact of the Raman response function on the SC generation process and serves as compelling evidence that the Raman response function obtained through the multiple vibrational mode model offers superior accuracy when compared to the single Lorentzian model. Consequently, this model presents a more precise approach for studying and simulating materials characterized by a multi-peaked Raman gain spectrum, which holds significant potential for enhancing the performance and output characteristics of Raman lasers as well as for facilitating the design and evaluation of fiber amplifiers.

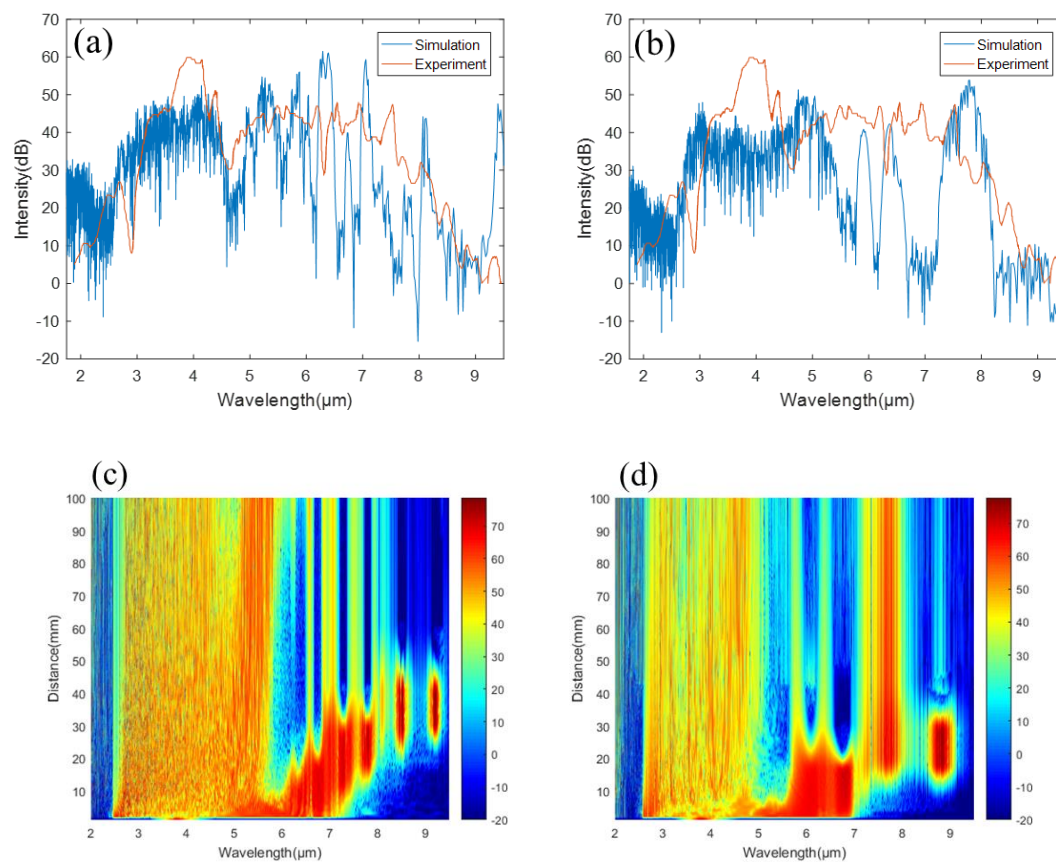


Figure 10. (a) Comparison of SC generated by simulation of Raman response function using the multiple-vibrational-mode model with experimental results. (b) Comparison of SC generated by simulation of Raman response function generated using the single Lorentzian model with experimental results. (c,d) represent the simulated SC spectral evolution of (a,b).

5. Conclusions

In this paper, we fabricated a type of Core-cladding Ge–Sb–Se glass fiber with a $\text{Ge}_{12.5}\text{Sb}_{15}\text{Se}_{72.5}$ core and $\text{Ge}_{15}\text{Sb}_{10}\text{Se}_{75}$ cladding and achieved a SC spectrum spanning from 2 μm to 9 μm by pumping the Ge–Sb–Se fiber with a core diameter of 11 μm using a femtosecond laser pump at 3.8 μm . We present the Raman response function of Ge–Sb–Se glasses calculated using a multiple vibrational mode model. Furthermore, we calculate the Raman gain spectrum using the Raman response function and compare it with the Raman gain spectrum obtained from experimentally measured Raman spectra. This comparative analysis serves to verify the accuracy of this model. The simulation of SC generation is performed based on the Raman response functions obtained from both the multiple vibrational mode model and the single Lorentzian model, incorporating relevant parameters, such as dispersion and nonlinearity coefficients. A comparison between the simulated SC and the experimental SC reveals that the multiple vibrational mode model exhibits a superior fit with the experimental data. This finding underscores the influence of the Raman response function on the generation of SC and highlights the critical importance of an accurate Raman response function for precise simulation of SC generation. To the best of our knowledge, this study represents the first report on Ge–Sb–Se fiber SC generation utilizing the Raman response function obtained with a multiple vibrational mode model. Moreover, we assert that this model can be applied to other materials characterized by a multi-peaked Raman gain spectrum. By accurately capturing the material-specific characteristics, this model provides a foundation for more precise simulation and optimization of SC generation as well as Raman laser output.

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