Engineered Raman Lasing in Photonic Integrated Chalcogenide Microresonators

Di Xia, Yufei Huang, Bin Zhang,* Pingyang Zeng, Jiaxin Zhao, Zelin Yang, Suwan Sun, Liyang Luo, Guiying Hu, Dong Liu, Zifu Wang, Yufei Li, Hairun Guo, and Zhaohui Li*

Photonic integrated Raman lasers have extended the wavelength range of chip-scale laser sources and have enabled applications including molecular spectroscopy, environmental analysis, and biological detection. Yet, the performance is strongly determined by the pumping condition and Raman shift value of nonlinear medias, leaving challenges to have a widely and continuously tunable Raman laser (e.g., over 100 nm). Here, photonic engineered Raman lasers based on chip-integrated chalcogenide microresonators are demonstrated. The home-developed chalcogenide photonic platform is of high nonlinearity, wide transparency, and low loss. The strong and broadband material Raman response has promised rich dynamics of Raman lasing. Indeed, both single-mode Raman lasing and a broadband Raman-Kerr comb, which are found engineered by tuning the dispersion of the chalcogenide microresonator, are demonstrated. The single-mode Raman laser, together with its cascaded modes, supports a gap-free tuning range over 140 nm, while the threshold power is as low as 3.25 mW. The results may contribute to the understanding of Raman and Kerr nonlinear interactions in dissipative and nonlinear microresonators, and on application aspect, may pave a way to integrated and efficient laser sources that is desired in spectroscopic applications in the infrared.

1. Introduction

Stimulated Raman scattering (SRS) is well known as an effective means to extend the spectral coverage of conventional semiconductor and rare-earth-doped laser sources,[1–4] in which a longer-wavelength signal (i.e., the Stokes radiation) can be generated from the initial pump wave, with a frequency offset equal to the molecular vibrational frequency underlying the material.[5,6] Recent development of Raman lasers based on photonic integrated microresonators have been successfully achieved with continuous-wave (CW) pumping and low power thresholds. Indeed, photonic integrated Raman lasers have been experimentally demonstrated in silicon racetrack resonators,[1,7–9] silicon photonic crystals,[2] on-chip diamond resonators,[10,11] photonic aluminum nitride (AlN) microresonators,[12] lithium niobate microresonators (LN),[13,14] silicon carbide,[15] and silica microcavities,[16–18] which enables microscale Raman lasing with high compactness, low energy consumption, and high design freedom in photonic integrated devices.[8–10,12] Moreover, through a tunable directional coupler to optimize both pump and signal coupling efficiency in the ring cavity, high-performance Raman lasing with a slope of up to 26% has been implemented.[9] Integrated SRS lasing based on the platforms mentioned above and techniques allow for potential applications such as optical amplification, spectroscopic sensing, archaeology, and clinical diagnosis.[5,7,19–24]

Yet, challenges remain in achieving a wide and continuous tunability of the photonic Raman laser. In general, discrete tuning of the Raman laser over a wide bandwidth is achievable by tuning the pump laser in the microresonators.[9,10,12] With additional cascaded Raman lasing, this tuning range can be further extended from the telecom band to the mid-IR range.[15] The interval in the tuning range is largely related to the mode spacing of the laser cavity, such that the mode hopping will leave a gap in the laser tunability. An additional challenge is on the material Raman spectrum. For instance, while significant Raman gain is accessible in crystalline cavities, the gain spectrum is always narrow-band. This requires precise control on the crystal
orientation and the cut, and control on the cavity resonant frequency to align with the Raman mode. To achieve a wide and gap-free tunability, strong and broadband material Raman gain is desired. In this way, the hopping-free tuning of each lasing mode could cover the spacing of the cavity mode, forming a wide and gap-free turning range of the Raman laser. Moreover, an unavoidable and strong interplay between Raman scattering and the Kerr effect has been observed in nonlinear materials.\(^\text{[11,25]}\) Most strategies have been proposed to suppress the Raman effects on the Kerr effect by controlling the FSR of microresonators.\(^\text{[11]}\) However, the impact of the Kerr effect on microresonators-based Raman laserings remains largely unexplored.\(^\text{[4,25]}\)

Chalcogenide glass (ChG) has attracted wide interest due to its significant Raman and Kerr nonlinearity while being resistant to two-photon absorption (TPA) and free-carrier absorption (FCA).\(^\text{[26–31]}\) It has wide transparency from the visible to the infrared region (up to \(\approx 25\ \mu m\)).\(^\text{[32–36]}\) Being amorphous, the material Raman gain becomes broadband, enabling a flexible design on the free spectral range (FSR) of integrated microresonators.\(^\text{[10,12,16]}\) Such properties in integrated photonic devices not only enable largely boost the performance of SRS and the Raman lasing at low optical powers but can also lead to nontrivial nonlinear interactions over a large wavelength.\(^\text{[13,37]}\) Promising applications such as trace-gas sensing, environmental monitoring and biomedical analysis could be achieved through a potential development of Raman lasers in the mid-infrared (mid-IR) wavelength band.\(^\text{[1,26]}\) Indeed, so far, the implementation of single-mode Raman lasing in the chip integrated ChG microresonators have yet to be explored.\(^\text{[38,39]}\) The reported Q-factors to date remain insufficient to support high-efficient nonlinear interactions as well as the SRS\(^\text{[30]}\).

In this work, we develop a photonic integrated high-Q ChG microresonator platform and demonstrate a highly efficient chip-scale Raman lasing at the telecommunication L band. Compared with ChG devices prepared with the typical Arsenic (As)-based components reported in our previous work,\(^\text{[41]}\) the integrated microresonators are based on environment-friendly \(\text{Ge}_{25}\text{Sb}_{10}\text{S}_{65}\) (GeSbS), which shows a nonlinear coefficiency \(n_2\) of \(\approx 2.0 \times 10^{-18}\ \text{m}^2\ \text{W}^{-1}\).\(^\text{[42]}\) It has a superior damage threshold and thermal stability and also reveals a low TPA \(\beta\) of 0.032 cm GW\(^{-1}\), which is more than one order of magnitude less than As\(_2\)Se\(_3\).\(^\text{[43–46]}\) The intrinsic Q-factor of our microresonators is as high as 2.2 \(\times 10^8\), and the threshold power for the Raman lasing is as low as 3.25 mW. We demonstrate both the single-mode and the cascaded Raman lasing, together with a broadband Raman–Kerr comb, by engineering the dispersion and the four-wave-mixing (FWM) phase matching of the microresonator. Moreover, the thermal tunability of the Raman laser is also demonstrated to cover a frequency range of over 200 GHz (more than one FSR). Our work marks, to the best of our knowledge, the first observation of tunable Raman lasers by engineering the Kerr effect to enhance the generation of Raman laserings in photonic chip-integrated platforms, which may contribute not only to the fundamental physics of Raman lasing in dissipative Kerr resonators, but also to spectroscopic applications by, e.g., supporting hyperspectral imaging.

### 2. Results and Discussions

#### 2.1. Design Principles

A schematic of the ChG microring resonator-based Raman lasering is shown in Figure 1a. For the purpose of Raman lasing, the GeSbS waveguide is designed to support normal cavity dispersion. This will suppress nonlinear parametric oscillations usually evoked by the spontaneous Kerr nonlinearity in the anomalous dispersion regime. As such, engineering the waveguide geometry (i.e., changing the core width and height) will mainly contribute to tuning the FWM phase-matching condition among the pump wave and the Raman-induced Stokes/anti-Stokes waves. Consequently, there are mainly two types of Raman lasing: single-mode and cascaded Raman lasers in the FWM phase-matching condition and a relative broadband Raman–Kerr comb in the presence of phase mismatch (Figure 1a(i),(ii)).

The Raman gain spectrum of the GeSbS film was measured by Raman spectrometer (Figure 1b), which shows a broadband spectral profile compared with crystalline materials.\(^\text{[47]}\) In particular, the main Raman gain is centered at the frequency offset of 340 cm\(^{-1}\) (\(\approx 10.2\ \text{THz}\)), with a linewidth of \(\approx 1.73\ \text{THz}\) (estimated by a single-Lorentzian fit of the gain profile).\(^\text{[48]}\) A complete analysis of all vibrational modes underlying the gain spectrum and the normalized temporal response are presented in the Supporting Information.

In the presence of a strong pump wave, the Raman lasing dynamic is enriched. Given that the nonlinear parametric oscillations are effectively suppressed (in the condition of overall normal cavity dispersion), dispersion engineering within a certain range would mainly alter the potential phase matching condition for the SRS and the Raman lasing. In particular, with respect to the generation of the second Stokes wave, there are FWM processes along with the Raman effect. One is the degenerate FWM process involving the pump wave, the first and the second Stokes waves. In this case, the pump energy is converted to the first Stokes wave because of the accompanied SRS effect, and the conversion between the first and the second Stokes waves is followed. This would lead to a strong second Stokes wave that may surpass the anti-Stokes wave in power (Figure 1c). The other is the non-degenerate FWM process involving the pump wave, the first and the second Stokes waves, and the anti-Stokes wave (Figures S3 and S4, Supporting Information). In microresonators, the phase mismatches \(\Delta \nu\) of these FWM processes are reflected by the difference of resonant frequencies of related modes,\(^\text{[33]}\) i.e.

\[
\Delta \nu^1 = 2\omega_{s,1} - \omega_0 - \omega_{s,2}
\]

\[
\Delta \nu^2 = \omega_0 + \omega_{s,1} - \omega_{s,1} - \omega_{s,2}
\]

where \(\omega_0\) indicates the resonant frequency of the pumped mode, \(\omega_{s,1}, \omega_{s,2}\) are resonant frequencies of modes that support the first and the second Stokes waves, respectively, \(\omega_{s,1}\) is the resonant frequency of the anti-Stokes mode. Clearly, the FWM phase-matching condition can be altered by tailoring the dispersion of the microresonator, which is implemented via tuning the core width of the GeSbS waveguides. In this work, we designed mainly two types of GeSbS microresonators, with a difference in the cav-
Figure 1. Principles of engineered Raman lasing in chip-scale ChG microresonators. a) A schematic of engineered Raman lasing in GeSbS microresonators, two types of Raman lasing can be generated: i) the cascaded Raman laser in the microresonator with FWM phase matching and ii) the Raman–Kerr comb state in the presence of FWM phase mismatch. b) Measured Raman spectrum of ChG film with a thickness of 0.8 μm. c) Schematic of the degenerate FWM between the pump wave, the first and the second Stokes waves. d) Calculated dispersion parameter D2 of the microresonators, with different waveguide widths (1.7 and 2.4 μm). e) Calculated phase mismatch for the degenerate FWM process (Δν1).

2.2. Low-Threshold Single-Mode Raman Laser

We characterized the Raman lasing in the fabricated GeSbS microresonators. A typical scanning electron microscopy (SEM) picture of the fabricated GeSbS microresonators is presented in Figure 2a, together with the picture of the magnified coupling area. In a microresonator with the FSR of ≈200 GHz, a typical resonance linewidth of ≈150 MHz was measured in the regime of the critical coupling (Figure 2b–e). This indicates a high intrinsic Q-factor (Qi) ≈2.2 × 1010. In exploring the Raman lasing dynamic, the GeSbS microresonator is deliberately designed to support undercoupled pump modes and overcoupled Stokes modes, such that the conversion efficiency is effectively elevated at the transmitted port of the microresonator.[9]

The Raman lasing threshold power in the fabricated GeSbS microresonators was tested. In the experiment, an amplified continuous wave (CW) tunable laser is coupled to the resonator. The insertion loss of fiber-to-chip coupling is ≈2.5 dB/facet for the fundamental TE mode. At the output, the optical signal is monitored by an optical spectral analyzer (OSA), in which the generated Raman lasing is quantitatively noted (cf. the Supporting Information for a detailed experimental setup). At a low pumping power, the threshold for the Raman lasing is identified. In this condition, only single-mode Raman lasers are observed (Figure 2f). When the microresonator is pumped at 1550 nm with the power of 3.5 mW, a significant Stokes laser is monitored at 1637 nm, in the transmitted port of the resonator. The frequency offset is ≈10.3 THz, which is close to the central peak of the Raman gain. In addition, a weak anti-Stokes laser is also monitored at the blue side of the pump with a similar frequency offset. Moreover, the transmitted power of this single-mode Stokes wave is traced as a function of the pump power, in which a threshold power of ≈3.25 mW can be identified (Figure 2g). Above the threshold, the Stokes wave is enhanced when the pump power is increased. The external slope efficiency is extracted to be 13.86%. We can further estimate the Raman gain coefficient (gR) of GeSbS material, which is ≈7.37 × 10−12 m W−1 (a value over 100 times higher than that in silica[17]). By comparing Raman laser characteristics based on different material platforms (Table S1, Supporting Information), our ChG-based Raman microresonator demonstrates efficient in the low pumping threshold (≈3.25 mW) and high slope efficiency (≈13.6%).

2.3. Experimental Results of Engineered Raman Lasing

Next, we verified the experimental results of engineered Raman lasing in ChG microring resonators at high pump power. As a result, distinguished Raman lasing dynamics were observed. In
Figure 2. Raman lasing threshold power in CHG microresonator. a) SEM image of a GeSbS microresonator, together with the image on the coupling region. The cross section of the GeSbS waveguide is 2.4 \( \mu \text{m} \times 0.8 \mu \text{m} \), and a radius of 100 \( \mu \text{m} \). The microring has a 1.3 \( \mu \text{m} \) wide bus waveguide and a 35° pulley-style coupler with a critical coupling gap of \( \approx 550 \text{nm} \). Simulated field distribution at the b) pump wave and c) first Stokes wave, respectively. The microring can support good spatial overlap between the pump and Stokes mode. d) Measured transmission spectra and e) resonance of fundamental TE modes of the microring. The Q factors of the 2.4-\( \mu \)m microresonator are \( Q_i = 2.2 \times 10^6, Q_L = 1.3 \times 10^6, Q_c = 0.8 \times 10^6, Q_S = 5.8 \times 10^6 \). f) Measured optical spectrum of the first Stokes wave at around 1637 nm, with the on-chip pump power estimated to be \( \approx 3.5 \text{mW} \). g) The power trace of the first Stokes wave as a function of the pump power. The slope efficiency is \( \approx 13.86\% \), and the threshold power is \( \approx 3.25 \text{mW} \).

In the case of a small phase mismatch, the FWM processes will be dominant, leading to single-mode and cascaded Raman lasing when the pump power is increased (Figure 3a,b). In contrast, in the case of large phase mismatch, a remarkable broadband comb spectrum is observed mainly around the pump wave and the first Stokes wave (Figure 3d,e,g). This is understood that when the energy conversion to the second Stokes wave is suppressed as essentially phasing mismatched, the accumulated phase in the first Stokes wave would surpass the FSR of the resonator and start to convert energy to the adjacent resonant modes. As a consequence, the spontaneous Kerr nonlinear process is triggered, which leads to the generation of “sub-Kerr-combs” around the first Stokes wave and around the pump wave. The FWM phase-matching conditions at different pump wavelengths are also estimated, cf. the Supporting Information, in which the phase mismatch value (\( \Delta \nu \)) is slightly decreased with an increase in the pump wavelength. As such smooth transition from the cascaded Raman lasing to the Raman–Kerr comb state is observed, cf. the Raman lasing spectra presented in Figure S4 (Supporting Information).

Indeed, such a Raman–Kerr comb has been previously reported in whispering gallery mode crystalline resonators, which was also attributed to the cavity dispersion effect.\(^{[25]}\) However, due to the fact that performing dispersion engineering in crystalline resonators is difficult, the pump frequency has to be carefully selected to meet the required dispersion condition. In this context, our work may contribute to providing a flexible photonic platform that allows for engineered Raman lasing performance.

We also performed numerical simulations in both dispersion conditions underlying the two microresonator structures (i.e., the waveguide width of 1.7 and 2.4 \( \mu \text{m} \)). The simulation is based on the Lugiato–Lefever equation (LLE) with the complete form of the Raman response included.\(^{[25]}\) As a result, both the cascaded Raman lasing and the Raman–Kerr comb are observed by simulations, which shows good agreement with experiment results (Figure 3c,e; for more details, see the Supporting Information). In addition, simulations also indicate that the cascaded Raman lasing represents a stable state in the cavity, while the Raman–Kerr comb is in a non-stable state (Figure S7, Supporting Information). As shown in Figure 3g, when increasing pump power to \( \approx 30 \text{mW} \) in 2.4-\( \mu \)m-width microresonators, the broadband Raman–Kerr comb covering first and anti-first comb lines can be obtained, indicating the versatility of our GeSbS platform.

2.4. Tunability of Raman Laser

We next demonstrated the tunability of the Raman lasers in the FWM phase-matching condition, which is using tuning the pump wave or by tuning the operating temperature on the photonic chip. As a result, by consecutively coupling the pump wave into the resonant modes of the microresonator, both the first and the second Stokes waves are shifted accordingly (Figure 4a–c). The tested tuning range for the first Stokes wave is 1615–1658 nm, and that for the second Stokes wave is 1720–1755 nm, while the pump wavelength is tuned within the telecommunica-
Figure 3. Engineered Raman lasing dynamics. a,b) The measured Raman spectra in the 1.7-μm microresonator, by narrowly tuning the laser frequency (i.e., changing the laser-cavity detuning). This operation is equivalent to the change in the cavity coupling power.\textsuperscript{[17]} The pump power is $\approx 35 \text{ mW}$ (corresponding intracavity power intensity is $345.1 \text{ mW cm}^{-2}$). The first Stokes and second Stokes lasing with Raman shifts of $\approx 10.22$ and $\approx 10.24$ THz are generated almost simultaneously, implying similar threshold power. d,e) The measured Raman spectra of the microring with 2.4 μm width by changing the detuning. The pump power is $\approx 20 \text{ mW}$ (corresponding intracavity power intensity is $273.7 \text{ mW cm}^{-2}$). c,f) Numerical simulations in both dispersion conditions of the microresonators with the width of 1.7 and 2.4 μm, respectively. g) The measured Raman spectrum when increasing the pump power to $\approx 30 \text{ mW}$ in the 2.4-μm microresonator.
Figure 4. Tunability of the Raman laser. Discrete tuning of a) first Stokes spectra and b) the pump power (red) and the corresponding Raman laser output power (blue) as well as c) second Stokes spectra by adjusting the pump wavelengths ranging from 1535 to 1570 nm. d) The pump frequency is tuned within a thermally red-shifted resonance. The output power is normalized to the peak emission at each pump wavelength. e) Individually measured first Stokes wavelengths of the microring resonator corresponding to the different temperatures. Continuous tuning of the Stokes wavelength over ≈220 GHz (more than 1-FSR).
tion C-band (1540–1565 nm). The laser output power at each first Stokes mode is presented in Figure 4b, corrected for 9.2 dB output optical path loss. The laser power can be maintained around $\approx -10$ dBm by tuning the pump power and the frequency. The required pump power is slightly increased due to a decrease in the Raman gain coefficient. The maximum second stokes output power can also reach $\approx -9$ dBm. However, the output power of the second stokes mode has a strong fluctuation as the output power can also reach $\approx -9$ dBm. Further improvement is expected by improving the intrinsic Q-factor and optimizing the coupling design.[9]

Moreover, the Raman laser can also be continuously tuned by tuning the temperature. It is observed that with an increase in the temperature, the first Stokes wave is redshifted (Figure 4d,e). Significantly, by tuning the temperature of $\pm 40$ °C, the frequency shift of the Raman lasing ($\approx 220$ GHz) surpasses the FSR ($\approx 210$ GHz) of the microresonator. Therefore, single-mode Raman lasing at an arbitrary wavelength in the range 1615–1755 nm is accessible with a combination of both the pump laser and the temperature tuning schemes. In principle, such a tunability on the Raman lasing is unlimited as the Stokes wave is only coupled to the pump wave with a fixed frequency offset. Nevertheless, we are limited by the operational bandwidth of our laser sources and the optical fiber amplifiers. On another aspect, the FWM phase-matching condition should always be carefully designed as it may drift over the wavelength.

3. Conclusion

In summary, we have presented an engineered Raman lasing in an integrated GeSbS microring resonator with a high Q-factor (above 10[4]). We investigate the effect of dispersion on the interaction of the Kerr–Raman effect in integrated ChG microresonators and determine the conditions required for enriched Raman lasering. The cascaded Raman lasing and a broadband Raman–Kerr comb were experimentally realized by tailoring the dispersion of the microresonator. Moreover, the tunability of such a chip-scale Raman laser is also demonstrated by adjusting the pump wavelength and changing the temperature on the chip. This allows for the access of single-mode lasing at arbitrary wavelengths in the range of 1615–1755 nm. Our results pave the way for the generation of versatile on-chip Raman lasers and Kerr frequency combs covering the NIR to MIR range based on the ChG photonic platform.

4. Experimental Section

The photonic integrated ChG waveguides and microresonators were fabricated based on an improved subtractive nanofabrication process that allows for stable and high-quality ChG microresonators.[43] Initially, GeSbS glass was synthesized from high-purity elements, including 6N germanium (Ge), 6N antimony (Sb), and 6N sulfur (S), by the conventional melt-quenching technique,[44] which was further purified by a modified physical and chemical purification technique.[45] The high-purity GeSbS was then deposited on a silicon wafer with a 3-µm SiO$_2$ layer via thermal evaporation, forming a film of 0.8-µm thickness, and was patterned by electron-beam lithography. Afterward, the pattern was etched in an inductively coupled plasma reactive ion etcher (ICP-RIE) with CH$_4$ gas. At last, a 3-µm layer of silica was deposited on the device via inductively coupled plasma chemical vapor deposition (ICP-CVD) as the cladding. Regarding this work, the ChG waveguides and the microresonators have a fixed height of 0.8 µm, and the width was flexibly tailored in the range 1.6–2.4 µm, which allows for dispersion engineering as well as engineered Raman lasing.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

Acknowledgements

D.X. and Y.H. contributed equally to this work. This work was supported by the National Key R&D Program of China (Grant No. 2019YFA0706301), Key Project in Broadband Communication and New Network of the Ministry of Science and Technology (MOST) (2018YFB1801003), National Science Foundation of China (NSFC) (U2001601, 61975242, 61525502, 11074234, 62035018), the Science and Technology Planning Project of Guangdong Province (2019A1515010774), and the Science Foundation of Guangzhou City (202002030103).

Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available in the Supporting Information of this article.

Keywords

chalcopyrite glasses, integrated Raman lasing, on-chip microresonators, Raman–Kerr comb

Received: August 9, 2021
Revised: December 20, 2021
Published online: January 30, 2022

[1] H. Rong, S. Xu, O. Cohen, O. Raday, M. Lee, V. Sih, M. Paniccia, Nat. Photonics 2008, 2, 170.
[2] Y. Takahashi, Y. Inui, M. Chihara, T. Asano, R. Terawaki, S. Noda, Nature 2013, 498, 470.
[3] M. Bernier, V. Fortin, M. El-Amraoui, Y. Messaddeq, R. Vallee, Opt. Lett. 2014, 39, 2052.
[4] P. J. Zhang, Q. X. Ji, Q. T. Cao, H. M. Wang, W. J. Liu, Q. H. Gong, Y. F. Xiao, Proc. Natl. Acad. Sci. USA 2021, 118, e2101605118.
[5] X. Shen, H. Choi, D. Chen, W. Zhao, A. M. Armani, Nat. Photonics 2019, 14, 95.
[6] Z. Gong, M. Li, X. Liu, Y. Xu, J. Lu, A. Bruch, J. B. Surya, C. Zou, H. X. Tang, Phys. Rev. Lett. 2020, 125, 183901.
[7] H. Rong, A. Liu, R. Jones, O. Cohen, D. Hak, R. Nicolaeascu, A. Fang, M. Paniccia, Nature 2005, 433, 292.
[8] Z. Zhou, B. Yin, J. Michel, Light: Sci. Appl. 2015, 4, e358.
[9] M. Ahmad, W. Shi, S. LaRochele, Optica 2021, 8, 804.
[10] P. Latwiew, V. Venkataraman, M. J. Burek, B. J. M. Hausmann, I. Bulu, M. Loncar, Optica 2015, 2, 924.
