Stimulated Raman scattering induced dark pulse and microcomb generation in the mid-infrared

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Abstract

We demonstrate that strong stimulated Raman scattering in silicon and germanium microresonators can induce stable and breathing dark pulses generation circumventing traditional complex approaches such as pump modulation and mode coupling. Although multi-photon absorption shows a small influence on the detuning value for stable dark pulse excitation, the concomitant free carrier will assist dark pulse excitation and broaden the excitation area of dark pulse thus making it easier to capture stable pulse. Furthermore, dark breather dynamics in Si and Ge are also observed, which shows distinct properties from the dark soliton breathers dominated solely by Kerr effect. Finally, we show that octave spanning mid-infrared (MIR) microcomb can be generated combining with high-order dispersion engineering, which in turn affects the breathing dynamics of dark pulses. Our findings provide another way for the initiation of dark pulses in group IV materials and broadband MIR microcomb generation for spectroscopy applications.

1. Introduction

Bright and dark solitons generally exist in microresonators featuring anomalous or normal dispersion, respectively. Triggered by plenty of applications such as astronomy [1, 2], optical atomic clocks [3], optical communication [4], spectroscopy [5–7], imaging and precision ranging [8–10], most of the reported solitons and microcombs operate in the anomalous dispersion regime as phase matching conditions are more accessible and favored for broadband optical frequency comb generation [11–14]. Correspondingly, the generated spectra are mainly located in the near-infrared band. Compared with bright solitons, dark pulses and microcombs generation in the normal dispersion regime exhibit higher pump to comb conversion efficiency [15]. Theoretically, conventional bright solitons form in the anomalous dispersion regime, ascribed to double balance of Kerr nonlinearity and dispersion on the one hand, as well as external driving and loss on the other. While dark pulse, as another kind of localized dissipative structure, its underlying mechanism lies on the interlocked switching waves or fronts connecting the upper and lower homogeneous steady-states of the bistable system [16, 17]. As the counterparts of bright solitons, dark pulses also provide a feasible route to generate microcombs in platforms where normal dispersion are dominated. However, the excitation of dark pulses usually requires additional assistance from mode coupling [18–21] or modulated pump [22, 23]. Besides the Kerr nonlinearity, another effect associated with soliton propagation is Raman interaction, which introduces a red shift of the soliton spectrum in microcavities [24]. Raman gain can produce optical amplification and laser action of waves in microresonators where the gain spectrum is narrow and strong [25, 26]. Especially, the Raman gain in crystalline microresonators such as magnesium fluoride, silicon [27], germanium [28], diamond [29], and aluminum nitride (AlN) [30] is strong and narrow. Stimulated Raman scattering (SRS) effect has
non-trivial impact on soliton formation in those platforms. Researchers have demonstrated the competition between SRS and Kerr nonlinearity in diamond and silicon microresonators [31]. Platonic generation initiated via Raman assisted four wave mixing in AlN microresonator have also been investigated [32]. While whether dark pulses can be excited by a narrow Raman gain in the mid-infrared (MIR) has remained unexplored.

Silicon, germanium and their alloy are extremely promising group IV material for significant MIR applications such as spectroscopy and biochemical-sensing [6, 33–35]. The transparency window of Si and Ge ranges from 1.1 to 8.5 μm [36] and 1.5 to 14.3 μm [33], respectively, while SiGe alloy expands more deeply into the MIR (potentially up to 15 μm). Covering the functional group (2.5–7.7 μm) and molecule ‘fingerprint’ regions (7.7–16.7 μm) [37]. In addition, Si and Ge exhibit much larger linear and non-linear refractive indices than conventional nonlinear platforms (e.g. SiN, and SiO2), which introduces well light-confinement and lower power threshold for parametric process. Recently, optical frequency comb generation in Si and Ge has attracted lots of interest [34, 38–40]. Group IV photonics materials also have large Raman gain coefficient [41, 42], constituting an opportunity to dark pulses excitation without additional techniques.

In this work, we investigate that dark pulses can be generated in Si and Ge microresonators with normal dispersion via the strong and narrow Raman gain spectrum. Stable and breathing dark pulses dynamics have been demonstrated with a single pump through linear detuning scanning in both cases. The widely existing multi-photon absorption in the two materials shows influence on the value of detuning associated with the emergence of stable or breathing dark pulses. Free carriers introduced by multi-photon absorption, will assist dark pulse excitation and extend the existing range of dark pulses thus making it easier to capture stable pulses. The properties of Raman effect dominated dark pulses have also been characterized and compared with their Kerr dominated counterparts. Furthermore, an octave spanning MIR microcomb is demonstrated with the assistance of SRS combining with high-order dispersion (HOD) engineering, which in turn affects the breathing dynamics of dark pulses. These findings pave the way for the excitation of dark pulses with simplified pump condition and broadband MIR microcomb generation for spectroscopy applications.

2. Theory

To model the dynamics of SRS induced dark pulses, the extended Lugiato–Lefever equation (LLE) incorporating HOD, three- and four-photon absorption (3PA and 4PA), free-carrier absorption (FCA), free-carrier dispersion (FCD) and SRS effects is applied for numerical simulations [43, 44]. Such extended LLE is a normalized form for generalization.

\[
\frac{\partial \psi}{\partial \zeta} = - \left(1 + i \Delta \right) \psi - \frac{1}{2} \left(1 + i K \right) \phi \psi - \frac{d_2}{2} \frac{\partial^2 \psi}{\partial \eta^2} + \sum_{n=0}^{\partial_2} \frac{d_n}{n!} \frac{\partial^n \psi}{\partial \eta^n} - \frac{A_1}{3} |\psi|^4 \psi
\]

\[
- \frac{A_1}{4} |\psi|^6 \psi + i \left(1 - f_0 \right) |\psi|^2 \psi
\]

\[
+ if_1 \left( \Re \langle |\psi|^2 \rangle \right) \psi + F,
\]

\[
\frac{\partial \phi}{\partial \zeta} = \theta_3 \langle |\psi|^6 \rangle + \theta_4 \langle |\psi|^4 \rangle - \frac{\phi_c}{\tau_c},
\]

\[
\Re(\eta) = H(\eta) \cdot a e^{-i n \sin(\phi)}.
\]

Here, the parameters

\[
\psi = E/\sqrt{\alpha/\gamma L}, \quad \zeta = \tau \delta / \alpha, \quad \eta = \tau \sqrt{\alpha/|\beta_2| L}, \quad \Delta = \delta_0 / \alpha, \quad K = \mu, \quad d_2, \quad d_n = \nu^{n+1} \left( \frac{\nu}{\rho} \right)^{n-1} \frac{\rho}{\nu^{n}},
\]

\[
A_3 = \frac{\alpha^2 \rho_0 A_{eff}}{\gamma L \alpha^2}, \quad A_4 = \frac{\alpha^2 \rho_0 A_{eff}}{\gamma L \alpha^2}, \quad F = \frac{E_{in}}{\sqrt{\alpha/|\beta_2| L}}, \quad \phi_c = \langle N_c \rangle \sigma L / \alpha, \quad \theta_3 = \frac{\alpha^2 \rho_0 A_{eff}}{4 \mu \gamma L \alpha^2}, \quad \theta_4 = \frac{\alpha^2 \rho_0 A_{eff}}{4 \mu \gamma L \alpha^2},
\]

\[
\tau_c = \tau_{eff} \alpha / \beta_2, \quad \alpha, \quad \tau, \quad T, \quad E_{in}, \quad f_0, \quad f_2, \quad F_2, \quad \gamma, \quad n_2, \quad \omega_0, \quad \omega_0, \quad \eta, \quad \theta.
\]

Kerr nonlinear coefficient \(\gamma = n_2 / \alpha / c A_{eff}\) with \(A_{eff}, \omega_0\) and \(n_2\) representing the effective mode area, angular frequency of the CW pump and nonlinear refractive index, respectively. In equation (2), brackets denote...
average operation over round-trip time: \( \langle |\psi|^6 \rangle = \left( 1/T_R \right) \int_{-T_R/2}^{T_R/2} |\psi|^6 \, d\eta \). We set \( d_1 = 1 \) for the normal dispersion regime. In equation (3), \( \mathcal{R}(\eta) \) is the normalized Raman kernel function with
\[
a = T_0 (\tau_1^2 + \tau_2^2) / (\tau_1 \tau_2),
b = T_0/\tau_2 \text{ and } c = T_0/\tau_1,
\]
where \( T_0 = \sqrt{\beta_2 L / \alpha} \) is the normalization factor. \( H \) is the Heaviside function. The relationships between \( \tau_{1,2} \) and Raman coefficients (material-dependent, see the next paragraph) are \( \tau_1 = 1/\sqrt{\Omega_R^2 - \Gamma_R^2} \) and \( \tau_2 = 1/\Gamma_R \). The Raman effect is calculated by the convolution theorem which is denoted as \( \mathcal{R} \otimes |\psi|^2 = \mathcal{F}^{-1}(\mathcal{F}[\mathcal{R}] \cdot \mathcal{F}[|\psi|^2]) \). Technically, the extended LLE can be numerically solved by the Runge–Kutta algorithm with split-step Fourier transform.

We first consider the SRS term of Si and Ge. For Si, the full width at half maximum of Raman gain spectrum is \( \Gamma_R/\pi = 105 \) GHz and the frequency shift is \( \Omega_R/2\pi = 15.6 \) THz. While Ge shows a larger Raman gain coefficient with a linewidth \( \Gamma_R/\pi = 216 \) GHz and Raman shift \( \Omega_R/2\pi = 9.02 \) THz. The Raman coefficients (\( \Gamma_R \) and \( \Omega_R \)) are applied to calculate the temporal coefficients \( \tau_{1,2} \), which then can be used to obtain \( \mathcal{R}(\eta) \). Thus the real and imaginary parts of Fourier transform of the normalized Raman kernel function is calculated. The normalized Raman response is depicted in figure 1 with the real and imaginary parts of \( \mathcal{F}[\mathcal{R}] \) calculated by
\[
\text{Re}(\mathcal{F}[\mathcal{R}]) = ac \left( b^2 + c^2 - \omega^2 \right)/Q(\omega)
\]
and
\[
\text{Im}(\mathcal{F}[\mathcal{R}]) = 2abc\omega/Q(\omega),
\]
respectively, where \( Q(\omega) = \left( b^2 + c^2 - \omega^2 \right)^2 + 4b^2c^2 \). The real part modifies the refractive index while the imaginary one represents the Raman gain [45].

3. Results and discussions

In the absence of additional mode interaction, dark pulses can be stimulated by SRS as depicted in figure 2. During the sweeping of laser frequency, the pump is tuned into the resonance and the intracavity power grows correspondingly. As SRS occurs spontaneously in the cavity once the threshold power is reached, cascaded four-wave mixing process between these Raman comb lines and pump will dominate by tuning the laser detuning linearly. The temporal waveform becomes nonstationary (the slow time \( \zeta \) before 550). When the laser is tuned carefully to a proper red-detuning region (\( \Delta = 13.3 \) in our case), the intracavity power undergoes a sharp decrease, corresponding to the dark pulse step. The breathing dark pulses is first observed, which shows periodic variation in pulse energy and width. Figures 2(a)–(c) show the total intracavity energy, temporal and spectral evolution, respectively. The pulse trajectory shows a negative slope in the retarded fast time window (figure 2(b)), due to the redshift of the center frequency by strong comb lines near the Raman gain peak. Such redshift results in a group velocity increase and the negative trajectory slope. Theoretically, the Raman induced breather is the result of competition between Kerr and SRS effects. The Raman dominated breather shows a breathing frequency of 20.6 MHz and has relatively large breathing
depth compared with Kerr dominated dark soliton breathers reported in [20], which is 14.5% when the detuning is $\Delta = 13.3$. In addition, this dark breather shows a strong breathing in the waveform top, which is distinct from strong breathing in the waveform hole of dark soliton breathers.

Further increasing the detuning, stable dark pulse will form with constant pulse energy and width as shown in figure 3. In silicon, the nonlinear loss from 3PA exhibits small influence on the laser location for stable dark pulses emerging. The detuning value for stable dark pulse excitation with 3PA ($\Delta = 13.47$) is smaller than that without 3PA ($\Delta = 14.45$). Those stable pulses exhibit similar pulse width and spectral characteristics. Such continuous tuning scheme requires precise control of the laser frequency to capture stable dark pulse, because the survival range of stable pulse is relatively narrow. Intriguingly, the FC effects are found to be beneficial for stable dark pulses capturing, which will greatly broaden the pulse capture area and improve the feasibility of the experiment. Figure 4(a) shows the implementation process of dark pulse capture with FC. Firstly, one can increase the detuning linearly until the energy in the cavity reaches a noisy state. Then keep the detuning unchanged ($\Delta = 8$) and turn on the FC via setting a proper external bias (the red arrow in figure 4(a) with $\tau_c = 0.18$). The nonlinear loss associated with FCA and nonlinear detuning attributed to FCD will work together and help to achieve a stable pulse. Figures 4(b) and (c) show the final spectrum and temporal waveform. Two pronounced wings in the spectrum (highlighted by the black dashed circle) around the pump are weaker than that in figure 3(h) (highlighted by the black dashed circle) due to the absence of FCA. To confirm the feasibility of the scheme, we stop the laser at five different positions, and obtain stable dark pulses by controlling the FC lifetime in all cases (see figure 4(d)). Apparently, the larger the detuning value, the shorter the carrier lifetime is required for stable dark pulses evolution. This approach is experimental friendly and different from a two-step method in [32]. FC assisted dark pulses generation with SRS will further promote the realization of microcombs in group IV materials such as silicon and germanium.
Figure 3. Dynamics of SRS induced stable dark pulses with/without 3PA. (a)–(c) and (d)–(f) Are the intracavity energy, temporal evolution and spectral evolution of SRS induced stable dark pulses in the absence (left column) and presence (right column) of 3PA. (g) Final temporal profiles and (h) spectra of dark pulses with (green curve) and without (blue curve) 3PA. The parameters are: $A_3 = 0.0014$, $\theta_3 = 0.0472$, $A_4 = 0$, $\theta_4 = 0$, $F = 6$, $\kappa = 0.016$, $d_2 = 1$, $d_4 = 0$, $\tau_c = 0.005$. The detuning is linearly tuned from $-0.001$ to 13.47 or 14.45 for the cases with or without 3PA, respectively.

Germanium, as another promising candidate of group IV photonics materials, exhibits larger Raman gain coefficient (about 4.5 times) \[46\] and nonlinear refractive index (about 7–8 times) \[28\] than silicon. Moreover, the transparency window of Ge covers most of the ‘fingerprint’ region, making it a potential platform for producing MIR microcombs. Stable and breathing dark pulses can also be realized utilizing the strong SRS effect. Owing to the larger Raman gain spectrum of Ge, a microcavity with a larger free spectral range (FSR) is needed to balance the competition between SRS and Kerr effects for dark pulse excitation. The 4PA in this wavelength range is negligible. Figures 5(a)–(c) illustrate the dynamics of stable dark pulse sharing a similar tuning method and principle as applied in Si. The breathing pulse emerges before the stable one during laser sweeping as given in figures 5(d)–(f). The breathing frequency is 16.1 MHz. The Raman dominated breather shows relatively large breathing depth, which is 24.7% when the detuning is $\Delta = 7.68$. Realization of dark pulse and microcomb in Ge could promote spectroscopy in the far wavelength range of MIR. These results also help to deepen the understanding of nonlinear dynamic processes in the germanium microcavity.

Those theoretical and numerical results illustrate the feasibility of dark pulses generation with the aid of SRS. Importantly, excitation of dark pulse with SRS has no specific requirements on the microresonator quality factor as reported in [47]. Combing with dispersion engineering in realistic microresonators, broadband MIR microcombs can be generated with a single CW laser source. The geometric size of the silicon microcavity is $500 \times 2550$ nm with a radius of 65 $\mu$m and propagation loss of 0.7 dB cm$^{-1}$. The pump wavelength is chosen as 2700 nm with pump power of 140 mW. Other parameters are set as: $\beta_2 = 4.599 \times 10^{-26}$ s$^2$ m$^{-1}$, $\beta_3 = 6.707 \times 10^{-40}$ s$^3$ m$^{-1}$, $\beta_4 = 4.086 \times 10^{-34}$ s$^4$ m$^{-1}$, $\beta_3PA = 8.939 \times 10^{-49}$ s$^{-1}$, $\beta_{4PA} = 2 \times 10^{-26}$ m$^3$ W$^{-2}$, $\beta_{4PA} = 3 \times 10^{-42}$ m$^5$ W$^{-3}$ [48, 49], $n_2 = 6 \times 10^{-18}$ m$^2$ W$^{-1}$, $\kappa = 0.0083$, $\sigma = 8.26 \times 10^{-21}$ m$^2$, $\mu = 3.16$ [50] and $\tau_{eff} = 5$ ps. The estimated normalized values are: $d_2 = 1$, $d_3 = 0.2723$, $d_4 = 0.0310i$, $d_5 = -0.0013$, $A_3 = 0.0014$, $\theta_3 = 0.0472$, $A_4 = 5.566 \times 10^{-6}$, $\theta_4 = 7.0838 \times 10^{-4}$, $F = 4.3$, $\tau_c = 0.005$. Figure 6 shows the dynamics of temporal and spectral evolution, in which the temporal drift (figures 6(a) and (c)) is the collective effect of SRS.
Figure 4. (a) Energy evolution with FC assisted dark pulse capture. The red arrow indicates the FC trigger point. (b) Final spectral and (c) temporal profiles of FC induced dark pulse. $A_3 = 0.0014, \theta_3 = 0.0472, A_4 = 0, \theta_4 = 0, F = 6, \kappa = 0.016, d_2 = 1, d_4 = 0, \tau_c = 0.18$. (d) FC lifetime required for different stopping value of detuning.

Figure 5. Dark pulse excitation with SRS in Ge microresonators. (a) Intracavity energy evolution (blue curve) and laser detuning (red dotted curve). (b) Temporal and (c) spectral evolution of SRS induced stable dark pulse. Right column shows dynamics of breathing dark pulses. The pump amplitude $F = 3.5$.

and HOD [43, 51]. In figure 6(c), the breather shows a breathing frequency of 12.4 MHz and breathing depth of 7.6%. The oscillation on the top of the pulse is more severe than that with only SRS (comparing figures 6(e) and 4(c)). The generated stable spectrum is favored for spectroscopy. More importantly, we provide a new idea for generating MIR optical frequency comb operating in the normal dispersion regime. Such mechanism could be exploited in materials exhibiting large Raman gain coefficient, including silicon, germanium and their derivatives. For stable dark pulse excitation, the FSR of the microresonators should be designed to controlling the competition between Raman gain and Kerr effect.
4. Conclusion

In conclusion, we propose a novel technique for dark pulse and MIR microcomb generation exploiting SRS effect in Si and Ge microresonators with normal dispersion, which circumvents traditional mode interaction and pump modulation schemes. Breathing and stable dark pulses will appear one after another during laser sweeping. Their evolution properties are different from the reported dark solitons dominated by Kerr effect. Interestingly, with the assistance of FC originated from 3PA or 4PA, the stable dark pulse capture range will be expanded. Therefore, the requirement for precise control of laser frequency is relaxed. Through microcavity cross-section designing, broadband MIR microcomb can be realized in realistic silicon microresonators. We expect that the proposed method will facilitate dark pulse excitation in group IV platforms as well as their practical MIR applications.

Disclosures

The authors declare no conflicts of interest.

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Data availability statement

Data underlying the results presented in this paper are not publicly available at the time but may be obtained from the author upon reasonable request.
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