Phase Synchronization in Stimulated Raman Process for Long Pulse Regime: Route for Wideband Coherent Spectrum Generation

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(Dated: February 19, 2019)

We investigate the evolution of coherence property of noise-seeded Stokes wave in short (< 1 ps) and long pulse (> 1 ps) regimes. Nonlinear equations describing the evolution of pump and Stokes waves are solved numerically for both the regions. Our statistical analysis explore the hidden coherence characteristics of Stokes wave in long pulse regime. Numerical results proclaim that noise-seeded stimulated Raman process, which plays the role in degradation of coherence in short pulse region, exhibits strong phase synchronization in long pulse regime. The manifestation of phase synchronization occurs by the transition of the Stokes wave from incoherent to coherent spectra. Finally, experiment is performed to validate our numerical results where sub-nanosecond pump is launched in the normal dispersion region of a commercially available fiber. Cascaded Raman based broadband spectrum is generated and exhibits high spectral coherence which is measured through f-2f interferometry.

Synchronization behavior is an omnipresent natural phenomena in physical, chemical and biological systems. Large amount of theoretical and experimental studies have been accomplished to understand various aspect of synchronization in classical system like flashing fireflies, Huygens clocks, pacemaker cells in mammalian hearts, as well as in quantum system like cold atom systems, nanomechanical resonators, spintronic and so on. More recently, the idea of synchronization has been extended to optical system where different mathematical models like Kuramoto model, reduced phase model, have been employed to understand different nonlinear phenomena such as phase locking, optical frequency comb and coherent beam combining. Researchers have harnessed nonlinear interactions through Kerr medium and utilize it to realize myriad of applications in several fields. Supercontinuum generation is the synergy of several nonlinear processes where spectral broadening occurs from normal band source and finds diverse applications. The sensitivity of the spectral broadening to the input pulse noise i.e, shot-to-shot stability of amplitude and phase becomes a paramount property from application aspect. Incoherent SC with large pulse-to-pulse fluctuations is suitable for optical coherence tomography, coherent anti-Stokes Raman scattering spectroscopy and many more. On the other hand, SC spectrum with high degree of coherence can be useful for many applications such as, optical frequency comb, frequency metrology. Several studies have been performed to achieve coherent SC generation. Coherence property of the generated SC spectrum relies on the nature of the pump source and the broadening mechanism. The degradation of coherence mainly happens due to the amplification of noise based phenomena like modulation instability, stimulated Raman scattering, Four-wave mixing and cross-phase modulation. Generally, femtosecond laser pulses are launched in all-normal dispersion fiber (ANDi) to generate coherent SC where soliton fission and MI are subsequently suppressed and spectral broadening occurs due to self-phase modulation (SPM) and optical wave breaking (WB). For long pulse regime, spectral broadening is dominated by noise-seeded SRS which causes incoherent spectrum. The detailed analysis explaining the transition between coherent and incoherent spectral broadening during SC generation has been reported in. This paper also reports how noise-seeded SRS process causes spectral decoherence of SC spectrum in long pulse regime (up to 5 ps).

In this work, we report a statistical analysis describing the evolution of the spectral coherence of noise-seeded Stokes wave for short and long pulse regimes in a unified manner. Our numerical results explore the inquisitive property of Stokes wave in long pulse regime, which reveals a strong synchronization between pump and noise-seeded Stokes pulse inside optical fiber. Phase synchronization is manifested by the gradual stabilization of the spectral coherence of the Stokes wave until it reaches to its maximum value. Experimental work is carried out to validate our analysis where cascaded Raman based broadband spectrum is generated by pumping with sub-nanosecond laser pulses into a commercially available fiber and exhibits high spectral coherence.

Short pulse regime (< 1 ps)—SRS is a non-linear process where energy from an incident pump field is transferred to redshifted Stokes field via molecular vibrations. A unified realistic model to study the growth of Stokes pulse through SRS process has been presented in. The evolution of pump and the Stokes wave is described by following coupled nonlinear equations,
\[
\frac{\partial u_p}{\partial z} + \frac{1}{v_{gp}} \frac{\partial u_p}{\partial t} + \frac{i}{2} \beta_{2p} \frac{\partial^2 u_p}{\partial t^2} - \frac{\beta_{3p}}{6} \frac{\partial^3 u_p}{\partial t^3} + \frac{\alpha_p}{2} u_p = \\
i\gamma_p (1 - f_R) u_p |u_p|^2 + i\gamma_p f_R u_p \\
\int_{-\infty}^{\infty} h_R(t - t') |u_p(t')|^2 + |u_s(t')|^2 dt' + \\
i\gamma_p f_R u_s \int_{-\infty}^{\infty} h_R(t - t') u_p(t') u_s^*(t') \exp[i\Omega_R(t - t')] dt' + \\
iu_s \int_{-\infty}^{\infty} h_p(t - t') f_N(z, t') \exp[i\Omega_R(t - t')] dt'
\]

(1)

\[
\frac{\partial u_s}{\partial z} + \frac{1}{v_{gs}} \frac{\partial u_s}{\partial t} + \frac{i}{2} \beta_{2s} \frac{\partial^2 u_s}{\partial t^2} - \frac{\beta_{3s}}{6} \frac{\partial^3 u_s}{\partial t^3} + \frac{\alpha_s}{2} u_s = \\
i\gamma_s (1 - f_R) u_s |u_s|^2 + i\gamma_s f_R u_s \\
\int_{-\infty}^{\infty} h_s(t - t') |u_s(t')|^2 + |u_s(t')|^2 dt' + \\
i\gamma_s f_R u_p \int_{-\infty}^{\infty} h_s(t - t') u_s(t') u_p^*(t') \exp[i\Omega_R(t - t')] dt' + \\
iu_p \int_{-\infty}^{\infty} h_s(t - t') f_N(z, t') \exp[i\Omega_R(t - t')] dt'
\]

(2)

In equations (1) and (2), \( \beta_{2j} \) is the group velocity dispersion (GVD), \( v_{gj} \) is the group velocity, \( u_j \) is the field amplitude, which is normalized as,

\[
u_j = \kappa A_j \int_{-\infty}^{\infty} T^2(x, y) dx dy)^{1/2}, \quad (j = p, s)
\]

(3)

where \( T(x, y) \) is the electric field distribution perpendicular to the direction of propagation, \( \kappa \) is a factor which normalize \( u_j \) such that \( |u_j|^2 \) represents power and defined as,

\[\kappa = \frac{1}{2} n_j (\varepsilon_0 / \mu_0)\]

(4)

where \( n_j \) is the linear index of the fiber. \( \gamma_j \) is the nonlinear coefficient and defined as,

\[
\gamma_j = \frac{\omega_j n_2}{c A_{eff} \omega^2}, \quad n_2 = \frac{3}{8 n_j} \chi_k (1 + \frac{2}{3} \frac{\chi_0}{\chi_k}),
\]

\[
A_{eff} = \frac{\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} T^2 dx dy \cdot T^2 dx dy}{\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} T^2 dx dy}, \quad \chi_R(t) = \chi_0 h(t)
\]

(5)

where \( n_2 \) is the nonlinear refractive index, \( c \) is the velocity of light in vacuum, \( A_{eff} \) is the effective mode area, \( \chi_k \) is the Kerr susceptibility, \( \chi_R \) is the third order time dependent nonlinear susceptibility leading to Raman scattering process and \( \chi_0 \) is the peak value of \( \chi_R(t) \). \( f_R \) is the fractional contribution of nuclei to the total nonlinear polarization, \( h_j \) is the response function for noise term, defined as,

\[h_j(t) = \frac{\omega_j R_N(t)}{4c n_j t_j}\]

(6)

where \( R_N(t) \) is the response function that accounts spontaneous scattering during propagation and convert a Langevin noise source function \( F_N \) which incorporates the random vibration of the silica molecule due to temperature \( T \), into susceptibility \( \chi_N \),

\[
\chi_N = \int_{-\infty}^{\infty} R_N(t - t') F_N(t') dt' \]

(7)

The noise force \( F_N \) can be expressed as,

\[F_N(z, t) = \frac{1}{2} [f_N(z, t) \exp(-i\Omega_R t) + c.c.]
\]

(8)

where \( f_N \) is the slowly varying function of random force and \( \Omega_R = \omega_p - \omega_s \) is the difference between carrier frequency of the pump and stokes wave.

To interpret equations (1) and (2), the first term on the left-hand side of both the equations represents the pulse envelope variation whereas the next three terms accounts the effect of group velocity, GVD and the third-order dispersion, respectively. The last terms on the left-hand side represent the fiber loss for both pulses. The first two terms on the right-hand side account the effect of SPM and XPM. The next two terms represent the molecular contribution of SPM and XPM, respectively and it also account the effect of inter-pulse self-frequency shift (SFS) and intra-pulse cross-frequency shift (CFS). The last term corresponds for the spontaneous Raman scattering process. To investigate the evolution of pump and the Stokes wave, we solve eq. (1) and eq. (2) using split-step Fourier method. The noise, which occurs due to thermal and quantum fluctuations of vibrational mode through spontaneous Raman scattering process, is modeled as a Markovian stochastic process with Gaussian statistics of zero mean and unit variance i.e. \( < f_N(z, t) > = 0 \). Due to the shot noise, the input pulse experiences fluctuations in power density and random phase of photon ranges within 0 to 2\( \pi \). This has been included in simulations. The input pump pulse has the form of, \( u_p(0, t) = \sqrt{P_0 \text{sech}(t/t_p)} \), where \( P_0 \) is the pump power and \( t_p \) is the pump pulse width. Initially, Stokes pulse is taken as white Gaussian noise distribution. To investigate coherence property of the generated Stokes wave, an ensemble of generated spectrum is simulated with varying input conditions. The input pump pulse is added with quantum shot noise where one photon per mode with random phase on each spectral discretization bin. An ensemble of 40 simulations are performed considering same identical input condition with different stochastic noise on the input laser pump pulse. The shot-to-shot fluctuation describing modulus of first-order coherence at zero path difference is given by [39],

\[|g_{12}^{(1)}(\lambda)| = \frac{< E_1(\lambda) E_2(\lambda) >}{\sqrt{< |E_1(\lambda)|^2 > < |E_2(\lambda)|^2 >}} \]

(9)

The angular bracket represents the ensemble average of the adjacent pair \( |E_1(\lambda), E_2(\lambda) | \) of generated spectrum from independent simulations. The degree of spectral
coherence varies between $0 \leq |g_{12}| \leq 1$ where 0 corresponds to incoherent spectrum representing high shot-to-shot fluctuation, whereas 1 corresponds to coherent spectrum representing perfect stability in phase and amplitude $^{40}$.

We solve equation (9) numerically several times to obtain the statistical evolution of coherence of generated Stokes wave. Parameters are taken from ref. $^{41}$ which is also compatible with our experimental conditions. The pump is Gaussian shaped pulse at 1064 nm. Both the Stokes pulse (1120 nm) and the pump fall in the normal dispersion region of the fiber. Fiber parameters for the pump wavelength are $\beta_2^p=25 \text{ ps}^2/\text{km}$, $\gamma^p=5.06 \text{ w}^{-1}\text{km}^{-1}$ and $g_p=2.34 \text{ w}^{-1}\text{km}^{-1}$. The value of these parameters for the Stokes pulse are scaled by the factor $\lambda_p/\lambda_s=0.95$. The walk-off length $d$ is 2.2 ps/m, where $d$ is given by,

$$d = \frac{1}{v_{gp}} - \frac{1}{v_{gs}} \quad (10)$$

In every simulation, fluctuating noise in the input leads to significant difference in Stokes properties. The statistical analysis of the evolution of coherence of Stokes with pulse width variation, input pump power and fiber length is performed from the simulated ensemble. Fig. 1 (a) and Fig. 1 (b) exhibit the spectral evolution and the corresponding spectral coherence, respectively, for the fiber length of 50 m. Very low spectral coherence is observed in this region which supports the results as discussed in $^{35}$. The amplification of noise causes high pulse-to-pulse fluctuation, resulting into poor coherence. As the pump pulse duration increases (fs to ps), SRS becomes more dominant and exhibits large fluctuation in phase and amplitude, leading to degradation of coherence as shown in Fig. 1(b).

To investigate the effect of input pump power on the coherence of the Stokes wave, the pump energy is varied. Fig. 2 (a) and Fig. 2 (b) depict the evolution of spectral intensity and spectral coherence of Stokes wave, respectively, over the propagation distance of 50 m long the fiber. It is observed that with the increase in power, Stokes spectrum also broadens but there is progressively degradation of spectral coherence. Fig. 3 (a) and Fig. 3 (b) show the spectral evolution and corresponding coherence of Stokes wave, respectively, when fiber length is varied. Pump power and pump pulse width are fixed at
1.8 kW and 0.6 ps, respectively. It is observed that Stokes spectrum gets compressed as it propagates through fiber. This is due to the narrowing of Raman gain with propagation. The central frequency of the Stokes wave are amplified over wings, which causes narrowing of Stokes spectrum \[11\]. The coherence also gradually degrades over the distance. The incoherent spectral evolution of noise seeded Stokes wave are discussed above. In long pulse region the scenario is totally different which we discuss in the next section.

**Long pulse regime** (\(>1\) ps)— The evolution of pump and Stokes waves governed by equations \([1] \) and \([2]\), relies on slowly varying envelope approximation. These equations are no longer valid for pulse width greater than 1 ps. For broad pulse width, the pulse envelope can be treated as constant in comparison with the time scale of Raman response function \(h(r)\). After rigorous simplification, the coupled nonlinear equations describing pump and Stokes evolution in long pulse regime can be given by \[36\],

\[
\begin{align*}
\frac{\partial u_p}{\partial z} + \frac{1}{v_g} \frac{\partial u_p}{\partial t} + \frac{\beta_2 p}{2} \frac{\partial^2 u_p}{\partial t^2} - \frac{\beta_3 p}{6} \frac{\partial^3 u_p}{\partial t^3} + \alpha_p u_p &= \gamma_p u_p |u_p|^2 + 2(f_R)|u_s|^2 + i/2 u_p u_s, \\
\int_{-\infty}^{\infty} g_p(t-t') u_s(t') \exp[i \Omega_R(t-t')] \, dt' + \\
iu_s \int_{-\infty}^{\infty} h_p(t-t') f_N(z,t') \exp[i \Omega_R(t-t')] \, dt' 
\end{align*}
\]

\[\text{(11)}\]

\[
\begin{align*}
\frac{\partial u_s}{\partial z} + \frac{1}{v_g} \frac{\partial u_s}{\partial t} + \frac{\beta_2 s}{2} \frac{\partial^2 u_s}{\partial t^2} - \frac{\beta_3 s}{6} \frac{\partial^3 u_s}{\partial t^3} + \alpha_s u_s &= \gamma_s u_s |u_s|^2 + 2(f_R)|u_s|^2 + i/2 u_p u_s, \\
\int_{-\infty}^{\infty} g_s(t-t') u_s(t') \exp[-i \Omega_R(t-t')] \, dt' + \\
iu_p \int_{-\infty}^{\infty} h_s(t-t') f_N^*(z,t') \exp[-i \Omega_R(t-t')] \, dt' 
\end{align*}
\]

\[\text{(12)}\]

where Raman gain coefficient \(g_j\) is defined as,

\[
g_j = 2f_R \gamma_j |\tilde{h}_r^j(\omega_R)|^2 \quad (j = p, s)
\]

\[\text{(13)}\]

\(h_r(\Omega)\) is the Raman response function at frequency \(\omega = \Omega_R\). The real \(\tilde{h}_r^j(\omega_R)\) and imaginary parts \(|\tilde{h}_r^j(\omega_R)|\) of \(h_r(\omega_R)\) stand for Raman induced index change and Raman gain, respectively.

In order to investigate the evolution of spectral coherence at long-pulse regime, equations \[(11)\] and \[(12)\] are solved numerically. The fiber parameters are similar to the case for short-pulse regime. Fig. \[4\] represents the simulated results for varying pump pulse duration. It is observed that with the increase in pulse width, the Stokes wave which builds from the random noise, gradually develops towards high spectral coherence and reaches to its maximum value. The dynamics of the pump and the Stokes wave for varying pulse width are also shown in the figure. The origin of the phase synchronization can be understood in terms of walk-off length. With the increase in pump pulse width, the corresponding walk-off length also increases as the walk-off length is proportional to the pulse width. The enhanced walk-off length allows the pump and the noise-seeded Stokes pulse to propagate together, providing phase synchronization. The initial random phase of the Stokes wave tends to stabilize gradually to its maximum value with the increase in pulse width and hence the coherence. The simulated results reveal the phase synchronization of the Stokes wave where the degree of spectral coherence of the Stokes wave gradually stabilize with the increase in pulse width and at 500 ps pulse width, very high spectral coherence is obtained.

**Experimental verification**— To validate our numerical analysis, we have performed experiment which is compatible with the simulated parameters taken in our analysis. We have used Corning\textsuperscript{\textregistered} LEAF fiber (CLF) which is pumped with a sub-nanosecond pulses in the normal dispersion region of the fiber. The pump pulse is a single-mode output of a Q-switched microchip laser producing pulses of 0.77 ns duration with a repetition rate of 20 kHz at 1064 nm. The experimental setup is shown in Fig. \[5\] The coupling is achieved using microscope objective (NA=0.4, 20X) and the combined power is controlled with the combination of HWPG and the PBS. As the pump wavelength is below the cut-off wavelength of the fiber, the fiber supports few-modes which may interact with each other through nonlinear processes. The desired spatial mode excitation is controlled by a three-axis translational stage. Output spectrum is recorded with the optical spectrum analyzer (OSA, AQ6319) whereas the spatial mode profile is captured through CCD camera as shown in Fig. \[5\]

To experimentally characterize the coherence property of the Stokes wave, we perform \(f-2f\) interferometry, where the output of the spectrum is sent to the asymmetric Michelson interferometer having unequal arms \[42\]. The gradual movement of one arm while the other arm is fixed, provides the control on time delay that corresponds to the pulse time period. The overlapping of the temporally delayed pulses generate interference fringes. The spectrally resolved interference spectrum is measured through OSA. The magnitude of the first order mutual coherence as a function of wavelength \(g_{12}^{(1)}(\lambda)\) is calculated as \[43\],

\[
|g_{12}^{(1)}(\lambda)| = \frac{V(\lambda)|I_1(\lambda) + I_2(\lambda)|}{2\sqrt{I_1(\lambda)I_2(\lambda)}}
\]

where, \(I_1(\lambda)\) and \(I_2(\lambda)\) are the generated spectrum through individual arm of Michelson interferometer, \(V(\lambda)\) is the fringe visibility given by, \(V(\lambda) = (I_{max} - I_{min})/(I_{max} + I_{min})\), where \(I_{max}\) and \(I_{min}\) are the maximum and the minimum intensity, respectively. We use
FIG. 4: Numerical simulation in long pulse region. Left column: (a), (d), (g), (j) and (m) are spectral evolution of the pump after 0.5 km propagation for pump pulse width of 5 ps, 50 ps, 100 ps, 250 ps and 500 ps, respectively. Middle column: (b), (e), (h), (k) and (n) represents the spectral evolution of the generated Stokes for the pump pulse width of 5 ps, 50 ps, 100 ps, 250 ps and 500 ps, respectively. Right column: (c), (f), (i), (l) and (o) are the simulated spectral coherence of the Stokes wave for pump pulse width of 5 ps, 50 ps, 100 ps, 250 ps and 500 ps, respectively.

FIG. 5: Schematic of the experimental set-up. $M_1$, $M_2$, $M_3$, $M_4$: silvered mirror, HWP: half wave plate, PBS: polarization beam splitter, $MO_1$, $MO_2$, $MO_3$, $MO_4$: microscope objective, $V_1$, $V_2$, $V_3$: V-grooves, BS: plate beam splitter, CP: compensator plate, P: pellicle (92:8), VA: variable attenuator, $L_1$: convex lens, CCD: charged coupled device, OSA: optical spectrum analyzer.

100 m length of CL fiber and the pump is launched in the $LP_{01}$ mode at 1064 nm. The pumping in the normal dispersion region makes the system free from soliton fission, modulation instability etc. which are adverse for the coherence. The generated spectrum at the end of the fiber for an average pump power of 47.5 mW is shown in Fig. 6(a). As the input pump power is well above the threshold value of Raman peak, noise-seeded cascaded Stokes wave is generated up to sixth order in the red-side of the input pump wavelength. In the blue-side of the pump, Raman anti-stokes and multiple additional peaks are also generated. The detailed investigation of the origin of all spectral peaks are discussed in.

FIG. 6: Experimental spectral coherence measurement using Michelson interferometry. (a) spectrum after 100 m CL fiber for average pump power 47.5 mW. (b) corresponding interference spectrum showing spectral fringe where the inset shows the fringe pattern in magnified form. (c) calculated first order mutual coherence as a function of wavelength.

44. Cascaded Raman and intermodal four-wave mixing is responsible for the generation of the broadband spectrum. The corresponding interference fringe pattern after
Michelson interferometer is shown in Fig. 6(b). Temporal delay is employed between the two arms of Michelson interferometer to attain high resolution over entire spectral range. Fig. 6(c) shows the calculated value of the first order mutual coherence followed by equation (14). It is observed that in the long pulse regime (input pump pulse width=0.77 ns), the red-side of the pump which is generated through noise-seeded cascaded Raman peaks, offers high spectral coherence. This agrees well with the theoretical predictions outlined before. Fig. 7(a) represents evolution of interference fringes at the output of Michelson interferometer after 100 m length of propagation for varying input pump power. The evolution of first order mutual coherence is depicted in Fig. 7(b). In the blue-side of the pump wavelength, spectrum is generated through cascaded intermodal-four wave mixing and offers poor spectral coherence. It is observed that Stokes peaks offer high spectral coherence and with the increase in pump power, number of Raman peaks increase which in turn broadens the coherent spectrum. The evolution of interference fringes for different fiber lengths are shown in Fig. 8(a). Average pump power is fixed at 50 mW. The corresponding spectral coherence is depicted in Fig. 8(b). It is observed that increase in fiber length generates cascaded Raman based broadband coherent spectrum.

Conclusion—We have investigated the detailed statistical evolution of spectral coherence of the noise-seeded Stokes wave in short and long pulse regimes in a unified manner. Simulation results largely reveal the undislosed coherence properties of the Stokes wave in long pulse regime. Noise-seeded Stokes pulse which offers poor spectral coherence in short pulse regime, exhibits gradual increment of spectral coherence with the increase in pulse width in long pulse regime. Experimental findings confirm the theoretical predictions where the Raman peaks offer high spectral coherence in long pulse operation. Increase in pump power and fiber length generates higher-order Raman peaks, resulting broadband coherent spectrum. The findings would enhance the prolonged ideas and will provide new insight.

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