# Bright–bright $2\pi$ solitons in stimulated Raman scattering

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We found conditions for excitation of bright–bright $2\pi$ solitons in stimulated Raman scattering that involves nutation of population at Raman quantum levels, for two (laser–Stokes) and three (e.g., laser–Stokes–anti-Stokes) components. The soliton components at all participating frequencies are bright solitons of the same, Lorentzian, shape, in contrast to the well-known bright–dark soliton combination in stimulated Raman scattering.

It is well known that stimulated Raman scattering (SRS) may result in the formation of peculiar solitons that combine a so-called bright (regular) soliton at the pump (laser) frequency and a so-called dark soliton (a deep minimum in the intensity profile) at the Stokes frequency. Mathematically identified as solitons in early research[1,2] and experimentally observed first in the research reported in Ref. 3, these formations (regarded also as transient solitons[4–6] having a $\pi$ phase jump at the Stokes frequency) have been extensively explored.[7] Following experimental observations, most of the theoretical research neglected the changes of populations at the Raman quantum transition. If the population at the Raman transition is an essential part of the pulse dynamics (and if there is a nonvanishing dispersion), it may result in $2\pi$ pulses (to the extent similar to self-induced-transparency solitons[8]) that consist in the case of only two components, of bright solitons at the pump and Stokes components, both having a surprisingly simple, Lorentzian envelope. Theoretically discovered many years ago,[2] these solitons have not been observed experimentally, which may be attributed to their threshold nature that imposes stringent limitations on the pulse frequency and length for the most of the materials traditionally used for SRS. In this Letter we suggest conditions for experimental excitation of these solitons and develop a theory for the case of three components (e.g., laser–Stokes–anti-Stokes or laser–Stokes–second-Stokes)[9] that may broaden possibilities of exploration of new solitons.

Consider a collinear propagation of radiation with frequencies $\omega_j = \omega_L + j\omega_0$, $j = 0, \pm 1$ (below we assign $j = -1$, $j = 0$, and $j = 1$ to the Stokes, $\omega_S$, laser, $\omega_L$, and anti-Stokes, $\omega_A$, components, respectively), where $\omega_0$ is the resonant frequency of a Raman quantum transition. In the plane-wave approximation the field components, $\mathrm{Re}[E_j(t,z)\exp(ik_jz - i\omega_jt)]$, have slowly varying envelopes, $E_j(t,z)$, where $z$ is the axis of propagation and $k_j = \omega_j n_j/c$, with $n_j = n(\omega_j)$ being the refractive index at frequency $\omega_j$. The Raman quantum transition is described by the density matrix with nondiagonal elements, $\rho_{12} = \rho_{21}^*$, and the difference $\Delta = \rho_{11} - \rho_{22}$ between populations of the lower (ground) level, $\rho_{11}$, and the upper (excited) level, $\rho_{22}$: $\rho_{11} + \rho_{22} = 1$. Assuming $\rho_{12} = i\sigma(t,z)\exp(ik_0z - i\omega_0t)$, where $k_0 = (k_L - k_S) \approx k_0 \equiv \omega_0/c$, and using, e.g., the procedure of Ref. 10 to find the respective nonlinear (Raman) polarizations, we obtain the Maxwell equations for the envelopes $E_j(t,z)$:

$$\partial E_j/\partial z + {\nu_j}^{-1}\partial E_j/\partial t = 2\pi\omega_j N_a(cn_j)^{-1}Q_j, \qquad j = S, L, A, \quad (1)$$

where $\nu_j = \mathrm{d}\omega_j/\mathrm{d}k_j$ is a group velocity at the frequency $\omega_j$, $N_a$ is the density number of Raman-active particles, $Q_S = -\alpha_{S,L}^*\xi_{S,L}^*\sigma^*E_L$, $Q_L = \alpha_{S,L}\xi_{S,L}\sigma E_S - \alpha_{L,A}^*\xi_{L,A}^*\sigma^*E_A$, $Q_A = \alpha_{L,A}\xi_{L,A}\sigma E_L$, $\xi_{j-1,j} = \exp[i(k_0 + k_{j-1} - k_j)z]$ is a phase mismatch factor, and

$$\alpha_{j-1,j} = \hbar^{-1}\sum_m[(\mathbf{d}_{1m}\mathbf{e}_j)(\mathbf{d}_{m2}\mathbf{e}_{j-1})(\omega_{m1} - \omega_j)^{-1} + (\mathbf{d}_{1m}\mathbf{e}_{j-1})(\mathbf{d}_{m2}\mathbf{e}_j)(\omega_{m1} + \omega_{j-1})^{-1}] \quad (2)$$

is the Raman polarizability[10,11] between the $j-1$ and $j$th components. Here $\mathbf{d}_{1m}$ ($\mathbf{d}_{m2}$) is a dipole matrix element for the resonant transitions between the lower (upper) Raman quantum level and the $m$th quantum level, $\omega_{m1}$ is the frequency of the $m \rightarrow 1$ transition, and $\mathbf{e}_j$ is a unity vector of the field polarization at the frequency $\omega_j$; the summation is performed over all the quantum levels except the Raman levels. Equation (1) (and the other equations below) are readily reduced to the two-component case by, e.g., setting $E_A = 0$. In Eq. (1) we neglected absorption. The generalized Bloch equations for $\sigma$ and $\Delta$ are

$$\partial\sigma/\partial t = -\tilde{\Omega}_R^*\Delta/2, \qquad \partial\Delta/\partial t = 2\,\mathrm{Re}(\sigma\tilde{\Omega}_R), \quad (3)$$

where $\tilde{\Omega}_R$ is the (in general, complex) Rabi frequency; using the approach of Ref. 10, we find it as

$$\tilde{\Omega}_R = 2\hbar^{-1}(\alpha_{S,L}\xi_{S,L}E_SE_L^* + \alpha_{L,A}\xi_{L,A}E_LE_A^*). \quad (4)$$

The first integral of Eqs. (3) is $\Delta^2 + 4|\sigma|^2 = \text{constant} = 1$. Because the solitons considered here are usually a few orders of magnitude shorter than the relaxation times of typical Raman transitions, the relaxation is neglected in Eqs. (3). Also, because the coherent lengths $(\tilde{k}_0 + k_{j-1} - k_j)^{-1}$ ($>> k_0^{-1}$) are usually much larger than the spatial

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size of the soliton (which is especially true in gases or vapors), we assume here that $\xi_{j-1,j} \approx 1$ (which is an exact relation on the case of only two components). As is usual in SRS theory, we neglected the Stark shift of the Raman frequency and related small corrections to the group velocities (see, e.g., Ref. 10).

A solitary wave, by definition, is a solution with all the envelopes $E_j$ propagating without change and with the same (unknown at this point) velocity $\tilde{\nu}$. Thus all these components are locked in (in contrast to the transient[4–6] bright–dark SRS solitons). Using retarded coordinates $\eta = t - z/\tilde{\nu}$ and $\zeta = z$ and stipulating that $\partial E_j/\partial\zeta = 0$, we transform Eq. (1) into

$$\delta_j \,\mathrm{d}\mathcal{E}_j \,\mathrm{d}\eta = \tilde{Q}_j, \qquad j = S, L, A, \tag{5}$$

where $\delta_j = 1/\nu_j - 1/\tilde{\nu}$ is a group-velocity dispersion parameter,[12] $\tilde{Q}_S = -w_{S,L}^*\sigma^*\mathcal{E}_L$, $\tilde{Q}_L = w_{S,L}\sigma\mathcal{E}_S - w_{L,A}^*\sigma^*\mathcal{E}_A$, $\tilde{Q}_A = w_{L,A}\sigma\mathcal{E}_L$, $w_{j-1,j} = 2\pi c^{-1}\alpha_{j-1}, N_a(\omega_{j-1}\omega_j/n_{j-1}n_j)^{1/2}$, and $\mathcal{E}_j \equiv E_j(n_jc/2\hbar\omega_j)^{1/2}$ are flux amplitudes, such that $\Phi_j = |\mathcal{E}_j|^2$ are photon fluxes of the respective components. Equations (3) and (5) yield two new integrals:

$$\sum \delta_j\Phi_j = \text{const.} \equiv I,$$
$$2(\delta_A\Phi_A - \delta_S\Phi_S) - \pi N_a\Delta(\eta) = \text{const.} \equiv J, \tag{6}$$

the first one being a Manley–Rowe-like integral. One can prove, using Eqs. (3) and (5), that the product $\sigma\tilde{\Omega}_R$ is real and that the phases of both $\sigma$ and $\tilde{\Omega}_R$ are constant, which allows one, without loss of generality, to replace $\tilde{\Omega}_R$ in Eqs. (3) with the real Rabi frequency and replace $\partial/\partial t$ with $\mathrm{d}/\mathrm{d}\eta$. The all-bright-solitons condition is that, for all the components $E_j(\eta) \to 0$, $\sigma(\eta) \to 0$ as $|\eta| \to \infty$. The solution for $\sigma$ and $\Delta$ in terms of the Rabi phase $\phi_R = \int \Omega_R(\eta)\,\mathrm{d}\eta$ is

$$\Delta = \pm\cos\phi_R, \qquad \sigma = (1/2)\sin\phi_R, \tag{7}$$

where the plus corresponds to the Raman system's being initially at the equilibrium $[\Delta(-\infty) = 1]$ and the minus corresponds to the initially inversed population difference $[\Delta(-\infty) = -1]$. In Eqs. (6), $I = 0$ and $J = -\pi N_a$, if the molecules were initially at the equilibrium, while $J = \pi N_a$, if the population difference was initially inverted.

Under appropriate conditions, Eqs. (3) and (5) allow for a solution with all the soliton components being the bright ones and of the same, Lorentzian, normalized shape: $S(\eta) = 1/(1 + \gamma^2\eta^2)$, where $\gamma \equiv 2/\tau$, $\tau$ being the duration of the soliton. When we introduce the total photon flux, $\Phi_\Sigma \equiv \sum\Phi_j$, the soliton solution in our terms is

$$\Phi_\Sigma = \Phi_0 S(\eta), \qquad \Delta(\eta) = \pm[1 - 2S(\eta)],$$
$$\sigma(\eta) = -\gamma\eta S(\eta), \tag{8}$$

where $\Phi_0$ is the peak total photon flux (see below). The solution for the Rabi phase is $\phi_R = 2c\tan^{-1}(\gamma\eta) + \pi/2$ $[\phi_R = 2\tan^{-1}(\gamma\eta) + \pi/2]$ in the case of the plus (minus) in Eq. (8) (assuming that the soliton peaks at $\eta = 0$), i.e., the total area (or Rabi phase) of the soliton is $\phi(\infty) - \phi(-\infty) = 2\pi$, indicating a $2\pi$ soliton, which is expected. (One can show that the Lorentzian solitons are the only possible solitons in the case of two and three components.) One has now $\Phi_j = |a_j|^2\Phi_\Sigma$, where $|a_j|^2 =$ constant ($\sum|a_j|^2 = 1$) are photon distribution coefficients (to be found). Substituting Eq. (8) into Eq. (5), we obtain a linear homogeneous set of equations for $a_j$ (with $\gamma$ as a parameter), which in the simplest case of only two components results in

$${\gamma_{(2)}}^2 = -|w_{S,L}|^2/\delta_S\delta_L \tag{9}$$

(see also Ref. 2) and in the three-component case (e.g., Stokes–laser–anti-Stokes) results in

$${\gamma_{(3)}}^2 = -(|w_{S,L}|^2/\delta_S\delta_L + |w_{L,A}|^2/\delta_L\delta_A). \tag{10}$$

[It is worth noting that one of the possible solutions of the Maxwell–Bloch equations is a continuous (i.e., nonsoliton) wave with two frequencies, e.g., $\omega_S$ and $\omega_A$, separated by $2\omega_0$, essentially similar to an optical balance[13] for four-wave mixing.] Using Eq. (5), we evaluate the coefficients $|a_j|^2$ for the two-component case as

$$|a_L|^2 = \delta_S/\delta_{SL} > 0, \qquad |a_S|^2 = -\delta_L/\delta_{SL} > 0, \tag{11}$$

where $\delta_{SL} = 1/\nu_S - 1/\nu_L$ and $\delta_{S(L)} = 1/\nu_{S(L)} - 1/\tilde{\nu}$, and for the three-component soliton as

$$|a_L|^2 = {\gamma_{(3)}}^2/W, \qquad |a_S|^2 = |w_{SL}|^2/{\delta_S}^2W,$$
$$|a_A|^2 = |w_{LA}|^2/{\delta_A}^2W, \tag{12}$$

where $W \equiv |w_{SL}|^2/\delta_S\delta_{SL} - |w_{LA}|^2/\delta_A\delta_{LA}$. Finally, the peak total photon flux, $\Phi_0$, is obtained from Eq. (6) for the two-component case as

$$\Phi_0 = \mp\pi N_a\delta_{SL}/\delta_S\delta_L, \tag{13}$$

where the signs correspond to those in Eqs. (8), and for the three-component soliton as

$$\Phi_0 \pm \pi N_a(|w_{SL}|^2/\delta_L\delta_{SL} - |w_{LA}|^2/\delta_A\delta_{LA})$$
$$/(|w_{SL}|^2/\delta_S - |w_{LA}|^2/\delta_A). \tag{14}$$

The total number of photons within the soliton pulse is $P_\Sigma \equiv \int_{-\infty}^{\infty}\Phi_\Sigma\,\mathrm{d}\eta = \pi N_a\tau/2$; for the two-component soliton,

$$P_\Sigma = \mp\pi N_a\delta_{SL}/(-\delta_S\delta_L)^{1/2}|w_{SL}|^2. \tag{15}$$

For the three-component soliton, $P_\Sigma$ is similarly obtained by Eqs. (10) and (14). All the values $\gamma^2, N_0$, and $|a_j|^2$ are positive. In the two-component case, this determines the following dispersion condition required for support of a soliton, if initially the particles are at the equilibrium:

$$\delta_{SL} > 0, \qquad \delta_S > 0, \qquad \delta_L < 0, \tag{16}$$

i.e., $\nu_S < \tilde{\nu} < \nu_L$; the reversed dispersion condition is required if initially the population was inverted. For the three-component soliton, if the Raman states are initially at the equilibrium the dispersion conditions are either

$$\delta_S > 0\,, \qquad \delta_L < 0\,, \qquad |\delta_A| > \delta_S |a_{L,A}|^2/|a_{S,L}|^2 \tag{17}$$

(in particular, $\nu_S < \tilde{\nu} < \nu_L$) or

$$\delta_A < 0\,, \qquad \delta_L > 0\,, \qquad |\delta_S| > -\delta_A |a_{S,L}|^2/|a_{L,A}|^2 \tag{18}$$

(in particular, $\nu_L < \tilde{\nu} < \nu_A$). This case provides one with much broader opportunities for choosing appropriate materials and lasers.

The major characteristic of the bright–bright SRS soliton is that it is a threshold soliton: its parameters must satisfy threshold conditions $\tau < \tau_{cr}$, $\Phi_0 > \Phi_{cr}$, and $P_\Sigma > P_{cr}$, where the respective threshold (critical) values for the two-component soliton are

$$\tau_{cr} = \delta_{SL}/|w_{SL}|\,, \qquad \Phi_{cr} = 4\pi N_a/\delta_{SL}\,,$$
$$P_{cr} = 2\pi N_a/|w_{SL}| = c\alpha_{SL}{}^{-1}(n_S n_L/\omega_S \omega_L)^{1/2}\,. \tag{19}$$

At the threshold, $1/\tilde{\nu} = (1/\nu_S + 1/\nu_L)/2$. Note that both $\Phi_{cr}$ and $P_{cr}$ depend only on spectral parameters and not on the concentration of Raman particles, since $\delta_{SL}, w_{SL} \propto N_a$. The threshold conditions for the three-component soliton are found in similar fashion.

The conditions, Eq. (19), impose stringent limitations on the parameters of both a medium and a driving field, in particular, on the soliton length $\tau$, which, to our knowledge, has not been addressed. Our estimates show that for most of the typical SRS media $\tau_{cr}$ may be as short as a few femtoseconds. Thus the major requirement is the selection of material and laser frequency. A natural choice is electronic SRS in alkali metal vapors, e.g., Cs vapor,[14] with its high (up to 50%) conversion efficiency. For the lasers used in Ref. 14 ($\lambda_L$ = 420–530 nm), one can show that $\tau_{cr} < 10$ fs, which is too short. $\tau_{cr}$ can be increased by resonant enhancement of $\delta_{SL}$. Such enhancement, however, is impossible if initially only the Cs ground level is populated. Indeed, the frequency of IR Stokes radiation is much lower than the frequency of any transition from the Cs ground level. On the other hand, if the laser frequency is close to the frequency of such a transition, then $\delta_{SL} < 0$ (this can be shown by the Sellmeier equation[11]). As a solution to this problem, we suggest tuning the pumping laser in such a way that the Stokes radiation is resonant to a $6p$–$5d$ transition and using another laser to populate the $6p$ level slightly. In particular, with 467.17-nm pumping ($\tilde{\nu} \approx 21\,406$ cm$^{-1}$, $\tilde{\nu}_S \approx 2870$ cm$^{-1}$) and the $6p_{3/2}$ population being ≈5% of that of the ground level, Eqs. (19) yield $\tau_{cr}$ ~8 ps and a critical area energy density of 0.02 J/cm$^2$. The SRS experiment[14] was conducted with a 1.3-ps, 500-$\mu$J dye laser focused confocally into a Cs vapor column 30 cm long. Our estimates show that all the conditions of Eqs. (19) would be fulfilled if an additional, 852.34-nm laser populated the Cs $6p_{3/2}$ level; this would also provide the control of the soliton. Another interesting opportunity could be presented by optical fibers, in which, because of larger dispersion, the limitations of Eqs. (19) on the soliton length could be relaxed.

In conclusion, we found conditions for excitation of bright–bright Lorentzian $2\pi$ solitons in SRS for two (laser–Stokes) and three (e.g., laser–Stokes–anti-Stokes) components. We also proposed an arrangement (including the medium and the laser) for the experimental observation of these solitons.

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## References


1. F. Y. F. Chu and A. C. Scott. Phys. Rev. A **12,** 2060 (1975); N. Tan-no, T. Shirahata, and K. Yokoto, and H. Inaba, Phys. Rev. A **12,** 159 (1975).
2. T. M. Makhviladze, M. E. Sarychev, and L. A. Shelepin, Sov. Phys. JETP **42,** 255 (1975); T. M. Makhviladze and M. E. Sarychev, Sov. Phys. JETP **44,** 471 (1976).
3. K. Druhl, R. G. Wenzel, and J. L. Carlsten, Phys. Rev. Lett. **51,** 1171 (1983).
4. C. R. Menyuk, Phys. Rev. Lett. **62,** 2937 (1989); Phys. Rev. A **47,** 2235 (1993).
5. D. J. Kaup, Physica **6D,** 143 (1983); **19D,** 125 (1986).
6. J. C. Englund and C. M. Bowden, Phys. Rev. Lett. **57,** 2661 (1986); Phys. Rev. A **42,** 2870 (1990); **46,** 578 (1992); P. C. MacPherson, R. C. Swanson, and J. L. Carlsten, Phys. Rev. Lett. **61,** 66 (1988); Phys. Rev. A **39,** 6078 (1989).
7. G. P. Agrawal and R. W. Boyd, eds., *Contemporary Nonlinear Optics* (Academic, New York, 1992), pp. 72–74.
8. S. L. McCall and E. L. Hahn, Phys. Rev. Lett. **18,** 908 (1967).
9. Bright–dark SRS solitons for more than two components (but with the change of population at the Raman transition neglected) were considered in J. R. Ackerhalt and P. W. Milonni, Phys. Rev. A **33,** 32 (1986); M. Scalora, S. Singh, and C. M. Bowden, Phys. Rev. Lett. **70,** 1248 (1993).
10. V. S. Butylkin, A. E. Kaplan, Yu. G. Khronopulo, and E. I. Yakubovich, *Resonant Nonlinear Interactions of Light with Matter* (Springer-Verlag, New York, 1989), pp. 206–209.
11. D. C. Hanna, M. A. Yuratich, and D. Cotter, *Nonlinear Optics of Free Atoms and Molecules* (Springer-Verlag, New York, 1979), Chap. 4.
12. The nonvanishing group-velocity dispersion is substantial. In the two-component case, for instance, the assumption of equal group velocity, $\nu_L = \nu_S$, even with the population change taken into account, would not result in the bright–bright soliton; see, e.g., H. Steudel, Physica **6D,** 155 (1983).
13. J. J. Wynne, Phys. Rev. Lett. **52,** 751 (1984); in *Multiphoton Processes,* S. J. Smith and P. L. Knight, eds. (Cambridge U. Press, Cambridge, 1988), p. 318.
14. R. Wyatt and D. Cotter, Opt. Commun. **37,** 421 (1981).