Subfemtosecond Pulses in Mode-Locked 2π Solitons of the Cascade Stimulated Raman Scattering

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We predict multifrequency 2π solitons in the cascade stimulated Raman scattering, mode locked by the dynamics of Raman quantum transition; they are "eigensolitons" of a general solitary wave solution. Soliton components at each individual frequency are "bright" pulses of the same, Lorentzian, shape; their interference gives rise to the train of well resolved high intensity subfemtosecond pulses.

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Generation of very short coherent pulses with high repetition rates has a great importance for various applications in both fundamental and applied physics. The shortest to date optical pulse length of 6 fs based on the pulse compression technique was achieved in 1987 [1]. New opportunity offered by a Fourier synthesizer [2] allows one to generate subfemtosecond pulses (SFP) by superposition of equidistant frequencies from separate lasers synchronized by nonlinear phase locking [3]. In this Letter we propose a new approach based on using multifrequency cascaded stimulated Raman scattering (CSRS) in which the equidistancy of frequencies is sustained automatically, and Raman components are mode locked within a 2π soliton reminiscent to the self-induced transparency (SIT) solitons [4] and related to the quantum dynamics at the Raman transition. We show that Raman active materials (with the transition frequency ω_0) can support solitons consisting of pump laser wave (with the frequency ω_L) and many Stokes and anti-Stokes components with their frequencies $\omega_j = \omega_L \pm j\omega_0$, $j = 1, 2, 3, \ldots$. These solitons have a new, very simple, Lorentzian intensity profile first found in Ref. [5] for two (laser + Stokes) components (and for three components in [6]). However, in contrast to [5,6], the new solitons consist of *many* mode-locked frequency components, whose coherent interference give rise to the train of ultrashort pulses (spaced by $2\pi/\omega_0$) with their length being as short as or even *shorter* than the pump cycle, $2\pi/\omega_L$. One can relate this effect (in which the 2π soliton plays a role of traveling shatter) to mode-locked laser pulses formed by the coherent interference of many modes. In the proposed effect, however, the pulse formation occurs at the time scale up to 5–6 orders of magnitude shorter than in the mode-locked lasers. The difference between this new phenomenon and the well known bright + dark SRS solitons [7] is that *all* the frequency components of the new soliton are *bright* pulses; all of them propagate with the same group velocity and, in the case of eigensolitons, have the *same* shape.

High-order CSRS required for the proposed phenomenon was first observed experimentally [8] and understood [8,9] in the early 1960s, with the total number of components up to ~10–20 and even higher [10]. The other required effect—"all-bright-SRS" solitons—has never, to the best of our knowledge, been observed in experiment. Their threshold nature stipulates properly chosen group dispersion, driving intensity and frequency, and sufficiently short driving pulse. Recent estimates [6] showed that the materials can be found to meet these requirements for two components, with the intensity requirements being very modest. Similarly, material and laser parameters could conceivably be found to satisfy the threshold conditions for CSRS solitons, with a reward for this effort being substantially high.

In the plane-wave approximation, the total CSRS field has $M = M_S + M_A + 1$ components (where M_S is the number of cascade Stokes and M_A the number of anti-Stokes components) colinearly propagating along the axis z, and can be written as $\mathrm{Re}[\sum_{j=-M_S}^{M_A} E_j(t,z)\exp(ik_j z - i\omega_j t)]$, where $E_j(t,z)$ is the envelope of the jth component, $k_j = \omega_j n_j/c$, and $n_j = n(\omega_j)$ is the refractive index at ω_j. The Raman quantum transition is descried 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, ρ_{11}, and upper (excited) level, ρ_{22}, $\rho_{11} + \rho_{22} = 1$. Assuming ρ_{12} in the form $\rho_{12} = (i/2)\sigma(t,z)\exp(i\tilde{k}_0 z - i\omega_0 t)$, where $\tilde{k}_0 = (k_{M_A} - k_{-M_S})/(M-1) \approx k_0 \equiv \omega_0/c$, the Raman-coupled Maxwell equations for the envelopes $E_j(t,z)$ are obtained by, e.g., using the procedure [11] as

$$\partial E_j/\partial z + v_j^{-1}\,\partial E_j/\partial t = \pi N_a \omega_j (\hbar c n_j)^{-1} \times \left(r_{j-1,j}\xi_{j-1,j}\sigma E_{j-1} - r_{j,j+1}^*\xi_{j,j+1}^*\sigma^* E_{j+1} \right), \quad (1)$$

where $v_j = d\omega_j/dk_j$ is a group velocity at ω_j [12]; $E_{-(M_S+1)} = E_{M_A+1} = 0$; N_a is the density number of Raman particles; $\xi_{j-1,j} = \exp[i(\tilde{k}_0 + k_{j-1} - k_j)z]$ is a phase mismatch factor, and $r_{j-1,j} = \sum_m [(\mathbf{d}_{1m}\cdot\mathbf{e}_j)(\mathbf{d}_{m2}\cdot\mathbf{e}_{j-1})(\omega_{m1} - \omega_j)^{-1} + (\mathbf{d}_{1m}\cdot\mathbf{e}_{j-1})(\mathbf{d}_{m2}\cdot\mathbf{e}_j)(\omega_{m1} + \omega_{j-1})^{-1}]$ is a nonlinear Raman coefficient [11,13]. Here $\mathbf{d}_{1m}$ ($\mathbf{d}_{m2}$) is a dipole moment of a transition between the lower (upper) Raman level and the mth quantum level, ω_{m1} is the frequency of $m \rightarrow 1$ transition, and $\mathbf{e}_j$ is the unity vector of polarization at ω_j; the summation is executed over all the quantum transitions except the Raman one. Equation (1) is valid when the "local" dispersion near each of the frequencies is much smaller than the "large

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scale" dispersion between any two adjacent frequencies which is usually the case in CSRS; we also neglected absorption assuming that all the CSRS frequencies are sufficiently far from resonances. The dynamics of the density matrix variables σ and Δ is governed by the generalized Bloch equations:

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

where $\tilde{\Omega}_R$ is a (in general, complex) "Rabi frequency" of the system; using, e.g., a "generalized two-level system" approach [11] it is found as

$$\tilde{\Omega}_R = (2/\hbar^2)\sum_{j=-M_S}^{M_A-1} r_{j,j+1}\xi_{j,j+1}E_jE_{j+1}^* . \tag{3}$$

The first integral of Eq. (2) is $\Delta^2 + |\sigma|^2 = \text{const} = 1$. Since the new solitons are usually a few orders of magnitude shorter than relaxation times of a typical Raman transition, the relaxation is neglected in Eq. (2). Similarly, since the coherent lengths $(\tilde{k}_0 + k_{j-1} - k_j)^{-1}$ $(\gg k_0^{-1})$ are usually much larger than the spatial length of the soliton (which is especially true in gasses or vapors), we will also assume $\xi_{j-1,j} \approx 1$ (for $M = 2$, i.e., laser + first Stokes, $\xi = 1$ is an exact relation). As usual in the SRS theory, we neglected in Eqs. (1) and (2) small corrections to the group velocities v_j and Stark shift of the Raman frequency ω_0, which if needed can easily be accounted for (see, e.g., [11,13]).

Defining a stationary wave (of which solitons are a particular case) as a solution with *all* the envelopes E_j, Δ, and σ propagating without change and with the same (unknown at this point) velocity $\tilde{v}$, using retarded coordinates $\eta = t - z/\tilde{v}$, $\zeta = z$, and stipulating $\partial E_j/\partial\zeta = 0$, we retain Eq. (2) (with $\partial/\partial t$ replaced by $d/d\eta$) and transform Eq. (1) into

$$\delta_j d\mathcal{E}_j/d\eta = w_{j-1,j}\sigma\mathcal{E}_{j-1} - w_{j,j+1}^*\sigma^*\mathcal{E}_{j+1}, \tag{4}$$

where $\delta_j = 1/v_j - 1/\tilde{v}$ is the group velocity dispersion parameter [12], $w_{j-1,j} = \pi r_{j-1,j}N_a(\omega_{j-1}\omega_j/n_{j-1}n_j)^{1/2} \times (c\hbar)^{-1}$, 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 (2) and (4) yield two more integrals:

$$\sum_{j=-M_S}^{M_A}\delta_j\Phi_j = \text{const} = I,$$
$$2\sum_{j=-M_S}^{M_A} j\delta_j\Phi_j(\eta) - \pi N_a\Delta(\eta) = \text{const} = J. \tag{5}$$

the first one being a Manley-Rowe-like integral. Amazingly, the set of $M + 2$ *nonlinear* equations, Eqs. (2) and (4), can be solved analytically. First, using Eq. (4) for evaluating $d(\mathcal{E}_j^*\mathcal{E}_{j+1})/d\eta$, and thus $d\tilde{\Omega}_R/d\eta$ due to Eq. (3), we find out, by using Eqs. (2) and (3), that $d(\sigma\tilde{\Omega}_R)/d\eta$ and thus $\sigma\tilde{\Omega}_R$ are real functions. Multiplying the first of Eqs. (2) by σ^*, we obtain that the phase of σ is invariant, i.e., if $\sigma = \rho_\sigma\exp(i\phi_\sigma)$ (with ρ_σ and ϕ_σ real), $\phi_\sigma = \text{const}$. [Applying this again to Eq. (4), starting from $j = -M_S$, one can see that all the cascade components of the stationary wave are phase locked to each other.] Without loss of generality, we can assume now $\phi_\sigma = 0$, i.e., that both σ and Ω_R are real functions. Using Eqs. (2) and the first integral, $\Delta^2 + |\sigma|^2 = 1$, Δ and σ can then be expressed as

$$\Delta = \pm\cos\phi_R, \qquad \sigma = \sin\phi_R. \tag{6}$$

where $\phi_R \equiv \int\Omega_R(\eta)\,d\eta$ is a Rabi phase, and the upper sign corresponds to the atoms being initially ($\eta \to -\infty$) at the equilibrium, $\Delta(-\infty) = 1$, whereas the lower sign to the an inverse population, $\Delta(-\infty) = -1$, at $\eta \to -\infty$. Introducing $\psi \equiv \int\sigma\,d\eta$, we reduce Eq. (4) to the set of M *linear* differential equations for the envelopes $\mathcal{E}_j$ by writing it as $\delta_j d\mathcal{E}_j/d\psi = w_{j-1,j}\mathcal{E}_{j-1} - w_{j,j+1}^*\mathcal{E}_{j+1}$. Once the eigenvalues γ_k of this set are evaluated, its solution is immediately found as

$$\mathcal{E}_j(\psi) = \sum_{k=1}^{M} c_{jk}e^{\gamma_k\psi}, \qquad c_{jk} = \text{const}. \tag{7}$$

Out of M^2 constants c_{jk} in Eq. (7), only M are truly independent. For the odd M's ($M \geq 3$) one of the eigenvalues is $\gamma = 0$ [14]. The equation for nonzero γ's have the form of polynomial in γ^2. Its solution in the simplest case of only two components, $M = 2$ (i.e., laser + Stokes) [5], is $\gamma^2 = -|w_{S,L}|^2/\delta_S\delta_L$; in the case $M = 3$ (e.g., Stokes + laser + anti-Stokes, or two Stokes + laser, etc.) it is [6] $\gamma^2 = -(|w_{S,L}|^2/\delta_S\delta_L + |w_{L,A}|^2/\delta_L\delta_A)$, whereas in the cases $M = 4$ and $M = 5$ it is found as $\gamma^2 = -(B_1 \pm (B_1^2 - B_2)^{1/2})$, where $B_1 = \sum_j b_{j,j+1}$, with $b_{j,k} \equiv |w_{j,k}|^2/\delta_j\delta_k$, and for $M = 4$, $B_2 = b_{-M_S,-M_S+1}b_{M_A-1,M_A}$, while for $M = 5$, $B_2 = b_{-M_S,-M_S+1}b_{M_A-2,M_A-1} + b_{-M_S,-M_S+1}b_{M_A-1,M_A} + b_{-M_S+1,M_S+2}b_{M_A-1,M_A}$, etc.

Using the Maxwell equations solution, Eq. (7), and noting that the Rabi frequency, $\Omega_R = (4/\pi N_a)\sum w_{j,j+1}\mathcal{E}_j\mathcal{E}_{j,j+1}^*$, can now be expressed as

$$\Omega_R(\psi) = \sum_{m,k=1}^{M} Q_{mk}e^{(\gamma_m+\gamma_k^*)\psi} \tag{8}$$

with $Q_{mk} = (4/\pi N_a)\sum_{j=1}^{M} w_{j,j+1}c_{jk}c_{j+1,m}^*$, $Q_{mk} = Q_{mk}^*$, we reduce the Bloch equations, Eq. (2), to

$$d^2\psi/d\eta^2 + [1 - (d\psi/d\eta)^2]^{1/2}\Omega_R(\psi) = 0, \tag{9}$$

whose general solution is readily found in the quadrature form:

$$\int\left(1 - \left[\int\Omega_R(\psi)\,d\psi\right]^2\right)^{-1/2} d\psi = \eta. \tag{10}$$

Once Eq. (10) is solved for $\psi(\eta)$, one obtains $\sigma = d\psi/d\eta$, $\Delta = \sqrt{1-\sigma^2}$, and, through Eq. (7), all the envelopes $\mathcal{E}_j$. In general, Eq. (10) gives rise to a rich family of solutions: solitary waves, stationary traveling fronts, bright and dark solitons, periodic stationary waves, etc. The solutions attributed to more than one eigenvalue γ may be regarded as higher-order solitons. The simplest and most fundamental are eigensolitons, i.e., those attributed to a *single* eigenvalue γ. (For $M = 2$ and $M = 3$ they are only feasible as all-bright solitons [5,6].) In such a case, all the coefficients c_{jk} but one in Eq. (7) vanish, such that $\mathcal{E}_j = \tilde{c}_j \exp(\gamma\psi)$. Thus, all the components of the eigenwave have the same time dependence. Of special interest is a *bright* soliton, i.e., solitary eigenwave with its CSRS components vanishing at the edges,

$$\mathcal{E}_j(\eta) \to 0 \quad \text{as } |\eta| \to \infty. \tag{11}$$

All these components have the same temporal profile, $|\mathcal{E}_j|^2 \propto S(\eta)$, found from Eq. (10) as

$$S(\eta) = \sin^2\phi(\infty)/2\{\cosh[2\tau^{-1}(\eta - \eta_0)\sin\phi(\infty)] - \cos\phi(\infty)\}; \tag{12}$$
$$\Omega_R = 4S(\eta)/\tau,$$

where η_0 can be assumed zero, $\phi(\infty)$ is the initial Rabi phase, and $\tau \equiv 1/\mathrm{Re}(\gamma)$ is the soliton length. The area of the eigensoliton is $\Delta\phi_R \equiv \phi_R(\infty) - \phi_R(-\infty) = 2[\pi - \phi(\infty)]$, i.e., not necessarily an integer of π, a situation unusual for SIT-like solitons. In the limiting case, $\phi_R = \pi/2$ (i.e., $\Delta\phi_R = \pi$, which corresponds to a π soliton), the eigensoliton has profile $S(\eta) = \frac{1}{2}\cosh(2\eta/\tau)$. However, this soliton is due to the most unlikely initial conditions: $\Delta(-\infty) = 0$ and $\sigma(-\infty) = -1$. For the "intermediate" solitons with $0 < \phi(\infty) < \pi/2$, the initial state of the system is $\Delta(-\infty) = \pm\cos\phi(\infty)$, and $\sigma(-\infty) = -\sin\phi(\infty) \neq 0$. Although to prepare initial states with $\sigma \neq 0$ may not be an easy experimental task, they are still feasible (even if the finite relaxation is taken into consideration) if another short pulse is used as a precursor. Fortunately, however, a regular initial state of the system [i.e., $\Delta(-\infty) = \pm 1$, $\sigma(-\infty) = 0$] can also give rise to the eigensoliton with $\phi(\infty) = 0$, i.e., 2π soliton ($\Delta\phi_R = 2\pi$); it has a very simple, Lorentzian profile:

$$S(\eta) = (1 + 4\eta^2/\tau^2)^{-1}, \quad \Delta = \pm[1 - 2S(\eta)], \quad \sigma = 4(\eta/\tau)S(\eta), \tag{13}$$

which again is unusual for SIT solitons. Under the condition Eq. (11), in Eq. (5) $I = 0$, while $J = -\pi N_a$ if initially the two-level Raman system is at equilibrium [i.e., $\Delta(-\infty) = 1$; upper sign in Eq. (6)], and $J = \pi N_a$ if the population difference was initially inversed [i.e., $\Delta(-\infty) = -1$; lower sign in Eq. (6)].

Each eigensoliton has a unique set of M components characterized by photon distribution eigencoefficients, $\alpha_j = \text{const}$, $\sum|\alpha_j|^2 = 1$, i.e., $\mathcal{E}_j(\eta) = \alpha_j\sqrt{\Phi_\Sigma}$, or $\Phi_j = |\alpha_j|^2\Phi_\Sigma$, where $\Phi_\Sigma \equiv \sum\Phi_j = \Phi_{\mathrm{pk}}S(\eta)$, with Φ_{pk} being a peak total photon flux. Using Eq. (4), $|\alpha_j|^2$ is evaluated as $|\alpha_j|^2 = |\beta_j|^2/\sum_{j=-M_S}^{M_A}|\beta_n|^2$, where $\beta_{-M_S-1} = 0$, $\beta_{-M_S} = 1$, and for $M_A \geq j > -M_S$, $\beta_{j+1} = (w_{j-1,j}\beta_{j-1} - \gamma\beta_j\delta_j)/w^*_{j,j+1}$. Using Eq. (9), Φ_{pk} is obtained as

$$\Phi_{\mathrm{pk}} = \mp\pi N_a / \sum_{j=-M_S}^{M_A} j\delta_j|\alpha_j|^2, \tag{14}$$

with the signs corresponding to those in Eq. (6). The total number of photons in the soliton is

$$P_\Sigma \equiv \int_{-\infty}^{\infty}\Phi_\Sigma\, d\eta = \pi\Phi_{\mathrm{pk}}/2\mathrm{Re}(\gamma). \tag{15}$$

With P_Σ or τ given one can find the velocity of the soliton $\tilde{v}$. The conditions $\mathrm{Re}(\gamma^2) > 0$ and $\Phi_{\mathrm{pk}} > 0$ impose limitations on the dispersion and nonlinearity of the system. For $M = 2$ (and initially equilibrium Raman states) the dispersion conditions are [5,6] $\delta_S > 0$, $\delta_L < 0$, i.e., $v_S < \tilde{v} < v_L$ (if the population is initially inversed, the signs are to be inversed, too); for $M = 3$, see [6]. For $M = 2$, $|\alpha_L|^2 = 1 - |\alpha_S|^2 = 1/(1 - \delta_L/\delta_S)$, $\tau \equiv 1/\gamma = \sqrt{-\delta_S\delta_L}/r_{SL}\pi N_a$, and $P_\Sigma = (\pi/r_{SL})\delta_{SL}/\sqrt{-\delta_S\delta_L}$, where $\delta_{SL} \equiv \delta_S - \delta_L = 1/v_S - 1/v_L$. The soliton length τ, peak photon flux Φ_{pk}, and the total number of photons P_Σ must satisfy some threshold conditions. The critical length τ_{cr} for $M = 2$ could be in the range from a few tens of femtoseconds to a few picoseconds, depending on material, whereas the critical energy density for, e.g., Cs vapor [6] could be ~ 0.02 J/cm^2.

To fully characterize CSRS soliton with $M \gg 1$, in particular the spectral distribution of the Stokes and anti-Stokes components in it, for a *specific* material, one has to account for a great number of nonlinear spectral characteristics of the material in a very wide range of frequencies, from the far infrared to far ultraviolet. Instead, in order to illustrate that the SFPs may appear for virtually *any* CSRS spectral distribution, we choose here three substantially different model spectral distributions whereby the photon fluxes (a) are equally distributed between the CSRS components, $\Phi_j = \text{const}$; (b) peak in the middle of the CSRS spectrum and fall off parabolically toward both of its ends, $\Phi_j/\Phi_{\max} = 1 - 4(j - j_{\max})^2/M^2$, where $j_{\max}$ is the position in the middle of the CSRS spectrum (which is usually below the driving frequency); and (c) vanish in the middle of the CSRS spectrum and rise up parabolically toward both of its ends, $\Phi_j/\Phi_{\max} = 4(j - j_{\max})^2/M^2$. We choose a line with $\lambda_0 \approx 2.4$ μm (as in a hydrogen gas) pumped by the third harmonics of Ti:Sp laser ($\lambda_L \approx 0.28$ μm), with 12 components. The oscillations in time domain for all these cases are depicted in Fig. 1. In our simulation we assumed that the spacing between the pulses is much shorter than the soliton length τ (slight irregularities seen in Fig. 1 are due to incommensurability of ω_L and ω_0). Distinct SFPs are evident in all the cases, and they are well separated. The length of each individual pulse is ~ 0.218 fs for the distribution (a), ~ 0.225 fs for (b), and ~ 0.199 fs for (c), although, as expected, the pulses with the most inhibited background correspond to the distribution (b), and with the least to (c). Assuming a significant portion of energy of the original laser pulse to be trapped within 2π CSRS soliton, there is a good potential for attaining high intensity SFP by compressing laser pulse into the SFP train. For the above example, assuming a 100 fs long laser pulse with the cross section 10^{-4} cm^2 and energy 10^{-3}–10^{-2} J, of which about 10% get trapped into a CSRS soliton of the same length, and that the major part of radiation is concentrated in SFP, as in case (b), one obtains the peak pulse intensity of the

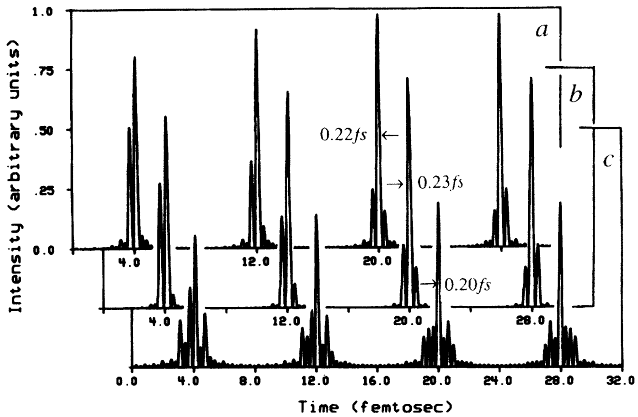

FIG. 1. Subfemtosecond pulses (intensity vs time) in a mode-locked CSRS soliton when the photon fluxes of the CSRS components (a) are equally distributed between the components, (b) peak in the middle of the CSRS spectrum and fall off toward both of its ends, and (c) vanish in the middle of the spectrum and rise up toward both of its ends.

same order of magnitude as the original laser pulse, i.e., 10^{14}–10^{15} W/cm^2—and even higher for better laser systems. Since in real Gaussian beams the anti-Stokes components are usually radiated away in the far-field area as narrow cones, the SFPs should be better observed in the near-field area; however, even if only Stokes components are left in the output beam, the pulse will be broadened by only 10%–20%.

In conclusion, we demonstrated that Raman-coupled multiple CSRS components can form a 2π soliton, with the coherent interference of mode-locked components giving rise to the SFP train of large intensity.

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