Theory of Pump Depletion and Spike Formation in Stimulated Raman Scattering

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By using the inverse spectral transform, the stimulated Raman scattering equations are solved and the explicit output data are given for arbitrary laser pump and Stokes seed profiles injected on a vacuum of optical phonons. For long duration laser pulses, this solution is modified so as to take into account the damping rate of the optical phonon wave. This model is used to interpret the experiments of Drühl, Wenzel, and Carlsten [Phys. Rev. Lett. **51**, 1171 (1983)], in particular, the creation of a spike of (anomalous) pump radiation. The related nonlinear Fourier spectrum does not contain discrete eigenvalues, hence this *Raman spike* is not a soliton.

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Stimulated Raman scattering (SRS) is one of the most studied three-wave interaction processes in nonlinear optics not only because it retains all the ingredients of any other stimulated process, but also because it has revealed many striking and sometimes unexplained phenomena. The theory of SRS has been developed on a semiclassical basis, for instance, by Shen and Bloembergen [1], Wang [2], or Carman *et al.* [3] by assuming a permanent pump intensity profile. But when "depletion of the laser power implies that the laser field may not be treated as a fixed constant parameter" [1], Stokes generation and amplification induces pump depletion, and it is a serious obstacle to the propagation of high intensity laser pulses in a Raman-active medium. However, it can also be thought of as a means to study (experimentally and theoretically) the fundamental properties of matter and radiation, and indeed Raymer and Mostowski [4] predict large (macroscopic) fluctuations of the Stokes pulse energy that are reminiscent of the small (quantum) fluctuations of the material dynamical variable (the polarization density state variable). These predictions were then checked on measurements of the statistical distribution of Stokes pulses in pressured H_2 gas by Wamsley and Raymer [5] and also by Fabricius, Natterman, and von der Linde [6].

At the same time, experiments of Drühl, Wenzel, and Carlsten [7] for long duration pump pulses (order of 100 ns) revealed that "occasionally the pump depletion is anomalously reversed for a short time interval generating a spike of pump radiation." They call this a *soliton* referring to the soliton solution of the undamped SRS equations given by Chu and Scott [8]. The lesson of these results is the spectacular fit of the experimental data to numerical simulations of the SRS equations, indicating that the model is quite adequate. Then the fundamental result is the discovery that the *spike* in the pump depletion occurs when a π phase shift is introduced in the Stokes seed.

In the absence of phase flip, the spike formation is still observed (on real experiments, not on simulations) but with much lower probability. This led Englund and Bowden [9] to propose that the spike is the macroscopic manifestation of quantum phase fluctuations of the Stokes wave. The subsequent experiments of MacPherson, Swanson, and Carlsten revealed that the anomalous pump radiation spike (what they call a *Raman soliton*) occurs in about 10.1% of the shots [10]. Englund and Bowden [11] developed a complete theoretical basis of such Raman spikes generated by quantum fluctuations of the initial Stokes vacuum, and they obtained a reasonable qualitative agreement with the previous experiments (they found a spontaneously generated spike in 13.6% of the shots).

The observed spikes of pump radiation acquire then importance also for fundamental studies, and they are always referred to as being solitons [12]. But it was already remarked on the first series of numerical simulations that the spike narrowing (as propagation distance is increased) indicates that they are "not solitary waves in the strict sense" [7]. In particular, the nonzero velocity of the Raman soliton implies, as shown by Menyuk, that "any solitonlike structures are subluminous and will ultimately disappear at the back end of the pulse" [13].

Consequently, the problem of the theoretical interpretation of the experiments of Drühl, Wenzel, and Carlsten [7] is still open, and we solve it here in terms of the inverse spectral transform theory (IST) extended to arbitrary boundary values [14]. We obtain the explicit global solution (output laser pulse) which maps perfectly the numerical simulation of SRS (as performed by MacPherson, Carlsten, and Druhl [15]).

Such an explicit analytic formula for the output [Eqs. (7) and (8) below] is important for physics in many aspects.

(1) It allows one to understand why the Stokes phase flip and the finiteness of the dephasing time (long pulses) are *both* essential to the spike formation, and to discover the precise nonlinear mechanism generating the Raman spike.

(2) It provides a powerful tool to analyze the experiments. Indeed, having the digitalized input, our formula readily gives the predicted output, and a comparison

with the data of [7] will be published later with other details [16].

(3) It unveils the nature of the Raman spike as its nonlinear Fourier spectrum does not contain isolated points (bound states) but consists only in the continuum (radiation). Apart from the fact that the spike is not a soliton, the important consequence of this is that it survives long propagation distances, which is important for applications.

The last point to mention is the fact that our solution, although being obtained from the infinite line case, is quite close to the finite line case solution (obtained through numerical simulations). This is easily seen by comparing, for instance, our Fig. 2 to the Fig. 2 of [15]. The reason for this is that the input data are of finite duration and the interaction is local (for any fixed time). We show in [16] that indeed the generated material excitation is localized in a small region (of length comparable with the duration of the amplified Stokes pulse).

The model of SRS can be taken, for instance, from [11] and reads, if we neglect the ground state depletion ($R_3 = -N$),

$$\partial_\zeta A_L = K_{LS} R A_S\,, \qquad \partial_\zeta A_S = -K_{LS}^* R^* A_L$$
$$\partial_\tau R + RL/(cT_2) = -K_{LS} N A_L A_S^*\,. \tag{1}$$

The spatial variable ζ lies in $[0, L]$, where L is the total beam path in the Raman cell, and the retarded time $\tau = t - \zeta/c$ is positive. A_L and A_S are the slow envelopes of the laser pump (frequency ω_L) and of the Stokes emission (frequency ω_S) which stimulate the material dynamical variable R (optical phonon, frequency $\omega_P = \omega_L - \omega_S$). K_{LS} is the complex coupling constant, T_2 is the mean collisional dephasing time, and N is a scaled density ($= \rho AL$ with ρ the density of Raman-active molecules and A the effective cross sectional area of the pump beam). The initial-boundary value problem associated to the system (1) is the following:

$$R(\zeta, 0) = 0\,, \quad A_L(0,\tau) = A_{L0}(\tau)\,, \quad A_S(0,\tau) = A_{S0}(\tau)\,, \tag{2}$$

where, to reproduce the experiments, A_{L0} is a Gaussian and A_{S0} is a fraction of A_{L0} with possibly a change of sign (phase flip) somewhere. The problem is to determine the output quantities $A_L(L,\tau)$ and $A_S(L,\tau)$.

Our model results from (1) by considering first an infinite line ($L \to \infty$) and taking into account the mismatched wave number of value $2k = k_P + k_S - k_L$, which results, for instance, from the Doppler effect due to molecular thermal motions. The resulting system is (1) but with A_S replaced by $A_S \exp[-2ik\zeta]$. To take into account the contributions of all values of k, we introduce the distribution $g(k)$ (centered in $k = 0$) of the relative coupling intensities. The resulting model equations are

$$\partial_x a_1 = q a_2\,, \qquad \partial_x a_2 - 2ik a_2 = -\bar{q} a_1\,,$$
$$\partial_t q + \gamma q = \int dk\, g(k) a_1 \bar{a}_2\,. \tag{3}$$

Here and in the following, an integral with no specified boundaries stands for $(-\infty, +\infty)$. Here we have made the following change of variables and scalings:

$$x = -\zeta\,, \quad t = \tau\,, \quad q = -K_{LS} R\,, \quad \gamma = L/(cT_2)\,, \tag{4}$$

$$(k = 0): \qquad a_1 = A_L/A_0\,, \quad a_2 = A_S e^{2ikx}/A_0\,, \tag{5}$$

where $A_0 = \max\{A_L(0,\tau)\}$. The distribution $g(k)$ is actually related to the inhomogeneous broadening and can be normalized to the coupling constant by setting $\int dk\, g(k) = K_{LS}^2 N |A_0|^2$. The related initial-boundary value problem corresponding to (2) reads here [note the sign in (4)],

$$q(x,0) = 0\,, \qquad a_1(k, +\infty, t) = I_1(k,t)\,,$$
$$a_2(k, +\infty, t) = I_2(k,t) e^{2ikx}\,, \tag{6}$$

where for $k = 0$ $I_1(t) = A_{L0}(\tau)/A_0$ and $I_2(t) = A_{S0}(\tau)/A_0$. The problem to be solved now is to compute the output data $a_1(k, -\infty, t)$ and $a_2(k, -\infty, t)$.

Note that, although t represents the retarded time, the initial value problem is physically meaningful because, the medium being initially in the ground state, we have set $q(x,0) = 0$. Another important remark is that the model (3) maps onto (1) in the limit when $g(k)$ becomes the Dirac distribution $\delta(k)$. However, on the infinite line, this is a singular limit [14] [in short it is not compatible with $q(x,t) \to 0$ as $x \to \pm\infty$], and hence $g(k)$ can be as sharp as we want but never a true delta function (which is physically quite reasonable).

It has been shown in [14] that the above initial-boundary value problem is solvable for $\gamma = 0$ and we shall use these results directly here. The solution is given by

$$a_1(k, -\infty, t) = I_1/\bar{\beta} + I_2 \bar{\alpha}/\bar{\beta}\,, \tag{7}$$

$$a_2(k, -\infty, t) e^{-2ikx} = I_2/\beta - I_1 \alpha/\beta\,. \tag{8}$$

The coefficients $\alpha(k,t)$ and $\beta(k,t)$ (the *spectral data*) can be computed explicitly from [14] and read for $\gamma = 0$,

$$\alpha(k,t) = -\pi g(k) e^{\phi(k,t)} \int_0^t dt'\, \bar{I}_1(k,t') I_2(k,t') e^{-\phi(k,t')}, \tag{9}$$

$$\phi(k,t) = \int_0^t dt' \left[\frac{1}{2} \pi g(k) U(k,t') - \frac{i}{2} \int g(\lambda) U(\lambda, t') \right], \tag{10}$$

$$U(k,t) = |I_1(k,t)|^2 - |I_2(k,t)|^2, \tag{11}$$

$$\beta(k,t) = \sqrt{1 + |\alpha(k,t)|^2}\, e^{i\theta(k,t)}, \tag{12}$$

$$\theta(k,t) = -\frac{1}{2\pi} \int (1 + |\alpha(\lambda,t)|^2)\,, \tag{13}$$

where the slashed integral denotes the Cauchy principal value. Analogous formulas can be found in [17] but in the context of resonant interaction of light with a two-level medium (and application to superfluorescence) for which the boundary value problem notably differs. The above result gives the exact solution to SRS for short pump pulses (for which $\gamma \sim 0$). We will report the discussion of this case in a forthcoming paper and consider now the case of long pump pulses for which the pump depletion can be anomalously reversed [7].

Our main argument is that both pump depletion and spike formation are described by the above solution of the boundary-value problem (6) for the system (3). Indeed, considering (7) and (12), we remark that

$$\alpha \to \infty \Rightarrow |\beta| \to \infty \Rightarrow |a_1(-\infty)| \to |I_2|, \quad (14)$$

$$\alpha \to 0 \Rightarrow |\beta| \to 1 \Rightarrow |a_1(-\infty)| \to |I_1|. \quad (15)$$

Then pump depletion will occur in the time region where $\alpha(k,t)$ is large (the pump output will be of the order of the Stokes *input* I_2), and pump radiation will occur in the time region where $\alpha(k,t)$ is close to zero (the pump output will be of the order of the pump *input* I_1).

In the case of long duration pump pulses, the effect of the dephasing time is included in our model by assuming instead of the evolution (9) the following:

$$\alpha(k,t) = -\pi g(k) e^{\phi(k,t)} \int_0^t dt' \, \bar{I}_1(k,t') I_2(k,t') \times e^{-\phi(k,t')} e^{\gamma(t'-t)}. \quad (16)$$

The added exponential factor above is justified by considering the linear limit in the spectral transform context. To that end it is convenient to write down the solution of (3) given by the spectral transform method [14],

$$a_1 = I_1 f_1 + I_2 e^{2ikx} f_2, \quad (17)$$

$$a_2 = -I_1 \bar{f}_2 + I_2 e^{2ikx} \bar{f}_1, \quad (18)$$

$$q = -\frac{1}{\pi} \int dk \, \bar{\alpha} e^{-2ikx} f_1, \quad (19)$$

where f_1 and f_2 are the solutions of

$$f_1(k,x,t) = 1 + \frac{1}{2i\pi} \int \frac{d\lambda}{\lambda + i0 - k} \times \alpha(\lambda,t) e^{2i\lambda x} f_2(\lambda,x,t), \quad (20)$$

$$f_2(k,x,t) = \frac{1}{2i\pi} \int \frac{d\lambda}{\lambda - i0 - k} \bar{\alpha}(\lambda,t) e^{-2i\lambda x} f_1(\lambda,x,t).$$

With the linear limit, obtained from the above solution by taking simply $f_1 = 1$, it can be verified that the evolution for q in (3) with $\gamma \neq 0$ has precisely the solution (16). We remark at this stage that the time evolution of the nonlinear Fourier transform $\alpha(k,t)$ bears the *linear* character of the evolution. Hence the nonlinearity enters only in the output expressions (7) and (8) through the nonlinear combinations of α with $I_{1,2}$.

In order to realize how the mechanism of pump depletion and spike formation is allowed by Eq. (16), we deal with all quantities evaluated at $k = 0$ [corresponding to a very sharp $g(k)$]. Then we assume a Gaussian shaped laser pump input [from (5) the amplitude is normalized to 1] and a small proportion of Stokes seed with possibly a π phase shift,

$$I_1(t) = \exp[-(t - t_1)^2/\tau_1^2], \quad I_2(t) = \tanh[(t - t_0)/\tau_0] I_1(t)/\Gamma. \quad (21)$$

With such input data, the pump repletion can be physically understood as resulting from a reversal of the Raman gain due to the change of sign of the Stokes input. This is precisely the behavior which is described by formula (16) where, if I_2 changes sign then α starts to decrease and the pump repletion limit (15) is approached.

We have drawn the energy $|a_1|^2$ of the pump output given by (7) in Figs. 1 and 2, for the above choices of $I_{1,2}$ with the parameter values $t_1 = 50$, $\tau_1 = 45.5$, $\Gamma = 10$, $g(0) = 100$, $\gamma = 160$, and no Stokes phase flip in Fig. 1 (that is $t_0 = 0$) and a phase flip in $t_0 = 50$ for Fig. 2 (with $\tau_0 = 1$). These parameters correspond in the physical world to a pump pulse of maximum amplitude A_0 of 6.36×10^6 V m^{-1} and a Stokes seed of 0.636×10^6 V m^{-1}, for the values (taken from Ref. [11]) for τ in nanoseconds: $N = 8.5 \times 10^{19}$, $|K_{LS}| = 5.8 \times 10^{-17}$, and $T_2 = 0.625$ ns. Consequently, from $\gamma = L/(cT_2) = 160$ the data would correspond to a beam path of 30 m.

We can now easily understand the different behaviors of the pump output in Figs. 1 and 2 just by inspection of (16). Indeed, in the first zone (up to 30 ns), α is small and we have no depletion. Then the growth of α (pump depletion) results from the factor $\exp[\phi]$ in (16) because U given by (11) is *positive*. Now if I_2 in (11) does not change sign (Fig. 1), α increases to when the damping term dominates again and we observe the pump radiation again (right-hand side hump in Figs. 1 and 2). If instead we introduce a phase flip in the Stokes seed, then the integral in (11) makes α decrease and possibly vanish, which constitutes the mechanism for reversal of pump depletion. However, this is allowed only if the growth of α with $\exp[\phi]$ is not too fast, and enters the role of the damping term $\exp[-\gamma t]$ here. Hence the spike of pump

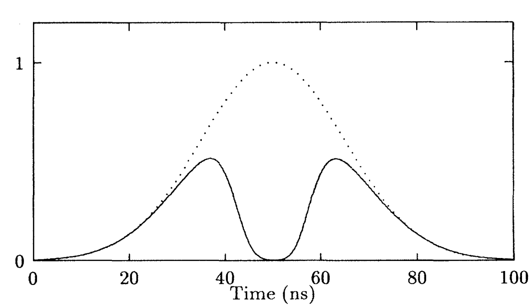

FIG. 1. Pump energy profile $|a_1|^2$ at $x = +\infty$ (input, dashed line) and at $x = -\infty$ (output, solid line) with unflipped phase of the Stokes seed.

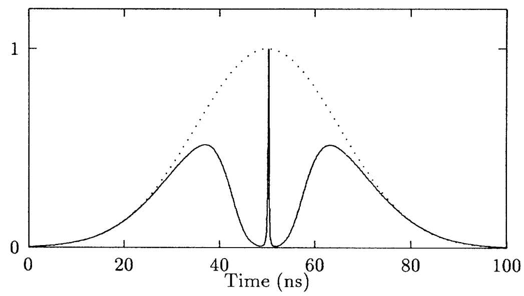

FIG. 2. Pump energy profile $|a_1|^2$ at $x = +\infty$ (input, dashed line) and at $x = -\infty$ (output, solid line) when a π phase shift of the Stokes seed is introduced at 50 ns.

radiation occurs as a result of the Stokes phase flip via a balance between Stokes amplification and optical phonon damping.

In conclusion, we have obtained the following set of results.

(1) An exact and explicit solution to the SRS equations for short pulses ($\gamma = 0$).

(2) An approximate solution for long pump pulses which describes perfectly in a unique formalism the pump depletion and the formation of a spike of pump radiation when the Stokes seed is given phase flip.

(3) The proof that the spike of pump radiation occurs as a balance between Raman gain and phonon damping, and that it can survive long propagation distances.

(4) A new mathematical structure, the *Raman spike*, related to zero of the reflection coefficient [our $\alpha(k,t)$], and which, in the IST scheme, is part of the continuous spectrum.

(5) The proof that the spike of pump radiation is not a soliton, namely, that it is not related to a discrete part of the (nonlinear Fourier) spectrum.

(6) An explicit formula for the description of transient SRS which can be used to study, for instance, the decay of the Raman spike, the generation of multispikes, the result of a stochastic phase in the generated Stokes wave, etc. These studies will be reported elsewhere (see [16]).

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