THEORY OF PUMP DEPLETION AND SPIKE FORMATION IN STIMULATED RAMAN SCATTERING

C. CLAUDE and J. LEON
Physique Mathématique et Théorique, CNRS-URA 768
Université Montpellier II, 34095 MONTPELLIER FRANCE

Abstract

By using the inverse spectral transform, the SRS equations are solved and the explicit output data is 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 such as to take into account the damping rate of the optical phonon wave. This model is used to interprete the experiments of Drühl, Wenzel and Carlsten (Phys. Rev. Lett., (1983) 51, 1171), in particular the creation of a spike of (anomalous) pump radiation. The related nonlinear Fourier spectrum does not contain discrete eigenvalue, hence this Raman spike is not a soliton.

PACS # 42.65Dr, 42.50 Rh,
Preprint #PM 94-16
To appear in Physical Review Letters

1
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 semi-classical basis, for instance by Shen and Bloembergen, Wang or Carman et al. 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", 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 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 $\text{H}_2$ gas by Wamsley and Raymer and also by Fabricius, Natterman and von der Linde.

At the same time, experiments of Drühl, Wenzel and Carlsten for long duration pump pulses (order of 100ns) revealed that "ocasionally the pump depletion is anomalously reversed for a short time interval generating a spike of pump radiation". They call it a soliton referring to the soliton-solution of the undamped SRS equations given by Chu and Scott. The lesson of these results is the spectacular fit of the experimental data with 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 $\pi$-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 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. Englund and Bowden 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. 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". In particular the non-zero 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 [15]. We obtain the explicit global solution (output laser pulse) which maps perfectly the numerical simulation of SRS (as performed by MacPherson, Carlsten and Druhl [17]).

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

1 - It allows 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 analyse 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 [14].

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 be seen by comparing for instance our figure 2 to the figure 2 of [17]. 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 [14] 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 (\( R_3 = -N \)):

\[
\begin{align*}
\partial_\zeta A_L &= K_{LS} R A_S, \\
\partial_\zeta^* A_S &= -K^*_{LS} R^* A_L \\
\partial_\tau R + RL/(cT_2) &= -K_{LS} N A_L A_S^*.
\end{align*}
\]  
(1)

The spatial variable \( \zeta \) 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 \( \omega_L \)) and of the Stokes emission (frequency \( \omega_S \)) which stimulate the material dynamical variable \( R \) (optical phonon, frequency \( \omega_P = \omega_L - \omega_S \)). \( K_{LS} \) is the complex coupling constant, \( T_2 \) the mean collisional dephasing time and \( N \) is a scaled density (= \( \rho A L \) with \( \rho \) 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:

\[
R(\zeta, 0) = 0, \quad A_L(0, \tau) = A_{L0}(\tau), \quad A_S(0, \tau) = A_{S0}(\tau),
\]  
(2)
where, to reproduce the experiments, $A_{L0}$ is a gaussian, $A_{S0}$ 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 mismatch 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 (2) but with $A_S$ replaced with $A_S \exp[-2ik\zeta]$. 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

\[
\partial_x a_1 = qa_2, \quad \partial_x a_2 - 2ika_2 = -qa_1
\]

\[
\partial_t q + \gamma q = \int dk g(k) a_1 a_2. \tag{3}
\]

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

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

\[
(k = 0) : \quad a_1 = A_L/A_0, \quad a_2 = A_S e^{2ikx}/A_0, \tag{4}
\]

where $A_0 = \text{Max}\{A_L(0, \tau)\}$. 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 (3) reads here (note the sign in (4))

\[
q(x, 0) = 0, \quad a_1(k, +\infty, t) = I_1(k, t), \quad a_2(k, +\infty, t) = I_2(k, t) e^{2ikx} \tag{5}
\]

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 is now 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 (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 [15] that the above initial-boundary value problem is solvable for $\gamma = 0$ and we shall use here directly these results. The solution is given by

\[
a_1(k, -\infty, t) = I_1/\beta + I_2 \bar{\alpha}/\bar{\beta}, \tag{6}
\]

\[
a_2(k, -\infty, t) e^{-2ikx} = I_2/\beta - I_1 \alpha/\beta. \tag{7}
\]

The coefficients $\alpha(k, t)$ and $\beta(k, t)$ (the spectral data) can be computed explicitly from (8) and read for $\gamma = 0$

\[
\alpha(k, t) = -\pi g(k) e^{\phi(k, t)} \int_0^t dt' I_1(k, t') I_2(k, t') e^{-\phi(k, t')} \tag{9}
\]
\[ \phi(k,t) = \int_0^t dt' \left[ \frac{1}{2} \pi g(k) U(k, t') - i \frac{1}{2} \int \frac{d\lambda}{\lambda-k} g(\lambda) U(\lambda, t') \right] \]

\[ U(k, t) = |I_1(k, t)|^2 - |I_2(k, t)|^2 \]

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

\[ \theta(k, t) = -\frac{1}{2\pi} \int \frac{d\lambda}{\lambda-k} \log(1 + |\alpha(\lambda, t)|^2) \]

where the slashed integral denotes the Cauchy principal value. Analogous formulae can be found in [16] 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 report the discussion of this case to forthcoming paper and consider now the case of long pump pulses for which the pump depletion can be anomalously reversed.

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

\[ \alpha \to \infty \Rightarrow |\beta| \to \infty \Rightarrow |a_1(-\infty)| \to |I_2|, \]

\[ \alpha \to 0 \Rightarrow |\beta| \to 1 \Rightarrow |a_1(-\infty)| \to |I_1|. \]

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 (10) the following one

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

The added exponential factor hereabove 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 (15):

\[ a_1 = I_1 f_1 + I_2 e^{2ikx} f_2, \]

\[ a_2 = -I_1 \bar{f}_2 + I_2 e^{2ikx} \bar{f}_1, \]

\[ q = -\frac{1}{\pi} \int dk \ \bar{\alpha} e^{-2ikx} f_1, \]

where \( f_1 \) and \( f_2 \) are the solution of

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

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

(20)

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 (14). 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 \( \alpha \) with \( I_{1,2} \).

In order to realize how the mechanism of pump depletion and spike formation is allowed by the equation (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 \( \pi \)-phase shift:

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

(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 this behavior which is described by the formula (16) where, if \( I_2 \) changes sign, then \( \alpha \) 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) on 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 the fig 2 (with \( \tau_0 = 1 \)). These parameters correspond in the physical world to a pump pulse of maximum amplitude \( A_0 \) of \( 6.36 \times 10^6 \, \text{V} \, \text{m}^{-1} \) and a Stokes seed of \( 0.636 \times 10^6 \, \text{V} \, \text{m}^{-1} \), for the values (taken from ref. [11]) for \( \tau \) in nanosec: \( N = 8.5 \times 10^{19}, \; |K_{LS}| = 5.8 \times 10^{-17} \) and \( T_2 = 0.625 \, \text{ns} \). Consequently, from \( \gamma = L/(cT_2) = 160 \) the data would correspond to a beam path of 30 m.
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 $\pi$-phase shift of the Stokes seed is introduced at 50ns.

We can now easily understand the different behaviors of the pump output on figs. 1 and 2 just by inspection of (10). Indeed, in the first zone (up to 30ns), $\alpha$ is small and we have no depletion. Then the growth of $\alpha$ (pump depletion)
results from the factor exp[\phi] in (10) because \( U \) given by (11) is positive. Now if \( I_2 \) in (11) does not change sign (fig 1), \( \alpha \) grows up to when the damping term dominates again and we observe the pump radiation again (right hand side hump in fig 1 and 2). If instead we introduce a phase flip in the Stokes seed, then the integral in (11) makes \( \alpha \) to decrease and possibly to vanish, which constitutes the mechanism for reversal of pump depletion. However this is allowed only if the growth of \( \alpha \) with exp[\phi] is not too fast, and here enters the role of the damping term \( \exp[-\gamma t] \). Hence the spike of pump radiation occurs as a result of 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 a 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 a 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 multi-
spikes, the result of a stochastic phase in the generated Stokes wave, etc... These studies will be reported elsewhere (see [14]).

References

[1] Y.R. Shen, N. Bloembergen, Phys. Rev. 137, A 1787 (1965)
[2] C.S. Wang, Phys. Rev. 182, 482 (1969)
[3] R.L. Carman, F. Shimizu, C.S. Wang, N. Bloembergen Phys. Rev. A 2, 60 (1970)
[4] M.G. Raymer, J. Mostowski, Phys. Rev. A 24, 1980 (1981)
[5] I.A. Wamsley, M.G. Raymer, Phys. Rev. Lett., 50, 962 (1983)
[6] N. Fabricius, K. Natterman, D. von der Linde, Phys. Rev. Lett., 52, 113 (1983)
[7] K. Drühl, R.G. Wenzel, J.L. Carlsten, Phys. Rev. Lett., 51, 1171 (1983)
[8] F.Y.F. Chu, A.C. Scott, Phys. Rev. A 12, 2060 (1975)

[9] J.C. Englund, C.M. Bowden, Phys. Rev. Lett., 57, 2661 (1986)

[10] D.C. MacPherson, R.C. Swanson, J.L. Carlsten, Phys. Rev. A 40, 6745 (1989)

[11] J.C. Englund, C.M. Bowden, Phys. Rev. A 42, 2870 (1990); Phys. Rev. A 46, 578 (1992)

[12] H. Steudel, Physica 6D, 155 (1983)
   D.J. Kaup, Physica 6D, 143 (1983); 19D, 125 (1986)
   K. Drühl, G. Alsing, Physica 20D, 429 (1986)
   D.J. Kaup, C.R. Menyuk, Phys. Rev. A 42, 1712 (1990)

[13] C.R. Menyuk, Phys. Rev. Lett., 62, 2937 (1989)

[14] C. Claude, F. Ginovart, J. Leon, Nonlinear Theory of Transient Stimulated Raman Scattering and its Application to Long-Pulses Experiments, Preprint PM 94-39 (Montpellier), submitted to Phys. Rev. A

[15] J. Leon, Phys. Rev. A 47, 3264 (1993)
   J. Leon, J. Math. Phys. 35, 1 (1994)

[16] I.R. Gabitov, V.E. Zakharov, A.V. Mikhailov, Teor. Mat. Fiz. 63, 11 (1985) [Theor. Math. Phys. 63, 328 (1985)]; Sov. Phys. JETP, 59, 703 (1984) [Zh. Eksp. Teor. Fiz. 37, 234 (1984)]

[17] D.C. MacPherson, J.L. Carlsten, K.J. Druhl, J. Opt. Soc. Am. B 4, 1853 (1987)