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Raman Solitons and Raman spikes

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Stimulated Raman scattering of a laser pump pulse seeded by a Stokes pulse generically leaves a two-level medium initially at rest in an excited state constituted of *static solitons* and radiation. The soliton birth manifests as sudden very large variations of the phase of the output pump pulse. This is proved by building the IST solution of SRS on the semi-line, which shows moreover that initial Stokes phase flips induce Raman spikes in the pump output also for short pulse experiments.

Introduction. Stimulated Raman scattering (SRS) of high energy Laser pulse in two-level medium has been intensively studied these last years (see e.g. the review [1]), more especially after the experiments by Drühl, Wenzel and Carlsten [2] showing that spikes of pump radiation (Raman spikes) occur spontaneously in the pump depletion zone, see also [3]. Consequently nonlinear Raman amplification was used as a means to observe at the macroscopic level the fluctuation of the phase of the initial Stokes vacuum [4].

The SRS equations possess a Lax pair [5] and, as a consequence, theoretical and experimental works were partly devoted to the search of the *Raman soliton* predicted in [5]. In particular it has been believed [6] that the *spike of pump radiation* observed in [2] is a soliton which, in the inverse spectral transform scheme (IST), would be related to the discrete part of the spectrum. However it has been proved that the observed Raman spike is not a soliton but merely a manifestation of the continuous spectrum [7].

Then the question of the creation a genuine Raman soliton is still an open problem (for 20 years now). We show here that the SRS equations, solved as a boundary value problem on the semi-line, do induce the generation of solitons by pair, and that, after the passage of the pulses, the solitons are static in the medium. Although the solitons are not seen in the output pump *intensity* (where only Raman spikes are seen), the signature of the soliton birth consists of very large fluctuations of the *phase* of the pump output, which remains to be experimentally observed.

Last we prove that the Raman spikes, observed in long pulse experiments (pulses with fwhm of 75 ns in [2] and 15 ns in [3]), where *damping* is essential in the model, must occur also for short pulses (no damping), and give the condition for their creation.

The efficiency of the method of solution serves not only as a basis for seeking the Raman soliton, but also to evaluate *exactly* the result of any Raman amplification. Indeed the SRS equations are solved for *arbitrary* values of the input ($x = 0$) pump and Stokes pulses on a medium with any initial ($t = 0$) state. The original problem is reduced to a Riccati equation (11) and linear in-

tegral equations (12). In many interesting cases the Riccati equation (11) can be explicitly solved (see (15)) and important physical information can be obtained without solving equations (12), such as the output (14).

The SRS equations. For a medium of length L , a detuning $\Delta\omega$ and a phase mismatch Δk , in the dimensionless variables $x = Z/L$, $t = (cT - \eta Z)/L$, where Z and T are the physical variables, the SRS equations for short pulses (infinite dephasing time) in the *slowly varying envelope approximation* read [9]

$$\begin{aligned}\partial_x a_L &= q a_S e^{-i\Delta\omega t} e^{2ikx}, \\ \partial_x a_S &= -\bar{q} a_L e^{i\Delta\omega t} e^{-2ikx}, \\ \partial_t q &= -g \int dk a_L a_S^* e^{i\Delta\omega t} e^{-2ikx}.\end{aligned}\quad (1)$$

The phase mismatch is represented there by the dimensionless parameter $k = \frac{1}{2}L(\Delta k - \frac{\eta}{c}\Delta\omega)$. The envelopes a_L (frequency ω_L) and a_S (frequency ω_S) are scaled to the peak intensity I_m of the input laser field and the dimensionless coupling constant is given by

$$g = \frac{L^2 N \alpha'_0 \epsilon_0 \omega_S}{16 \eta m c^2 \omega_V} I_m^2, \quad (2)$$

where α'_0 is the differential polarizability at equilibrium, c/η is the light velocity in the medium of transition frequency ω_V , and N is the density of oscillators of mass m . We have considered in (1) the cooperative interaction of all k -components with the medium and then the input ($x = 0$) values of the light waves are also function of k sharply distributed around $k = 0$

$$a_L(k, 0, t) = J_L(k, t), \quad a_S(k, 0, t) = J_S(k, t). \quad (3)$$

Finally the electromagnetic field is obtained as

$$E = \frac{I_m}{2} a_L e^{i(k_L Z - \omega_L T)} + \frac{I_m}{2} \sqrt{\frac{\omega_S}{\omega_L}} a_S e^{i(k_S Z - \omega_S T)} + c.c.$$

The system (1) can be solved on the infinite line for arbitrary asymptotic boundary values [8] and it has been proved in [7] that this allows the interpretation of the experiments of [2]. We will demonstrate that it can be solved completely on the semi-line $x \in [0, \infty)$, which actually furnishes the solution at any point $x = L$ and hence solves the finite interval case as a *free end problem*.

Lax pair. The time evolution in (1) results as the compatibility condition $U_t - V_x + [U, V] = 0$ for the Lax pair ($x \geq 0, t \geq 0$)

$$\varphi_x = U\varphi + ik\varphi\sigma_3, \quad (4)$$

$$\varphi_t = V\varphi + \varphi e^{-ik\sigma_3 x} \Omega e^{ik\sigma_3 x}, \quad (5)$$

where the *dispersion relation* Ω is x -independent and

$$U = \begin{pmatrix} -ik & q \\ -\bar{q} & ik \end{pmatrix},$$

$$V = \frac{g}{4i} \int \frac{d\lambda}{\lambda - k} \begin{pmatrix} |a_L|^2 - |a_S|^2 & 2a_L \bar{a}_S e^{-2i\lambda x} \\ 2\bar{a}_L a_S e^{2i\lambda x} & |a_S|^2 - |a_L|^2 \end{pmatrix}. \quad (6)$$

$V(k)$ is discontinuous, therefore only the limits $V^\pm = V(k \pm i0)$ are defined on the real k -axis. Such is also the case for Ω , and Ω^\pm will have to be determined from the boundary conditions on the solution φ .

Scattering problem. Following standard methods in spectral theory, see e.g. [10], we define the solutions φ^+ and φ^- according to

$$\begin{pmatrix} \varphi_{11}^+ \\ \varphi_{21}^+ \end{pmatrix} = \begin{pmatrix} 1 \\ 0 \end{pmatrix} - \begin{pmatrix} -\int_0^x d\xi q \varphi_{21}^+ \\ \int_0^x d\xi \bar{q} \varphi_{11}^+ e^{2ik(x-\xi)} \end{pmatrix}$$

$$\begin{pmatrix} \varphi_{12}^+ \\ \varphi_{22}^+ \end{pmatrix} = \begin{pmatrix} 0 \\ 1 \end{pmatrix} - \begin{pmatrix} \int_x^\infty d\xi q \varphi_{22}^+ e^{-2ik(x-\xi)} \\ \int_0^x d\xi \bar{q} \varphi_{12}^+ \end{pmatrix}$$

$$\begin{pmatrix} \varphi_{11}^- \\ \varphi_{21}^- \end{pmatrix} = \begin{pmatrix} 1 \\ 0 \end{pmatrix} + \begin{pmatrix} \int_0^x d\xi q \varphi_{21}^- \\ \int_x^\infty d\xi \bar{q} \varphi_{11}^- e^{2ik(x-\xi)} \end{pmatrix}$$

$$\begin{pmatrix} \varphi_{12}^- \\ \varphi_{22}^- \end{pmatrix} = \begin{pmatrix} 0 \\ 1 \end{pmatrix} + \begin{pmatrix} \int_0^x d\xi q \varphi_{22}^- e^{-2ik(x-\xi)} \\ -\int_x^\infty d\xi \bar{q} \varphi_{12}^- \end{pmatrix}. \quad (7)$$

These solutions obey the *reduction* $\bar{\varphi}_1^+(\bar{k}) = i\sigma_2 \varphi_2^-(k)$, the Riemann-Hilbert relations

$$\begin{aligned} \varphi_1^- &= \varphi_1^+ - e^{2ikx} \rho^* \varphi_2^-, \\ \varphi_2^+ &= \varphi_2^- + e^{-2ikx} \rho \varphi_1^+, \end{aligned} \quad (8)$$

and the bounds

$$x = 0 : \varphi^+ = \begin{pmatrix} 1 & \rho \\ 0 & 1 \end{pmatrix}, \quad \varphi^- = \begin{pmatrix} 1 & 0 \\ -\rho^* & 1 \end{pmatrix},$$

$$x \rightarrow \infty : \varphi^+ \rightarrow \begin{pmatrix} 1/\tau & 0 \\ e^{2ikx} \rho^*/\tau^* & \tau \end{pmatrix},$$

$$\varphi^- \rightarrow \begin{pmatrix} \tau^* & -e^{-2ikx} \rho/\tau \\ 0 & 1/\tau^* \end{pmatrix}. \quad (9)$$

which define the reflection coefficient ρ and the transmission coefficient τ ($\rho^*(k)$ stands for $\bar{\rho}(\bar{k})$). Note that $\det\{\varphi(x)\} = \det\{\varphi(\infty)\} = 1$, hence for real k $|\tau|^2 = 1 + |\rho|^2$.

The column vectors φ_1^+ and φ_2^- are entire functions in the k -plane, with good behaviors as $k \rightarrow \infty$ in the upper half-plane for φ_1^+ (and the lower one for φ_2^-). The vector φ_2^+ is meromorphic in the upper half-plane with a finite number N of simple poles k_n (and φ_1^- in the lower one with poles \bar{k}_n). Consequently τ and ρ have meromorphic extensions in the upper half-plane where they possess the N simple poles k_n (the bound states locations).

Solution of SRS. From the boundary data (3) we have readily (set $\Delta\omega = 0$ for simplicity because it can be scaled off in a_S)

$$\begin{pmatrix} a_L \\ a_S e^{2ikx} \end{pmatrix} = J_L \varphi_1^+ + J_S \varphi_2^- e^{2ikx}. \quad (10)$$

After careful computations of the boundary values of (5), using separately φ^+ and φ^- , we obtain both limits Ω^+ and Ω^- of Ω and the following time evolution of the scattering coefficient $\rho(k, t)$:

$$\rho_t = -\rho^2 C^+[m^*] - 2\rho C^+[\phi] - C^+[m]. \quad (11)$$

$$C^+[f] = \frac{1}{\pi} \int \frac{d\lambda}{\lambda - (k + i0)} f(\lambda),$$

$$m = \frac{i\pi}{2} g J_L J_S^*, \quad \phi = \frac{i\pi}{4} g (|J_L|^2 - |J_S|^2).$$

The reconstruction proceeds through the solution of the Riemann-Hilbert problem (8) which reads

$$\varphi_1^+(k) = \begin{pmatrix} 1 \\ 0 \end{pmatrix} + \frac{1}{2i\pi} \int_{C_-} \frac{d\lambda}{\lambda - k} \rho^*(\lambda) \varphi_2^-(\lambda) e^{2i\lambda x},$$

$$\varphi_2^-(k) = \begin{pmatrix} 0 \\ 1 \end{pmatrix} + \frac{1}{2i\pi} \int_{C_+} \frac{d\lambda}{\lambda - k} \rho(\lambda) \varphi_1^+(\lambda) e^{-2i\lambda x}, \quad (12)$$

where C_+ (resp. C_-) is a contour from $-\infty + i0$ to $+\infty + i0$ (resp. from $-\infty - i0$ to $+\infty - i0$) passing over all poles k_n of $\rho(k)$ (resp. under \bar{k}_n). Remember the notation $\rho^*(\lambda) = \bar{\rho}(\bar{\lambda})$.

Since m and ϕ are continuous bounded functions of k , the coefficients of the Riccati equation (11) are analytic functions in the upper half k -plane, where precisely ρ is meromorphic at time $t = 0$. Consequently ρ remains meromorphic at later times, which allows to prove that the reconstructed fields do obey the system of equations (1), or in short that the reconstructed φ does obey (9). Indeed from (12), the Cauchy theorem leads to

$$\forall x < 0 : \varphi_1^+ = \begin{pmatrix} 1 \\ 0 \end{pmatrix}, \quad \varphi_2^- = \begin{pmatrix} 0 \\ 1 \end{pmatrix}. \quad (13)$$

The formulae (11) and (12) with (10) furnish the solution of (1), in particular the output field values at $x = L$ (whatever be q for $x > L$). In the limit $L \rightarrow \infty$, and by (9), the solution becomes *explicit*, and for instance the output pump reads

$$x \rightarrow \infty : a_L \rightarrow \frac{1}{\tau} J_L - \frac{\rho}{\tau} J_S. \quad (14)$$

Evolution of laser pulses. We are interested here in the physically relevant case when the medium is initially in the ground state, that is $\rho(k, 0) = 0$. Next we choose a Stokes wave as a portion of the pump wave, both with Lorentzian lineshape, namely, for $\kappa > 0$,

$$|J_L|^2 = |A(t)|^2 \frac{\kappa}{\pi(k^2 + \kappa^2)}, \quad J_S = e^{-\gamma - i\theta(t)} J_L$$

where $A(t)$ is the pulse shape of duration t_m ($t > t_m \Rightarrow A(t) = 0$). The parameter γ (real positive constant) measures the ratio pump/Stokes inputs. Considering the case when the input Stokes wave experiences one phase flip at some time t_0 , we choose the phase $\theta(t) = 0$ for $t < t_0$ and $\theta(t) = \pi$ for $t > t_0$.

It follows from contour integration that the Riccati equation (11) can be rewritten as

$$\rho_t = -\frac{i}{2} \frac{ge^{-\gamma}}{k + i\kappa} |A|^2 [\rho^2 e^{-i\theta} - 2\rho \sinh \gamma - e^{i\theta}].$$

Taking into account the phase flip, the general solution with zero initial datum reads

$$\begin{aligned} t \leq t_0 : \rho &= \frac{\sinh \delta}{\cosh(\delta - \gamma)}, \\ t \geq t_0 : \rho &= \frac{\rho_0 \cosh(\delta - \delta_0 + \gamma) - \sinh(\delta - \delta_0)}{\cosh(\delta - \delta_0 - \gamma) - \rho_0 \sinh(\delta - \delta_0)}, \end{aligned} \quad (15)$$

with $\rho_0 = \rho(k, t_0)$, $\delta_0 = \delta(k, t_0)$ and the definitions

$$\delta(k, t) = \frac{iT(t)}{k + i\kappa}, \quad T(t) = \frac{1}{4} g(1 + e^{-2\gamma}) \int_0^t dt' |A|^2. \quad (16)$$

Soliton generation and signature. Let us consider first the case with no phase flip, that is $t_0 > t_m$. The spectral transform ρ , first function in (15), has an infinite set of moving poles $k_n(t)$, $n \in \mathbb{Z}$, given by

$$k_n = -i\kappa + \frac{T(t)}{(n + \frac{1}{2})\pi - i\gamma}, \quad (17)$$

which are associated with solitons as soon as they lie in the upper half plane (in the lower half plane they are the resonances).

As t evolves, and for a given linewidth κ , these poles move from the point $-i\kappa$ and they may cross the real axis (and generate solitons) if $T(t)$ is large enough, which means enough energy in the input pulses. Moreover, since $k_n = -\bar{k}_{-n-1}$, solitons are created by pair. After the passage of the pulse, ($t > t_m$), m and ϕ vanish in (11), $T(t)$ is constant, and hence the whole solution becomes t -independent. Consequently the laser pulses leave in the medium a *finite number of static bi-solitons*.

The question of their observation is not straightforward. Indeed, taking for simplicity the case of an infinite medium, the pump output is given in (14) where the coefficients ρ and τ are understood for real values

of k (the essential mismatch). The main point here is that both $1/\tau$ and ρ/τ are holomorphic functions in the upper half-plane of k , and hence an integrated intensity like $\int dk |a_L|^2$ will not show the presence of the poles k_n . Moreover, if at $t = t_s$ a resonance crosses the real axis at k_s to become a bound state (soliton birth), the output *intensity* at that particular value k_s will simply show a full depletion ($1/\tau(k_s) = 0$ and $|\rho(k_s)/\tau(k_s)| = 1$).

However, the *phase* of the transmission coefficient $\tau(k)$ diverges for all real k precisely at $t = t_s$. Indeed the *trace formula* (with no pole on the real axis i.e. just before pole crossing) reads for $k \in \mathbb{R}$ [11]

$$\frac{\tau(k)}{|\tau(k)|} = \prod \frac{k - \bar{k}_n}{k - k_n} \exp\left[-\frac{i}{2\pi} \int \frac{\ln(1 + |\rho(\lambda)|^2)}{\lambda - k}\right],$$

where the integral over λ is understood as the Cauchy principal value, and where the product concerns only the bound states if any. It is then clear that, for every $k \neq k_s$, the phase of $\tau(k)$ diverges as $t \rightarrow t_s$ because $\rho(\lambda)$ has a pole in $\lambda = k_s$. The observation of true Raman solitons should go then with a measure of the phase of the pump output.

We remark here that, in the *sharp line limit* $\kappa \rightarrow 0$ and with absence of a flip in the boundary condition, the infinite set of poles k_n lie in the upper half-plane and $|k_n|$ is proportional to $T(t)$ (17), which implies that the solution becomes self-similar (the self-similar solution of SRS is analyzed in [12]).

Stokes phase flip and Raman spike. As shown in [7], the Raman spike observed in [2] and largely studied later, occurs at time t_r for which $\rho(k, t_r) = 0$. Indeed, then $|\tau(k, t_r)| = 1$ and from (14) the pump (intensity) is fully depleted.

The second expression in (15) gives that ρ vanishes at t_r for

$$\coth(\delta_0) - \coth(\delta_r - \delta_0) = 2 \tanh \gamma, \quad (18)$$

which relates t_0 (instant of phase flip) and t_r (instant of Raman spike) to the pulse area through (16).

To get a simple insight in that formula, we may consider the dominant term $k = 0$ (resonance) for which δ_0 and $\delta_r - \delta_0$ are real positive. Then a Raman spike will be allowed if (at $k = 0$)

$$\coth(\delta_m - \delta_0) > \coth(\delta_0) - 2 \tanh \gamma > 0,$$

where $\delta_m = \delta(k, t_m)$, and t_m is the pulse duration. This is obtained without damping, which suggests the search of Raman spikes in short pulse experiments (short with respect to the medium dephasing time).

We note finally that the occurrence of a phase flip alters the denominator of ρ , which can be used to change the discrete spectrum and hence to modify the soliton part of the solution.

Conclusion. The following results have been obtained:

1 - The proof that genuine Raman solitons do occur in SRS experiments and cannot be detected in the energy of the output but rather in its phase divergence.

2 - The *spontaneous* generation of solitons out of vacuum in an integrable system.

3 - The complete solution by IST of the boundary value problem for SRS on the semi-line. This solution relies on the analytical properties of the nonlinear Fourier transform $\rho(k, t)$ which are *conserved* in the time evolution.

4 - The proof that Raman spikes are *generic* as they occur as well for short duration pulses, as suspected in the experimental work [3].

5 - The proof that in the sharp line limit and with the proportional pump/Stokes input on a medium initially at rest, the solution is self-similar.

Comments. 1 - The IST solution of SRS on the semi-line has been considered in [13] for the sharp line case. Following the same procedure in our *inhomogeneously broadened* case, and in particular neglecting some terms in the spectral data evolution, would lead to an evolution for ρ with non-analytic coefficients, in contradiction with the necessary meromorphy of ρ .

2 - IST on the semi-line for the nonlinear Schrödinger equation has been considered in [14], and the problem is considerably more difficult mainly because of the necessity of the knowledge of additional constraints in $x = 0$ (e.g. the time derivative of the field).

3 - Boundary conditions for nonlinear evolution equations are known to be a source for soliton generation [15] [14], but this is the first instance of an integrable system which shows *explicitely* the spontaneous generation of solitons, as motion of poles from the lower to the upper half-plane, from resonances to bound states.

4 - At the moment we are not able to extract, from the explicit form of the spectral data, the explicit form of the *potential* (the medium excitation field) because on the semi-line no pure soliton exist (would ρ vanish on the real axis that it would vanish everywhere and no bound states would be allowed). However the method provides the explicit form of the physically relevant quantities which are the light pulses output values.

5 - The manifestation of the soliton birth as sudden large variation of the phase of the output was quite unsuspected and should lead to reconsider the measures of the output in SRS experiments.

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