Transient Solitons in Stimulated Raman Scattering

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It is shown that permanent, solitonlike structures in which the pump retains a large fraction of its initial energy cannot exist for finite-energy pulses. Ultimately, solitonlike structures propagate to the back end of the pulse and disappear. A method for propagating pump energy over long distances is proposed.

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Since the early work of Wang,[1] Carman *et al.*,[2] and Mack *et al.*,[3] the time-dependent effects in stimulated Raman scattering have been the continued focus of attention. These effects are thought to play a role in a wide variety of media, including plasmas,[4] liquids,[5] glasses,[6] and gases.[7,8] These effects are also of interest to mathematical physicists who study solitons.[9-13] Recently, solitons, or at least solitonlike pulses, have been observed experimentally by Drühl, Wenzel, and Carlsten[14] and Wenzel, Carlsten, and Drühl.[15] Pulses of long duration compared to T_2, the natural damping time of the material excitation, are injected into a Raman-active medium. In most cases, the pump depletes completely, giving up its energy to the Stokes wave. However, when a rapid phase flip is present in the Stokes wave, a small amount of energy can remain in the pump, leading to a soliton of duration less than T_2. Of course, only a small fraction of the pulse's initial energy remains in the pump.

In recent experiments of Reintjes and co-workers at the Naval Research Laboratory, stimulated Raman scattering has been studied with pulses short compared to T_2.[7,8] It is thus natural to ask whether it is possible to create a soliton in which both the pump and Stokes pulses are short compared to T_2 and the pump retains a large fraction of its initial energy. The chief purpose of this Letter is to show that it cannot be done. The pump will always deplete after some distance—even in the limit $T_2=\infty$. In this Letter, we also interpret this result physically and obtain the soliton lifetime.

Assuming that diffraction can be ignored, ground-state depletion of the material excitation can be ignored, and the slowly varying envelope approximation holds, the basic equations become after normalization,[1-15]

$$\frac{\partial A_1}{\partial \chi} = -XA_2, \tag{1a}$$

$$\frac{\partial A_2}{\partial \chi} = X^* A_1, \tag{1b}$$

$$\frac{\partial X}{\partial \tau} + \gamma X = A_2^* A_1, \tag{1c}$$

where A_1 and A_2 are the complex amplitudes of the pump and Stokes waves, X is the complex amplitude of the material excitation, and χ and τ represent distance along the Raman cell and time. Finally, $\gamma \propto T_2^{-1}$ represents the damping rate. In physical systems, $X=0$ at any χ before the arrival of radiation; hence, $X \to 0$ as $\tau \to -\infty$ at all χ and Eq. (1c) integrates to yield

$$X = \int_{-\infty}^{\tau} A_2^* A_1 \exp[-\gamma(\tau - \tau')]\, d\tau'. \tag{2}$$

We shall always assume that

$$\int_{-\infty}^{\infty} (|A_1|^2 + |A_2|^2)\, d\tau' = I_\infty < \infty \tag{3}$$

so that Eq. (2) is well defined even when $\gamma=0$.

From Eq. (1), it follows that

$$|A_1(\chi,\tau)|^2 + |A_2(\chi,\tau)|^2 = K(\tau), \tag{4}$$

which is independent of χ, so that if Eq. (3) holds at $\chi=0$, it holds for all χ. We shall assume that $A_1(\chi,\tau)$ and $A_2(\chi,\tau)$ are known at $\chi=0$ and then evolve according to Eq. (1) at later χ, corresponding to the experimental fact that known pulses are injected into the Raman-active medium at one end. We find now from Eqs. (2)-(4) and the Cauchy-Schwartz inequality that $X(\chi,\tau) \le I_\infty/2$ for all χ and τ.

The invariance of $K(\tau)$ is a strong constraint. If permanent solitons exist, then they must have zero velocity, as otherwise they will travel to the back end or front end of the pulse and disappear. A similar argument holds for breathers. Hence, to exclude the possibility of permanent solitons or breathers, it suffices to show that no stable stationary or quasiperiodic solutions exist for which the pump is nonzero. We shall proceed by showing that $X \to 0$ uniformly as $\chi \to \infty$ at all τ. From the continuity of A_1 and A_2 as functions of χ, it then follows that the only allowed stationary or quasiperiodic solution is one in which $A_1 A_2^* = 0$ at all τ. This solution is unstable unless $A_1 = 0$ at all τ.

To proceed, we first note from Eq. (1) that

$$\frac{\partial R}{\partial \chi} \le -|X|^2, \tag{5}$$

where $R = \int_{-\infty}^{\tau} |A_1|^2 d\tau'$. Equality is obtained in Eq. (5) when $\gamma=0$. Noting as well that $R \ge 0$ for all χ, while $-|X|^2 \le 0$, it follows from mathematical analysis[16] that $\int_\chi^\infty |X|^2 d\chi' \to 0$ as $\chi \to \infty$. In the opposite case, R would have to become negative. From Eq. (1), we obtain

$$\left|\frac{d|X|^2}{d\chi}\right| = \left|X^* \int_{-\infty}^{\tau} X[|A_1|^2 - |A_2|^2]\exp[-\gamma(\tau-\tau')]d\tau' + \text{c.c.}\right| < I_\infty^3 . \tag{6}$$

We now fix τ and suppose that $|X| = |X|_m$ at $\chi = \chi_m$. It follows that

$$\int_{\chi_m}^{\infty} |X|^2 d\chi' > |X|_m^4 / I_\infty^3 . \tag{7}$$

Equation (7) is only consistent with our previous result if $X \to 0$ as $\chi \to \infty$ for all τ. The precise theorem that we wish to obtain is as follows: *Given any $\epsilon > 0$, there exists a χ_1 (independent of τ, but depending on ϵ) such that if $\chi > \chi_1$, then $|X(\chi,\tau)| < \epsilon$.* The argument we have just given is not quite sufficient to show that χ_1 is independent of τ and that therefore $X \to 0$ uniformly. To remedy this defect, we consider a compact region $[-T,T]$ of τ, chosen so $\int_T^\infty K(\tau)d\tau < \epsilon$ and $\int_{-\infty}^{-T} K(\tau)d\tau < \epsilon$. It follows from Eq. (2) and the Cauchy-Schwartz inequality that $|\Delta X|$, the contribution to $|X|$ from either the region $(-\infty,-T)$ or the region (T,∞), is less than or equal to $\epsilon/2$. Inside the compact region, we now choose χ_1 to equal the maximum χ value beyond which $|X|$ is bounded below $\epsilon/2$. Beyond this χ value, $|X|$ is bounded below ϵ for all τ.

Since $|X| \to 0$ uniformly, it follows that the only possible stationary or quasiperiodic solution is one where $X=0$. It follows from Eq. (2) that $A_2^* A_1 = 0$. It is worth stressing that this result does not show that $A_1 A_2^* \to 0$ as $\chi \to \infty$ in the general, nonstationary case as it is possible for the derivative of $A_1 A_2^*$ to become singular. In fact, numerical results[17,18] indicate that both occur: $A_1 A_2^* \to 0$ while its derivative becomes singular.

We turn now to considering the stability of the stationary solution $A_1 A_2^* = 0$. We first consider some region $[\tau_1, \tau_2]$ in τ where $A_2 = 0$ while $A_1 \neq 0$, and we perturb our stationary solution slightly in this region. We determine the perturbed quantities $\delta A_1(\chi,\tau)$, $\delta A_2(\chi,\tau)$, and $\delta X(\chi,\tau)$, given $\delta A_1(0,\tau)$ and $\delta A_2(0,\tau)$, by linearizing Eq. (1) and solving the linearized equations. We obtain[2]

$$\begin{aligned}
\delta A_1(\chi,\tau) &= \delta A_1(0,\tau) , \\
\delta A_2(\chi,\tau) &= \delta A_2(0,\tau) \\
&\quad + \chi^{1/2} A_1(\tau) \int_{-\infty}^{\tau} A_1^*(\tau')\delta A_2(0,\tau')\exp[-\gamma(\tau-\tau')][I(\tau)-I(\tau')]^{-1/2} I_1(2\{\chi[I(\tau)-I(\tau')]\}^{1/2}) d\tau' , \\
\delta X(\chi,\tau) &= \int_{-\infty}^{\tau} A_1(\tau')\delta A_2^*(0,\tau')\exp[-\gamma(\tau-\tau')] I_0(2\{\chi[I(\tau)-I(\tau')]\}^{1/2}) d\tau' ,
\end{aligned} \tag{8}$$

where

$$I(\tau) = \int_{-\infty}^{\tau} K(\tau')d\tau' . \tag{9}$$

The quantities $I_0(x)$ and $I_1(x)$ are modified Bessel functions. The perturbation is unstable as δA_2 and δX both grow exponentially. We now consider a stationary solution where $A_1 = 0$ at all τ. In this case we find[18]

$$\begin{aligned}
\delta A_1(\chi,\tau) &= \delta A_1(0,\tau) - \chi^{1/2} A_2(\tau) \int_{-\infty}^{\tau} A_2^*(\tau')\delta A_1(0,\tau')\exp[-\gamma(\tau-\tau')][I(\tau)-I(\tau')]^{-1/2} J_1(2\{\chi[I(\tau)-I(\tau')]\}^{1/2}) d\tau' , \\
\delta A_2(\chi,\tau) &= \delta A_2(0,\tau) , \\
\delta X(\chi,\tau) &= \int_{-\infty}^{\tau} A_2^*(\tau')\delta A_1(0,\tau')\exp[-\gamma(\tau-\tau')] J_0(2\{\chi[I(\tau)-I(\tau')]\}^{1/2}) d\tau' ,
\end{aligned} \tag{10}$$

where $J_0(x)$ and $J_1(x)$ are standard Bessel functions. To verify the stability of this result, it is useful to write the equation for δA_1 in Stieltjes form,

$$\delta A_1(\chi,\tau) = \delta A_1(0,\tau) - A_2(\tau) \int_{-\infty}^{\tau} [\delta A_1(0,\tau')/A_2(\tau')]\exp[-\gamma(\tau-\tau')] dJ_0(2\{\chi[I(\tau)-I(\tau')]\}^{1/2}) . \tag{11}$$

If we assume that $\delta A_1/A_2$ is bounded, which physically amounts to saying that the perturbation is always small, then $\delta A_1(\chi,\tau) \to 0$ at all τ. In the worst case, $|\delta A_1|$ can grow no faster than linearly with χ. We conclude that the perturbation is stable.

Combining all our previous results, we conclude that the only stable stationary solution is one for which $A_1 = 0$. Hence, there are no permanent solitons.

From a mathematical standpoint, it may be somewhat surprising at first that there are no permanent solitons when $\gamma=0$. Equation (1) has a Lax pair in this limit;[9-12] indeed, when A_1 and A_2 are both real, Eq. (1) reduces to the sine-Gordon equation. The crucial role played by the boundary conditions we have chosen cannot be overemphasized. As previously shown by Chu and Scott,[9] if one considers the time evolution of A_1, A_2, and X from an initial condition where $X(\chi)\rightarrow 0$ rapidly at $\chi=\pm\infty$, and if one demands that $A_1A_2^*\rightarrow 0$ rapidly at $\chi=\pm\infty$ for all τ, then permanent solitons exist. We have considered the spatial evolution where $|A_1|^2+|A_2|^2\rightarrow 0$, and we have demanded that $X\rightarrow 0$ rapidly at $\tau=-\infty$. Our boundary conditions are appropriate to model the experiments which have been carried out to date.

The standard spectral transform method, described, for example, in Ref. 19, cannot be directly applied to the solution of Eq. (1) with our boundary conditions. The difficulty is that the evolution of the spectral data depends on X in the limit $\tau\rightarrow\infty$. The behavior of X in this limit is not known *a priori* if $\gamma=0$. Kaup[10] has found a clever solution to this problem when I_∞ in finite. One in essence neglects the contribution to the spectral data evolution which depends on X at $\tau=+\infty$. The resulting pseudospectral data yield the correct answers for A_1, A_2, and X as functions of χ when substituted into the Marchenko equation. The poles of the kernel do not correspond to permanent solitons in Kaup's approach.

From a physical standpoint, the result we have obtained is eminently reasonable. One can gain considerable insight into the problem by considering the transient "soliton" solution to Eqs. (1) and (2) in the limit $\gamma=0$,

$$\begin{aligned}
A_1&=[K(\tau)]^{1/2}\operatorname{sech}\left[\alpha\chi-\frac{1}{\alpha}\int_0^\tau K(\tau')d\tau'\right],\\
A_2&=[K(\tau)]^{1/2}\tanh\left[\alpha\chi-\frac{1}{\alpha}\int_0^\tau K(\tau')d\tau'\right],\\
X&=\alpha\operatorname{sech}\left[\alpha\chi-\frac{1}{\alpha}\int_0^\tau K(\tau')d\tau'\right].
\end{aligned}\tag{12}$$

When $K(\tau)$ is set equal to a constant K_0, this solution corresponds to the single-soliton solution found by Chu and Scott.[9] When $K(\tau)$ is chosen such that $\int_{-\infty}^{\infty}K(\tau)\times d\tau=I_\infty<\infty$ and thus Eq. (12) satisfies our boundary conditions as well as those of Chu and Scott, then the solution of Eq. (12) closely resembles the single-soliton solution of Chu and Scott over some length if $\alpha\ll I_\infty$. Hence, it seems appropriate to refer to the solution of Eq. (12) as a transient soliton, although it is not a soliton in the standard sense. There is another viewpoint which sheds further light upon these transient solitons. Making the transformations $T=\int_0^\tau K(\tau')d\tau'$, $A_1'=A_1/[K(\tau)]^{1/2}$, and $A_2'=A_2/[K(\tau)]^{1/2}$, and replacing τ, A_1, and A_2 with these new variables, one finds that Eq. (1) is invariant. The solution of Eq. (12) can be viewed as a standard soliton in these new variables. However, the physical values of T are located in the range $(T_{\min},T_{\max})$, where $T_{\min}=-\int_{-\infty}^{0}K(\tau)d\tau$ and $T_{\max}=\int_0^\infty K(\tau)d\tau$, so that at large χ the soliton propagates outside the physical time domain. Thus, the soliton's presence in the physical time domain is transient.[20]

The reason that it is not possible to create a permanent solitonlike structure in which the pump does not deplete is now clear. In any such structure the pump energy propagates subluminously to the back of the pulse and disappears. If $\alpha\ll I_\infty$, these pulses can persist for a great length, $\chi\sim I_\infty/\alpha^2$. In the long-pulse experiments of Drühl and co-workers[14,15] transient solitons can easily persist the length of the experiment, while that is not true in the short-pulse experiments of Reintjes and co-workers.[7,8]

A certain complementarity is evident in these results. The ratio of the width of the pump pulse to the width of the Stokes pulse in these transient solitons is given by α/I_∞. Hence, transient solitons which persist for long lengths can only contain a small fraction of the pulse energy and vice versa. A potential solution to this dilemma is to stack a series of solitons as close to each other as possible. In effect, one finds that Eq. (1) has the cnoidal wave solution

$$\begin{aligned}
A_1&=[K(\tau)]^{1/2}\operatorname{cn}\left[\alpha\chi-\frac{1}{m\alpha}\int_0^\tau K(\tau')dt'\,|\,m\right],\\
A_2&=[K(\tau)]^{1/2}\operatorname{sn}\left[\alpha\chi-\frac{1}{m\alpha}\int_0^\tau K(\tau')dt'\,|\,m\right],\\
X&=\alpha\operatorname{dn}\left[\alpha\chi-\frac{1}{m\alpha}\int_0^\tau K(\tau')d\tau'\,|\,m\right],
\end{aligned}\tag{13}$$

assuming $\gamma=0$. When $\gamma\neq0$, a solution with roughly this form will still exist whenever $m\alpha\gamma<K(\tau)$. Using long pulses so that $I_\infty\gg\alpha$, it should be possible to retain a large fraction of the initial pulse energy in the pump while propagating long distances. This basic idea has already been discussed by Duncan *et al.*[8] Their experiments indicate that it may be possible to achieve a quasi-steady-state like that of Eq. (13). Of course, Eq. (1) does not contain physical effects such as higher-order Stokes and anti-Stokes generation, which in practice are almost certain to limit the longevity of pulses like those of Eq. (13).

In this Letter, we have shown that pump pulses will always deplete due to the effect of stimulated Raman scattering. We have shown that any solitonlike structures are subluminous and will ultimately disappear at the back end of the pulse. We propose the use of cnoidal wave solutions in order to retain a large fraction of the initial pulse energy in the pump while propagating long distances.

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[16]See, e.g., T. M. Apostol, *Mathematical Analysis* (Addison-Wesley, Reading, MA, 1974), p. 185. Refer to Theorem 8.6. To apply the theorem, we let $R_n = R(\xi = n\Delta)$, where Δ is any real number greater than 0 and $n = 0,1,2,\ldots$. We note that $R_{n+1} \leq R_n$, but that $R_n \geq 0$ for all n. Hence, the theorem applies.

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[19]See, e.g., M. J. Ablowitz and H. Segur, *Solitons and the Inverse Scattering Transform* (SIAM, Philadelphia, 1981), Chap. 1 for a discussion of spectral transform methods.

[20]This picture of solitons propagating outside the physical time domain was provided by David Kaup.