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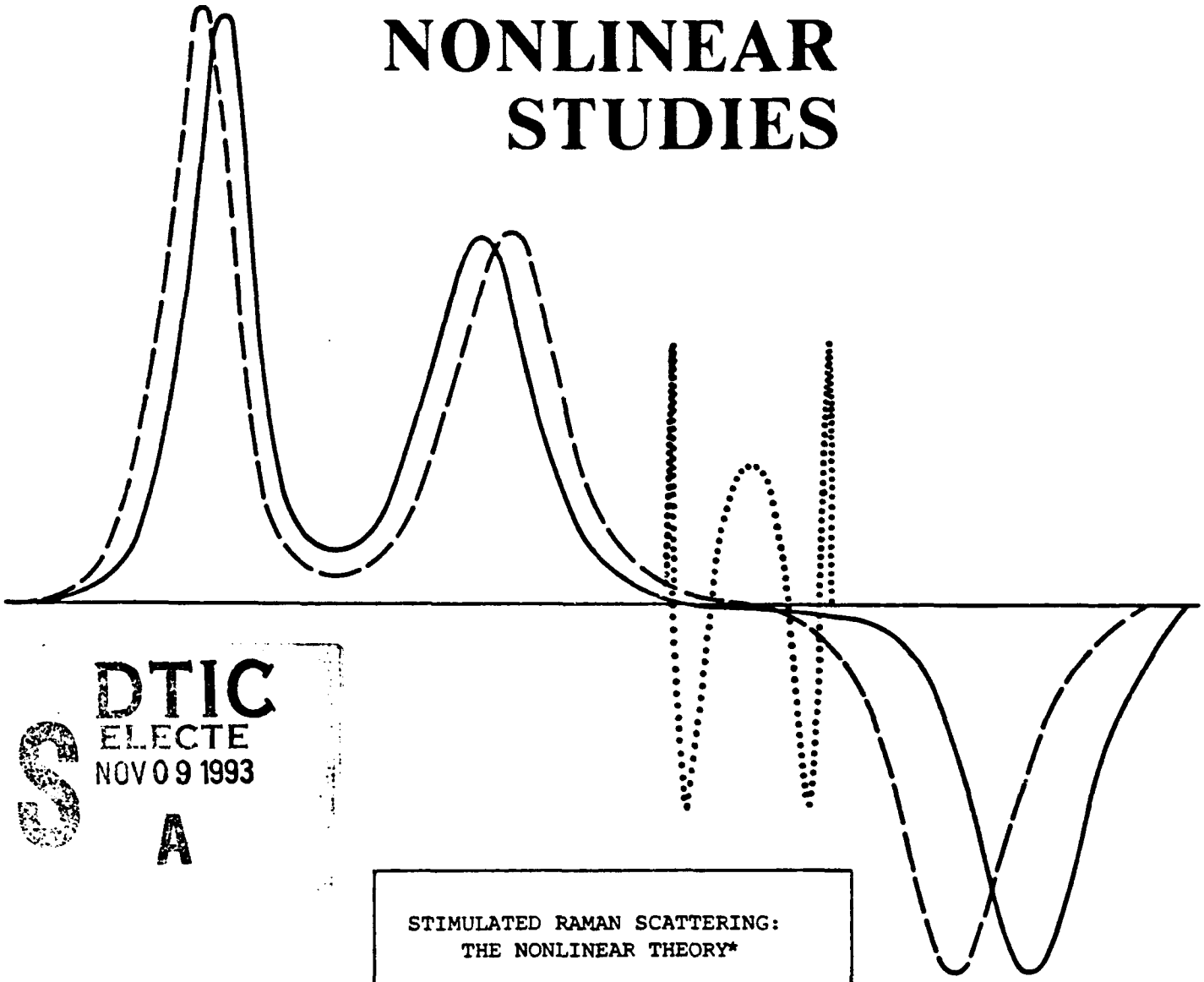

STIMULATED RAMAN SCATTERING:
THE NONLINEAR THEORY*

by

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Abstract

We give a simple review of some of the theoretical aspects of Stimulated Raman Scattering with particular emphasis on those features which would be of experimental interest.

Ever since Chu and Scott[1] first demonstrated that the Stimulated Raman Scattering (SRS) equations were integrable, these equations have proven themselves to be quite interesting from a theoretical point of view. These equations have been intensely studied[2-9] theoretically and numerically. We shall discuss here some of the more elementary and interesting aspects of these equations, with particular emphasis on those simple features that would be observed experimentially. We shall avoid as much as possible, the complexities of the Inverse Scattering Transform (IST) and restrict our discussions to those points that experimentalists would be interested in.

There are two forms of these equations. The first is the more general case where it is possible for the ground state of the molecules could become depleted[3,4]. This case, at the present, cannot be achieved experimentally, so we shall not consider or discuss it here. The usual experimential situation is where only a very small fraction of the available molecules can be excited. This is the case originally studied by Scott and Chu[1] and is the only case that we shall consider here. These reduced equations are what one usually calls the SRS equations. There have been some very enlightening numerical studies[6-8] of these equations in addition to all the analytical studies. And there have been very interesting analytical and experimental studies of SRS soliton evolution in the presence of damping[5,9]. The standard form of the reduced SRS equations are

$$\partial_x A_1 = -X A_2 \tag{1a}$$

$$\partial_\chi A_2 = X^* A_1 \tag{1b}$$

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

where we have allowed for a phenomenological molecular damping factor, γ. For pulses with a sufficiently short width, one can neglect this damping factor. We shall need to do this when we discuss the soliton solutions but for now, we shall retain this term. This damping factor will be used only in the first part of our discussions, after which we shall set it equal to zero. In (1), $A_1(A_2)$ is the envelope of the pump (Stokes) wave and X is the material excitation. χ and τ are the comoving coordinates

$$\chi = z \tag{2a}$$

$$\tau = t - z/u \tag{2b}$$

where z is the spatial coordinate, t is the laboratory time and u is the common group velocity of the pump and Stokes envelopes.

The nonlinearities in (1) are quadratic nonlinearities and there is no linear dispersion. Thus these equations are identical in form to the standard 3-wave parametric equations, called the 3-wave resonant interaction[10] (3WRI). However the SRS equations are a degenerate case of these equations since the group velocities of the pump and the Stokes are usually taken to be equal. Because of this, the SRS equations do not fit into the standard classification scheme[10] of the 3WRI equation, but rather they fall between two of the 3WRI cases. The classification scheme for the 3WRI cases depends on the ordering of the group velocities of the three waves and whether the pump wave (the high frequency wave) lies between the other two group velocities or not. If it does lie between them, then we have the the soltion decay case and if it does not, then we have the stimulated backscatter case. With the above, it is rather easy to see that the SRS equations are a degenerate case. Just note that if the group velocity of the Stokes wave were slightly higher than that of the pump, then the pump would have the middle group velocity and the SRS equations could then be classified as the soliton decay case. However if we just slightly shifted the group velocity of the Stokes pulse to be just below that of the pump, then we would have the other case of stimulated backscatter. Of course, in actuality, any experiment would fit into one or the other of

these case. But also in any experiment, the interaction length would never be long enough for the extremely small difference in the group velocities to have any effect. Thus in effect what we have is really a hybrid of the features of both of these two fundamental three-wave interactions. This demonstrates itself in that we see decay of the pump into daughter waves (the dominate feature of soliton decay) and also growth of nonlinear oscillations (the dominate feature of the stimulated backscatter case). On the other hand, some of the standard features of the standard cases will be absent. For example, the soliton decay case always has a threshhold area in order for decay to occur. However due to the approximate equality of the group velocities, there is no threshhold area for decay of the pump in SRS because the envelopes copropagate and never separate. (In soliton decay, the threshhold area can be interpreted as the width necessary in order for the interaction to build up before the envelopes separate.)

One of the main features of the SRS equations is the decay of the pump into a Stokes wave. This starts as a linear Raman instability and can best be seen by considering (1) in the limit of very slowly varying (in time) envelopes. In this case, we can ignore the time derivative in (1c) since the damping term dominates. Then we can then solve directly for X, obtaining $X = A_1 A_2^*/\gamma$. From (1a) and (1b) we obtain

$$\partial_x A_1 + \frac{1}{\gamma}(A_2^* A_2)A_1 = 0 \tag{3a}$$

$$\partial_x A_2 - \frac{1}{\gamma}(A_1^* A_1)A_2 = 0 \tag{3b}$$

Clearly (3a) is decaying while (3b) is growing. Consequently, the Stokes wave will grown from any noise at the expense of the pump.

In general, the stable configuration will always be a Stokes wave with no pump wave remaining. It is interesting to note also that it is only on such a background that one can have a soliton solution. Of course, solitons do not generally exist in the presence of any damping, so let us now assume that the time derivative will be so large (short pulse widths) that the damping constant, γ, can be neglected. Then a soliton solution of (1) is

$$A_1 = \frac{A(\tau)e^{i\phi}}{\cosh Z} \tag{4a}$$

$$A_2 = A(\tau)\tanh Z \tag{4b}$$

$$X = \frac{2\eta e^{i\phi}}{\cosh Z} \tag{4c}$$

where the arguement of the hyperbolic functions is

$$Z = 2\eta\chi - \frac{1}{2\eta}\int_{-\infty}^{\tau} A^2(s)ds \tag{5}$$

In the above, $A(\tau)$ is a real function of τ (otherwise arbitrary) and η and ϕ are real constants. This solution represents a localized disturbance on a Stokes background wherein the Stokes amplitude passes through zero and changes sign. This solution also has the feature that

$$A_1^* A_1 + A_2^* A_2 = A^2(\tau) \tag{6}$$

which is nothing more than the conservation of photon number. (From the derivation of (1), one can determine that the squared magnitude of the scaled amplitudes A_n are indeed photon numbers fluxes.) For every pump photon that is lost, a Stokes photon is created and vice versa. One can then interpret $A^2(\tau)$ as the total photon number flux as a function of the retarded time. If we fix χ, then $A^2(\tau)$ is simply the total flux of photons seen at χ as a function of t. And as the pump and Stokes envelopes copropagate through the medium, this profile of $A^2(\tau)$ never changes simply because for every photon lost in one envelope, another one has appeared in the other.

Another feature of these soliton solutions is that they are in general transient solutions. Given any pulse of finite duration, eventually any soliton solution will vanish. To see this, let us take an initial pulse profile where $A(\tau)^2$ has a finite area and also has some knid of a slowly decaying tail structure. The exact structure is not important, only that it eventually vanishes. Then as we shall now show, any soliton solution will move toward the back of the pulse, eventually falling into the tail region of $A^2(\tau)$ and vanishing. To show this, just look at the motion of the center of the soliton, which is at $Z = 0$. From (2) and (5), upon setting $Z = 0$ and differentiating, one obtains

$$\frac{dz}{dt} = u\frac{A^2}{A^2 + \eta^2 u} < u \tag{7}$$

Note that this velocity is always less than u, the group velocity of the electromagnetic waves. Thus the center of the soliton always moves slower than the wave envelopes which move at the group velocity, u. Consequently the soliton will always move to the back of the pulse and must eventually fall into the tail region of the pulse. In the tail region, $A(\tau)$ becomes smaller, and the soltion therefore also moves even slower. If $A(\tau)$ vanishes, then the soliton becomes stationary and by (4),

the envelope amplitudes of the soliton become zero. However, note that even if the field amplitudes vanish, one still will have the medium excitation [see Eq. (4c)] nonzero. What has happened is that the energy of the soliton has been deposited in the medium by leaving some of the molecules excited ($X \neq 0$). And the position of these excited molecules will become stationary. As long as $A(\tau)$ is nonzero, by (5), we have that the SRS soliton (and the material excitations) will travel forward, albeit slower than the pump and Stokes. However as one approaches the tail region of $A(\tau)$ where $A(\tau)$ is approaching zero, the soliton slows down [see Eq. (7)], eventually becoming stationary in the laboratory frame when τ is such that $A(\tau) = 0$. At the same time the electromagnetic components of the soliton, A_1 and A_2, have vanished.

Of course this brings to mind another way to create SRS solitons. Namely, create an excitation in the medium where the area of X is at least $\pi/2$. Now before this excitation can decay, inject the pump and/or a Stokes pulse. One will then see this soliton emerging on the background of the Stokes. This is also easily predicted from the analogy between SRS and SIT, presented by Dr. H. Steudel[11] in these same proceedings.

Let us now consider some more of the consequences of (1). As we have already remarked, the total photon number

$$A^2 = A_1^* A_1 + A_2^* A_2 \tag{8}$$

is conserved, not only for the soliton, but also for the general solution. ¿From (1a), (1b) and (8), it follows that

$$\partial_\chi A = 0 \rightarrow A = A(\tau) \tag{9}$$

and A will be only a function of τ. Then interpretating A^2 as the total photon number gives us the conservation of photon number as the pulses propagate through a Raman active medium.

Another point is that the evolution rate scales as the total photon number. Thus is we double the number of photons, the interaction proceeds twice as fast. This follows directly from (1c). In general we may scale A_1 and A_2 by $A(\tau)$. In Eqs. (1a) and (1b), since $\partial_\chi A = 0$, these factors of $A(\tau)$ just factor out, leaving the equations form invariant. However in (1c) they give (for $\gamma = 0$)

$$\partial_\tau X = A^2(\tau)[(A_1/A)(A_2/A)^*] \tag{10}$$

Now define a new (nonlinear) time, T by

$$T = \int_{-\infty}^{\tau} A^2(\tau)d\tau \tag{11}$$

then (10) becomes

$$\partial_T X = (A_1/A)(A_2/A)^* \tag{12}$$

which is now independent of the total photon number. So the only effect of $A(\tau)$ is to scale the evolution rate in (10). It has no effect on (1a) or (1b). And if we use the nonlinear time T to "clock" the evolution as in (12), then all effects except two become independent of $A(\tau)$. (Let's also note that back in Eq. (5), the last term in Z is exactly this nonlinear time, T.) Now these two effects that are dependent on $A(\tau)$ are i) the "clock" rate as discussed above and ii) how "long" the interaction will continue. To see this latter, first note that there is an upper limit to T given by

$$T_\infty = \int_{-\infty}^{\infty} A^2(s)ds \tag{13}$$

Now let us only consider the case where the pulse profiles are finite in extent and contain a finite number of photons. Consider the soliton solution, (4), when this T_∞ is finite. Then there will be a finite value of $\chi(= z)$, beyond which the soliton will not be found. This value is dependent on the total number of photons. If we double the number of photons, then T_∞ is doubled and the soliton will double the distance that it will propagate before stopping. The value of T_∞ (along with the amplitude η) determines the "clock" limit of the interaction. As soon as the nonlinear time T reaches this limit of T_∞, the interaction simply ceases even though the laboratory time is continuing. This is true of the general solution as well. Let us represent the pulse components by

$$A_1 = A(\tau)e^{i\alpha}\cos(\frac{1}{2}\beta) \tag{14a}$$

$$A_2 = A(\tau)e^{i\alpha}e^{-i\theta}\sin(\frac{1}{2}\beta) \tag{14b}$$

Then (1) becomes

$$\partial_\chi[e^{i\alpha}\cos(\frac{1}{2}\beta)] = -Xe^{i\alpha}e^{-i\theta}\sin(\frac{1}{2}\beta) \tag{15a}$$

$$\partial_\chi[e^{i(\alpha-\theta)}\sin(\frac{1}{2}\beta)] = X^*e^{i\alpha}\cos(\frac{1}{2}\beta) \tag{15b}$$

$$\partial_T X = \frac{1}{2} e^{i\theta} \sin\beta \tag{15c}$$

Now the equations are clearly independent of $A(\tau)$ except through the nonlinear time T. The initial value problem to be solved in SRS is given α, β and θ at $\chi = 0$ and X for $\tau \to -\infty$, construct the solution for A_1, A_2 and X. Integration of (15) will give this solution. Note that this solution will be the same for any profile $A(\tau)$ as long as these initial values are the same. The only manner in which $A(\tau)$ affects the solution is i) how fast the "ticks" of the nonlinear clock are compared to "ticks" of the laboratory clock $[\Delta T = A^2(\tau)\Delta t]$ and ii) the maximum time that the nonlinear clock is to run $(T \leq T_\infty)$.

The decomposition in (14) also illustrates that the dynamical part of SRS is simply the dynamics of the phases; α, β and θ. The overall amplitude $A(\tau)$ scales out and simply serves as a background on which the phases evolve. Of course, it is necessary to measure A^2 to determine the nonlinear clock rate and limit. But outside of this, there is no other need of A^2. Of these phases, the most important one appears to be β which determines the relative ratio of the Stokes to the pump. In most experiments, initially β is very small and as the pump converts into the Stokes, β rotates to either $+\pi$ or $-\pi$. The other two phases, α and θ, seem to play a lesser role and tend to be slaved to the major phase, β. However it is also more difficult to measure these phases, since one must measure the actual phase of the envelopes in order to obtain these. The most that we can say at the moment about these phases is that the presence of gradients in these phases probably will make it more difficult for SRS solitons to form.

Lastly, let me make a general remark in regard to the inverse scattering solution for the SRS equations, Eq. (1) with $\gamma = 0$ and try to explain simply how the IST works for any integrable system. To do this it may be simplest to take the original forms of Scott and Chu[1], which are

$$v_{1\chi} + i\zeta v_1 = X v_2 \tag{16a}$$

$$v_{2\chi} - i\zeta v_2 = -X^* v_1 \tag{16b}$$

$$v_{1\tau} = \frac{i}{4\zeta}(A_1^* A_1 - A_2^* A_2) v_1 - \frac{i}{2\zeta} A_1 A_2^* v_2 \tag{17a}$$

$$v_{2\tau} = \frac{-i}{2\zeta} A_1^* A_2 v_1 - \frac{i}{4\zeta}(A_1^* A_1 - A_2^* A_2) v_2 \tag{17b}$$

where ζ is called the spectral parameter. Note that (16) and (17) are an overdetermined set of equations. One may use (16) to construct the solutions for v_1 and v_2, given appropriate boundary values. Of course, one may use (17) to do the same thing. Now the key question is can one find a *common* solution to both (16) and (17). This now is a question of integrability. The way that it is answered is to cross-differentiation these two sets of equations, requiring that $\partial_\chi \partial_\tau v_n - \partial_\tau \partial_\chi v_n = 0$ for $n = 1$ and 2. In other words, a common solution to (16) and (17) will exist if and only if this cross-differentiation does give zero. (A simple example of this is that a force field can be given as a gradient of a potential if and only if the curl of the force field vanishes.) A simple exercise that anyone may do is to cross-differentiate (16) and (17). What you will find is that a common solution will exist, for arbitrary ζ, if and only if the SRS equations, (1), are true. This has a very simple and important consequence. If by some means we can construct a common solution for v_1 and v_2 which satisfies both (16) and (17), for arbitrary ζ, then *the potentials in (16) and (17) must and will satisfy the SRS equations.* Let us restate this as follows: *We solve the SRS equations by not solving the SRS equations.* Instead, we solve the *linear equations* (16) and (17). If we find any solution for v_1 and v_2 which satisfies both (16) and (17), then we are guarenteed that A_1, A_2 and X will satisfy (1) for $\gamma = 0$. If there is any secret or mystery about the IST, it is contained in this above statement.

A brief outline of how this method of solution is executed is as follows. One first chooses one of the above pair of equations to be the "eigenvaue problem". For SRS we choose (17) to be this eigenvalue problem. Since this problem is linear, we can use superposition and all the other known techniques for constructing solutions to this linear problem. In particular, we find it very useful to use scattering data to represent the general solution of (17), for arbitrary A_1 and A_2. Note that there is a "spectral parameter", ζ. This parameter serves as a "label" or "index" of the scattering data. Thus we express the general solution of (17) in terms of scattering data.

Now the last trick is to determine how the general scattering data will evolve in χ by using (16). Once this is done, then one has a general solution for v_1 and v_2 which satisfies both linear equations (16) and (17). Now one can reconstruct the potentials from the scattering data. We do that. Since both (16) and (17) are true, it therefore follows that the potentials will satisfy (1) and one has then constructed the solution of the nonlinear SRS equations (and done so by *not* solving the nonlinear equations). Of course, this has been only a simple description of how it is done. There are many technical details in do such and I shall simply refer you to the quoted literature for the mathematical details.

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