Spontaneous Generation of Raman Solitons from Quantum Noise

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Large phase shifts that arise during the quantum initiation of stimulated Raman scattering are shown to induce solitons in the pump intensity. These phase shifts are associated with phase waves, which were previously discovered in quantum superfluorescence theory.

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Although soliton solutions for stimulated Raman scattering (SRS) have been known for over a decade,[1] it is only relatively recently that solitons were observed experimentally by Drühl, Wenzel, and Carlsten.[2,3] This delay reflects the fact that the initial conditions required to reach the soliton steady states were unknown. In this regard, Ref. 2 is also of significant theoretical interest, for it furnished the necessary initial conditions. Specifically, it was shown that an instantaneous π phase shift in the Stokes source would lead to a subsequent, brief repletion of the pump intensity, in the form of a soliton. This discovery, which was suggested by an analysis of the experiment and confirmed by the numerical analysis of a semiclassical model, has since been substantiated analytically, by means of the inverse-scattering method,[4] and has been exploited in the generating (theoretically) of solitons by four-wave-mixing SRS.[5]

In this Letter, we report that Raman solitons may also be generated from quantum noise (see Fig. 1). These are associated with large ($\sim\pi$), rapid phase shifts in the Stokes field, which arise during quantum initiation. Thus, in contrast to the situation in Refs. 2, 3, and 5, where phase shifts were introduced externally through a Stokes seed pulse, solitons are here generated *spontaneously*. That this is not entirely unexpected can be appreciated by our recalling similar phase shifting seen in superfluorescence[6,7] and in swept-gain amplifiers.[8] We discuss this point further below.

The modeling of quantum SRS[9] is facilitated by a well-known method.[10–13] Rather than dealing with operator dynamics, one replaces the Heisenberg equations by stochastic equations that satisfy the following requirements: (1) The two models must yield identical field statistics during initiation (this is confirmed through linearization about the initial values); (2) the stochastic model must reduce to the corresponding nonlinear semiclassical model when the intensities become macroscopic. In addition, one must also specify a correspondence rule that relates the stochastic averages to a particular *ordering* of field operators. Since we are interested here only in the macroscopic reaction fields, the particular ordering prescription is irrelevant (i.e., from a computational standpoint). In the following, we adopt the antinormal-ordering correspondence.[12,13]

Under these conditions, we describe the pump field $A_L(\zeta,\tau)$, the Stokes field $A_S(\zeta,\tau)$, and the medium polarization $R(\zeta,\tau)$ and population inversion $R_3(\zeta,\tau)$ of the Raman-connected states by the c-number equations

$$\partial_\zeta A_L(\zeta,\tau) = -K_{LS}R^*(\zeta,\tau)A_S(\zeta,\tau), \tag{1}$$

$$\partial_\zeta A_S(\zeta,\tau) = K_{LS}^* R(\zeta,\tau)A_L(\zeta,\tau), \tag{2}$$

$$\partial_\tau R(\zeta,\tau) = -K_{LS}A_L^*(\zeta,\tau)A_S(\zeta,\tau)R_3(\zeta,\tau) - \gamma R(\zeta,\tau) + G(\zeta,\tau), \tag{3}$$

$$\partial_\tau R_3(\zeta,\tau) = 2[K_{LS}A_L^*(\zeta,\tau)A_S(\zeta,\tau)R^*(\zeta,\tau) + \text{c.c.}]. \tag{4}$$

Here, K_{LS} is a complex coupling constant, ζ is the distance along the propagation axis, and τ is the retarded time, defined so that $\tau=0$ coincides with the leading edge of the pump pulse for all ζ. Phenomenological, non-Hamiltonian terms representing collisional dipole dephasing are also present in (3), with the decay rate γ connected to the corresponding noise source $G(\zeta,\tau)$ through the fluctuation-dissipation relations[14]

$$\langle G(\zeta,\tau)\rangle_R = \langle G(\zeta,\tau)G(\zeta',\tau')\rangle_R = 0, \quad \langle G^*(\zeta,\tau)G(\zeta',\tau')\rangle_R = \gamma[N+R_3(\zeta,\tau)]\delta(\zeta-\zeta')\delta(\tau-\tau'), \tag{5}$$

where N is the number of atoms or molecules composing the sample, and where the subscripts R indicate that the averages are reservoir averages only. To complete the model, we must also specify the initial and boundary conditions.

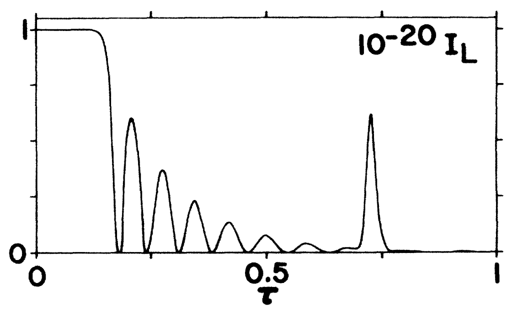

FIG. 1. SRS soliton (right) generated from quantum noise. Here, the pump intensity $I_L \equiv |A_L|^2$ is plotted as a function of the retarded time τ, for the parameters $N=10^{22}$, $K_{LS}=3\times10^{-20}$, $\gamma=4$, and $\Omega_L=10^{10}$. The values of τ and γ are scaled by the sample transit time, while the pump field is scaled such that $|\Omega_L|^2$ is the number of photons injected into the sample within a transit time. (The magnitude of K_{LS} reflects these scalings.)

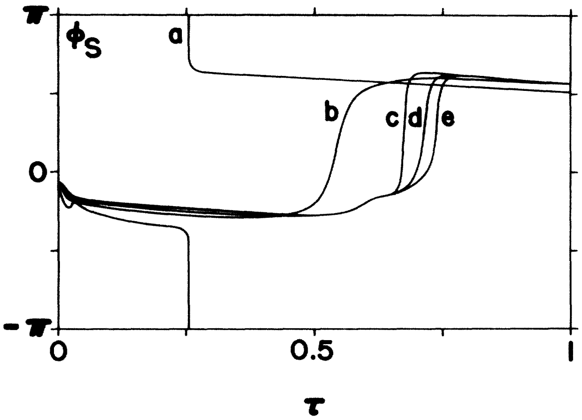

FIG. 2. Propagation of the Stokes phase ϕ_S. Curves a–e correspond to $\zeta=0.2$, 0.4, 0.6, 0.8, and 1.0, respectively. The parameter ζ is scaled by the sample length.

These are

$$A_L(0,\tau)=\Omega_L H(\tau), \tag{6}$$

$$\langle A_S(0,\tau)\rangle=\langle A_S(0,\tau)A_S(0,\tau')\rangle=0,$$
$$\langle A_S^*(0,\tau)A_S(0,\tau')\rangle=\delta(\tau-\tau'), \tag{7}$$

$$R(\zeta,0)=0, \tag{8}$$

and

$$R_3(\zeta,0)=-N, \tag{9}$$

where $H(\tau)$ is the Heaviside function. The averages in (7) are ensemble averages, and include all degrees of freedom. Since the Stokes field at $\zeta=0$ consists entirely of the vacuum, the relations (7) are derived directly from the free-field evolution, with the antinormal-ordering correspondence rule.

According to (6), (8), and (9), a constant pump field encounters an unpolarized medium, consisting of molecules exclusively in their ground states, at $\tau=0$. Because of (5) and (9), the dephasing fluctuations cannot induce a dipole moment via Eq. (3); rather, it is the vacuum source $A_S(0,\tau)$ whose statistics are Gaussian[12,13] and delta-correlated [as in (7)] that, scattering off the pump field in (3), provides quantum initiation. This interpretation is a consequence of our having chosen to model the antinormally ordered field averages.[12,13] As in theories of spontaneous emission,[15,16] however, there is a complementary interpretation—one may also formulate the quantum initiation in terms of dipole fluctuations[6–11] (radiation reaction), in which case the stochastic averages correspond to normally ordered operator averages. Let us note finally that Eqs. (1)–(4) are equivalent, in the semicalssical limit, to those studied in Refs. 2 and 3 if it is further assumed that the medium ground state remains undepleted: $R_3(\zeta,\tau)\equiv -N$.

Equations (1)–(4) were simulated numerically, and solved for an ensemble of 500 members. One of these is represented by Fig. 1. There, we see the pump intensity, initially constant, first deplete, then undergo relaxation oscillations, and finally exhibit a soliton, just as in Refs. 2 and 3. [Because of the photon-number conservation law inherent in Eqs. (1)–(4), it is clear that the Stokes intensity exhibits a corresponding solitary *trough*.] In comparison with the experiments, where a Stokes seed pulse was used, a much larger Raman gain is required to ensure pump depletion and, hence, soliton formation; this requirement is reflected in our choice of the incident pump intensity.

Evidence of solitons was seen in twelve members of the ensemble. Because the position of such solitons is random, some of these occurred amid the relaxation oscillations. However, if one propagates such examples further through the medium, the solitons move away from the leading edge of the pump field, while the oscillations move in the opposite direction—thus, these solitons are eventually resolved.

To demonstrate the connection mentioned above with phase shifting, we have plotted in Fig. 2 the phase $\phi_S(\zeta,\tau)$ of the Stokes field at various propagation distances, for the case represented by Fig. 1. A large ($\sim\pi$) phase shift is clearly evident for each value of ζ. (Within the interval $0.2<\zeta<0.4$, the shift undergoes a transition from one of clockwise to one of counterclockwise rotation.) Notice that, as the phase shift propagates, it moves away from the leading edge ($\tau=0$) of the pump intensity and accelerates.

In Fig. 3, we have depicted also, in addition to $\phi_S(\zeta,\tau)$, the pump and polarization phases [respectively, $\phi_L(\zeta,\tau)$ and $\phi_R(\zeta,\tau)$] at a single value of ζ. Notice here that the Stokes phase shift is "echoed," after a short delay, in $\phi_R(\zeta,\tau)$. If we define the relative phase $\phi(\zeta,\tau)\equiv\arg[K_{LS}R^*(\zeta,\tau)A_S(\zeta,\tau)]$, then it is clear, as shown in the inset, that $\phi(\zeta,\tau)$ peaks at $\approx 3\pi/4$, immediately fol-

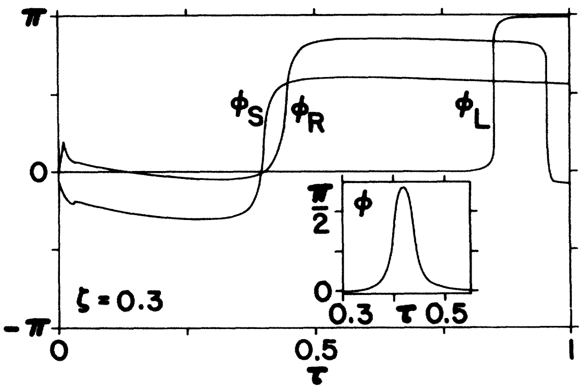

FIG. 3. Stokes, pump, and polarization phases (ϕ_S, ϕ_L, ϕ_R) at $\zeta=0.3$, plotted as functions of the retarded time τ. Inset: A phase wave (see text).

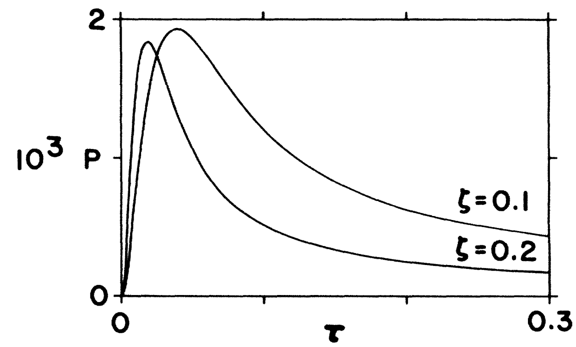

FIG. 4. The probability P of finding $|\phi| > 3\pi/4$, plotted as a function of τ, at two values of ζ.

lowing the shift in $\phi_S(\zeta,\tau)$. Such features have been called *phase waves*[6–8] (see below). Before leaving this figure, let us point out that the phase shift seen in $\phi_L(\zeta,\tau)$ is of a wholly different origin. This, in fact, corresponds merely to the first of the relaxation oscillations. In contrast to the solitary phase shift evident in $\phi_S(\zeta,\tau)$, this feature propagates toward $\tau=0$, and is followed by a regular series of similar shifts in the pump phase; $\phi_L(\zeta,\tau)$ eventually resembles a square wave, except near the soliton peak.

It is obviously a difficult, if not impossible, task to characterize the soliton statistics analytically. We have seen, however, that the phase shifts needed to generate solitons are associated with phase waves in $\phi(\zeta,\tau)$, and one may therefore follow Hopf and Overman[6,8] in analyzing the phase-wave statistics. Examples relating to the previous plots are shown in Fig. 4. There, we see a pronounced peak in the relative-phase probability, which propagates (like the point of quantum initiation) towards the front edge of the pump pulse. Results like these should be valuable in identifying factors that enhance phase-wave, and hence soliton, generation.

To conclude this Letter, let us clarify an important point. Drühl has discovered,[17] in investigations of the semiclassical dynamics, that when instantaneous Stokes phase shifts of magnitude other than *exactly* π are introduced, the solitonlike structures induced in the pump intensity invariably decay. It is clear that in our model, as in experiments, perfect π phase shifts almost certainly never occur. Therefore, the solitons discussed here cannot be considered solitons in a mathematical sense. Rather, they are transient structures that appear to stabilize over *finite* domains of space and time. One can draw an analogy with a ball rolled to the crest of a frictionless hill—as the ball nears its steady-state position (i.e., the crest) it slows and thus appears to stabilize, but eventually rolls away. In spite of this instability, we have chosen, for historical reasons (like the authors of Ref. 5), to retain the term "soliton" in referring to these structures, while emphasizing their transience.

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