Observation of Solitary Waves with Different Phase Behavior in Stimulated Raman Forward Scattering

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Solitary waves originating from quantum noise in stimulated Raman forward scattering are interferometrically investigated. The experimental data obtained proves the existence of solitary waves of different character. This finding is also supported by numerical calculations. The observed solitary pulses are interpreted as either a solitary wave, an antisolitary wave, or a breather. [S0031-9007(98)07954-X]

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Solitary waves in stimulated Raman forward scattering (SRFS) which are different from solitons in fibers have been considered for almost 20 years [1]. Originally of more academic interest, they have become a subject of growing attention, both experimentally and theoretically, after the first evidence of their occurrence [2]. They basically result from a phase jump in the Stokes field and manifest themselves as spikes in the depleted pump with a lifetime long compared to the medium dephasing time T_2 but with a duration shorter than or about equal to T_2. The phase jump can be deterministically generated [3,4], or originates from quantum-noise fluctuations of the Stokes wave and the medium at the beginning of the scattering process [5–7]. The phase behavior of the Stokes wave and the mutual phase relations between the pump and Stokes fields and medium polarization have been thoroughly investigated by a number of authors [8–13]. However, to the best of our knowledge, the question of whether the phase of the solitary wave occurring in the depleted pump has only one form of appearance at its disposal or perhaps more has not received very much attention so far [1]. The study of this phase behavior is the subject of the present paper.

Our interferometric measurements prove that there are three different types of solitary waves originating from quantum noise. This is an observation which has been unknown in SRFS before. The quantity used to distinguish the various waves from each other is their phase relative to that of the undepleted pump. The first two types of waves differ from each other in that their phases are opposite in sign and the third wave type has an oscillating phase. The numerical calculations of SRFS from quantum noise agree with these experimental findings. The waves may be viewed as solitary, antisolitary, and breather (for the introduction of these terms, see, e.g., Eilenberger [14]).

The experimental setup used to analyze the phase structure of the solitary waves is essentially a modified nonsymmetrical Mach-Zehnder interferometer with the Raman cell located in one of its two arms (see Fig. 1). SRFS is excited by the second harmonic of a single frequency Q-switched Nd:YAG laser emitting pulses of 15-ns FWHM duration and roughly Gaussian shape, both temporally and spatially. The 2ω pulse energy runs up to 10 mJ. The pump beam is directed into the Raman cell filled with molecular hydrogen through an aperture of 1-mm diameter (Fig. 1). The Fresnel number of the interaction volume, $N_F = A/(\lambda L)$, is about one. Here A denotes the cross section of the pump beam, λ the wavelength of the Stokes wave, and $L = 140$ cm the effective interaction length. The low N_F value ensures that only a single transverse mode of the Stokes beam is excited. Raman scattering is due to the excitation of the $Q(1)$ vibrational line of H_2 with a wave number of 4155 cm^{-1} yielding a Stokes wavelength of $\lambda = 683$ nm. The hydrogen pressure amounts to 5×10^6 Pa with a corresponding relaxation time of the molecular vibrations, T_2, of 0.15 ns. Under these conditions, forward scattering strongly dominates backward scattering and anti-Stokes scattering.

The solitary waves are recorded in two different ways. The first observation channel monitors the depleted pump intensity using the photodiode 12, the delay cable 13, and

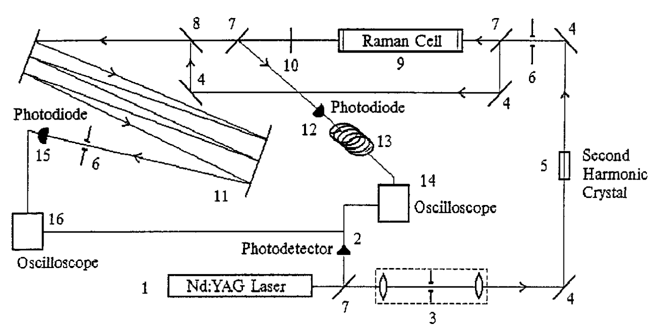

FIG. 1. Experimental setup. 1—Nd:YAG laser; 2—photodetector for triggering both oscilloscopes 14 and 16; 3—spatial filter; 4—highly reflecting mirrors for 1064 or 532 nm; 5—second harmonic generator; 6—apertures; 7—beam splitters; 8—50% mirror; 9—high-pressure Raman cell; 10—glass filter absorbing at 683 nm and transparent at 532 nm; 11—optical delay line; 12 and 15—photodetectors; 13—cable delay line.

the oscilloscope 14 (see Fig. 1) with a time resolution of about 0.8 ns. The second channel receives the interferometric signal. This is generated by superimposing portions of the undepleted and depleted pump pulses via mirror 8. The optical delay line 11 (in air) in conjunction with photodiode 15 and oscilloscope 16 provides a time resolution of 0.2 ns. A proper choice of the length of the two delay lines enables a simultaneous observation of the signals produced by the solitary wave in each channel and hence any correlation between them. Because of the somewhat low time resolution of the first channel, fine details of the solitary wave on the time scale of T_2 remain unresolved.

In 90% of the shots, the pump pulses are emitted in single mode, both longitudinally and transversely. Single longitudinal mode operation is checked in a separate interferometric measurement by looking for modulations of the pump pulse intensity at each shot. Pulses exhibiting even small amounts of modulations are excluded from further consideration.

A typical input pump pulse (upper trace) and a depleted pump pulse recorded in separate measurements with the second channel and containing three solitary waves generated simultaneously (lower trace) are pictured in Fig. 2(a). The number of solitary pulses per shot, the positions where they show up in the depleted pump, and their intensity courses change statistically from shot to shot [7]. In a series of 420 shots, about 470 solitary pulses were registered. Most of them had small intensities, which is in accordance with previous results [6,7]. Figure 2(b) shows the depleted pump pulse intensity (upper trace) recorded with the first channel and the interference signal (lower trace) resulting from the superposition of the input and output pump pulses and received via channel two. In this shot, two solitary waves of opposite phase can be clearly distinguished. The unambiguous existence of such solitary waves can be proven only in a single-shot experiment. Only in this case, it can be definitely ruled out that mechanical vibrations which may change the phase relation between the depleted and undepleted pump pulses in the interferometer perturb the measurements. This is due to the fact that the time scale of the mechanical vibrations is larger by orders of magnitude than the few-ten nanosecond time scale of the Raman scattering process. Solitary waves with a phase opposite to that of the pump pulse are observed only in 4% or 17 out of the 420 evaluated shots. The phase of the solitary wave hence usually coincides with that of the exciting wave. Occasionally, solitary waves are registered whose phase abruptly changes during their time of existence, as shown in Fig. 2(c). At present, we have no real explanation at hand why the in-phase solitary waves occur so much more frequently than those with opposite phase. We will pay attention to this point, which may provide information on the nature of the quantum fluctuations in a future investigation.

The observations as described above can be reproduced by a plane-wave theory of SRFS when the Stokes wave is generated from quantum noise [11,12]. The model

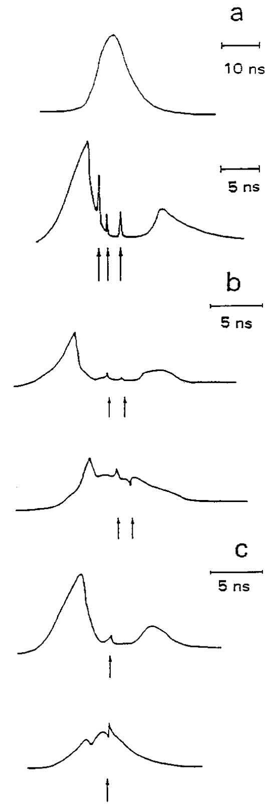

FIG. 2. (a) Intensity traces of the input pump pulse (top) and depleted output pump pulse (bottom) with three solitary waves (marked by arrows) recorded by detector 15 (see Fig. 1), when the corresponding arm of the interferometer was closed. The time scales correspond to 10 ns (top) and 5 ns (bottom). (b) Occurrence of two spikes in the depleted pump: intensity trace (top) recorded with detector 12 and interferometric trace (bottom) recorded with detector 15 and showing that the two spikes correspond to two solitary waves of opposite phases. The time scale is 5 ns. (c) A solitary wave changing its phase during its duration: intensity trace (top) recorded by detector 12 and interferometric trace (bottom) recorded by detector 15. The time scale is 5 ns.

considers the mutual interaction between the slowly varying pump and Stokes field envelopes, $E_P(z,t)$ and $E_S(z,t)$, and the molecular medium of polarizability, $Q(z,t)$, in terms of the following coupled nonlinear equations [11]:

$$\partial E_P/\partial z = (\pi/c) i\omega_P N (\partial\alpha/\partial q)_0 Q E_S ,$$
$$\partial E_S/\partial z = (\pi/c) i\omega_S N (\partial\alpha/\partial q)_0 Q^* E_P ,$$
$$\partial Q/\partial t = -Q/T_2 + (i/4m\omega_0)(\partial\alpha/\partial q)_0 E_P E_S^* .$$

Here $\partial\alpha/\partial q$ denotes the molecular differential polarizability; m is the mass of the H_2 molecule, ω_0 is the resonance frequency of the Raman transition, $\omega_{P,S}$ are the carrier frequencies of the pump and Stokes fields, and N is the concentration of the H_2 molecules.

In the simplest case, it is sufficient to introduce the quantum noise into the SRFS equations solely through the Stokes field noise at the input boundary, $z = 0$ [11]. At this position, the Stokes field, $E_S(z = 0, t) = E_{S0}(t)$, is treated as a complex Gaussian quantum-noise source with $\langle E_{S0}(t)\rangle = \langle E_{S0}(t)E_{S0}(t')\rangle = 0$ and $\langle E_{S0}(t)E^{*}_{S0}(t')\rangle = \delta(t - t')$. If $E_{S0}(t)$ is represented by $A(t)e^{i\varphi(t)}$ then $A(t)$ has a Gaussian and $\varphi(t)$ a uniform distribution. For the field envelope of the pump pulse, we took the boundary condition $E_P(z = 0, t) = A_P \exp(-t^2/t_0{}^2)$ with the peak intensity (proportional to A_P^2) ranging from 0.1 to 0.5 GW/cm^2 and a pulse duration t_0 of 10 ns. The medium is assumed to always remain in the ground state (depletion neglected) and to be completely unpolarized prior to interaction leading to the initial condition $Q(z, t = 0) = 0$. Dispersion is neglected, too.

First, the depleted pump pulse was calculated at the output of the Raman cell for conditions close to those used in the experiment. Almost each depleted output pump pulse contained one solitary spike and often more. Figure 3 demonstrates some results of our numerical calculations for the real part of the depleted pump field (the real part is chosen because its sign is reversed when a π-jump occurs) and the interference signal. The insets show the intensities of the depleted pump (solid line) and Stokes (dashed line) pulses for the moment of the solitary wave creation. The phase behavior of the three pictured solitary waves corresponds to that experimentally observed. We can hence conclude that both experiment and numerical simulation prove the existence of solitary waves exhibiting opposite phases and an oscillating phase.

Another question of interest is how these solitary waves develop when they further move through the Raman medium beyond the distance of one meter as investigated so far. Does a solitary wave of a certain type once originated from noise remain unchanged upon propagation, or are transformations possible? This point is studied in Fig. 4 for a spike with an initially oscillating field envelope. The changes occurring with the distance traveled resemble breatherlike behavior (see, e.g., [1]).

Another type of change is pictured in Fig. 5. Over a distance of about 2 m, an initially fluctuating field envelope is then transformed back again into an antisolitary wave whose negative phase stabilizes for distances beyond 2 m. The increase of the depleted pump region upon propagation is accompanied by newly developing field oscillations becoming the more prominent the longer the distance traveled. The occurrence of a transition regime prior to stabilization seems to be quite a general

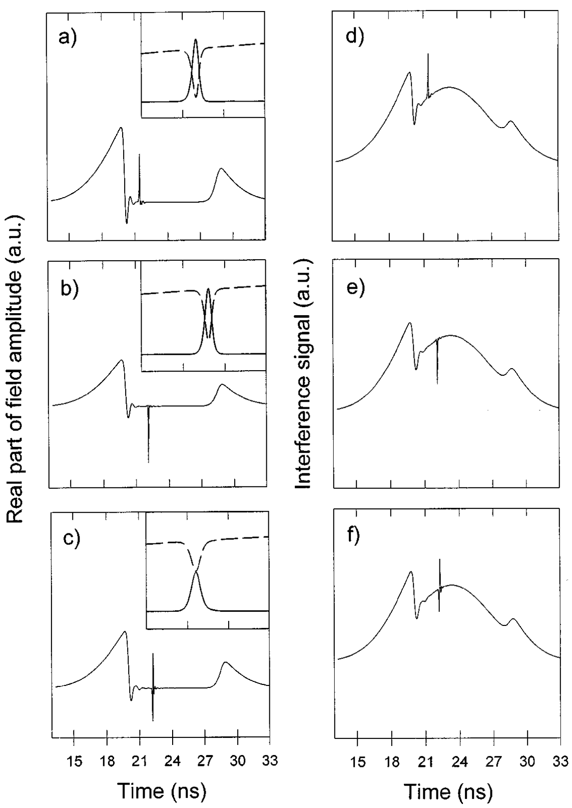

FIG. 3. Results of numerical calculations for SRFS originating from quantum noise. (a)–(c): Real part of the field amplitude for the depleted pump pulses at the output of the Raman cell. The insets show the intensities of the depleted pump (solid line) and Stokes (dashed line) pulses. The pump peak and the Stokes dip are not equal because the associated photon energies are different. (d)–(f): Corresponding interference signals resulting from the superposition of portions of the input and depleted pump pulses. Peak pump intensity 0.4 GW/cm^2. Length of the Raman cell, $L = 1$ m.

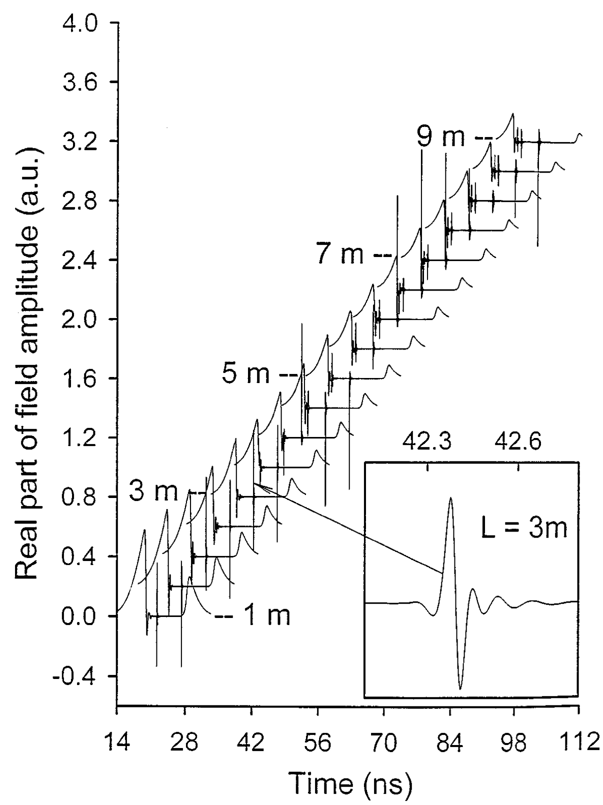

FIG. 4. Breatherlike behavior of a solitary wave upon propagation. The first trace in the plot on the lower left portrays the pump pulse after traveling a distance of 1 m in the Raman medium. The distance between neighboring traces corresponds to 0.5 m. Peak pump intensity 0.4 GW/cm^2. The inset is a magnified representation of the solitary wave after traveling a distance of 3 m in the Raman medium.

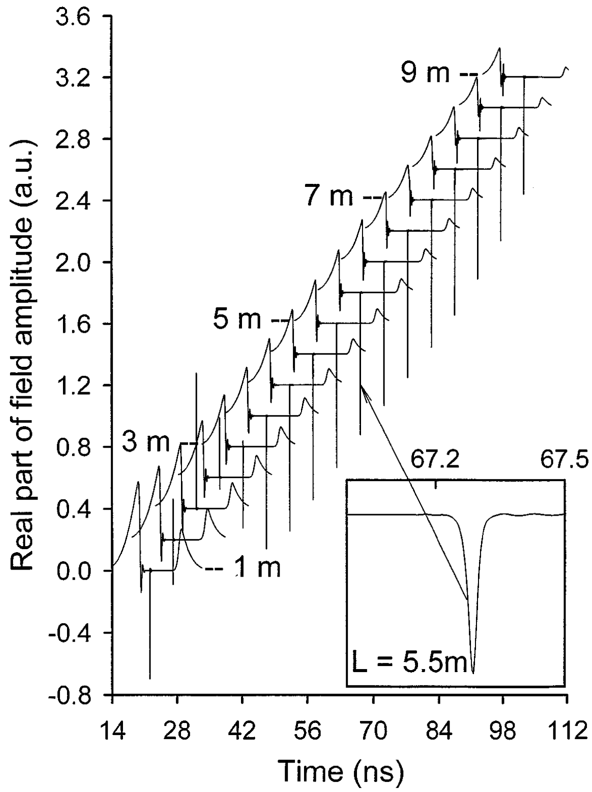

FIG. 5. Development of an antisolitary wave. Peak pump intensity, first trace, and distance between neighboring traces as in Fig. 4. The inset is a magnified representation of the solitary wave after traveling a distance of 6 m in the Raman medium.

phenomenon. The propagation behavior which is qualitatively similar to that described above was also observed for a positive phase solitary wave which after undergoing some transformations finally moves on as a solitary wave. Each type of solitary waves exists over distances much longer than $cT_2 = 4.5$ cm, confirming the solitary nature of these objects. When a π-phase jump is artificially introduced in the Stokes field as realized in [4], no transition region occurs. Obviously, a π-phase jump yields a stable solitary wave.

In conclusion, we have investigated the phase behavior of solitary waves in SRFS originating from quantum noise. This behavior turns out to be rather complex according to our experimental and numerical data and cannot be observed by considering the intensity only. Some typical scenarios were singled out showing the existence of three different types of solitary waves which can be in phase, out of phase, or oscillate relative to the input pump field. The in-phase waves are the solitary waves mainly studied in the past. To the best of our knowledge, the out-of-phase waves or antisolitary waves as well as the waves with an oscillating phase also called breather (see, e.g., [14]) were not observed in SRFS experiments before. The individual wave types occur with different probabilities. While solitary waves were found in almost any depleted pump pulse, antisolitary waves and breathers could be detected only in a small percentage of the evaluated shots.

There exists some similarity between the solitary waves as described here and the solitary solutions found for the integrable sine-Gordon equation: kink, antikink, and breather. However, the SRFS equations as used here and the sine-Gordon equation are mathematically not completely equivalent [11,15] due to the presence of the dephasing term Q/T_2 in our equations which makes them analytically nonintegrable. Our findings are also useful in that they provide new information on the nature of quantum noise.

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