Statistics of the energy fluctuations of Stokes pulses in stimulated Raman scattering in various situations[1)]

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(Submitted 26 December 1985)
Pis'ma Zh. Eksp. Teor. Fiz. **43**, No. 3, 118–122 (10 February 1986)

A change in the statistics of the energy of Stokes pulses has been discovered upon an intensification of a monoenergetic pump. The change is from a distribution with a mode near zero to a stabilized distribution with a nonzero mode. A region of bistability with a bimodal distribution has been found. A sharp edge has also been found in the low-energy wing of the distribution in nonlinear stimulated Raman scattering in a broad-band pump field.

1. During stimulated Raman scattering, the energy of the Stokes scattered pulses undergoes pronounced fluctuations. The statistical properties of these fluctuations, which arise from quantum noise and which are amplified to macroscopic levels, have recently been the subject of active research, both theoretical[1–7] and experimental.[8–11] Raymer and Mostowski[1] have predicted, and Walmsley and Raymer[8] and Fabricius *et al.*[9] have observed, that under conditions (a)–(c) listed just below, the statistical properties of the 'nucleating" spontaneous photons are conserved during amplification, and the fluctuations in the energy of the Stokes pulses are described by an exponential distribution function

$$\mathcal{P}(W) = \langle W\rangle^{-1}\exp(-W/\langle W\rangle), \qquad (1)$$

where $\langle W\rangle$ is the mean energy of the Stokes pulses. For a distribution of this type, there are significant fluctuations in the energy, with a standard deviation of 100%, even when the pump pulses are completely reproducible.[9] According to distribution (1), the most probable event corresponds to a Stokes pulse with an approximately zero energy.

Here are the three conditions:

(a) The stimulated Raman scattering are time-varying processes ($\gamma = \tau_{\text{pulse}}/T_2 g_s I_p L \lesssim 1$, where τ_{pulse} is the length of the pump pulse, T_2 is the vibrational relaxation time, g_s is the gain of the stimulated Raman scattering, I_p is the intensity of the pump pulse, and L is the length of the stimulated-Raman-scattering medium).

(b) There is no reduction of the pump or of the level populations (linear stimulated Raman scattering; the coefficient of the conversion of pump energy W_L into the energy of Stokes pulses, W, i.e., $\eta = W/W_L$, is much less than unity).

(c) The pump is unimodal in terms of spatial characteristics (the Fresnel number $F = A/\lambda_s L$ of the active zone for the stimulated Raman scattering is less than unity; here A is the cross-sectional area of the zone, and λ_s is the wavelength of the Stokes radiation) and also in terms of temporal characteristics ($\Delta\omega_p T_2 \ll 1$, where $\Delta\omega_p$ is the spectral width of the pump).

If any of conditions (a)–(c) is violated, the distribution $\mathcal{P}(W)$ will not be the exponential distribution in (1). For example, during the excitation of several spatial modes ($F > 1$) the distribution $\mathcal{P}(W)$ will be approximately a gamma distribution[11]

$$\mathcal{P}_N(W) = \frac{1}{(N-1)!}\,\frac{W^{N-1}}{\langle W\rangle^N}\exp(-W/\langle W\rangle), \qquad (2)$$

where N is the number of statistically independent spatial modes. Raymer *et al.*[7] have derived a theory incorporating the transverse distribution of the pump.

On the other hand, as the pump intensity is increased, condition (b), i.e., the linearity of the conditions for the stimulated Raman scattering, is violated. This circumstance changes the statistical properties of the nucleating photons in the propagation process and thus stabilizes the fluctuations of W with a nonzero mode (see the experiments of Ref. 10 and the theoretical explanation in Refs. 5 and 6). Just how the transformation from distribution (1) to a stabilized distribution occurs, however, remains an open question. We have carried out an experimental study of the statistics of the energy of Stokes pulses in the course of a transformation of this sort, specifically, in the transition from a linear, time-varying stimulated Raman scattering to a nonlinear version of this scattering with increasing intensity of the pump, with either a broad or narrow spectrum.

2. The first Stokes component of the stimulated Raman scattering ($\lambda_s = 683$ nm) is excited in a cell filled with molecular hydrogen (hydrogen pressure $p = 50$ atm,

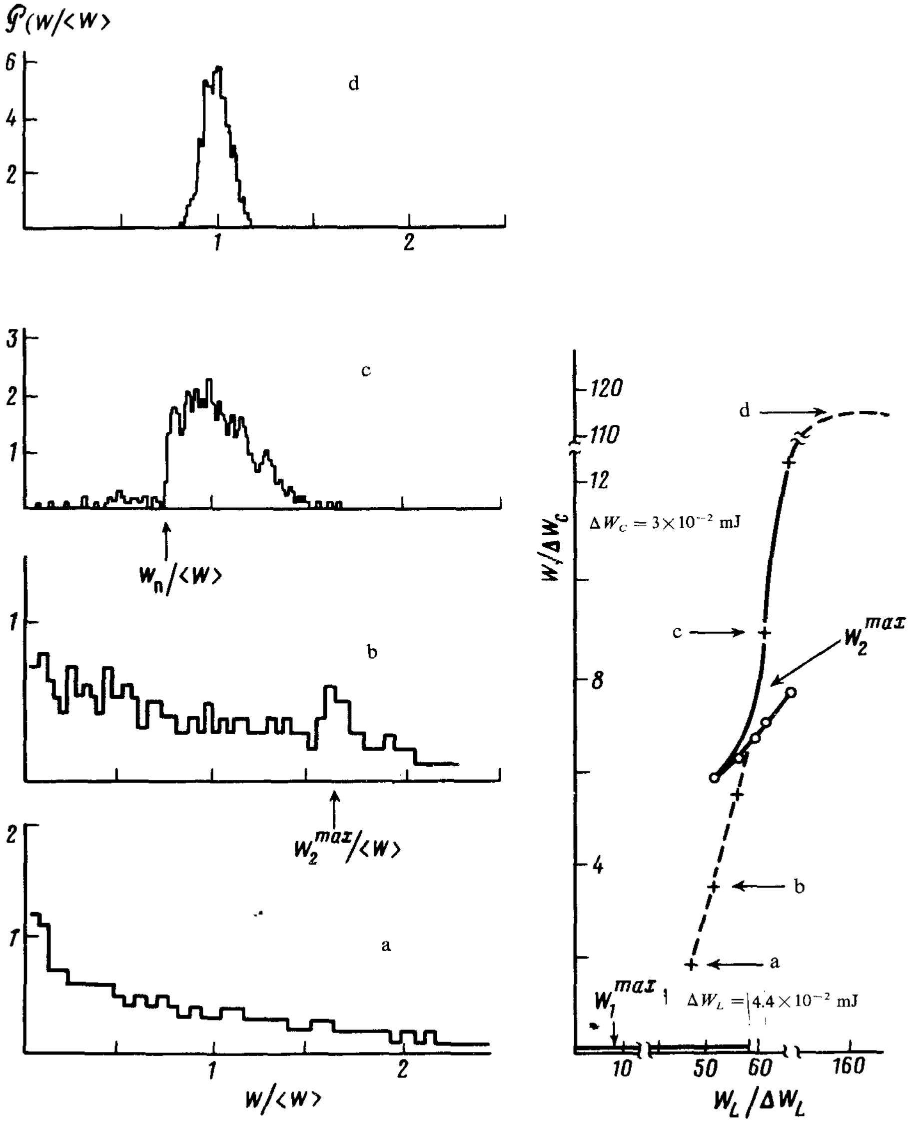

FIG. 1. Mean energy of the Stokes pulses, $\langle W\rangle$ (dashed line; the plus signs show experimental data), energy of the second peak in the distribution, $W_2^{\max}$ (solid line), and energy of the sharp edge, W_t (circles), versus the energy of the laser pulses, W_L; histograms of the energy distribution of the Stokes pulses for various efficiencies of the conversion of the broad-band pump ($\Delta\omega_p \sim 0.2$ cm^{-1}) in the regime of a time-varying ($\gamma \approx 1$–3) stimulated Raman scattering. Efficiencies: a—$\eta = 1\%$; b—$\eta = 4\%$; c—$\eta = 6\%$; d—$\eta = 46\%$.

$L = 120$ cm, $T_2 = 0.148$ ns). The exciting light is the second harmonic ($\lambda_p = 532$ nm) from an Nd:YAG laser with a pulse length $\tau_{\text{pulse}} = 10$–15 ns, an energy up to 10 mJ, and a pulse repetition frequency of 10 Hz (Ref. 12). The oscillator has an unstable resonator with a polarization output; the output light is amplified in an amplifier. The width ($\Delta\omega_p$) of the laser light is reduced by means of a Fabry-Perot etalon to 0.2 cm^{-1} through electrooptic Q switching or to 0.06 cm^{-1} through passive switching with a LiF crystal with F^{2-}-centers. The exciting light, whose intensity is varied over the range $I_p = 10^7$–10^9 W/cm^2 by means of a Glan prism or filters, is focused into the cell by a lens with $f = 30$ cm. The light emerging from the cell is collected by a lens ($f = 50$ cm) and resolved by a glass prism into its spectral components. The Stokes pulse is selected from these components by means of a diaphragm and filters. The energies of each Stokes pulse and of the laser pulse that excites it are measured by photodiodes. The electrical pulses are converted into digital form by an analog-to-digital converter and sent to a Mera-60 minicomputer, operated as a two-dimensional analyzer. The measurement system distributes the energies of each laser pulse and the Stokes pulse that it excites into one of the 256 intervals (channels) and thereby makes it possible to obtain a set of histograms of the energies of the Stokes pulses. Each histogram corresponds to a definite value of the energy of the laser pulses (within 0.5–1%, depending on the particular channel$^{2)}$). The sample volume for each histogram ranges from 10^3 to 10^4.

3. From the experimental results, shown in Figs. 1 and 2, we draw the following conclusions:

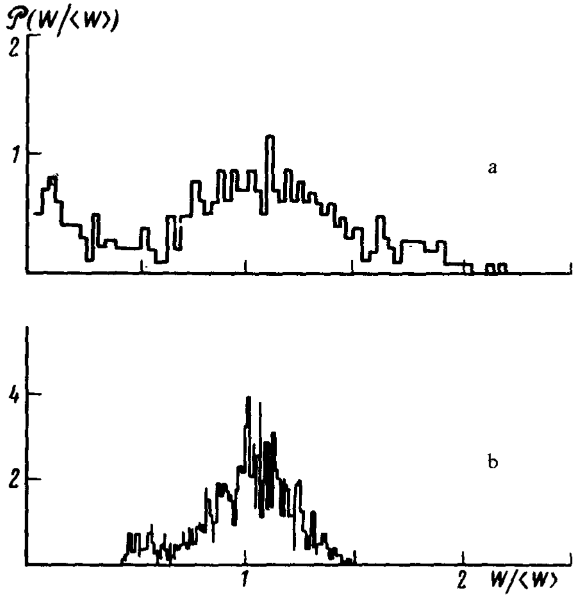

FIG. 2. Histograms of the energy distribution of the Stokes pulses for a narrow-band pump ($\Delta\omega_p \approx 0.06$ cm^{-1}). a—$\eta \approx 5\%$; b—$\eta \approx 20\%$.

- At relatively low pump intensities [$I_p \lesssim 10^8$ W/cm^2, with an average conversion efficiency $\eta < 1\%$) the energies of the Stokes pulses are distributed with a mode value $W = W_1^{\max}$ of approximately zero$^{3)}$ [Fig. 1(a)]. The histograms found in this case differ from the exponential form in (1) because the Fresnel number of the excited region of the cell varies over the range 1–5, depending on the diameter of the diaphragm in the laser beam.
- At efficiencies of the stimulated-Raman-scattering conversion for which the reduction of the pump cannot be ignored ($\eta > 1\%$), a peak appears in the distribution $\mathscr{P}(W)$ at nonzero values of $W = W_2^{\max}$ [Figs. 1(b) and 2(a)]. There is an interval of pump intensities in which these two peaks are present simultaneously (a bistability).
- At higher values of the conversion coefficient ($\eta \approx 5$–6%), the first peak, at $W_1^{\max}$ disappears. The second peak has a sharp edge (a threshold) in the low-energy part of $\mathscr{P}(W)$ at the point $W = W_t$ [Fig. 1(c)]. This "threshold" occurs only in the case of a broad-band pump [the threshold is observed at $\Delta\omega_p \approx 0.2$ cm^{-1}; it is not found at $\Delta\omega_p \approx 0.06$ cm^{-1} (Fig. 2)]. It should be noted that W_t corresponds to a constant conversion efficiency ($\eta = \eta_t$). For this particular experiment, we find $\eta_t \approx 5.6\%$.
- For the very nonlinear version of stimulated Raman scattering ($\eta \approx 40\%$), the fluctuations in the energy of the Stokes pulses stabilize near $W_2^{\max}$ [Figs. 1(d) and 2(b)]. The smallest value found by us for a standard deviation of a stabilized distribution in these measurements is 4%. For the histogram in Fig. 1(d) the stabilization is 8%.

4. Such aspects of the energy distribution of the Stokes pulses of stimulated Raman scattering as a distribution with an approximately zero mode value in a time-varying linear regime and a stabilization of the distribution in a nonlinear regime of stimulated Raman scattering agree well with the theoretical predictions of Refs. 2, 5, and 7. On the other hand, the bistability phenomena found in the region of the transition to the nonlinear regime and the existence of a low-threshold edge do not yet have an adequate theoretical explanation. It might be suggested that the sharp edge appears in the distribution because of phase locking during broad-band pumping,[13] while the bistability results from the pronounced sensitivity of the distribution to spatial fluctuations of the pump.[7,11]

It should also be noted that these studies are pertinent to a range of problems broader than simply the problems of stimulated Raman scattering, because the system studied by us is an example of an unstable nonlinear quantum system in which microscopic quantum fluctuations are amplified to macroscopic, observable levels. Another example of this sort would be fluctuations in the delay time of the superradiance pulses observed in Refs. 14 and 15.

We wish to thank P. A. Apanasevich, V. A. Orlovich, G. V. Venkin, Yu. E. D'yakov, and S. Yu. Nikitin for useful discussions of these results. One of us (S. K.) wishes to thank D. von der Linde and K. Nattermann for furnishing papers before their publication.

[1] This study was reported at the Twelfth Conference on Coherent and Nonlinear Optics, held in Moscow in August 1985.

[2] The error in the energy measurements by the measurement apparatus was determined in independent experiments in which laser pulses were sent to both channels of a two-dimensional analyzer.

[3] The first channel was not used in these experiments, since it received signals from instrumental noise.

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Translated by Dave Parsons