Pulse-energy statistics in the linear regime of stimulated Raman scattering with a broad-band pump

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The results of the investigation of Stokes energy statistics in the linear regime of stimulated Raman scattering with a broad-band pump are presented. The pulse-energy distributions and their standard deviations were measured. The comparison of the experimental data with known ones for coherent pump shows the strong difference in the distribution shape and the standard deviation value. This difference is explained as a manifestation of the non-Gaussian (super-Gaussian) statistics in stimulated scattering with a broad-band pump. The results of numerical calculations for statistics of Stokes intensity are in a qualitative agreement with the experimental findings. [S1050-2947(97)03808-0]

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Stimulated Raman scattering (SRS) is an interesting example of a quantum optical process. Large-scale fluctuations of the Stokes pulse energy in the linear regime of SRS are evidence of a macroscopic manifestation of quantum noise [1–4]. Theoretical and experimental data show that corresponding fluctuations of a spontaneously generated Stokes field can be described by Gaussian statistics, a synonym for chaotic behavior [5]. These results were obtained for a coherent pump, with the pump noise minimized.

Usually the laser radiation is not coherent, and contains additional noise due to multimode operation. In many cases the multimode laser statistics can be characterized as Gaussian [6]. It is known that nonlinear optical effects with a multimode or broad-band pump can transform the statistics from Gaussian to non-Gaussian. This transformation can be seen in second-harmonic generation [7,8] and coherent anti-Stokes Raman scattering [9,10]. By increasing the intensity fluctuations these statistics can be characterized as super-Gaussian (see, for example, Ref. [11]). Non-Gaussian statistics are also seen in spontaneous scattering, where it is a result of scattering of the fluctuating field on the fluctuations of the medium [12].

The investigation of SRS with a broad-band pump has a long history, beginning in the middle of the 1960s. The broad-band nature of the pump laser is responsible for a large forward-backward asymmetry of the Raman amplification [13], the existence of the critical intensity I_{cr} needed to overcome the decorrelating effects of dispersion [14,15], and the increase of the conversion efficiency into the Stokes wave [15]. Effects caused by pump noise in SRS can result in interesting statistical properties of SRS radiation. At the same time, a detailed knowledge of SRS statistics with a coherent pump [5] gives a good background for a comparative analysis of the broad-band case. The main statistical data concerning coherent pump were obtained by measuring the pulse-energy statistics [2–5]. An investigation of the pulse-energy statistics for broad-band SRS was reported in Ref. [16], where attention was mainly paid to the transition from the linear to nonlinear regimes. Details of the statistics in the linear regime of broad-band SRS were not considered.

In this Brief Report we focus on an observation of non-Gaussian statistics in SRS with a broad-band pump. Our conclusions are the result of a comparison of our data with the known examples of processes with Gaussian statistics: Stokes generation with a coherent pump in the linear regime, and dye laser generation near the threshold. The difference found in the pulse-energy distribution shapes for broad-band and coherent pumps demonstrates non-Gaussian behavior in broad-band SRS. Additional comparison of standard deviations of Stokes and dye lasers shows an increase in the fluctuations for Stokes radiation with respect to the Gaussian light of the dye laser, which is evidence of super-Gaussian behavior. Single-shot spectral measurements allow for an estimation the intensity fluctuations time scales participating in broad-band SRS. Finally, a comparison of experimental data for pulse-energy statistics with the results of the numerical calculations for the intensity statistics will be offered.

In the experiment the superluminescent Rh6G dye laser was pumped by a frequency doubled, nanosecond pulsed Nd:YAG (yttrium aluminum garnet) laser. The figure-8-shaped 50-cm cavity dye laser has no spectral selecting elements, and consisted of three highly reflective mirrors and an output mirror with a reflection coefficient of 10%. The cavity dye cell was oriented at the Brewster angle with respect to the generated beam. The longitudinal scheme of a pump provides conditions close to superluminescent lasing with multiple passing of the multimode amplified radiation through the active medium, and allows one to obtain an output laser beam with a divergence of 0.8 mrad. The maximum energy of a 5.5-ns (full width at half maximum, or FWHM) dye laser pulse is 15 mJ. The spectrum of the dye laser is centered at 563 nm with a bandwidth of approximately 290 cm^{-1} (FWHM).

The dye laser beam is directed through a 2-mm aperture and forming optics (lens with focal length of 2.7 m and 2× telescope) into 140-cm-long Raman cell of molecular hydrogen at 50 atm. The dye laser radiation generated the first Stokes component of vibrational SRS in hydrogen. The second Stokes component of SRS and backward SRS were not excited.

The Stokes pulse radiation is centered at 734 nm, and was spectrally selected by a prism. For spectral measurements some parts of Stokes and passed pump pulses were diverted by a mirror to a monochromator. Pulse spectra were mea-

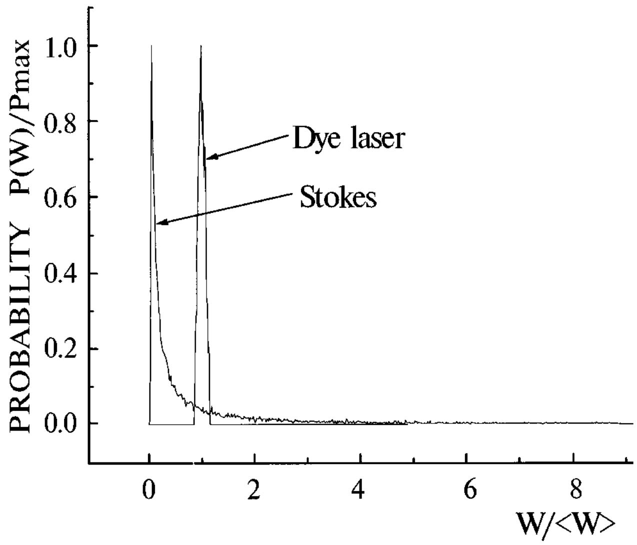

FIG. 1. Stokes and pump laser pulse energy distributions. The vertical scale is normalized to the maxima, and the horizontal scale is normalized to the mean energies.

sured with a CCD array connected to a personal computer.

Photodiodes were used to measure the input pump and Stokes energies. The pump and Stokes signals were digitized by a CAMAC system. The computer performed the data acquisition and analysis. In the analysis the registered energies were considered in the two-dimensional (pump and Stokes) plane [4]. The energy range of every dimension consisted of 512 channels. As a result, the Stokes energy distributions could be analyzed both for very narrow and very broad (as the data sum for some neighboring channels) pump energy intervals. The main parameters of the distribution such as mean value and standard deviation were calculated.

The linear regime of SRS with the efficiency of Stokes generation of $0.9\times10^{-3}\%$ was realized at a mean dye laser pulse energy equal to 3.7 mJ. The Fresnel number for the SRS process $F=S/(z\lambda)$ (where z is the Raman cell length, λ is the Stokes wavelength, and S is the beam cross section) was in the range of 0.3–0.5<1, i.e., one spatial Stokes mode was excited [5]. The steady-state condition for SRS is $t_p > G_0T_2$, where t_p is the pump pulse duration, $G_0=I_pgz$ is the Raman gain, I_p is the pump intensity, g is the steady-state gain coefficient, and T_2 is the dephasing time of the Raman medium (150 ps). For our experiment $t_p\sim G_0T_2$, i.e., SRS was in the intermediate regime. The SRS threshold intensity was $I_{\rm th}=240\ {\rm MW/cm^2}$. The number of the pump modes can be estimated as $N=\Delta\omega/\Delta\nu\sim 30\,000$, where $\Delta\omega$ is the laser spectral bandwidth, $\Delta\nu=1/2L$, and L is the laser resonator length.

The Stokes and laser energy distributions for these conditions are shown in Fig. 1. The pump laser distribution contains 40 000 shots registered in 49 channels. The distribution is centered near the mean energy with a normalized standard deviation of 0.09. The standard deviation was calculated by $\sigma=\sqrt{(1/n)\Sigma_{i=1}^{n}(W_i-\langle W\rangle)^2}$, where n is the number of events, W_i is the energy accumulated in ith histogram column, and $\langle W\rangle$ is the mean energy. It was normalized to mean energy. The Stokes energy distribution contains 16 000 shots corresponding to the sum of 20 central channels of the laser distribution. In a separate consideration we checked the effect of the summation over laser channels on the Stokes distribution. No qualitative difference in the shape of the distribution was found for the pump energy intervals used. The Stokes energy distribution has a maximum strongly shifted from the mean energy to small energy channels. The maximum of the distribution is resolved, and it is slightly shifted from the zero value.

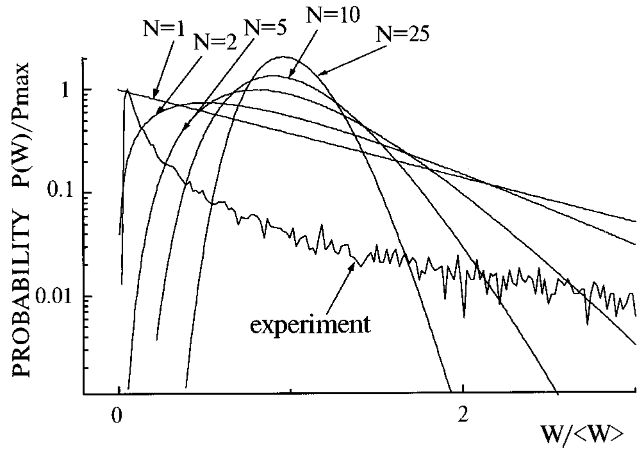

FIG. 2. Comparison of the experimental Stokes pulse energy distribution with the calculated distributions for Gaussian statistics (γ distribution) with increasing the mode number N.

A detailed description of the Stokes pulse energy statistics in the linear regime of SRS with a coherent pump is given by Raymer and Walmsley [5]. They showed that the Stokes field statistics are Gaussian. When the single mode of the Stokes radiation is excited, the shape of the pulse-energy distribution is close to exponential,

$$P(W)=(1/\langle W\rangle)e^{-W/\langle W\rangle}, \tag{1}$$

where P is probability density. If the Stokes mode number N is increased, the pulse-energy distribution corresponds approximately to the γ distribution,

$$P(W)=\frac{1}{(N-1)!}\,\frac{W^{N-1}}{\langle W\rangle^N}e^{-W/\langle W\rangle}, \tag{2}$$

with the maximum near $\langle W\rangle$. For $N\rightarrow\infty$ $P(W)$ converges to a normal distribution, in accordance with the central limit theorem. Here one assumes that the mean energies of the Stokes modes are equal. A more detailed consideration of the pulse-energy statistics, taking into account a quantum description of the Stokes wave propagation, does not give a qualitative difference from this behavior [5]. The transformation of the calculated distribution shape (2) with increasing Stokes modes number is shown in Fig. 2. A comparison of these distributions for the case of a coherent pump and the distribution obtained in our experiment shows a qualitative difference. The experimental distribution corresponds to many modes, and cannot be compared with the normal distribution because it has a long high-energy tail, and the maximum is shifted from the mean to the low-energy channels. The long tail is different from the exponential (see the calculated distribution for $N=1$ in Fig. 2). As a result, we can conclude that the Stokes field statistics corresponding to the pulse-energy distribution measured with a broad-band pump

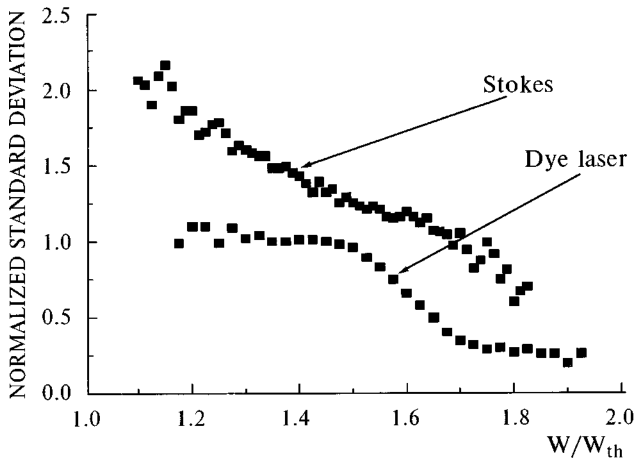

FIG. 3. Normalized standard deviations for the Stokes and dye laser pulse energies vs the energies of pump (dye laser and second-harmonic radiation) W normalized to the threshold values of the pump energy W_{th} for SRS and dye laser generation, correspondingly. The standard deviations are normalized to the mean energies of the Stokes and dye laser radiation.

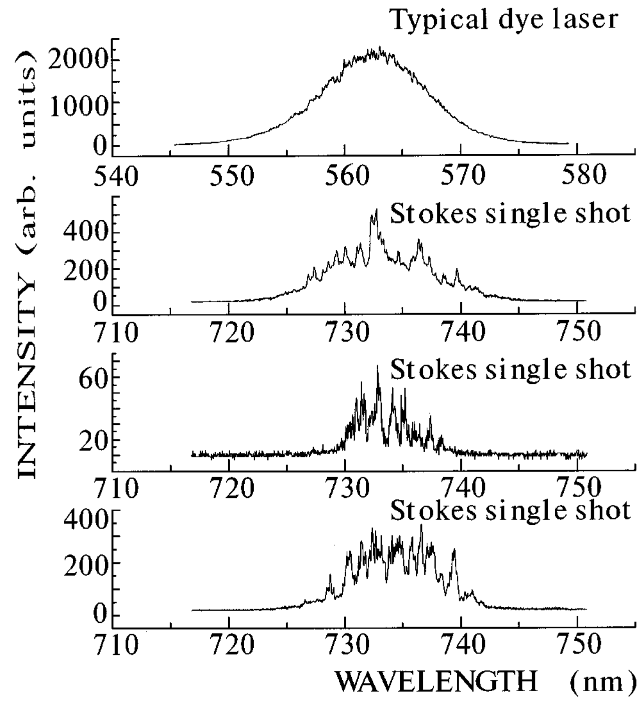

FIG. 4. Single-shot spectra of pump laser and Stokes generations.

are not a Gaussian. We will call the statistics with energy distributions, which are different from that predicted by Eq. (2), non-Gaussian statistics.

To characterize the non-Gaussian behavior as super- or sub-Gaussian (fluctuations are larger or smaller than for Gaussian or thermal field, correspondingly [11]), we compared the standard deviation coefficients for Stokes and dye lasers near threshold (Fig. 3). The data obtained for the linear and nonlinear regimes give fluctuation scales of different natures for both processes. Near threshold the multimode dye laser radiation has a normalized standard deviation up to 100%, which is in agreement with the Gaussian statistics. It should be pointed out that the Stokes pulse energy with a coherent pump has 100% fluctuations as well [2]. The fluctuations of the broad-band SRS radiation are considerably larger, up to 200%, which is evidence of the super-Gaussian or superchaotic behavior in the linear regime. Decreasing fluctuations with increasing energy is connected with the strong pump depletion due to increasing the generated fields [4,5].

In our measurements of the pulse energy, intensity fluctuations are partially averaged out. The spectral data can be used for an estimation of the fluctuation time scales existing under our conditions, and are registered experimentally in correlation measurements at the nonlinear regime [18]. The results of single-shot spectral measurements for pump laser and Stokes radiations are shown in Fig. 4. In the upper part the typical pump laser spectrum is pictured. The pump spectra had good reproducibility, with only minor changes from pulse to pulse. The other three examples represent the typical Stokes spectra. The difference from shot to shot is well defined. The spectrum fluctuates in the FWHM, and the fine structure is also irreproducible. The spectra demonstrate the existence of several fluctuation time scales in Stokes intensity [18]. The whole spectrum band gives the shortest fluctuation time scale, which is estimated as about a hundred femtoseconds. We suppose that these short-time fluctuations were averaged out in our measurements. Fluctuations with time scales of tens and hundreds (close to T_2) of picoseconds, corresponding to the narrow spectral structure in the Stokes spectrum, were measured in the experiment. As a result, the averaging partially smoothed out the fluctuations, but even under these limitations the non-Gaussian character of the Stokes field statistics is evident.

To our knowledge, a theoretical consideration of the Stokes pulse-energy statistics for broad-band pump has not been developed. A description of the broad-band SRS is very complex, and analytical solutions are an exception. For simplicity we consider Stokes intensity statistics with time fluctuations comparable to or longer than T_2.

The Stokes intensity in the linear regime of broad-band SRS for a dispersionless medium can be approximately given as $I_S = I_{SO}\exp(I_P g z)$ [19]. Two terms are responsible for fluctuations, I_{SO} and I_P. I_{SO} introduces the spontaneous fluctuations of the Stokes seed, and I_P describes the pump fluctuations. For the numerical simulation we used the approach developed in Ref. [7]. The probability distribution of the generated Stokes intensity can be calculated as

$$P(I_S) = \int_0^\infty \int_0^\infty P_{SO}(I_{SO}) P_{PO}(I_{PO}) \times \delta(I_S - I_{SO}\exp(gzI_{PO}))\, dI_{SO}\, dI_{PO}, \qquad (3)$$

where the Dirac δ function stands for the condition $I_S = I_{SO}\exp(gzI_{PO})$. A mutual independence of the input signals with the normal probability distributions $P_{SO}(I_{SO})$ and $P_{PO}(I_{PO})$ is assumed. The parameters are Raman gain $G_0 = 10$ and input Stokes $I_{SO} = 10^{-12}\, I_P^{\max}$, and the number of Stokes input modes was equal to 40. The comparison of the calculated and experimental distributions shows a good agreement (see Fig. 5). The maximum of the calculated distribution is resolved, and shifted from the mean value to low Stokes intensities, and the decay has the same nonexponen-

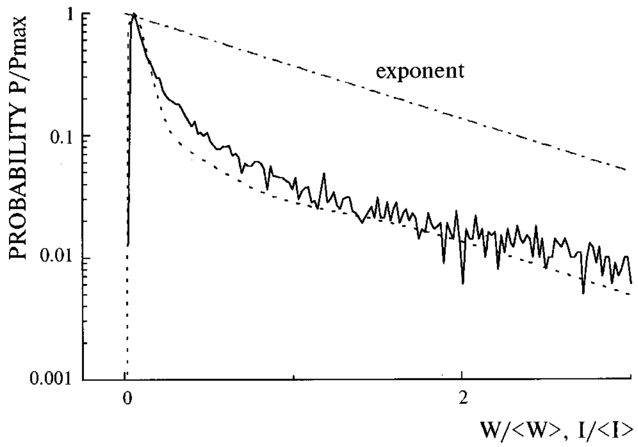

FIG. 5. The comparison of the experimental Stokes pulse-energy distribution $P(W/\langle W\rangle)$ (solid line) and calculated Stokes intensity distribution $P(I/\langle I\rangle)$ (dashed line) in the linear regime of SRS.

tial type. In the case of a stabilized laser with a δ distribution corresponding to coherent pump and normal distributed Stokes input, the output Stokes intensity was exponentially distributed in agreement with the previous results for coherent pump [2–4].

To analyze the reasons for the manifestation of the non-Gaussian statistics, we calculated the output Stokes distribution for the stabilized input Stokes signal. The stabilized Stokes input was described by the δ function. The same type of distribution shape (as in Fig. 5) in the case of the stabilized Stokes input was obtained. It shows that the reason for non-Gaussian statistics can be a nonlinear transformation of the pump noise in Raman amplification. An analogous role can be played by the interaction of two noise sources, Stokes seed and pump, because only the linear combination of the Gaussian processes results in a Gaussian, which is not the case under our conditions. It should be pointed out that an intermode connection in the coherent regime of broad-band SRS can also be responsible for the non-Gaussian statistics [14,15].

The experimental data obtained for the pulse-energy statistics of Stokes radiation in the linear regime of SRS with a broad-band pump are in contradiction with the known data for the distribution shape obtained for a coherent pump. The difference in the distribution shape, and the increase of Stokes fluctuations demonstrated by the standard deviation measurements, give evidence of the non-Gaussian statistics of the Stokes radiation for the case of a broad-band pump, mainly the super-Gaussian one. Numerical calculations of the Stokes intensity distributions are in qualitative agreement with experimental findings for energy statistics. To our knowledge, such observations of the super-Gaussian statistics were unknown for stimulated scattering and can be of importance for applications.

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[1] M. G. Raymer, K. Rzazewski, and J. Mostowski, Opt. Lett. **7**, 71 (1982).
[2] I. A. Walmsley and M. G. Raymer, Phys. Rev. Lett. **50**, 962 (1983).
[3] N. Fabricius, K. Nattermann, and D. von der Linde, Phys. Rev. Lett. **52**, 113 (1984).
[4] P. A. Apanasevich, D. E. Gakhovich, A. S. Grabtchikov, S. Ja. Kilin, V. P. Kozich, B. L. Kontsevoi, and V. A. Orlovich, Kvant. Elektron. **19**, 884 (1992) [Sov. J. Quantum Electron. **22**, 822 (1992)].
[5] M. G. Raymer and I. A. Walmsley, Prog. Opt. **XXVIII**, 183 (1990).
[6] A. V. Masalov, Prog. Opt. **XXII**, 145 (1985).
[7] A. A. Grutter, H. P. Weber, and R. Dandliker, Phys. Rev. **135**, 629 (1969).
[8] Y. Qu and S. Singh, Phys. Rev. A **51**, 2530 (1995).
[9] L. A. Rahn, R. L. Farrow, and R. P. Lucht, Opt. Lett. **9**, 223 (1984).
[10] R. J. Hall, Opt. Commun. **56**, 127 (1985).
[11] S. Ja. Kilin, *Quantum Optics. Fields and Their Detection* (Nauka, Minsk, 1990).
[12] B. Grosignani, P. Di Porto, and M. Bertolotti, *Statistical Properties of Scattered Light* (Academic, New York, 1975).
[13] A. Z. Grasuk, I. G. Zubarev, and N. V. Sujasov, Pis'ma Zh. Eksp. Teor. Fiz. **16**, 237 (1972) [JETP Lett. **16**, 166 (1972)].
[14] I. G. Zubarev, A. B. Mironov, and S. I. Mikhailov, Pis'ma Zh. Eksp. Teor. Fiz. **23**, 697 (1976) [JETP Lett. **23**, 642 (1976)].
[15] S. A. Akhmanov, Yu. E. D'yakov, and L. I. Pavlov, Zh. Eksp. Teor. Fiz. **66**, 520 (1974) [Sov. Phys. JETP **39**, 259 (1974)].
[16] A. S. Grabtchikov, S. Ja. Kilin, V. P. Kozich, and N. M. Iodo, Pis'ma Zh. Eksp. Teor. Fiz. **43**, 118 (1986) [JETP Lett. **43**, 151 (1986)].
[17] M. G. Raymer, Z. W. Li, and I. A. Walmsley, Phys. Rev. Lett. **63**, 1586 (1989).
[18] A. I. Vodchitz, V. P. Kozich, P. A. Apanasevich, and V. A. Orlovich, Opt. Commun. **125**, 243 (1996).
[19] G. P. Djotian, Ju. E. D'yakov, and I. G. Zubarev, Zh. Eksp. Teor. Fiz. **73**, 822 (1977) [Sov. Phys. JETP **73**, 822 (1977)].