Macroscopic Manifestation of Quantum Fluctuations in Transient Stimulated Raman Scattering

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The statistical distribution of the Stokes pulse energy is measured for transient stimulated Raman scattering in pressurized hydrogen gas. Large fluctuations of the Stokes energy obeying an exponential distribution law are observed. These macroscopic energy fluctuations are caused by quantum noise in the initial state of the system.

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When an intense light beam interacts with a Raman-active medium, strong optical amplification at a Stokes shifted frequency may occur. This stimulated Raman process was discovered about two decades ago.[1] To date, a rather complete understanding of the phenomena associated with stimulated Raman scattering (SRS) has been achieved[2,3] and numerous applications exist.

Amplification of Stokes light by the stimulated Raman process can be theoretically described with use of a semiclassical approach[4] which gives very good quantitative agreement with experiments. *Generation* of Stokes light, on the other hand, is not possible from the semiclassical point of view unless a Stokes input wave is supplied. In many typical experimental situations, however, the Raman gain is so large that even without input a spontaneous buildup of strong Stokes waves from "noise" is observed. A proper description of the initiation of the Stokes generation is beyond the scope of the semiclassical theory and requires a more rigorous quantum theoretical treatment.

In a recent detailed theoretical analysis Raymer and Mostowski[5] have described the spontaneous initiation of stimulated Raman scattering in terms of microscopic random fluctuations originating from quantum noise associated with the dynamical variable $Q(t)$ which is active in the Raman process, i.e., a molecular normal-mode coordinate. It has been predicted[6,7] that in a pulsed SRS experiment the Stokes pulses which may be comparable in energy with the fundamental laser pulses should exhibit large *macroscopic* energy fluctuations reminescent of the stochastic nature of their initiation.

This fluctuation aspect of SRS is physically very interesting for at least two reasons. First, from a basic physics point of view SRS represents a situation in which fundamental quantum fluctuations may be observed on a macroscopic energy scale. In fact, the close connection between SRS and two-level superfluorescence has been pointed out by several authors.[8,9] The statistical properties of superfluorescent pulses which are triggered off by quantum fluctuations and grow to macroscopic energies have received considerable attention.[10] Second, the fluctuations of the Stokes pulses are also important in a more practical sense when SRS is used in pulse generation schemes or for the generation of intense coherent material excitations.[2]

In the discussion of stimulated Raman scattering two different time regimes must be distinguished: (i) a *transient situation* where the duration t_p of the exciting laser pulses is much shorter than the dephasing time[3] T_2 which describes the damping of Q, and (ii) a *steady-state situation* where t_p is much longer than T_2.

The distinction between the two cases is also important in the discussion of Stokes pulse fluctuations. The quantum theory of SRS[5] shows that in a transient situation the Stokes field operator is proportional to the initial Heisenberg operator $Q(0)$ describing the state of the system just before the laser pulse is turned on ($t=0$). In this case the Stokes pulse statistics is determined by quantum uncertainties associated with the random Raman polarization of the initial state.[6] In the steady-state case, on the other hand, the Stokes field is proportional to the Langevin force accompanying the dephasing process. As a result the Stokes pulse statistics is then governed by the properties of the Langevin operator, and, for observation times much longer than T_2, the fluctuations of the Stokes energy are considerably smaller than in the transient regime.[7] By a proper choice of t_p/T_2 it is thus possible to distinguish the two cases and to study the statistics of Stokes pulses resulting from either situation.[11]

Here, we wish to report the probability distribution of the energy of the Stokes pulses for $t_p/T_2<0.12$, i.e., in highly transient SRS.[11] We believe that in our experiment we have observed macroscopic quantum fluctuations due to the initial-state uncertainties.

Very recently, the Stokes pulse statistics has been measured[12] for $t_p/T_2 \gtrsim 20$, i.e., much closer to the steady-state conditions,[11] where the probability distribution is expected to be governed by the damping mechanism.

Experimentally we work with the $Q_{01}(1)$ vibrational transition of molecular hydrogen (H_2). The vibrational frequency corresponds to $\bar{\nu}_v = 4155$ cm^{-1}. Measurements are made with gas pressure ranging from 12 to 25 bars. In this pressure regime the Raman linewidth is determined by homogeneous pressure broadening, and the corresponding dephasing time varies from $T_2 = 526$ ps at 12 bars to $T_2 = 253$ ps at 25 bars.[13] It is thus obvious that picosecond light pulses must be used to realize transient conditions for SRS in H_2. High quality and excellent reproducibility of these picosecond pulses is absolutely essential because the Stokes pulse statistics of interest could easily be blurred by fluctuations due to irreproducible excitation pulses.

We use a Nd-doped yttrium-aluminum-garnet laser system consisting of an actively and passively mode-locked oscillator, a pulse switching system for the selection of a single pulse from the mode-locked pulse train, and two laser amplifier stages. Combination of active and passive mode locking is important because of the superior pulse stability that can be achieved with such a scheme. The amplified single picosecond pulses are frequency doubled by use of a partially deuterated potassium dihydrogen phosphate crystal. The temporal shape of the final pump pulses at 532 nm is approximately Gaussian with a full width at half maximum (FWHM) of 30 ps. The spatial beam profile is also Gaussian.

For the generation of Stokes radiation the laser beam is collimated to a diameter of 175 μm (FWHM) and passed through a 16-cm-long H_2 pressure cell. The experimental geometry corresponds to a Fresnel number of about 0.4.

The Stokes pulses are separated from the pump pulses with use of a dispersion prism and suitable optical filters. The Stokes energy is measured by a photomultiplier followed by signal digitizing and processing equipment. The random error of the Stokes signal which results from the photomultiplier statistics and the finite digitizer resolution was measured to be about 1%.

The probability distribution of the Stokes pulse energy is determined as follows. We measure the energy of each incident laser pulse and the corresponding Stokes energy. The individual events are then grouped into classes with less than 2% variation of the laser energy within each class. As an additional criterion we also measured the unsaturated conversion efficiency of a separate second-harmonic generator which represents a sensitive measure of fluctuations of the pulse width and of the spatial properties of the laser beam. Events are rejected if the deviation of the second-harmonic efficiency exceeds 10% of the class average. The probability distribution of the Stokes energy is then evaluated with typically a few thousand acceptable events in each class.

Before considering these distributions let us discuss Fig. 1 which shows the Stokes energy as a function of pump energy for four different gas pressures. Each data point represents the average of the Stokes energies of all events of the respective 2% energy class. The solid curves are calculated from transient SRS theory[5] with use of the known Raman gain coefficient[14] of H_2, $g = 2.1\times10^{-9}$ cm/W. For the calculation of curve a, the absolute Stokes energy and the effective area of the pump beam are treated as adjustable parameters. The Stokes energy scale established by this fit is consistent within an order of magnitude with the estimated energy sensitivity of the Stokes detector, while the effective beam area from the fit agrees within 20% with the measured beam cross section at FWHM of $A = 2.4\times10^{-4}$ cm^2.

With use of the parameters obtained from the fit of curve a the Stokes energy is then calculated for 18, 15, and 12 bars. Figure 1 shows that with the very same parameters an excellent

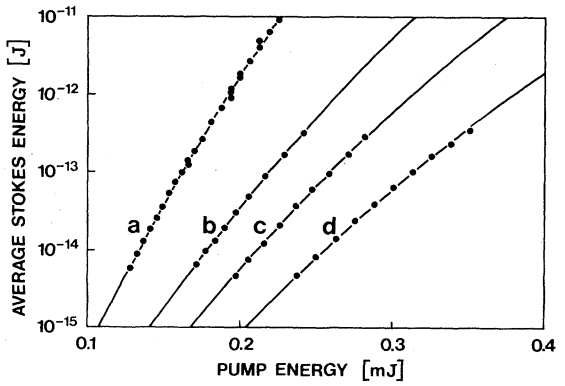

FIG. 1. Average Stokes energy vs laser pump energy. The H_2 gas pressures and the t_p/T_2 values are as follows: curve a, 25 bars, 0.12; curve b, 18 bars, 0.085; curve c, 15 bars, 0.071; curve d, 12 bars, 0.057. The solid curves are calculated with use of transient SRS theory.

agreement with the experimental data is obtained in all cases. Actually, additional experimental data at 25 bars not shown in Fig. 1 cover nine orders of magnitude of the Stokes energy completely consistent with transient SRS theory. We conclude that the transient character of the Stokes process is firmly established, and that transient SRS theory provides a sound basis for our discussion of the probability distribution of the Stokes energy.

Figure 2 depicts, in the form of histograms, four examples out of several dozens of measured Stokes distributions. They correspond to average Stokes energies of 18, 30, 48, and 70 fJ. Note that the Stokes conversion efficiency is only a few times 10^{-10}. Nevertheless, we work in the high-gain limit of SRS well above the spontaneous scattering level with a transient gain coefficient $G_T = 2(2gIlt_p/T_2)^{1/2}$ of 20 to 30 (I is the laser intensity and l is the cell length).

A striking feature of all distributions is the fact that the most probable events correspond to Stokes energies near zero. In fact, the semilogarithmic plot of Fig. 2 shows that the data are very well accounted for by a straight line, indicating an exponential decrease of the probability P with Stokes energy W_s. It follows that the probability can be represented as

$$P(W_s) = \langle W_s \rangle^{-1} \exp(-W_s/\langle W_s \rangle).$$

Strong Stokes energy fluctuations obeying such an exponential probability law[6,9] have been predicted to occur as a result of fundamental quantum fluctuations in a Raman generator operating in the transient regime.

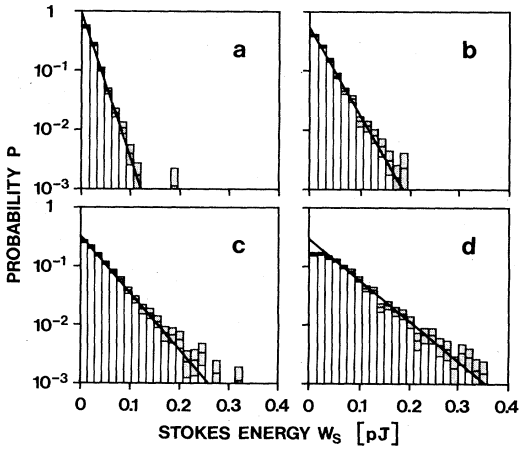

FIG. 2. Examples of Stokes energy distributions for $t_p/T_2 = 0.085$. The shaded areas represent the statistical error (Ref. 15). The laser pump energies are (a) 187 μJ; (b) 196 μJ; (c) 204 μJ; (d) 214 μJ. The total number of events in each distribution is $N = 1800$.

For an exponential probability law the standard deviation is equal to the average value, e.g., $\Delta W_s = [\langle W_s^2 \rangle - \langle W_s \rangle^2]^{1/2} = \langle W_s \rangle$. Experimentally we find that $\Delta W_s/\langle W_s \rangle$ is between 0.9 and 1, indicating a slight deviation from a pure exponential. A careful inspection of the experimental data shows that the measured probability for Stokes energies close to zero is slightly less than what one expects for an exponential distribution. For example, the first column near zero in the histograms of Figs. 2(c) and 2(d) are off by several error bars[15] from the straight line. In fact, it has been pointed out that this type of deviation is in agreement with improved three-dimensional calculations[16] which predict a value of $\Delta W_s/\langle W_s \rangle$ close to 0.9 for a Fresnel number of the order of unity.

In conclusion, we have measured the statistical distribution of the Stokes pulse energies in stimulated Raman scattering under highly transient conditions using picosecond excitation pulses. We find large fluctuations of the Stokes energy corresponding to an exponential distribution with a standard deviation very nearly equal to the average Stokes energy. The analysis shows that these fluctuations represent the macroscopic manifestation of the fundamental quantum noise which is responsible for the initiation of the transient Stokes generation.

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