THE EFFECT OF SPACE AND TIME FACTORS ON THE ENERGY CHARACTERISTICS OF SRS

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The effect of space and time structures of the exciting light beam on the energy characteristics of stimulated Raman scattering (SRS) is investigated theoretically and experimentally. It is shown that, contrary to Bloembergen's analysis[7], variation of the focal length of the lens focusing exciting radiation on the specimen (liquid nitrogen) significantly affects SRS energy characteristics. It is found that if the real exciting pulse length τ_p is replaced by effective time $\tau^*_{1,2}$ the energy characteristics can be described by the same relations that hold for intensity characteristics. The obtained experimental results are in satisfactory agreement with theory.

INTRODUCTION

ALTHOUGH stimulated Raman scattering of light (SRS) was discovered a long time ago[1] and considerable experimental and theoretical work was done in this area, there is still a lack of complete agreement between theory and experiment. Particular difficulties arise from the interpretation of the energy characteristics of SRS. In addition to the complications introduced by related phenomena (stimulated Mandel-shtam-Brillouin scattering (SMBS), self-focusing, etc.) the problem also involves difficulties due to the SRS phenomenon itself. The reason is that almost all the theoretical analyses of SRS have been performed in terms of intensities and only for the case of single-mode operation of the exciting laser source. Furthermore the exciting beam was always considered parallel. However, it is not possible to satisfy these conditions in practice. As a rule the exciting radiation used in the experiments has a complex spectral composition with time-dependent intensity distribution over the beam cross section. Lenses are often used to increase radiation density by focusing the exciting radiation on the specimen.

The effect of spectral composition of the exciting radiation on SRS was the subject of experimental research[2,3] showing that the effect is weak. The theoretical interpretation of the problem was given in[4]. The effect of spatial structure of the exciting radiation on energy characteristics of SRS was discussed in[5]. The theoretical analysis of SRS energy as a function of exciting energy taking the higher Stokes components into account was presented in[6]. However these calculations were performed without considering the geometry and parametric mechanisms that may prove to be significant.

Bloembergen[7] considered the effect of focusing exciting radiation in a qualitative manner and showed that the energy characteristics of SRS do not depend on the focal length of the focusing lens. However a comparison of the results of[8] and[9] indicates that the focal length of the lens can affect SRS characteristics.

In the present work we consider the effect of the geometric factors and the time dependence of the exciting pulse on SRS characteristics.

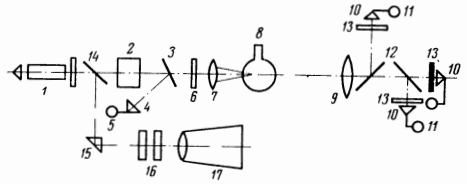

FIG. 1. Experimental setup. 1—laser; 2—attenuator; 3, 12, 14—beam splitters; 4, 5—control calorimeter and galvanometer; 6—calibrated attenuating filter; 7—focusing lens; 8—specimen; 9—collecting lens; 10, 11—measuring calorimeters with corresponding galvanometers; 13—component separation filters; 15—beam turning prism; 16—Fabry-Perot interferometer; 17—UF-85 camera.

EXPERIMENTAL METHODOLOGY

The experimental setup used is shown in Fig. 1.

We used ruby laser 1 Q-switched by a rotating prism. The laser emitted single pulses with a half-altitude length ~ 25 nsec and total energy per pulse up to 1 J, beam diameter ~ 3 mm, and divergence $\Delta\varphi = 4.6 \times 10^{-3}$. To stabilize the operation of the entire setup, the laser operated with constant pumping and ~ 3 min intervals between the pulses. Attenuator 2 in the form of a glass cell with a water solution of $CuSO_4$ was used to vary the intensity of exciting radiation. By varying the concentration of $CuSO_4$ we could vary the intensity of radiation incident on the specimen. A portion of radiation that passed through the attenuator was split off by glass plate 3 and allowed to fall on calibrated calorimeter 4 whose signal was transmitted to an M17 mirror galvanometer 5. The calorimeter sensitivity was 3.5×10^{-3} J per scale division of the galvanometer. To increase the accuracy of measuring the exciting energy under conditions of high incident beam attenuation, attenuating filters 6 whose transmission was known to sufficient accuracy (not worse than 5%) were placed after the beam splitter 3.

The exciting radiation was focused on specimen 8 by lenses 7 having focal lengths of f = 40, 135, 225, and 500 mm. The radiation scattered in specimen 8 was

collected by lens 9 with focal length of 250 mm and aperture 1 : 2, placed two focal lengths from the specimen, and delivered to three calorimeters 10 whose sensitivity together with M17 galvanometers 11 reached 2×10^{-6} J. One calorimeter was situated on the optical axis of the setup and the two others received portions of the radiation split off by plates 12. Filters 13 were placed in front of these calorimeters to separate the necessary radiation components. For spectral composition studies a portion of the exciting radiation was split off by plate 14 and turned by prism 15 in the direction of Fabry-Perot interferometer 16 where it was photographed with a UF-85 camera 17.

For the measurements of time dependence calorimeters 10 and system 16, 17 could be replaced with FÉK-15 coaxial photocells driving I2-7 oscilloscopes (not shown in the drawing). The time resolution was not worse than 3×10^{-9} sec.

Investigation of the spectral composition of the exciting radiation showed that the spectrum as a rule contained one intense and three or four weak modes spaced ~ 0.02 cm^{-1} apart (interferometer thickness of 1 cm).

The SRS investigation was performed with liquid nitrogen. No Mandel'shtam-Brillouin scattering components were detected in the entire range of exciting intensities used in the experiments. The SRS spectrum as a rule contained one spectral component and only in the saturation region we observed another one. (For these measurements one calorimeter 10 was replaced by an interferometer).

The specimen was held in a spherical dewar with an internal diameter of about 8 cm. Liquid nitrogen was selected as specimen because its intense Raman scattering is not accompanied by the undesirable phenomena of self-focusing and stimulated Mandel'shtam-Brillouin scattering[10]. The relatively large value of vibrational frequency (~ 2330 cm^{-1}) facilitates the separation of the necessary harmonics. It is also important that the specimen "does not deteriorate" when exposed to high-power radiation.

We note that all calorimeters and filters were calibrated directly in situ. In the processing of experimental results special attention was paid to the fact that the filters used to separate the investigated frequency components also passed the neighboring components even though the "parasitic" transmission was small. Attention was also paid to all reflections in order to determine energy values directly at the input and output of the material.

EXPERIMENTAL RESULTS

The experiment was performed to study the energies of the first four Stokes components (E_{1S}), the first anti-Stokes component (E_{1as}), and the exciting radiation E_e leaving the specimen as functions of the exciting energy E_0 at the input to the specimen when focused by lenses having various focal lengths. The general nature of these functions is similar to those obtained earlier[8,10]: there are regions of considerable rise of all SRS components and regions of saturation. However the variation of focal length of the lens contributes systematic changes to these functions.

Table I

f, mm	Stokes component threshold, j			
	1 s	2 s	3 s	4 s
40	0.025	0.049	0.100	0.17
135	0.015	0.040	0.06	0.10
225	0.011	0.025	0.05	0.25
500	0.020	0.087	0.27	0.5

Table II

f, mm	q_f from (6)	q_f^E	E_{es}, j
40	0.96	0.74	0.165
135	1	1	0.138
225	1.1	1.5	0.114
500	2.33	0.74	0.120

For Stokes components as f increases we first observe a decrease of the experimental threshold and then as f changes from 225 mm to 500 mm the threshold increases. An exception is the fourth Stokes component whose threshold increases as f changes from 135 mm to 225 mm (see Table I). In practice however thresholds furnish scant physical information about the phenomenon. The slopes of the rectilinear segments (in semilog scale) have a greater physical significance. It is to be expected that we should then observe a picture that is the reverse of the threshold case: as f increases the slopes increase, reach a maximum, and then fall (see Fig. 2). The "saturation" energy of the first Stokes component also rises, but the role of higher order components diminishes. The Stokes components above the first do not show saturation apparently because of the relatively moderate power of our laser.

The saturation energy E_{es} of the exciting radiation that passed through the specimen at first falls and then rises with increasing f. The values of saturation energy for lenses with f = 40, 135, 225, and 500 mm are $E_{es} = 0.165$, 0.138, 0.114, and 0.120 J respectively.

The analysis of energy balance shows that within limits of experimental error the entire converted radiation travels in the direction of propagation of the exciting light (more precisely it forms an angle of the order of 30° determined by lens 9 (see Fig. 1)).

EFFECT OF SPACE AND TIME FACTORS

A. Time characteristics. It is useful to precede the discussion of experimental results by the consideration of the quantities that should be measured in the study of SRS energy characteristics in order to avoid ambiguities upon comparison of the obtained results with theory. This problem arises because of the fact that practically all theoretical papers on SRS deal with intensities while experimentalists measure energies which is much simpler.

In the simplest case the intensity I_{1S} of the first Stokes component of SRS as a function of exciting radiation intensity can be represented in the form (see for example[10,11])

$$I_{1s} = I_{1s}^0 e^{I_0 s l}, \quad (1)$$

where I_{1S}^0 is an analog of spontaneous Raman scatter-

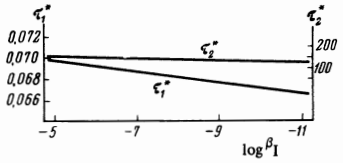

FIG. 2. Effective times $\tau_1^* = a_I/a_E$ and $\tau_2^* \times 5 \times 10^3 = b_E/b_I$ as functions of computed parameter b.

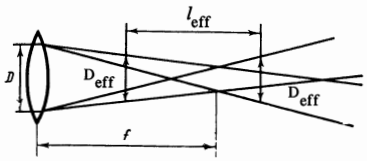

FIG. 3. Experimental geometry analysis.

ing, l is the path of light in the medium, I_0 is the exciting light intensity, and g is gain determined by the properties of the medium (we neglect all anti-Stokes and Stokes components higher than the first order). In this case the straight line slope is gl on a semilog scale. The same formula is also valid for energy when the exciting pulse is rectangular both in time and in cross section provided we replace I_i by $E_i/\tau S$ where E_i are the corresponding energies, τ is the length of the exciting pulse, and S is beam cross section. This substitution is invalid for pulses of other shapes.

A more exact formula that takes into account the transition from spontaneous to stimulated Raman scattering and, in rough approximation, the transition to saturation is given in[12,13].

This formula has the form

$$I_{1s} = \frac{b[\exp\{a(I_0+b)\}-1]}{1+(b/I_0)\exp\{a(I_0+b)\}}. \quad (2)$$

We performed numerical integration of this expression with respect to time in order to clarify the effect of nonlinearity of the SRS process. This was done on the assumption that the intensity of exciting radiation depends on time according to the law $I_0 = I_0^0 \exp\{-t^2/\tau^2\}$ (see[8] for example). The parameters a and b varied within broad limits. In Eq. (2), b characterizes spontaneous Raman scattering and $a = gl$. It was also assumed that the exciting radiation intensity is constant over the beam cross section S.

The results of the computation showed that the general nature of the dependence of SRS energy on the exciting light energy is analogous to the dependence of SRS intensity on the exciting light intensity corresponding to (2)[1)]. It is significant that both curves can be transformed into one another by the introduction of two effective parameters $\tau_{1,2}^*$ that relate quantities a_1 and b_1 of the intensity formula to parameters a_E and b_E respectively of the energy expression. According to Fig. 2 τ_1^* and τ_2^* are practically unchanged within a broad range of the a_1 and b_1 parameters. For example $\tau_1^* = 0.068$ and $\tau_2^* = 0.0288$ (averaged values); the computed value of τ was taken as 0.0312 corresponding to the half-amplitude pulse length of 26 nsec. We see that both τ and τ_2^* coincide within the limits of accuracy of the computation results. Incidentally this can be expected since the b parameter defines stimulated Raman scattering that closely follows the exciting pulse.

Consequently the consideration of the complicated time dependence of the exciting radiation intensity in the transition from intensities to energies reduces to the simple introduction of two effective parameters τ_1^* and τ_2^* that can be readily found for any experimental conditions.

B. Consideration of the real geometry of the experiment. The SRS theory is usually developed on the assumption that the exciting radiation propagates in the medium in the form of a parallel beam. In actual practice this requirement is as a rule not satisfied because the necessary power can be achieved only in a focused beam. Strictly speaking even in an unfocused beam the exciting radiation is always more or less divergent.

Bloembergen[7] found that SRS intensity does not depend on the focal length of the focusing lens. The problem is that SRS intensity is determined by a factor proportional to l/S where l is the effective interaction length and S is beam cross section; in the case of an ideal lens and strictly parallel incident beam this quantity does not depend on f. However this again is valid only for the linear process and furthermore, even if an ideal lens is assumed, the divergence of the exciting beam remains. Because of the practical significance of the effect of real geometry of the experiment on SRS characteristics we attempted to take this effect into account.

We assume that the caustic is represented by a hyperboloid of revolution

$$(z^2+y^2)/B^2 - x^2/A^2 = 1.$$

(The vertex of the hyperbola is in the center of the cell, the x axis coincides with the light propagation axis, $B = f\Delta\varphi$, and $A = f^2\Delta\varphi/2D$. Here f is the focal length of the lens, $\Delta\varphi$ is the divergence of the exciting beam, and D is beam diameter at the lens). In this case if pumping level is small (low SRS conversion coefficients) we obtain gain a as a function of the experimental geometry

$$a \infty \frac{4}{\pi D\Delta\varphi}\operatorname{arctg}\frac{lD}{f^2\Delta\varphi} \quad (3)$$

(here l is the length of the specimen). Hence it follows that for $lD/f^2\Delta\varphi \gg 1$ gain $a \sim 2/D\Delta\varphi$, i.e., gain depends neither on the length of the medium nor on f. In the other limiting case of $lD/f^2\Delta\varphi \ll 1$ we have $a \sim 4l/\pi(f\Delta\varphi)^2$, i.e., $a \sim f^{-2}$ for $l = \text{const}$. In our case $lD/\Delta\varphi \approx 5.2 \times 10^2\ \text{cm}^2$. Therefore for $f < 7$ cm a is practically independent of f and for $f > 50$ cm $a \sim f^{-2}$.

All this however is only valid for a symmetric caustic which as a rule does not occur in practice (see[14] for example). We now try to take this asymmetry into account.

In the geometric optics approximation we obtain (see Fig. 3)

$$l_{\text{eff}}/S_{\text{eff}} = \frac{8}{\pi\Delta\varphi}\,\frac{k}{(k+1)^2}\,\frac{D}{D^2-(f\Delta\varphi)^2}. \quad (4)$$

[1)]We note that after integration over t the curve of E_{1s} dependence on E_0 in semilog scale consists of (in different sectors) not one but two straight lines with slightly differing slopes. In the analysis of results we took a slope that was an arithmetic mean of the two.

Here $(1+k)^2$ is a coefficient relating beam diameter in the lens focus to the diameter of effective cross section S_{eff}, $D_{eff} = (k+1)f\Delta\varphi$, and l_{eff} is the length of the effective interaction region. The nonlinearity of the process is not taken into account in (4) (as in[7]). The process nonlinearity can be taken into account in the most interesting practical case when the SRS conversion coefficient is low. We then obtain for the quantity l_{eff}/S_{eff}

$$\frac{l_{eff}}{S_{eff}} = \frac{8}{\pi\Delta\varphi}\,\frac{D}{D_{eff}}\,\frac{D_{eff}-f\Delta\varphi}{D^2-(f\Delta\varphi)^2}, \tag{5}$$

or, considering as before that $D_{eff} = (k+1)f\Delta\varphi$, we obtain

$$q_l = \frac{l_{eff}}{S_{eff}} = \frac{8}{\pi\Delta\varphi}\,\frac{D}{D^2-(f\Delta\varphi)^2}\,\frac{k}{k+1}. \tag{6}$$

The above analysis leads to the obvious conclusion that the energy characteristics of SRS are significantly affected by the time-dependent pulse shape and by the spatial structure of the exciting beam and that this effect must be taken into account in a comparison of theory with experiment. The next section presents a discussion of this problem and a comparison with experimental results.

DISCUSSION OF RESULTS

A. The first Stokes component. We first consider the effect of geometric factors on gain. Using the values for $D \approx 3$ mm and $\Delta\varphi \approx 4.6\times10^{-3}$ rad given in the preceding section together with (3) and (5) we find the ratios a_{f_i}/a_{f_k} (see Table II). We see that if the dependence of a_f on f qualitatively coincides with the computed value for lenses with focal lengths of 40, 135, and 225 mm, there is a sharp difference between the experimental and computed values for f = 500 mm. This can be attributed to the fact that for the f = 500 mm lens the effective interaction length exceeds the size of the dewar ($l_{eff} > 8$ cm).

If the laser beam diameter at the specimen entry point represents D_{eff}, simple calculation based on (5) and the computer data yields a_f values of 77 and 279 J^{-1} (the experimental values are 154 and 316 J^{-1} respectively) for a_{500} and a_{225} respectively (the subscript indicates focal length of the lens in mm). Considering the approximate nature of the computation the experimental and theoretical results may be said to coincide satisfactorily.

In the numerical computation of a_f we took into account the fact that the formulas for energy relationships contain effective pulse lengths $\tau^*_{1,2}$ that differ from the real pulse length τ_p of the exciting radiation.

Consequently in the SRS energy measurements the transition from intensities to energies is accomplished by a substitution of the quantity a_1 by $a_E = a_1/\tau^*_1$ and caustic asymmetry must be considered if we take experimental geometry into account.

The interpreters of experimental results concerning the energy relationships of the first Stokes component sometimes say that (see[10] for example) the region of the steepest increase of intensity of this component corresponds to generation rather than to amplification of SRS. Feedback necessary for generation can be supplied by "parasitic" reflection and scattering. However it seems to us that under our conditions there is no generation because, first, the obtained results are in good enough agreement with computations based on (2) and, second, there is no back scattering. The problem of SRS generation is in itself of considerable interest although work on this problem is only just beginning[3,11,15-17]. We note that Alekseev and Sobel'man[15] imply that the "forward" and "backward" SRS emission beams are equal if the reflection coefficients of mirrors at the ends of the resonator are equal. The great proponderance of the "forward" radiation in our experiments shows that generation apparently played a negligible role in our case.

B. Higher-order Stokes components, first anti-Stokes component, and saturation of exciting radiation energy that passed through the specimen. The effect of focal length on the energy characteristics of SRS Stokes components of higher orders (second, third, and fourth Stokes components) is more pronounced than in the case of the first component. In a qualitative sense this is understandable since the energy of these components receives contributions from phase matching mechanisms as well as from the consecutive excitation mechanism.

The problem is that the processes involved in the phase matching condition require that the exciting radiation contain a definite set of angles[18]. Consequently the role of these processes increases with decreasing focal length of the focusing lens, i.e., with increasing divergence of the exciting radiation. Apparently this can also explain the dependence of the thresholds of the third and fourth Stokes components on the focal length of the focusing lens.

We note that the gain of the second Stokes component (in terms of the consecutive mechanism of SRS excitation, i.e., when the second component is excited by the first) obtained by applying the method of least squares to the experimental data for the 500 mm lens was 92.2 J^{-1}, fairly close to the gain of the first Stokes component (150 J^{-1}), especially if we consider the frequency dependence of gain. It is not possible at this time to compare this value with theory.

The experimental data allow us to draw some conclusions about the principal mechanism of excitation of the first anti-Stokes component. The excitation of this component is due to two processes:

$$2\mathbf{k}_e = \mathbf{k}_{-1} + \mathbf{k}_{+1}, \tag{7}$$

$$\mathbf{k}_e + \mathbf{k}_{-1} = \mathbf{k}_{-2} + \mathbf{k}_{+1}. \tag{8}$$

In our previous work[8] we gave preference to mechanism (8); in the present experiment however (distinguished from that in[8] by a higher sensitivity of the recording system) we show that the first Stokes and the anti-Stokes components occur "simultaneously" (in any case the first anti-Stokes component appears earlier than the second Stokes component). This indicates that process (7) is the controlling one in the excitation of the first anti-Stokes component.

In conclusion we consider the saturation region of the exciting energy that passed through the specimen. The saturation of exciting energy that passed through the specimen is usually attributed to higher order SRS components (see[5] for example). In fact an elementary

limiting transition in (2) shows that saturation intensity $I_{es} \to 0$ when $I_0 \to \infty$. However in regard to a beam with Gaussian distribution of power along the radius we can show that

$$\frac{2\pi r_0^2 \tau}{a}\left(1+\frac{1}{ab}\right) > E_{es} > \frac{2\pi r_0^2 \tau}{a}. \quad (9)$$

Substituting corresponding values for a, b, and r_0 for τ = 30 nsec we obtain 10^9 J > E_e > 10^{-2} J; our experiment yields $E_{es} > 10^{-1}$ J.

It follows from (9) that the lower boundary for $E_{es} \sim 1/a$. It follows from experimental data that the values of $E_{es\,135}/E_{es\,f}$ are 0.84, 1.2, and 1.15 respectively for f = 40, 225, and 500 mm. Experimental measurements of a_f yield values of these ratios of 0.74, 1.5, and 0.74 respectively. Consequently taking the dependence of exciting radiation intensity on beam radius into account allows us to explain the saturation of exciting radiation energy that passed through the specimen.

In conclusion we thank V. V. Davydov for help with the experiments.

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