

ENERGY AND TIME CHARACTERISTICS OF STIMULATED RAMAN SCATTERING OF LIGHT

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The energy of four Stokes and three anti-Stokes components of stimulated Raman scattering of light (SRS) in liquid nitrogen is measured as a function of the exciting-radiation energy. The intensities of the first anti-Stokes components are studied as functions of the first Stokes component intensities in carbon disulfide and liquid nitrogen. This dependence is exponential. The shape and length of the SRS emission pulses in liquid nitrogen are investigated for the first and second Stokes components and the first anti-Stokes component as functions of exciting radiation energy. A qualitative interpretation of the obtained relationships is given. The role of the various mechanisms in the formation of the Stokes and anti-Stokes SRS components of various orders is discussed.

THE dependence of the stimulated Raman scattering (SRS) line intensity on the pumping conditions (primarily on the exciting radiation power) is of considerable interest if we wish to clarify the mechanism of the phenomenon and its practical applications. However the pertinent experimental data that are available at this time are quite limited and concern mainly the 1st Stokes component^[1-4].

It should be noted that the actual SRS process is very complex since the high power input to a medium gives rise to other optical and mechanical processes (self-focusing, stimulated Mandel'shtam-Brillouin scattering, etc.) in addition to SRS. As a result, simple concepts of the origin of SRS are not always acceptable. A detailed investigation of the phenomenon under various conditions is necessary for the clarification of the actual mechanisms responsible for the excitation of any particular SRS component. It is also important to study the time dependence of the phenomenon. This is of interest from yet another point of view. Since the energy conversion in SRS is a time-dependent function, the power and energy conversion coefficients can be significantly different. We know (see^[5] for example) that the pulse length of the scattered radiation can be two or three times shorter than that of the exciting radiation; i.e., the power conversion coefficients can be two or three times larger than the energy conversion coefficients.

The present paper reports on the results of the study of energy distribution among the SRS components and the time dependence of their intensity under various conditions of excitation.

THE MEASUREMENT METHOD

The SRS spectral characteristics were studied in a setup illustrated in Fig. 1.

The SRS spectrum was excited by a Q-switched ruby laser 1. The Q-switch was a rotating prism. Attenuator 2 consisting of a stack of identical plane-parallel glass plates was placed in the path of the laser beam. In some cases a neutral filter with a transmittance of ~ 0.4 was added to the stack. The transmittance of the stack as a function of the number of glass plates was measured experimentally. A portion of energy ($\sim 10\%$) that passed

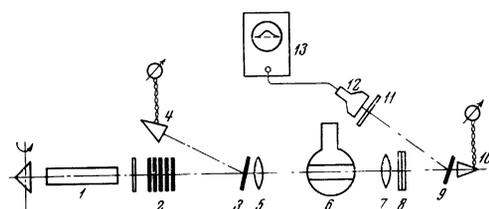


FIG. 1. Experimental setup. 1 – Q-switched laser; 2 – glass plate stack to measure pulse energy; 3 – reflecting glass plate; 4 – control calorimeter; 5 – lens focusing laser emission on specimen; 6 – dewar with liquid nitrogen; 7 – lens focusing SRS emission on measuring calorimeter; 8 – filters separating SRS components; 9 – reflecting glass plate; 10 – measuring calorimeter; 11 – neutral filters for the attenuation of light; 12 – photocell; 13 – I2-7 oscilloscope.

through the attenuator was diverted by glass plate 3 to control calorimeter 4. The latter, together with a mirror galvanometer serving as the measuring instrument, was calibrated against two optical power meters (IMO-1). The sensitivity of the control calorimeter determined in this manner with an accuracy of $\sim 5\%$ reached 10^{-3} J per scale division for the laser output energy. The laser emission was focused by lens 5 with a focal length of $f \approx 150$ mm at the center of scatterer cell 6.

Liquid nitrogen and carbon disulfide in which the SRS is effective enough^[6] were selected as objects of the investigation. The cell for liquid nitrogen was a dewar in the form of a flask with an internal diameter of ~ 8 cm. The dewar was silvered except for input and output radiation windows. Carbon disulfide was studied in a simple cylindrical cell 3 cm long. The scattered radiation was focused by lens 7 with a focal length of $f \approx 150$ mm at recording calorimeter 10 or at the slit of the spectroscope. A suitable filter stack 8 was placed directly behind the second lens in order to separate the required spectral region for calorimetric recording. The recording calorimeter with a mirror galvanometer serving as the meter had a sensitivity of $\sim 1.7 \times 10^{-5}$ J. The accuracy of measurements was $\sim 7\%$. In front of the calorimeter there was a glass plate 9 diverting a small portion of the energy ($\sim 10\%$) to coaxial photocell 12 connected to an I2-7 high-speed oscilloscope 13. Whenever necessary, neutral filters 11 were placed in front of the photocell.

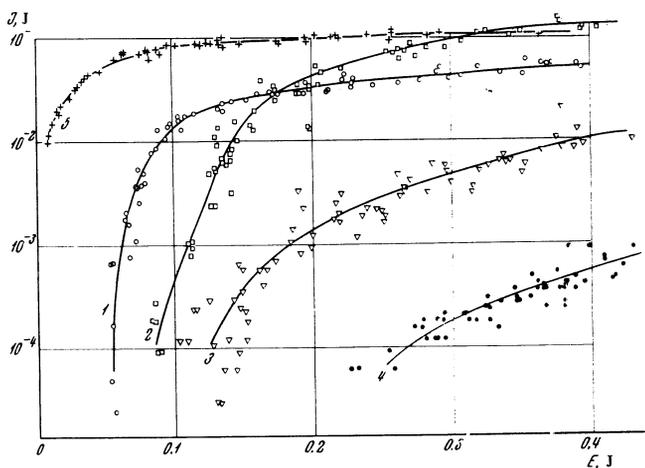


FIG. 2. Energy J or SRS Stokes components in liquid nitrogen as a function of exciting energy E . 1 — first Stokes component; 2 — second; 3 — third; 4 — fourth; 5 — exciting radiation at the output of specimen.

A characteristic curve plotted for the laser emission was used for intensity measurements of the photographs. The characteristic curve was plotted from emulsion density marks obtained with a set of special diaphragms having a number of calibrated holes uniformly distributed over the diaphragm cross sections. Laser light was diffused by a layer of magnesium oxide powder to secure a uniform illumination of the diaphragms. The spectral sensitivity of the setup was determined from density marks obtained with the aid of a calibrated ribbon lamp. We note that the slopes of the characteristic curves of the ribbon lamp and of the laser are significantly different.

MEASUREMENT RESULTS

The results of measurements in liquid nitrogen are shown in Figs. 2 and 3. These measurements cover the spectral region from ~ 4700 to $\sim 19,700 \text{ \AA}$ or, in frequency terms, from $\sim 21,300$ to $\sim 5080 \text{ cm}^{-1}$. Owing to the experimental conditions, of course, this spectral region is covered in comparatively large steps of $\sim 2330 \text{ cm}^{-1}$.

By comparing the data for the individual SRS components we can discern a fairly general pattern of their behavior. The energy J of all the SRS components smoothly

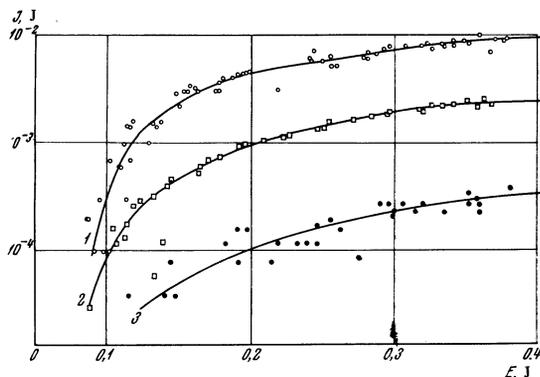


FIG. 3. Energy of SRS anti-Stokes components in liquid nitrogen as a function of exciting radiation energy. 1 — first anti-Stokes component; 2 — second, 3 — third.

Component	Coefficient of transformation	
	in terms of energy, %	in terms of the number of photons, %
1st Stokes	17	20
2nd Stokes	32	47
3rd Stokes	2.5	4.8
4th Stokes	0.14	0.4
1st anti-Stokes	2.5	2.1
2nd anti-Stokes	0.6	0.5
3rd anti-Stokes	0.08	0.05

risers when the exciting energy E is small. As the exciting energy increases the rate of increase of the SRS components drops and a "saturation" effect is observed. We note that with increasing exciting energy the rise of SRS energy is slower for each successive component; i.e., the fastest rise is observed in the case of the first Stokes component and it slows down for the next component. An analogous picture is seen with the anti-Stokes components: the rise slows down with increasing harmonics.

We consider some features of energy distribution among the components.

The first feature is the comparative constancy of the exciting radiation energy that passed through the specimen. This energy varies only by 25% in response to a four-fold change in the energy of the exciting radiation that entered the specimen.

The second feature is the considerable energy of the second Stokes component of the SRS. As the exciting energy is increased the second Stokes component rapidly rises and approaches the energy of the first Stokes component when the exciting energy is only ~ 0.17 – 0.18 J . When the exciting energy is further increased the second Stokes component overtakes the 1st and becomes comparable with the energy of the exciting radiation that passed through the specimen (at the energy of ~ 0.33 – 0.34 J). A still further increase of the exciting energy causes the energy of the second Stokes component to exceed the energy of the exciting line that passed through the specimen. At this point the Stokes energy amounts to $\sim 0.125 \text{ J}$ for an incident exciting energy of 0.4 J . We did not observe any anomalous behavior of other SRS components at the exciting energies used.

The third feature of energy distribution in an SRS spectrum is the considerable energy increase in components of increasing order that accompanies the rise of the exciting energy.

Of interest (see the table) are the maximum conversion coefficients (or maximum efficiencies) obtained for the individual components (except for the first Stokes component, all our results refer to the exciting energy of 0.4 J). We emphasize that the data cited here concern comparatively low exciting energies. A boost in the power of the exciting laser will doubtlessly permit us to increase the conversion coefficients of higher-order SRS components.

The energy relationships obtained in the experiment were analyzed from the viewpoint of the energy balance in the stimulated Raman scattering observed along the direction of propagation of the exciting radiation. The energy balance is satisfied fairly well according to the analysis although there is some difference between the total incident radiation energy and the sum of the meas-

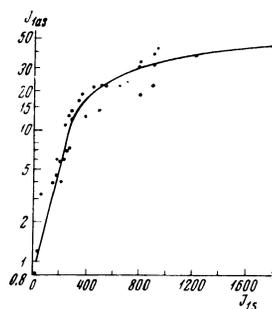


FIG. 4. Intensity of the first anti-Stokes component of SRS as a function of the first Stokes component in carbon disulfide. Intensities are given in arbitrary units.

ured radiation components that emerge from the specimen in the forward direction within the limits of the angle of $20\text{--}25^\circ$. This difference increases with increasing exciting energy reaching approximately 25% for an exciting energy of 0.4 J. However these discrepancies can be considered as within the limits of experimental accuracy if the energy transition into the vibrational energy of molecules is taken into account, i.e., if the quantum balance rather than energy balance is being discussed.

The correlation of intensities of the first Stokes and first anti-Stokes SRS components is of particular interest in the attempt to clarify the basic mechanisms of SRS. This correlation was investigated in detail by photographic methods in carbon disulfide (the 656 cm^{-1} line) and in a somewhat lesser detail in liquid nitrogen. These measurements were performed by recording the scattered radiation within angles of the order of $20\text{--}25^\circ$ so that the angle-integrated intensity was automatically recorded for the case of the anti-Stokes radiation.

The data obtained for carbon disulfide are shown in Fig. 4. We see that the intensity of the first anti-Stokes component is at first an exponential function of the intensity of the first Stokes component. Beginning with some value of intensity this function deviates from the exponential, the growth rate decreases, and a visible tendency toward saturation appears. The departure from the exponential relationship appears at an exciting power $P \sim 6.5\text{ MW}$. The SRS observational threshold corresponded to the power $P \sim 5\text{ MW}$.

Time-dependent characteristics in liquid nitrogen were taken for the exciting radiation in front of and behind the specimen cell and for the frequencies of the 1st

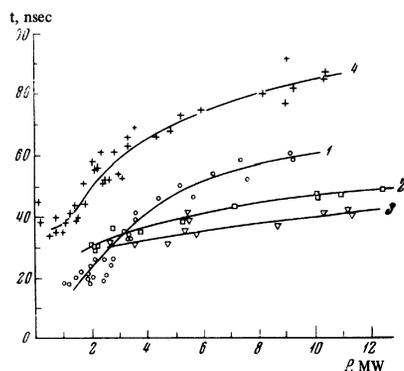


FIG. 5. Pulse length of SRS components in liquid nitrogen as a function of exciting radiation power. 1 — first Stokes component; 2 — second Stokes component; 3 — first anti-Stokes component; 4 — pulse length of exciting radiation that passed through the specimen.

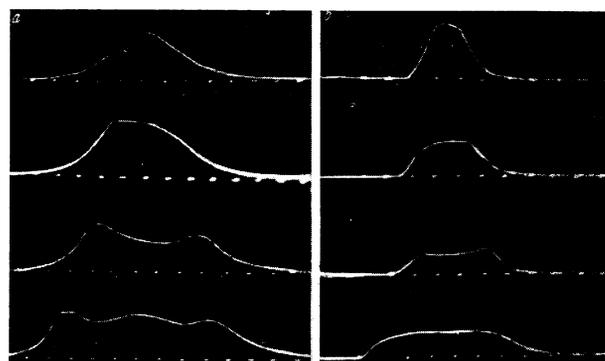


FIG. 6. Variation of radiation pulse shape as a function of exciting energy at the input. a — for the exciting radiation that passed through the specimen; b — for the first Stokes component (energy increases along the top-to-bottom sequence of curves). Time marks every 10 nsec.

and 2nd Stokes and the 1st anti-Stokes components. These measurements were carried out at various energies of the exciting radiation (the term “energy of exciting radiation” denotes energy that entered the specimen cell). The half-height pulse length of the exciting radiation was $\sim 35\text{ nsec}$. The maximum pulse power used in our measurements was 20 MW. The maximum power delivered directly to the specimen was 13 MW. We note that the pulse shape of the exciting radiation was very close to the gaussian.

The measured radiation pulse lengths t are given in Fig. 5. The results show that the pulse of exciting light that passed through the specimen significantly differs from the incident pulse. The pulse length of the exciting radiation that passed through the material varies from ~ 35 to $\sim 85\text{ nsec}$ when the incident pulse power is increased to 12 MW (curve 4). The pulse of the first Stokes component is the most singular. At low exciting power levels it is shorter than the exciting pulse and amounts to $\sim 18\text{ nsec}$. At high power levels this length increases to $\sim 60\text{ nsec}$ (curve 1). The second Stokes and 1st anti-Stokes components reveal no singularities. The pulse length of the second Stokes component varies from ~ 30 to $\sim 50\text{ nsec}$ and that of the 1st anti-Stokes component, from ~ 30 to $\sim 40\text{ nsec}$.

Figure 6 shows the results of investigating the variation in pulse shape. The pulse of the exciting radiation that passed through the specimen changes from the usual dome shape to a two- or even three-peaked curve when the exciting radiation power is increased. The variation in the pulse shape of the first Stokes component is almost identical. No significant change in the pulse shape of the second Stokes and first anti-Stokes components was observed.

DISCUSSION OF RESULTS

In the case of low energy of SRS components the theory of consecutive excitation of Stokes components leads to the conclusion that the energy of the n -th Stokes component is an exponential function of the energy of the $(n-1)$ -st component^[7]. This conclusion agrees with the experimental data for the first and second Stokes components (initial regions of curves 1 and 2 in Fig. 7). We note that the exponent of the second Stokes component is somewhat higher than the corresponding

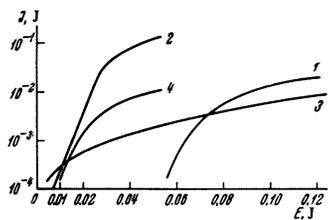


FIG. 7. Energy of the n -th component of SRS as a function of the energy of the $(n-1)$ -st component in liquid nitrogen. Curve 1 – energy of the first Stokes component as a function of the exciting energy; 2 – energy of the second Stokes component as a function of the energy of the first Stokes component; 3 – energy of the third Stokes component as a function of the energy of the second Stokes component; 4 – energy of the first anti-Stokes component as a function of the energy of the first Stokes component.

exponent of the first Stokes component (compare the slopes of curves 1 and 2 in Fig. 7). Therefore we can assume that the second Stokes component contains a contribution from scattered radiation due to other mechanisms.

In the consecutive excitation mechanism the n -th Stokes component can appear only when the energy E_{thresh} of the $(n-1)$ -st component (acting as the exciting line) reaches a noticeable magnitude. The experiment (see Fig. 7) yields the following values for this magnitude (we adopt the value of 10^{-4} J for the threshold sensitivity of the apparatus):

$$E_{\text{пор}} \quad n\text{-й КОМПОНЕНТЫ, Дж:} \quad \begin{array}{ccc} 1 & 2 & 3 \\ \sim 0,056 & \sim 0,006 & \sim 0,0001 \end{array}$$

The systematic depression of E_{thresh} with increasing number of the Stokes harmonic indicates that other mechanisms of SRS excitation participate in the formation of these components.

Thus the mechanism of consecutive excitation seems to play a significant role in the generation of Stokes components but is supplemented by other mechanisms.

We now consider the data obtained for the anti-Stokes components of SRS. A theoretical analysis of the four-photon process satisfying the matching condition

$$2k = k_{-n} + k_n, \quad (1)$$

explains the appearance of anti-Stokes components. Such an analysis yields an exponential dependence of the first anti-Stokes component intensity on the first Stokes intensity^[7]. As we see in Fig. 4 (data for carbon disulfide) and Fig. 7, the experimental data indeed verify the exponential relationship within a broad range of energies. Consequently the main role in the formation of the first anti-Stokes component seems to be assigned to the four-photon SRS process which occurs when there are enough photons with the first Stokes frequency.

The four-photon processes can also be very important in the formation of the second Stokes component of SRS. The most interesting from this point of view are those processes that can be characterized by the following wave vector relations:

$$2k_{-1} = k_{-2} + k, \quad k + k_{-1} = k_{-2} + k_1, \quad (2)$$

where k , k_{-1} , k_{-2} , k_1 are wave vectors of the exciting radiation, the 1st and 2nd Stokes components, and the first anti-Stokes component respectively. It is difficult to estimate at this time the relative contribution from

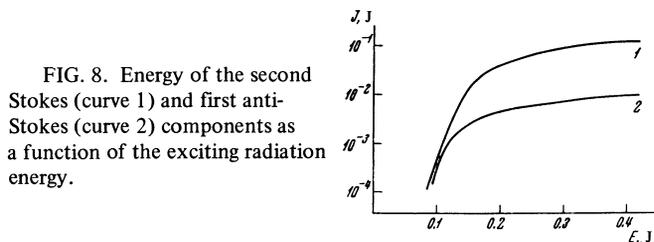


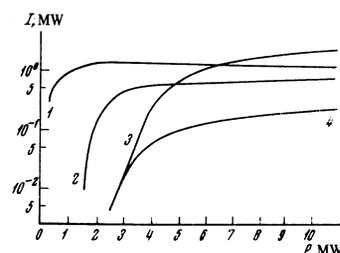
FIG. 8. Energy of the second Stokes (curve 1) and first anti-Stokes (curve 2) components as a function of the exciting radiation energy.

these processes and from the stepwise excitation to the second Stokes component. It is of considerable interest however to compare the dependence of the first anti-Stokes component intensity and the second Stokes intensity on the energy of the first Stokes component (Fig. 7) and of the exciting radiation (Fig. 8). These relationships are similar in the initial region near the experimental threshold. An analogous situation is observed in the corresponding power characteristics (Fig. 9). These results indicate that the mechanisms responsible for the initial region intensities of the 2nd Stokes and 1st anti-Stokes components are similar (possibly representing type two in (2)).

In analyzing the results obtained for the energy yield of the individual components of SRS our data were compared with those of Ragul'skiĭ and Faizullov^[8]. They measured the SRS component energies in liquid nitrogen as a function of the exciting radiation energy. The coefficients of transformation of laser energy into the SRS components given in^[8] are markedly lower than our own. Apparently this is due to the different geometries of the experiments. Short-focus lenses used in^[8] could have reduced the working volume. The sensitivity of our apparatus was 10–15 times higher than that of^[8] permitting us to observe the SRS phenomenon in greater detail with comparatively low level of exciting radiation.

The variation in shape and length of the exciting radiation pulse can be understood by considering that the energy transfer into the first Stokes component is most effective at time instants corresponding to the maximum power of the exciting light. As a result the pulse peak disappears and the pulse assumes a trapezoidal shape. The greater the exciting energy the greater this change. The appearance of a maximum in the center of the pulse at high exciting radiation energies may be due to the reduction of the 1st Stokes component energy because of conversion into the second Stokes component. The behavior of the first Stokes component pulse is due to the same causes operating in the case of the exciting pulse that passed through the specimen. When the exciting light energy is low only the peak of the pulse is effective and the resulting SRS pulse is short. As the exciting energy increases, an ever larger portion of the exciting pulse becomes effective and the SRS pulse

FIG. 9. Emission power I of SRS components and exciting radiation that passed through the specimen as a function of power P of exciting radiation. 1 – exciting radiation that passed through the specimen; 2 – first Stokes component; 3 – second Stokes component; 4 – first anti-Stokes component.



widens. The next process then coming into play is the energy transfer from the first Stokes component to the second Stokes component. A plateau and then a dip form in the center of the pulse of the first Stokes component. A change in pulse shape of the exciting radiation as it propagated within the specimen may also play a significant role.

No significant narrowing was observed in the second Stokes and first anti-Stokes components. This can be explained by the fact that the formation of these components at low exciting light energies is chiefly due to four-photon interaction effective over the entire pulse length of the corresponding exciting radiation. Pulse broadening with pumping is due to the broadening of the exciting pulses. No change in pulse shape of these components is observed because of the negligible energy transfer into the successive components.

In view of the obtained data it is of interest to study the variation of the emission pulse shape in greater detail particularly in those cases when the pulse shape is trapezoidal. We assume that the exciting radiation pulse incident on the specimen is described by a time-dependent function $f(t)$. We do not specify this function although it is known in this case since the pulse has a Gaussian shape. We merely consider that the exciting pulse is transformed only into the first Stokes component and there is no reverse energy transfer. The model under consideration is close to that described in^[2].

The equations determining the pulse shape of the exciting radiation and that of the 1st Stokes component have the form¹⁾

$$\frac{1}{c} \frac{\partial n_{-1}}{\partial t} + \frac{\partial n_{-1}}{\partial x} = (B + An_{-1})n, \quad (3)$$

$$\frac{1}{c} \frac{\partial n}{\partial t} + \frac{\partial n}{\partial x} = -(B + An_{-1})n. \quad (4)$$

Here n_{-1} is the number of quanta per cm^3 at the frequency of the first Stokes component, n is the number of quanta of the exciting light per cm^3 , A characterizes the SRS process, and B characterizes the spontaneous Raman scattering. These equations should satisfy the initial conditions at the input boundary of the specimen

$$n(0, t) = f(t), \quad n_{-1}(0, t) = 0. \quad (5)$$

Integration of (3) and (4) yields an expression for the pulse shape of the exciting radiation at the output from the specimen (the pulse of the first Stokes component is

not considered in this case):

$$n(t) = f(t) \left\{ \frac{A}{B} f(t) + 1 \right\} \left\{ \frac{A}{B} f(t) + e^{[B+An_{-1}]l} \right\}, \quad (6)$$

where l is the thickness of the specimen plate. The above equation readily shows that the exciting radiation pulse at the specimen output has a power minimum ($n(t)$ has a minimum) at time instants corresponding to high pump powers ($f(t)$ has the largest value).

A dip forms in the central portion of the bell-shaped pulse of the exciting input radiation and the pulse thus assumes a double-peaked shape.

The results obtained in the investigation of time characteristics made it possible to compare time-resolved emission power characteristics of three SRS components and the exciting radiation passed through the specimen cell with their energy characteristics. The factual data concerning these characteristics are given in Fig. 9. The general character of emission power variation as a function of the pump power coincides with the behavior of the energy characteristics. The saturation however is much more pronounced in the power than in the energy characteristics.

The figures characterizing the coefficient of transformation of the exciting power into the corresponding SRS component are of interest. The maximum values of the transformation coefficients are: 1st Stokes component—15%; second Stokes component—24%; first anti-Stokes component—2%.

In conclusion we note that when the exciting power is high the power transformation coefficients are markedly lower than the energy transformation coefficients.

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¹⁾An analogous problem was solved in [9] but without taking spontaneous Raman scattering into account.