

ENERGY AND TIME CHARACTERISTICS OF STIMULATED RAMAN SCATTERING OF LIGHT IN A DISPERSE MEDIUM AT VARIOUS TEMPERATURES

G. V. PEREGUDOV, E. N. RAGOZIN and V. A. CHIRKOV

P. N. Lebedev Physics Institute

Submitted March 3, 1972

Zh. Eksp. Teor. Fiz. **63**, 421–430 (August, 1972)

The effect of temperature on the energy and time characteristics of stimulated Raman scattering (SRS) of the 1593 and 977 cm^{-1} lines in stilbene powder is experimentally studied. The measurements were performed over a broad range of SRS intensities: in the range of spontaneous Raman scattering and amplification, in the generation range and in the saturation range. Data on the effect of temperature on the generation thresholds are obtained. The diffusion approximation is used to describe SRS in disperse media. The dependence of the power, line width, and pulse duration of the SRS radiation on exciting light intensity is calculated.

STIMULATED Raman scattering (SRS) in powders^[1] is of scientific and practical significance. Interest in SRS in powders is due to a number of causes: the possibility of investigating the SRS and the phenomena accompanying it in solids without resorting to laborious growing of single crystals^[2-5], and the relatively simple procedure for obtaining radiation power up to several megawatts in the visible and infrared regions, and the possibility of obtaining the required wavelength by choosing a powder material with appropriate vibrational frequency. Such a light source can be used in various problems of nonlinear optics or in other applications, e.g., high-speed infrared photography at the required wavelength. On the other hand, SRS in powders is of interest as a branch of the spectroscopy of light-scattering substances^[6].

Processes that occur in SRS of light in powders can be treated in analogy with the processes that take place in a laser using a dispersed medium^[7,8], in which the scattering by the faces of the crystallites plays the role of a nonresonant feedback. The mean free path of the photon in the substance is in this case much smaller than the working region. Such lasing was predicted in^[7] and experimentally observed with SRS in^[9]. The development of the SRS in a powder, as in a homogeneous medium, is connected with the gain at the Stokes frequency. This gain, induced by the exciting light propagating in the medium, is proportional to the ratio $\sigma/\delta\nu_S$ of the cross section of the spontaneous Raman scattering of the medium to the width of the spontaneous line; both quantities in the ratio are affected by temperature. Thus, by varying the temperature of the medium it is possible to change the gain of the Stokes radiation of the SRS, and consequently its temporal and energy parameters (pulse duration, generation threshold, saturation level, etc.). In addition, the temperature dependence of the parameters of the SRS radiation in powders can yield additional information on the connection between SRS and spontaneous Raman scattering.

We have investigated the power and duration of SRS radiation in stilbene powder as functions of the exciting radiation in a wide range of intensities at temperatures from 100 to 293° K.

1. The experimental setup is shown in Fig. 1. The Raman scattering was excited by a pulse of a ruby laser with passive Q switching (1). The laser pulse duration was $\tau_L = 20$ nsec and the width of the spectral line was $\delta\nu_L = 1.5 \times 10^{-2} \text{ cm}^{-1}$. To increase the flux density of the exciting radiation, we used a converging lens ($f = 18.5$ cm), and the power of the exciting radiation was varied with an aqueous solution of CuSO_4 (2). The front surface of the cell 4 with the investigated powder was located 20 mm away from the focus of the lens. The diameter of the illuminated spot on the surface of the cell was 3 mm. We used stilbene powder with crystallite dimensions 0.25–0.50 mm. The cell with the powder was placed in a Dewar in which liquid nitrogen was poured for the low-temperature experiments.

The scattered radiation was gathered from the same powder surface on which the exciting light was incident. The scattered light passed through a set of neutral attenuating light filters (SNF), and was incident on the entrance slit of a DFS-12 double monochromator. The spectral width of the entrance and exit slits of the monochromator was 40 cm^{-1} . The receiver of the Stokes radiation was an ELU-FT-09 photomultiplier. The elec-

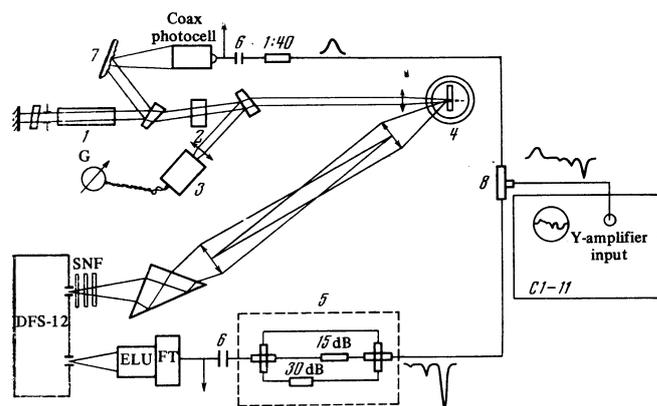


FIG. 1. Diagram of experimental setup: 1—laser, 2—cell with CuSO_4 solution in water, 3—thermoelectric calorimeter, G—control galvanometer, 4—cell with investigated powder, placed in a Dewar, 5—attenuation for extending the dynamic range of the oscilloscope, 6—voltage-dividing capacitors, 7—scattering surface, 8—splitter.

tric signal from the photomultiplier was fed to the vertical-deflection amplifier of an S1-11 oscilloscope; the oscillogram was photographed. Each oscillogram displayed, in addition to the SRS pulse, also the exciting-radiation pulse arriving from a coaxial photocell with a time delay. The laser pulse duration was determined from the oscillogram and its energy was registered with a thermocouple calorimeter 3; the accuracy with which the laser power was measured was 10%.

Figure 2 shows a plot of the power and duration of the SRS pulses of the $\Delta\nu = 1593 \text{ cm}^{-1}$ line on the pump power. Curves 1 on Figs. 2a and 2b correspond to 120° K , and curves 2 to 290° K . We see that the curves have similar shapes at the two temperatures. Four regions can be separated: spontaneous scattering, amplification, generation, and saturation. At exciting-radiation powers $P_L = 0.5\text{--}1.0 \text{ MW}$, the Raman scattering has a spontaneous character. The dependence of the Stokes-radiation power P_S on P_L in this region is linear (Fig. 3), and there is no noticeable decrease of the SRS pulse duration τ_S (Fig. 2b), while the Stokes-radiation intensities are the same for both temperatures, in agreement with the fact that the integral intensity of the $\Delta\nu = 1593 \text{ cm}^{-1}$ line of stilbene does not vary in the temperature region $120\text{--}293^\circ \text{ K}$. When P_L exceeds 1 MW , the dependence of P_S on P_L becomes nonlinear and further on, at $P_L = P_L^t = 4.8 \text{ MW}$ for 293° K and $P_L^t = 2.4 \text{ MW}$ for 120° K , there is a region of sharp increase of P_S (by five orders of magnitude), which we identify with the threshold of the SRS generation. The generation threshold corresponds to a sharp decrease in the duration of the SRS pulse. Lowering the temperature of the powder leads to a decrease of the threshold power P_L^t of the exciting radiation. Figure 2c shows the experimental temperature dependence of P_L^t for the $\Delta\nu = 1593 \text{ cm}^{-1}$ line (solid curve) and Fig. 4 shows the same for the lines $\Delta\nu = 1192$ and 997 cm^{-1} . At powers $P_L > P_L^t$, the growth of P_S slows down and saturation sets in. The plot of P_S against P_L for the $\Delta\nu = 997 \text{ cm}^{-1}$ line is similar (Fig. 5).

2. When the photon scattering mean free path is much less than the dimensions of the region filled with the radiation, the propagation of light in a dispersive medium can be considered by the methods of diffusion theory^[7,8,10]. The radiation is then described by a

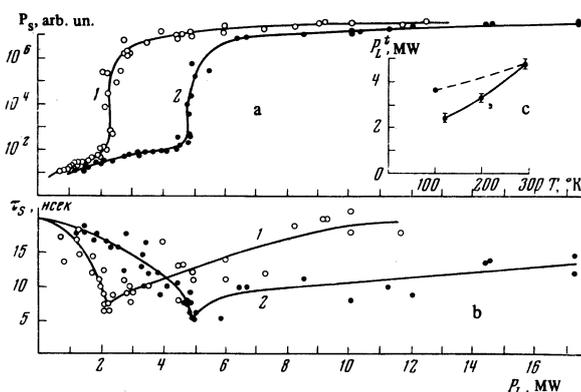


FIG. 2. Power P_S (a) and duration τ_S (b) of the SRS of the $\Delta\nu = 1593 \text{ cm}^{-1}$ stilbene line vs. the exciting radiation power P_L ; c - threshold power P_L^t vs. temperature. Curves: 1 - $T = 120^\circ \text{ K}$, 2 - $T = 290^\circ \text{ K}$, dashed - theoretical plot.

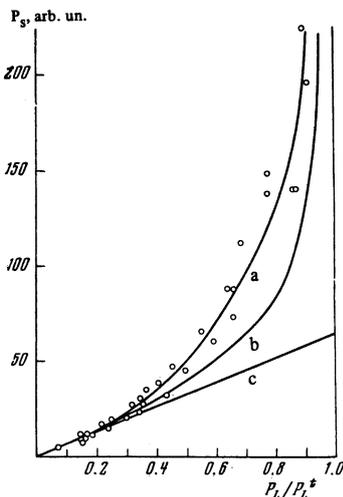


FIG. 3

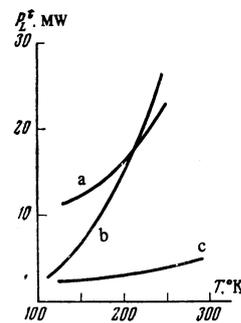


FIG. 4

FIG. 3. Raman-scattering power vs. exciting-radiation power below the lasing threshold (a), theoretical plot (b), and tangent to the curves a and b (c); the latter indicates the spontaneous Raman scattering power.

FIG. 4. Temperature dependence of P_L^t : $\Delta\nu = 1593 \text{ cm}^{-1}$, b - $\Delta\nu = 997 \text{ cm}^{-1}$, c - $\Delta\nu = 1192 \text{ cm}^{-1}$.

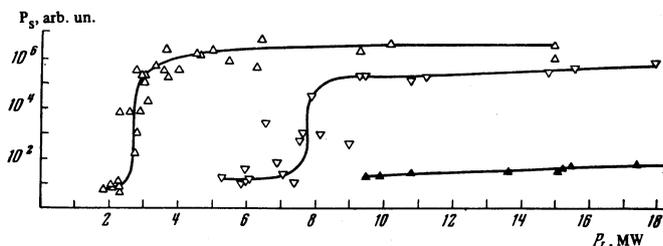


FIG. 5. SRS power of the $\Delta\nu = 997 \text{ cm}^{-1}$ line vs. the pump power at different temperatures: Δ - 115° , ∇ - 150° , \blacktriangle - 293° K .

scalar flux Φ , i.e., by the product of the density of the quanta by their average velocity. In the case of SRS in powder, the joint propagation of the exciting radiation and of the first Stokes component is described by two coupled diffusion equations:

$$\frac{1}{v_L} \frac{\partial \Phi_L}{\partial t} = D_L \Delta \Phi_L - K_L^a \Phi_L - \int K_s^g \Phi_s^g dv, \quad (1)$$

$$\frac{1}{v_s} \frac{\partial \Phi_s^g}{\partial t} = D_s \Delta \Phi_s^g - (K_s^a - K_s^g) \Phi_s^g + f, \quad (2)$$

where t is the time, ν (in cm^{-1}) is the wave number of the radiation at the Stokes frequency, Φ_L (in $\text{cm}^{-2} \text{ sec}^{-1}$) is the exciting-radiation flux integrated over the spectrum, Φ_s^g (in $\text{cm}^{-1} \text{ sec}^{-1}$) is the spectral component of the Stokes radiation flux, $v_{L,S}$ is the average velocity of the photons in the powder, $D_{L,S}$ is the diffusion coefficient of the radiation (the subscripts L and S pertain respectively to the exciting and Stokes radiations), Δ is the Laplace operator (∇^2), $K_{L,S}^a$ (in cm^{-1}) is the coefficient of light absorption in the powder, K_S^g (in cm^{-1}) is the gain of the Stokes radiation in the powder, and f is a function describing the spontaneous Raman scattering. Equations (1) and (2) must be supplemented with equations that establish the connection between K_S^g , f , and Φ_L .

In the derivation of the system (1) and (2) we used the assumption that the spectral width of the exciting radiation is much less than the width of the SRS line ($\delta\nu_L \ll \delta\nu_{\text{SRS}}$), and also the fact that the relaxation time of the molecular oscillations, τ_R , a measure of which is the reciprocal width of the spontaneous Raman scattering spectral line ($\tau_R \approx (2\pi c\delta\nu_S)^{-1} \approx 10^{-12}$ sec) is much shorter than the duration of the pulses of the exciting and Stokes radiations ($\tau_L = 2 \times 10^{-8}$ sec, $\tau_S = 10^{-8} - 10^{-9}$ sec) and the photon diffusion time τ_D ($\tau_D = (v_{L,S} K_{L,S}^a)^{-1} \approx 10^{-10}$ sec).

If $\tau_{L,S} \gg \tau_D$, then we can put $\partial\Phi/\partial t = 0$ and solve the stationary problem. We consider the one-dimensional problem for the case when the exciting light is incident from the left on a half-space $x > 0$ filled with powder, and the conversion of the exciting radiation into SRS is small, i.e., $\int \Phi_S^0 d\nu \ll \Phi_L$. We assume also that the absorption and diffusion coefficients are the same at the frequencies of the exciting and Stokes radiations: $K_L^a = K_S^a = K_a$; $D_L = D_S = D$. Then Eqs. (1) and (2) take the form

$$Dd^2\Phi_L/dx^2 - K_a\Phi_L = 0, \quad (1')$$

$$Dd^2\Phi_S^*/dx^2 - (K_a - K_s^s)\Phi_S^* + f = 0, \quad (2')$$

where $K_S^g = \Phi_L \rho g$, $f = \Phi_L \sigma^\nu N \rho$, $g = I(\nu) N \sigma / 4\pi^2 c \nu_S^2 n^2 \delta\nu_S$ (cf. [11]), $I(\nu) = (\delta\nu_S/2)^2 / [(\nu - \nu_S)^2 + (\delta\nu_S/2)^2]$ is the contour of the spontaneous Raman scattering spectral line, has a dispersion shape, and is normalized to unity intensity at the maximum of the line; $\delta\nu_S$ (cm^{-1}) is the width of the spontaneous scattering spectral line; ν_S (cm^{-1}) is the wave number of the center of the spontaneous Raman scattering line; ρ is the packing coefficient of the powder in the volume; N (cm^{-3}) is the density of the scattering molecules; σ (cm^2) is the spontaneous Raman scattering cross section in 4π sr per molecule of the medium, integrated over the frequencies of the line, for unpolarized exciting radiation; c is the speed of light; n is the refractive index of the powder material, $\sigma^\nu = 2\sigma I(\nu)(\pi\delta\nu_S)^{-1}$ is the cross section for spontaneous Raman scattering in a unit spectral interval.

It is assumed that the flux to the powder material is $\rho\Phi$ and the flux to the air gaps between the crystallites is $(1 - \rho)\Phi$, which is true in the case of not too large a refractive index of the powder material.

Equation (1') has the following solution that decreases as $x \rightarrow +\infty$:

$$\Phi_L(x) = \Phi_L(0) \exp(-x \sqrt{K_a D^{-1}}), \quad (3)$$

$\Phi_L(0)$ is obtained from the illumination condition on the powder boundary [10]:

$$\begin{aligned} \frac{1}{4}\Phi_L(0) - \frac{1}{2}Dd\Phi_L/dx|_{x=0} &= E_L, \\ \frac{1}{4}\Phi_L(0) + \frac{1}{2}Dd\Phi_L/dx|_{x=0} &= RE_L; \end{aligned} \quad (4)$$

E_L ($\text{cm}^{-2}\text{sec}^{-1}$) is the number of exciting-radiation quanta incident on the powder surface $x = 0$ and R is the coefficient of diffuse reflection from the half-space filled with the powder. Solving the system (12), we obtain $\Phi_L(0) = 2(R+1)E_L$. If we put $\sqrt{K_a D^{-1}} = L$ and substitute (3) in one of the equations (4), we can obtain an expression for the coefficient R of reflection from the half-space in terms of L and D :

$$R = (1 - 2DL) / (1 + 2DL). \quad (4')$$

Substituting (3) in (2'), we obtain an equation for the flux of the quanta at the Stokes frequency:

$$Dd^2\Phi_S^*/dx^2 - [K_a - K_s^s e^{-xL}] \Phi_S^* + 2(R+1)E_L \sigma^\nu N \rho e^{-xL} = 0, \quad (5)$$

where

$$K_s^s \equiv K_s^s(x)|_{x=0} = 2(R+1)E_L g \rho. \quad (5')$$

We divide Eq. (5) by D , make a change of variable $u = 2L^{-1}\sqrt{K_g D^{-1}}e^{-xL}/2 = 2\sqrt{K_g K_a^{-1}}e^{-xL}/2$, and introduce the symbol $B = \sigma^\nu N g^{-1}$. For a dispersion contour of the spontaneous Raman scattering spectral line we have $B = 8\pi\nu_S^2 n^2 c$. We then obtain an inhomogeneous Bessel equation of second order for Φ_S^0 as a function of u :

$$u^2 d^2\Phi_S^0/du^2 + ud\Phi_S^0/du + (u^2 - 4)\Phi_S^0 + Bu^2 = 0, \quad (6)$$

which has a solution in the form (cf. [12])

$$\Phi_S^0(u) = c_1 J_2(u) + c_2 Y_2(u) - B(1 + 4u^{-2}), \quad (7)$$

where $J_2(j)$ and $Y_2(u)$ are Bessel functions of second order of first and second kind, respectively.

From the boundary conditions $\Phi_S^0(x = \infty; x = 0) = 0$ we obtain first $c_2 = -\pi B$, and then

$$c_1 = \frac{\pi B Y_2(\eta) + B(1 + K_a K_s^{-1})}{J_2(\eta)}, \quad \eta = 2\sqrt{K_g K_a^{-1}}.$$

Using the solution (7), we can find the dependence of the intensities of the spontaneous and stimulated Raman scattering on the intensity of the exciting light, the line width, and the durations of the SRS pulses.

We consider two cases.

1. The intensity of the exciting radiation, and consequently the gain at the Stokes frequency, is small: $K_g K_a^{-1} \rightarrow 0$. The function Φ_S^0 tends then to zero like $\frac{1}{2} K_g K_a^{-1} B L x e^{-xL}$. Substituting here the expression for B and K_g and integrating with respect to the frequency of the spontaneous Raman scattering spectral line, we obtain an expression for the total scalar flux of the spontaneous Raman scattering in the powder:

$$\Phi_s(x) = \sigma N \rho E_L (R+1) L K_a^{-1} x e^{-xL}, \quad \Phi_s = \int \Phi_s^0 d\nu \quad (8)$$

(cf. curve 2 of Fig. 6). The radiation from a single surface of the powder $E_S(0)$ can be obtained by using the formula from [10]

$$E_s(0) = D |\text{grad } \Phi_s(x)|_{x=0} = D |d\Phi_s(x)/dx|_{x=0}. \quad (9)$$

Putting $DK_a^{-1} = L^{-2}$, we get

$$E_s(0) = (R+1)L^{-1}\sigma N \rho E_L. \quad (10)$$

Let us compare the intensity of the SRS of the powder and of a single crystal of the same substance. We can show, using formula (10), that at identical illumination with the exciting light, E_L , the intensity of the spontaneous Raman scattering from a unit surface of a powder having specified parameters R , L , and ρ is approximately the same as the intensity of the spontaneous Raman scattering from a unit area of the illuminated face of a single crystal of thickness $l_c = 2(R+L)\rho L^{-1}$. Since the largest contribution to the spontaneous Raman scattering in the powder is made by a layer of thickness $l_p \approx 2L^{-1}$, to obtain the same spontaneous Raman scat-

tering radiation intensity from a single crystal and from a layer of powder it is necessary to have approximately the same amount of matter.

2. When $2\sqrt{K_g K_a^{-1}} \equiv \eta$ becomes equal to the first root of the Bessel function $J_2(\eta)$, the denominator in the expression for C_1 in formula (7) vanishes, and the flux Φ_S^v becomes infinite. This occurs at $\eta \approx 5.14^{[13]}$, and then

$$K_g = K_g^t \approx 6.6 K_a. \quad (11)$$

If $c_1 \gg c_2$ in (7) (this takes place when $6.6 > K_g K_a^{-1} > 5$), then formula (7) can be replaced, using the data of the tables in $^{[13]}$, by

$$\Phi_S^v(x) = \frac{0.68BJ_2[\eta(v)e^{-xL/2}]}{5.14 - \eta(v)}, \quad (12)$$

$$\eta(v) = 2 \left(\frac{K_g(v)}{K_a} \right)^{1/2}$$

(curve 3 of Fig. 6), from which we readily obtain an expression for the spectral component of the flux density $E_S^v(0)$ (in $\text{sec}^{-1}\text{cm}^{-1}$) of the SRS radiation from a unit surface of the powder, using formula (9):

$$E_S^v(0) = D \left| \frac{d\Phi_S^v(x)}{dx} \right|_{x=0} = \frac{1.15DBL\eta(v)}{5.14 - \eta(v)}. \quad (13)$$

We can write, in accordance with (5') and (11),

$$K_g / 6.6 K_a = E_L / E_L^t = P_L / P_L^t,$$

where E_L^t is the powder illumination at which generation sets in, and P_L and P_L^t are the power and threshold power of the laser. We substitute $D = K_a L^{-2}$ in (13). Then, after making the appropriate transformations, we get

$$E_S^v = \frac{1.14BK_a L^{-1} A}{1 - A}, \quad A = [P_L I(v) (P_L^t)^{-1}]^{1/2}, \quad (14)$$

where P_L^t is the threshold laser power for the excitation of generation at the center of the line. From this formula we can easily derive a relation for the line width of the SRS end of the spontaneous Raman scattering:

$$\delta\nu_{\text{SRS}} = \delta\nu_S \sqrt{1 - P_L / P_L^t}. \quad (15)$$

Multiplying the expression for the intensity of the spectral component of the SRS (14) by the expression for the SRS line width (15), we obtain an approximate formula for the total intensity of the SRS line in the powder:

$$E_S(0) \approx \frac{2.3BKL^{-1} \delta\nu_S \sqrt{P_L / P_L^t}}{\sqrt{1 - P_L / P_L^t}}. \quad (16)$$

To obtain an expression for the power P_S of the Stokes radiation it is necessary to multiply E_S by the radiating area S of the powder surface and by the quantum energy $\hbar\omega_S$ at the Stokes frequency.

It can be shown that when SRS is excited by a laser pulse in the form $I(t) = I_0 \exp[-\ln 2(2t/\tau_L)^2]$, where τ_L is the pulse width at half height, the SRS pulse duration τ_S depends on the power of the exciting radiation in the following manner:

$$\tau_S \approx \tau_L [\ln(4 - 3P_L / P_L^t)]^{1/2} (\ln 2)^{-1/2}. \quad (17)$$

We note once more that formulas (14)–(17) are valid for $6.6 > K_g K_a^{-1} > 5$, and consequently for $P_L^t > P_L > 0.75 P_L^t$.

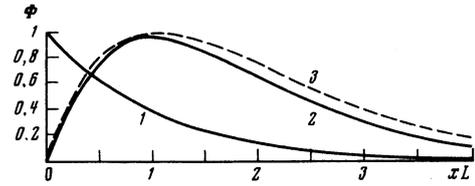


FIG. 6. Distribution of the quantum fluxes over the thickness of the powder: 1—for the exciting radiation (formula (3)); 2—for the SRS radiation near the generation threshold (formula (12)); 3—for the spontaneous Raman scattering radiation (formula (8)). The functions are normalized to their maxima.

3. A criterion for the applicability of the diffusion equations (1) and (2) to powder is satisfaction of the condition that the absorption coefficients and the threshold gain K_a and K_g^t be small in comparison with the scattering coefficient $K_{\text{SC}} \approx (3D)^{-1}$. The values for D , K_g^t , and K_a can be obtained from the relations $R = (1 - 2DL)(1 + 2DL)^{-1}$, $L = \sqrt{K_a D^{-1}}$, $K_g^t = 6.6 K_a$, if one measures R and L . The value $L = 4.6 \text{ cm}^{-1}$ for stilbene was taken from $^{[15]}$, and our measurements yielded $R = 0.7$. Then $D = 0.019 \text{ cm}$, $K_{\text{SC}} = 17.8 \text{ cm}^{-1}$, $K_a = 0.4 \text{ cm}^{-1}$, and $K_g^t = 2.6 \text{ cm}^{-1}$, and consequently $K_{\text{SC}} \gg K_g^t, K_a$.

Let us estimate the ratio $E_S(0)/E_L$ for Raman scattering. Recalculating the spontaneous scattering cross section for the line $\Delta\nu = 1593 \text{ cm}^{-1}$ $^{[14]}$ for the red region of the spectrum ($\sigma = 8.3 \times 10^{-27} \text{ cm}^2$) and recognizing that $N = 3.6 \times 10^{21} \text{ cm}^{-3}$ and $\rho = 0.5$, we get from (10) $E_S(0)/E_L = 2.5 \times 10^{-6}$. If we use the ratio of the SR radiation energy in the saturation and spontaneous scattering sections, which is equal to 10^5 , and the data of $^{[15]}$ for the coefficient of conversion into SRS in the saturation section ($\sim 20\%$), then we obtain $E_S(0)/E_L = 2 \times 10^{-6}$, which results in good agreement between the estimates based on formula (10) and the experimental results.

Let us examine the dependence of the threshold power P_L^t on the powder temperature. On the basis of $^{[11]}$ we can write $K_g \sim E_L \sigma (\delta\nu_S)^{-1}$. It was reported in $^{[16]}$ that σ for the stilbene line $\Delta\nu = 1593 \text{ cm}^{-1}$ remains unchanged in the interval $100\text{--}300^\circ \text{K}$ (and the value of $\delta\nu_S$ is 3.7 cm^{-1} for 100° and 4.8 cm^{-1} for 300°K), so that we can write $K_g(T) \sim E_L / \delta\nu_S(T)$. To maintain K_g constant with changing temperature, it is necessary to change $E_L \propto \delta\nu_S(T)$. It follows from this, in particular, that the threshold illumination is $E_L^t = P_L^t(T) (S \hbar \omega_L)^{-1} \sim \delta\nu_S$, where S is the illuminated area of the powder and $\hbar \omega_L$ is the energy of the exciting-radiation quantum, or that $P_L^t(T) \sim \delta\nu_S(T)$. Our measurements have shown that the powder parameters R and L remain unchanged in the temperature interval $100\text{--}300^\circ \text{K}$, and thus do not affect the temperature dependence of P_L^t . This might mean an expected dependence of the type $P_L^t(T) \sim \delta\nu_S(T)$. In the experiments, however, the temperature has a stronger influence (cf. Fig. 2c). A similar result was obtained in $^{[17]}$ for the temperature dependence of the SRS threshold in calcite.

An even stronger temperature dependence of $P_L^t(T)$ is observed for the $\Delta\nu = 997 \text{ cm}^{-1}$ line, for which the value of σ remains constant at temperatures $100\text{--}300^\circ \text{K}$, and $\delta\nu_S$ decreases on cooling by not more than a factor

1.5^[16], whereas the generation threshold decreases by more than 20 times (cf. curve b of Fig. 4). Such a behavior of the threshold for this line can be attributed to a decrease in the SRS line owing to the appreciable transfer of the energy of the exciting radiation to the first SRS Stokes component of the $\Delta\nu = 1593 \text{ cm}^{-1}$ line, the generation for which sets in earlier than for the $\Delta\nu = 997 \text{ cm}^{-1}$ line.

As to the dependence of the Stokes radiation P_S on the laser intensity P_L , a comparison of the experimental data with the results of a calculation by formulas of the diffusion approximation shows that it describes well the experimental results.

Figure 3b shows the theoretical dependence of the Stokes radiation P_S on the ratio P_L/P_L^t , calculated with the aid of formulas (10), (14), and (16). The same figure shows the experimental points pertaining to the first Stokes component of the stilbene line $\Delta\nu = 1593 \text{ cm}^{-1}$. To obtain identical scales on the ordinate axis, we chose the same slope for the theoretical and experimental plots at the origin, i.e., in the region corresponding to the spontaneous Raman scattering. The calculated curve agrees qualitatively with the experimental relation. The difference between them is apparently due to some difference between the theoretical premissis and the experimental conditions (the powder constituted a layer rather than a half-space, and a limited section of the surface was illuminated).

In the stationary case the SRS line width tends to zero when the generation threshold is approached (cf. formula (17)), but it is difficult to observe this narrowing at $P_L < P_L^t$, owing to the low intensity of the SRS in this region. After the generation threshold is reached, i.e., at $P_L < P_L^t$, the development of the SRS in the powder has a nonstationary character. In the nonstationary case, according to^[8], the spectral width of the emission line of a generator with nonresonant feedback narrows down in accordance with the law

$\delta\nu \approx \delta\nu_0(K_g vt)^{-1/2}$. Substituting for K_g the mean gain in a layer of thickness L^{-1} at threshold pumping

($\overline{K_g^t} = 1.4 \text{ cm}^{-1}$), substituting for t the duration τ_S of the SRS pulse at an exciting radiation power $P_L > P_L^t$ ($\tau_S = 6 \text{ nsec}$) and substituting $\delta\nu_S$ for $\delta\nu_0$, we obtain $\delta\nu_{\text{SRS}} = 0.35 \text{ cm}^{-1}$. This value agrees with the data

of^[18], where a value $\delta\nu_{\text{SRS}} = 0.37 \text{ cm}^{-1}$ was obtained for the $\Delta\nu = 1593 \text{ cm}^{-1}$ line at $P_L > P_L^t$. The experimentally obtained plot of the SRS pulse duration τ_S is described qualitatively by formula (19) in the region $0.75 \cdot P_L^t < P_L < P_L^t$, and at $P_L \ll P_L^t$ (the region of the spontaneous Raman scattering) we have $\tau_S \approx \tau_L$.

Let us estimate the laser power P_L^t (in $\text{erg}\cdot\text{sec}^{-1}$), needed to excite SRS generation. The illumination of the powder surface (in terms of the number of quanta) is $E_L^t = P_L^t (S\hbar\omega_L)^{-1}$, where S is the area of the illuminated powder surface and $\hbar\omega_L$ is the energy of the laser quanta. Using the solution, we obtain with the aid of formulas (t') and (11) the expression

$$P_L^t = \frac{3.3K_g S \hbar\omega_L}{(R+1)g(v,\rho)}$$

An estimate with the aid of this formula gives values $P_L^t = 4.2$ and 3.1 MW for generation of the powder at temperatures 300 and 100° K , respectively. Experiment has yielded the values $P_L^t = 4.8$ and $P_L^t = 2.4 \text{ MW}$, which is in satisfactory agreement with the calculated data.

In conclusion, the authors are indebted to A. V. Kraiskii for help with practical advice during the experiments and also to N. I. Lipatov for supplying the radiation receivers.

¹V. A. Zubov, G. V. Peregudov, M. M. Sushchinskiĭ, V. A. Chirkov, and I. K. Shuvalov, *ZhETF Pis. Red.* 5, 188 (1967) [*JETP Lett.* 5, 150 (1967)].

²Ya. S. Bobovich and A. V. Bortkevich, *Zhur. Prikl. Spektrosk.* 10, 992 (1969).

³Ya. S. Bobovich, A. V. Bortkevich, and V. V. Kryukov, *Opt. Spekt.* 30, 257 (1971).

⁴Ya. S. Bobovich and A. V. Bortkevich, *ZhETF Pis. Red.* 7, 20 (1968) [*JETP Lett.* 7, 13 (1968)].

⁵Ya. S. Bobovich and A. V. Bortkevich, *Usp. Fiz. Nauk* 103, 3 (1971) [*Sov. Phys.-Uspekhi* 14, 1 (1971)].

⁶A. P. Ivanov, *Optika rassivayushchikh sred (Optics of Scattering Media)*, Nauka i tekhnika, Minsk, 1969.

⁷V. S. Letokhov, *ZhETF Pis. Red.* 5, 262 (1967) [*JETP Lett.* 5, 212 (1967)].

⁸V. S. Letokhov, *Zh. Eksp. Teor. Fiz.* 53, 1442 (1967) [*Sov. Phys.-JETP* 26, 835 (1968)].

⁹E. K. Kazakova, A. V. Kraiskii, M. M. Sushchinskiĭ, and I. K. Shuvalov, *FIAN Kratk. soobshch. po fizike* 7 (1970).

¹⁰A. M. Weinberg and E. P. Wigner, *Physical Theory of Nuclear Chain Reactors*, Univ. of Chicago Press, 1958.

¹¹M. Maier, W. Kaiser and J. A. Giordmaine, *Phys. Rev.* 177, 588, 1969.

¹²G. N. Watson, *Theory of Bessel Functions*, Cambridge Univ. Press.

¹³E. Jahnke and F. Emde, *Tables of Functions*, Dover, 1945.

¹⁴M. M. Sushchinskiĭ, *Spektry kombinatsionnogo rassenyaniya molekul i kristallov (Raman Spectra of Molecules and Crystals)*, Nauka, 1969.

¹⁵V. A. Zubov, A. V. Kraiskii, and M. M. Sushchinskiĭ, *FIAN Preprint No. 189, 1968: FIAN Preprint No. 145, 1970.*

¹⁶V. A. Chirkov, V. S. Gorelik, G. V. Peregudov, and M. M. Sushchinskiĭ, *Paper at Fourth All-union Symposium on Nonlinear Optics*, Kiev, October, 1968.

¹⁷W. Heinicke and G. Winterling, *Appl. Phys. Lett.* 11, 231, 1967.

¹⁸V. A. Chirkov, V. S. Gorelik, G. V. Peregudov and M. M. Sushchinskiĭ, *ZhETF Pis. Red.* 10, 416 (1969) [*JETP Lett.* 10, 267 (1969)].