

INFLUENCE OF DAMPING ON THE TEMPORAL CHARACTERISTICS OF STIMULATED RAMAN SCATTERING OF LIGHT

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In the interaction between the exciting radiation and the first Stokes component of the stimulated Raman scattering of light (SRS) it is usually considered that at large transformation coefficients the polarizability of the medium follows the field instantaneously, i.e., the process is regarded as quasistationary. This approach makes it possible to explain the most characteristic features of SRS. In the present paper, for the case of large transformation coefficients, the first approximation is taken to be the finite time of establishment of the oscillations in the medium. In this case the solution has an asymmetrical form for a time-symmetrical pump pulse. The correction is most noticeable in the case of exciting radiation passing through matter; there is then a dip in the center of a Gaussian pulse and the height of the first hump is greater than the height of the second. With increase of pumping power, the distance between the humps increases, and its magnitude is a linear function of the relative difference between the hump heights. These conclusions are confirmed by our experiments.

THE finite time of establishment of the process of stimulated Raman scattering of light (SRS) has been taken into account in a number of recent papers.<sup>[1, 2]</sup> This is connected with the possibility of exciting SRS by picosecond radiation pulses and with the possibility of formation of ultrashort radiation pulses with the aid of the SRS phenomenon.<sup>[3]</sup> It turns out that the relaxation time also plays a role in the excitation of SRS by nanosecond pulses of radiation.

An experimental investigation of the temporal characteristics of SRS at large transformation coefficients has been carried out in<sup>[4]</sup>. The most interesting aspect is the waveform of the exciting-radiation pulse passing through matter: if the exciting-radiation pulse entering the cell is Gaussian in time, it acquires a dip at its center after passing through the medium, i.e., it acquires a two-hump shape, with the height of the first maximum being larger than that of the second. It follows from the model assumed in<sup>[4]</sup> for the phenomenon that the heights of the maxima should be equal. Allowance for the difference between the propagation velocities of the exciting radiation and of the first Stokes component<sup>[5]</sup> does not explain this fact, since for such changes in the pulse it is necessary that the dispersion of the medium be anomalous, something not very likely for liquid nitrogen when working with a ruby laser. This effect can be attributed to the finite time of establishment of the SRS process, and a possibility is uncovered of determining the relaxation time from the waveform of the pulse. The calculation can be carried out within the framework of the model developed in a number of papers on SRS.<sup>[6-10]</sup> Account is taken of the exciting radiation and of the first Stokes component, and it is assumed that the amplitudes of the waves vary sufficiently slowly. The vibrations of the molecule are described by an equation that connects the change of the vibrational coordinate with the derivative of the polarizability of the molecule. A slow time dependence is also assumed for the amplitude of the vibrations. Within the framework of this model, the equa-

tions for the field amplitudes of the exciting radiation  $E_0(z, t)$  and of the Stokes radiation  $E_S(z, t)$  and the amplitude of the vibrations of the molecule  $Q(z, t)$  take the form

$$\frac{n}{c} \frac{\partial E_0}{\partial t} + \frac{\partial E_0}{\partial z} = i \frac{\pi N \omega_0}{cn} \frac{\partial \alpha(q)}{\partial q} Q E_0, \tag{1a}$$

$$\frac{n}{c} \frac{\partial E_S}{\partial t} + \frac{\partial E_S}{\partial z} = i \frac{\pi N \omega_s}{cn} \frac{\partial \alpha(q)}{\partial q} Q^* E_0, \tag{1b}$$

$$\frac{\partial Q}{\partial t} + \frac{1}{\tau} Q = i \frac{1}{4\mu\Omega} \frac{\partial \alpha(q)}{\partial q} E_0 E_S^*. \tag{1c}$$

In these expressions  $n$  is the refractive index,  $\omega_0$  and  $\omega_s$  the frequencies of the exciting and of the Stokes radiation,  $\Omega$  the vibration frequency,  $\tau$  the time of establishment,  $N$  the density of the molecules of the medium,  $\partial \alpha(q)/\partial q$  is the derivative of the polarizability of the molecule with respect to the normal coordinate, and  $\mu$  is the reduced vibrating mass. It is usually assumed that  $\tau$  is negligibly small, i.e.,  $\partial Q/\partial t \ll Q/\tau$ . The solution of (1c) is then

$$Q = i \frac{\tau}{4\mu\Omega} \frac{\partial \alpha}{\partial q} E_0 E_S^*. \tag{2}$$

When  $\tau$  is taken into account we have in the next higher order approximation, with allowance for the initial conditions  $E_0(z, t_0) = 0$  and  $E_S(z, t_0) = 0$ :

$$Q = i \frac{\tau}{4\mu\Omega} \frac{\partial \alpha}{\partial q} \left[ E_0 E_S^* - \tau \frac{\partial}{\partial t} (E_0 E_S^*) \right]. \tag{3}$$

Taking into account the smallness of the time  $\tau$ , we have confined ourselves to only two terms. We see that the first term in (3) corresponds to the solution (2), the second term corresponds to the perturbation connected with allowance, in the first approximation, of the finite establishment time  $\tau$ . Substituting expression (3) in (1a) and (1b) and changing over to the quantum density:

$$n_0 = \frac{n^2}{8\pi\hbar\omega_0} E_0 E_0^*, \quad n_s = \frac{n^2}{8\pi\hbar\omega_s} E_S E_S^*,$$

we obtain

$$\frac{n}{c} \frac{\partial n_0}{\partial t} + \frac{\partial n_0}{\partial z} = -\sigma n_0 n_s + \frac{\sigma \tau}{2} \frac{\partial}{\partial t} (n_0 n_s), \quad (4a)$$

$$\frac{n}{c} \frac{\partial n_s}{\partial t} + \frac{\partial n_s}{\partial z} = \sigma n_0 n_s - \frac{\sigma \tau}{2} \frac{\partial}{\partial t} (n_0 n_s), \quad (4b)$$

where

$$\sigma = \frac{4\pi^2 N \omega_0 \omega_s \tau \hbar}{\mu \Omega c n^3} \left( \frac{\partial \alpha}{\partial q} \right)^2.$$

We seek the solution in the form

$$n_0 = n_0^0 (1 + \beta_0), \quad n_s = n_s^0 (1 + \beta_s). \quad (5)$$

$n_0^0(z, t)$  and  $n_s^0(z, t)$  is the solution of the unperturbed problem ( $\tau = 0$ ) with boundary conditions  $n_0^0(0, t) = f_0(t)$  and  $n_s^0(0, t) = f_s(t)$ ,<sup>[4, 5]</sup> and  $\beta_0$  and  $\beta_s$  are functions of the time and of the coordinate and take the perturbation into account.

We shall confine ourselves henceforth to a consideration of  $n_0(z, t)$ —the exciting radiation passing through the sample—since it is precisely for this case that the experiments reveal the largest peculiarities, namely the two-hump structure.<sup>[4]</sup> From (4) and (5) we obtain an equation for  $\beta_0(z, t)$ . The solution of this equation, neglecting the terms containing  $\tau\beta_0$  and  $\beta_0^2$ , with allowance for the boundary condition  $\beta_0(0, t) = 0$ , takes the form

$$\beta_0(z, t) = \left\{ [\exp \Phi(z, C)] \int_0^z [\exp(-\Phi(x, C))] \cdot \frac{\sigma \tau}{2n_0^0(x, C)} \left[ \frac{\partial}{\partial t} [n_0^0(x, t) n_s^0(x, t)] \right] \Big|_{t=C+nz/c} dx \right\} \Big|_{t=C-nz/c}, \quad (6)$$

where

$$\Phi(z, C) = \sigma \int n_0^0(z, C) dz.$$

An exact calculation of this integral is impossible, and we therefore introduce certain simplifications and limitations of the calculation region, so that together with the simplifications we separate the characteristics that are most convenient for estimates and for comparison with experiment.

The first simplification is the assumption that  $f_s(t) = \gamma f_0(t)$ , where  $\gamma$  is small. Then the solution of the unperturbed problem takes the form

$$n_0^0(z, t) = \frac{f_0(t_z)}{1 + \gamma \exp[\sigma z f_0(t_z)]}, \quad (7)$$

$$n_s^0(z, t) = \frac{f_0(t_z) \gamma \exp[\sigma z f_0(t_z)]}{1 + \gamma \exp[\sigma z f_0(t_z)]},$$

where we introduce the notation  $t_z = t - nz/c$ . It should be noted that this simplification is not too critical, and is important only to make  $f_s(t) \ll f_0(t)$ . This is seen from the fact that perfectly analogous results are obtained if it is assumed that  $f_s(t) = \text{const } f_0(t)$ .

The second limitation is connected with the fact that we are considering only the region of the maxima for the exciting radiation passing through the sample. These maxima are clearly pronounced and it is possible to use them for a comparison with experiment and for estimates of the establishment time. It can be assumed that allowance for  $\beta_0$  does not shift the maxima strongly, and acts mainly on their relative magnitude. Therefore the condition for the maxima is best obtained from the first relation in (7). For exciting radiation from a sample with working length  $l$ , the condition of the maxima with respect to time has the form

$$\gamma [\exp(\sigma l f_0(t_i))] [f_0(t_i) \sigma l - 1] = 1.$$

We take into consideration the fact that the gains are large. This is due to the fact that we are considering a region of appreciable conversion of the energy of the exciting radiation, i.e.,  $f_0(t_l) \sigma l \gg 1$ . Then

$$\gamma \sigma l f_0(t_i) \exp[\sigma l f_0(t_i)] = 1 \quad (8)$$

and

$$\gamma \exp[\sigma l f_0(t_i)] \ll 1.$$

We thus have for the region of the maxima in time from (7)

$$n_0^0 \text{max} = f_0(t_{\text{max}}) = f_0(t_{\text{max}}),$$

$$n_s^0 \text{max} = \gamma f_0(t_{\text{max}}) \exp[\sigma z f_0(t_{\text{max}})].$$

These relations are valid so long as it can be assumed that

$$\gamma \exp[f_0(t_i) \sigma z] \ll \gamma \exp[f_0(t_i) \sigma l] \ll 1.$$

This means that near the maxima the conversion of the energy of the exciting radiation into SRS is negligible. Leaving a sample with working length  $l$ , taking into account the derived relations, we obtain from (6)

$$\beta_0(l, t) \text{max} = \frac{1}{4} \sigma \tau l \left. \frac{\partial f_0(t_i)}{\partial t} \right|_{t_i=t_{\text{max}}}. \quad (9)$$

This relation makes it possible to assess the change of the waveform of the pulse of the exciting radiation passing through the sample as a result of the finite time of development of the process,  $\tau$ . We see that on the leading front of the pulse of the exciting radiation we have  $\partial f_0/\partial t > 0$  and  $\beta_0 \text{max}$  is positive, and on the trailing edge of the pulse  $\partial f_0/\partial t < 0$  and  $\beta_0 \text{max}$  is negative. Thus, we obtain the experimentally observed asymmetry of the pulse: the front maximum is higher and the rear maximum is lower.

Let us take into account the concrete form of the exciting-radiation pulse—a Gaussian pulse with duration  $2T$  at the  $1/e$  height:  $f_0(t) = n_0 \exp[-(t/T)^2]$ . It is assumed that the maximum of the pulse corresponds to the time  $t = 0$ . Calculation in accordance with (9) yields

$$\beta_0(l, t)_{\text{M}} = \frac{1}{2} \sigma \tau \frac{|t_{\text{M}}|}{T^2} f_0(t_{\text{M}}). \quad (10)$$

In this expression  $2t_{\text{M}}$  is the time distance between the maxima, and  $2\beta_0(l, t)_{\text{M}}$  is the difference between the heights of the maxima.

Since  $f_0(t_{\text{M}})$  is the solution of the algebraic equation determining the maximum (8), this quantity is constant. The obtained relation (10) establishes a direct proportionality between the time separation of the maxima  $2t_{\text{M}}$  and the difference between the heights of the maxima  $2\beta_0(l, t)_{\text{M}}$ . In addition, the obtained relation makes it possible to determine the establishment time  $\tau$ :

$$\tau = 2 \frac{1}{\sigma l} \frac{T^2}{|t_{\text{M}}|} \frac{\beta_0(l, t)_{\text{M}}}{f_0(t_{\text{M}})} = 2 \frac{S}{\sigma l} \frac{T^2}{|t_{\text{M}}|} \frac{\beta_0(l, t)_{\text{M}}}{P(t_{\text{M}})}. \quad (11)$$

This expression contains quantities that are determined experimentally. From the observed waveform of the exciting-radiation pulse passing through the sample we determine  $2\beta_0(l, t)_{\text{M}}$ —the relative difference between the heights of the maxima, and  $2t_{\text{M}}$ —the time distance between the maxima. Measurement of the waveform of the pulse of the exciting radiation at the input gives  $2T$ —the

duration of the pulse at height  $1/e$  and  $P_0(t_M)$ —the intensity of the radiation at the instant of time  $t_M$ , where  $S$  is the cross section of the beam. The quantity  $\sigma'l/S$  can be determined from the dependence of the peak intensity of the first Stokes component on the peak intensity of the exciting radiation under conditions when the system operates with a given pump, i.e., with a small transformation coefficient. For this case we have, in analogy with (7),

$$P_s^0(l, t)_p = P_0(t)_p \gamma \exp \left[ P_0(t)_p \frac{\sigma'l}{S} \right].$$

The slope of the plot constructed in a semilogarithmic scale:

$$\ln P_s^0(l, t)_p = P_0(t)_p \sigma'l/S + \ln P_0(t)_p + \ln \gamma,$$

is given by

$$\frac{d \ln P_s^0(l, t)_p}{d P_0(t)_p} = \frac{\sigma'l}{S} \left[ 1 + \frac{1}{P_0(t)_p \sigma'l/S} \right] \approx \frac{\sigma'l}{S},$$

since

$$P_0(t)_p \frac{\sigma'l}{S} = f_0(t) \sigma l \gg 1.$$

To verify the correctness of the approximations and simplifications adopted here, we measured experimentally the waveform of the pulse of exciting radiation passing through the sample. The characteristics of the radiation were measured with a previously described setup.<sup>[4]</sup> The object of the investigation was liquid nitrogen. The measurement conditions were chosen such that the energy of the first Stokes component was sufficiently high, i.e., the conversion coefficient was high. At the same time, the energy of the second Stokes component was a small fraction of the energy of the first Stokes component and could be neglected. In the experiment, however, in contrast to the calculation, the exciting radiation was focused into a cell with the sample. Another departure from the calculation is that the distribution of the exciting radiation over the cross section differed somewhat from rectangular. Thus, a comparison of the calculations with the experiment can characterize only the general situation and cannot claim high accuracy.

We first measured the dependence of the energy of the first Stokes component on the energy of the exciting radiation. This made it possible, on the one hand, to choose for the time measurements the region of SRS of interest to us, and on the other hand to determine the quantity  $\sigma'l/S$ . In the chosen region, we measured the waveform of the radiation pulses photoelectrically and simultaneously monitored the energy of the exciting radiation. The characteristics indicated above were measured. The investigations were made both with the laser operating in the multimode regime and with the laser operating in the single-mode regime. No differences were observed in the results.

An example of the waveform of the exciting-radiation pulse passing through the sample is shown in Fig. 1. One sees clearly the aforementioned characteristic change in the form of the pulse, consisting in formation of two maxima with different heights. Figure 2 shows, for comparison with the calculation, a plot of the relative difference in the heights of the maxima against the

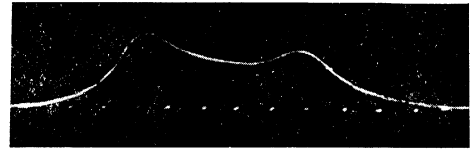


FIG. 1. Waveform of radiation pulse passing through the medium.

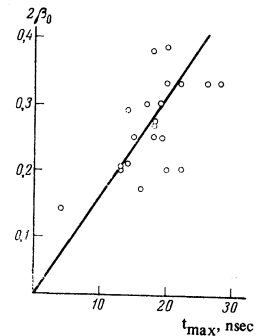


FIG. 2. Dependence of the difference of the heights of the humps, referred to the height of the first hump, on the distance between the humps.

time distance between the maxima. The abscissas represent the distances between the maxima, and the ordinates the differences between the heights of the maxima, referred to the height of the first maximum. It can be assumed that there is a proportionality between the relative difference of the heights of the maxima and the time interval between the maxima (in accordance with relation (10)).

On the basis of the experiments we can estimate the establishment time  $\tau$ . Measurements give the following parameters:  $2t_M = 40$  nsec,  $2\beta_0(l, t)_M = 0.3$ ,  $2T = 42$  nsec,  $\sigma'l/S = 10$  MW<sup>-1</sup>,  $P(t_M) = 1.56$  MW. Calculation gives  $\tau = 0.45$  nsec or  $\Gamma = 2/c\tau = 0.15$  cm<sup>-1</sup>. Usually it is assumed<sup>[10]</sup> that the time of development of the process is connected with the width of the line in the spectrum of the ordinary Raman scattering. For liquid nitrogen this width is  $\sim 0.07$  cm<sup>-1</sup>.<sup>[11]</sup> If we recognized the possibility of processes such as generation due to feedback resulting from the scattering of the radiation, then the time of development may differ from that indicated.

In our case, for purposes of evaluating the approximations introduced, it is possible to compare these quantities in some sense. We see that in order of magnitude the results coincide. The difference is perfectly understandable, if account is taken of the approximate character of the model in question on the one hand, and, on the other, the difference under the conditions of calculation and experiment. The main factors here are apparently the deviation of the energy distribution of the cross section from rectangular, focusing of the exciting radiation into a cell with the sample, and the crude allowance for the vibrations of the molecule.

It is interesting to note that in spite of the short establishment time of the process ( $\tau = 0.45$  nsec) compared with the pulse duration ( $2T \approx 40$  nsec), its influence on the waveform of the pulse is appreciable: the relative difference in the heights of the humps, which are separated by approximately 40 nsec, reaches  $\sim 0.3$ .

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