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# STIMULATED RAMAN SCATTERING OF LIGHT ABSORBING MEDIA EXCITED BY ULTRASHORT LASER PULSES

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16. Abstract

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# STIMULATED RAMAN SCATTERING OF LIGHT IN ABSORBING MEDIA EXCITED BY ULTRASHORT LASER PULSES

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The fluctuation-dissipation theory of spontaneous and stimulated vibration Raman scattering is worked out taking into account the dissipation losses at frequencies of laser pump and scattering radiation. General expressions are found, which describe the absolute intensities and shape, energy and duration of scattered pulses in terms of the parameters of the medium and the parameters of the input laser pulses. The general regularities are analyzed in detail. Conditions are found for the realization of spontaneous or stimulated Raman scattering and its dependence on absorption, pulse duration and other parameters of the problem.

## Introduction /1746*

A good study has been made by now [1 - 3 and others] of stimulated Raman scattering (SRS) of light in transparent media and nonstationary mode which is realized with pumping by ultrashort laser pulses. However in many cases (for example, with resonance or quasi-resonance excitation of SRS) we have to deal with a fairly significant linear absorption in the medium at laser excitation frequencies $\omega_l$ and Stokes' radiation $\omega_s$. Stationary SRS in dissipative media has already been previously studied (see, for example [4]), however nonstationary SRS has not been studied, which could impair, in particular, an interpretation of experimental results.

We consequently developed a theory for nonstationary oscillating SRS in dissipative media, whose main conclusions are presented in this work. The theory is constructed in the framework of the known fluctuation-dissipation technique [4]. Because of spatial damping and temporal modulation of the waves, the Stokes' "seed" sources determining the emergence of scattered radiation are not equivalent in different points of the medium and different moments in time, so that it is necessary to have a correct consideration for the spatial-temporal relationship of fluctuation polarization and field with frequency $\omega_{l,s}$, which we have done.

*Numbers in margin indicate pagination in original foreign text.

The theory presented below is a quantum theory. It was constructed for simplicity as applied to scattering on nonpolar eigen fluctuations in crystals. The results are generalized without difficulty for the case of nonresonance SRS on polaritons, as well as for the case of SRS in liquids; for this in the final formulas it is sufficient to replace the phenomenological parameter of the task, the amplification coefficient, by its corresponding value (compare with [3]).

All the field quantities for the Stokes' wave and the scattering excitations are Heisenberg operators, and the pumping field is a classic linear-polarized flat nonmonochromatic wave propagating along the z axis with electrical field $E_1(z, t) = e_1 A_1(z, t) \times \exp[i(k_{1z} - \omega_{1t})] + k.s. = e_1(E_1^{(+)} + E_1^{(-)})$, $E_1^{(+)} = A_1 \exp[i(k_{1z} - \omega_{1t})]$, $E_1^{(-)} = (E_1^{(+)})^*$. The indexes 1 and s designate quantities referring to the pumping wave and the Stokes' wave respectively. e--real unit vectors of polarization, $\alpha = \omega\varepsilon''/cn$ --energy coefficients of absorption, $n = \sqrt{\varepsilon'}$, $\varepsilon = \varepsilon' + i\varepsilon''$ --complex dielectric permeability of the medium, $k = \omega n/c$. It is assumed that $\alpha_{l,s} \ll k_{l,s}$, however the quantities $\alpha_{1,s}L$ (L is the thickness of the scattering layer) can be random. Definite polarization of the Stokes' wave $e_s$ is considered active in scattering. Generalization to the case of other wave polarizations $\omega_{1,s}$ is reduced to the corresponding replacement of the amplification coefficient. /1747

## 1. Calculating Scattering Intensity

When light is scattered in a layer, the observed quantum-statistical average quantities depend only on the variables z and t. However in problems of the scattering theory, energy (quadratic for the fields) characteristics must be average, while the values of the field are determined with regard for the presence in the medium of distributed fluctuation sources, and consequently, also depend on the variables x and y.

We will present the Stokes' field $E_s(r, t) = e_s E_s(r, t)$, as well as the oscillating coordinate $Q_\sigma(r, t)$ (the index $\sigma$ designates the mutually degenerate oscillations with specified frequency $\alpha_f$, if these are available), normed for one elementary cell, in the form of Fourier expansions with respect to variables x and y:

$$E_s(r,t) = E_s^{(+)}(\mathbf{r},t) + E_s^{(-)}(\mathbf{r},t), \quad E_s^{(-)} = (E_s^{(+)})^-,$$
$$E_s^{(+)}(\mathbf{r},t) = \int d^2k_s^\tau \xi_s(z, \mathbf{k}_s^\tau, t) \exp[i(\mathbf{k}_s\mathbf{r} - \omega_s t)], \quad \omega_s = \omega_l - \omega_f,$$
$$Q_\sigma(\mathbf{r},t) = Q_\sigma^{(+)}(\mathbf{r},t) + Q_\sigma^{(-)}(\mathbf{r},t), \quad Q_\sigma^{(-)} = (Q_\sigma^{(+)})^-,$$
$$Q_\sigma^{(+)}(\mathbf{r},t) = \int d^2k_\sigma^\tau \xi_\sigma(z, \mathbf{k}_\sigma^\tau, t) \exp[i(\mathbf{k}_\sigma\mathbf{r} - \omega_f t)].$$

Here $k_s^{x,y}$ and $k_\sigma^{x,y}$--arguments of the Fourier transform, $\xi_s(z, k_s^\tau, t)\exp[i(k_s^z z - \omega_s t)]$ and $\xi_\sigma(z, \mathbf{k}_\sigma^\tau, t)\exp[i(\mathbf{k}_\sigma^\tau z - \omega_f t)]$ --Fourier-patterns, $k^\tau_{s,\sigma}$ --two-dimensional vectors with components $k^x_{s,\sigma}$ and $k^y_{s,\sigma}$; $k_{s,\sigma} = (k^\tau_{s,\sigma}, k^z_{s\sigma})$, and $k_s^z = [k_s^2 - (k_s^\tau)^2]^{1/2}$, while $k^z_\sigma = k_l - k^z_s$. It is clear from considerations of symmetry that waves with $k^\tau_s = -k^\tau_\sigma$ interact, therefore with fixed $k_s$ we can assume $k_\sigma = w$, where $w = k_l - k_s$.

The system of equations

$$\frac{\partial \xi_s}{\partial z} + \frac{1}{v_s \cos\theta}\frac{\partial \xi_s}{\partial t} + \frac{\alpha_s}{2}\xi_s = b_0 A_l \exp(-\alpha_l z/2)\sum_\sigma \alpha_\sigma \xi_\sigma^{(+)}, \tag{1}$$

$$\frac{\partial \xi_\sigma^{(+)}}{\partial t} + \frac{\gamma}{2}\xi_\sigma^{(+)} = -\frac{i\alpha_\sigma^*}{\hbar}A_l^* \exp(-\alpha_l z/2)\xi_s - \frac{i}{2\omega_f}K_\sigma^{(+)}\exp(-i\omega_f t) \tag{2}$$

(where $b_0 = 2\pi i\omega_s N/cn_s\cos\theta$, $\theta$ --scattering angle, N--concentration of cells, $\alpha_\sigma = e_s^i\alpha_{ij}^\sigma e_l^j$, $\alpha_{ij}^\sigma$ --normal tensor of spontaneous Raman scattering in calculation for one cell) is converted into that examined in publication [3] if we assume $\alpha_l = \alpha_s = 0$. As in [4], we ignore the eigen radiation at frequency $\omega_s$, considering it to be weak. Excluding $\xi_\sigma^{(+)}$, we obtain one equation of the second order in relation to $\xi_s$. As in [3], it assumes solution by the Riemann method. The transition to the observed characteristics of scattered radiation is also made similarly [3] in the framework of the procedure described there for quantum-statistical averaging.

We will examine a more interesting and important case where only local nonstationariness is significant, governed by the finiteness of the relaxation time $\tau_0 = 1/\gamma$. The wave nonstationariness governed by difference in the rates of propagation of pulses $v_{l,s}$ will be ignored; this is formally equivalent to identification of the quantities $v_l$ and $v_s \cos\theta$. The surface brightness $B_s(\theta, t)$ of the output edge at frequency $\omega_s$ is a fairly complete and adequate energy characteristic in this case. We will immediately write the final expression for $B_s$: /1748

$$B_s = B_0 \frac{\exp(-\alpha_s L)}{L}\left[e^{-\frac{\eta-\eta_0}{\tau_0}}\int_0^L dz e^{-\beta z} I_0^2(\varphi_1) + \int_0^L dz e^{-\beta z}\int_{\eta_0}^{\eta} d\eta' e^{-\frac{\eta'-\eta}{\tau_0}} I_0^2(\varphi_2)\right]. \qquad (3)$$

here

$$B_0 = B_s^0 \frac{\pi}{2\tau_0} g_0 \rho(\eta_L)(\bar{n}_j + 1) L, \qquad B_s^0 = \frac{\hbar\omega_s^3 n_s^2}{8\pi^3 c^2}, \quad \beta = \alpha_l - \alpha_s,$$

$$\varphi_1 = \left[\psi \int_{\eta_0}^{\eta} \rho(\eta_1)\, d\eta_1\right]^{1/2}, \quad \psi = \frac{G}{\tau_0 a_l}\left(e^{-a_l\zeta} - e^{-a_l}\right), \quad G = g_0 L, \qquad (4)$$

$$a_l = \alpha_l L, \quad \zeta = z/L,$$

$$\varphi_2 = \left[\psi \int_{\eta'}^{\eta} \rho(\eta_1)\, d\eta_1\right]^{1/2}, \quad \eta = t - L/v_l, \quad g_0 = \frac{8\pi\omega_s \alpha^2 N (|A_l^0|^2)_{\max}}{c n_s \hbar\gamma\cos\theta}$$

where $\alpha^2 = \sum_\sigma |\alpha_\sigma|^2$ --amplification coefficient in the center of the scattering line of the stationary SRS with $A_l$ equal to peak value of the input amplitude $A_l(0, t)$. Function $\rho(\eta) = |A_l^0(\eta)|^2/(|A_l^0(\eta)|^2)_{\max}$ describes the shape of the pumping pulse, $A_l(\eta)$ is determined by the correlation $A_l(z,t) = A_l^0(\eta)\exp(-\alpha_l z/2)$, $A_l^0(\eta)$ --value of the amplitude without consideration for damping. Finally, $I_0$ is the modified Bessel function of zero order, $\bar{n}_f = [\exp(\hbar\omega_f/kT) - 1]^{-1}$, $\eta_0 = t_0$ --moment that the pumping pulse approaches the front edge of the crystal. (In the case of a nonrectangular pumping pulse, selection of the moment is conditional to a certain degree and is dictated by calculation accuracy).

With $\alpha_{l,s} = 0$, expression (3) becomes the corresponding expression from [3].

Analysis of the Results

It is apparent from (3) that, as in the case of nondissipative media, only the amplitude (but not the phase) modulation of pumping pulses is significant. Analysis of some characteristic specific laws of nonstationary SRS will be made further on the assumption that the pumping pulses are fairly short ($\tau_l \ll \tau_0$), have square shape and the difference in absorption coefficients at frequency $\omega_{l,s}$ is small ($L|\alpha_l - \alpha_s| \ll 1$). In this case one can assume $\alpha_s \approx \alpha_l = \alpha$, as a result (3) is transformed as follows:

$$B_s = B_0 \frac{e^{-a}}{L} \int_0^1 I_0^2(\varphi)\, d\zeta, \quad \varphi = \sqrt{\psi\eta}, \quad a = \alpha L. \qquad (5)$$

The quantities $\psi$ and $\eta$ are determined by formulas (4). Figure 1 presents the dependences of the shape of the scattered radiation pulse on absorption calculated from (5).

We will further discuss the question of the SRS threshold from peak intensity of pumping and its dependence on the absorption level. The absorption coefficient in (5) is contained in the pre- /1749
exponential factor exp(-a) and in the argument of the Bessel function $I_0$ (with $a \ll 1$ the absorption is insignificant, therefore it is expedient to consider that $a \gtrsim 1$). If $G\tau_1/a\tau_0 \ll 1$, then $\varphi \ll 1$, $I_0^2 \approx 1$ and $B_s = B_0 \exp(-a)$, i.e., a mode of spontaneous Raman scattering (PRS)

is realized, insofar as $B_s \sim L$, while the factor exp(-a) transmits simple damping of pumping and scattered radiation during propagation in the media. The transition to the SRS mode could occur with disruption in the condition $\tilde{G}\tau_1/a\tau_0 \sim 1$.

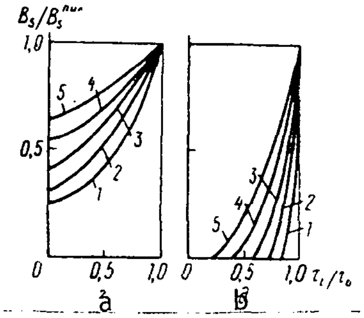



Figure 1. Temporary Shape of the Stokes' Impulses with Different Absorption Quantities

Key:
1. a = 0.5
2. 1
3. 1.5
4. 2
5. 2.5

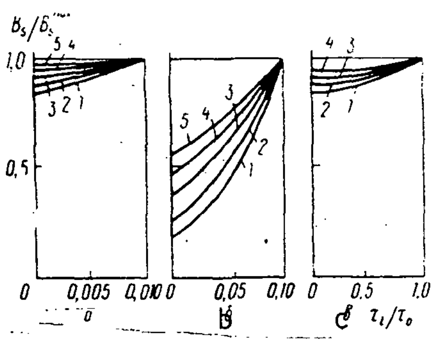
a. $\tau_1 = 0.01\ \tau_0$, $G = 100$

b. $\tau_1 = 0.1\ \tau_0$, $G = 100$

c. $\tau_1 = \tau_0$, $G = 1$

d. $\tau_1 = \tau_0$, $G = 10$

e. $\tau_1 = \tau_0$, $G = 100$



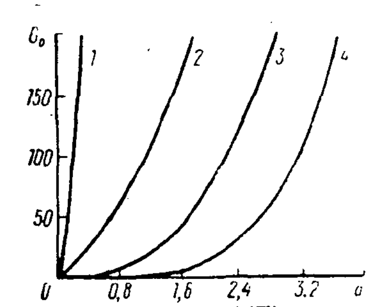




Figure 2. Dependence of Threshold Amplification Coefficient $G_0$ on Absorption with Different Durations of the Pumping Pulse

Key:
1. $\tau_1 = 0.01\ \tau_0$
2. $0.1\ \tau_0$
3. $\tau_0$
4. $\tau_1 >> \tau_0$

More accurate identification of the threshold conditions is possible based on appropriate analysis of the SRS threshold. This concept, however, is conditional to a significant measure; a detailed discussion of different methods for determining it is contained, for example, in [4]. One of the possibilities for the analysis corresponds to the approach which is traditional for quantum electronics based on compensation for losses by amplification. In the case of SRS it is specified as follows (compare with [4]): amplification must be sufficient for the output Stokes' radiation[whose intensity is determined by formula (5)]to be the same as it would be in the absence of dissipative losses. The output radiation of PRS is Stokes' radiation of minimum intensity. It is natural that there is rapid increase in the generation intensity, starting with the threshold intensity of pumping defined in this manner. In the case of nonstationary SRS, it is expedient to identify the peak intensity $B_s^{max}$ and $(B_s^{PRS})^{max}$. The aforementioned analysis of the threshold results in the following transcendent equation for the threshold value $G_0$:

$$e^{-a}\int_0^1 I_0^2(\tilde{\varphi})\,d\zeta = 1, \quad \tilde{\varphi}=\sqrt{\tau_l}\psi.$$

Its solution with different $\tau_1$ is illustrated in Figure 2 (curves 3 and 4 were constructed from common formulas of section 1). For comparison we present curve 4 which corresponds to the threshold of stationary SRS ( $\tau_1 >> \tau_0$). These data allow us to judge the degree of /1750 increase in the threshold with the transition from stationary to significantly nonstationary mode depending on $\tau_1$. Similar behavior of the SRS threshold occurs with other methods of its analysis (see [4]).

We will further examine the energy of the SRS pulses $E_s$. It can be obtained by integration of expression (5) over time, as a result

$$\mathcal{E}_s = B_0\tau_l e^{-a}\int_0^1 [I_0^2(\tilde{\varphi}) - I_1^2(\tilde{\varphi})]\,d\zeta. \qquad (6)$$

By using (5) and (6) we can introduce effective duration of the Stokes' pulses $\tau_s = E_s/B_s^{max}$ ($B_s^{max} = B_s(\eta = \tau_1)$). We are easily

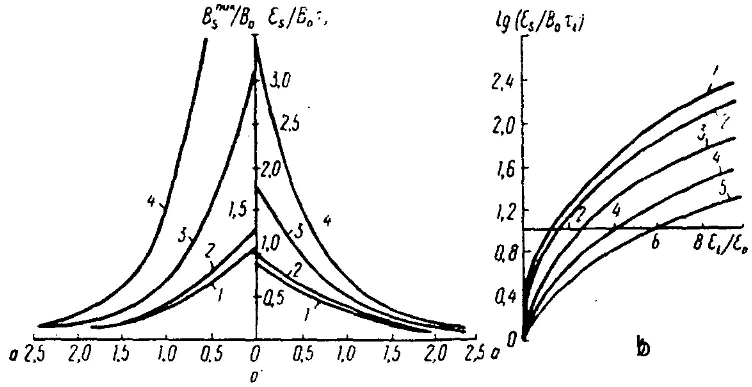


Figure 3. a) dependence of energy and peak intensity on absorption with different energy of the pumping pulse

Key:
1. $E_1 = 0.1\ E_0$
2. $E_0$
3. $5\ E_0$
4. $10\ E_0$ ($E_0 = J_1^0 \tau_1$)

b) dependence of energy of Stokes' pulses on energy of pumping pulses with different absorption quantities

1. a = 0.25
2. 0.5
3. 1
4. 1.5
5. 2

convinced that $\tau_s \leq \tau_1$. The formulas for $E_s$ and $\tau_s$ are simplified in the case of moderately short pulses with $\tau_0/G \ll \tau_l \ll \tau_0$. Here $\tilde{\varphi} \gg 1$, so that we can use the asymptotic presentation of the Bessel function [5], which yields

$$\mathcal{E}_s = B_0\tau_0 \exp(-a + 2\tilde{\xi})/2\pi\tilde{\xi},\ \tilde{\xi} = (\tilde{\tau}_l G')^{1/2},\ \tilde{\tau}_l = \tau_l/\tau_0,$$
$$G' = g_0 L',\ L' = (1 - e^{-a})/\alpha,\ \tau_s = \tau_l/\tilde{\xi}. \tag{7}$$

With weak absorption (a ≪1), taking into consideration that $\tilde{\xi} = \xi_0 = (\tilde{\tau}_l G)^{1/2} \gg 1$, we obtain from (6)

$$\mathscr{E}_s = B_0 \tau_l \exp(-\xi_0 \alpha L/2)/2\pi\xi_0.$$

The influence of absorption is characterized by the factor exp $(-\xi_0 \alpha L/2)$, so that it is stronger the greater the pumping intensity.

With strong absorption ($a > 1$) it follows from (6) that

$$\mathscr{E}_s = B_0 \tau_l \sqrt{a} \exp(-a + 2\xi_0/\sqrt{a})/2\pi\xi_0.$$

The SRS process can evidently occur fairly effectively, if $\xi_0 > a^{3/2}/2$; in this case the dependence on absorption is determined mainly by the factor $\exp(2\xi_0/\sqrt{a})$. The listed laws are partially illustrated in Figure 3. In Figure 3, a, dependences of $B_s^{max}$ and $E_s$ on a are presented with different $E_l$. The nonmonotonic nature of the dependence of $E_s$ on $E_l$ is also visible from Figure 3, b. In conclusion we present the formulas following from (3) for the case where $\alpha_l \ll \alpha_s$, which can be realized with scattering pumping which is quasi-resonance for frequency [6]: /1751

$$B(\eta) = B_0 \frac{L''}{L} e^{-\alpha_s L} [I_0^2(\varphi_3) - I_1^2(\varphi_3)], \quad \varphi_3 = (g_0 L'' \eta/\tau_0)^{1/2},$$

$$L'' = (1 - e^{-\alpha_l L})/\alpha_l,$$

$$\mathscr{E}_s = 2B_0 \frac{L''}{L} \tau_l e^{-\alpha_s L} [I_0^2(\varphi_4) - I_1^2(\varphi_4) - I_0(\varphi_4) I_1(\varphi_4)/\varphi_4], \quad \varphi_4 = (g_0 L'' \tau_l/\tau_0)^{1/2}.$$

The influence of absorption is characterized here by the effective length L" defining the area of pumping existence. In the case of moderately short pulses, as we are easily convinced, the threshold condition adopts the appearance $L'' = 4L \ln^2 \varphi_0/\varphi_0^2$, where $\varphi_0$ is the value $\varphi_4$ with threshold $g_0$, while the duration of the Stokes' impulses is defined by formula (7) as before.

The findings could prove useful in interpreting experimental data in studying SRS in the field of ultrashort pumping pulses. In particular, they indicate the significant increase in the nonstationary threshold of SRS with a decrease in the duration of the pumping pulse, as well as increase in the role of absorption with increase in the pumping pulse energy.

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