

# Stimulated light scattering and four-photon processes

D. N. Klyshko

Moscow State University

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Small-angle stimulated Raman scattering and scattering at the wing of the Rayleigh line are considered by using a very simplified model. In the small-angle region, the interaction of the Stokes (s) and antistokes (a) field components is important. As sources, both the distributed fluctuations, which are proportional to the energy dissipation of the field in the medium, and the vacuum fluctuations of the field at the boundary ( $z = 0$ ) between the linear and nonlinear regions of the medium are taken into account. It is shown that the distributed fluctuation sources at conjugated ( $\omega_s + \omega_a = 2\omega_0$ ) s- and a-frequencies are completely correlated and this strongly affects the scattered field spectrum. The frequency-angular spectrum permits the transition to zero pumping intensities  $S_0$  and for extreme values of the parameters includes linear spontaneous scattering and four-photon scattering, which latter is quadratic with respect to  $S_0$  ("scattering of light by light"). An interesting result of the numerical calculation is that the a-radiation is due to fluctuations of vacuum and not of molecules for  $\hbar(\omega_a - \omega_0) \gtrsim kT$  and large  $S_0$  ( $T$  is the temperature of the sample). The contribution of vacuum fluctuations to the Stokes part of the spectrum does not exceed  $\sim 3\%$ . Calculation of the spectrum of higher s- and a-components shows that the model employed (with an infinite cross section of the pumping beam) does not explain the clear-cut ring angular structure of the components.

One of the most unexpected phenomena accompanying stimulated Raman scattering is the radiation directed along the cone generatrix at the antistokes frequency.<sup>[1]</sup> This radiation has been interpreted<sup>[2]</sup> as diffraction of the incident laser beam (the "pump") by the traveling lattice which is formed by the stimulated oscillations of the molecules. In photon language, this process, which is also called parametric or coherent, corresponds to a four-photon transition, in which two laser photons with frequency  $\omega_0$  decay into a pair of photons with frequencies  $\omega_s$  and  $\omega_a$ . Analogous secondary processes lead to radiation of higher components.<sup>[3,4]</sup>

Bloembergen and Shen<sup>[3,4]</sup>, as well as the authors of many other works on stimulated light scattering, restricted themselves essentially to analysis of the amplification factor  $g$ , which is the root of the quadratic dispersion equation obtained from Maxwell's equations for plane and monochromatic s- and a-waves. Such a consideration,<sup>[1]</sup> which is based on HOMOGENEOUS EQUATIONS (without sources), is frequently too crude (in particular, it does not permit the limit transition to spontaneous scattering). This inadequacy is also characteristic of researches devoted to the study of s- and a-interactions in scattering at the "wing" of the Rayleigh line.<sup>[6,7]</sup> In<sup>[5,8-10]</sup>, inhomogeneous equations were used; however, other limitations appear in these works—thus, the correlation of s- and a- sources was not considered in them, nor quantum fluctuations of the field, nor the reverse effect of a- and s-waves. An s-source was used in<sup>[9]</sup> that was monochromatic and localized at the origin. We also consider the paper of Arbatskaya et al.,<sup>[11]</sup> in which an attempt was made to use 4-photon processes to explain the observed effect of the repetition, in the Stokes frequency-angular spectrum, of the pump spectrum. However, we note that there is a contradiction in the conservation law given in<sup>[11]</sup>—the energies of the two incident (as also of the two scattered) photons enter into it with different signs. After correction of the signs, the process has the form  $\omega_0 + \omega_s \rightarrow \omega'_0 + \omega'_s$ , and leads only to the mutual broadening of the spectra of the pump

and the Stokes component (this process corresponds to a nonlinear susceptibility  $\chi(\omega'_s = \omega_s + \omega_0 - \omega'_0)^{[4]}$ ). We also emphasize that in the resonance region, it is impossible to separate the four- and two-photon processes, although this is often done. The constants characterizing these processes ( $\gamma$  and  $\beta$  in the expressions given below) enter nonadditively into the propagation constant  $g$  (see (9) below). Both the input (vacuum) fluctuations of the s- and a-fields, which are responsible for "pure" four-photon scattering far from the combination frequencies  $\omega_0 \pm \Omega_0$ , and the distributed molecular fluctuations, which produce "pure" Rayleigh ( $\Omega_0 = 0$ ) and Raman scattering, are taken into account in the present work. The analysis is carried out in the simplest—scalar stationary and parametric—approximations. The nonlinear properties of the medium are described phenomenologically by means of the cubic polarizability  $\chi$ .<sup>[3,4]</sup> The initial equations (Sec. 1) differ from those used in<sup>[3,4,6,7]</sup> only by the addition of Langevin forces, which are determined with the help of the fluctuation-dissipation theorem (FDT) or its nonlinear analogs,<sup>[12,13]</sup> and by account of input noise. A similar model has been used by Strizhevskii and coworkers<sup>[14]</sup> for the description of stimulated polariton and parametric scattering, and also by Bepalov et al.<sup>[15]</sup> for the description of Rayleigh scattering (but without account of s-a interaction and input noise). The general features of the spectrum are discussed in Sec. 2 and the contribution of vacuum (external) noise is considered in more detail in Sec. 3, and the molecular (internal) noise in Sec. 4.

Let us now consider briefly the question of the ring-shaped angular spectrum of the higher components, which has been observed in a number of experiments (for example,<sup>[16-18]</sup>). There exist at least three different explanations for the nonisotropy of the higher components. For example, let us consider the second Stokes component (ss), which can arise as a result of two different parametric processes with the following conditions of synchronism:

$$k_{ss} = k_s + k_0 - k_a, \quad (1a)$$

$$\mathbf{k}_{ss} = \mathbf{k}_s + \mathbf{k}'_s - \mathbf{k}_0. \quad (1b)$$

Chiao and Stoicheff<sup>[17]</sup> (see also<sup>[4]</sup>) explained the diameter  $V_{SS}$  of the rings observed by them by the condition (1a), evidently assuming that all the vectors are coplanar. However, Lugovoi<sup>[9]</sup> directed attention to the fact that in the case of isotropic s-radiation, the condition (1a) does not give a definite scattering angle  $\nu_{SS}$  inasmuch as the pairs of vectors  $\mathbf{k}_{SS}$ ,  $\mathbf{k}_S$  and  $\mathbf{k}_0$ ,  $\mathbf{k}_a$  do not necessarily lie in a single plane. Lugovoi<sup>[9]</sup> developed a theory which leads to the condition (1b) for  $\mathbf{k}'_S = \mathbf{k}_S$  and later verified it experimentally, together with Ataev.<sup>[18]</sup> However, the theory of<sup>[9]</sup> is not sufficiently convincing, inasmuch as a point source is used in it, which also leads to the additional condition  $\mathbf{k}'_S = \mathbf{k}_S$ . We note that the lack of correlation of the waves emitted by individual molecules does not prevent their four-photon transformation. Finally, there is still another, third explanation,<sup>[16]</sup> based on the assumption that the s field consists of two parts— isotropic and directed ( $\mathbf{k}'_S \parallel \mathbf{k}_0$ ). Fixing of the direction  $\mathbf{k}'_S$  in (1b) leads to single-valuedness of  $\nu_{SS}$ .

Calculation of fields of higher components by means of our one-dimensional model with two types of sources (which we shall not give here in view of its cumbersome nature) showed that neither one of the processes (1) gives a ring spectrum. Obviously, one can describe it only with the help of a three-dimensional model, which takes into account the boundedness of the cross section of the pump beam.

## 1. INITIAL EQUATIONS

We consider an isolated nonlinear layer of thickness  $l$  in an unbounded transparent medium. The pump wave

$$E_0 \exp(i k_0 z - i \omega_0 t) + \text{c.c.} \quad k_0 = k_{0l} + \beta_0,$$

propagates perpendicular to the layer, where  $\beta_0$  takes into account the "self-force," and  $k_{0l}$  is the linear wave vector. We represent the scattering field in the form of a superposition of plane waves with definite frequencies and transverse wave-vector components  $\mathbf{q} = \mathbf{k}_\perp$  and with amplitudes slowly dependent on  $z$  (incomplete—three-dimensional—Fourier transformation):

$$E(r, t) = \int d\omega \iint d\mathbf{q}_\perp d\mathbf{q}_\parallel E_{\omega\mathbf{q}} \exp(i\mathbf{k}\mathbf{r} - i\omega t) + \text{H.c.} \quad (2)$$

$$\mathbf{k} = \mathbf{q} + z\mathbf{k}_z, \quad k_z = \sqrt{k^2 - q^2} = k \cos \theta, \quad k = n\omega / c.$$

The amplitudes of the field for  $z \leq 0$  do not depend on  $z$  and are connected with the ordinary Fourier components  $E_{\mathbf{k}}$  by the relation  $E_{\omega\mathbf{q}0} = E_{\mathbf{k}}/u_z$ ; their correlators are equal to

$$\langle E_{\omega\mathbf{q}}^+ E_{\omega'\mathbf{q}'} \rangle = \frac{\hbar \omega N_\omega}{4\pi^2 c n |\cos \theta|} \delta(\omega - \omega') \delta(\mathbf{q} - \mathbf{q}'), \quad (3)$$

$$N_\omega = [\exp(\hbar\omega / kT) - 1]^{-1} = -N_{-\omega} - 1.$$

(The "antinormal" correlator  $\langle EE^+ \rangle$  differs by the substitution  $N_\omega \rightarrow -N_{-\omega}$ ).

The interaction of the waves is described by the nonlinear polarizations

$$P_{\omega\mathbf{q}} = \{ \chi(\omega_0 - \omega_0 + \omega) E \exp(i\mathbf{k}_z z) + \chi(2\omega_0 - \bar{\omega}) E^+ \exp[i(2k_0 - \bar{k}_z)z + 2\varphi_0] \} |E_0|^2, \quad (4)$$

$$\bar{\omega} \equiv 2\omega_0 - \omega, \quad \bar{\mathbf{q}} = -\mathbf{q}, \quad \bar{k}_z \equiv (\bar{k}^2 - \bar{q}^2)^{1/2},$$

$$E_0 \equiv |E_0| e^{i\varphi_0}, \quad E = E_{\omega\mathbf{q}}, \quad E^+ \equiv E_{\bar{\omega}\bar{\mathbf{q}}}$$

Substituting (3) and (4) into Maxwell's equation and neglecting the second derivatives, we obtain the following system for the "conjugate" amplitudes (with respect to the frequency and the transverse momentum):

$$dE/dz - i\beta E - i\gamma E^+ \exp[i\{(2\beta_0 - \Delta)z + 2\varphi_0\}] = if \exp[i(k_0 - k_z)z], \quad dE/dz - i\beta E - i\bar{\gamma} E^+ \exp[i\{(2\beta_0 - \Delta)z + 2\varphi_0\}] = i\bar{f} \exp[i(k_0 - \bar{k}_z)z]; \quad (5)$$

$$\Delta = k_z + \bar{k}_z - 2k_{0z}, \quad \beta = b\chi(\omega_0 - \omega_0 + \omega) |E_0|^2,$$

$$b \equiv 2\pi\omega^2 / c^2 k_z^2, \quad \gamma \equiv b\chi(2\omega_0 - \bar{\omega}) |E_0|^2$$

( $\bar{\beta}$ ,  $\bar{\gamma}$  are similar coupling constants for the conjugate wave; up to now we have not established whether  $\omega$  refers to the s-wave and  $\bar{\omega}$  to the a-wave or vice versa). On the right sides of (5) we have added the Langevin forces, which are proportional to the external polarizations:

$$f = f_{\omega\mathbf{q}} = bP_{\omega\mathbf{q}}^{\text{ext}} \exp[-i(k_0 z + \varphi_0)]. \quad (6)$$

This polarization arises as a consequence of the interaction of the molecular oscillations with the thermostat. As follows from the "nonlinear FDT<sup>[12,13]</sup> in a transparent<sup>2)</sup> (in the linear approximation) medium with cubic polarizability, fluctuations of the polarization develop under the action of the pump, with the spectrum

$$\langle P_{\omega\mathbf{r}'}^+ P_{\omega'\mathbf{r}'} \rangle = \frac{\hbar}{2\pi i} N_\omega [\chi(\omega + \omega_0 - \omega_0)_{i\beta\Omega} - \chi(\omega + \omega_0 - \omega_0)_{j\Omega}] \times E_{\Omega\mathbf{r}'} E_{\Omega\mathbf{r}'}^* \delta(\omega' - \omega) \delta(\mathbf{r}' - \mathbf{r}), \quad \Omega = \omega - \omega_0 \quad (7)$$

(the antinormal-ordered correlator differs by the substitution  $N_\Omega \rightarrow -N_{-\Omega}$ ).

For the calculation of scattering with account of s-a interaction, it is also necessary to know the mutual correlator of the forces  $f$  and  $\bar{f}$ . As is easily shown by following<sup>[12]</sup>, another variety of nonlinear FDT arises in the cubic medium and determines the mutual correlation of the conjugated modes:

$$\langle P_{\omega\mathbf{r}'}^+ P_{\omega'\mathbf{r}'} \rangle = \frac{\hbar}{2\pi i} N_{-\omega} [\chi(-\omega + 2\omega_0)_{i\beta\Omega} - \chi(-\bar{\omega} + 2\omega_0)_{j\Omega}] \times E_{\Omega\mathbf{r}'} E_{\Omega\mathbf{r}'}^* \delta(\omega' - \bar{\omega}) \delta(\mathbf{r}' - \mathbf{r}). \quad (8)$$

The correlator  $\langle P^+ P^+ \rangle$  is equal to the complex conjugate of the right side of (8) with the substitution of  $N_{-\Omega}$  by  $-N_\Omega$ .

The general solution of the system (5) is conveniently represented by means of the coefficients of amplification  $G$  and transformation  $F$ :

$$E_i = G_i E_0 + F_i E_0^+ + i \int_0^i dz [f_z G_{i-z} - \bar{f}_z^+ F_{i-z} \exp(-2i\varphi_0)] \times \exp[i\{(k_0 - k_z)z + \varphi_0\}],$$

$$G_i = \exp[i(\beta - \Delta_{nl}/2)z] (\text{ch } gz + i\Delta_{nl} \text{sh } gz) / 2g, \quad (9)$$

$$F_i = \exp[i(\beta - \Delta_{nl}/2)z + 2i\varphi_0] i\gamma \text{sh } gz / g,$$

$$\Delta_{nl} = \Delta + \beta + \beta^* - 2\beta_0, \quad g = (\gamma\bar{\gamma}^* - \Delta_{nl}^2/4)^{1/2}.$$

The normally ordered square of the solution (9), together with the correlators of the external (3) and internal (8) noise, determines the intensity of the scattering field to the right of the nonlinear layer. However, before writing down the explicit results, we make some further simplifications to decrease the cumbersome nature of the results and the number of parameters.

Up to now, we have not specified the frequency dependence of  $\chi$ . Let us now consider two simple cases, which correspond in some approximation to scattering by 1) the internal molecular oscillations with eigenfrequencies  $\Omega_0 \ll \omega_0$  and 2) the wing of the Rayleigh line (polarization component). Let the electron frequencies be much greater than  $\omega_0$  and  $\omega/\bar{\omega} \sim 1$ , then the interaction constants in (9) satisfy the following relations<sup>[4,6,7]</sup>

$$\beta^* = \beta = \beta_0 + \beta_a, \quad \beta_0 = \beta_{el} + \beta_{\text{Ral}}, \quad (10)$$

$$\bar{\gamma}^* = \gamma = \beta_{el} + \beta_a,$$

where  $\beta_{el}$  is proportional to the electron nonlinearity,  $\beta_{\text{Ray}}$  is the part of  $\beta$  that is independent of  $\Omega$ , due to

orientation of the molecules by the pump field, and  $\beta_\Omega$  is the resonance part of  $\beta$ , equal in the two cases to

$$\beta_\alpha = \beta_{\text{Ral}} / (1 - ix), \quad x = \Omega\tau, \quad (11a)$$

$$\beta_\alpha = -\text{sign } \Omega \cdot \beta_{\text{Ram}} / (x + i), \quad x = (\Omega - \Omega_0 \text{ sign } \Omega)\tau, \quad (11b)$$

where  $\tau$  is the corresponding relaxation time.

The nonlinear wave detuning in (9) does not now depend on the nonresonant part of  $\chi$ :

$$\Delta_{\text{nl}} = \Delta + 2\beta_\alpha, \quad (12)$$

while the gain is equal to

$$g = [\beta_{\text{el}}(\beta_{\text{el}} + 2\beta_\alpha) - \Delta(\beta_\alpha + \Delta/4)]^{1/2}. \quad (13)$$

Bloembergen<sup>[4]</sup> adduced a slightly different expression for the Raman case (11b) in connection with the neglect of the quantity  $\beta_0$ . In the Rayleigh case, (13) coincides with the expression obtained by Starunov<sup>[7]</sup> and Chiao et al., for  $\beta_{ll} = 0$ <sup>[6]</sup>. The linear wave detuning in the Raman case depends on the frequency and the angle of observation in the following way:

$$\Delta = (d^2k/d\omega^2)_0(\Omega^2 - \Omega_0^2) - k_1(\theta^2 - \theta_0^2). \quad (14)$$

In the Rayleigh region, we must set  $\Omega_0 = \nu_0 = 0$  and, as a rule, we can neglect the dependence of  $\Delta$  on the frequency.

Further, from the equality of the imaginary parts of the Fourier components of  $\chi$  follows the equality of the scalar characteristic and mutual correlators for external polarizations, so that for  $b/b \sim \omega/\tilde{\omega} \sim 1$  it follows from (7), (8) that

$$\langle f_{\omega q z}^+ f_{\omega' q' z'} \rangle = \langle f_{\omega q z} f_{\omega' q' z'}^+ \rangle = \hbar N_{\Omega} b \beta^*. \quad (15)$$

It follows from (15) that one can set the external forces equal in (5) and (9):  $\tilde{f}^+ = f$ , which materially simplifies the formula for the distributed noise.

## 2. FREQUENCY-ANGULAR SPECTRUM OF THE SCATTERED FIELD

The following expression for the spectral brightness<sup>3</sup> of the radiation scattered at the frequency  $\omega$  and at the angle  $\nu = \arccos(q/k)$  follows from (9) with account of (3), (15):

$$\frac{d^2S(\omega, q)}{d\omega d\Omega} = \frac{\omega^2 n^3 \cos \theta}{2\pi c} (E_l^2)_{\omega q} \equiv I_0 N_{\omega q}, \quad (16)$$

$$N_{\omega q} \equiv N_\omega |G|^2 + (N_\omega + 1) |F|^2 + N_\omega \cdot 2\beta'' \int_0^l dz |G_z - F_z|^2,$$

where  $I_0 = \hbar\omega k^2/8\pi^3$  is the "spectral brightness of the vacuum" to the left of the nonlinear layer, and  $N_{\omega q}$  is the number of photons in the mode to the right.

Thanks to the simplicity of the model used, Eq. (16) clearly shows the relative role of the external (the first two components) and internal (the third component) fluctuations. Moreover, (16) permits us to determine the region of applicability of the "homogeneous method" (see footnote 1), which is limited to analysis of the quantity  $g$  or at best,  $G$  and  $F$ .

The first term in (16) is the result of amplification (or attenuation) by a factor of  $|G|^2$  of the initial number of photons (without zero-point fluctuations) and does not play a role in ordinary experiments.<sup>4</sup> In the following, we shall consider  $N_\omega = N_{\tilde{\omega}} = 0$ .

## 3. VACUUM NOISE

The second term in (16), which is equal to

$$N_{\text{vac}} = |\beta_{\text{el}} + \beta_\alpha|^2 (\text{ch } 2g'l - \cos^2 g''l) / 2|g|^2, \quad (17)$$

can be interpreted as a result of parametric (coherent) transformation of (doubled) zero-point fluctuations from the conjugated mode  $(\tilde{\omega}, \tilde{q})$  to the observed  $(\omega, q)$  mode. This term, which is the same for the s- and a-frequencies and is appreciable only in the range of angles and frequencies connected by the synchronism condition  $\Delta \sim 0$ , describes, for  $\beta'' = 0$ , the "pure" four-photon scattering ("scattering of light by light"—see, for example,<sup>[20]</sup>). On the approach to resonance, the contribution of the vacuum noise (17) increases; however, it is comparable with the contribution of molecular noise only at sufficiently far a-frequencies, at which  $N_\Omega \lesssim 1$  (see below). In the case of Rayleigh scattering, the vacuum noise plays practically no role. The spectrum (17) for  $\beta_{\text{el}} = 0$  is shown in Fig. 1. For large wave detuning ( $|\Delta/\beta| \gg 1$ )

$$g \approx -\beta'' + i\Delta/2, \quad (18)$$

so that (17) goes over into nonlinear scattering for weak pumping ( $|\beta''|l \ll 1$ ):<sup>[20]</sup>

$$N_{\text{vac}} = |\beta_{\text{el}} + \beta_\alpha|^2 \left( \frac{\sin \Delta l/2}{\Delta/2} \right)^2 \quad (19a)$$

and into

$$N_{\text{vac}} = \left| \frac{\beta_{\text{el}} + \beta_\alpha}{\Delta} \right|^2 \exp(2|\beta''|l). \quad (19b)$$

for strong pumping. The weak (power-law) dependence of (19b) on  $\Delta$  probably explains the frequently observed longitudinal ( $x = 0$ ) stimulated antistokes scattering.

In the region of synchronism ( $|\Delta/\beta| \lesssim 1$ ) and for strong pumping  $|\Omega| \sim \Omega_0$ , the function  $N_{\text{vac}}(\Delta)$  has two sharp maxima, separated by a dip (Fig. 1c), as was predicted by Bloembergen and Shen.<sup>[3]</sup> The value of the splitting is of the order of  $4\beta_{\text{Ram}} l \equiv 2A$ . However, for  $A \lesssim 5$ , the dip disappears (Figs. 1a and 1b). We also note that, besides the angular splitting of the spectrum, a frequency splitting also occurs (of the order of half the spontaneous width—see Fig. 1).

According to a numerical calculation for  $A \gtrsim 5$  and  $\beta_{\text{el}} = 0$ , one can use the following formula to estimate the maximum value of  $N_{\text{vac}}$ :

$$N_{\text{vac max}} \approx 0.05 e^A. \quad (20)$$

Thus, the vacuum noise is smaller by about 1–2 orders of magnitude than the maximum molecular noise (for  $|N_\Omega| = 1$ ), which is equal to  $(e^A - 1)$  photons/mode.

## 4. MOLECULAR NOISE

Let us consider further the third term in (16), which depends on the temperature of the sample. For  $\beta_{ll} = 0$  its contribution (in photons/mode) is equal to

$$N_{\text{mol}} = N_\alpha \cdot 2\beta'' \int dz |\text{ch } gz - i\Delta \text{ sh } gz/2g|^2 \\ = N_\alpha \left[ 1 - \left| \frac{g''}{g} \right|^2 \text{ch } 2g'l - \frac{g''}{\Delta} \left( 1 + \left| \frac{\Delta}{2g} \right|^2 \right) \text{sh } 2g'l \right. \\ \left. - \left| \frac{g'}{g} \right|^2 \cos 2g''l - \frac{g'}{\Delta} \left( 1 - \left| \frac{\Delta}{2g} \right|^2 \right) \sin 2g''l \right], \quad (21)$$

where  $g$  and  $\beta$  are defined in (13) and (10). The relief of the function  $N_{\text{mol}}(x, \Delta)$  for some values of the parameters is shown in Fig. 2.

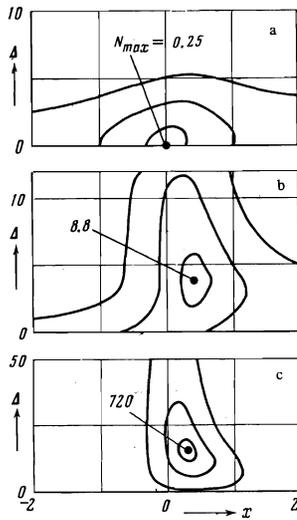


FIG. 1

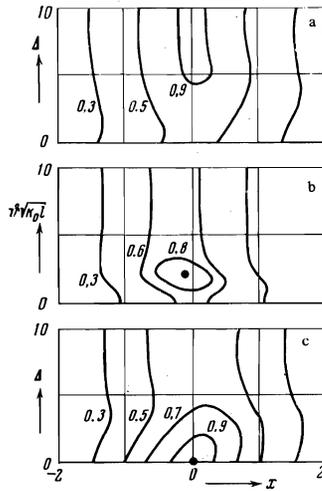


FIG. 2

FIG. 1. Frequency-angular spectrum of the vacuum part of the anti-Stokes scattering for various pump intensities ( $A = 2\beta_{\text{Ram}}l$ ),  $x = (\omega - \omega_0 - \Omega_0)\tau$ ,  $\Delta$  is proportional to the departure of the scattering angle from synchronism. The contour lines are drawn at the 0.1, 0.5 and 0.9 levels of  $N_{\text{max}}$ . It is assumed that  $\chi_{e1} = 0$ ; therefore the spectrum is invariant relative to inversion of the coordinates. a— $A = 1$ , b— $A = 5$ , c— $A = 10$ .

FIG. 2. Spectrum of the molecular part of the scattering for  $A = 1$  and  $\chi_{e1} = 0$  about a) the Stokes-Raman resonance; b) the Rayleigh resonance and c) the anti-Stokes Raman resonance. The maximum intensities are respectively equal to  $1.2(N_{\Omega_0} + 1)$ ,  $1.3kT\tau/h$  and  $1.0N_{\Omega_0}$  photons/mode.

For large wave detunings, (18) is satisfied and the usual isotropic expression follows from (21):

$$N_{\text{mol}}(x, \infty) = N_0[1 - \exp(-2\beta''l)]. \quad (22)$$

Generally, because of the factor  $\beta''$  (which is due to the FDT (7)), the function  $N_{\text{mol}}$  is important only in the resonance regions, and its behavior is very different in the three resonance regions: Rayleigh,  $\Omega \sim 0$  (Fig. 2b); Raman-Stokes,  $\Omega \sim -\Omega_0$  (Fig. 2a); and anti-Stokes,  $\Omega \sim \Omega_0$  (Fig. 2c). In the latter case, for not too weak pumping, the function  $N_{\text{mol}}/N_{\Omega}$  is surprisingly similar to  $N_{\text{vac}}$  (see (17)) (in any event for  $\beta_{II} = 0$ )—it has the same two maxima (Fig. 1c), and its maximum values are less than  $N_{\text{vac max}}$  (20) by only  $\sim 10\%$ . This correspondence is explained by the fact that both functions depend exponentially on the same function  $g'(x, \Delta)$ . For weak pumping however, their behavior is strikingly different:  $N_{\text{mol}}$  tends toward the ordinary linear isotropic scattering (Fig. 2c and (22)  $\beta'' > 0$ ), and  $N_{\text{vac}}$  tends toward the quadratic coherent scattering (19a).

Thus the anti-Stokes stimulated ( $A \geq 5$ ) scattering for  $\hbar\Omega_0 \sim kT$  is about half due to the zero-point vacuum fluctuations (in the conjugated Stokes modes of the left half-space) and half to the thermal fluctuations of the molecules.

In the region of the Stokes resonator ( $\Omega \sim -\Omega_0$ ) the principal role of the a-s interaction is reduced to disappearance of the scattering around the synchronism directions relative to (22) (Fig. 2a). As has been shown in [3,4] on the basis of the analysis of the function  $g'(\Delta)$  in the angular spectrum of the Stokes radiation, there appears a dark ring with a diameter determined by the synchronism condition. As  $|\Delta|$  increases, the intensity approaches the trivial value (22) almost monotonically

(Fig. 2a). The ring disappears upon a decrease in the pumping (see (23)).

Finally, in the case of Rayleigh resonance (Fig. 2b), the dark ring degenerates into a dip in forward scattering (for  $\Delta = \nu = 0$ ) and in contrast to the Stokes resonance, a bright ring borders on it. For strong pumping, the absolute maximum of the stimulated Rayleigh scattering, which is equal to  $\sinh A$  photons/mode,<sup>5</sup> occurs at  $\Omega = 0$ ,  $\Delta = -2\beta_{\text{Ray}}$  ( $\nu_{\text{max}} = \sqrt{2\beta_{\text{Ray}}/k_0l}$ ) which is identical with the position of the maximum of the function  $g'(x, \Delta)$ .<sup>[6,7]</sup> However, on a decrease in the pumping, the diameter of the ring increases (and reaches  $\sim 2\nu_{\text{max}}$  for  $A = 1$ , see Fig. 2b).

Thus, the model of a plane scattering layer leads to the following fundamental conclusions.

1. The scattering spectrum consists of two independent parts, which are determined by the temperatures of the sample  $T_{\text{mol}}$  and of the left half-space  $T_{\text{vac}}$ . In the region of Rayleigh and Stokes combination resonances for  $T_{\text{vac}} = 0$ , the contribution of the vacuum noise is negligible, while outside of resonance and in the region of sufficiently far-removed anti-Stokes resonances, it is basic.

2. The scattering spectrum is simply expressed by the amplification coefficient of the plane waves (see (16)) and has a complicated frequency-angular structure in the region of Bragg scattering angles (see Figs. 1, 2).

3. The spectrum of the higher components does not have strong angle maxima.

<sup>1</sup>This approach in [5] (which is specifically devoted to its inadequacies) is called the "plane-wave method." This term was unfortunate, since it is not the properties of the basis functions that are important, but the lack of consideration of the distributed character of the fluctuations (i.e., the inhomogeneity of the equations).

<sup>2</sup>Thus the case of Rayleigh scattering due to linear absorption is excluded here. (See, for example, the review of [15,19].)

<sup>3</sup>Usually the spectral intensity of the light measured in the far zone can be estimated roughly by multiplying (16) by the cross section of the pump beam  $a^2$ . We also note that in the framework of the model used, one can consider only angles bounded by the limits  $\vartheta, \pi - \vartheta \ll a/l$ .

<sup>4</sup>The case  $N_{\Omega} \neq 0$ ,  $\Omega > 0$  corresponds to the so-called "back" scattering.

<sup>5</sup>This exceeds the intensity of the back ( $\vartheta = 180^\circ$ ) scattering by a factor  $0.5e^{A/2}$ . It is surprising that only in one experimental work of Zaitsev [21] is there any mention of the existence of this ring.

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