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STIMULATED LIGHT SCATTERING INDUCED BY ABSORPTION

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I. INTRODUCTION

IN 1967, Herman and Gray[1] considered a new type of stimulated scattering of light (SS), which differed qualitatively from those previously known. The mechanism of this SS is peculiar to absorbing media and is due entirely to the presence of absorption.

The occurrence of the additional SS in absorbing media (we shall henceforth use the abbreviation SSA) is easiest to explain in the following manner. Assume that two waves propagate in an isotropic medium: a powerful laser wave $\mathbf{E}_L(\mathbf{r})\exp(-i\omega_L t)$ and a "signal" wave $\mathbf{E}_S(\mathbf{r})\exp(-i\omega_S t)$. If the absorption spectrum is broad enough and the absorption coefficient β is constant in all the frequency intervals of interest to us, then the power $Q(\mathbf{r}, t)$ released per unit volume is proportional to the instantaneous value of the square of the modulus of the total field $|\mathbf{E}(\mathbf{r}, t)|^2$. Therefore the expression for $Q(\mathbf{r}, t)$ contains the interference term

$$Q'(\mathbf{r}, t) = (cn\beta/8\pi)\,\mathbf{E}_S(r)\,\mathbf{E}_L^*(\mathbf{r})\exp(i\Omega t), \tag{1}$$

where n is the refractive index and $\Omega = \omega_L - \omega_S$. This term, obviously, accounts for the periodic variation (with frequency Ω) of the properties of the medium, including the change of dielectric constant ϵ.

Without specifying concretely the real processes responsible for the change of ϵ (these may be the thermal expansion of the medium, the change of the populations of the molecular levels, etc.—see Ch. II) and assuming for simplicity that the fields $\mathbf{E}_L(\mathbf{r})$ and $\mathbf{E}_S(\mathbf{r})$ are linearly polarized plane waves:

$$\mathbf{E}_L(\mathbf{r}) \sim \exp(i\mathbf{k}_L\mathbf{r}), \quad \mathbf{E}_S(\mathbf{r}) \sim \exp(i\mathbf{k}_S\mathbf{r}),$$

we write down the connection between $\delta\epsilon$ and Q' in general form

$$\delta\varepsilon(\mathbf{r}, t) = \hat{L}\left(\frac{\partial}{\partial t}, \frac{\partial}{\partial \mathbf{r}}\right) Q'(\mathbf{r}, t) = (cn\beta/8\pi)\exp(i\Omega t)\,\mathbf{E}_S(\mathbf{r})\,\mathbf{E}_L^*(\mathbf{r})\,L(i\Omega, i\mathbf{q}), \tag{2}$$

where L is a linear operator determined by the properties of the medium, and $\mathbf{q} = \mathbf{k}_S - \mathbf{k}_L$.

As a result of the scattering of the laser wave by the increment $\delta\epsilon$, there appears in the electric-induction vector an additional term

$$\mathbf{D}' = \delta\varepsilon\mathbf{E}_L(\mathbf{r})\exp(-i\omega_L t) = (cn\beta/8\pi)(\mathbf{e}_S\mathbf{e}_L)\,\mathbf{e}_L\,|\mathbf{E}_L(\mathbf{r})|^2 L(i\Omega, i\mathbf{q})\,\mathbf{E}_S(\mathbf{r})\exp(-i\omega_L t + i\Omega t), \tag{3}$$

where $\mathbf{e}_L$ and $\mathbf{e}_S$ are polarization unit vectors. Inasmuch as $\omega_L - \Omega = \omega_S$, the right side of (3) is proportional to the signal field $\mathbf{E}_S(\mathbf{r})\exp(-i\omega_S t)$. We therefore have for the components of the total electric-induction vector at the frequency ω_S

$$(\mathbf{D}_S)_i = \sum_k \varepsilon_{ik}^{\text{eff}}(\mathbf{E}_S)_k, \tag{4}$$

where

$$\varepsilon_{ik}^{\text{eff}} = \varepsilon_0\delta_{ik} + (cn\beta/8\pi)\,|\mathbf{E}_L|^2(\mathbf{e}_L\mathbf{e}_S)(\mathbf{e}_L)_i(\mathbf{e}_S)_k L(i\Omega, i\mathbf{q}) \tag{5}$$

and ϵ_0 is the dielectric constant of the medium without the field. Thus, the propagation of the "signal" wave in the presence of the laser field can be described by the effective dielectric constant $\epsilon_{ik}^{\text{eff}}$ from (5). The addition to ϵ_0 is proportional to $\beta|\mathbf{E}_L|^2$, i.e., it is essentially connected with the presence of absorption and increases linearly with increasing laser-field power. We assume further for simplicity that $\mathbf{e}_L \parallel \mathbf{e}_S$ and

$$\varepsilon^{\text{eff}} = \varepsilon_0 + (cn\beta/8\pi)\,|\mathbf{E}_L|^2 L(i\Omega, i\mathbf{q}). \tag{6}$$

As is well known, the imaginary part of the dielectric constant is determined by the dissipation of the electromagnetic energy in the medium. Since $\text{Im}\,\epsilon_0 = cn\,\beta/\omega$, it follows from (6) that

$$\text{Im}\,\varepsilon^{\text{eff}} = (cn\beta/\omega)\,[1 + (\omega/8\pi)\,|\mathbf{E}_L|^2\,\text{Im}\,L]. \tag{7}$$

The quantity Im L can be either positive or negative. In the latter case, the second term in (7) at low intensities $|\mathbf{E}_L|^2$ leads to partial compensation of the absorption, and it may become larger than the first term when $|\mathbf{E}_L|^2$ increases. The absorption then gives way to amplification.

For the corresponding threshold intensity of the laser field $I_L^{\text{thr}} = (cn/8\pi\hbar\omega)\,|\mathbf{E}_L|^2$(quanta/cm^{2}sec) and the gain $g \sim \text{Im}\,\epsilon^{\text{eff}}$ we obtain from (7)

$$I_L^{\text{thr}} = (cn/\hbar\omega)\,(-\omega\,\text{Im}\,L)^{-1}, \tag{8}$$

$$g = \beta[(I_L/I_L^{\text{thr}}) - 1]. \tag{9}$$

From (8) and (9) we get a number of remarkable properties of the considered SS mechanism, which disting-

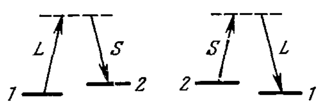

FIG. 1. Elementary quantum processes describing the spontaneous and stimulated scatterings of "ordinary" type. In process a) a laser quantum is absorbed and a signal quantum is generated, while the excess energy is transferred to the medium; process b) is the inverse.

uish it strongly from all other (ordinary) types of SS. Before we proceed to discuss the peculiarities of SSA, it is useful to recall a number of basic properties of ordinary types of SS. (For convenience, Appendix 1 contains a derivation of the corresponding formulas.)

The "ordinary" types of spontaneous and corresponding stimulated scattering are described in quantum mechanics by the transition n_L, $n_S \to n_L - 1$, $n_S + 1$, where n_L and n_S are the numbers of the quanta of the laser and signal fields. The corresponding probability is given by the square of the modulus of the transition amplitude $M \propto n_L^{1/2}(n_S + 1)^{1/2}$ (Fig. 1a). The medium goes over thereby into a state with energy $E_2 = E_1 + \hbar(\omega_L - \omega_S) = E_1 + \hbar\Omega$. The probability of such a transition is given by $w = AI_L(n_S + 1)$, i.e., it contains two parts: spontaneous AI_L and simulated $AI_L n_S$. As a result, the following universal connection exists between the intensity of the spontaneous scattering and the gain g_0 for the SS

$$g_0 = [(2\pi)^3/k^2]\, I_L\, (dR/do\, d\omega)\, [1 - \exp(-\hbar\Omega/kT)]; \qquad (10)$$

here $dR/do d\omega$ is the differential effective cross section for scattering of light by a unit volume (dR has the dimension of cm^{-1}), and $\Omega = \omega_L - \omega_S$ is the frequency difference between the incident and scattered photons. The last factor in (10) appears as a result of the balance of the direct (Fig. 1a) and inverse (Fig. 1b) processes ($\omega_L \to \omega_S$, $\omega_S \to \omega_L$). Because of this factor, gain is possible only in the Stokes region $\Omega > 0$.

The role of absorption in the case of ordinary types of SS reduces simply to a decrease of the effective amplification, so that the true gain is $g = g_0 - \beta$. Therefore, say in an absolutely transparent medium ($\beta = 0$), the gain of the Stokes component of scattering takes place for arbitrarily small values of I_L (i.e., $I_L^{thr} = 0$). When $\beta \neq 0$, we have $I_L^{thr} \sim \beta$. Therefore, although $g(I_L)$ is given by formula (9) also for ordinary types of SS, the value of g does not depend on β when $I_L \gg I_L^{thr}$.

The SSA process considered above does not have such properties.

First, according to (8), the threshold intensity I_L^{thr} of the SSA effect does not depend on β, and the amplification of the signal wave has itself principally a threshold character. At $I_L > I_L^{thr}$, on the other hand, the gain g for the SSA is proportional to β.

Second, depending on the form of the function $L(i\Omega, iq)$, the gain connected with the SSA can occur in both the Stokes and the anti-Stokes region. As shown in Ch. II, in a number of concrete cases the gain is realized in the anti-Stokes region.

Third, the SSA phenomenon does not correspond to any spontaneous-scattering process connected with it by relation (10).

Relation (10) is derived (see Appendix 1) from the most fundamental principles of quantum mechanics. This raises very acutely the question of the very nature of the SSA effect within the framework of the quantum-mechanical description, and whether the classical analysis presented above contradict quantum mechanics. The present article is devoted to a discussion of this entire group of problems connected with the specific nature of the SSA phenomenon.

First, in Ch. II, we consider within the framework of the classical scheme described above the different concrete SSA mechanisms connected with thermal conductivity, hypersound, and excitation of individual molecules. In Ch. III we present a quantum-mechanical description of the phenomenon. It is shown here that this phenomenon can be described as the result of a unique interference between the matrix elements of the first and third orders of perturbation theory. This in fact is the cause of all the differences from the ordinary types of scattering. Chapter IV is devoted to a clarification of the role of absorption in spontaneous-scattering processes. It is shown that the SSA process corresponds to a definite noise scattering which does not vanish at $I_S = 0$, and therefore plays the role of a spontaneous process. This scattering, however, turns out to be proportional to I_L^2 and is connected essentially with the shot noise of the quantum-absorption process.

Throughout the article, the field of the laser is assumed to monochromatic. A generalization to the case of a non-monochromatic laser field can be found in[1,2]. The result of this generalization reduces to a convolution of the gain (and also of ϵ_{eff} from (6)) with the spectrum of the laser. It should be noted that such a procedure for taking into account the laser spectrum in the case of large gain ($\exp(gl) \gg 1$) is, in general, insufficiently well founded. We shall not touch upon this question, all the more since it is not peculiar to the SSA effect, and arises in practically all stimulated-scattering problems.

We shall likewise not touch at all upon problems of propagation and generation of the "signal" field in space and in time. The status of this question is discussed in detail in a recent paper by Starunov and Fabelinskiĭ [2], devoted to a detailed analysis of different aspects of stimulated scattering.

II. CONCRETE TYPES OF SSA

In this chapter we discuss briefly three concrete mechanisms of stimulated scattering induced by absorption: temperature (entropy) scattering, scattering in the region of the Mandel'shtam-Brillouin doublet, and scattering connected with the change of the polarizabilities of individual molecules as they are excited.

1. Stimulated Temperature Scattering Connected with Absorption (STS-II)

The simplest type of SSA is connected with the heat-induced change of the dielectric constant of the medium. We shall consider heating of the medium only as a result of absorption of light. The corresponding scattering mechanism is designated STS-II in the review [2], to distinguish it from scattering occurring when the medium is heated by the electrocaloric effect (SRS-I). The STS-I phenomenon pertains to the "ordinary" type of stimulated scattering; it is connected genetically by relation (10) with the unshifted (entropy) component of the spontaneous scattering. We shall therefore not consider it.

It is customary to refer to stimulated temperature (entropy) scattering of light in the case when the changes of the temperature occur at constant pressure. In order for the pressure to have time to become equalized it is necessary to satisfy the relation $\tau \gg l/v_{ac}$, where v_{ac} is the speed of sound in the medium, $\tau = |\Omega|^{-1}$, and $l = |q|^{-1} \equiv |k_S - k_L|^{-1}$ are the characteristic temporal and spatial scales of the temperature variation. Thus, we assume that

$$|\Omega| \ll \Omega_{MB}, \tag{11}$$

where $\Omega_{MB} = |q| V_{ac}$ is the frequency-shift upon the Mandel'shtam-Brillouin scattering. When (11) is satisfied, the change of ϵ is given by the relation

$$\delta\varepsilon = \left(\frac{\partial\varepsilon}{\partial T}\right)_p \delta T, \tag{12}$$

and the change of the temperature obeys the thermal-conductivity equation

$$\frac{\partial T}{\partial t} - \varkappa \nabla^2 T = \frac{1}{\rho_0 c_p} Q(\mathbf{r}, t); \tag{13}$$

here κ is the temperature conductivity coefficient, and ρ_0 and c_p are the density and the specific heat of the medium.

It follows from (12) and (13) that the operator $\hat{L}$, which connects $\delta\epsilon$ and Q (see formula (2) of the introduction (Ch. I)), is equal to

$$L = \frac{1}{\rho_0 c_p}\left(\frac{\partial\varepsilon}{\partial T}\right)_p \frac{1}{i\Omega + \Gamma} \equiv \frac{1}{\rho_0 c_p}\left(\frac{\partial\varepsilon}{\partial T}\right)_p \frac{-i\Omega + \Gamma}{\Omega^2 + \Gamma^2}, \tag{14}$$

where $\Gamma = \Gamma(q) = \kappa q^2$ is the damping constant of the thermal wave with wave vector $q = k_S - k_L$.

In most cases $(\partial\epsilon/\partial T)_p < 0$, corresponding to thermal expansion of the medium; then the gain (Im L < 0) is realized in the anti-Stokes region ($\Omega < 0$). For the threshold intensity we obtain from (8) and (14)

$$I_L^{thr} = \frac{cn}{\hbar\omega} \frac{\rho_0 c_p}{\left(\frac{\partial\varepsilon}{\partial T}\right)_p} \frac{\Gamma^2 + \Omega^2}{\omega\Omega}, \tag{15}$$

and the gain is given by the general formula (19). The minimum value of I_L^{thr} (or what is equivalent, the maximum gain) is reached at $|\Omega| = \Gamma$. The sign of Ω at which the gain occurs coincides with the sign of $(\partial\epsilon/\partial T)_p$. The frequency dependence of the gain at $I_L \gg I_L^{thr}$ is shown schematically in Fig. 2a, where the STS-II phenomenon corresponds to the central part of the diagram. For comparison, Fig. 2b shows the form of the gain corresponding to the "ordinary" types of SS, and the central part of the diagram describes STS-I. We note that the functional form of $g(\Omega)$ is the same for STS-I and STS-II; for scattering in the region of the Mandel-shtam-Brillouin doublet, as will be seen from the next section, the situation is different.

We note that when light is absorbed by molecules it may turn out that a noticeable fraction of the absorbed energy goes into chemical transformations, and only a fraction $\mu < 1$ goes directly into heat. In this case it is necessary to substitute in (13) (and also in (20) below μQ in place of Q, and as a result the threshold intensity increases by a factor μ^{-1}.

2. SSA by Sound Waves (Mandel'shtam-Brillouin Doublet)

Let us consider now the SSA connected with the excitation of sound waves as a result of thermal expansion of the medium in inhomogeneous heating ($\exp(i\Omega t + i\mathbf{q}\cdot\mathbf{r})$). In analogy with the case of thermal waves considered above, the excitation of sound waves may be connected with other mechanisms, namely, with thermal expansion of the medium upon absorption of light and with the electrostriction effect; the latter corresponds to "ordinary" stimulated Mandel-shtam-Brillouin scattering, which we shall not consider.

The change of ϵ in the sound wave can be represented in the form

$$\delta\varepsilon = \left(\frac{\partial\varepsilon}{\partial\rho}\right)_s \delta\rho(\mathbf{r}, t). \tag{16}$$

The linearized equation of hydrodynamics is

$$\frac{\partial^2\rho}{\partial t_2} = \nabla^2 p, \tag{17}$$

where p is the pressure. Neglecting the initial damping (we shall introduce it later), we write

$$\delta p = \left(\frac{\partial p}{\partial\rho}\right)_s \delta\rho + \left(\frac{\partial p}{\partial s}\right)_\rho \delta s; \tag{18}$$

here $(\partial p/\partial\rho)_s = v_{ac}^2$, and the change of entropy δs is connected with the absorbed power by the relation

$$\delta s = T^{-1}\int dt Q(\mathbf{r}, t), \tag{19}$$

where T is the absolute temperature. The equation for $\delta\rho(\mathbf{r}, t)$ is obtained by substituting (18) and (19) in (17):

$$\frac{\partial^2\rho}{\partial t^2} - v_{ac}^2\nabla^2\rho - R\frac{\partial}{\partial t}\nabla^2\rho = \frac{1}{T}\left(\frac{\partial p}{\partial s}\right)_\rho \nabla^2 \int dt Q(\mathbf{r}, t); \tag{20}$$

we have added here a term proportional to R, describing the absorption of sound (see, for example,[3], Sec. 5). It follows from (20) that the operator L has in this case the form

$$L = A\Omega_{MB}/[i\Omega(\Omega^2 - \Omega_{MB}^2 - iR\Omega q^2)], \tag{21}$$

where

$$A = \frac{1}{Tv_{ac}^2}\left(\frac{\partial p}{\partial s}\right)_\rho \left(\frac{\partial\varepsilon}{\partial\rho}\right)_s. \tag{22}$$

$$\Omega_{MB} = v_{ac}|\mathbf{q}|. \tag{23}$$

The effective excitation of the sum occurs at $|\Omega| \approx \Omega_{MB}$. Putting $\Omega = \pm\Omega_{MB} + \Delta$, we obtain from (21)

$$L \approx (A/2)(i\Delta + \Gamma)^{-1}; \tag{21a}$$

in this case the threshold power is equal to

$$I_L^{thr} = (2cn/\hbar\omega A)\frac{(\Delta^2 + \Gamma^2)}{\omega\Delta}. \tag{24}$$

The constant A can be transformed to more convenient variables, using the thermodynamic identities. First

$$\frac{1}{v_{ac}^2 T}\left(\frac{\partial p}{\partial s}\right)_\rho = -\frac{1}{\rho_0 c_p}\left(\frac{\partial\rho}{\partial T}\right)_p. \tag{25}$$

Further

$$\left(\frac{\partial\varepsilon}{\partial\rho}\right)_s = \left(\frac{\partial\varepsilon}{\partial\rho}\right)_T + \left(\frac{\partial\varepsilon}{\partial T}\right)_\rho \left(\frac{\partial T}{\partial\rho}\right)_s \tag{26a}$$

$$\left(\frac{\partial\rho}{\partial T}\right)_p^{-1}\left[\left(\frac{\partial\varepsilon}{\partial T}\right)_p-\frac{c_p}{c_v}\left(\frac{\partial\varepsilon}{\partial T}\right)_\rho\right]. \quad (26b)$$

In this case the two terms in (26a) describe the changes of the dielectric constant ϵ due respectively to the oscillations of the density and of the temperature in the sound wave. Taking (25) and (26) into account, we obtain

$$A=-\frac{1}{\rho_0 c_p}\left[\left(\frac{\partial\varepsilon}{\partial T}\right)_p-\frac{c_p}{c_v}\left(\frac{\partial\varepsilon}{\partial T}\right)_\rho\right]. \quad (27)$$

The approximate form of the dependence of the gain g on the frequency shift $\Omega = \omega_L - \omega_S$ is shown in Fig. 2a. It should be noted that unlike the ordinary SMBS (see Fig. 2b), for SSA-MB gain takes place both in the Stokes ($\Omega = +MB + \Delta$) and in the anti-Stokes ($\Omega = -\Omega_{MB} + \Delta$) regions. Another difference between SSA-MB and "ordinary" SMBS is that for SSA-MB the maximum of the gain occurs not at the center of the line ($\Delta = 0$), but at the point $|\Delta| = \Gamma_{MB}$. A clear explanation of this circumstance is that the thermal expansion and the electrostriction give rise to sound waves having a relative phase shift $\pi/2$. The phases of the corrections to the dielectric constants, necessitated by these two effects, are therefore different (for "ordinary" SMBS the quantity $\delta\epsilon$ is proportional to $i\Omega/(i\Delta + \Gamma)$ as against $1/(i\Delta + \Gamma)$ for SSA-MB).

For greater clarity, we have considered separately stimulated scattering by isobaric changes of ϵ, i.e., by thermal conductivity waves (see Sec. 1 of Ch. II), and stimulated scattering by adiabatic changes of ϵ, i.e., by acoustic waves. In a more rigorous approach, these changes must be taken into account simultaneously (see[1,2]). Naturally, the results of the rigorous analysis for $|\Omega| \ll \Omega_{MB}$ and for $|\Omega| \approx |\Omega_{MB}|$ coincide with those given above.

We note that recent papers[4] and [5] are devoted to concrete types of SSA. The authors of[4] consider the possibility of observing SSA by second-sound waves. In[5] is discussed the influence of the finite time of thermalization (conversion of absorbed energy into heat) on the course of the SSA processes.

3. SSA Following Excitation of Molecules

We now consider the SSA connected with the difference between the polarizabilities of the individual molecules in the ground state (a) and in the excited state (b).

This example is of interest to us also because we

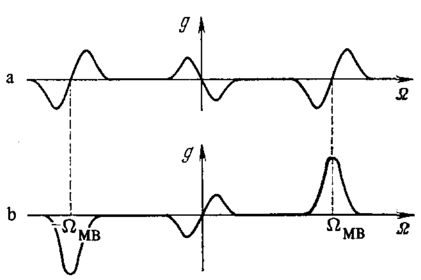

FIG. 2. Schematic form of the dependence of the signal gain g on the frequency shift $\Omega = \omega_L - \omega_S$ at $I_L \gg I_L^{thr}$: a) for SSA, b) for ordinary types of stimulated scattering. The simple components correspond to stimulated temperature scattering, and the lateral components to the Mandel'shtam-Brillouin doublet.

shall use an analogous model in Ch. III below, devoted to the quantum mechanical description of the SSA phenomenon. For the number n_b (cm^{-3}) of the excited molecules we can write the equation

$$\frac{\partial n_b}{\partial t}+\Gamma n_b=\mu Q(r, t)/\hbar\omega; \quad (28)$$

here the dimensionless parameter $\mu \ll 1$ describes the efficiency of excitation of the state (b) of interest to us upon absorption of a quantum, while the constant Γ describes the relaxation of the population from the level (b) to the level (a).

From (28) we obtain

$$L=\frac{\mu}{\hbar\omega}\frac{\partial\varepsilon}{\partial n_b}\frac{1}{i\Omega+\Gamma}, \quad (29)$$

$$I_L^{thr}=\frac{nc}{\mu}\left(\frac{\partial\varepsilon}{\partial n_b}\right)^{-1}\frac{\Gamma^2+\Omega^2}{\omega\Omega}. \quad (30)$$

As an approximate estimate we can take for $\partial\epsilon/\partial n_b$ an expression that follows from the Lorenz-Lorentz formula

$$\left(\frac{\partial\varepsilon}{\partial n_b}\right)=4\pi(\alpha_b-\alpha_a)\left(\frac{n^2+2}{3}\right)^2, \quad (31)$$

where α_a and α_b are the polarizability of the molecule in the states a and b respectively, and $n = \epsilon^{1/2}$ is the refractive index. Then (30) takes the form

$$I_L^{thr}=\frac{n}{\mu[(n^2+2)/3]^2}\frac{c}{4\pi(\alpha_b-\alpha_a)}\frac{\Gamma^2+\Omega^2}{\Omega\omega}. \quad (30a)$$

We note Mack[6] observed a stimulated scattering connected with absorption with picosecond pumping. The interpretation of this scattering, given in[6], is close to the scheme described above for SSA by molecules.

III. QUANTUM MODEL OF SSA

As was noted in the introduction (Ch. I) and illustrated by the concrete examples in Ch. II, the SSA phenomenon differs greatly in its properties from the "ordinary" processes of stimulated scattering. This pertains in particular to violation of the universal connection between the gain and the cross section for spontaneous scattering (formula (10)).

This means that the gain of the signal wave in the SSA phenomenon certainly cannot be due to simple two-photon scattering processes of the $n_L, n_S \to n_{L-1}, n_S + 1$ type (see Fig. 1), for in the latter case relation (10) must be satisfied (see Appendix 1).

On the other hand, for any process in which more than one quantum ω_L takes part, the square of the modulus of the matrix elements proportional to $n_L^2 \propto I_L^2$ and to higher powers of I_L, in contrast to the linear dependence $g(I_L)$ in (9). Nonetheless, as will be shown below, the SSA phenomenon with all its characteristic features finds a relatively simple explanation within the framework of the quantum-mechanical description of the processes of proton absorption and production.

In this chapter we present a quantum-mechanical analysis of the scattering of the fields ω_L and ω_S by an individual atom (molecule) and show that the SSA effect arises even for such a very simple system in the case when the frequencies ω_L and ω_S fall in the absorption line $a \to b$.

Let us consider the increments of the signal-field

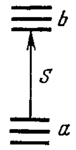

FIG. 3. Absorption of signal quantum in first order of perturbation theory. The molecule goes over from the state a to the state b.

energy due to the interaction with the molecule. We shall assume that prior to the turning on of the interaction the molecule is in the ground state a. In the absence of the field ω_L, the interaction reduces to absorption of the photon ω_S, described by the matrix element $M_{ab}^{(-)} = V_{ab}^{(1)} n_S^{1/2}$ (Fig. 3). In the presence of a strong field, there are added a number of processes, including:

1) the scattering $\omega_L \rightarrow \omega_S$ and $\omega_S \rightarrow \omega_L$ by the ground state of the molecule (see Fig. 1);

2) different multiphoton processes accompanied by a → b transitions of the molecule (including processes in which the photons ω_S are both absorbed and created).

Processes 1) will not be considered below, since they correspond to the "ordinary" types of scattering and have been thoroughly investigated; where necessary, we shall make use of published results. We shall focus attention on molecular transitions a → b corresponding to the absorption line, and ascertain the effect to which the presence of the strong field ω_L leads.

Processes 2) alter the matrix element $M_{ab}^{(-)}$ of signal-photon absorption, inasmuch as the transition (a, n_S) → (b, $n_S - 1$) can now be accompanied by virtual vanishing and creation of photons ω_L. We therefore have for the matrix element $M_{AB}^{(-)}$

$$M_{ab}^{(-)} = V_{ab}^{(1)} n_S^{1/2} + V_{ab}^{(3)} n_L n_S^{1/2} + \ldots, \quad (32)$$

where $V_{ab}^{(m)}$ is the matrix element of m-th order of perturbation theory; the quantities $n_L \pm 1$ have been replaced by n_L.

In addition, in the strong field ω_L, the molecular transition a → b can correspond not to vanishing but to creation of a photon ω_L. For example, one signal photon is produced and two ω_L photons are absorbed in the third-order process a, n_S, n_L → b, $n_S + 1$, $n_L - 2$.

The matrix element of the a → b transition accompanied by the production of a photon ω_S is

$$M_{ab}^{(+)} = U_{ab}^{(3)} n_L (n_S + 1)^{1/2} + \ldots \quad (33)$$

Examples of transitions corresponding to $V_{ab}^{(3)}$ and $U_{ab}^{(3)}$ are shown in Figs. 4 and 5.

The energy increment at the signal-field frequency ω_S is

$$Q_S = \hbar\omega (w^{(+)} - w^{(-)}), \quad (34)$$

where $w^{(\pm)} \propto |M_{ab}^{(\pm)}|^2$. Using (32) and (33) we obtain, accurate to terms of third order inclusive,

$$Q \sim -n_S \{ |V^{(1)}|^2 + 2n_L \operatorname{Re}(V^{(1)*}V^{(3)}) + n_L^2 [|V^{(3)}|^2 - |U^{(3)}|^2] \} + n_L^2 |U^{(3)}|^2. \quad (35)$$

The first correction for the strong field ω_L in (35), proportional to $n_L n_S$, is given by the interference of the terms of the first and third orders. The correction term proportional to $n_L^2 n_S$ contains the difference $|V^{(3)}|^2 - |U^{(3)}|^2$ and, as will be shown below, does not play an important role in the model under consideration.

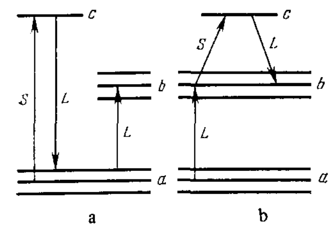

FIG. 4. Absorption of a signal quantum in third-order perturbation theory (in this case the laser quantum is virtually absorbed and emitted).

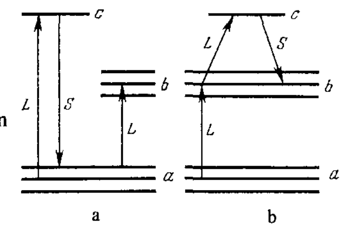

FIG. 5. Emission of a signal quantum in third-order perturbation theory with absorption of two laser quanta.

Depending on the sign, the interference term can lead to either enhancement or to weakening of the absorption. At sufficiently large n_L, the absorption can give way to amplification, corresponding to the SSA phenomenon considered in the introduction (Ch. I). Indeed, this gain, which is linear in n_L and proportional to the cross section of the ordinary absorption, has no corresponding spontaneous scattering; the spontaneous production of the photons ω_S (at $n_S = 0$) is proportional here to n_L^2.

Thus, the SSA process can be described in the language of photon emission and absorption as a change of absorption as a result of interference of terms of first and third orders of perturbation theory (see Figs. 3 and 4). We recall that the usual types of SS (and the associated ordinary types of spontaneous scattering) are described by the square of a second order term (see Fig. 1).

Before we proceed to further discussions, let us obtain concrete formulas describing the processes in question. Let the molecule have two continuous groups of levels a and b with a density of states

$$\frac{dN_a}{dE} = \frac{\Gamma_a}{\pi\hbar \{[(E - E_a)^2/\hbar^2] + \Gamma_a^2\}}, \quad \frac{dN_b}{dE} = \frac{\Gamma_b}{\pi\hbar \{[(E - E_b)^2/\hbar^2] + \Gamma_b^2\}}. \quad (36)$$

We assume that the absorption in the a → b transition is the result of a sufficiently weak forbidden transition, and that the polarizability of the molecule in the states a and b is determined mainly by the levels c, which are dipole-coupled to levels a and b. Under these assumptions, the contribution of the a → b transition to the polarizabilities α_a and α_b is small: $|\delta\alpha\ (a \longleftrightarrow b) \ll |\alpha_a|\alpha_b|$, and the quantities α_a and α_b are real. We shall henceforth assume also that the average frequency of the a → b transition coincides with the frequency of the laser field: $(E_b - E_a)/\hbar = \omega_L$.

Let the signal field contain n_S quanta and the laser field n_L quanta, $n_L \gg 1$. It is convenient to express n_L in the final formulas in terms of $|E_L|^2$.

Under the foregoing assumptions, it can be readily shown that the absorption in the aforementioned absorption and emission matrix elements, which correspond to transitions from the initial state a with energy $E_1 = E_a + \hbar\nu$, are equal to

$$M_{ab}^{(-)} = M_{ab}^{(-)(1)} + M_{ab}^{(-)(3)} = H\,(n_S)^{1/2}\left[1 - \frac{\alpha_a\,|E_L|^2}{4\hbar\,(\nu - \Omega + i\Gamma_a)} - \frac{\alpha_b\,|E_L|^2}{4\hbar\,(\nu + i\Gamma_b)}\right], \tag{37}$$

$$M_{ab}^{(+)} = M_{ab}^{(+)(3)} = H\,(n_S + 1)^{1/2}\left[-\frac{\alpha_a\,|E_L|^2}{4\hbar\,(\nu + \Omega + i\Gamma_a)} - \frac{\alpha_b\,|E_L|^2}{4\hbar\,(\nu + i\Gamma_b)}\right], \tag{38}$$

where the first-order matrix element H is equal to

$$H = m_{ab}\,(8\pi\hbar\omega/V)^{1/2}/2. \tag{39}$$

Calculating further the probabilities of these transitions with the aid of the standard formulas of perturbation theory and averaging them over the distribution (36) for the initial state a, we get

$$w^{(\pm)} = \int \frac{dN_a}{dE_1}\,dE_1\,\frac{dN_b}{dE_2}\,dE_2\,|M^{(\pm)}|^2\,\frac{2\pi}{\hbar^2}\,\delta(\nu \mp \Omega - \mu), \tag{40}$$

where $E_1 = E_a + \hbar\nu$ and $E_2 = E_b + \hbar\mu$. Substituting (37) and (38) in (34), we obtain an expression for the transfer of energy to the signal field. We write this expression in the form similar to (35):

$$\begin{aligned} Q_S = -\frac{2\pi\omega H^2}{\hbar}\,\frac{\Gamma}{\pi}\,\frac{1}{\Omega^2 + \Gamma^2}\,n_S \Big\{ 1 &+ \frac{|E_L|^2}{2\hbar}\left[\frac{\alpha_a\Omega\Gamma_b\,(\Gamma_b + 3\Gamma_a)}{(\Omega^2 + 4\Gamma_a^2)\,\Gamma^2} - \frac{\alpha_b\Omega\Gamma_a\,(\Gamma_a + 3\Gamma_b)}{(\Omega^2 + 4\Gamma_b^2)\,\Gamma^2}\right] \\ &+ |E_L|^4 F(\Omega) - |E_L|^4 F(\Omega) \Big\} + \frac{2\pi\omega H^2}{\hbar}\,\frac{\Gamma}{\pi}\,\frac{1}{\Omega^2 + \Gamma^2}\,|E_L|^4 F(\Omega); \end{aligned} \tag{41}$$

we have introduced here the absorption line width for the a → b transition $\Gamma = \Gamma_a + \Gamma_b$, and $F(\Omega)$ is a function of the frequency, but its explicit form will not be needed.

Expression (41) contains two parts: one proportional to n_S and responsible for the stimulated transition, and one independent of n_S namely the spontaneous noise. The first of the terms proportional to n_S corresponds to the ordinary absorption (the term $|V^{(1)}|^2$ from (35)). The second term determines the contribution of the interference of the matrix elements with the first and third orders; depending on the parameters that enter in this term, it may turn out to be either positive or negative. The term proportional to $n_S|E_L|^4$, corresponding to the difference $|V^{(3)}|^2 - |U^{(3)}|^2$ from (35), vanishes.

Such an exact compensation in the model in question is connected with the concrete choice $\hbar\omega_L = E_b - E_a$.

As already mentioned, the gain connected with the SSA arises when the interference term in (41) becomes larger than the first term. Simultaneously, the term $|M_3^{(-)}|^2$ is also of the order of the first two. This term, however, is completely cancelled out in (41) by the square of the matrix element $|M_3^{(+)}|^2$, which is responsible for the photon production process.

Thus, the sign and magnitude of the gain are determined only by the ratio of the first and interference terms in the absorption probability, terms proportional to n_S but not to $n_S + 1$, as would be the case for the production probability. This indeed corresponds to the violation of the universal connection between the spontaneous scattering (the term "1" in $(n_S + 1)$) and the gain. All that is left of the matrix element for the production of the photon ω_S in (41) is the last "spontaneous" term, which is furthermore proportional to $|E_L|^4 \sim I_L^2$. The spontaneous noise will be discussed in greater detail in the next chapter.

Let us determine the threshold intensity of the SSA phenomenon. Assuming, for simplification of the formulas, that $\Gamma_a = \Gamma_b$, we get from (41)

$$I_L^{\text{thr}} = \frac{c}{4\pi}\,\frac{\Omega^2 + \Gamma^2}{\omega\Omega}\,\frac{1}{\alpha_b - \alpha_a}. \tag{42}$$

Formula (42) coincides with the result (30a) obtained for SSA on molecules in the semiclassical analysis, if Γ is taken to mean the relaxation constant*.

In concluding this chapter, let us discuss the relation between the SSA effect and other strong-field effects discussed many times in the literature.

It is well known that in the presence of a strong field ω_L that is resonant with one of the transitions of the molecule, the interaction of the atom with other (weak) fields changes in a radical manner: the spectral lines are split, amplification becomes possible in those regions where initially absorption takes place, etc. All these effects, however, are essentially connected with the saturation of the resonant transition and occur at intensities

$$I_L \gg I_{\text{sat}} \sim \frac{1}{\sigma T_1} \sim \frac{c}{\alpha''\omega T_1}, \tag{43}$$

where α'' is the imaginary part of the polarizability of the frequency $\omega = \omega_{ab}$, $\sigma \sim \alpha''\omega/c$ is the absorption cross section, and $T_1 = \Gamma^{-1}$ and is the time of longitudinal relaxation. A description of these effects is based on the solution of the problem of the mixing of states of a two-level system in a strong field.

For the SSA phenomenon, to the contrary, it is important that the strong field mixes into the levels a and b all the other levels of the system (the levels c from Figs. 4 and 5). For this reason, the characteristic intensities corresponding to SSA are determined by other parameters:

$$I_L \gg I_{\text{SSA}}^{\text{thr}} \sim \frac{c}{\omega T_1}\,\frac{1}{|\alpha_a - \alpha_b|}. \tag{44}$$

If the a → b transition is sufficiently weak (i.e., $\alpha''_{ab} \ll |\alpha_a - \alpha'_b|$), then the SSA threshold can be much smaller than the saturation intensity (43).

Thus, the separation of this effect from all the remaining strong-field effects is perfectly justified physically. For this reason, in a derivation of the concrete formulas (37) and (38) above, we did not take into account the contribution of a large number of transition amplitudes not connected specifically with the SSA effect, of the type shown in Fig. 6.

For the same reason, we could confine ourselves in the foregoing analysis only to terms up to third order inclusive. If we neglect the spontaneous noise and describe the fields E_S and E_L classically, then the SSA theory can be developed in a form analogous to the theory of other effects of strong fields. Namely, it is sufficient to write for the density matrix of the mole-

*In the semiclassical analysis it is precisely the longitudinal-relaxation constant which is important, and the line width is assumed there to be large and has no influence whatever on the threshold. Such a disparity between the results of this chapter and the quasiclassical model is connected with the fact that we use an oversimplified quantum model. For our purposes it was sufficient to demonstrate the existence of the SSA effect.

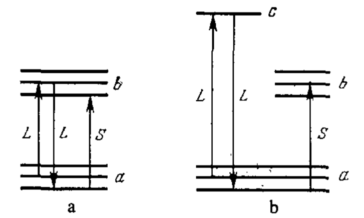

FIG. 6. Examples of transitions whose contributions are either insignificant (a) or have no bearing on the SSA phenonenon (b).

cule the standard two-level approximation equations cf. Eq. (28) of Ch. II). On the other hand, for the dipole moment, in place of the usual expression

$$\langle d\rangle = \mathrm{Sp}\,(\hat{d}\,\hat{\rho}) = d_{ab}\rho_{ba} + d_{ba}\rho_{ab} \tag{45}$$

it is necessary to assume

$$\langle d\rangle = [\alpha_a\rho_{aa}(t) + \alpha_b\rho_{bb}(t)]\,E(t). \tag{46}$$

Whereas (45) describes mixing of states a and b, expression (46), corresponding to SSA, contains the polarizabilities, connected with the presence of other levels c, of the states a and b.

IV. SPONTANEOUS PROCESS CORRESPONDING TO SSA

From the quantum model of the SSA (see Ch. III, formula (41)), it follows that Q_S does not vanish when $n_S = 0$. Thus, the SSA process can be set in correspondence to the certain spontaneous scattering process. This process differs essentially from the ordinary types of spontaneous scattering, primarily in the fact that the intensity of the scattered light is proportional not to the intensity of the pump, but to its square. In spite of this, we shall use the term "spontaneous scattering." As will be shown below, this scattering has the simple physical meaning which does not depend on the concrete model of Ch. III.

Within the framework of the semiclassical description, the indicated spontaneous scattering should be connected with fluctuations $\delta\epsilon$ arising in the absence of a signal wave. In the presence of absorption of a powerful laser wave, there is always at least one source of such fluctuations—the analog of the shot noise connected with the discrete character of the absorption of energy from the field (the energy is absorbed only in batches $\hbar\omega$). Such a noise can be taken into account by substituting in the right side of (2) $Q(\mathbf{r}, t)$ in the form

$$Q(\mathbf{r}, t) = \langle Q(\mathbf{r}, t)\rangle + (\hbar\omega\langle Q(\mathbf{r}, t)\rangle)^{1/2} f(\mathbf{r}, t), \tag{47}$$

where the random function $f(\mathbf{r}, t)$ is characterized by the properties*

$$\langle f(\mathbf{r}, t)\rangle = 0, \quad \langle f(\mathbf{r}', t')\, f(\mathbf{r}'', t'')\rangle = \delta^{(3)}(\mathbf{r}'-\mathbf{r}'')\,\delta(t'-t''). \tag{48}$$

By virtue of (2), such a random energy release leads to fluctuations of $\delta\epsilon$ with $\langle\delta\epsilon(\mathbf{r}, t)\rangle = 0$ and

$$\langle\delta\varepsilon(\mathbf{r}', t')\,\delta\varepsilon(\mathbf{r}'', t'')\rangle = [\beta P_L\hbar\omega/(2\pi)^4]\int d^3\mathbf{q}\,d\Omega\,\exp(i\mathbf{q}\mathbf{r} + i\Omega t)\,|L(i\Omega, i\mathbf{q})|^2, \tag{49}$$

where

$$\mathbf{r} = \mathbf{r}' - \mathbf{r}'', \quad t = t' = t'', \quad P_L = I_L\hbar\omega.$$

The scattering of a monochromatic laser wave by fluctuations of the type (49) is described by a scattering cross section per unit volume

$$dR/do\,d\omega = (1/2^5\pi^3)(\omega/c)^4\,|\mathbf{e}_S\mathbf{e}_L^*|^2\,\beta P_L\hbar\omega\,|L(i\Omega, i\mathbf{q})|^2. \tag{50}$$

Since $dR/dod\omega \sim P_L$, the power of the scattered light is proportional to P_L^2 in accord with the statement made above.

Interest attaches to the value of the spontaneous noise, expressed in terms of the number of noise quanta λ per mode (see Appendices 1 and 2). Using formulas (1.3), (6), and (42), we can easily get

$$\lambda = \frac{1}{4}\,\frac{x^2}{x-1}\,\frac{|L|^2}{(\mathrm{Im}\,L)^2}\,\frac{1}{|\mathbf{e}_S\mathbf{e}_L|^2}, \quad x = \frac{I_L}{I_L^{\mathrm{thr}}}. \tag{51}$$

We have taken here a case when, generally speaking $|\mathbf{e}_S\cdot\mathbf{e}_L^*| = 1$. It is interesting to note that λ does not depend on the temperature of the medium and is determined only by the parameters x and $|(\mathrm{Im}\,L)/L|$.

The minimum of λ as a function of the power I_L is reached at double the threshold $(x = 2)$, and is equal to (at $|\mathbf{e}_S\cdot\mathbf{e}_L| = 1$)

$$\lambda(x=2) = |L|^2/(\mathrm{Im}\,L)^2. \tag{52}$$

In all the examples considered by us (see Ch. II)

$$L \sim [\Gamma + i(\Omega - \Omega_1)]^{-1}.$$

The maximum gain is realized in this case at $|\Omega - \Omega_1| = \Gamma$, corresponding to $\lambda = 2$. When $|\Omega - \Omega_1| \gg \Gamma$, the gain decreases and the noise tends to the maximum value $\lambda_{\min} = 1$. The latter circumstance demonstrates in addition that the semiclassical approach used above apparently reflects correctly the main properties of the phenomenon. In any case, it does not lead to a contradiction to the known (see Appendix 2) quantum-mechanical noise minimum of "one photon per mode." By way of an illustration of the general formula (50), let us consider the concrete case of SSA on the thermal conductivity. Using formulas (14) and (15), we obtain

$$\frac{dR}{do\,d\omega} = \frac{\Gamma}{\pi}\,\frac{1}{\Omega^2+\Gamma^2}\left[\frac{1}{\rho_0 c_p}\left(\frac{\partial\varepsilon}{\partial T}\right)_p\right]^2\frac{\hbar\omega\beta P_L}{2\Gamma(4\pi)^2}\left(\frac{\omega}{c}\right)^4|\mathbf{e}_S\mathbf{e}_L^*|^2. \tag{53}$$

As seen from (53),

$$dR/do \equiv \int d\omega\cdot dR/dod\omega \sim \Gamma^{-1},$$

i.e., dR do is proportional to the characteristic time $\tau_0 = \Gamma^{-1}$.

The spontaneous process described by formula (50) is, in essence, a nonlinear inertial scattering. By inertia we mean here the fact that for incident light with characteristic intensity-variation times $\tau < \Gamma^{-1}$, the intensity of the scattered light $I_{sc}(t)$ depends on the entire prior behavior of $I_L(t')$ at $0 \leq -t' \lesssim \Gamma^{-1}$. In other words, in the scattering there become manifest fluctuations of the numbers of the absorbed quanta accumulated during the time $\sim\tau_0 = \Gamma^{-1}$.

If only a fraction of the energy absorbed in each act goes to heat release, then the cross section from (53) must be multiplied by μ^2.

In a real situation, the spontaneous nonlinear scat-

*The concrete value of the coefficient f(r, t) can be obtained, for example, in the following manner. The number of quanta absorbed in the volume V during a time T is equal to $N = \iint d^3r\,dt\,Q(r, t)(\hbar\omega)^{-1}$. By stipulating the satisfaction of Poisson's law $\langle N^2\rangle - \langle N\rangle^2 = \langle N\rangle$, we obtain expression (47).

tering (53), as a rule, is small compared with the linear scattering by ordinary thermal fluctuations of the quantity ϵ. We present for comparison the cross section of the linear scattering in the region of the central component of the Rayleigh line:

$$\frac{dR}{do\,d\omega}=\frac{\Gamma}{\pi}\frac{1}{\Omega^2+\Gamma^2}\left(\frac{\partial\varepsilon}{\partial T}\right)_p^2\frac{T_0^2 k}{c_p\rho_0}\frac{1}{(4\pi)^2}\left(\frac{\omega}{c}\right)^4|\mathbf{e}_L\mathbf{e}_S^*|^2, \tag{54}$$

where T_0 is the absolute temperature and k is Boltzmann's constant.

It is easy to see that the cross sections (53) and (54) become comparable at a pump intensity P_L equal to

$$P_L=2\Gamma\rho_0 c_p T_0^2 k/\hbar\omega\beta\mu^2. \tag{55}$$

This intensity is quite large, making it difficult to observe the nonlinear spontaneous scattering. In particular, as shown by elementary estimates with the aid of formula (55), difficulties with overheating of the medium are unavoidable when working with such intensities.

In this connection, a characteristic situation is one in which the noise us due to the ordinary spontaneous scattering (54) even in those cases when the decisive role in the amplification is played by the SSA process. The effective factor λ_{eff} in this case is larger than given by (51), but smaller than $\lambda(SS) = kT/\hbar\Omega$ (as would be the case for ordinary SS at $kT \gg \hbar\Omega$). Namely, from formula (1.3) of Appendix 1 we obtain

$$\lambda_{\text{eff}}\,(\text{SSA})\sim\lambda\,(\text{SS})\,I_{\text{thr}}\,(\text{SSA})/I_{\text{thr}}\,(\text{SS})\ll\lambda\,(\text{SS}). \tag{56}$$

The spontaneous process corresponding to SSA on acoustic waves (see Sec. 2 of Ch. II) is described in perfect analogy with the spontaneous process described above, corresponding to SSA on thermal waves; the same holds also for the remark on the role of ordinary linear scattering.

The noise source considered above (see formula (47)) in the general case is not at all unique. In various concrete situations, other types of sources may be added to it. This is seen, in particular, with SSA by molecules as an example. It can be shown that the corresponding spontaneous processes analogous to the ordinary Rayleigh scattering by a gas of molecules with effective polarizability $\alpha_{eff} = \alpha_b - \alpha_a$ and density $\bar{n}_b = \mu\beta I_L/\Gamma$. The corresponding extinction coefficient is

$$dR/do\,d\omega=\bar{n}_b\,d\sigma_{\text{рэл}}/do\,d\omega=$$
$$=\left(\frac{\Gamma}{\pi}\right)(\Omega^2+\Gamma^2)^{-1}\beta I_L\mu\Gamma^{-1}(\alpha_b-\alpha_a)^2\left(\frac{\omega}{c}\right)^4|\mathbf{e}_S\mathbf{e}_L^*|^2\,|(n^2+2)/3|^4. \tag{57}$$

The last factor in (57) corresponds to allowance for the acting field in accordance with the Lorenz-Lorentz formula. The calculation of the equivalent noise for the SSA on molecules by formula (1.3) of Appendix 1 yields

$$\lambda=\frac{2}{\mu}\frac{1}{4}\frac{x^2}{x-1}\frac{\Omega^2+\Gamma^2}{\Omega^2}\frac{1}{|\mathbf{e}_S\mathbf{e}_L^*|^2}, \tag{58}$$

which is $2/\mu$ times larger than the value obtained from the general formula (51). The deviation of $|\mathbf{e}_S\cdot\mathbf{e}_L^*|$ and of μ from unity leads to additional fluctuations, which by analogy with shot noise can be called separation noise (cf.[7]). For an estimate of the minimal noise we put $\mu = 1$ and $|\mathbf{e}_S\cdot\mathbf{e}_L| = 1$. In this case the physical cause of the doubling of the noise in (58) compared with the general expression (51) is as follows.

In the case of SSA on molecules, not only is the process of molecule excitation discrete (and introduces by the same token fluctuations), but so is also the process of their de-excitation. From the formal point of view this means that the kinetic operator L from (28) contains itself an additional noise part. It is easy to see that both indicated sources of fluctuations give identical contributions to $\langle\delta\epsilon^2\rangle$, and this leads to a doubling of the noise. On the other hand, in the case of SSA on thermal and acoustic waves, the buildup of fluctuations is not accompanied by additional noise.

APPENDICES

1. AMPLIFICATION NOISE AND CONNECTION BETWEEN THE CROSS SECTION OF THE SPONTANEOUS SCATTERING AND THE GAIN IN SS

As is well known, in the absence of an input signal, the process of stimulated scattering can be regarded as a result of amplification of the amplifier noise. This noise corresponds to spontaneous scattering of the incident pump wave. The relation between the induced and spontaneous parts of the signal-energy increment, for ordinary types of scattering, can be obtained from general principles. The gain for ordinary types of SS is therefore uniquely expressed in terms of the cross section of the spontaneous scattering and the temperature of the medium. We present the derivation of the corresponding formulas.

Assume that a "signal" wave propagates in the direction of the z axis. The intensity of the signal can be characterized by the flux of quanta I_S (quanta/cm^{2}sec); we introduce also the average occupation number n_S of the quantum states of the signal field. Then

$$I_S=n_S v_S\Delta N=n_S v_S k^2\Delta k\Delta o/(2\pi)^3=n_S\,[k^2/(2\pi)^3]\Delta\omega\Delta o; \tag{1.1}$$

here $v_S = \partial\omega_S/\partial k$ is the group velocity of the signal wave, and $\Delta N = k^2\Delta k\Delta o/(2\pi)^3$ is the number of modes per unit volume in the solid angle Δo of interest to us and in the frequency interval $\Delta\omega = v_S\Delta k$. In a real situation, the values of Δo and $\Delta\omega$ are determined usually by the geometry of the experiment and by the bandwidth of the amplifier respectively; we do not need them in explicit form. Moreover, in considering a coherent signal wave (i.e., neglecting noise), we can put $\Delta o \to 0$ and $\Delta\omega \to 0$.

The variation of the flux I_S with the coordinate z is connected with the action of three mechanisms: 1) a decrease due to the ordinary linear absorption, 2) an increase due to the spontaneous scattering, and 3) amplification connected with the SS. Accordingly, we can write

$$dI_S/dz=-\beta I_S+\Delta R I_L+g_0 I_S=g\,(I_S+g^{-1}\Delta R I_L). \tag{1.2}$$

We have introduced here the resultant gain (with allowance for losses) $g = g_0 - \beta$. The quantity $\Delta R = (dR/do\,d\omega)\Delta o\Delta\omega$ is the extinction coefficient (i.e., the cross section for scattering by a unit volume), corresponding to spontaneous scattering in a solid angle Δo and a frequency interval $\Delta\omega$ and in a definite type of polarization (the dimension of ΔR is cm^{-1}). An expression of the type (1.2) is valid for all types of SS;

it is convenient to rewrite it in terms of n_S:

$$dn_S/dz = g(n_S+\lambda), \quad \lambda = (I_L/g)(2\pi)^3 k^{-2}\, dR/do\, d\omega. \tag{1.3}$$

Here λ is the equivalent number of noise quanta per mode; the corresponding spectral intensity of the noise power per mode with a definite transverse index is $\hbar\omega\lambda$, and the minimum value of λ compatible with the principles of quantum mechanics is $\lambda_{min} \approx 1$ (see Appendix 2).

Let us consider the usual types of SS, not connected with absorption, and neglect first the linear absorption. In the language of quantum mechanics, such types of scattering are described by the schemes of Fig. 1a. The probability of emission of a signal quantum, corresponding to such a scheme, is proportional to $w^{(+)} \sim \rho_1(n_S+1)n_L$, where ρ_1 is the population of level 1 at equilibrium. The probability of absorption of a signal quantum is $w^{(-)} \sim \rho_2 n_S(n_L+1)$ (see Fig. 1b); when $n_L \gg 1$, we can approximately put $n_L \approx n_L + 1 \sim I_L$. As a result, the energy transferred to the signal wave is proportional to

$$dn_S/dz \sim w^{(+)} - w^{(-)} \sim I_L[n_S + \rho_1(\rho_1-\rho_2)^{-1}]. \tag{1.4}$$

For an equilibrium medium, however, $\rho_1/(\rho_1-\rho_2) = \nu + 1 = [1-\exp(-\hbar\Omega/kT)]$, where $\nu = [\exp(\hbar\Omega/kT) - 1]^{-1}$ is the average number of "quanta" of frequency $\Omega = \omega_L - \omega_S$ at a temperature T. Comparing (1.4) with (1.3) we obtain for g_0

$$g_0 = I_L \frac{(2\pi)^3}{k^2}\frac{dR}{do\, d\omega}\left[1-\exp\left(-\frac{\hbar\Omega}{kT}\right)\right]. \tag{1.5}$$

Allowance for the linear absorption leads to the conclusion that amplification is possible only when $I_L > I_L^{thr}$, where the threshold pump power is determined by the condition $g_0(I_L^{thr}) = \beta$:

$$I_L^{thr} = [k^2/(2\pi)^3]\,\beta\,[1-\exp(-\hbar\Omega/kT)]^{-1}(dR/do\, d\omega)^{-1}. \tag{1.6}$$

The value of λ, from (1.3), is then

$$\lambda = (\nu+1)\frac{x}{x-1} = \left[1-\exp\left(-\frac{\hbar\Omega}{kT}\right)\right]^{-1}\frac{x}{x-1}, \quad x = \frac{I_L}{I_L^{thr}}. \tag{1.7}$$

From (1.5) and (1.6) there follow a number of important consequences for the ordinary types of SS. First, inasmuch as $\nu + 1 > 0$ only if $\Omega > 0$, amplification ($g_0 > 0$) is possible only in the Stokes region ($\Omega > 0$). Further, when $I_L \gg I_L^{thr}$ the amplification noise is determined only by the temperature of the medium and reaches at low temperature ($kT \ll \hbar\Omega$) the quantum minimum $\lambda = 1$. We note that on approaching the threshold the spontaneous scattering itself does not experience any singularity; only λ, which characterizes the properties of the amplifier, tends to infinity. Finally, for ordinary types of SS, the value of λ decreases monotonically with increasing $x = I_L/I_L^{thr}$.

2. DESCRIPTION OF NOISE IN QUANTUM MECHANICS. QUANTUM MINIMUM OF AMPLIFICATION NOISE

We present for reference purposes the main premises of the description of noise in quantum mechanics of a mode of an electromagnetic field. Two approaches are possible here. One of them corresponds to the Schrödinger picture of quantum mechanics and was considered in[8]. In the present appendix we present a description of the Heisenberg picture, following the fundamental papers of Senitzky[9] and the book of Louisell[10]. This picture is convenient for a comparison with the classical Maxwell's equations.

The equation for the Heisenberg annihilation operator $\hat{a}(t)$ in the presence of linear damping is of the form

$$(d\hat{a}(t)/dt) + (\gamma/2)\hat{a}(t) = \hat{f}(t). \tag{2.1}$$

It is assumed here that the fast dependence on the time ($\sim\exp(-i\omega t)$) has already been separated, and γ is the damping constant. The operator $\hat{f}(t)$ describes a random Langevin force and corresponds to an aggregate of quantum and thermal noises connected with the damping.

We shall show first that neglect of the noises leads to a contradiction. In fact, if $\hat{f}(t) \equiv 0$, then $\hat{a}(t) = \hat{a}(0)\exp(-\gamma t/2)$, and we would obtain for the commutator

$$[\hat{a}(t), \hat{a}^+(t)] \overset{?}{=} [\hat{a}(0), \hat{a}^+(0)]\exp(-\gamma t). \tag{2.2}$$

But in the correct theory the commutator of $\hat{a}$ and $\hat{a}^+$ should be equal to unity at any instant of time t. We see therefore that the noise cannot be neglected even at zero temperature of the dissipative subsystem (thermostat).

It follows from (2.1) that

$$\hat{a}(t) = \hat{a}(0)\exp(-\gamma t/2) + \int_0^t dt'\hat{f}(t')\exp[-\gamma(t-t')/2]. \tag{2.3}$$

If the dissipative subsystem has a broad spectrum, then f(t) can be regarded as a δ-correlated random operator process. This means that

$$\langle\hat{f}(t)\rangle = 0,$$
$$\langle\hat{f}(t_1)\hat{f}^+(t_2)\rangle = A\delta(t_1-t_2), \quad \langle\hat{f}^+(t_2)\hat{f}(t_1)\rangle = B\delta(t_1-t_2); \tag{2.4}$$

it is easy to see that in this case $A \geq 0$ and $B \geq 0$. Let us assume that the commutators $\hat{f}$ and $\hat{f}^+$ are c-numbers (and not an operator), i.e., that

$$[\hat{f}(t_1), \hat{f}^+(t_2)] = C\delta(t_1-t_2), \quad C = A - B. \tag{2.5}$$

If we now stipulate that the commutator $[\hat{a}(t)\hat{a}^+(t)]$ be equal to unity at all t, then we get from (2.3) and (2.5)

$$[\hat{a}(t), \hat{a}^+(t)] = \exp(-\gamma t) + C\gamma^{-1}[1-\exp(-\gamma t)] = 1,$$

from which it follows that

$$C = \gamma. \tag{26}$$

An analogous relation can be obtained by considering the average number of quanta $\langle\hat{n}(t)\rangle = \langle\hat{a}^+(t)\hat{a}(t)\rangle$:

$$\langle\hat{n}(t)\rangle = \langle\hat{a}^+(t)\hat{a}(t)\rangle:$$
$$\langle\hat{n}(t)\rangle = \langle\hat{n}(0)\rangle\exp(-\gamma t) + B\gamma^{-1}[1-\exp(-\gamma t)]. \tag{2.7}$$

If at $t = 0$ the mode was in equilibrium with the thermostat at a temperature T, then $\langle\hat{n}(0)\rangle = \nu \equiv [\exp(\hbar\omega_0/kT) - 1]^{-1}$ and equilibrium should be retained also at $t > 0$. Hence

$$B = \gamma\nu \quad A = B + C = \gamma(\nu+1). \tag{2.8}$$

Equations (2.4)–(2.6) and (2.8) constitute the simplest formulation of the fluctuation-dissipation theorem in the quantum case: the noise intensity coefficients A, B, and C are proportional to the damping constant γ.

Attention must be called to the characteristic features inherent in the quantum description of the noise:

1) Even at zero temperature of the thermostat ($T = 0$, $\nu = 0$) there is quantum noise: $A = C = \gamma$, $B = 0$.

2) It is necessary to observe rigorously the order of the operators in the expressions of the type (2.4) for the averages of the products of the random forces; namely, the coefficient B corresponds to "normally ordered" noise, and the coefficient A corresponds to "antinormally ordered" noise, with $A \neq B$.

The field energy (the number of quanta in the mode is determined by the normally-ordered expression $\langle \hat{n} \rangle = \langle \hat{a}^+ \hat{a} \rangle$. At zero temperature ($\nu = 0$) the intensity B of the "normally-ordered" i.e., energy part of the noise is equal to zero: $B = \gamma\nu = 0$. We can therefore say that at $\nu = 0$ the quantum noise does not excite real oscillations of the mode, and serves only to maintain the operator $[a, a^+]$ constant (or, what is approximately the same, to maintain the vacuum fluctuations of the quantities $\hat{p}$ and $\hat{x}$).

If the dissipative subsystem (thermostat) is not in the equilibrium state, then in principle amplification is also possible, i.e., $\gamma < 0$. Such a non-equilibrium state may be connected with population inversion or, in our case, with the propagation of a powerful pump beam in the medium. From the conservation of the commutation relations we get as before $C = \gamma$, but now $C < 0$. Since $B = A - C$ and $A \geq 0$, we obtain immediately the minimum value of the energy, i.e., "normally-ordered" noise of a quantum amplifier:

$$B \geqslant |\gamma| \quad \text{if} \quad \gamma < 0. \tag{2.9}$$

From (2.7) we find that at the minimum B

$$d\langle \hat{n}(t)\rangle/dt = +|\gamma|(\langle \hat{n}(t)\rangle + B|\gamma|) = |\gamma|(\langle \hat{n}\rangle + 1). \tag{2.10}$$

This means that the minimum noise of a quantum amplifier corresponds formally to an amplification of the noise energy of one photon per mode. The flux of the noise power corresponding to this photon is equal to

$$P_n = v_{gr}(\hbar\omega/L)\,\Delta N = v_{gr}(\hbar\omega/L)\,L\Delta k/2\pi = \hbar\omega\Delta\omega/2\pi = \hbar\omega\Delta f. \tag{2.11}$$

We have considered here one-dimensional propagation with group velocity $v_{gr} = \partial\omega/\partial k$ for modes with definite transverse index; Δf is the bandwidth. We note that this value of P_n is twice as large than the so-called power of the vacuum fluctuations $\hbar\omega\Delta f/2$.

More rigorous proofs of this conclusion for arbitrary quantum amplifiers are given in[11] (on the basis of the Heisenberg-equation method) and in[12] (on the basis of the method of quantum characteristic functions).

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