STIMULATED EMISSION OF RADIATION IN A STRONG ELECTROMAGNETIC FIELD

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Stimulated emission of radiation of an atom (molecule) placed in a strong field is calculated by taking into account saturation, the Doppler effect and field fluctuations in the incident light wave. It is shown that the amplification mode in a medium with negative absorption and the character of the spectral narrowing may depend on the concentration of the active centers. The existence of a narrowing limit connected with saturation is considered.

THE intensity of stimulated emission is proportional to the intensity of the incident light only in the linear approximation, when the stimulating field is sufficiently weak. In a strong electromagnetic field, the level populations change and this leads to a redistribution of the intensity of the stimulated emission and to a broadening of the spectrum of the amplified frequencies (saturation effect, see, for example [1]). The question arises of how the saturation influences the operation of quantum amplifiers and generators, and whether saturation leads to the existence of some limit to the narrowing of the spectral lines.

In order to answer these questions to some degree of approximation, we solve below the following problem. An atom (molecule)[1)] in the excited state is introduced into a strong electromagnetic field (traveling wave) at a certain instant of time. The frequency of the atomic transition is close to the fundamental frequency of the field, and both these frequencies are much larger than the frequency shifts due to the Doppler effect, the line width connected with the damping, and the broadening due to saturation. The transition scheme is shown in Fig. 1. The spectral composition of the stimulated emission is calculated. In order to obtain the light amplification coefficient in an active medium consisting of a set of such atoms, the result obtained is averaged over the time of formation of the excited state t_0 and over the velocity of the atoms (allowance for the Doppler effect). The spontaneous emission does not exert any influence on the field growth mechanism and on the line narrowing and is therefore not calculated in the present paper. It is assumed that the upper and lower states in the atom attenuate with equal rate (equal to $\Gamma/2$).

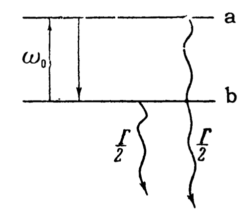

FIG. 1. Two-level quantum mechanical system. The wavy arrows denote transitions to all the lower levels (damping).

We first establish more accurately the limits of applicability of the semi-classical calculation method and discuss the role played by fluctuations of the incident field due to the shot effect; we then formulate the equations of the problem and analyze individual cases that have a bearing on experimentally realized laser schemes.

SEMICLASSICAL METHOD

The stimulated emission should, strictly speaking, be calculated using the formulas of the quantum theory of radiation, as was done for example by Rautian and Sobel'man [2]. It is possible to calculate the atomic transitions quantum mechanically, and to calculate the field classically. Such a "classical" method was used by Karplus and Schwinger [1] and by Basov [3]. It is clear from general considerations that the intensities of the electromagnetic field can be regarded as c-numbers in the case when the number of photons per cell of phase space is sufficiently large, $n_\lambda \gg 1$. In ordinary light sources the radiation density is such that n_λ rarely exceeds unity. Nonetheless, in linear optics the use of the semiclassical method is permissible. The spectrum of the incident radiation can always be represented in the form of a sum of monochromatic components, for which obviously $n_\lambda = \infty$. If the

[1)]We shall refer specifically to atoms, but the results obtained pertain to any two-level system.

system is nonlinear, the condition $n_\lambda \gg 1$ becomes essential. In light beams emitted by quantum generators this number is quite large, $n_\lambda \cong 10^{11}-10^{12}$. The condition for the applicability of the semiclassical method is then satisfied with large margin.

FLUCTUATIONS

It is known that the electromagnetic field in a light wave fluctuates over a time on the order of $1/\Delta$, where Δ is the frequency interval of the incident spectrum. In stimulated emission, the fluctuations are determined by the shot effect. The existing light sources can be regarded in the majority of cases as systems consisting of a large number of independently radiating objects. The result of addition of many waves with arbitrary phases is conveniently described by representing the field in the form of a Fourier series with $-T/2 < t < T/2$, where $T \to \infty$. The phases of the Fourier components E_ω of the summary field intensity vector are random quantities, which assume with equal probability all values from 0 to 2π. The intensity $|E_\omega|^2$ is the Fourier transform of the correlation function, and, being already an averaged quantity, does not fluctuate. In linear optics the undetermined phases of the complex E_ω drop out from the final formulas, which are quadratic in the field vector, and come into play only in the coherence or incoherence phenomena.

To calculate linear systems such as optical generators it is necessary to know not only the intensity but also the law governing the phase distribution. It is known that the statistical properties of the generated signal are determined by the noise in the system. In optical generators, the photon-multiplication mechanism is such that the photons, increasing in number, maintain their phases and consequently the phase relations are not disturbed. We therefore assume throughout that the statistical properties of the incident field do not differ from the statistical properties of the "bare" field of the spontaneous emission, that is, that the phases are distributed with equal probability from 0 to 2π. In particular, we shall retain this assumption also in the case when the incident field coincides with the field of the optical generator itself.

The saturation effect is usually calculated for monochromatic light only. It is assumed here that the amplitude and the phase of the field-intensity vector are strictly fixed. Such calculations give the correct result only in the case when the time of observation is much shorter than the characteristic time of the field fluctuations (inertialess observation). In ordinary light sources the width of the spectral lines is $\Delta \sim 10^9$ sec^{-1} and larger. The fluctuation time $1/\Delta$ is very small, and it is clear that the optical instruments register only quantities that are averaged over the fluctuations.

It is easy to understand that in calculations with monochromatic radiation the statistical properties of the light can be taken into account without repeating all the derivations anew. It is sufficient to carry out the averaging in the final formulas. Let φ be a quantity proportional to the field in the incident wave, ω the frequency of the incident light ($\omega \approx \omega_0$), V the volume occupied by the field, and p_{ab} the transition matrix element. According to equation (34) of the paper of Karplus and Schwinger [1] the stimulated emission of the atom placed in the field φ changes intensity by an amount

$$\delta|\varphi|^2 = \frac{4\pi}{\hbar V}\,|p_{ab}|^2\,\frac{\Gamma}{\Gamma^2+(\omega_0-\omega)^2+|\varphi|^2}\,|\varphi|^2. \tag{1}$$

Account must be taken of the fact that φ depends little on the time and fluctuates in such a way that the Fourier components of the function $\varphi(t)$ have indeterminate phases. In order to carry out the averaging, it is necessary to know the probability that the value $|\varphi|^2$ (which has a specified mean) assumes a certain value. This probability was calculated by Rayleigh as long ago as in 1880 (see, for example, [6]). It amounts to

$$w(|\varphi|^2)\,d(|\varphi|^2) = \exp\{-|\varphi|^2/\overline{|\varphi|^2}\}\,d(|\varphi|^2)/\overline{|\varphi|^2}. \tag{2}$$

The results of integration of formula (1) with weight (2) are shown in the figure (lower curves). The upper-dashed curves have been plotted in accordance with (1), without account of the statistical properties. We see that the greatest role is played in the fluctuations in the case of average saturation, and the discrepancy does not exceed 20 per cent. It will be shown below that in spite of the large value of the effect, under certain conditions the fluctuations can greatly influence the gain, preventing the narrowing down of the spectral lines.

POPULATION EQUATIONS

If the light wave is a traveling wave, then the Doppler effect leads only to a shift in the frequencies of the spectrum in the coordinate frame fixed in the c.m.s. of the atom. In quantum generators, however, a standing wave is produced. It is easily understood that in a coordinate frame fixed in the moving atom the standing wave loses it monochromaticity. This effect was analyzed in detail by Rautian and Sobel'man [7]. In our paper we shall consider only the traveling wave.

Having made these preliminary remarks, let us proceed to describe the fundamental equations. The field is conveniently characterized by the function

$$\varphi(t) = 2\hbar^{-1}|p_{ab}|\,E(t)\,e^{i\omega_0 t},$$

which has a dimensionality $\sec^{-1}$. Here $E(t)$ is a variable electric field, and p_{ab} the matrix element of the transition. In the approximation $\Gamma \ll \omega_0$ and $\Delta \ll \omega_0$, where Δ is the width of the incident spectrum, we can neglect the double frequencies and assume that $\varphi(t)$ is a complex function with a characteristic interval of variation $1/\Delta$.

Let ζ be the frequency shift due to the first-order Doppler effect. In the coordinate system fixed at the c.m.s. of the atom, the function $\varphi(t)$ goes over into the product $\varphi(t)\exp(i\zeta t)$. Let a and b be the amplitudes of states "a" and "b," which depend on the time. The known procedure[3] leads to semi-classical equations describing the stimulated emissions with allowance for the Doppler effect

$$i\left(\dot{a} + \frac{\Gamma}{2}a\right) = \frac{\varphi}{2}\,be^{i\zeta t},$$

$$i\left(\dot{b} + \frac{\Gamma}{2}b\right) = \frac{\varphi^*}{2}\,ae^{-i\zeta t}. \qquad (3)$$

The initial conditions $a = 1$ and $b = 0$ are specified at the instant of time t_0, when the atom is introduced into the system. Summation over many atoms leads to averaging over t_0, and therefore, calculating the contribution of one atom, we shall average beforehand over t_0. The amplitudes a and b depend both on the time t and on t_0. Let us determine the overpopulation of the upper level compared with the lower one, averaged with respect to t_0,

$$N(t) = \Gamma\int_{-\infty}^{t}(|a|^2 - |b|^2)\,dt_0.$$

It follows from (3) that

$$-\Gamma + \frac{dN(t)}{dt} + \Gamma N(t) = -\operatorname{Re}\int_{-\infty}^{t}\varphi(t)\,\varphi^*(t')\exp\{-\Gamma(t-t') + i\zeta(t-t')\}\,N(t')\,dt'. \qquad (4)$$

We expand the field $\varphi(t)$ in a Fourier series with $-T/2 < t < T/2$, $T \to \infty$:

$$\varphi(t) = \sum_\lambda \varphi_\lambda e^{i(\omega_0 - \omega_\lambda)t}.$$

In order to find the stimulated emission within the framework of the semiclassical theory, it is sufficient to calculate the polarization vector. The component of the polarization Fourier vector has in the laboratory frame the form

$$p_\lambda = p_{ab}\,\overline{a^* b\exp\{-i(\omega_0 - \omega_\lambda)t - i\zeta t\}}. \qquad (5)$$

The superior bar denotes here averaging over the infinitely large time T. The change in intensity at the frequency ω_λ is proportional to the real part of the product $\varphi_\lambda^* p_\lambda$. The amplitudes a and b in the right half of (5) are conveniently replaced by substituting the formal integrals (3). We find that the radiation intensity at the frequency ω_λ increases by an amount

$$\delta|\varphi_\lambda|^2 = \frac{4\pi}{\hbar V}|p_{ab}|^2 \times \operatorname{Re}\left(\varphi_\lambda^*\,\overline{N(t)\,\varphi(t)\,e^{i(\omega_0-\omega_\lambda)t}}\right)/(\Gamma + i(\omega_0 - \omega_\lambda) + i\zeta). \qquad (6)$$

The mathematical difficulty lies in the fact that the function $\varphi(t)$, contained in (4), is a random function. One must solve Eq. (4) for arbitrary $\varphi(t)$, and then substitute the solution in (6) and average over the time and over the phases (statistical properties).

In general form, unfortunately, it is impossible to carry out the derivations. It is possible, however, to obtain a solution for the most important case, when the incident field has a spectral composition close to monochromatic. Let us assume that the width of the spectrum of the incident radiation is much smaller than the damping width Γ. In this case the function $\varphi(t)$ in equation (4) depends little on the time and has the obvious solution $N(t) = \text{const}$. For the stimulated emission we obtain the well known formula (1). The gain $\delta|\varphi_\lambda|^2/|\varphi_\lambda|^2$ at the fundamental frequency has a maximum at $\omega_\lambda = \omega_0$, thus determining the "oscillation mode with maximum Q." The averaging of (1) over the Doppler shifts with mean square ζ_0^2 yields

$$\frac{\delta|\varphi_\lambda|^2}{|\varphi_\lambda|^2} = \frac{4\pi}{\hbar V}|p_{ab}|^2\int_0^\infty \frac{\Gamma\,dt}{\sqrt{1 + 2\zeta_0^2 t}} \times \exp\left[-\left(\Gamma^2 + |\varphi_\lambda|^2 + \frac{(\omega_\lambda - \omega_0)^2}{1 + 2\zeta_0^2 t}\right)t\right]. \qquad (7)$$

It is seen that for all relations between the parameters of the problem the gain remains a maximum when $\omega_\lambda = \omega_0$.

This fact, however, is still not sufficient to enable us to draw any conclusion concerning the spectral narrowing. It may turn out that the monochromatic solution is not stable (that is, with maximum Q). If the gain at the sideband frequencies exceeds the gain at the fundamental frequency, the radiation spectrum will start broadening (there is a limit on the narrowing). In this connection it is necessary to solve also the stability problem: let the incident wave represent a superposition of an intense field frequency ω_λ and a weak field with frequency ω_μ.

We shall take the weak field into account in first approximation, assuming that the overpopulation "flickers" slightly with a difference frequency $\omega_\mu - \omega_\lambda$.

It follows from (4) and (6) that the gain at the frequency is determined by the expression

$$\frac{\delta|\varphi_\mu|^2}{|\varphi_\mu|^2} = \frac{4\pi}{\hbar V}|p_{ab}|^2 \operatorname{Re}\left[\frac{\Gamma^2 + (\omega_0 - \omega_\lambda + \zeta)^2}{(\Gamma^2 + (\omega_0 - \omega_\lambda + \zeta)^2 + |\varphi_\lambda|^2)(\Gamma + i(\omega_0 - \omega_\mu + \zeta))}\right.$$
$$\left.\times\left(1 - \frac{|\varphi_\lambda|^2}{2}\frac{(\Gamma - i(\omega_0 - \omega_\lambda + \zeta))^{-1} + (\Gamma + i(\omega_0 - \omega_\mu + \zeta))^{-1}}{\Gamma + i(\omega_\lambda - \omega_\mu) + |\varphi_\lambda|^2\Gamma/\{[\Gamma + i(\omega_\lambda - \omega_\mu)]^2 + (\omega_0 - \omega_\lambda + \zeta)^2\}}\right)\right]. \tag{8}$$

Here $|\varphi_\lambda|^2$ is the intensity of the monochromatic line (at the fundamental frequency). As $\omega_\mu \to \omega_\lambda$ the foregoing formula yields

$$\frac{\delta|\varphi_\mu|^2}{|\varphi_\mu|^2} = \frac{4\pi}{\hbar V}|p_{ab}|^2 \frac{\Gamma[\Gamma^2 + (\omega_0 - \omega_\mu + \zeta)^2]}{[\Gamma^2 + (\omega_0 - \omega_\mu + \zeta)^2|\varphi_\lambda|^2]^2}. \tag{9}$$

The gain at the weak sideband frequencies turns out to be as a rule smaller than the gain at the fundamental frequency [see formula (1)], and near the fundamental frequency the gain changes abruptly (this effect was observed by Rautian and Sobel'man [2]). However, if we take into account the statistical nature of the light, expressions (8) and (9) must still be averaged in accordance with the probability distribution (2). It is easy to verify that the integrals of (1) and of (9) coincide identically, and the field fluctuations in the incident light wave lead to a vanishing of the jump in the amplification and deteriorate the conditions under which the spectral line narrows down.

FUNDAMENTAL PARAMETERS AND PARTICULAR CASES

Let us consider a plane wave propagating in a homogeneous medium with inversely populated levels. The character of the variation of the spectral composition under these conditions is determined by four parameters, namely, Γ —the damping constant of the atomic levels, ζ_0 —the mean square Doppler shift, and the average field at $(\overline{|\varphi|^2})^{1/2}$ in $\sec^{-1}$. The other important parameter depends on the concentration of the active centers, namely the rate of increase of the signal along the beam β.

By definition of the quantity β, the intensity increases as $\exp(\beta x/c)$, where x is the distance along the beam. In optical generators in the steady-state mode, the value of β is determined by the losses and is simply equal to $c(1-r)/l$, where r is the reflection coefficient and l the distance between mirrors. Within a time on the order of $1/\beta$, the field can increase by a factor of several times, so that the processes which occur at speeds much smaller than β have no time to occur. Let the spectrum of the incident radiation have a width $\Delta \ll \beta$. Under these conditions, the field fluctuations do not influence the amplification and generation. When $\beta \ll \Delta$, to the contrary, it is essential to take the fluctuations into account. In gases usually $\Gamma \ll \zeta_0$. In dense media, as is well known, the thermal motion of the atoms does not lead to the Doppler effect, and therefore it is necessary to put for crystals $\zeta = 0$. For illustration, let us give the values of the main parameters of a laser operating with a mixture of helium and neon [5]:

$$\Gamma \sim 10^7\,\sec^{-1},\quad \zeta_0 \sim 2\cdot 10^9\,\sec^{-1},\quad \varphi \sim 4\cdot 10^8\,\sec^{-1},$$
$$\beta \sim 2\cdot 10^6\,\sec^{-1},\qquad \Delta \sim 10^4\,\sec^{-1}.$$

We proceed now to analyze particular cases. We assume that the spectrum of the incident radiation is close to monochromatic, $\Delta \ll \Gamma$.

<u>Dense medium, $\zeta = 0$, very narrow line, $\Delta \ll \beta$.</u> This case was considered in detail by Rautian and Sobel'man [2]. The gain at the fundamental frequency is determined by formula (1); the gain at the weak sideband frequencies is apparently smaller by approximately the magnitude of the jump, that is, by the difference between the radiation in accordance with (1) and (9).

<u>Gas medium, $\zeta_0 \gg \Gamma$, very narrow line, $\Delta \ll \beta$.</u> The radiation at the fundamental frequency is determined by expression (7). The jump in the gain guarantees stability of the monochromatic solution.

<u>Dense medium, $\zeta = 0$, width $\Delta \gg \beta$.</u> In this case the fluctuations of the light field are significant. The gain at the fundamental frequency is obtained by averaging formula (1) (Figs. 2 and 3). The jump in the gain drops out. On going to the sideband frequencies the gain decreases monotonically (stability). A typical curve at a medium degree of saturation, when $\overline{|\varphi|^2} = \Gamma^2$, is shown in Fig. 4.

<u>Gas medium, $\zeta_0 \gg \Gamma$, width $\Delta \gg \beta$.</u> An analysis of the fundamental formula (8) shows that the monochromatic solution is unstable even for weak de-

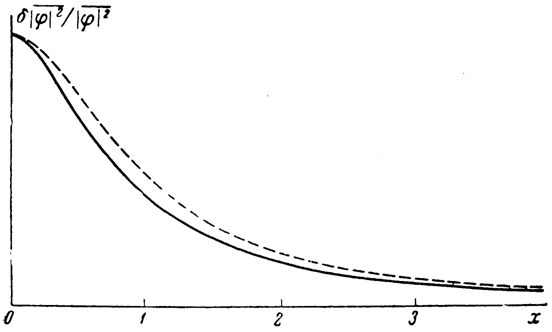

FIG. 2. Decrease in the coefficient of stimulated emission resulting from saturation. The abscissas represent the average quantity $x = (\overline{|\varphi|^2}/\Gamma^2)^{1/2}$, which is proportional to the electromagnetic field incident on the atom. The ordinates represent the intensity of the stimulated emission, divided by the intensity of the incident field, averaged over the fluctuations. The upper curve is constructed without account of fluctuations

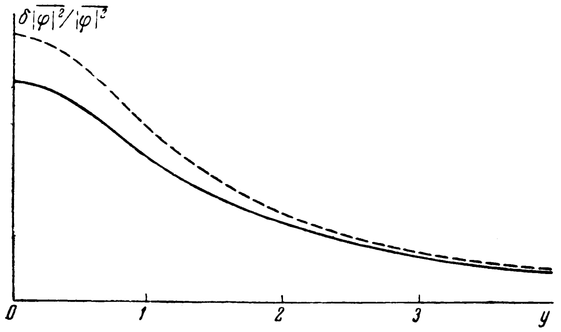

FIG. 3. Decrease in coefficient of stimulated emission resulting from detuning. The abscissas represent the detuning in units of $y = |\omega_0 - \omega_\lambda|/\Gamma$. The degree of saturation is such that $\overline{|\varphi|^2} = \Gamma^2$. The upper curve has been plotted without allowance for fluctuations.

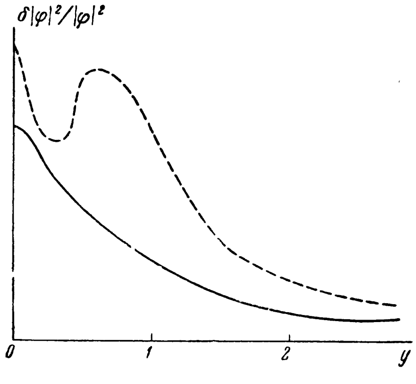

FIG. 4. Coefficient of stimulated emission as a function of the weak-field frequency. Case $\beta \ll \Delta \ll \Gamma$, $\zeta_0 = 0$. The abscissas represent the detuning $|\omega_0 - \omega_\lambda|/\Gamma$. Here $\omega_0 = \omega_{ab}$ is the frequency of the main strong monochromatic field and ω_μ is the frequency of the weak field. The degree of saturation is such that $\overline{|\varphi|^2} = \Gamma^2$. The upper dashed curve is plotted without account of fluctuations ($\Delta \ll \beta$).

grees of saturation. In weak saturation we have in first approximation

$$\frac{\delta\,|\varphi_\mu|^2}{|\varphi_\mu|^2} \cong \frac{4\pi}{\hbar V}\,|p_{ab}|^2 \sqrt{\frac{\pi}{2\zeta_0^2}} \left(1 - \frac{|\varphi_\lambda|^2}{\Gamma^2 + (\omega_\lambda - \omega_\mu)^2}\right). \quad (10)$$

Let us assume that the frequency of the fundamental field coincides with the atomic frequency ω_0. At very weak fields the gain will decrease with increasing difference $\omega_0 - \omega_\mu$ as a result of the detuning. The greatest stability is obtained when $\omega_\mu \approx \omega_0$. In the approximation of linear optics we have

$$\frac{\delta\,|\varphi_\lambda|^2}{|\varphi_\lambda|^2} \cong \frac{4\pi}{\hbar V}\,|p_{ab}|^2 \sqrt{\frac{\pi}{2\zeta_0^2}} \left(1 - \frac{(\omega_0 - \omega_\mu)^2}{2\zeta_0^2}\right). \quad (11)$$

Comparing (10) with (11) we see that if the intensity of the main field exceeds $\Gamma^4/2\zeta_0^2$, the gain curve at the sideband frequencies has a minimum when $\omega_\mu = \omega_0$ and the solution becomes unstable. In generators $|\varphi_\lambda|^2$ is always larger than $\Gamma^4/2\zeta_0^2$, and therefore the monochromatic curve will spread at least to the damping width Γ.

It must be noted that the influence of the fluctuations always decreases the gain, and therefore a very narrow line with $\Delta \ll \beta$ is known to be under more favorable conditions. Nonetheless, it may turn out that the calculations for a strong monochromatic field will lead to certain "natural spectra" with $\Delta \sim \Gamma$, which will be stable relative to weak perturbations. This will mean that in gaseous media for the same parameters of the medium there are possible two oscillation modes, in one of which the spectrum width of the emitted light is of the order of the damping width.

In conclusion I take this opportunity to thank S. G. Rautian, I. I. Sobel'man, and L. I. Gudzenko for numerous discussions.

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