

Polarization Characteristics of Secondary Radiation from a Gas System

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The secondary radiation produced under the action of arbitrarily polarized intense light \mathbf{E}_L in resonance with the atomic transition $A \rightarrow B$ on a gas system located in a stationary magnetic field \mathbf{H} is investigated theoretically. The secondary radiation is regarded as being the result of interaction between atoms of the system and photon vacuum. Expressions for the frequency spectra of secondary radiation of arbitrary polarization involving the $A \rightarrow B$, $A \rightarrow C$ and $B \rightarrow D$ transitions in terms of the atomic density matrices in prescribed classical fields \mathbf{E}_L and \mathbf{H} are derived by means of the Konstantinov-Perel' diagram technique. The expressions obtained are employed for investigating the structure arising on the spontaneous emission Doppler contours for the $A \rightarrow B$ transition when \mathbf{E}_L is a traveling wave of low intensity. For certain polarizations of the field \mathbf{E}_L and secondary radiation, terms appear in the structure which are due to gas system atoms being in coherent superposition in the Zeeman sublevels (Hertz coherence). These terms possess properties which are characteristic of both the ordinary secondary radiation and the Hanle effect. They depend in a resonant manner on the magnetic field strength \mathbf{H} , and are negligible when the Zeeman splitting exceeds the radiative width of the upper level. When the \mathbf{E}_L field polarization and secondary radiation polarization are linear, the structure magnitude at $\mathbf{H} = 0$ is modulated as $\sim \cos 2\psi$, where ψ is the angle between the polarizations. For the $2S_2 \rightarrow 2P_1$ transition, $\lambda = 1.52 \mu$, in Ne the percentage modulation may reach 100%.

INTRODUCTION

THE interaction of atoms with an intense optical field that is resonant with one of the atomic transitions leads to a number of distinct effects both in stimulated and in spontaneous emission^[1-9]. In the case of spontaneous or secondary radiation¹⁾, the resonant field causes deformation of the frequency spectrum of the immobile atoms^[1,2]. The spectrum of the atoms participating in the thermal motion depends on the angle θ between the directions of observation and of the propagation of the light field^[2,4,8]. In a broad Doppler line ($\gamma \ll ku$), even a rather weak resonant light field leads to the appearance of a structure with width γ on the order of the radiative width when observed at angles θ close to zero or π , and the structure can have different forms for $\theta = 0$ and $\theta = \pi$ ^[3-6].

This paper investigates the polarization characteristics of the frequency spectra of the secondary radiation of the gas system in a magnetic field. The gas system is regarded as an ensemble of atoms situated in a classical electromagnetic field \mathbf{E}_L that is resonant with the atomic dipole transition $A \rightarrow B$. A constant magnetic field \mathbf{H} is applied to the system and leads to a linear Zeeman effect. It is assumed that each atomic level K can be characterized by a total angular momentum j_K . Homogeneous and isotropic incoherent pumping is applied to the levels A and B . The secondary radiation is regarded as a result of the interaction of the ensemble of the atoms with the photon vacuum.

In addition to the already mentioned scalar effects, effects connected with the vector character of the field \mathbf{E}_L and with the Hertz coherence of the ensemble of atoms can appear in secondary radiation of arbitrary polarization. Indeed, when the field \mathbf{E}_L consists of a mixture of σ^+ , σ^- , and π polarized components, it trans-

forms the atoms into states that are coherent superpositions of Zeeman sublevels. A magnetic field applied to the system destroys the Hertz coherence of the ensemble and the radiation of light by such a system leads to resonant effects such as the Hanle effect^[11-13]. At the same time, an investigation of the secondary radiation in a polarization orthogonal to the polarization of the intense field \mathbf{E}_L may be convenient from the experimental point of view, since it permits an additional discrimination of the secondary radiation from the field \mathbf{E}_L with the aid of polarization.

In Sec. I we derive general expressions for the power of arbitrarily polarized secondary radiation as a function of its frequency on the transitions $A \rightarrow B$, $A \rightarrow C$, and $B \rightarrow D$ (Fig. 1) (formulas (10) and (12)). These expressions, which reduce the determination of the spectra in a field \mathbf{E}_L of arbitrary intensity to a calculation of atomic density matrices, acquire a particularly simple form when the field \mathbf{E}_L constitutes a single traveling wave (formula (15)). The dependence of expressions (10) and (12) for the spectra on the density matrix elements $\rho_{a'a_1}$ which are not diagonal with respect to the Zeeman sublevels of the level A is a formal confirmation of the statement made above concerning the influence of the Hertz coherence of the ensemble of atoms on the secondary-radiation spectra.

In Sec. II we investigate in detail the spectrum of the secondary radiation on the resonant transition $A \rightarrow B$ in the approximation of a weak field \mathbf{E}_L , when $(\mathbf{E}_L \cdot \mathbf{d}_{ab})^2 \ll \gamma_a \gamma_b$. We determine the structure which appears on broad spontaneous-emission Doppler contours in a longitudinal magnetic field when the levels A and B have equal g factors, and which is proportional to the intensity of the arbitrarily polarized traveling wave $|\mathbf{E}_L|^2$. In the general case, the structure consists of an incoherent part and a coherent part due to the Hertz coherence of the ensemble of atoms; these parts differ noticeably in their properties (formulas (16) and (17)).

¹⁾We use the term "secondary radiation" ^[10] in place of the more cumbersome "spontaneous emission in an external field," reserving the expression "spontaneous emission" for the case when there is no external field.

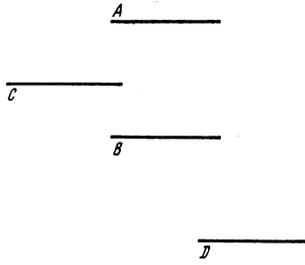


FIG. 1

The frequency dependence of the structure in the particular case of linear polarizations of the field \mathbf{E}_L and of the secondary radiation in the $\theta = 0$ direction is shown in Fig. 2 (in accordance with formula (18)).

The position of the minima of the structure can be explained by using concepts analogous to those of the "own" and "foreign" Bennett dips^[14]. The structure is due to the fact that the only atoms effectively interacting with the field \mathbf{E}_L are those whose velocities satisfy the resonance condition

$$\omega_L - k v_q = \omega_{ab} + q\Omega, \quad (1)$$

where ω_L and \mathbf{k} are the frequency and wave vector of the wave \mathbf{E}_L , ω_{ab} is the frequency of the $A \rightarrow B$ transition at $\mathbf{H} = 0$, $q = \pm 1$, and $\Omega = \mu g\mathbf{H}$ is the frequency distance between the Zeeman sublevels.

The atoms with velocities \mathbf{v}_q emit at the frequencies

$$\omega_{\mu^q} - \mathbf{k}_{\mu} \cdot \mathbf{v}_q = \omega_{ab} + q_1 \Omega, \quad (2)$$

where \mathbf{k}_{μ} is the wave vector of the secondary-radiation photon μ . Putting $\mathbf{k}_{\mu} \cdot \mathbf{v}_q = \mathbf{k} \cdot \mathbf{v}_q$, we obtain from (1) and (2) the frequencies at which the "own" dips appear at $q_1 = q$ and the "foreign" dips appear at $q_1 = -q$:

$$\omega_{\mu^q} = \omega_L + (q_1 - q)\Omega.$$

From this we obtain at $q_1 = q$ the condition for the central minimum, and at $q_1 = -q$ the condition for the two side minima at $\omega_{\mu} = \omega_L \pm 2\Omega$ (see Fig. 2).

The magnetic field \mathbf{H} influences the structure (18) in two ways. First, it causes both types of dips to be split by 2Ω (a consequence of the Zeeman effect), and second, it destroys the Hertz coherence of the ensemble of atoms, leading to the resonant dependence, characteristic of the Hanle effect, of the coherent part of the structure on Ω . Unlike the incoherent part, the coherent part of the structure also depends on the angle ψ between the linear polarizations of the field \mathbf{E}_L and of the secondary radiation. This dependence acquires the particularly simple form of a structure modulation proportional to $\cos 2\psi$ at $\mathbf{H} = 0$ (formula (19)). At certain values of j_A and j_B , for example, for the $2S_2-2P_1$ transition in Ne

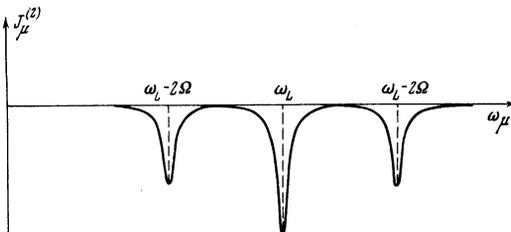


FIG. 2

($\lambda = 1.52 \mu$), the depth of such modulation is 100%. If the linear polarizations are equal ($\psi = 0$) and $\mathbf{H} = 0$, the results coincide with those of the scalar theory^[2,4].

I. DERIVATION OF GENERAL FORMULA FOR THE SECONDARY-RADIATION SPECTRUM

1. We consider an ensemble of noninteracting atoms, which will be assumed during the first stage of the calculations to be immobile. We assume also that the collective effects are negligible. The atoms interact with the classical fields \mathbf{E}_L and \mathbf{H} and with the photon vacuum.

The photon field is assumed quantized in traveling plane waves. The field oscillator with frequency ω_{μ} , wave vector \mathbf{k}_{μ} , and unit polarization vector $\mathbf{e}^{(\mu)}$ will be characterized by a single index μ . The secondary-radiation photons in the state μ can be emitted as a result of the interaction of the atoms with the field oscillator μ .

We are interested in the power of secondary radiation having a certain polarization $\mathbf{e}^{(\mu)}$ in the frequency interval $d\omega_{\mu}$ about ω_{μ} and in a solid angle $d\Omega$ about \mathbf{k}_{μ} . The specified limits contain $\mathcal{V} d\mathbf{k}_{\mu} / (2\pi)^3$ field oscillators (\mathcal{V} is the normalization volume), and the sought power is given by the expression

$$I_{\mu} d\omega_{\mu} d\Omega = \hbar \omega_{\mu} \frac{dN_{\mu}}{dt} \frac{\mathcal{V} d\mathbf{k}_{\mu}}{(2\pi)^3}, \quad (3)$$

$$\frac{dN_{\mu}}{dt} = \int_{-\infty}^t \frac{\partial n_{\mu}}{\partial t} dt_0. \quad (4)$$

Here n_{μ} is the average number of photons of type μ emitted by the instant of time t by atoms excited by the pump at the instant of time t_0 (the pump is turned on at $t = -\infty$).

2. We introduce the density matrix F of the quantum system consisting of the photon vacuum and the atoms excited by the instant of time t_0 to the levels A and B of atoms situated in the classical fields \mathbf{E}_L and \mathbf{H} . Then

$$n_{\mu} = \text{Sp } F a_{\mu}^{\dagger} a_{\mu}, \quad (5)$$

where a_{μ}^{\dagger} and a_{μ} are the operators for the creation and annihilation of photons in the state μ .

The density matrix F satisfies the equation

$$i\hbar \partial F / \partial t = [\mathcal{H}, F]. \quad (6)$$

The Hamiltonian of the system is $\mathcal{H} = H_0 + \mathcal{H}'$. The unperturbed part $\mathcal{H}_0 = \mathcal{H}_{ph} + \mathcal{H}_{at}$ consists of the Hamiltonians of the photon field and of the atoms in the magnetic field. The interaction Hamiltonian $\mathcal{H}' = \hbar(V + U)$ includes the interaction with the classical field $\hbar V = -\mathbf{d} \cdot \mathbf{E}_L$ and with the photon vacuum $\hbar U = -c^{-1} \mathbf{d} \cdot \mathbf{A}$, where \mathbf{d} is the operator of the time derivative of the dipole moment and \mathbf{A} is the operator of the vector potential of the photon field.

The formal solution of (6) can be written in the form

$$F(t) = e^{-i\mathcal{H}_0 t / \hbar} S(t, t_0) e^{i\mathcal{H}' t_0 / \hbar} F(t_0) e^{-i\mathcal{H}' t_0 / \hbar} S^{\dagger}(t, t_0) e^{i\mathcal{H}' t / \hbar}. \quad (7)$$

Here $S(t, t_0)$ is the operator of the evolution of the system in the interaction representation. It can be represented in the form of the formal series

$$S(t, t_0) = 1 + \sum_{m=1}^{\infty} \left(-\frac{i}{\hbar} \right)^m \int_{t_0}^t dt_m \tilde{\mathcal{H}}'(t_m) \int_{t_0}^{t_m} dt_{m-1} \tilde{\mathcal{H}}'(t_{m-1}) \dots \int_{t_0}^{t_1} dt_1 \tilde{\mathcal{H}}'(t_1),$$

$$\tilde{\mathcal{H}}'(t) = e^{i\mathcal{H}_0 t / \hbar} \mathcal{H}' e^{-i\mathcal{H}_0 t / \hbar}.$$

We choose the initial density matrix in the form²⁾

$$F(t_0) = \Phi\rho(t_0). \quad (8)$$

Here Φ is the density matrix of the photon vacuum, $\rho(t_0)$ is the atomic density matrix describing the state produced by the pump at the instant of time t_0 , namely, at each Zeeman sublevel k of the level $K = A$ or B there are λ_k atoms, with $(2j_K + 1)\lambda_k$ the number of atoms excited to the level K per unit time. Substituting (7) in (5), we obtain

$$n_\mu = \text{Sp} F(t_0) S^+(t, t_0) a_\mu^+(t) a_\mu(t) S(t, t_0). \quad (9)$$

3. The quantities n_μ are calculated with the aid of the diagram technique of Konstantinov and Perel'^[12,15,16]. The interaction \mathcal{H}' is taken into account in all orders of perturbation theory, retaining in each order the diagrams that make the main contribution at times comparable with the characteristic radiative lifetime of the considered atomic levels γ^{-1} . The ratio of the discarded diagrams to those taken into account will then be of order γ/ω , where ω is the characteristic frequency distance between the considered atomic levels^[12]. The only limitation imposed on the classical field is $(V/\omega)^2 \ll 1$, so that the field \mathbf{E}_L can be regarded as interacting with only one atomic transition $A \rightarrow B$.

Under the foregoing assumptions, the energy conservation law is approximately satisfied at each vertex of the diagrams that are taken into account^[12]. The secondary radiation on each of the transitions $A \rightarrow B$, $A \rightarrow C$, and $B \rightarrow D$ must be regarded separately, assuming each time that the emission of the secondary-radiation photons in the state μ of interest to us is the result of the corresponding atomic transitions.

For our purposes, the most convenient is the diagram-technique variant developed in^[12], where the motion of the atoms is considered classically. We neglect here the momentum of the recoil produced when a photon is emitted by the atom.

We illustrate the determination of the quantity n_μ with an example of secondary radiation on the transition $A \rightarrow B$ for immobile atoms³⁾.

Figure 3 shows one of the diagrams corresponding to the expansion (9). The solid horizontal lines are atomic and the wavy lines are photonic. The atomic line l going from the point t_1 to the point t_2 corresponds to the factor $\exp[-i\epsilon_l(t_2 - t_1)/\hbar]$, and the photonic line ν corresponds to the factor $\exp[-i\omega_\nu(t_2 - t_1)]$. Here ϵ_l and $\hbar\omega_\nu$ are the energies of the Zeeman sublevel l and of the photon ν , described by the atomic and photonic lines, respectively. All the atomic and photonic lines are regular, i.e., the starting point of the line lies earlier on the Konstantinov-Perel' contour than its end point^[12,16]. The interaction with the classical field V is denoted by crosses, and that with the photon vacuum U by circles. The cross at the point t' is set in correspondence with the matrix element $V_{ik}(t')$, where k is the index of the incoming atomic line and i is the index of the outgoing one. A circle with an incoming photonic line ν (absorption of a photon ν) corresponds to a factor

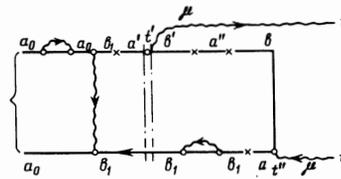


FIG. 3

$(2\pi/\gamma\hbar\omega_\nu)^{1/2} \cdot \mathbf{d}_{lm}(\nu)$, and one with an outgoing line (emission of a photon ν) corresponds to a factor $(2\pi/\gamma\hbar\omega_\nu)^{1/2} \cdot \mathbf{d}_{lm}(\nu)^*$, where m and l are the indices of the incoming and outgoing atomic lines. A vertical line joining the atomic lines on the right signifies that the trace is taken over the atomic indices. According to the right-hand return rule^[12,15], it is drawn beyond the vertex at the point t'' . The curly bracket on the left-hand end of the diagram corresponds to the initial atomic density matrix $\rho_A = \lambda_a \hat{E}_A$, where \hat{E}_A is a unit matrix of order $2j_A + 1$. The expansion (9) also contains diagrams with initial density matrix ρ_B in accordance with the representation of $\rho(t_0)$ from (8) in the form of a submatrix

$$\rho(t_0) = \begin{pmatrix} \rho_A & 0 \\ 0 & \rho_B \end{pmatrix}.$$

Each vertex on the upper atomic line is assigned a factor $-i$, and on the lower one a factor i . Summation is carried out over all the internal indices and integration is carried out over all the times characterizing the vertices in the same order as the arrangement of the vertices on the diagram.

The internal photon lines can be of two types.

1) The start and end of the photon line lie on the upper (lower) atomic line l . The summation of such self-energy parts in all orders of perturbation theory in U causes the corresponding Zeeman sublevel l to acquire a radiative width (we neglect the shift)^[12,16]

$$\gamma_l = \sum_k \gamma_{l \rightarrow k}, \quad \gamma_{l \rightarrow k} = \frac{4\omega_{lk}^3 |j_L \parallel d \parallel j_K|^2}{3\hbar c^3 (2j_L + 1)},$$

where ω_{lk} is the central frequency of the $L \rightarrow K$ transition and $(j_L \parallel d \parallel j_K)$ is the reduced dipole-moment matrix element.

2) The start of the photon line lies on the upper atomic line and the end on the lower one. For the analysis that follows it is important that in the approximation $\gamma/\omega \ll 1$ under consideration, such photon lines can be regarded as vertical^[10,17]. The fact that the photon lines are vertical makes it possible to break up each diagram into two fragments, as shown arbitrarily in Fig. 3 by the dash-dot lines. Each of the fragments constitutes a diagram corresponding to a certain term in the series expansion (with respect to \mathcal{H}') of the matrix element of the atomic density matrix obtained under definite initial conditions.

In accordance with the general rule, the atomic density matrix is defined as a trace of the total density matrix of the system over all the photon indices. By summing the diagrams of the expansion (9) over all orders of perturbation theory, we find that the graphic expression for n_μ can be represented in the form of a sum of four diagrams. Each of the diagrams consists of two fragments (blocks), which are the elements of the atomic density matrices in the external fields \mathbf{E}_L and \mathbf{H} .

²⁾The form of $F(t_0)$ does not affect the final result^[12,16].

³⁾The diagram technique developed in^[12] makes it possible to take the motion of the atoms into account. We consider immobile atoms to simplify the derivation. Allowance for the motion is discussed later on.

4. Changing over in accordance with (4) to an ensemble of atoms with constantly acting pump, we obtain in the transition $A \rightarrow B$ the graphic expression shown in Fig. 4 for the average number dN_{μ}/dt of the photons of type μ emitted by such an ensemble in a unit time.

In each of the diagrams, the first block (with arrows coming from the left) is the density matrix element of the ensemble of atoms with the pump. Thus, in the first diagram this block corresponds to the quantity $\rho_{a'a_1}(t')$. The second blocks are the density matrix elements of one atom. In the first diagram this is $\rho_{b'a_1}; ba(t', t)$.

The first two subscripts of $\rho_{b'a_1}; ba(t', t)$ denote the initial conditions specified at the instant of time t' , namely $\rho_{b'a_1}; ba(t', t) = \delta_{b'b} \delta_{a_1 a}$. The "remainder" of the incoming photon line at the circle in the lower right corner of each diagram indicates that this is the vertex for the absorption of the photon μ .

With the aid of (3) we obtain for the power of the secondary radiation the fundamental equation determining the spectrum of the immobile atoms on the transition $A \rightarrow B$ ⁴:

$$I_{\mu} = C_{AB} \sum_{aa'bb'} \left\{ D_{aa'}^{bb'}(e^{(i\mu)}) \int_{-\infty}^t dt' \left[\sum_{a_1} \rho_{aa_1}(t') \rho_{b'a_1; ba}(t', t) \right. \right. \\ \left. \left. + \sum_{b_1} \rho_{a'b_1}(t') \rho_{b'b_1; ba}(t', t) \right] e^{-i\omega_{\mu}(t-t')} \right\} + \text{c.c.} \quad (10)$$

Here

$$C_{AB} = 3\hbar\omega_{\nu} \gamma_{a \rightarrow b} (2j_A + 1) / 16\pi^2 |j_A \parallel d \parallel j_B|^2,$$

and $D_{aa'}^{bb'}(e^{(i\mu)}) = (d_{ab} \cdot e^{(i\mu)}) (d_{b'a'} \cdot e^{(i\mu)*})$ determines the angular characteristics of the radiation.

The graphic expressions for dN_{μ}/dt on the transitions $A \rightarrow C$ and $B \rightarrow D$ are analogous to that shown in Fig. 4. The only difference is that the radiation of the photons μ occurs on the transitions $A \rightarrow C$ or $B \rightarrow D$, and not $A \rightarrow B$ as shown in Fig. 4. The formula for I_{μ} on the transition $A \rightarrow C$ is thus obtained from (10) by replacing the indices b and b' by c and c' , and for the $B \rightarrow D$ transition a, a', b and b' must be replaced by b, b', d , and d' , respectively.

In all the considered transitions, the formal structure of the expression for the spectra as functions of the atomic density matrices is one and the same, and the determination of the secondary-radiation spectra reduces to a standard procedure of calculating the atomic density matrices in specified classical fields E_L and H .

5. Formula (10) and the analogous expressions for the transitions $A \rightarrow C$ and $B \rightarrow D$ contain density matrices of two types.

1) The density matrix of an ensemble of atoms with pumping; the matrix element is $\rho_{ik}(t')$. The matrix satisfies the well-known equation

$$i\hbar \partial \rho / \partial t = [(\mathcal{H}_{at} + \hbar V), \rho] - 1/2 i \hbar (\Gamma \rho + \rho \Gamma) + i \hbar \Lambda. \quad (11)$$

In the energy representation, the radiative-decay matrix Γ and the pump matrix Λ are diagonal, with Λ having nonzero matrix elements λ_a and λ_b only for the Zeeman sublevels of the levels A and B .

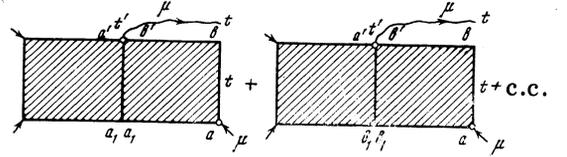


FIG. 4

2) The density matrix describing the state of one atom; the matrix element is $\rho_{ik}; l_m(t', t)$. This matrix satisfies the homogeneous part of Eq. (11) (at $\Lambda = 0$). We recall that the initial conditions posed at the instant of time t' are specified by the relation $\rho_{ik}; l_m(t', t') = \delta_{il} \delta_{km}$.

6. Formula (10) gives the secondary-radiation spectrum for an ensemble of immobile atoms. Let us consider now the case when the atoms are in motion. Assume that the pumping causes atoms to be produced at the levels A and B with a certain velocity distribution $f(v)$. It can be assumed that expression (10) has been written out for a subensemble of atoms having the same velocity v in the coordinate system in which these atoms are at rest. On going over to the laboratory frame, the power I_{μ} becomes a function of the velocity $I_{\mu}(v)$: the frequency ω_{μ} is replaced by the Doppler-shifted frequency $\omega'_{\mu} = \omega_{\mu} - \mathbf{k}_{\mu} \cdot \mathbf{v}$, and the dependence on the velocity appears also in the atomic density matrices. The expression for $I_{\mu}(v)$ contains, in addition, an integration that is not contained in (10), with respect to the positions \mathbf{r}' of the atoms at the instant of time t' . It is obvious that by averaging $I_{\mu}(v)$ over the velocities with a distribution $f(v)$ we obtain the sought power of the secondary radiation of an ensemble of moving atoms

$$I_{\mu} = \int_{-\infty}^{\infty} I_{\mu}(v) f(v) dv.$$

Then for the transition $A \rightarrow B$ we have

$$I_{\mu} = C_{AB} \sum_{aa'bb'} \left\{ D_{aa'}^{bb'}(e^{(i\mu)}) \int_{-\infty}^t dv \int_{-\infty}^t dt' \int d\mathbf{r}' \left[\sum_{a_1} \rho_{aa_1}(\mathbf{r}', v, t') \right. \right. \\ \left. \left. \times \rho_{b'a_1; ba}(\mathbf{r}', t'; \mathbf{r}, t; v) + \sum_{b_1} \rho_{a'b_1}(\mathbf{r}', v, t') \rho_{b'b_1; ba}(\mathbf{r}' t'; \mathbf{r}, t; v) \right] \right. \\ \left. \times \exp \{-i(\omega_{\mu} - \mathbf{k}_{\mu} \cdot \mathbf{v})(t - t')\} \right\} + \text{c.c.} \quad (12)$$

The system of equations for $\rho_{ik}(\mathbf{r}, \mathbf{v}, t)$ is obtained from (11) by making the substitutions $\partial/\partial t \rightarrow \partial/\partial t + \mathbf{v} \nabla$ and $\Lambda \rightarrow \Lambda f(\mathbf{v})$. From the homogeneous system we obtain the density matrix elements of one atom, $\rho_{ik}; l_m(\mathbf{r}', t'; \mathbf{r}, t; v)$, each of which is a solution of the initial problem under the condition that at the initial instant of time t' we have $\rho_{ik}; l_m(\mathbf{r}', t'; \mathbf{r}, t'; v) = \delta(\mathbf{r} - \mathbf{r}') \delta_{il} \delta_{km}$. In fact, the quantities $\rho_{ik}; l_m(\mathbf{r}', t'; \mathbf{r}, t; v)$ are the elements of the matrix Green's function of this system of equations^[4].

Formula (12) is valid for the case when the atomic collisions are immaterial and the velocity of each atom is constant. It can be shown that when the collisions are taken into account we get

$$I_{\mu} = C_{AB} \sum_{aa'bb'} \left\{ D_{aa'}^{bb'}(e^{(i\mu)}) \int_{-\infty}^t dv \int_{-\infty}^t dv' \int_{-\infty}^t dt' \int d\mathbf{r}' \left[\sum_{a_1} \rho_{aa_1}(\mathbf{r}', v', t') \right. \right. \\ \left. \left. \times \rho_{b'a_1; ba}(\mathbf{r}', v', t'; \mathbf{r}, v, t) + \sum_{b_1} \rho_{a'b_1}(\mathbf{r}', v', t') \rho_{b'b_1; ba}(\mathbf{r}' v', t'; \mathbf{r}, v, t) \right] \right\}$$

⁴Formula (10) gives the secondary radiation from a certain specified point of space \mathbf{r} , at which the radiating atoms are located. The density matrices depend on \mathbf{r} as a parameter.

$$\times \exp \{-i[\omega_\mu(t-t') - \mathbf{k}_\mu(\mathbf{r}-\mathbf{r}')]\} + \text{c.c.} \quad (13)$$

Naturally, it is necessary here to introduce the collision terms into the atomic density matrices, and

$$\rho_{ik}; l_m(\mathbf{r}', \mathbf{v}', t'; \mathbf{r}, \mathbf{v}, t) = \delta(\mathbf{r}-\mathbf{r}')\delta(\mathbf{v}-\mathbf{v}')\delta_{il}\delta_{km}.$$

7. Formulas (10) and (11), as well as the analogous expressions for the transitions $A \rightarrow C$ and $B \rightarrow D$, are the main result of the foregoing analysis. They generalize the scalar theory developed in^[2,4,8] for spontaneous emission in an external field to include the case of emission in a magnetic field for arbitrary polarizations of the incident classical field and of the secondary radiation. At $\mathbf{E}_L = \mathbf{H} = 0$ these expressions describe the ordinary spontaneous emission of atoms. Thus, for example, for immobile atoms we then have

$$\begin{aligned} \rho_{a'a_i}^{(0)} &= (\lambda_a/\gamma_a)\delta_{a'a_i} & \rho_{b'a_i; ba}^{(0)} &= \exp\{-(\gamma_{ab} - i\omega_{ab})(t-t')\}\delta_{b'a_i}\delta_{a'a}, \\ \rho_{a'b_i}^{(0)} &= 0; & \gamma_{ab} &= \frac{1}{2}(\gamma_a + \gamma_b) \end{aligned}$$

and from (10) we readily obtain

$$J_\mu^{(0)} = \frac{\hbar\omega_\mu}{8\pi^2} \frac{\gamma_{a \rightarrow b} \lambda_a (2j_A + 1)}{\gamma_a} \frac{\gamma_{ab}}{\gamma_{ab}^2 + (\omega_\mu - \omega_{ab})^2}$$

The spectrum described by formula (12) for moving atoms will obviously be a convolution of the Doppler and Lorentz contours. The introduction of the magnetic field \mathbf{H} causes splitting of the spectrum into several components with different polarizations (the Zeeman effect). The classical field \mathbf{E}_L changes not only the quantities ρ_{aa} (Bennett dips appear) and $\rho_{ba}; ba$, but also lead to the appearance of terms ρ_{ab} connected with the optical coherence of the ensemble of atoms^[4,8]. In addition, if the polarization of the field \mathbf{E}_L is coherent⁽⁵⁾, the interaction V changes the atoms of the ensemble from states with definite values of the projection of the total angular momentum, i.e., from the Zeeman sublevels on which they fall as a result of the incoherent pumping, into a superposition of such states. This Hertz coherence of the ensemble of atoms leads to new effects that appear in the secondary-radiation spectrum. Usually in the study of resonant phenomena connected with the Hertz coherence of atomic states, such as the Hanle effect, level crossing, scattering of modulated light, etc. one is interested in the power of a definite polarization averaged over the frequency^[11-13]

$$P_\mu = \int_0^\infty J_\mu d\omega_\mu.$$

Since the secondary-radiation spectra J_μ are concentrated in the optical band, the lower limit of the integration can be replaced by $-\infty$. It then follows from (12) that

$$P_\mu = 2\pi C_{AB} \sum_{aa'b} D_{aa'}^{bb}(\mathbf{e}^{(u)}) \int_{-\infty}^\infty \rho_{a'a}(\mathbf{r}, \mathbf{v}, t) d\mathbf{v}.$$

We have obtained the well-known formula for the radiation intensity of atoms in a superposition of states, and

$$\sum_b D_{aa'}^{bb}(\mathbf{e}^{(u)})$$

is the density matrix^[11].

8. Expressions (10) and (12) are valid for a classical

field \mathbf{E}_L of arbitrary form. The only requirement is resonance with the transition $A \rightarrow B$. When the field \mathbf{E}_L is monochromatic, i.e.,

$$\mathbf{E}_L = \mathbf{E}\varphi(\mathbf{r})e^{-i\omega_L t} + \mathbf{E}^*\varphi^*(\mathbf{r})e^{i\omega_L t},$$

where $\varphi(\mathbf{r})$ is a certain function of \mathbf{r} , then the power of the secondary radiation can be shown to be independent of the time. The expression for the power is particularly simple in the important particular case when the field \mathbf{E}_L constitutes one traveling wave:

$$\mathbf{E}_L = \mathbf{E} \exp\{-i(\omega_L t - \mathbf{k}\mathbf{r})\} + \mathbf{E}^* \exp\{i(\omega_L t - \mathbf{k}\mathbf{r})\}. \quad (14)$$

It is convenient here to change from the density matrix elements to the following system of quantities:

$$\begin{aligned} r_{ki} &= \rho_{ki} \exp\{iq_{ki}(\omega_L t - \mathbf{k}\mathbf{r})\}, \\ r_{b'i'; ki} &= \rho_{b'i'; ki} \exp\{iq_{ki}(\omega_L t - \mathbf{k}\mathbf{r}) - iq_{b'i'}(\omega_L t' - \mathbf{k}\mathbf{r}')\}, \\ r_{c'iv'; ci} &= \rho_{c'iv'; ci} \exp\{iq_{ci}(\omega_L t - \mathbf{k}\mathbf{r}) - iq_{b'i'}(\omega_L t' - \mathbf{k}\mathbf{r}')\}, \\ r_{d'iv'; di} &= \rho_{d'iv'; di} \exp\{iq_{di}(\omega_L t - \mathbf{k}\mathbf{r}) - iq_{b'i'}(\omega_L t' - \mathbf{k}\mathbf{r}')\}. \end{aligned}$$

Here $\mathbf{k}, i, i' = a, b; q_{ab} = 1; q_{ba} = -1; q_{aa'} = q_{bb'} = 0; \mathbf{k}(\mathbf{r}-\mathbf{r}') = \mathbf{k} \cdot \mathbf{v}(t-t')$.

It is easy to verify by direct substitution that the introduced quantities satisfy, in the resonance approximation, systems of linear differential equations with constant coefficients. Consequently, the elements of the matrix Green's function $r_{lm}; l'm'$ depend only on the time difference $t-t'$, and the undamped solutions of the inhomogeneous system r_{jk} do not depend on the time at all. Turning to formula (12), we find that the power of the secondary radiation is independent of the time and can be expressed in terms of the stationary values r_{jk} and the unilateral Fourier transformations of the quantities $r_{lm}; l'm'(t-t')$:

$$\begin{aligned} J_\mu &= C_{AB} \sum_{aa'bb'} \left\{ D_{aa'}^{bb'}(\mathbf{e}^{(u)}) \int_{-\infty}^\infty d\mathbf{v} \left[\sum_{q_i} r_{a'a_i}(\mathbf{v}) r_{b'i'; bi}(\omega_\mu' - \omega_L') \right. \right. \\ &\quad \left. \left. + \sum_{b_i} r_{a'i'; bi}(\mathbf{v}) r_{b'i'; ba}(\omega_\mu' - \omega_L') \right] \right\} + \text{c.c.} \quad (15) \end{aligned}$$

Here

$$\begin{aligned} r_{lm}; l'm'(\omega_\mu' - \omega_L') &= \int_0^\infty e^{-i(\omega_\mu' - \omega_L')\tau} r_{lm}; l'm'(\tau) d\tau, \\ \omega_\mu' &= \omega_\mu - \mathbf{k}_\mu \mathbf{v}, \quad \omega_L' = \omega_L - \mathbf{k}\mathbf{v}. \end{aligned}$$

The expressions for J_μ on the adjacent transitions $A \rightarrow C$ and $B \rightarrow D$ are obtained from (15) as well as from (10), except that ω_L' is replaced by $-\omega_L'$ for the $B \rightarrow D$ transition.

The systems of linear differential equations with constant coefficients for the quantities r can be solved exactly, i.e., without limitations on the value of the field \mathbf{E}_L . If the velocity distribution is Maxwellian, then in the Doppler limit ($\gamma/\mathbf{k}u \ll 1$) the integration with respect to the velocities in (15) reduces to integration of a rational fraction between infinite limits. Consequently, when the classical field \mathbf{E}_L is a single traveling wave, the secondary-radiation spectra can be obtained in principle for a field \mathbf{E}_L of arbitrary intensity, but their form may be quite complicated.

II. SPECTRUM OF SECONDARY RADIATION ON THE TRANSITION $A \rightarrow B$

1. It is well known that it is impossible to solve exactly the systems of equations for the atomic density-

⁵⁾Coherent polarization is that of a field consisting of a mixture of at least two of the σ^+ , σ^- and π polarized components^[11].

matrix elements in the formulas for the spectra if the field $\mathbf{E}_L(\mathbf{r}, t)$ is arbitrary. But even in the exactly solvable case, when \mathbf{E}_L is a single traveling wave, it is difficult to extract concrete information concerning the gas system from the spectra, since the latter are complicated. At the same time, interesting changes in the spontaneous-emission spectra, due to the classical field \mathbf{E}_L , appear already when \mathbf{E}_L is sufficiently small and $V^2 \ll \gamma^2$. In this case, the atomic density matrices can be found by using perturbation theory in terms of the interaction V . The power of the secondary radiation is then represented as a series in V^2

$$J_\mu = J_\mu^{(0)} + J_\mu^{(2)} + J_\mu^{(4)} + \dots$$

As already mentioned above, J_μ describes the usual spontaneous emission in a magnetic field. We shall henceforth confine ourselves to an investigation of the structure of $J_\mu^{(2)}$, which is proportional to the intensity of the classical field; the latter is chosen in the form of the traveling wave (14).

2. We introduce two right-hand Cartesian coordinate systems: a fixed system xyz which is rigidly connected with the gas system, and a movable system $\xi\eta\zeta$ with ζ axis along the variable direction of observation of the secondary radiation ($\mathbf{k}_\mu = k_\mu \mathbf{e}_\zeta$). The rotation of the system $\xi\eta\zeta$ relative to xyz is specified by the Euler angles φ, θ, ψ , which are defined in accordance with^{[18]6)}. We direct the z axis along the magnetic field \mathbf{H} and choose it to be the quantization axis. We assume that the field \mathbf{E}_L propagates in the same direction, i.e., $\mathbf{k} = k\mathbf{e}_z$.

As is well known, the increment of the spontaneous emission $J_\mu^{(2)}$ due to the field \mathbf{E}_L acquires the form of a sharp structure only when the secondary radiation is observed in a direction that coincides with the propagation direction of the wave \mathbf{E}_L , and in the direction opposite to it^[2-6]. The reason for this is that under the resonance condition (1), which determines the atoms that interact effectively with the field \mathbf{E}_L , only the projection of the atom velocity on the propagation direction of the field \mathbf{E}_L is of importance. We shall therefore consider two cases: 1) $\theta = 0$ and 2) $\theta = \pi$, when $\varphi = 0$ and ψ is arbitrary. More accurately, the angle θ between the wave vectors \mathbf{k} and \mathbf{k}_μ should not differ from zero or π by more than γ/ku ^[2]. In the Doppler limit of interest to us we have $\gamma/ku \ll 1$.

Without going into the details of the calculations, we present the final results obtained for the $A \rightarrow B$ transition under the assumption that all the g factors of the levels in question are equal⁷⁾:

1) In the case $\theta = 0$ we have

$$J_\mu^{(2)} = \mathcal{A} \sum_{q=\pm 1} \left\{ |E_q|^2 \left[\frac{G_0(j_A, j_B) |e_q^{(\mu)}|^2 \gamma_{ab}}{\gamma_a [\gamma_b^2 + (\omega_\mu - \omega_L)^2]} + \frac{G_1(j_A, j_B) |e_{-q}^{(\mu)}|^2 \gamma_{ab}}{\gamma_b [\gamma_b^2 + (\omega_\mu - \omega_L + 2q\Omega)^2]} \right] \right. \\ \left. + \operatorname{Re} E_{-q}(E_q)^* e_q^{(\mu)} (e_{-q}^{(\mu)})^* \frac{G_2(j_A, j_B) e^{-2iq\varphi}}{(\gamma_a - 2iq\Omega) [\gamma_b + i(\omega_\mu - \omega_L)]} \right\}. \quad (16)$$

2) In the case $\theta = \pi$ we have

$$J_\mu^{(2)} = \mathcal{A} \sum_{q=\pm 1} \left\{ |E_q|^2 \left[\frac{G_0(j_A, j_B) |e_q^{(\mu)}|^2 \gamma_{ab}}{\gamma_a [4\gamma_{ab}^2 + (\omega_\mu + \omega_L - 2\omega_{ab} - 2q\Omega)^2]} \right. \right. \\ \left. \left. + \frac{G_1(j_A, j_B) |e_q^{(\mu)}|^2 \gamma_{ab}}{\gamma_a [4\gamma_{ab}^2 + (\omega_\mu + \omega_L - 2\omega_{ab})^2]} \right] + \operatorname{Re} E_{-q}(E_q)^* e_{-q}^{(\mu)} (e_q^{(\mu)})^* \right. \\ \left. \times \frac{G_2(j_A, j_B) e^{2iq\varphi}}{(\gamma_a - 2iq\Omega) [2\gamma_{ab} + i(\omega_\mu + \omega_L - 2\omega_{ab} - 2q\Omega)]} \right\}. \quad (17)$$

Here

$$\mathcal{A} = \frac{-3\gamma_{a \rightarrow b} |j_A||j_B| |^2 N(2j_A + 1)\omega_\mu}{4\pi^3 \hbar k u}, \quad N = \frac{\lambda_a}{\gamma_a} - \frac{\lambda_b}{\gamma_b}, \quad \gamma_{ab} = \frac{\gamma_a + \gamma_b}{2}$$

$G_\alpha(j_A, j_B)$ are functions of the total angular momenta j_A and j_B ⁸⁾, Ω is the frequency distance between the Zeeman sublevels, $\mathbf{E}_{\pm 1} = \mp (\mathbf{E}_x \mp i\mathbf{E}_y)/\sqrt{2}$ and $\mathbf{e}_{\pm 1}^{(\mu)} = \mp (e_\xi^{(\mu)} \mp ie_\eta^{(\mu)})/\sqrt{2}$. The polarizations of the field \mathbf{E}_L and of the secondary radiation, characterized by the index q , are considered in two different coordinate systems connected with the directions of the wave vectors \mathbf{k} and \mathbf{k}_μ . Here \mathbf{E}_q and $\mathbf{e}_q^{(\mu)}$ correspond to identical circular polarization when $\theta = 0$, and \mathbf{E}_q and $\mathbf{e}_{-q}^{(\mu)}$ when $\theta = \pi$.

The structure $J_\mu^{(2)}$ described by formulas (16) and (17) consists of two essentially different parts. The terms proportional to G_0 and G_1 correspond to the emission of photons μ with right- or left-hand circular polarization by atoms at definite Zeeman sublevels. The terms $\sim G_2$ occur for radiation by atoms in a coherent superposition of Zeeman states, and differ from zero only at coherent polarizations of the field \mathbf{E}_L and of the secondary radiation. It is interesting to note that a contribution to the coherent part of the structure ($\sim G_2$) is made not only by the terms containing directly the Hertz coherence of the ensemble of atoms ($\rho_{a'a}^{(2)}$ in formulas (10) and (12) for the $A \rightarrow B$ transition), but also by the terms corresponding to the emission of the photons μ by atoms in a superposition of Zeeman states. In (10) and (12) these are the terms for which $a \neq a'$.

3. We consider first the case when the polarization of at least one of the two fields (\mathbf{E}_L or the secondary radiation) is not coherent. From (16) and (17) we see that in this case there remain only the terms $\sim G_0$ and G_1 , which we shall call noncoherent. These noncoherent terms are Lorentz dips (we assume for concreteness that $N > 0$) of width $2\gamma_b$ when the secondary radiation is observed in the direction $\theta = 0$ and $4\gamma_{ab}$ in the direction $\theta = \pi$.

If the field \mathbf{E}_L is circularly polarized ($\mathbf{E}_q \neq 0, \mathbf{E}_{-q} = 0$), then observation of $J_\mu^{(2)}$ in the same circular polarization gives rise to a dip $\sim G_0$ with a minimum at the classical-field frequency ω_L at $\theta = 0$ and at the frequency $2(\omega_{ab} + q\Omega) - \omega_L$ at $\theta = \pi$. Observation in circular polarization orthogonal to \mathbf{E}_L shows a dip $\sim G_1$ at the frequency $\omega_L - 2q\Omega$ at $\theta = 0$ and $2\omega_{ab} - \omega_L$ at $\theta = \pi$. We note that dips having the same circular polarization when observed in the direction $\theta = 0$ are symmetrical with respect to the central transition frequency ω_{ab} to the dips of the orthogonal circular polarization at $\theta = \pi$.

On the other hand, if the field \mathbf{E}_L consists of a sum of σ^+ and σ^- polarized components ($\mathbf{E}_q \neq 0, \mathbf{E}_{-q} \neq 0$), then observation of the secondary radiation in each of

⁶⁾These angles are designated α, β , and γ in^[18].

⁷⁾Without this assumption, it is impossible to sum the expression for $J_\mu^{(2)}$ at arbitrary values of the total angular momenta j_A and j_B over the projections of these momenta. When this condition is not satisfied, it is necessary to carry out separate calculations for each value of j_A and j_B .

⁸⁾They are given in explicit form in^[18], in the notation of which

the circular polarizations will each reveal two dips $\sim G_0$ and $\sim G_1$. The distance between these dips is 2Ω for both $\theta = 0$ and $\theta = \pi$. The positions of the minima of the structure coincide with the positions of the "own" and "foreign" Bennett dips^[14] on the spontaneous-emission Doppler contours. (See the Introduction).

The dependence of the structure of $J_\mu^{(2)}$ on the level scheme, i.e., on the values of the total angular momenta j_A and j_B , is contained in the quantities $G_\alpha(j_A, j_B)$; $\alpha = 0, 1, 2$. Thus, the quantity $G_1(j_A, j_B)$ determines the relative influence of the field \mathbf{E}_L of one circular polarization on the secondary radiation of the other circular polarization at specified j_A and j_B . Obviously, if at the level A there is not a single Zeeman sublevel from which both the σ^+ and σ^- transitions begin, then there is no such influence, since waves of opposite circular polarizations are connected with different atomic σ transitions. From the explicit expressions for $G_1(j_A, j_B)$ we see that $G_1 = 0$ for the transitions $1 \rightarrow 0$, $3/2 \rightarrow 1/2$, and $1/2 \rightarrow 1/2$ ^[19], which are transitions of precisely this type.

4. We consider now the case of coherent polarizations of the field \mathbf{E}_L and of the secondary radiation. Let these polarizations be linear and let the angle between them be ψ . Then, directing the x axis along \mathbf{E} and the ξ axis along $\mathbf{e}(\mu)$, we obtain from (16) at $\theta = 0$

$$J_\mu^{(2)} = \frac{\mathcal{A} |E_x|^2}{4} \left\{ \frac{2G_0(j_A, j_B) \gamma_b}{\gamma_a [\gamma_b^2 + (\omega_\mu - \omega_L)^2]} + \sum_{q=\pm 1} \frac{G_1(j_A, j_B) \gamma_b}{\gamma_a [\gamma_b^2 + (\omega_\mu - \omega_L + 2q\Omega)^2]} + \frac{2G_2(j_A, j_B) (\gamma_a \gamma_b \cos 2\psi + 2\gamma_b \Omega \sin 2\psi)}{(\gamma_a^2 + 4\Omega^2) [\gamma_b^2 + (\omega_\mu - \omega_L)^2]} \right\}. \quad (18)$$

The structure (18) as a function of the frequency ω_μ at fixed Ω constitutes three Lorentz dips of equal width $2\gamma_b$, a central one with the minimum at the classical-field frequency ω_L and two side minima symmetrical with respect to the former at distances $\pm 2\Omega$. The side dips are due to the noncoherent terms $\sim G_1$ and the central one is a sum of the noncoherent $\sim G_0$ and coherent $\sim G_2$ terms. With increasing magnetic field, the contribution of the coherent term decreases this being due to the destruction of the coherence between the Zeeman sublevels of the level A by the magnetic field. Indeed, as already mentioned, the appearance of the coherent term $\sim G_2$ is due to the Hertz coherence induced in the gas system. It therefore possesses peculiarities typical both of the secondary radiation (the character of the dependence on ω_μ) and of the Hanle effect (the character of the dependence on Ω). The coherent term as a function of Ω changes from a Lorentz contour of width $2\gamma_a$ to a dispersion contour when the angle ψ changes from zero to $\pi/4$. The dependence on the angle ψ is another peculiarity possessed only by the coherent term. Its contribution to the frequency spectrum can reverse sign when ψ is suitably changed. This makes it possible, in particular, to separate the coherent part of the structure (18) experimentally. It suffices to make the measurements at two values of ψ that differ by $\pi/2$, and subtract one result from the other.

The dependence of the structure on the angle ψ between the polarizations becomes particularly simple when $\mathbf{H} = 0$. Then

$$J_\mu^{(2)} = \frac{\mathcal{A} |E_x|^2}{2\gamma_a} \{G_0 + G_1 + G_2 \cos 2\psi\} \frac{\gamma_b}{\gamma_b^2 + (\omega_\mu - \omega_L)^2}. \quad (19)$$

The depth of the Lorentz dip is modulated when the angle ψ between the polarizations is varied⁹⁾. This appears particularly clearly for the $1 \rightarrow 0$ transition (for example the $2S_2 - 2P_1$ transition with $\lambda = 1.52 \mu$ in Ne), when $G_0 = G_2 = 1/9$, $G_1 = 0$, and the depth of modulation reaches 100%.

Observation of secondary radiation in coherent polarization, which is necessary for the appearance of the coherent part of the structure $J_\mu^{(2)} \sim G_2$, is possible when there exist Zeeman sublevels of the lower level B, at which both the σ^+ and σ^- transitions can terminate. When j_A and j_B are such that there are no such sublevels, $G_2(j_A, j_B)$ and there is no coherent term. From the expressions for $G_2(j_A, j_B)$ we see that $G_2 = 0$ for the transitions $0 \rightarrow 1$, $1/2 \rightarrow 3/2$, and $1/2 \rightarrow 1/2$ ^[19].

An expression analogous to (18) can readily be obtained for the case $\theta = \pi$ from formula (17), but will not be presented here.

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⁹⁾Such a modulation was observed experimentally in the case of amplification of a "weak" signal in the presence of a "strong" one^[9].

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