Spontaneous transitions of polarization

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A new type of spontaneous transition is considered, the spontaneous transition of polarization (or the off-diagonal element of the density matrix or the dipole moment or the optical coherence), similar to the spontaneous transitions of particles and magnetic coherence. The example of a close doublet is used to analyze the role of a spontaneous polarization cascade in forming the spectra of absorption, refraction, and spontaneous emission. The spontaneous polarization transition is found to provide an interference contribution to the spectrum similar to nonlinear interference effects. The spectral and amplitude properties of the doublet are established. Finally, the possibility of gain without population inversion at the expense of the energy of the medium is demonstrated. © *1995 American Institute of Physics.*

1. INTRODUCTION

Spontaneous radiative processes can be broken down into three types. First, we have the spontaneous transitions of atoms (particles) from one stationary state to another, or what is known as Bohr transitions (see, e.g., Ref. 1). Transitions of the second type consist in the transfer to a lower level of the coherence (correlation) between the magnetic levels of an upper state.[2–6] In Ref. 7 the present author pointed out the existence of spontaneous radiative transitions of a third type, the transfer of polarization (or the dipole moment or the optical coherence) from one transition to another.

In all three types the transitions occur downward, i.e., from a state with a higher energy to a state with a lower energy. In other words, spontaneous transitions are of the cascade category, in contrast to similar collisional processes, which may occur in both directions, upward and downward.

Despite the cascade nature, only spontaneous transitions of the first type are accompanied by an increase in the energy of the electromagnetic field: the intensity of spontaneous emission related to Bohr transitions is given by the well-known relationship

$$I=\hbar\omega_{mn}A_{mn}N_m, \tag{1.1}$$

where ω_{mn} and A_{mn} are the Bohr frequency and the first Einstein coefficient for the $m-n$ transition, and N_m is the population of the upper level m. By themselves transitions of the second and third types do not give rise to photon emission; they affect only the shape of the emission spectrum and do not change the total emission intensity. In other words, transitions of the second and third types determine the interference effects in the emission, absorption, and scattering spectra.

The main goal of this paper is to analyze the interference phenomena related to a spontaneous cascade of polarization.

2. THE MAIN RELATIONSHIPS

We start with the kinetic equation for the one-particle density matrix in the JM-representation (see, e.g., Refs. 4 and 5):

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\cdot\nabla+i\omega_{mn}\right)\rho_{mn}(MM')=R_{mn}(MM')+S_{mn}(MM')+i(\rho V-V\rho)_{mM,nM'}. \tag{2.1}$$

The labels m and n number the stationary states. The second and third terms on the right-hand side of Eq. (2.1) describe the contributions of collisions $S_{mn}(MM')$ and the external field to the element $\rho_{mn}(MM')$ of the density matrix. At this point we are interested in the radiative relaxation matrix $R=-R^{(1)}+R^{(2)}$, which consists of the outgoing term $-R^{(1)}$ and the incoming term $R^{(2)}$. It is the latter that describes spontaneous cascades of polarizations from the m_1-n_1 to the $m-n$ transitions:

$$R^{(1)}_{mn}(MM')=\Gamma_{mn}\rho_{mn}(MM'),\quad \Gamma_{mn}=(\Gamma_m+\Gamma_n)/2, \tag{2.2}$$

$$R^{(2)}_{mn}(MM')=\sum_{m_1M_1n_1M_1'}A(mMnM'|m_1M_1n_1M_1')\times\rho_{m_1n_1}(M_1M_1'), \tag{2.3}$$

$$A(mMnM'|m_1M_1n_1M_1')=\sqrt{A_{m_1m}A_{n_1n}}\sum_\sigma\langle J_mM1\sigma|J_{m_1}M_1\rangle\langle J_nM'1\sigma|J_{n_1}M_1'\rangle.$$

Here Γ_i and J_i are the rate of spontaneous decay and the total angular momentum of the i state; i, M, M_1, etc. are the corresponding magnetic numbers, and $\langle\cdots|\cdots\rangle$ is a vector addition coefficient. According to (2.3), the polarization $\rho_{m_1n_1}(M_1M_1')$ in the m_1-n_1 transitions can generate the polarization $\rho_{mn}(MM')$ on the $m-n$ transition if the Einstein coefficients A_{m_1m} and A_{n_1n} are nonzero. In Fig. 1 this process is depicted symbolically by the two dashed arrows.

The transfer coefficient $A(mMnM'|m_1M_1n_1M_1')$ results from the interaction of each field oscillator with the two

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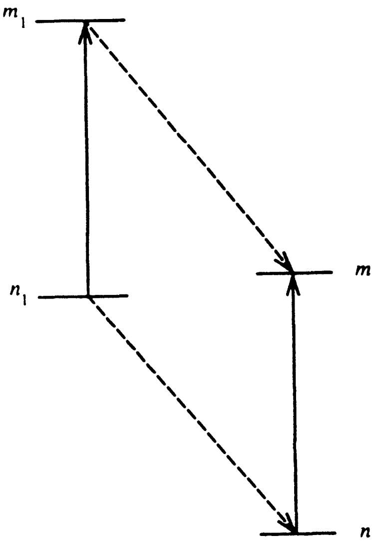

FIG. 1. A system of four optically coupled levels. The dashed arrows depict polarization transfer.

transitions, m_1-m and n_1-n, of the atom, i.e., it reflects the interference of these transitions. The geometric mean of the Einstein coefficients A_{m_1m} and A_{n_1n} emphasizes the interference nature of the polarization transfer $m_1-n_1\rightarrow m-n$. This is also shown by the fact that the transfer coefficient can be either positive or negative.

Equation (2.3) can be considered the most general. The rates of spontaneous cascades of the first and second types can be obtained from it as particular cases. If we put $m_1=n_1$ and $m=n$, we arrive at the following formula for the magnetic coherence transfer $m_1M_1M_1'\rightarrow mMM'$ (see Refs. 2–6):

$$R_{mm}^{(2)}(MM')=A_{m_1m}\sum_{\sigma M_1M_1'}\langle J_mM1\sigma|J_{m_1}M_1\rangle \times\langle J_mM'1\sigma|J_{m_1}M_1'\rangle\rho_{m_1m_1}(M_1M_1'). \quad (2.4)$$

The transfer of populations of magnetic sublevels is described by Eq. (2.4) if in it we put $M=M'$ and $M_1=M_1'$:

$$R_{mm}^{(2)}(MM)=A_{m_1m}\sum_{\sigma M_1}\langle J_mM1\sigma|J_{m_1}M_1\rangle^2\rho_{m_1m_1}(M_1M_1). \quad (2.5)$$

In contrast to (2.3) and (2.4), the transfer coefficients in (2.5) are positive, as they should be.

In the linear approximation (in field intensity) it is convenient to deal with what has become known as the κq-representation of the density matrix, which is defined by the following relationships:

$$\rho_{mn}(\kappa q)=\sum_{MM'}(-1)^{J_n-M'}\langle J_mMJ_n-M'|\kappa q\rangle\rho_{mn}(MM'), \quad (2.6)$$

$$\rho_{mn}(MM')=\sum_{\kappa q}(-1)^{J_n-M'}\langle J_mMJ_n-M'|\kappa q\rangle\rho_{mn}(\kappa q). \quad (2.7)$$

The incoming term $R_{mn}^{(2)}(\kappa q)$ in the κq-representation has the form

$$R_{mn}^{(2)}(\kappa q)=\sum_{m_1n_1}A(mn|m_1n_1,\kappa)\rho_{m_1n_1}(\kappa q),$$
$$A(mn|m_1n_1,\kappa)=\sqrt{A_{m_1m}A_{n_1n}}\,(-1)^{J_m+J_{n_1}+1+\kappa}\times\sqrt{2J_{m_1}+1}\,\sqrt{2J_{n_1}+1}\begin{Bmatrix}J_{m_1} & J_{n_1} & \kappa\\ J_n & J_m & 1\end{Bmatrix}. \quad (2.8)$$

Obviously, this term is diagonal in κq and $A(mn|m_1n_1,\kappa)$ is independent of q because the field oscillators responsible for the spontaneous transitions are isotropic. For $\kappa=1$, the most important case in linear spectroscopy, an additional symmetry property emerges:

$$K=\frac{\sqrt{2J_m+1}}{2J_{m_1}+1}\frac{A(mn|m_1n_1,1)}{\sqrt{A_{m_1m}A_{n_1n}}}=\frac{\sqrt{2J_{n_1}+1}}{2J_{m_1}+1}\frac{A(n_1n|m_1m,1)}{\sqrt{A_{m_1n_1}A_{mn}}}=(-1)^{J_m+J_{n_1}}\sqrt{2J_m+1}\,\sqrt{2J_{n_1}+1}\begin{Bmatrix}J_{m_1} & J_{n_1} & 1\\ J_n & J_m & 1\end{Bmatrix}. \quad (2.9)$$

Thus, K proves to be symmetric with respect to interchange of the levels, $m\leftrightarrow n_1$, or else the rates of the polarization cascades $m_1-n_1\rightarrow m-n$ and $m_1-m\rightarrow n_1-n$ are linked by a simple relationship.

The rates of transfer of magnetic coherence of rank κ and population ($\kappa=0$) corresponding to Eqs. (2.4) and (2.5) can be found from (2.8):

$$A(mm|m_1m_1,\kappa)=A_{m_1m}(-1)^{J_{m_1}+J_m+1+\kappa}(2J_{m_1}+1)\times\begin{Bmatrix}J_{m_1} & J_{m_1} & \kappa\\ J_m & J_m & 1\end{Bmatrix}, \quad (2.10)$$

$$A(mm|m_1m_1,0)=A_{m_1m}\left(\sqrt{2J_{m_1}+1}/\sqrt{2J_m+1}\right), \quad (2.11)$$

which agrees with the results obtained in Refs. 2–6.

Note that for the spontaneous coherence transfer $m_1-n_1\rightarrow m-n$ the transitions m_1-m and n_1-n must be optically allowed ($A_{m_1m}\neq0$ and $A_{n_1n}\neq0$). On the other hand, the m_1-n_1 and $m-n$ transitions may be either allowed or forbidden. The transfer relationships (2.3) and (2.8) are valid irrespective of the mechanism of formation of the transferred coherence in the m_1-n_1 transition, the one important thing being that $\rho_{m_1n_1}\neq0$. If the m_1-n_1 transition is optically allowed, $\rho_{m_1n_1}$ can be generated by a one-photon process. For a forbidden transition $\rho_{m_1n_1}$ can be generated by multiphoton processes. An example of transfer of coherence of an optically forbidden transition is the transfer of magnetic coherence (the $m_1M_1-m_1M_1'$ transition is optically forbidden).

Generally speaking, it is clear *a priori* that polarization transfer (e.g., $m_1-n_1\rightarrow m-n$; see Fig. 1) is effective if the

Bohr frequencies $\omega_{m_1n_1}$ and ω_{mn} differ little. The largest possible effect should be expected when the following conditions are met:

$$|\omega_{m_1n_1}-\omega_{mn}|=|\Delta|<\Gamma_{mn},\ \Gamma_{m_1n_1}. \qquad (2.12)$$

However, even for larger values of $|\Delta|$ a spontaneous polarization cascade may be noticeable.

3. THE STRUCTURE OF SPECTRAL DOUBLETS

Let us take a system of four levels m_1, n_1, m, and n (Fig. 1) in which the m_1-n_1, $m-n$, m_1-m, and n_1-n transitions are allowed. If we assume that each state evolves independently, the emission or absorption spectrum of such a system consists of four single lines with the central frequencies ω_{mn}, $\omega_{m_1n_1}$, ω_{m_1m}, and ω_{n_1n}. Suppose that these lines are pairwise coincident or almost coincident and the frequency differences

$$\omega_{m_1n_1}-\omega_{mn}=\omega_{m_1m}-\omega_{n_1n}=\Delta \qquad (3.1)$$

are fairly small, i.e., the spectrum of the system consists of two close doublets. We want to establish how a spontaneous coherence cascade changes the profiles of these doublets.

We assume that the simplest conditions are met: isotropic excitation, absence of collisions, the dipole nature of the interaction V with the field,[1,4,5] and a weak monochromatic field of frequency ω, which has a negligible effect on level population. In the linear approximation it is convenient to use the κq-representation (2.6) and (2.7), in which the equations for the desired elements $\rho_{ij}(\kappa q)$ of the density matrix assume the form

$$[\Gamma_1-i(\Omega-\Delta)]\rho_{m_1n_1}(1q)=iG_{1q}N_{n_1m_1},\quad \Gamma_1\equiv\Gamma_{m_1n_1}, \qquad (3.2)$$

$$(\Gamma-i\Omega)\rho_{mn}(1q)=iG_qN_{nm}+A(mn|m_1n_1,1)\rho_{m_1n_1}(1q),$$

$$\Gamma\equiv\Gamma_{mn}, \qquad (3.3)$$

where we have introduced the notation

$$\Omega=\omega-\omega_{mn},\quad G_q=\frac{d_{mn}}{2\sqrt{3}\,\hbar}E_q,\quad G_{1q}=\frac{d_{m_1n_1}}{2\sqrt{3}\,\hbar}E_q, \qquad (3.4)$$

$$N_{n_1m_1}=\frac{\rho_{n_1n_1}(00)}{\sqrt{2J_{n_1}+1}}-\frac{\rho_{m_1m_1}(00)}{\sqrt{2J_{m_1}+1}},$$

$$N_{nm}=\frac{\rho_{nn}(00)}{\sqrt{2J_n+1}}-\frac{\rho_{mm}(00)}{\sqrt{2J_m+1}}. \qquad (3.5)$$

Here d_{mn} is the reduced matrix element of the dipole moment for the $m-n$ transition, and E_q is the circular component of the field. The quantities N_{ij} are the differences in the populations of the magnetic sublevels of the states i and j. Spontaneous population cascades are included in the N_{ij}.

By combining (3.2) and (33) we find the absorbed field power P (the work done by the field per unit time):

$$P=-2\hbar\omega\mathrm{Re}\left\{i\sum_q[\rho_{m_1n_1}(1q)G_{1q}^*+\rho_{mn}(1q)G_q^*]\right\}$$

$$=2\hbar\omega\mathrm{Re}\sum_q\left\{\frac{N_{nm}}{\Gamma-i\Omega}|G_q|^2+\frac{N_{n_1m_1}}{\Gamma_1-i(\Omega-\Delta)}\times\left[|G_{1q}|^2+\frac{G_q^*G_{1q}}{\Gamma-i\Omega}A(mn|m_1n_1,1)\right]\right\}. \qquad (3.6)$$

According to this expression, the absorption spectrum contains two standard Lorentzian lines with central frequencies $\Omega=0$ and $\Omega=\Delta$, which corresponds to the doublet $\omega_{mn}, \Omega_{m_1n_1}$, and an additional term caused by the polarization cascade. This last term is proportional to $N_{n_1m_1}$ and is independent of the population difference N_{nm}. This fact emphasizes the cascade origin of the third term in P.

Above, the process of a spontaneous polarization cascade was closely linked with the interference of the m_1-m and n_1-n channels, which is reflected in the structure of the coefficient $A(mn|m_1n_1,1)$ [Eq. (2.8)]. The cascade term in (3.6) exhibits another type of interference, that of the m_1-n_1 and $m-n$ channels, indicated by the product of matrix elements $G_q^*G_{1q}$ and the Lorentzian profiles. It is this interference process that leads to a situation in which the integral of the cascade term with respect to ω is zero (which is easy to prove). Thus, the cascade term in (3.6) can be said to be a "double-interference term."

We employ the well-known relationship between the reduced matrix elements d_{ij} and the Einstein coefficients A_{ij} (see Ref. 1),

$$|d_{ij}|^2=|d_{ji}|^2=\frac{3\hbar c^3}{4\omega^3}(2J_j+1)A_{ji}, \qquad (3.7)$$

to transform (3.6) in the following manner:

$$P=\alpha(\Omega)\frac{c}{8\pi}|E|^2,\quad |E|^2=\sum_q|E_q|^2,$$

$$\alpha(\Omega)=\frac{\lambda^2}{4\pi}\left\{\bar{N}_{nm}\frac{A_{mn}\Gamma}{\Gamma^2+\Omega^2}+\bar{N}_{n_1m_1}\times\left[\frac{A_{m_1n_1}\Gamma_1}{\Gamma_1^2+(\Omega-\Delta)^2}+\frac{A^2K}{\Gamma\Gamma_1}f(\Omega)\right]\right\} \qquad (3.8)$$

$$\bar{N}_{nm}=(2J_m+1)N_{nm},\quad \bar{N}_{n_1m_1}=(2J_{m_1}+1)N_{n_1m_1}, \qquad (3.9)$$

$$A^2=\sqrt{A_{m_1m}A_{n_1n}A_{m_1n_1}A_{mn}}, \qquad (3.10)$$

$$f(\Omega)=\mathrm{Re}\frac{\Gamma\Gamma_1}{(\Gamma-i\Omega)[\Gamma_1-i(\Omega-\Delta)]}=\frac{\Gamma\Gamma_1[\Gamma\Gamma_1-\Omega(\Omega-\Delta)]}{(\Gamma^2+\Omega^2)[\Gamma_1^2+(\Omega-\Delta)^2]}. \qquad (3.11)$$

The quantity $\alpha(\Omega)$ is the absorption coefficient (cm^{-1}). Equation (3.8) contains the usual factors $\bar{N}_{ij}A_{ji}$, typical of absorption coefficients in single lines. One is also struck by the presence in the cascade term of a remarkable

TABLE I. Values of K.

J_{m_1}	J_{n_1}	J_m	J_n	K
J	$J+1$	$J+1$	$J+2$	1
J	J	J	J	$1-1/J(J+1)$
J	J	$J+1$	$J+1$	$\sqrt{1-1/(J+1)^2}$
$J+1$	J	$J+2$	$J+1$	$\sqrt{1-4/(2J+3)^2}$
J	J	J	$J+1$	$1/(J+1)$
$J+1$	$J+1$	$J+1$	J	$-1/(J+1)$
J	J	$J+1$	J	$-\sqrt{2J+3}/(J+1)\sqrt{2J+1}$
$J+1$	$J+1$	J	$J+1$	$\sqrt{2J+1}/(J+1)\sqrt{2J+3}$
J	$J+1$	$J+1$	J	$1/(J+1)(2J+1)$
$J+1$	J	J	$J+1$	$1/(J+1)(2J+3)$

and unique combination of four Einstein coefficients A^2, which clearly indicates the double interference that occurs in the formation of this spectral structure.

The factor K is the result of a complex interplay of various M-channels and their interference. Bearing in mind the symmetry properties of K and the properties of the $6j$-symbols [see Eq. (2.9)], we arrive at the following conclusions. It appears that K can assume either positive or negative values, with

$$-1\leqslant K\leqslant 1. \tag{3.12}$$

There are 10 possible combinations of J_{m_1}, J_{n_1}, J_m, and J_n that yield different algebraic expressions for K. These are listed in Table I, which shows, among other things, that the cases $K=1$ and $K=-1$ also exist. There is a combination $(J,\ J+1,\ J+1,\ J+2)$ which yields $K=1$ for any value of J. As J grows, the value of K approaches either 1 or 0; the sequence of the particular cases in Table I is constructed according to this tendency. Note that $K=0$ holds either in the limit $J\to\infty$ or by selection rules (the later case is not included in Table I). Thus, the interference of transitions for finite values of J never yields exactly zero.

Let us now examine the spectral properties of the cascade term, which are specified by the function $f(\Omega)$. At $\Delta=0$ and $\Omega=0$ we have $f=1$, which yields

$$\alpha(\Omega=0)=\frac{\lambda^2}{4\pi}\{\bar{N}_{nm}A_{mn}\Gamma^{-1}+\bar{N}_{n_1m_1}\Gamma_1^{-1}\times[A_{m_1n_1}+KA^2\Gamma^{-1}]\}. \tag{3.13}$$

Thus, if the Bohr frequencies of two transitions, $\omega_{m_1n_1}$ and ω_{mn}, accidentally coincide, all three terms in the expression for $\alpha(\Omega)$ are of the same order of magnitude. This is also true when $|\Delta|\lesssim\Gamma,\ \Gamma_1$. Earlier we noted that a polarization cascade alters the relative contribution to $\alpha(\Omega)$ of precisely the "upper" transition (by a quantity of order $A^2/A_{m_1n}\Gamma\sim 1$).

When $\Delta=0$ holds, we have

$$f(\Omega)=\frac{\Gamma\Gamma_1(\Gamma\Gamma_1-\Omega^2)}{(\Gamma^2+\Omega^2)(\Gamma_1^2+\Omega^2)}. \tag{3.14}$$

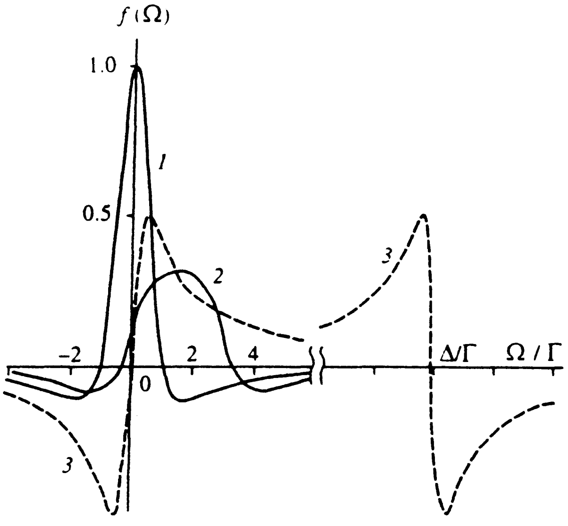

FIG. 2. The graphs of the function $f(\Omega)$: curve *1*, $\Delta=0$; curve *2*, $\Delta=3\Gamma$; and curve *3*, $\Delta=\Gamma$. The scale of curve *3* along the vertical axis is increased by a factor Δ/Γ-fold.

The function $f(\Omega)$ has a peak near the point $\Omega=0$, vanishes at $\Omega=\pm\sqrt{\Gamma\Gamma_1}$, and for large values of $|\Omega|$ becomes negative (curve *1* in Fig. 2). A simple calculation shows that at the minimum points $|f(\Omega)\leqslant 1/8|$ holds.

Now suppose that $\Delta\neq 0$. In an interval of values of $|\Delta|$ of order several halfwidths Γ and Γ_1, the function $f(\Omega)$ exhibits a broad maximum in the interval $0<\Omega<\Delta$ and retains negative "wings" (curve *2* in Fig. 2). When $|\Delta|/\Gamma$ and $|\Delta|/\Gamma_1$ are fairly large, a minimum appears at the center of the doublet $(\Omega\approx\Delta/2)$, and in the vicinity of the points $\Omega=0$ and $\Omega=\Delta$ a structure shaped like dispersion curves is formed (curve *3* in Fig. 2):

$$f(\Omega)\approx 4\Gamma\Gamma_1/\Delta^2,\quad \Omega=\Delta/2,$$
$$f(\Omega)\approx\frac{\Gamma_1}{\Delta}\frac{\Gamma\Omega}{\Gamma^2+\Omega^2},\quad |\Omega|\sim\Gamma, \tag{3.15}$$
$$f(\Omega)\approx-\frac{\Gamma}{\Delta}\frac{\Gamma_1(\Omega-\Delta)}{\Gamma_1^2+(\Omega-\Delta)^2},\quad |\Omega-\Delta|\sim\Gamma_1.$$

We see that for $|\Delta|\gg\Gamma,\Gamma_1$ at points $\Omega=\pm\Gamma$ and $\Omega-\Delta=\pm\Gamma_1$ we have $|f|\approx\Gamma_1/2|\Delta|,\ \Gamma/2|\Delta|$, while at the center of the doublet f is of second order in the small parameter $(\Gamma\Gamma_1/\Delta^2)$.

In some problems (e.g., in calculating absorption in so-called transparency windows) the behavior of $f(\Omega)$ in the extreme limbs, i.e., for $|\Omega|\gg|\Delta|$, is of interest. In such conditions Eq. (3.8) yields

$$\alpha(\Omega)=\frac{\lambda^2}{4\pi}\frac{1}{\Omega^2}\{\bar{N}_{nm}A_{mn}\Gamma+\bar{N}_{n_1m_1}[A_{m_1n_1}\Gamma_1-A^2K]\}. \tag{3.16}$$

Hence in the extreme limbs the splitting of the doublet plays no role, which is quite natural. However, the cascade effect does not disappear and the contributions to $\alpha(\Omega)$ of the cascade and population terms are of the same order of magnitude, which was also the case when the splitting is small.

The results of analysis of the function $f(\Omega)$ and the well-known properties of "pure" Lorentzian contours in Eq. (3.8) for $\alpha(\Omega)$ lead us to the following conclusion. When the splitting is large ($|\Delta|\gg\Gamma$, Γ_1), the components of the doublet are well-resolved, and polarization cascades cause a slight shift of their peaks and a small asymmetry. If we have $K>0$, the components of the doublets shift toward one another, i.e., the cascade effect "attracts" components. When we have $K<0$, the components "repel" each other. When $|\Delta|$ decreases to a magnitude on the order of a few halfwidth Γ and Γ_1, these manifestations of the polarization cascade are retained: the resolution of the doublet increases for $K<0$ and decreases for $K>0$. In the limit $|\Delta|\ll\Gamma$, Γ_1 the case $K>0$ means that the central peak of the line becomes sharper. But if $K<0$ holds, in certain conditions a minimum may be retained at point $\Omega=0$. Indeed, it is easy to show that under the conditions

$$K<0,$$

$$\Gamma_1^{-3}\bar{N}_{n_1m_1}A_{m_1n_1}\left[\left(1+\frac{\Gamma_1}{\Gamma}+\frac{\Gamma_1^2}{\Gamma^2}\right)\frac{A^2|K|}{A_{m_1n_1}\Gamma}-1\right]>\Gamma^{-3}\bar{N}_{nm}A_{mn}, \qquad (3.17)$$

we have $d^2\alpha/d\Omega^2>0$ at the point $\Omega=0$. More than that, if the condition is more stringent, namely

$$K<0,$$

$$\Gamma_1^{-1}\bar{N}_{n_1m_1}A_{m_1n_1}[A^2|K|A_{m_1n_1}\Gamma-1]>\Gamma^{-1}\bar{N}_{nm}A_{mn}, \qquad (3.18)$$

$\alpha(\Omega)$ is negative near the center of the line.

These tendencies are illustrated by Fig. 3, which gives α (in arbitrary units) as a function of Ω for $\Delta=0$ and $\Gamma=\Gamma_1$. In this case

$$\alpha(\Omega)\propto\frac{1}{1+x^2}\left(1+\eta\frac{1-x^2}{1+x^2}\right),$$

$$x=\frac{\Omega}{\Gamma},\quad \eta=\frac{\bar{N}_{n_1m_1}A^2|K|/\Gamma}{\bar{N}_{n_1m_1}A_{m_1n_1}+\bar{N}_{nm}A_{mn}}. \qquad (3.19)$$

At this point it is proper to mention the role of the amplitude parameter $\xi=\bar{N}_{n_1m_1}A_{m_1n_1}/\bar{N}_{nm}A_{mn}$: the higher its value the greater the relative role of a polarization cascade [see, e.g., Eqs. (3.17)–(3.19)]. The same is true of the region $|\Omega|\sim\Gamma$ in the limit $|\Delta|\gg\Gamma$. Here

$$\alpha(\Omega)\propto\frac{1}{1+x^2}+\xi\frac{\Gamma}{\Delta}\frac{x}{1+x^2},\quad \xi=\frac{\bar{N}_{n_1m_1}A_{m_1n_1}}{\bar{N}_{nm}A_{mn}}, \qquad (3.20)$$

and the large value of ξ can compensate for the smallness of $\Gamma/|\Delta|$, so that the cascade term proves to be the leading one.

Negative absorption may exist not only at the center but also in the extreme limbs of the line. Equation (3.16) implies that for $|\Omega|\gg|\Delta|$ we have $\alpha<0$ if

$$K>0,\quad \Gamma_1\bar{N}_{n_1m_1}A_{m_1n_1}[A^2K/A_{m_1n_1}\Gamma_1-1]>\Gamma\bar{N}_{nm}A_{mn}. \qquad (3.21)$$

Note that in the inequalities (3.18) and (3.21) both N_{nm} and $N_{n_1m_1}$ are assumed positive, i.e., on both transitions,

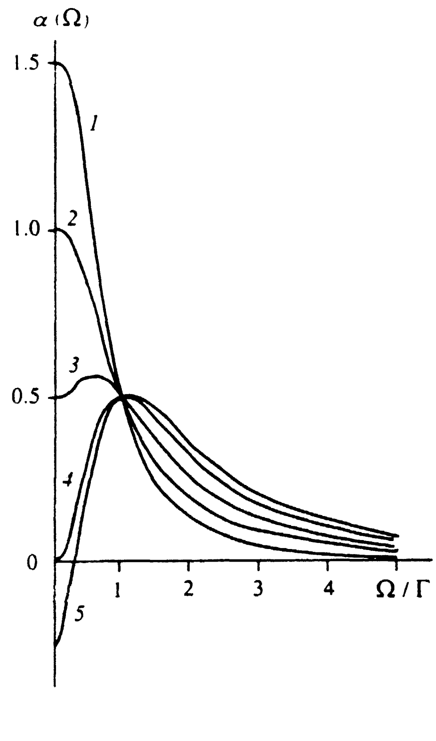

FIG. 3. The absorption coefficient curves according to Eq. (3.19) (arbitrary units) at $\Gamma=\Gamma_1$ and $\Delta=0$ for different values of the parameter η: curve *1*, $\eta=1/2$; curve *2*, $\eta=0$; curve *3*, $\eta=-1/2$, curve *4*, $\eta=-1$; and curve *5*, $\eta=-5/4$.

$m-n$ and m_1-n_1, the populations are not inverted. Thus, a polarization cascade may ensure radiation gain without population inversion.

The possibility of amplifying an electromagnetic field without population inversion is a characteristic feature of nonlinear interference effects. According to general theorems,[4,5] the integrated (over ω) contribution of nonlinear interference effects to the absorption spectrum is zero. Hence in some segments of the frequency scale this contribution is positive, while in others the contribution is necessarily negative. If in the latter frequency intervals the nonlinear interference contribution proves to be greater than the contribution of population effects, the net reaction of the medium is reduced to amplifying the field.

Gain without inversion was predicted in Ref. 8, was observed experimentally by Bonch-Bruevich *et al.*,[9] and lately has attracted much attention (see, e.g., the review by Agap'ev *et al.*)[10] All these numerous studies consider the amplification of a wave E_1 when the medium is simultaneously subjected to another laser or microwave field E_2. The increase in the energy of wave E_1, or its gain, can occur only at the expense of the energy of wave E_2. In this respect, in relation to the source of energy for amplification, gain without inversion is similar to lasing with multiple, difference, and sum frequencies.

In our problem an atom interacts with a monochromatic field. The interference effect is caused by a spontaneous cascade transition of polarization $\rho_{m_1n_1}\rightarrow\rho_{mn}$, which occurs, as it were, all by itself; more exactly, the zero-point vibrations of the field oscillators act as the second external wave. In this sense a polarization cascade may be considered the simplest nonlinear interference effect possible. The nonlinear nature of this interference effect can be judged from Eqs. (3.6) and (2.8), according to which the amplitude of the interference term contains the combination $G_q^* G_{1q}\sqrt{A_{m_1m}\,A_{n_1n}}$ proportional to the intensity of the external field, $(G_q^* G_{1q})$, and the intensity of the zero-point vibrations of the field oscillators, $(\sqrt{A_{m_1m}A_{n_1n}})$.

A natural question arises about the source of energy required for amplifying the external field. Since zero-point vibrations cannot supply the energy, it is obvious in general that the field is amplified at the expense of the energy of the medium. The paradox is that according to ordinary ideas a medium can give its energy to a field only in the presence of population inversion, while in our case the populations are not inverted.

First we note that the interaction of the levels m_1M_1 and n_1M_1' of the "upper transition" m_1-n_1 and the external field does not depend on whether there are levels mM and nM'. Hence the transition m_1-n_1 will absorb the field as in a simple two-level system with $\bar{N}_{n_1m_1}>0$, and atoms will go from level n_1 to level m_1. Hence if there is amplification of the field, it is related to the levels mM and nM'.

Equation (3.6) gives the work done by the external field (G_1, G_{1q}) on the current $(i\rho_{mn}, i\rho_{m_1n_1})$ induced by that field. If the induced current is in phase with the "driving force," there is absorption. But if the phase difference between field and current is changed by π, absorption changes to gain. Equations (3.2) and (3.3) imply that

$$\rho_{mn}(1q)=\frac{iG_q}{\Gamma-i\Omega}N_{nm}+\frac{iG_{1q}}{\Gamma_1-i(\Omega-\Delta)}N_{n_1m_1}\frac{A(mn|m_1n_1,1)}{\Gamma-i\Omega}. \tag{3.22}$$

The first term on the right-hand side yields the in-phase component of the current. The transport coefficient $A(mn|m_1n_1,1)/(\Gamma-i\Omega)$ in the second term is complex-valued and may contain various phase shifts. If $A(mn|m_1n_1,1)>0$ holds, then for $\Delta=0$ and small Ω the second term also describes in-phase current, which corresponds to absorption. However, for $|\Omega|>|\Delta|$ the phase of the second term changes by π, which means that it provides a negative contribution to P, i.e., induces $m\rightarrow n$ transitions of atoms notwithstanding the absence of population inversion. If $A(mn|m_1n_1,1)$ is negative, the second term is in phase in the limbs and in antiphase at the center of the doublet.

Thus, gain is possible if the contribution of the interference term to P is fairly large. In accordance with what we have said earlier, the energy needed to amplify the field is taken here from the medium at the expense of the $mM\rightarrow nM'$ transitions, which prevail over inverse transitions notwithstanding the absence of population inversion.

The above analysis prompts the following interesting general conclusion: gain without population inversion because of nonlinear interference may occur, in the presence of a second external field (E_2), not only at the expense of the energy of this field but also at the expense of the energy of the medium.

The absorption (emission) spectrum of a four-level system, m_1, n_1, m, and n, contains two doublets with central frequencies $\omega_{m_1n_1},\omega_{mn}$ and $\omega_{m_1m},\omega_{n_1n}$ and with the same splitting Δ [see Eq. (3.1)]. Phenomena similar to those considered above for the vicinity of the $\omega_{m_1n_1},\omega_{mn}$ doublet slso occur in the vicinity of the other doublet, $\omega_{m_1m},\omega_{n_1n}$: we need only substitute n_1 for m and m for n_1 in all the expressions and interpret Γ and Γ_1 as, respectively, the halfwidths Γ_{n_1n} and Γ_{m_1m} of the n_1-n and m_1-m transitions [see the definitions of Γ and Γ_1 in Eqs. (3.2) and (3.3)]. Note that the factor K does not change its value under the substitution $m\leftrightarrow n_1$, i.e., it is the same for both doublets. The total absorption spectrum is given by the sum of (3.8) and its analog with these substitutions.

4. THE REFRACTIVE INDEX

As is known, the imaginary part of the expression in braces in Eq. (3.6) is proportional to the contribution of the $m-n$ and m_1-n_1 transition to the refractive index, or the real part of the dielectric constant, $\delta\varepsilon'$, namely

$$\delta\varepsilon'=\frac{\lambda^3}{8\pi^2}\beta(\Omega), \tag{4.1}$$

$$\beta(\Omega)=\frac{\bar{N}_{nm}A_{mn}}{\Gamma}\frac{\Gamma\Omega}{\Gamma^2+\Omega^2}+\bar{N}_{n_1m_1}\times\left[\frac{A_{m_1n_1}}{\Gamma_1}\frac{\Gamma_1(\Omega-\Delta)}{\Gamma_1^2+(\Omega-\Delta)^2}+\frac{A^2K}{\Gamma\Gamma_1}\varphi(\Omega)\right], \tag{4.2}$$

$$\varphi(\Omega)=\operatorname{Im}\frac{\Gamma\Gamma_1}{(\Gamma-i\Omega)[\Gamma_1-i(\Omega-\Delta)]}=\frac{\Gamma\Gamma_1[\Gamma_1\Omega+\Gamma(\Omega-\Delta)]}{(\Gamma^2+\Omega^2)[\Gamma_1^2+(\Omega-\Delta)^2]}. \tag{4.3}$$

The first two terms in $\beta(\Omega)$ are ordinary dispersion functions. The third, the interference term, is essentially new. If we have $\Delta=0$, then $\varphi(\Omega)$ is an antisymmetric function of Ω, as the dispersion terms are, but with extrema at the points $|\Omega|\approx\sqrt{\Gamma\Gamma_1/3}$ and with more rapidly decaying wings (Ω^{-3}). Hence far from the doublet the cascade effect in ε' does not manifest itself, in contrast to ε'' or $\alpha(\Omega)$. If we have $\Delta\neq0$ and $\Gamma=\Gamma_1$, the function $\varphi(\Omega)$ is antisymmetric about to the point $\Omega=\Delta/2$ (the middle of the doublet). If $|\Delta|\gg\Gamma$, Γ_1 holds, then in the vicinity of the central frequencies of the double we have the following approximate expressions:

$$\varphi(\Omega)\approx-\frac{\Gamma_1}{\Delta}\frac{\Gamma^2}{\Gamma^2+\Omega^2},\quad |\Omega|\sim\Gamma,\ |\Delta|\gg\Gamma,\ \Gamma_1, \tag{4.4}$$

$$\varphi(\Omega)\approx\frac{\Gamma}{\Delta}\frac{\Gamma_1^2}{\Gamma_1^2+(\Omega-\Delta)^2},\quad |\Omega-\Delta|\sim\Gamma_1,\quad |\Delta|\gg\Gamma,\ \Gamma_1. \tag{4.5}$$

According to these relationships, $\varphi(\Omega)$ has different signs near the points $\Omega=0$ and $\Omega=\Delta$ and is described by symmetric Lorentzian contours.

The difference in the symmetry properties of the cascade and "ordinary" terms leads to more or less obvious departures from the standard Ω-dependence of $\delta\varepsilon'$, departures that become more obvious as $|\Delta|$ decreases.

5. DOPPLER BROADENING

The Doppler effect caused by the thermal motion of atoms can be taken into account if in the above formulas we make the substitution

$$\Omega \rightarrow \Omega' = \Omega - kv \tag{5.1}$$

and average over the appropriate distribution of the atoms in the projections of velocities v on the wave vector $\mathbf{k}$. We take the Maxwellian distribution

$$W(v)=\frac{1}{\sqrt{\pi}\bar{v}}\exp\left(-\frac{v^2}{\bar{v}^2}\right), \quad \bar{v}^2=\frac{2T}{m}, \tag{5.2}$$

where m and T are the mass of an atom and the temperature. It can be shown that

$$\begin{aligned}\langle f(\Omega')+i\varphi(\Omega')\rangle &= \int \frac{\Gamma\Gamma_1 W(v)dv}{[\Gamma-i(\Omega-kv)][\Gamma_1-i(\Omega-\Delta-kv)]} \\ &= \sqrt{\pi}\frac{\Gamma\Gamma_1}{(k\bar{v})^2}\frac{(k\bar{v})}{\Gamma_1+i\Delta-\Gamma}\left[w\left(\frac{\Gamma-i\Omega}{k\bar{v}}\right) - w\left(\frac{\Gamma_1+i\Delta-i\Omega}{k\bar{v}}\right)\right],\end{aligned} \tag{5.3}$$

where the function $w(z)$ is related to the error integral of a complex-valued argument in the following manner:[11]

$$w(z)=\exp(z^2)[1-\Phi(z)], \quad \Phi(z)=\frac{2}{\sqrt{\pi}}\int_0^z \exp(-t^2)dt. \tag{5.4}$$

For $\Gamma=\Gamma_1$ and $\Delta=0$ we have

$$\langle f(\Omega')+i\varphi(\Omega')\rangle = 2\left(\frac{\Gamma}{k\bar{v}}\right)^2[1-\sqrt{\pi}\,zw(z)], \quad z=\frac{\Gamma-i\Omega}{k\bar{v}}. \tag{5.5}$$

Note the presence of the factor $\Gamma\Gamma_1/(k\bar{v})^2$ in Eqs. (5.3) and (5.5), which means that when Doppler broadening is large, the interference effect is essentially suppressed. The reason for suppression is easily understood. When in the limit $k\bar{v} \gg \Gamma$ we average an isolated Lorentzian contour, the latter is assumed proportional to a δ function,

$$\left\langle \frac{\Gamma^2}{\Gamma^2+(\Omega-kv)^2}\right\rangle = \sqrt{\pi}\frac{\Gamma}{k\bar{v}}\exp\left\{-\frac{\Omega^2}{(k\bar{v})^2}\right\},$$

and because of averaging the amplitude of the profile decreases by a factor of $\Gamma/k\bar{v}$. For an interference profile such reasoning is meaningless since its integrated intensity is zero. A more accurate calculation leads to the additional factor $\Gamma/k\bar{v}$.

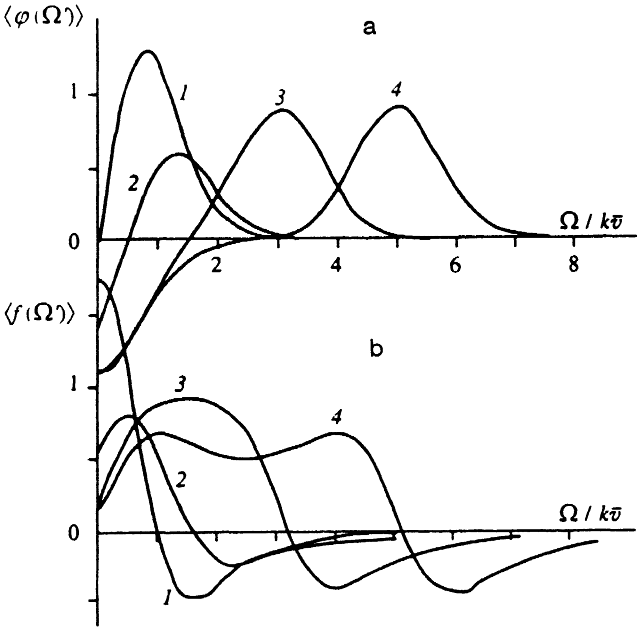

FIG. 4. The graphs of the functions (a) $\langle f(\Omega')\sqrt{\pi}\,10^2\rangle$ and (b) $\langle\varphi(\Omega')\rangle\sqrt{\pi}\,10^2$: curve *1*, $\Delta=0$ and $\Gamma=k\bar{v}/10=\Gamma_1/2$; curve *2*, $\Delta=k\bar{v}$ and $\Gamma=\Gamma_1=k\bar{v}/10$; curve *3*, $\Delta=3k\bar{v}$ and $\Gamma=\Gamma_1=k\bar{v}/10$; and curve *4*, $\Delta=5k\bar{v}$ and $\Gamma=\Gamma_1=k\bar{v}/10$. The curves *3* and *4* are multiplied by three and five, respectively.

In accordance with what has been said earlier, population effects are specified in order of magnitude by the factors $\bar{N}_{ji}A_{ij}/k\bar{v}$, while the cascade effect is specified by the factor $\bar{N}_{n_1m_1}A^2/(k\bar{v})^2$, where A^2 is a quadratic function of the Einstein coefficients. Hence the interference term is further suppressed by $A_{ij}/k\bar{v}$ ratio.

The Doppler width is proportional to ω, whereas A_{ij} is proportional to ω^3. Hence cascade polarization effects play a greater role when the doublet frequencies differ considerably and $\omega_{m_1m} \gg \omega_{mn}$.

Figure 4 depicts the results of calculating $\langle f\rangle$ and $\langle\varphi\rangle$ for several values of the parameters Δ/Γ and Γ_1/Γ. Qualitatively the curves have the same shape as in the case of immobile atoms, but here $k\bar{v}$ rather than Γ serves as the scale along the horizontal axis.

As is well known, the not-too-distant limbs of Doppler-broadened lines are actually Lorentzian.[11] Hence for $|\Omega| \gtrsim 2k\bar{v}$ and $|\Omega-\Delta| \gtrsim 2k\bar{v}$ we can ignore Doppler broadening and use the results of Secs. 3 and 4 (for rough calculations). For one thing, even in the near limbs the population and cascade terms are of the same order of magnitude, while the Doppler suppression manifests itself indirectly, because of the necessity of dealing with large values of $\Delta/k\bar{v}$ and $\Omega/k\bar{v}$.

6. SPONTANEOUS EMISSION

According to general rules, we can use the expression for the work P done by the field to calculate the spectral density of spontaneous emission:[4,5] in Eq. (3.6) we discard the populations of the lower levels n_1 and n in the quantities

$N_{n_1m_1}$ and N_{nm}, normalize the matrix elements G_q and G_{1q} in the proper manner, and change the sign. As a result the spectral intensity density of spontaneous emission, $I(\Omega)$, is given by

$$I(\Omega)=\frac{\hbar\omega}{\pi}\left\{N_m\frac{A_{mn}\Gamma}{\Gamma^2+\Omega^2}+N_{m_1}\left[\frac{A_{m_1n_1}\Gamma_1}{\Gamma_1^2+(\Omega-\Delta)}+\frac{A^2K}{\Gamma\Gamma_1}f(\Omega)\right]\right\}. \qquad (6.1)$$

The quantities

$$N_m=\sqrt{2J_m+1}\,\rho_{mm}(00),$$
$$N_{m_1}=\sqrt{2J_{m_1}+1}\,\rho_{m_1m_1}(00) \qquad (6.2)$$

are the total populations of the levels m and m_1.

Generally, the spectral properties of $I(\Omega)$ obey the laws discussed above in connection with the absorption coefficient $\alpha(\Omega)$. The quantities $a(\Omega)$ and $I(\Omega)$ differ because the population and interference terms enter Eqs. (3.6) and (6.1) in different proportions, fixed either by the population difference or by the populations of the upper levels. For one thing, this leads to a situation in which $I(\Omega)$ is positive (as it should be), while $\alpha(\Omega)$ can also be negative (at the center of the line for $K<0$ and in the limbs for $K>0$). Let us check whether $I(\Omega)$ is indeed positive.

An important fact here is that the m_1-m transition is optically allowed. Hence in addition to the polarization cascade, which is explicitly present in Eq. (6.1), there is also the $m_1\rightarrow m$ cascade of particles or populations. We can therefore write N_m as

$$N_m=\tilde{N}_m+N_{m_1}A_{m_1m}/\Gamma_m, \qquad (6.3)$$

where $\tilde{N}_m$ is the part of the population of the level m not related to the $m_1\rightarrow m$ cascade and caused by other excitation mechanisms. As a result,

$$I(\Omega)=\frac{\hbar\Omega}{\pi}\left\{\tilde{N}_m\frac{A_{mn}\Gamma}{\Gamma^2+\Omega^2}+N_{m_1}\psi(\Omega)\right\},$$
$$\psi(\Omega)=\frac{A_{m_1m}A_{mn}\Gamma}{\Gamma_m(\Gamma^2+\Omega^2)}+\frac{A_{m_1n_1}\Gamma_1}{\Gamma_1^2+(\Omega-\Delta)^2}+\frac{A^2K}{\Gamma\Gamma_1}f(\Omega). \qquad (6.4)$$

Clearly, the factor $\psi(\Omega)$ of N_{m_1} is positive. Suppose, for instance, that we have $K>0$ and $|\Omega|\gg|\Delta|$. Then it follows that

$$\psi(\Omega)=\frac{A_{m_1n_1}\Gamma}{\Omega^2}\left[\left(\frac{A^2}{\Gamma_1A_{m_1n_1}}\right)^2\frac{\Gamma\Gamma_1}{\Gamma_mA_{n_1n}}+1-\frac{A^2K}{\Gamma_1A_{m_1n_1}}\right]>0, \qquad (6.5)$$

since the discriminant $K^2-4\Gamma\Gamma_1/\Gamma_mA_{n_1n}$ of the quadratic trinomial (in the parameter $A^2/\Gamma_1A_{m_1n_1}$) is negative in view of the inequalities

$$K<1,\quad \Gamma=\frac{1}{2}(\Gamma_m+\Gamma_n)>\frac{\Gamma_m}{2},$$
$$\Gamma_1=\tfrac{1}{2}(\Gamma_{m_1}+\Gamma_{n_1})>\tfrac{1}{2}\Gamma_{n_1}\geq\tfrac{1}{2}A_{n_1n}. \qquad (6.6)$$

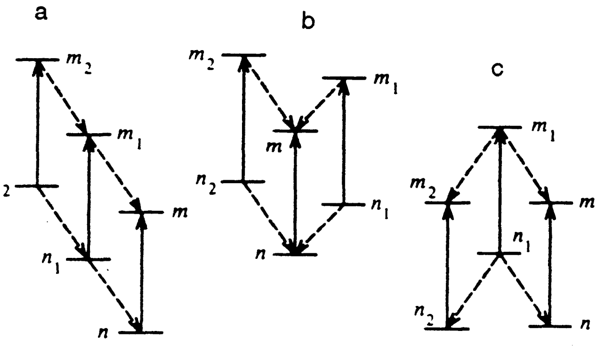

FIG. 5. The diagrams of spontaneous polarization cascades in triplets.

But if $K<0$ and $\Delta=0$ hold, then at the center of the line ($\Omega=0$) we have

$$\psi(\Omega=0)=\frac{A_{m_1n_1}}{\Gamma_1}\left[\left(\frac{A^2}{\Gamma A_{m_1n_1}}\right)^2\frac{\Gamma\Gamma_1}{\Gamma_mA_{n_1n}}+1-|K|\frac{A^2}{\Gamma A_{m_1n_1}}\right]>0, \qquad (6.7)$$

in view of the same inequalities (6.6).

Thus, because of a spontaneous polarization cascade in the spontaneous emission spectrum the doublet's components may change their shape, become asymmetric, and move together ($K>0$) or apart ($K<0$); "false splitting" as shown in Fig. 3 (curve *3*) may also occur.

7. DISCUSSION

Our study has revealed several effects in the spectra of absorption (gain), refraction, and spontaneous emission caused by a spontaneous polarization cascade. When two lines with different excitation potentials accidentally coincide (exactly or approximately), the spontaneous polarization cascade introduces an interference component whose spectral properties differ considerably from those of ordinary, population, profiles. In certain conditions favorable for interference manifesting itself the shape of close doublets may change considerably: the doublet components become asymmetric, the distance between the components changes, false splitting, and even amplification instead of absorption with noninverted populations may occur.

The system of four levels considered here (Fig. 1) is only one possible manifestation of a spontaneous polarization cascade: we selected it for its relative simplicity. With this example the physics of the process can be studied more easily. Examples of complex systems are also obvious: a double sequential polarization cascade $m_2-n_1\rightarrow m_1-n_1\rightarrow m-n$ (Fig. 5a), a V-type double parallel cascade $m_2-n_2\rightarrow m-n$, $m_1-n_1\rightarrow m-n$ (Fig. 5b), and a Λ-type double parallel cascade $m_1-n_1\rightarrow m-n$, $m_1-n_1\rightarrow m_2-n_2$ (Fig. 5c). Here we are speaking of triplets of accidentally coinciding lines and the exchange of polarization between them. Also there can be complications of another type, related, say, to the ideas and typical notions of nonlinear spectroscopy.

At present it is difficult to judge what applications the effects of polarization cascades will lead to. However, it can certainly be said that these effects must be taken into account in identifying spectral lines, in measuring the energies of stationary states, and in establishing oscillator strengths.

An important result, we believe, is the broadening of the physical picture of spontaneous transitions and the role these transitions play in spectroscopy. In this connection it is proper to recall the role of other mechanisms by which polarization is exchanged between various transitions. One mechanism is provided by collisions. Collisional polarization exchange or, as it is called, spectral exchange leads, for instance, to the collapse of inhomogeneously broadened spectral structures (for one thing, of the Q-branch of a rotational–vibrational spectrum)[4,5,12,13] and to Dicke narrowing of Doppler-broadened lines.[1,4,5,12,14] In contrast to the spontaneous cascade mechanism, collisional exchange is reciprocal, i.e., the right-hand side of Eq. (3.2) is expected to contain a term proportional to the polarization $\rho_{mn}(1q)$ of the $m-n$ transition. It is this reciprocal exchange that ensures the collapse of spectral structures. In addition to collisional stochastic exchange, there are many examples of dynamic exchange of polarizations due either to interaction with external electromagnetic fields[3–6,15] or to internal interactions (e.g., the Fermi resonance in the spectra of multiatomic molecules). On the phenomenological level our case is special in two respects: the simplicity and universality of the mechanism of polarization transfer, and the cascade nature of the process.

The above classification of spontaneous transitions into three types, the transitions of particles, magnetic coherence, and optical coherence (see Sec. 1), allows for an obvious expansion: an arbitrary coherent superposition of stationary states may spontaneously transform into another coherent superposition. Actual realization of a spontaneous cascade coherence transition of an arbitrary type with disintegration of coherence is, of course, associated with certain resonant conditions, specific for each case. These resonant conditions may be met only for a limited number of objects. Nevertheless, the widespread belief that optical coherence is irreversibly destroyed in spontaneous transitions cannot be considered universal and has its limits of application.

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