Collapse of spectral structures caused by stimulated radiative polarization exchange

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A theory is developed that explains the relaxation of an atom interacting with a random electromagnetic field. The radiative relaxation matrix incorporating particle transitions as well as the spontaneous and stimulated transfers of optical and magnetic coherence is calculated. Finally, the problem of radiative collapse of spectral doublets and magnetooptical resonances is discussed. © *1996 American Institute of Physics.* [S1063-7761(96)00908-0]

1. INTRODUCTION

The collapse of inhomogeneously broadened spectral structures is fairly widespread. The first area of research where this phenomenon was observed was, apparently, NMR spectroscopy, where it became known as motional narrowing of spectral lines (caused by molecular motion; see, e.g., Ref. 1). In the optical range collapse was discovered in the form of the Dicke decrease in the contribution of the Doppler effect to the linewidth[2–6] and as the collapse of the rotational structure of the spectra of IR absorption and Raman scattering of light in dense gases and in liquids.[4,7] The radiophysics analog of the collapse phenomenon is the transition from the "instrumental" broadening of the oscillation spectrum to the "natural."

Phenomenologically, collapse can be explained by frequency modulation as a result of a rapid change in frequency or, in spectral terms, by intense exchange of polarizations between the spectral components of the inhomogeneously broadened structure. From this viewpoint the above examples fall into a single pattern, the only difference being the exchange mechanism. Dicke narrowing and the collapse of the rotational structure are caused by exchange of polarizations in collisions. A similar reason may be considered the collisions of atoms with the walls of a vessel much smaller than the wavelength.[8,9] The narrowing of NMR lines is related to the translational motion of molecules, which ensures the averaging of inhomogeneities in the external molecular interactions.[1,10] Such averaging is the temporal equivalent of the spectral exchange pattern.

There is also one more collapse mechanism that has never been discussed before, spectral polarization exchange stimulated by "noise" radiation with a broad spectrum (e.g., thermal radiation). The present paper analyzes the radiative collapse mechanism.

Radiative relaxation includes three processes. First, the well-known spontaneous and stimulated transitions in atoms, processes introduced by N. Bohr and A. Einstein and accompanied by photon emission and absorption. These processes shorten the coherence lifetime and, hence, broaden the spectral lines, but do not lead to a collapse of the lines.[10–14] Collapse is caused by the transfer of polarization (or coherence) and not of particles. Spontaneous transfer of magnetic coherence (the correlations between the magnetic sublevels of the states) has been known for more than 30 years,[13–17] while the existence of spontaneous transfer of optical coherence was predicted not so long ago by the present author.[18–20] The spontaneous processes of coherence transfer lead to various interference phenomena, but they cannot be the reason for collapse either. The thing is that spontaneous transfer is of a "one-way" nature, since on the energy scale of stationary states it proceeds "downward" and no inverse spontaneous processes are possible. But collapse requires that there be mutual exchange of polarizations between the components of the structure, i.e., polarization transfer must occur "upward" and "downward." Only stimulated processes guarantee that there is mutual radiative exchange of polarizations. It is obvious that spontaneous polarization transfer, as any spontaneous process, must have a stimulated analog. Nevertheless, the existing theories of radiative relaxation do not mention stimulated polarization transfer.

Section 2 is devoted to a theory of radiative relaxation that incorporates both processes; spontaneous coherence transfer and stimulated coherence transfer. Section 3 examines stimulated radiative collapse of the simplest doublet spectral structure. Section 4 analyzes the role of radiative relaxation in the structure of magnetooptical resonances.

2. THE RADIATIVE RELAXATION MATRIX

We write the quantum kinetic equation for the one-particle density matrix ρ in the form[5,6]

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\cdot\nabla\right)\rho=R+S-i(V\rho-\rho V), \tag{2.1}$$

where $\mathbf{v}$ is the velocity of the atom, V is the Hamiltonian of interaction with coherent fields, and S and R are the collision integral and the radiative relaxation matrix. This section is devoted to the matrix R. Its calculation is done in the Appendix, while here we give only the results of calculations.

We start with the simplest diagram of four levels depicted in Fig. 1. The labels m, m_1, n, and n_1 number the stationary states of an isolated atom. The transitions m_1-m and n_1-n are assumed allowed in the dipole approximation, while the transitions m_1-n_1 and $m-n$ can be either allowed or forbidden. The Bohr transition frequencies are marked off on the horizontal axis in the lower half of Fig. 1. The following obvious relationships hold:

$$\omega_{m_1m}-\omega_{n_1n}=\omega_{m_1n_1}-\omega_{mn}\equiv\Delta. \tag{2.2}$$

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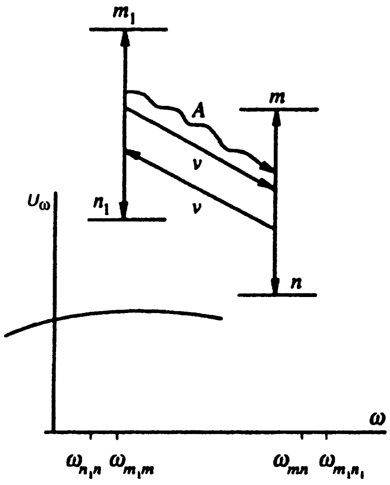

FIG. 1. A four-level system and the diagrams of spontaneous and stimulated polarization transfers.

The "noise" radiation with a broad spectrum $\Delta\omega$ is described by an average spectral bulk density U_ω. The spectrum expands over a frequency range that encompasses the $\omega_{m_1m}, \omega_{n_1n}$ doublet (Fig. 1).

The matrix R can be represented by two terms,

$$R=-R^{(1)}+R^{(2)}, \tag{2.3}$$

which by analogy with the collision integral are called the outgoing ($R^{(1)}$) and incoming ($R^{(2)}$) terms. In the model of nondegenerate states, the matrix elements R_{mn} and $R_{m_1n_1}$, which are present in Eq. (2.1) for ρ_{mn} and $\rho_{m_1n_1}$, can be written as follows:

$$\begin{aligned} R_{mn}^{(1)}&=\frac{1}{2}(\Gamma_m+\nu_m)\rho_{mn}+\rho_{mn}\frac{1}{2}(\Gamma_n+\nu_n),\\ R_{mn}^{(2)}&=(A_{mn,m_1n_1}+\nu_{mn,m_1n_1})\rho_{m_1n_1}e^{-i\Delta t}, \end{aligned} \tag{2.4}$$

$$\begin{aligned} R_{m_1n_1}^{(1)}&=\frac{1}{2}(\Gamma_{m_1}+\nu_{m_1})\rho_{m_1n_1}+\rho_{m_1n_1}\frac{1}{2}(\Gamma_{n_1}+\nu_{n_1}),\\ R_{m_1n_1}^{(2)}&=\nu_{m_1n_1,mn}\rho_{mn}e^{i\Delta t}. \end{aligned} \tag{2.5}$$

Here the Γ_j are the rates of spontaneous decay of the states $j=m,n,m_1,n_1$. The radiative outgoing frequencies ν_j are

$$\begin{aligned} \nu_m&=B_{mm_1}U_\omega, \quad \nu_n=B_{nn_1}U_\omega,\\ \nu_{m_1}&=B_{m_1m}U_\omega, \quad \nu_{n_1}=B_{n_1n}U_\omega, \end{aligned} \tag{2.6}$$

where the B_{ij} are the Einstein coefficients for the stimulated transition $i\rightarrow j$. Thus, the radiative outgoing frequencies in $R_{ij}^{(1)}$ are given by the arithmetic mean of the outgoing frequencies for the populations of levels i and j.

The incoming terms $R_{mn}^{(2)}$ and $R_{m_1n_1}^{(2)}$ describe the transfer of polarizations, or optical coherence, $\rho_{m_1n_1}$ and ρ_{mn} from the m_1-n_1 and $m-n$ transitions to the $m-n$ and m_1-n_1 transitions, respectively. The term A_{mn,m_1n_1} corresponds to spontaneous polarization transfer[18–20] and is present only in $R_{mn}^{(2)}$, since by convention the energies E_{m_1} and E_{n_1} of the levels m_1 and n_1 are higher than the energies E_m and E_n (Fig. 1). The terms ν_{mn,m_1n_1} and $\nu_{m_1n_1,mn}$ specify the polarization transfer stimulated by the radiation, and this process is of a reciprocal nature (as absorption and stimulated emission).

Coherence transfer processes are shown schematically in Fig. 1. The vertical double-tipped arrows connecting the pairs of levels m_1,n_1 and m,n symbolize polarization induced in the m_1-n_1 and $m-n$ transitions. The slanting single-tipped arrows connecting the vertical arrows symbolize coherence transfer processes: the wavy arrow (A) stands for the spontaneous process, and the straight arrows (ν) for the stimulated processes. The transitions of particles usually depicted by arrows connecting the levels are not shown in Fig. 1.

The incoming frequencies are given by the following formulas:

$$\begin{aligned} A_{mn,m_1n_1}&=\sqrt{A_{m_1m}A_{n_1n}}K,\\ \nu_{mn,m_1n_1}&=\nu_{m_1n_1,mn}-\sqrt{B_{m_1m}B_{n_1n}}U_\omega K, \end{aligned} \tag{2.7}$$

i.e., the frequencies are proportional to the geometric mean of the first and second Einstein coefficients. The factor K is of order unity, but in the model of nondegenerate states it is somewhat arbitrary, since actually it depends on the degeneracy of the states.

The matrix elements $R_{m_1m_1}$, $R_{m_1m_1}$, etc., can be obtained from Eqs. (2.4)–(2.7) by obvious label substitutions.

The above model of nondegenerate states is described by the simple kinetic scheme (2.4) and (2.5), which yields a graphic picture of the role of radiative relaxation. However, the results of calculations done in this model are only qualitative. For quantitative analysis and, the more so, for the case where polarization effects and external fields are taken into account, we must allow for the degeneracy of real states.

Let us examine states with total angular momenta J_j, $j=m,n,m_1,n_1$. In the JM-representation, the quantities ρ_{ij} and R_{ij} and the frequencies are matrices with respect to magnetic quantum numbers:

$$\begin{aligned} (\rho_{mn})_{MM'}&=\rho(mMnM'),\\ (\rho_{m_1n_1})_{M_1M_1'}&=\rho(m_1M_1n_1M_1'),\\ (R_{mn})_{MM'}&=R(mMnM'),\\ (R_{m_1n_1})_{M_1M_1'}&=R(m_1M_1n_1M_1'),\\ (\nu_j)_{MM'}&=\nu(jMjM'),\\ (\nu_{mn,m_1n_1})_{MM'M_1M_1'}&=\nu(mMnM'|m_1M_1n_1M_1'). \end{aligned} \tag{2.8}$$

In the Appendix we will see that R_{mn} and $R_{m_1n_1}$ in the JM-representation are given by the same formulas (2.4) and (2.5), but all the quantities in R_{mn} and $R_{m_1n_1}$ must be interpreted as MM'-matrices. Explicit expressions for the outgoing and incoming frequencies will be given below. Here we write the relationships for the matrix elements R_{mm} and $R_{m_1m_1}$ responsible for the radiative relaxation of the magnetic coherence of the $\rho(jMjM')$ type:

$$R^{(1)}_{mm}=\frac{1}{2}(\Gamma_m+\nu_m)\rho_{mm}+\rho_{mm}\frac{1}{2}(\Gamma_m+\nu_m),$$
$$R^{(2)}_{mm}=(A_{mm,m_1m_1}+\nu_{mm,m_1m_1})\rho_{m_1m_1}, \tag{2.9}$$

$$R^{(1)}_{m_1m_1}=\frac{1}{2}(\Gamma_{m_1}+\nu_{m_1})\rho_{m_1m_1}+\rho_{m_1m_1}\frac{1}{2}(\Gamma_{m_1}+\nu_{m_1}),$$
$$R^{(2)}_{m_1m_1}=\nu_{m_1m_1,mm}\rho_{mm}. \tag{2.10}$$

Similar equations hold for R_{nn} and $R_{n_1n_1}$. Comparison of the formulas (2.4) and (2.5) with (2.9) and (2.10) suggests that the radiative transfers of optical and magnetic coherences follow similar laws.

Equations (2.4), (2.5), (2.9), and (2.10) are valid if the change in the density matrix ρ in the course of the radiation correlation time $\tau_c=1/\Delta\omega$ is negligible.[10,11,16] Under certain conditions this imposes restrictions on the atomic characteristics, say, $\Delta\omega\gg\Gamma_j,|\Delta|$, etc.

The outgoing terms in R are well-known[11] and provide no new information. Our main goal is to study the incoming terms $R^{(2)}_{mn}$, $R^{(2)}_{m_1n_1}$, $R^{(2)}_{mm}$, and $R^{(2)}_{m_1m_1}$, which describe the exchange of polarizations. Notwithstanding its total randomness, an external perturbation is able to transfer both particles and coherence. The explanation lies in the fact that the same field oscillator mixes the atomic wave functions, both ψ_m,ψ_{m_1} and ψ_n,ψ_{n_1}, so that the random phases of the field oscillators do not manifest themselves in R. What is important here is the difference Δ of the Bohr frequencies of the "atomic oscillators" that interact with the field oscillator. When $|\Delta|$ is large, the incoming terms rapidly oscillate and prove to be unimportant. This fact, apparently, served as a psychological basis for ignoring the radiative transfer of optical coherence in previous research. Note that there are no oscillations in the $R^{(2)}_{jj}$ matrices when magnetic coherence is transferred in the absence of a static field. The ideas developed here also refer to the spontaneous and stimulated parts of $R^{(2)}$.

If the atom is in an external static field (magnetic or electric), the ω_{ij} depend on M and M' according to well-known laws,[21] and Δ is a matrix with respect to the magnetic quantum numbers. In particular, in the case of a magnetic field, for $\nu(mMnM'|m_1M_1n_1M_1')$ we have

$$\Delta=\omega_{m_1m}-\omega_{n_1n}+(g_{m_1}M_1-g_mM-g_{n_1}M_1'+g_nM')\mu H, \tag{2.11}$$

while for $\nu(mMmM'|m_1M_1m_1M_1')$ we have

$$\Delta=[g_{m_1}(M_1-M_1')-g_m(M-M')]\mu H. \tag{2.12}$$

Here H and g_j are the strength of the magnetic field and the g factor of the state j. We see that a magnetic field, as expected, destroys magnetic coherence transfer. In relation to optical coherence transfer a magnetic field may play an opposite role: the term proportional to H in Eq. (2.11) may balance the difference in the Bohr frequencies and decrease the oscillations in $R^{(2)}_{ij}$. A similar situation exists for a cascade of magnetic coherence in the presence of hyperfine level splitting.[22]

Note the structural similarity of the collision integral S in the model of relaxation constants (collisions do not change the atomic velocity) and the matrix R. The outgoing term $R^{(1)}$ consists of two terms: in one ρ is multiplied by ν from the left, and in the other the multiplication is from the right. The diagonal ($R^{(1)}_{ii}$ and $R^{(1)}_{jj}$) and off-diagonal ($R^{(1)}_{ij}$) matrix elements contain the same matrices ν_i and ν_j. The incoming terms contain a "supermatrix" that acts on both variables of the density matrix. The collision integral has the same properties.

According to Eqs. (2.4), (2.5), (2.9), and (2.10), radiative relaxation is not accompanied by a change in atomic velocity. If recoil is taken into account, the incoming term describes velocity mixing,[5,6,23] and radiative relaxation resembles collisional relaxation even more.

The formal similarity between R and S deserves attention also because of the seeming dissimilarity of the physical conditions of collisional and radiative perturbations of an atom. In deriving the kinetic equation with a collision integral of the Boltzmann type it is assumed that a short collision time is followed by a fairly long period of free motion. In the case of radiative relaxation, however, the external radiation represents a random stationary process without prolonged pauses. Notwithstanding such dissimilarity in conditions, radiative and collisional relaxation are described by similar terms in the kinetic equation.

As is the case with the collision integral, the radiative outgoing and incoming frequencies depend to a great extent on the symmetry of the perturbation. When the radiation is spherically symmetric, i.e., unpolarized and nondirectional, Eqs. (A21) and (A22) imply that

$$\nu(mMmM')=\nu_m\delta_{MM'},\quad \nu_m=B_{mm_1}U_\omega,$$
$$B_{mm_1}=|C_{mm_1}|^2, \tag{2.13}$$
$$\nu(mMnM'|m_1M_1n_1M_1')=C^*_{m_1m}C_{n_1n}U_\omega\sum_\sigma\langle J_mM1\sigma|J_{m_1}M_1'\rangle\times\langle J_nM'1\sigma|J_{n_1}M_1'\rangle,$$

$$C_{ij}=\frac{2\pi d_{ij}}{\hbar\sqrt{3}\sqrt{2J_i+1}}. \tag{2.14}$$

The spontaneous incoming rate is given by the following expression:[18–20]

$$A(mMnM'|m_1M_1n_1M_1')=\sqrt{A_{m_1m}A_{n_1n}}\sum_\sigma\langle J_mM1\sigma|J_{m_1}M_1\rangle\times\langle J_nM'1\sigma|J_{n_1}M_1'\rangle. \tag{2.15}$$

Thus, in the isotropic case the outgoing frequencies are specified by the rates of stimulated transitions of particles, are diagonal in the magnetic quantum numbers, and are independent of these numbers. On the other hand, there is similarity between the dependence of the spontaneous and stimulated incoming frequencies on the magnetic quantum numbers. The products of vector addition coefficients reflect interference between the $m_1M_1 \to mM$ and $n_1M_1' \to nM'$ transitions.

As is known,[16,24] when the perturbation of an atom is isotropic, the convenient representation for the density matrix is the κq-representation, which is specified by the following relationships:

$$\rho(mn\kappa q) = \sum_{MM'} (-1)^{J_n - M'} \langle J_m M J_n - M' | \kappa q \rangle \rho(mMnM'),$$

$$\rho(mMnM') = \sum_{\kappa q} (-1)^{J_n - M'} \langle J_m M J_n - M' | \kappa q \rangle \rho(mn\kappa q). \tag{2.16}$$

The quantities $\rho(mn\kappa q)$ are called the polarization moments of the level ($n=m$) or polarization moments ($n \neq m$) of rank κ.

Since in the isotropic case the $\nu(jMjM')$ are diagonal in M and are independent of M, in the κq-representation they are diagonal in κq, are independent of κq, and coincide with the values given by (2.13). The incoming frequencies are given by the following formulas:

$$\nu(mn\kappa q|m_1n_1\kappa_1q_1) = \delta_{\kappa\kappa_1}\delta_{qq_1}\nu(mn|m_1n_1,\kappa),$$

$$\nu(mn|m_1n_1,\kappa) = C^*_{m_1m}C_{n_1n}U_\omega K_\kappa, \tag{2.17}$$

$$A(mn\kappa q|m_1n_1\kappa_1q_1) = \delta_{\kappa\kappa_1}\delta_{qq_1}A(mn|m_1n_1,\kappa),$$

$$A(mn|m_1n_1,\kappa) = \sqrt{A_{m_1m}A_{n_1n}}K_\kappa, \tag{2.18}$$

$$K_\kappa = (-1)^{1+\kappa+J_{m_1}+J_n}\sqrt{2J_{m_1}+1} \times \sqrt{2J_{n_1}+1}\begin{Bmatrix} J_{m_1} & J_{n_1} & \kappa \\ J_n & J_m & 1 \end{Bmatrix}, \tag{2.19}$$

i.e., the result is diagonal in κq and independent of q. For $R^{(2)}_{mm}$ and $R^{(2)}_{m_1n_1}$ the following is true:

$$\nu(mm\kappa q|m_1m_1\kappa_1q_1) = \delta_{\kappa\kappa_1}\delta_{qq_1}\nu_{mm_1\kappa},$$

$$\nu_{mm_1\kappa} = B_{m_1m}U_\omega K_{1\kappa}, \tag{2.20}$$

$$A(mm\kappa q|m_1m_1\kappa_1q_1) = \delta_{\kappa\kappa_1}\delta_{qq_1}A_{mm_1\kappa},$$

$$A_{mm_1\kappa} = A_{m_1m}K_{1\kappa}, \tag{2.21}$$

$$K_{1\kappa} = (-1)^{1+\kappa+J_{m_1}+J_m}(2J_{m_1}+1)\begin{Bmatrix} J_{m_1} & J_{m_1} & \kappa \\ J_m & J_m & 1 \end{Bmatrix}. \tag{2.22}$$

Equations (2.21) are known from magnetooptical resonance theory[15–17] and are written here to make the picture complete. The frequencies of direct and reverse transitions obey the following relationships:

$$(2J_m+1)\nu_m = (2J_{m_1}+1)\nu_{m_1},$$

$$(2J_n+1)\nu_n = (2J_{n_1}+1)\nu_{n_1}, \tag{2.23}$$

$$\nu(mn|m_1n_1,\kappa) = \nu(m_1n_1|mn,\kappa), \quad \nu_{mm_1\kappa} = \nu_{m_1m\kappa}. \tag{2.24}$$

An analysis of the explicit expressions for $6j$ symbols shows that $K_{1\kappa}$ monotonically decreases as κ grows:

$$\nu_{mm_1\kappa} < \nu_{mm_10}. \tag{2.25}$$

Note that as κ increases, $K_{1\kappa}$ changes sign at $J_m = J_{m_1}$. The factor K_κ depends on four angular momenta, and its properties as a function of κ are complicated. For

$$J_{m_1} = J_m, \quad J_{n_1} = J_n; \quad J_{m_1} = J_m \pm 1, \quad J_{n_1} = J_n \pm 1, \tag{2.26}$$

the factor K_κ monotonically decreases as κ grows. But if we take

$$J_{m_1} = J_m, \quad J_{n_1} = J_n \pm 1; \quad J_{m_1} = J_m \pm 1, \quad J_{n_1} = J_n, \tag{2.27}$$

then for fairly large values of J_m and J_n the dependence of K_κ on κ may be nonmonotonic.

The orthonormality of the $6j$ symbols[21] implies that

$$K_\kappa^2 \leqslant (2J_{m_1}+1)(2J_m+1), \quad K_\kappa^2 \leqslant (2J_{n_1}+1)(2J_n+1). \tag{2.28}$$

These inequalities lead to a relationship between the incoming frequencies:

$$\nu(mn|m_1n_1,\kappa) \leqslant \left[\frac{(2J_{m_1}+1)(2J_n+1)}{(2J_m+1)(2J_{n_1}+1)}\right]^{1/4}\sqrt{\nu_{mm_10}\nu_{nn_10}}. \tag{2.29}$$

Equations (2.25) and (2.29) are also valid for collision frequencies,[5,6] which emphasizes once again the similarity of the properties of the collision integral and the stimulated radiative relaxation matrix.

Above we studied the case of isotropic radiation. If the perturbing radiation is anisotropic, the outgoing and incoming frequencies in the κq-representation are not diagonal in κq (see Eqs. (A24)–(A27)). In other words, anisotropic radiative relaxation mixes the polarization moments of different orders.

3. COLLAPSE OF A SPECTRAL DOUBLET

In the absence of polarization transfer the spectrum of absorption (emission, scattering, gain) of the four-level system of Fig. 1 consists of four lines with central frequencies ω_{ij}. When $|\Delta|$ is small, the lines are grouped into two doublets: $\omega_{mn}, \omega_{m_1n_1}$ and $\omega_{n_1n}, \omega_{m_1m}$. Let us examine the contour of the $\omega_{mn}, \omega_{m_1n_1}$ doublet in the case where the isotropic radiation is in resonance with the other doublet, $\omega_{m_1m}, \omega_{n_1n_1}$, and stimulates polarization transfer between the $m_1 - n_1$ and $m - n$ transitions.

In an approximation that is linear in the intensity of the monochromatic field and in which the levels m_1, n_1, m, and n are isotropically populated, solving the absorption-spectrum problem requires knowing the off-diagonal elements of the density matrix $\rho(ij\kappa q)$ for $\kappa = 1$ (the dipole approximation). At this point it is advisable to introduce the following simplifying notation:

$$\rho_q \equiv \rho(mn1q), \quad \Gamma \equiv \frac{\Gamma_m+\Gamma_n}{2}, \quad \nu \equiv \frac{\nu_m+\nu_n}{2},$$

$$\rho_{1q} \equiv \rho(m_1 n_1 1q), \quad \Gamma_1 \equiv \frac{\Gamma_{m_1}+\Gamma_{n_1}}{2}, \quad \nu_1 \equiv \frac{\nu_{m_1}+\nu_{n_1}}{2},$$

$$\tilde{A} \equiv A(mn|m_1 n_1,1), \tag{3.1}$$

$$\tilde{\nu} \equiv \nu(mn|m_1 n_1,1) = \nu(m_1 n_1|mn,1).$$

The system of equations for ρ_q and ρ_{1q} can be obtained from Eq. (2.1) in which the matrix elements of the interaction V are

$$V(mMnM') = -\sum_\sigma (-1)^{J_n-M'} \langle J_m M J_n - M'|1\sigma\rangle G_\sigma,$$

$$V(m_1 M_1 n_1 M_1') = -\sum_\sigma (-1)^{J_{n_1}-M_1'} \times \langle J_{m_1} M_1 J_{n_1} - M_1'|1\sigma\rangle G_{1\sigma},$$

where $G_\sigma = d_{mn}\mathscr{E}_\sigma/2\sqrt{3}\hbar$, and $G_{1\sigma} = d_{m_1 n_1}\mathscr{E}_\sigma/2\sqrt{3}\hbar$, with $\mathscr{E}_\sigma$ the circular component of the field. Ignoring collisions ($S=0$) and assuming that the atom is at rest, we arrive at the following system of equations for ρ_q and ρ_{1q}:

$$(\Gamma+\nu-i\Omega)\rho_q - (\tilde{A}+\tilde{\nu})\rho_{1q} = iG_q N,$$
$$-\tilde{\nu}\rho_q + (\Gamma_1+\nu_1-i\Omega_1)\rho_{1q} = iG_{1q}N_1, \tag{3.2}$$

where $\Omega = \omega - \omega_{mn}$, $\Omega_1 = \omega - \omega_{m_1 n_1} = \Omega - \Delta$, ω is the frequency of the field, and N and N_1 are the differences of populations of the magnetic sublevels of the states m,n and m_1,n_1. In the absence of isotropic radiation, $\tilde{\nu}=0$ and the system of equations (3.2) becomes decoupled: spontaneous polarization transfer (the term $\tilde{A}\rho_{1q}$) proceeds only in one direction, downward. This case was studied by the present author in Refs. 19 and 20, where it was shown that transferring polarization ρ_{1q} to the $m-n$ transition generates an interference component in the spectral contour of the doublet. But for $\tilde{\nu} \neq 0$, polarization transfer occurs in both directions and the contour changes considerably.

The solution of the system of equations (3.2),

$$\rho_q = \frac{i[(\Gamma_1+\nu_1-i\Omega_1)G_q N + (\tilde{A}+\tilde{\nu})G_{1q}N_1]}{D}, \tag{3.3}$$

$$\rho_{1q} = \frac{i[(\Gamma_1+\nu-i\Omega)G_{1q}N + \tilde{\nu}G_q N]}{D}, \tag{3.4}$$

$$D = (\Gamma+\nu-i\Omega)(\Gamma_1+\nu_1-i\Omega_1) - \tilde{\nu}(\tilde{A}+\tilde{\nu}), \tag{3.5}$$

makes it possible to calculate the work P done by the field and the field-absorption coefficient $\alpha(\Omega)$:

$$P = -2\hbar\omega \mathrm{Re}\sum_q i(G_q^*\rho_q + G_{1q}^*\rho_{1q})$$
$$= \alpha(\Omega)\sum_q \frac{c}{8\pi}|\mathscr{E}_q|^2, \tag{3.6}$$

$$\alpha(\Omega) = \frac{\lambda^2}{4\pi}\mathrm{Re}\frac{aN(\Gamma_1+\nu_1-i\Omega_1) + a_1 N_1(\Gamma+\nu-i\Omega) + \sqrt{aa_1}[\tilde{A}N_1 + \tilde{\nu}(N+N_1)]}{D}, \tag{3.7}$$

$$a = A_{mn}(2J_m+1), \quad a_1 = A_{m_1 n_1}(2J_{m_1}+1). \tag{3.8}$$

The general structure of Eq. (3.7) is traditional for the problem of doublet collapse due to polarization exchange:[4–6,10] the first two terms on the right-hand side of Eq. (3.7) represent the contributions of the $m-n$ and m_1-n_1 transitions, while the combination $\sqrt{aa_1}$ emphasizes the interference nature of the third term; here we are dealing with intratomic interference because both a and a_1 contain the characteristics of a single atom, while the properties of an atomic ensemble (the atomic densities N and N_1) are additive. Notwithstanding the traditional nature of the general expression (3.7), radiative polarization exchange has remarkable properties, which we discuss below.

The spectral properties of $\alpha(\Omega)$ are related, as usual, to the determinant D, which can be written as

$$D = [\gamma_1 - i(\Omega - \Delta/2)][\gamma_2 - i(\Omega - \Delta/2)],$$

$$\gamma_{1,2} = \gamma_0 \mp c, \quad \gamma_0 = \frac{\Gamma+\nu+\Gamma_1+\nu_1}{2}, \tag{3.9}$$

$$c = \sqrt{(\Gamma+\nu-\Gamma_1-\nu_1-i\Delta)^2/4 + \tilde{\nu}(\tilde{A}+\tilde{\nu})}.$$

The "kinetic part" of the determinant D,

$$(\Gamma+\nu)(\Gamma_1+\nu_1) - \tilde{\nu}(\tilde{A}+\tilde{\nu}) = \Gamma(\Gamma_1+\nu_1) + \nu\nu_1 - \tilde{\nu}^2 + \Gamma_1\nu - \tilde{A}\tilde{\nu}, \tag{3.10}$$

is positive since the inequalities (2.28) yield

$$\nu\nu_1 \geq \tilde{\nu}^2, \quad \Gamma_1\nu \geq \tilde{A}\tilde{\nu}. \tag{3.11}$$

We also note that $\tilde{\nu}\tilde{A}$ is positive for all signs of $\tilde{\nu}$ and $\tilde{A}$, since these two quantities contain the factor K_κ determining their signs.

The absorption coefficient $\alpha(\Omega)$ can be expanded in partial fractions, i.e., represented by a sum of two Lorentzians:

$$\alpha(\Omega) = \frac{\lambda^2}{4\pi}\mathrm{Re}\left[\frac{C_1}{\gamma_1 - i(\Omega-\Delta/2)} + \frac{C_2}{\gamma_2 - i(\Omega-\Delta/2)}\right]. \tag{3.12}$$

The amplitudes C_1 and C_2 are given by the following formulas:

$$C_{1,2} = C_0 \pm C, \quad C_0 = \frac{aN + a_1 N_1}{2},$$

$$C=\frac{1}{4c}\{(a_1N_1-aN)(\Gamma+\nu-\Gamma_1-\nu_1-i\Delta)+2\sqrt{aa_1}[\tilde{A}N_1+\tilde{\nu}(N+N_1)]\}. \quad (3.13)$$

When the exchange rate is low, or

$$4\tilde{\nu}\tilde{A}\ll|\Gamma+\nu-\Gamma_1-\nu_1-i\Delta|^2,\quad |\tilde{\nu}|\ll|\tilde{A}|, \quad (3.14)$$

the Lorentzians are centered at the frequencies $\Omega=0$ and $\Omega_1=0$ [see Eq. (3.5)] and have halfwidths $\Gamma+\nu$ and $\Gamma_1+\nu_1$. In the expressions for the amplitudes C_1 and C_2 we discard the term proportional to $\tilde{\nu}$ but keep the combination $\tilde{A}N_1$, which describes a spontaneous polarization cascade.[19,20] A detailed analysis of $\alpha(\Omega)$ will be carried out with the simplifying assumptions

$$\nu=\nu_1,\quad \Gamma=\Gamma_1,\quad a=a_1,\quad N=N_1, \quad (3.15)$$

in which case the interference of the doublet components manifests itself most vividly. When the conditions specified in (3.15) are met, we have

$$\gamma_{1,2}=\Gamma+\nu\mp d,\quad C_{1,2}=aN[1\pm(\tilde{\nu}+\tilde{A}/2)/2],$$
$$d=\sqrt{\tilde{\nu}(\tilde{A}+\nu)-\Delta^2/4}. \quad (3.16)$$

If we have $\tilde{\nu}\neq 0$ but $\tilde{\nu}(\tilde{A}+\tilde{\nu})<\Delta^2/4$, then d is imaginary, the Lorentzians in (3.12) are centered at different frequencies, and the distance between them is

$$\mathrm{Im}(\gamma_2-\gamma_1)=2\,\mathrm{Im}\,d=\sqrt{\Delta^2-4\tilde{\nu}(\tilde{A}+\tilde{\nu})}.$$

As $|\tilde{\nu}|$ grows, the components of the doublet shift toward one another, and if

$$|\tilde{\nu}|\geqslant(\sqrt{\Delta^2+\tilde{A}^2}-\tilde{A})/2\equiv\nu_0, \quad (3.17)$$

the doublet collapses: both Lorentzians are at the central frequency $\Omega=\Delta/2$ but have different halfwidths γ_1 and γ_2 and amplitudes. The Lorentzian with the smaller halfwidth γ_1 has a greater amplitude C_1, while the Lorentzian with the larger halfwidth γ_2 has a smaller (in absolute value) negative amplitude C_2.

Figure 2 depicts $\mathrm{Im}\gamma_{1,2}$ (an arc of a circle) and $\mathrm{Re}\gamma_{1,2}$ (a hyperbola) as functions of $|\tilde{\nu}|$. One can clearly see how fast the Lorentzians shift toward one another and how the difference of their halfwidths ($|\tilde{\nu}|>\nu_0$) increases in a fairly narrow interval near ν_0.

A remarkable case here is the limit

$$(\tilde{\nu}+\tilde{A}/2)^2\gg(\Delta^2+A^2)/4, \quad (3.18)$$

where the difference in the Lorentzians is at its maximum:

$$\gamma_1=\Gamma-\frac{|\tilde{A}|}{2}+\frac{\tilde{A}^2}{8|\tilde{\nu}|}+\nu-|\tilde{\nu}|+\frac{\Delta^2}{8|\tilde{\nu}|},\quad C_1=2aN, \quad (3.19)$$

$$\gamma_2=\Gamma+\frac{|\tilde{A}|}{2}+\nu+|\tilde{\nu}|\gg\gamma_1,$$
$$-C_2=\frac{aN(\Delta^2+\tilde{A}^2)}{8|\tilde{\nu}|^2}\ll C_1. \quad (3.20)$$

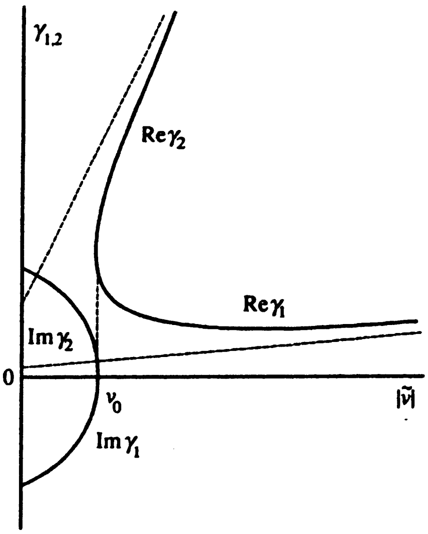

FIG. 2. Halfwidths and shifts of the Lorentzians as functions of the incoming frequency $|\tilde{\nu}|$.

Here the contour $\alpha(\Omega)$ contains a sharp and strong component (γ_1,C_1) and a broad and weak negative component (γ_2,C_2).

The expression (3.19) for γ_1 contains terms of different types. The difference $\nu-|\tilde{\nu}|$ of the outgoing and incoming frequencies is a typical feature of a well-developed collapse:[5,6] the contribution of the stimulated transitions $m\leftrightarrow m_1$ and $n\leftrightarrow n_1$ to the halfwidth γ_1 is balanced, either partially or completely, by polarization exchange. The component $\Delta^2/8|\tilde{\nu}|$ of the halfwidth γ_1 can be called the residual halfwidth. It is similar to the diffusion halfwidth k^2D, where k is the wave vector, and D is the diffusion coefficient, in the problem of Dicke narrowing of a Doppler contour.[2–6] Clearly, the term $\Delta^2/8|\tilde{\nu}|$ is important only in the limit $\nu-|\tilde{\nu}|\ll\nu$.

The component $\Gamma-|\tilde{A}|/2+\tilde{A}^2/8|\tilde{\nu}|$ of the halfwidth γ_1 is related primarily to spontaneous processes. In contrast to collisional collapse, where the spontaneous halfwidth remains constant, in radiative collapse the contribution of spontaneous processes changes. Instead of the natural halfwidth $\Gamma=(\Gamma_m+\Gamma_n)/2$, the halfwidth γ_1 contains the difference $\Gamma-|\tilde{A}|/2$, which is interpreted as the spontaneous analog of the difference $\nu-|\tilde{\nu}|$. The term $\tilde{A}^2/8|\tilde{\nu}|$ in γ_1 should be interpreted as the analog of the diffusion halfwidth. This term is significant when the spontaneous outgoing process is nearly perfectly balanced by the spontaneous incoming process $(\Gamma-|\tilde{A}|/2\ll\Gamma)$.

In the theory of spectral line broadening the contribution of the spontaneous decay of states is always assumed to be a universal quantity, i.e., a quantity that any additional perturbation of the radiating atom is unable to change. The radiative collapse mechanism proves to be a fundamental exception in the general physical sense. Indeed, the universality of the spontaneous part of the linewidth is related to the irreversible nature of the relaxation of the dipole moment caused by spontaneous decay and to the fact that the later is statistically independent from other reasons for relaxation. In the four-level diagram of Fig. 1 the spontaneous decay of the polarization $\rho_{m_1n_1}$ on the m_1-n_1 transition is not irreversible since the reverse stimulated transition $\rho_{mn}\rightarrow\rho_{m_1n_1}$ conserves coherence on the m_1-n_1 transition. Hence, within the system of two transitions, m_1-n_1 and $m-n$, coherence decays only because the balance is partial ($|\tilde{A}|<A_{m_1m}$) and spontaneous decay occurs through "third" levels that differ from those in Fig. 1. The quantity $\Gamma-|\tilde{A}|/2$ gives the rate of decay due to residual effects.

The inequality $|\tilde{\nu}|\gg|\tilde{A}|$ is almost equivalent to the conditions $\nu\gg\Gamma$ and $\nu_1\gg\Gamma_1$ under which saturation on the $m-m_1$ and $n-n_1$ sets in. We can therefore assume that irrespective of the excitation mechanism the populations are always approximately equal, $\rho_m=\rho_{m_1}$ and $\rho_n=\rho_{n_12}$, with the result that $N=N_1$.

When a two-level nondegenerate system interacts with a monochromatic field, the saturation factor contains the combination $\Gamma_m-A_{mn}+\Gamma_n$ (see, e.g., Refs. 5 and 6). The emerging difference Γ_m-A_{mn} is similar, in both form and physical meaning, to the difference $\Gamma-|\tilde{A}|/2$ in our problem.

In Refs. 19 and 20 it was found that for certain parameter ratios a spontaneous polarization cascade can lead to gain without population inversion. Let us examine the respective effect on this process of stimulated radiative relaxation at the center of the line and in the wings. If $|\Omega|$ is much larger than Δ and the relaxation constants, we have

$$\alpha(\Omega)=\frac{\lambda^2}{4\pi\Omega^2}\{aN\Gamma+a_1N_1\Gamma_1-\sqrt{aa_1}\tilde{A}N_1+aN\nu +a_1N_1\nu_1-\sqrt{aa_1}\tilde{\nu}(N+N_1)\}. \tag{3.21}$$

The first three terms on the right-hand side of this equation are related to spontaneous processes, and the remaining terms are related to stimulated processes. According to Refs. 19 and 20, the inequality

$$aN\Gamma+a_1N_1\Gamma_1-\sqrt{aa_1}\tilde{A}N_1<0$$

can be met, with the result that at $\nu=\nu_1=\tilde{\nu}=0$ we have $\alpha(\Omega)<0$. In determining the sign of the sum of the last four terms on the right-hand side of Eq. (3.21) it is advisable to use (3.21), in view of which

$$aN\nu+a_1N_1\nu_1-\sqrt{aa_1}\tilde{\nu}(N+N_1)>\tfrac{1}{2}[a\nu+a_1\nu_1 -2\sqrt{a\nu a_1\nu_1}](N+N_1)+\tfrac{1}{2}(a\nu+a_1\nu_1)(N-N_1). \tag{3.22}$$

The quantity in square brackets is positive. The remaining term may be either positive or negative, but for high saturations the difference $N-N_1$ is inversely proportional to $1/\nu$ and $1/\nu_1$, so that this term must be of the order of the spontaneous term.

Now let $\Delta=0$, $\tilde{\nu}<0$, and $\tilde{A}<0$. For the center of the line we have

$$\alpha(0)=\frac{\lambda^2}{4\pi D}\{aN(\Gamma_1+\nu_1)+a_1N_1(\Gamma+\nu) -\sqrt{aa_1}[|\tilde{A}|N_1+|\tilde{\nu}|(N+N_1)]\}. \tag{3.23}$$

If $\nu\gg\Gamma$, $\nu_1\gg\Gamma_1$, and $|\tilde{\nu}|\gg|\tilde{A}|$, we can say the same things about the expression in braces in (3.23) as we did about the right-hand side of (3.22). Thus, stimulated radiative relaxation does not exclude the possibility of gain (or negative absorption) without population inversion.

4. EFFECT OF STIMULATED RADIATIVE RELAXATION ON MAGNETOOPTICAL RESONANCE

Let us examine magnetooptical resonances on the $m-n$ transition and take into account radiative relaxation, which mixes the levels m and n. We are dealing, therefore, with two excited levels that are split by a magnetic field H and interact with broad-band radiation quasi-resonant to the $m-n$ transition. The radiation is assumed isotropic, so that Eqs. (2.13)–(2.22) are applicable. The problem consists primarily in calculating the polarization moments $\rho(jj\kappa q)$ of the levels that determine magnetooptical resonances in absorption, refraction, spontaneous emission, etc.[15,17]

To simplify matters we introduce the following notation:

$$\rho(mm\kappa q)\equiv\rho_{m\kappa q},\quad \rho(nn\kappa q)\equiv\rho_{n\kappa q},\quad \nu_{mn\kappa}\equiv\nu_\kappa,\quad A_{mn\kappa}\equiv A_\kappa. \tag{4.1}$$

The system of kinetic equations for $\rho_{m\kappa q}$ and $\rho_{n\kappa q}$ has the following form:

$$(\Gamma_m+\nu_m-i\Omega_mq)\rho_{m\kappa q}-\nu_\kappa\rho_{n\kappa q}=Q_{m\kappa q},$$
$$-(A_\kappa+\nu_\kappa)\rho_{m\kappa q}+(\Gamma_n+\nu_n-i\Omega_nq)\rho_{n\kappa q}=Q_{n\kappa q}. \tag{4.2}$$

The stimulated outgoing frequencies ν_m and ν_n are given by Eq. (2.13), and the spontaneous and stimulated incoming frequencies (A_κ and ν_κ) are given by Eqs. (2.20)–(2.23) to within level notation, with

$$(2J_m+1)\nu_m=(2J_n+1)\nu_n,\quad A_\kappa=A_{mn}K_{1\kappa},\quad \nu_k=\nu_mK_{1\kappa},$$
$$K_{1\kappa}=(-1)^{1+\kappa+J_m+J_n}(2J_m+1)\begin{Bmatrix}J_m & J_m & \kappa\\ J_n & J_n & 1\end{Bmatrix}. \tag{4.3}$$

The products Ω_jq in Eqs. (4.2) describe magnetic splitting:

$$\Omega_j=g_j\mu H, \tag{4.4}$$

where g_j is the g factor of the levels $j=m,n$. The right-hand sides of system (4.2), $Q_{m\kappa q}$ and $Q_{n\kappa q}$, are the level pump rates, with the pump radiation generally being anisotropic. The solutions of the system of equations (4.2) have the standard form

$$\rho_{m\kappa q}=\frac{(\Gamma_n+\nu_n-i\Omega_nq)Q_{m\kappa q}+\nu_\kappa Q_{n\kappa q}}{D_{\kappa q}}, \tag{4.5}$$

$$\rho_{n\kappa q}=\frac{(A_\kappa+\nu_\kappa)Q_{m\kappa q}+(\Gamma_m+\nu_m-i\Omega_{m\kappa q})Q_{n\kappa q}}{D_{\kappa q}}, \tag{4.6}$$

$$D_{\kappa q}=(\Gamma_m+\nu_m-i\Omega_mq)(\Gamma_n+\nu_n-i\Omega_nq) -\nu_\kappa(A_\kappa+\nu_\kappa). \tag{4.7}$$

The second part of the problem consists in calculating the experimentally measurable quantities via $\rho_{j\kappa q}$. Here we are interested in the absorption coefficient $\alpha(\omega,H)$ of the monochromatic field (with frequency ω and wave vector $\mathbf{k}$) that is in resonance with the $m-n$ transition:

$$\alpha(\omega,H)=\frac{\lambda^2}{4\pi}A_{mn}(2J_m+1)$$

$$\times\mathrm{Re}\left\langle\frac{1}{\Gamma-i\Omega'}\sum_{\kappa q}(a_{n\kappa}\rho_{n\kappa q}-a_{m\kappa}\rho_{m\kappa q})i^*(\kappa q)\right\rangle,$$

$$i(\kappa q)=\sum_{\sigma\sigma_1}(-1)^{1-\sigma}\langle 1\sigma_1\, 1-\sigma|\kappa q\rangle$$

$$\times\mathscr{E}_{\sigma_1}\mathscr{E}^*_{\sigma}\Big/\sum_{\sigma}|\mathscr{E}_{\sigma}|^2,\quad \Omega'=\omega-\omega_{mn}-\mathbf{k}\cdot\mathbf{v},\tag{4.8}$$

$$a_{n\kappa}=3(-1)^{1+J_m+J_n}\begin{Bmatrix}J_n & J_n & \kappa\\ 1 & 1 & J_m\end{Bmatrix},$$

$$a_{m\kappa}=3(-1)^{1+J_m+J_n}\begin{Bmatrix}J_m & J_m & \kappa\\ 1 & 1 & J_n\end{Bmatrix},$$

where the angle brackets stand for averaging over the velocities $\mathbf{v}$, and $i(\kappa q)$ is the normalized field polarization tensor. In (4.8) we did not allow for Zeeman line splitting, which is by assumption negligible in comparison to the Doppler width.

The fact that the dependence of $\rho_{j\kappa q}$ and $\alpha(\omega,H)$ on the magnetic field H is of a resonant nature is due to the form of the determinants $D_{\kappa q}$. In the absence of stimulated relaxation ($\nu_j=\nu_\kappa=0$), the magnetooptical resonances are described by the following Lorentzians:

$$\frac{1}{\Gamma_m-iQ_mq},\quad\frac{1}{\Gamma_n-iQ_nq}.\tag{4.9}$$

The width of each contour depends on the property of only one level, (Γ_j,g_j). In this respect the interference term caused by spontaneous magnetic coherence transfer [A_κ in (4.6)] is not an exception. The properties of magnetic resonances are well-studied.[15,17] Even if for some reason stimulated magnetic coherence exchange can be ignored ($\nu_\kappa=0$), the overall structure of the magnetic resonances is retained: they are described by the Lorentzian contours

$$\frac{1}{\Gamma_m+\nu_m-i\Omega_mq},\quad\frac{1}{\Gamma_n+\nu_n-i\Omega_nq},\tag{4.10}$$

which are similar to (4.9) but have different widths. Thanks to the reciprocity of the exchange, stimulated magnetic coherence transfer leads to a characteristic collapse of the contours (4.10).

All the magnetic resonances are centered at a zero magnetic field ($\Omega_j=0$) and differ only in width. It is therefore natural to introduce an average g factor and renormalize the relaxation frequencies:

$$\bar g=\frac{g_m+g_n}{2},\quad\bar\Omega=\frac{\Omega_m+\Omega_n}{2}=\bar g\mu H,\quad\bar\Gamma_j=\frac{\Gamma_j\bar g}{g_j},$$

$$\bar\nu_j=\frac{\nu_j\bar g}{g_j},\quad\bar\nu_\kappa=\frac{\nu_\kappa\bar g}{\sqrt{g_mg_n}},\quad\bar A_\kappa=\frac{A_\kappa\bar g}{\sqrt{g_mg_n}}.\tag{4.11}$$

The determinants $D_{\kappa q}$ are expressed in terms of the parameters (4.11) in the following manner:

$$D_{\kappa q}=\frac{(\Gamma_{1\kappa}-i\bar\Omega q)(\Gamma_{2\kappa}-i\bar\Omega q)g_mg_n}{\bar g^2},$$

$$\Gamma_{1\kappa}=\bar\Gamma+\bar\nu-c_\kappa,\quad\Gamma_{2\kappa}=\bar\Gamma+\bar\nu+c_\kappa,$$

$$\bar\Gamma=\frac{\bar\Gamma_m+\bar\Gamma_n}{2},\quad\bar\nu=\frac{\bar\nu_m+\bar\nu_n}{2},\tag{4.12}$$

$$c_\kappa=\sqrt{\frac{1}{4}((\bar\Gamma_m+\bar\nu_m-\bar\Gamma_n-\bar\nu_n)^2)+\bar\nu_\kappa(\bar A_\kappa+\bar\nu_\kappa)}.$$

The absorption coefficient $\alpha(\omega,H)$ can be represented by the following sum of Lorentzians:

$$\alpha(\omega,H)=\mathrm{Re}\sum_{\kappa q}\left[\frac{C_{1\kappa q}}{\Gamma_{1\kappa}-i\bar\Omega q}+\frac{C_{2\kappa q}}{\Gamma_{2\kappa}-i\bar\Omega q}\right].\tag{4.13}$$

According to the properties of $6j$ symbols, the order κ of the polarization moments in Eqs. (4.8) and (4.9) assumes the values $\kappa=0,1,2$, while q varies in the interval $|q|\leq\kappa$. The terms with $q=0$ are independent of H. Hence the resonant dependence on H is contained in six pairs of terms: $\kappa=1$ and $q=\pm1$ or $\kappa=2$ and $q=\pm1,\pm2$.

According to the system of equations (4.2), stimulated radiative transfer of magnetic coherence mixes only two contours. The reason for this, obviously, is the fact that the radiation is isotropic. When relaxation is anisotropic, polarization moments with different values of κ mix and a greater number of contours may collapse.

The amplitudes $C_{1\kappa q}$ and $C_{2\kappa q}$ depend on a large number of various parameters, which makes it natural to analyze them for specific cases. Here we are interested only in the resonance halfwidths $\Gamma_{1\kappa}$ and $\Gamma_{2\kappa}$.

Clearly, both $\Gamma_{1\kappa}$ and $\Gamma_{2\kappa}$ as functions of the outgoing frequency $\bar\nu$ represent two branches of the hyperbola (Fig. 3)

$$(\Gamma_\kappa-\Gamma-\bar\nu)^2-(a_\kappa\bar\nu+b_\kappa)^2=\bar\gamma^2-b_\kappa^2,\tag{4.14}$$

where the parameters $\bar\gamma$, a_κ, and b_κ do not depend on $\bar\nu$ and are given by the following formulas:

$$\bar\gamma=\frac{\bar\Gamma_m-\bar\Gamma_n}{2},\quad a_\kappa=\sqrt{\xi_\kappa^2+\eta^2},\quad b_k=\frac{\eta\bar\gamma+\xi_\kappa\bar A_\kappa}{2},$$

$$\xi_\kappa=\frac{\bar\nu_\kappa}{\bar\nu},\quad\eta=\frac{\bar\nu_m-\bar\nu_n}{2\bar\nu}.\tag{4.15}$$

The asymptotes of the hyperbola (4.14) are the straight lines (the dotted lines in Fig. 3)

$$\Gamma_{1\kappa}=\bar\Gamma-|b_\kappa|+(1-a_\kappa)\bar\nu,\quad\Gamma_{2\kappa}=\bar\Gamma+|b_\kappa|+(1+a_\kappa)\bar\nu,\tag{4.16}$$

which intersect the vertical axis at $\bar\Gamma\pm|b_\kappa|$. The hyperbolas intersect the vertical axis at $\bar\Gamma\pm|\bar\gamma|$. Depending on the sign of $b^2-\bar\gamma^2$, the hyperbola lies either in the region between the asymptotic lines ($b^2>\bar\gamma^2$) or outside that region ($b^2<\bar\gamma^2$;

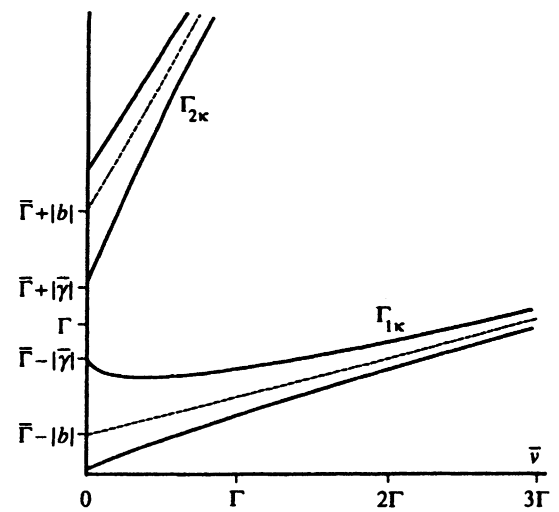

FIG. 3. Halfwidths $\Gamma_{1\kappa}$ and $\Gamma_{2\kappa}$ of the Lorentzian components of the magnetooptical resonance as functions of the frequency $\bar{\nu}$.

here we are dealing only with the quadrant where $\Gamma_\kappa>0$ and $\bar{\nu}>0$ in Fig. 3). In turn the definitions (4.15) imply that $b^2>\bar{\gamma}^2$ or $b^2<\bar{\gamma}^2$ depending on whether

$$\left|\frac{\bar{\Gamma}_m-\bar{\Gamma}_n}{\bar{A}_{mn}-\eta}\right|<\sqrt{1+\eta^2},\quad \bar{A}_{mn}=\frac{A_{mn}\bar{g}}{\sqrt{g_m g_n}}(b^2>\gamma^2) \qquad (4.17)$$

or

$$\left|\frac{\bar{\Gamma}_m-\bar{\Gamma}_n}{\bar{A}_{mn}-\eta}\right|>\sqrt{1+\eta^2}(b^2<\gamma^2). \qquad (4.18)$$

Both cases may occur. Interestingly, the conditions (4.17) and (4.18) do not contain the factor $K_{1\kappa}$, i.e., they are valid for all values of κ: the hyperbolas for $\kappa=1$ and $\kappa=2$ are distinct but have the same asymptotic lines.

Let us examine the simple case where $g_m=g_n$ and $\nu_m=\nu_n=\bar{\nu}$. The hyperbola (4.14) assumes the form

$$(\Gamma_\kappa-\Gamma-\nu_m)^2-(\nu_\kappa+A_\kappa/2)^2=\bar{\gamma}^2-A_\kappa^2/4. \qquad (4.14a)$$

In conditions of a well-developed collapse we have

$$\Gamma_{1\kappa}=\frac{\Gamma_m+\Gamma_n-|A_\kappa|}{2}+\nu_m-|\nu_\kappa|+\frac{A_\kappa^2-(\Gamma_m-\Gamma_n)^2}{8|\nu_\kappa|},$$
$$\Gamma_{2\kappa}=\frac{\Gamma_m+\Gamma_n+|A_\kappa|}{2}+\nu_m+|\nu_\kappa|, \qquad (4.19)$$

where $|\nu_\kappa|\gg|A_\kappa|,|\Gamma_m-\Gamma_n|$. Well-developed collapse is characterized by a partial balance between the spontaneous and stimulated components of the halfwidth $\Gamma_{1\kappa}$ and by a considerable increase in $\Gamma_{2\kappa}$. The condition $\nu_m=\nu_n$ means that $J_m=J_n$. If we use the explicit expression for a $6j$ symbol,[21] we find that

$$1-|\nu_\kappa|/\nu_m=1-|A_\kappa|/A_{mn}$$
$$=1-|K_{1\kappa}|=1-|1-\kappa(\kappa+1)/2J_m(J_m+1)|. \qquad (4.20)$$

Thus, radiative transfer of magnetic coherence always balances departure, but the extent of such balancing decreases as κ grows and increases with J_m. Note also the extensive analogy between the collapse of magnetooptical resonances and that of a spectral doublet (Sec. 3).

Callas and Chaĭka[25] described experiments that demonstrate the effect of radiation emitted by a plasma on magnetooptical resonances (see also Refs. 15 and 17). Their interpretation of the results was based on the idea that the radiation changes the rates of excitation of the polarization moments [$Q_{j\kappa q}$ in the notation adopted in (4.2)]. Such an effect must certainly exist. It is possible, however, that to a certain extent changes in the relaxation matrix also contribute.

5. DISCUSSION

The main achievement of the general theory developed in Sec. 2 is the prediction of radiation transfer of optical and magnetic coherence. The applied method of derivation coincides at crucial points with the methods used earlier. Hence the achievements of the theory are not related to the method but rather to the statement of the problem, i.e., by drawing on the analogy between stimulated and spontaneous transfer processes, on the one hand, and on the analogy between the transfer of magnetic and optical coherence, on the other.[18–20]

Explaining optical coherence transfer means introducing a complicated diagram of the levels participating in the radiative process, a diagram consisting of at least four levels (Fig. 1). Of course, transfer between stationary states involving any characteristics of the system leads to a broadening of the set of "essential" states. However, in relation to optical coherence such "broadening" seemed to be unnecessary. In other words, analysis points to certain limitations in the applicability of the canonical two-level system even in the resonance approximation. Up till now the limited nature of the two-level system was related only to the resonance approximation.

When the pump radiation is isotropic, the radiative relaxation matrix in the κq-representation is diagonal in κq, in accordance with general ideas.[16,24] The outgoing frequencies are proportional to the second Einstein coefficients, while the incoming frequencies for polarization are the geometric mean of two Einstein coefficients for transitions from the combining levels. In accordance with the above-mentioned analogy, the rates of spontaneous and stimulated polarization transfers follow similar patterns in their dependence on the moments of the states and the rank κ. When the pump radiation is anisotropic, stimulated radiative relaxation "mixes" polarization moments of different orders.

The existence of direct and reverse polarization transfers and the absence of a phase shift (the incoming frequencies are real-valued) cause tight spectral structures to collapse. In the above example of a doublet, collapse imposes certain restrictions on the relationship between the doublet splitting and the outgoing and incoming frequencies, and the restrictions are typical of other collapse mechanisms as well. A remarkable feature of radiative collapse is that in the limit of a well-developed collapse the width of the narrow component of the line may prove to be smaller than the spontaneous width of the initial doublet components.

There is another difference between radiative collapse and collisional collapse worth noting. It is an established fact that in gas-kinetic conditions collisions mix a group of states whose energies differ little (fine splitting of atomic levels, and rotational splitting of molecular levels). On the other hand, radiation may initiate transitions between states located in an arbitrary manner on the energy scale. Hence radiative coherence exchange may lead to a collapse of a structure in which the collisional mechanism is ineffective. If we ignore the problem of how the radiative and collisional transfer mechanisms are related and remain on purely phenomenological grounds, collisions can be taken into account by introducing certain terms in the expressions for the outgoing and incoming frequencies. Collisions may enhance the collapse process, but they may also hinder it.

Similar to the case of the collapse of spectral structures, stimulated radiative relaxation leads to the collapse of magnetooptical resonances by mixing the polarization moments of the levels. In this case, too, in conditions of a well-developed collapse the widths of magnetooptical resonances may probe to be smaller than the natural spontaneous values.

In Secs. 3 and 4 we discussed the manifestation of radiative relaxation in the coefficient of absorption (gain) of a weak monochromatic field. Similar effects appear in the spectra of emission, refraction, and scattering. The Doppler effect plays an important role here, as shown by the example of a spontaneous polarization cascade.[20]

Polarization induced between two levels can be considered the simplest type of coherence. This naturally leads to the problem of spontaneous and stimulated transfers of one state of an arbitrary type into another. Undoubtedly such a "generalized" transfer is possible in principle, but its effectiveness depends on the specific conditions and is determined by transfer coefficients similar to the Einstein coefficients and the conditions of resonance of the interfering coherent states of the quantum system.

In conclusion we note that the effects of stimulated radiative transfer of optical coherence manifest themselves in conditions where the rates of stimulated transitions are comparable to those of spontaneous transitions or are higher. This occurs at radiation brightness temperatures of order $\hbar\omega$. The real values of intensities are therefore different in the ultraviolet, visual, and infrared ranges of the spectrum.

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APPENDIX

Let us introduce the matrix $\sigma = aa^+$ consisting of the column a of probability amplitudes of the stationary states of an isolated atom and the Hermitian conjugate row a^+. The density matrix ρ can be obtained from σ by averaging over the random parameters:

$$\rho = \bar{\sigma}. \tag{A1}$$

The system of equations for finding the off-diagonal elements σ_{ij} of the four-level system of Fig. 1 has the form

$$\begin{aligned}
\dot{\sigma}_{mn} &= -\Gamma_{mn}\sigma_{mn} + i\sum_\lambda (G^\lambda_{mm_1} e^{i\Omega_\lambda t}\sigma_{m_1 n} - \sigma_{mn_1} G^\lambda_{n_1 n} e^{-i\Omega'_\lambda t}),\\
\dot{\sigma}_{m_1 n_1} &= -\Gamma_{m_1 n_1}\sigma_{m_1 n_1} + i\sum_\lambda (G^\lambda_{m_1 m} e^{-i\Omega_\lambda t}\sigma_{mn_1} - \sigma_{m_1 n} G^\lambda_{nn_1} e^{i\Omega'_\lambda t}),\\
\dot{\sigma}_{m_1 n} &= -\Gamma_{m_1 n}\sigma_{m_1 n} + i\sum_\lambda (G^\lambda_{m_1 m} e^{-i\Omega_\lambda t}\sigma_{mn} - \sigma_{m_1 n_1} G^\lambda_{n_1 n} e^{-i\Omega'_\lambda t}),\\
\dot{\sigma}_{mn_1} &= -\Gamma_{mn_1}\sigma_{mn_1} + i\sum_\lambda (G^\lambda_{mm_1} e^{i\Omega_\lambda t}\sigma_{m_1 n_1} - \sigma_{mn} G^\lambda_{nn_1} e^{i\Omega'_\lambda t}),
\end{aligned} \tag{A2}$$

$$\Omega_\lambda = \omega_\lambda - \omega_{m_1 m}, \quad \Omega'_\lambda = \omega_\lambda - \omega_{n_1 n}, \quad \Gamma_{ij} = \Gamma_i + \Gamma_j/2. \tag{A3}$$

Here ω_λ is the frequency of the λth mode of the "noise" radiation, and the quantities G^λ_{ij} and σ_{ij} are matrices with respect to the magnetic quantum numbers of states i and j. In the dipole approximation,

$$G^\lambda_{ij}(MM') = \frac{d_{ij}}{2\sqrt{3}\hbar}\sum_\sigma (-1)^{J_j - M'} \times \langle J_i M J_j - M' | 1\sigma\rangle E^\lambda_\sigma, \tag{A4}$$

where E^λ_σ is the circular component of the field in the λth mode.

Further calculations are done according to the following scheme. First the formal solutions of Eqs. (A2) for $\sigma_{m_1 n}$ and σ_{mn_1} are constructed, say

$$\sigma_{m_1 n}(t) = i\sum_\lambda \int_{t_0}^t e^{-\Gamma_{m_1 n}(t-t')} dt' [G^\lambda_{m_1 m} e^{-i\Omega_\lambda t'}\sigma_{mn}(t') - \sigma_{m_1 n_1}(t') G^\lambda_{n_1 n} e^{i\Omega'_\lambda t'}]. \tag{A5}$$

Then these expressions for $\sigma_{m_1 n}$ and σ_{mn_1} are plugged into the equations for $\sigma_{m_1 n_1}$ and σ_{mn} and form two sets of four similar terms there. Here is one of these terms:

$$-\sum_{\lambda\lambda_1} G^\lambda_{mm_1} e^{i\Omega_\lambda t}\int_{t_0}^t e^{-\Gamma_{m_1 n}(t-t')} G^{\lambda_1}_{m_1 m} e^{-i\Omega_{\lambda_1} t'}\sigma_{mn}(t')dt'. \tag{A6}$$

Bearing in mind the averaging procedure (A1) and the fact that the phases E^λ_σ are random, we should leave only the terms with $\lambda = \lambda_1$ in the double sum over λ and λ'. Summation over λ is replaced by integration with respect to ω_λ, which results in the appearance of $2\pi\delta(t-t')$. As a result the term (A6) becomes

$$-2\pi\rho_\lambda G^\lambda_{mm_1} G^\lambda_{m_1 m}\sigma_{mn}(t), \tag{A7}$$

where ρ_λ is the spectral density of the number of modes. In the averaging procedure we adopt the "decoupling" hypothesis:

$$\overline{G^\lambda_{mm_1} G^\lambda_{m_1 m}\sigma_{mn}} = \overline{G^\lambda_{mm_1} G^\lambda_{m_1 m}}\rho_{mn}. \tag{A8}$$

As a result of the above calculations we get

$$R^{(1)}_{mn} = \frac{1}{2}(\Gamma_m + \nu_m)\rho_{mn} + \rho_{mn}\frac{1}{2}(\Gamma_n + \nu_n), \tag{A9}$$

$$R^{(2)}_{mn} = (A_{mn,m_1n_1} + \nu_{mn,m_1n_1})\rho_{m_1n_1} e^{-i\Delta t}, \tag{A10}$$

where we have introduced the following notation:

$$\nu_m = 2\pi\rho_\lambda \overline{G^\lambda_{mm_1} G^\lambda_{m_1 m}}, \quad \nu_n = 2\pi\rho_\lambda \overline{G^\lambda_{nn_1} G^\lambda_{n_1 n}}, \tag{A11}$$

$$\nu_{mn,m_1n_1} = 2\pi\rho_\lambda \overline{G^\lambda_{mm_1} \otimes G^\lambda_{n_1 n}}, \quad \Delta = \omega_{m_1 m} - \omega_{n_1 n}. \tag{A12}$$

Here the symbol $\otimes$ stands for a direct product. The term A_{mn,m_1n_1} in (A10) represents spontaneous processes and is not present in our derivation scheme; it has been introduced here on the basis of the results of Refs. 18–20. Similar expressions can be obtained for the matrices $R_{m_1n_1}$, R_{mm}, and $R_{m_1m_1}$:

$$R^{(1)}_{m_1n_1} = \frac{1}{2}(\Gamma_{m_1} + \nu_{m_1})\rho_{m_1n_1} + \rho_{m_1n_1}\frac{1}{2}(\Gamma_{n_1} + \nu_{n_1}), \tag{A13}$$

$$R^{(2)}_{m_1n_1} = \nu_{m_1n_1,mn}\rho_{mn} e^{i\Delta t}, \quad \nu_{m_1n_1,mn} = \nu_{mn,m_1n_1}, \tag{A14}$$

$$R^{(1)}_{mm} = \frac{1}{2}(\Gamma_m + \nu_m)\rho_{mm} + \rho_{mm}\frac{1}{2}(\Gamma_m + \nu_m), \tag{A15}$$

$$R^{(2)}_{mm} = (A_{mm,m_1m_1} + \nu_{mm,m_1m_1})\rho_{m_1m_1}, \tag{A16}$$

$$R^{(1)}_{m_1m_1} = \frac{1}{2}(\Gamma_{m_1} + \nu_{m_1})\rho_{m_1m_1} + \rho_{m_1m_1}\frac{1}{2}(\Gamma_{m_1} + \nu_{m_1}), \tag{A17}$$

$$R^{(2)}_{m_1m_1} = \nu_{m_1m_1,mm}\rho_{mm}, \quad \nu_{m_1m_1,mm} = \nu_{mm,m_1m_1}, \tag{A18}$$

$$\nu_{m_1} = 2\pi\rho_\lambda \overline{G^\lambda_{m_1 m} G^\lambda_{mm_1}}, \quad \nu_{n_1} = 2\pi\rho_\lambda \overline{G^\lambda_{n_1 n} G^\lambda_{nn_1}}, \tag{A19}$$

$$\nu_{mm,m_1m_1} = 2\pi\rho_\lambda \overline{G^\lambda_{mm_1} \otimes G^\lambda_{m_1 m}}. \tag{A20}$$

The matrices R_{nn} and $R_{n_1n_1}$ can be obtained from R_{mm} and $R_{m_1m_1}$ by replacing the label m with n and the label m_1 with n_1.

Above we assumed that there is only one pair of levels, m_1 and n_1, from which transitions to the levels m and n occur. If there are other such pairs of levels, each contributes to R. For instance, for R_{mn} this is reduced to summing over m_1 and n_1 in Eqs. (A10)–(A12). Similarly, several pairs of levels similar to m and n can contribute to $R_{m_1n_1}$, ν_{m_1}, and ν_{n_1}, a fact that can be taken into account by summing over m and n in Eqs. (A14) and (A19).

Let us write the expressions for the outgoing and incoming frequencies in the JM-representation:

$$\nu(mMmM') = 2\pi |d_{m_1m}/2\hbar|^2 \sum_{\kappa q} (-1)^{1+J_m+J_{m_1}} \times \begin{Bmatrix} J_m & J_m & \kappa \\ 1 & 1 & J_{m_1} \end{Bmatrix} (-1)^{J_m-M'} \times \langle J_m M J_m - M' | \kappa q\rangle I(\kappa q), \tag{A21}$$

$$\nu(mMnM'|m_1M_1n_1M_1') = 2\pi \frac{d^*_{m_1m} d_{n_1n}}{(2\hbar)^2\sqrt{2J_{m_1}+1}\sqrt{2J_{n_1}+1}} \times \sum_{\sigma\sigma_1\kappa q} \langle J_m M 1\sigma | J_{m_1} M_1\rangle \langle J_n M' 1\sigma_1 | J_{n_1} M_1'\rangle \times (-1)^{1-\sigma}\langle 1\sigma_1\, 1-\sigma|\kappa q\rangle I(\kappa q), \tag{A22}$$

where $I(\kappa q)$ is the polarization tensor of the noise radiation:

$$I(\kappa q) = \sum_{k_\lambda\sigma\sigma_1} (-1)^{1-\sigma}\langle 1\sigma_1\, 1-\sigma|\kappa q\rangle \overline{E^{\lambda *}_\sigma E^\lambda_{\sigma_1}}\rho_\lambda. \tag{A23}$$

The other outgoing and incoming frequencies can be obtained from (A21) and (A22) by label substitutions. In the κq-representation for R_{mn} we have

$$R^{(1)}(mn\kappa q) = \sum_{\kappa_1 q_1} \nu(mn\kappa q|mn\kappa_1 q_1)\rho(mn\kappa_1 q_1), \tag{A24}$$

$$R^{(2)}(mn\kappa q) = \sum_{\kappa_1 q_1} \nu(mn\kappa q|m_1n_1\kappa_1 q_1)\rho(m_1n_1\kappa_1 q_1), \tag{A25}$$

where the outgoing and incoming frequencies are given by the following relationships:

$$\nu(mn\kappa q|mn\kappa_1 q_1) = 2\pi \sum_{\kappa_2 q_2} (-1)^{1+\kappa_1+\kappa_2}\sqrt{2\kappa_1+1}\sqrt{2\kappa_2+1} \times \langle \kappa_1 q_1 \kappa_2 q_2|\kappa q\rangle I(\kappa_2 q_2) \Bigg[(-1)^{J_{m_1}-J_n} \times \left|\frac{d_{m_1m}}{2\hbar}\right|^2 \begin{Bmatrix} \kappa_1 & \kappa_2 & \kappa \\ J_m & J_n & J_m \end{Bmatrix} \begin{Bmatrix} J_m & J_m & \kappa_2 \\ 1 & 1 & J_{m_1} \end{Bmatrix} + (-1)^{J_{n_1}-J_m} \left|\frac{d_{n_1n}}{2\hbar}\right|^2 \begin{Bmatrix} \kappa_1 & \kappa_2 & \kappa \\ J_n & J_m & J_n \end{Bmatrix} \times \begin{Bmatrix} J_n & J_n & \kappa_2 \\ 1 & 1 & J_{n_1} \end{Bmatrix} \Bigg], \tag{A26}$$

$$\nu(mn\kappa q|m_1n_1\kappa_1q_1)=2\pi\frac{d^*_{m_1m}d_{n_1n}}{(2\hbar)^2}\sum_{\kappa_2q_2}\langle\kappa_1q_1\kappa_2q_2|\kappa q\rangle \times I(\kappa_2q_2)(-1)^{\kappa_1+\kappa_2}\sqrt{2\kappa_1+1} \times\sqrt{2\kappa_2+1}\begin{Bmatrix} J_{m_1} & \kappa_1 & J_{n_1} \\ 1 & \kappa_2 & 1 \\ J_m & \kappa & J_n \end{Bmatrix}. \tag{A27}$$

The other elements of the matrix R in the κq-representation can be derived from Eqs. (A24)–(A27) by appropriate label substitutions.

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