Spectral line collapse as a result of radiation-induced transfer of polarization

S. G. Rautian
Institute of Automatic Systems and Electrometry, Siberian Branch of the Russian Academy of Sciences, 630090 Novosibirsk, Russia

(Submitted 9 August 1995)
Pis'ma Zh. Éksp. Teor. Fiz. **62**, No. 6, 460–465 (25 September 1995)

It is shown that wide-spectrum radiation, just like collisions, can bring about exchange of optical coherence between transitions with close Bohr frequencies. If the exchange rate is sufficiently high, close spectral structures undergo collapse. © *1995 American Institute of Physics.*

The existence of cascade transfer of polarization as a result of a spontaneous process was proved in Refs. 1 and 2 . The crux of the matter is as follows. For simplicity and definiteness we shall consider four atomic levels m, n, m_1, and n_1 (see Fig. 1) and assume that the transitions $m-n$, m_1-n_1, m_1-m, and n_1-n are dipole-allowed and give in the emission or absorption spectrum four lines with the Bohr frequencies ω_{mn}, $\omega_{m_1n_1}$, ω_{m_1n}, and ω_{n_1n}. Let these lines form a pair of close doublets; i.e., the difference

$$\omega_{m_1n_1}-\omega_{mn}\cong\omega_{m_1m}-\omega_{n_1n}\equiv\Delta \tag{1}$$

is quite small. The interaction with the zero-point vibrations of the electromagnetic field will then give rise, in addition to a cascade of particles and magnetic coherence, to a transfer of optical coherence (polarization) from one transition (m_1-n_1) to another $(m-n)$.

The possibility of coherence transfer by means of a typically stochastic mechanism is explained by the fact that the same field oscillator mixes the atomic wave functions ψ_{m_1} and ψ_m, as well as the wave functions ψ_{n_1} and ψ_n. As a result, the randomness of the phases of the field oscillators is not manifested in the summation over the field modes. Only the difference Δ of the Bohr frequencies of the transitions with which this field oscillator interacts simultaneously is significant. In this respect, the analogy to collision-induced transfer of coherence is obvious.

E. V. Podivilov called my attention to the possibility of an induced analog of polarization transfer. Indeed, the ideas presented above remain valid when the occupation numbers of the field modes are different from zero and the radiation spectrum is quite wide. The induced processes should therefore lead to exchange of polarizations between closely situated spectral lines. In contrast to spontaneous exchange,[1,2] induced exchange has a reciprocal character. As a result, it can give rise to a collapse of spectral doublets (and, in general, to a collapse of close spectral structures).

We employ the notation of Refs. 3 and 4 . We write the kinetic equation for the one-particle density matrix ρ in the form

$$(\partial/\partial t+\mathbf{v}\cdot\nabla)\rho=R+S-i(V\rho-\rho V), \tag{2}$$

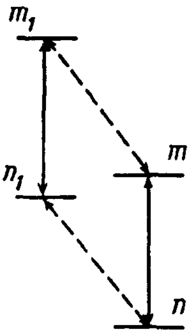

FIG. 1.

where **v** is the velocity of the atom, V is the interaction with an external coherent field, S is the collision integral, and R is the radiative-relaxation matrix. We shall consider the matrix R. Let

$$G^{\lambda}=dE_{\lambda}/\hbar \tag{3}$$

be the operator of the interaction of an atom and the λ mode of the wide-spectrum radiation, for example, thermal radiation, in the dipole approximation (d is the dipole moment). For simplicity, we shall describe the field classically. The following expressions can be obtained for the elements of the matrix R by the standard procedure (see, for example, Refs. 3 and 4) employed for analyzing spontaneous relaxation:

$$R=-R^{(1)}+R^{(2)}, \tag{4}$$

$$R_{mn}^{(1)}=\frac{1}{2}(\Gamma_m+\Gamma_n)\rho_{mn}+\pi\sum_{\mathbf{k}}\frac{d\lambda}{d\omega}(G_{mm_1}^{\lambda}G_{m_1m}^{\lambda}\rho_{mn}+\rho_{mn}G_{nn_1}^{\lambda}G_{n_1n}^{\lambda}), \tag{5}$$

$$R_{mn}^{(2)}=A_{mnm_1n_1}\rho_{m_1n_1}e^{-i\Delta t}+2\pi\sum_{\mathbf{k}}\frac{d\lambda}{d\omega}G_{mm_1}^{\lambda}\rho_{m_1n_1}G_{n_1n}^{\lambda}e^{-i\Delta t}. \tag{6}$$

All quantities, except the spontaneous relaxation rates Γ_j, are matrices with respect to the magnetic quantum numbers:

$$(\rho_{mn})_{MM'}=\rho(mMnM'),\quad (\rho_{m_1n_1})_{M_1M_1'}=\rho(m_1M_1n_1M_1'), \tag{7}$$

$$(R_{mn}^{(i)})_{MM'}=R^{(i)}(mMnM'),\quad i=1,2, \tag{8}$$

$$(G_{m_1m}^{\lambda})_{M_1M}=\frac{d_{m_1m}}{2\sqrt{3}\hbar}\sum_{\sigma}(-1)^{J_m-M}\langle J_{m_1}M_1J_m-M|1\sigma\rangle E_{\sigma}^{\lambda}, \tag{9}$$

$$(G_{n_1n}^{\lambda})_{M_1'M'}=\frac{d_{n_1n}}{2\sqrt{3}\hbar}\sum_{\sigma}(-1)^{J_n-M'}\langle J_{n_1}M_1'J_n-M'|1\sigma\rangle E_{\sigma}^{\lambda}, \tag{10}$$

$$A(mMnM'|m_1M_1n_1M_1')=\sqrt{A_{m_1m}A_{n_1n}}\sum_\sigma \langle J_mM1\sigma|J_{m_1}M_1\rangle\langle J_nM'1\sigma|J_{n_1}M_1'\rangle. \tag{11}$$

Here d_{ij} are the reduced matrix elements of the dipole moment for the transition $i-j$, A_{ij} are the Einstein coefficients for spontaneous emission, J and M are the angular momenta and the magnetic quantum numbers of the stationary states of the atom, $\langle ...|...\rangle$ are vector addition coefficients, and E_σ^λ are the circular components of the electric field intensity of the λ mode of the radiation. In the expressions (5) and (6) integration over the frequencies of the λ modes has already been performed. The sums over **k** denote summation over the directions of propagation of the modes. The levels are enumerated in accordance with Fig. 1, so that

$$\omega_{m_1m}>0,\quad \omega_{n_1n}>0,\quad \omega_{mn}>0,\quad \omega_{m_1n_1}>0. \tag{12}$$

According to Eq. (5), the term $R_{mn}^{(1)}$ which expresses the loss of the radiative relaxation matrix contains spontaneous decay and a sum over **k**, which is associated with the depletion of the levels m and n as a result of the transitions (absorption) $m\to m_1$ and $n\to n_1$ induced by radiation with a continuous spectrum. These effects are well known. The arrival term $R_{mn}^{(2)}$ consists of a spontaneous polarization cascade, which was studied in Refs. 1 and 2 [the first term on the right-hand side of Eq. (6)], and an induced cascade, in which we are mainly interested.

An expression for $R_{m_1n_1}$ is obtained from Eqs. (5) and (6) by interchanging the indices, but without the spontaneous component in $R_{m_1n_1}^{(2)}$ [by virtue of the conditions (12)]:

$$R_{m_1n_1}^{(1)}=\frac{1}{2}(\Gamma_{m_1}+\Gamma_{n_1})\rho_{m_1n_1}+\pi\sum_{\mathbf{k}}\frac{d\lambda}{d\omega}(G_{m_1m}^\lambda G_{mm_1}^\lambda\rho_{m_1n_1}+\rho_{m_1n_1}G_{n_1n}^\lambda G_{nn_1}^\lambda), \tag{13}$$

$$R_{m_1n_1}^{(2)}=2\pi\sum_{\mathbf{k}}\frac{d\lambda}{d\omega}G_{m_1m}^\lambda\rho_{mn}G_{nn_1}^\lambda e^{i\Delta t}. \tag{14}$$

As noted above, in contrast to spontaneous exchange, the induced radiative exchange of polarizations occurs not only "from top to bottom" $(m_1n_1\to mn)$ but also from "bottom to top" $(mn\to m_1n_1)$.

The matrix elements R_{jj}, which describe the radiative loss and gain for the levels $j=m$, m_1, n, and n_1, have a similar form. They incorporate the spontaneous and induced exchange of populations and magnetic coherences (see, for example, Refs. 3–6).

The expressions written out for R presume that there are no external static fields and that the stationary states of the atom are degenerate. Otherwise, the energy of the M sublevel depends on the magnetic quantum number and the quantities Δ in Eqs. (6) and (14) depend on M, M_1, etc., according to well-known laws (the Zeeman and Stark effects; see, for example, Ref. 7). In all other respects, Eqs. (5)–(14) remain valid.

It was also assumed above that wide-spectrum radiation is quasiresonant only with the transitions $m_1 - m$ and $n_1 - n$. If the indicated radiation mixes the states m_1, m, n_1, and n with other levels, then summation over all interacting states must occur in Eqs. (5), (6), (13), and (14).

Induced radiative relaxation depends strongly on the degree of polarization and directedness of the "noise" radiation. We shall give in more specific terms the relations for unpolarized isotropic radiation presented above. In this case the field can be characterized by the volume spectral energy density

$$U_\omega = \frac{1}{8\pi} \sum_{\sigma \mathbf{k}} |E_\sigma^\lambda|^2 \frac{d\lambda}{d\omega}, \tag{15}$$

and the matrix R can be greatly simplified. For an isotropic perturbation of the atom the so-called κq representation is especially useful:

$$\begin{aligned} \rho(JM \cdot J'M') &= \sum_{\kappa q} (-1)^{J'-M'} \langle JMJ' - M' | \kappa q \rangle \rho(JJ'\kappa q), \\ \rho(JJ'\kappa q) &= \sum_{MM'} (-1)^{J'-M'} \langle JMJ' - M' | \kappa q \rangle \rho(JMJ'M'). \end{aligned} \tag{16}$$

In this representation, in the case of isotropic radiation the elements $R(J_m J_n \kappa q)$ and $R(J_{m_1} J_{n_1} \kappa q)$ of the matrix R are

$$E^{(1)}(J_m J_n \kappa q) = [1/2(\Gamma_m + \Gamma_n) + \nu_{mn}] \rho(J_m J_n \kappa q), \tag{17}$$

$$R^{(2)}(J_m J_n \kappa q) = [A(mn|m_1 n_1, \kappa) + \tilde{\nu}(mn|m_1 n_1, \kappa)] \rho(J_{m_1} J_{n_1} \kappa q) e^{-i\Delta t}, \tag{18}$$

$$R^{(1)}(J_{m_1} J_{n_1} \kappa q) = [1/2(\Gamma_{m_1} + \Gamma_{n_1}) + \nu_{m_1 n_1}] \rho(J_{m_1} J_{n_1} \kappa q), \tag{19}$$

$$R^{(2)}(J_{m_1} J_{n_1} \kappa q) = \tilde{\nu}(m_1 n_1 | mn, \kappa) \rho(J_m J_n \kappa q) e^{i\Delta t}, \tag{20}$$

where the quantities ν_{ij} and $\tilde{\nu}(mn|m_1 n_1, \kappa)$ are given by the relations

$$\nu_{mn} = 1/2(B_{mm_1} + B_{nn_1}) U_\omega, \quad \nu_{m_1 n_1} = 1/2(B_{m_1 m} + B_{n_1 n}) U_\omega, \tag{21}$$

$$A(mn|m_1 n_1, \kappa) = \sqrt{A_{m_1 m} A_{n_1 n}} K_\kappa,$$

$$K_\kappa = (-1)^{1+\kappa+J_m+J_{n_1}} \sqrt{2J_{m_1}+1} \sqrt{2J_{n_1}+1} \begin{Bmatrix} J_m J_n \kappa \\ J_{n_1} J_{m_1} 1 \end{Bmatrix}, \tag{22}$$

$$\tilde{\nu}(mn|m_1 n_1, \kappa) = C^*_{m_1 m} C_{n_1 n} U_\omega K_\kappa = \tilde{\nu}(m_1 n_1 | mn, \kappa). \tag{23}$$

Here $\{...\}$ denotes a $6j$ symbol, and

$$C_{ij} = \frac{2\pi}{3} \frac{d_{ij}}{\hbar \sqrt{2J_i + 1}}, \quad B_{ij} = |C_{ij}|^2. \tag{24}$$

The quantities B_{ij} are the Einstein coefficients for induced emission in the transition $i - j$.[7]

In accordance with the general considerations, which are associated with the isotropy of the perturbation of the atom,[8] the loss rates ν_{mn} and $\nu_{m_1n_1}$ do not depend on κ, in contrast to the arrival rates $\tilde{\nu}$, which do exhibit such a dependence. We note also the structural similarity of the matrix R, defined by Eqs. (17)–(20), and the collision integral (see Refs. 3 and 4). In the case of an anisotropic perturbation, the similarity to the collision integral remains, but the expressions for the elements of the matrix R become more complicated (similar to the pump matrix[6]).

Let us now consider the contour of a doublet in the absorption spectrum of a weakly monochromatic field near ω_{mn}, $\omega_{m_1n_1}$ when the atom is subjected to an isotropic radiative perturbation. We simplify the notation as follows:

$$\rho_q \equiv \rho(J_mJ_n1q), \quad \Gamma \equiv (\Gamma_m+\Gamma_n)/2, \quad \nu \equiv \nu_{mn}, \quad \Omega = \omega - \omega_{mn},$$

$$\rho_{1q} \equiv \rho(J_{m_1}J_{n_1}1q), \quad \Gamma_1 \equiv (\Gamma_{m_1}+\Gamma_{n_1})/2, \quad \nu_1 \equiv \nu_{m_1n_1}, \quad \Omega_1 = \Omega - \Delta, \tag{25}$$

$$A \equiv A(mn|m_1n_1,1), \quad \tilde{\nu} \equiv \tilde{\nu}(mn|m_1n_1,1).$$

The equations for the off-diagonal elements ρ_q and ρ_{1q} are

$$(\Gamma+\nu-i\Omega)\rho_q-(A+\tilde{\nu})\rho_{1q}=iG_qN, \quad G_q=d_{mn}\mathscr{E}_q/2\sqrt{3}\hbar,$$

$$(\Gamma_1+\nu_1-i\Omega_1)\rho_{1q}-\tilde{\nu}\rho_q=iG_{1q}N_1, \quad G_{1q}=d_{m_1n_1}\mathscr{E}_q/2\sqrt{3}\hbar, \tag{26}$$

where $\mathscr{E}_q$ are the circular components of the monochromatic field, and N and N_1 are the differences in the populations of the magnetic sublevels in the states n, m and n_1, m_1. In contrast to isotropic irradiation $\tilde{\nu}=0$ and the system of equations (26) decouples, i.e., polarization exchange proceeds "in one direction," —in a cascade fashion. This is the case of Ref. 2 . If $U_\omega \neq 0$ and $\tilde{\nu} \neq 0$, the exchange of the polarizations of the transitions $m-n$ and m_1-n_1 becomes coupled and the contour of the doublet can change substantially. In this respect, induced exchange of coherences is completely analogous to collisional exchange, which can bring about collapse of the doublet (see, for example, Ref. 9). In our case, there are some interesting features which justify the following, almost standard analysis.

The absorption coefficient $\alpha(\Omega)$ is proportional to the work performed by the field

$$\alpha(\Omega) \propto -\mathrm{Re}\, i \sum_q (G_q^*\rho_q+G_{1q}^*\rho_{1q}).$$

In our case it can be represented as a sum of two Lorentzians

$$\alpha(\Omega) \propto \mathrm{Re}\left[\frac{C_1}{\gamma_1-i(\Omega-\Delta/2)}-\frac{C_2}{\gamma_2-i(\Omega-\Delta/2)}\right], \tag{27}$$

with the complex parameters γ_1, C_1 and γ_2, C_2. Solving the system (26), we can analyze expression (27) for arbitrary values of the parameters. We consider a simple case in which

$$\Gamma=\Gamma_1, \quad \nu=\nu_1, \quad N=N_1, \quad G_q=G_{1q} \tag{28}$$

and the interference phenomena are most striking. If the conditions (28) hold, we find

$$\gamma_{1,2}=\Gamma+\nu\mp D,\quad 2C_{1,2}=(\tilde{\nu}+A/2\pm D)/D,\quad D=\sqrt{\tilde{\nu}(\tilde{\nu}+A)-\Delta^2/4}. \tag{29}$$

If $\tilde{\nu}=0$, then $D=i\Delta/2$ and expression (27) describes a doublet with interference structure.[2] As $\tilde{\nu}$ increases, the components of the doublet begin to approach each other, and for sufficiently large $\tilde{\nu}$,

$$2\tilde{\nu}>\sqrt{\Delta^2+A^2}-A=\Delta^2/(\sqrt{\Delta^2+A^2}+A), \tag{30}$$

we obtain Im$D=0$ and Eq. (27) reflects the collapse of the doublets: Both Lorentzians have the same central frequency $\Omega-\Delta/2=0$ and different half-widths γ_1 and γ_2. The Lorentzian with the smaller half-width γ_1 has the larger amplitude (positive) and the Lorentzian with the larger half-width γ_2 has a negative amplitude which is lower in modulus. For $\tilde{\nu}\gg|\Delta|+A$, we find

$$\gamma_1=\Gamma+\nu-\tilde{\nu}-A/2,\quad C_1\approx 1,\quad \gamma_2=\Gamma+\nu+\tilde{\nu}+A/2,\quad C_2=(\Delta^2+A^2)/8\tilde{\nu}^2\ll 1. \tag{31}$$

The intense polarization exchange thus forms from a doublet a single, narrow line whose width for $\nu=\tilde{\nu}$ does not depend on ν. Remarkably, the spontaneous width $(\Gamma-A/2)$ is also partially canceled. The latter circumstance is evidently associated with the fact that the transfer of coherence from the transition m_1-n_1 to the transition $m-n$ is both induced and spontaneous. The subsequent reverse transfer of polarization $(m-n\rightarrow m_1-n_1)$ increases the effective lifetime of the coherence and therefore narrows the line. The fact that the minimum half-width γ_1 of the collapsed line is smaller than the spontaneous half-width Γ distinguishes the collapse which is due to the radiative exchange from the collapse due to the collisional exchange: In the latter case the spontaneous half-width Γ is the minimum half-width.

In summary, a wide-spectrum external field gives rise to diverse processes: transitions of particles and transfer of magnetic and optical coherence. The induced transfer of coherence, just as its spontaneous analog, is not associated with a change in the energy of the atom and the field. By analogy with the collisional variant, the induced transfer of coherence proceeds in both directions $(m-n\leftrightarrow m_1-n_1)$. This situation can, in particular, give rise to the collapse of spectral structures.

We have considered above the case in which the radiative processes $m_1\leftrightarrow m$ and $n_1\leftrightarrow n$ bring about the transfer of polarization of the dipole-allowed transitions m_1-n_1 and $m-n$. The transfer of coherence proceeds in much the same way in those cases in which the transitions m_1-n_1 and $m-n$ are forbidden.

Close spectral structures which are prone to collapse should be comparatively rare in the line spectra of atoms with a relatively small number of lines. There are many lines, however, in the spectra of atoms with partially filled d and f shells and the probability of random coincidences should not be too small. This is especially true for molecular spectra with an extended rotational-vibrational structure and for transitions between Rydberg states. We note that transitions induced by thermal radiation typically affect strongly the lifetimes of the Rydberg states.[10]

This work was supported by the programs "Optics. Laser Physics" and "Russian Universities."

[1]S. G. Rautian, JETP Lett. **60**, 481 (1994).
[2]S. G. Rautian, JETP Lett. **61**, 461 (1995).
[3]S. G. Rautian, G. I. Smirnov, and A. M. Shalagin, *Nonlinear Resonances in the Spectra of Atoms and Molecules* [in Russian], Nauka, Novosibirsk, 1979.
[4]S. G. Rautian and A. M. Shalagin, *Kinetic Problems of Nonlinear Spectroscopy*, North-Holland Publishing Co., Amsterdam, 1991.
[5]M. P. Chaĭka, *Interference of Degenerate Atomic States*[in Russian], Leningrad State University Press, Leningrad, 1975.
[6]E. B. Aleksandrov, G. I. Khvostenko, and M. P. Chaĭka, *Intereference of Atomic States* [in Russian], Nauka, Moscow, 1991.
[7]I. I. Sobel'man, *Introduction to the Theory of Atomic Spectra*, Pergamon Press, N. Y. [Russian original, Nauka, Moscow, 1979].
[8]M. I. D'yakonov and V. I. Perel', Zh. Éksp. Teor. Fiz. **47**, 1483 (1964) [Sov. Phys. JETP **20**, 997 (1965)].
[9]L. A. Vaĭnshteĭn, I. I. Sobel'man, and E. A. Yukov, *Excitation of Atoms and Broadening of Spectral Line*, Springer-Verlag, N. Y., 1981 [Russian original, Nauka, Moscow, 1979].
[10]R. F. Stebbings and F. B. Dunning [Eds.], *Rydberg States of Atoms and Molecules*, Cambridge University Press, N. Y., 1989 [Russian translation, Mir, Moscow, 1985].

Translated by M. E. Alferieff