Spectral lineshape in a polarization cascade

S. G. Rautian

Institute of Automation and Electrometry, Siberian Branch of the Russian Academy of Sciences, 630090 Novosibirsk, Russia

(Submitted 6 February 1995)
Pis'ma Zh. Éksp. Teor. Fiz. **61**, No. 6, 461–464 (25 March 1995)

An interference effect due to a radiative (spontaneous) cascade of polarization is predicted in the absorption spectrum of a doublet corresponding to transitions between four levels. Spectral and amplitude properties of the effect are analyzed. Under certain conditions there can be amplification without a population inversion. © *1995 American Institute of Physics.*

Numerous interference phenomena in atomic and molecular spectroscopy are known to result from various types of correlations between transitions (see, for example, Refs. 1–6). In this letter we wish to call attention to an interference effect which is arguably the simplest in a sense but which has not previously been described.

We consider a system of four levels, m_1, n_1, m, and n, between which the following transitions are allowed: m_1-n_1, m_1-m, n_1-n, and $m-n$ (Fig. 1). The absorption (emission) spectrum is usually described by four individual lines with central frequencies $\omega_{m_1n_1}$, ω_{mn}, ω_{m_1m}, and ω_{n_1n}. We now assume that these lines coincide or nearly coincide in pairs, i.e., that the differences

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

are quite small. Under these conditions there can be mutual effects (an interference) of these four lines, as we show below.

The interaction with vacuum zero modes, in addition to transitions of particles and a corresponding emission of photons, causes a transfer of dipole moments (or polarization or optical coherence). According to Ref. 7, the term representing a radiative (spontaneous) transition in the kinetic equation for the density-matrix element $\rho(mMnM')$ contains a term

$$\sum_{M_1M_1'} A(mMnM'|m_1M_1n_1M_1')\rho(m_1M_1n_1M_1'),$$

$$A(mMnM'|m_1M_1n_1M_1')=\sqrt{A_{m_1m}A_{n_1n}}\sum_{\sigma}\langle J_mM1\sigma|J_{m_1}M_1\rangle\langle J_nM'1\sigma|J_{n_1}M_1'\rangle. \tag{2}$$

This term describes transfer of the polarization $\rho(m_1M_1n_1M_1')$ from the m_1-n_1 transition to the $m-n$ transition (A_{m_1m} and A_{n_1n} are Einstein coefficients, J_i and M are the

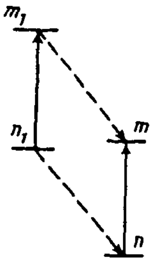

FIG. 1. Scheme of levels and transitions. The dashed arrows show the polarization transfer $m_1-n_1 \rightarrow m-n$.

angular momenta and magnetic quantum numbers, and $\langle ...|...\rangle$ are the coefficients of the vector summation). A cascade polarization transfer occurring in accordance with Eq. (2) causes an interference of the doublet components $\omega_{m_1n_1}, \omega_{mn}$.

Let us consider the absorption of a weak monochromatic field of frequency ω, which is close to the frequency of the doublet. In the steady state, the relevant density-matrix elements are given by

$$[\Gamma_1 - i(\Omega-\Delta)]\rho(m_1M_1n_1M_1') = iG(m_1M_1n_1M_1')(\rho_{n_1}-\rho_{m_1}),$$

$$(\Gamma - i\Omega)\rho(mMnM') = iG(mMnM')(\rho_n - \rho_m) + \sum_{M_1M_1'} A(mMnM'|m_1M_1n_1M_1')\rho(m_1M_1n_1M_1'), \tag{3}$$

$$\Omega = \omega - \omega_{mn}, \quad G(mMnM') = \frac{d_{mn}}{2\sqrt{3}\hbar}\sum_\sigma (-1)^{J_n - M'}\langle J_m M J_n - M'|1\sigma\rangle E_\sigma. \tag{4}$$

Here Γ and Γ_1 are relaxation constants, d_{mn} is the reduced dipole matrix element, E_σ is the circular component of the field, and ρ_j is the population of sublevel M of state j, which is independent of M ($j=m,n,m_1,n_1$). An expression analogous to (4) holds for $G(m_1M_1n_1M_1')$. Using the known relations between the d_{ij} and the A_{ij} (Ref. 1), we find the following expression for the absorbed power P:

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

$$\alpha(\Omega) = \frac{\lambda^2}{4\pi}\left\{N_{nm}A_{mn}\frac{\Gamma}{\Gamma^2+\Omega^2} + N_{n_1m_1}A_{m_1n_1}\left[\frac{\Gamma_1}{\Gamma_1^2+(\Omega-\Delta)^2} + \frac{KC}{\Gamma\Gamma_1}f(\Omega)\right]\right\}, \tag{5}$$

$$C = \sqrt{A_{m_1m}A_{n_1n}A_{mn}/A_{m_1n_1}}, \quad K = (-1)^{J_m+J_{n_1}}\sqrt{2J_m+1}\sqrt{2J_{n_1}+1}\begin{Bmatrix} J_m & J_n & 1 \\ J_{n_1} & J_{m_1} & 1 \end{Bmatrix}, \tag{6}$$

$$f(\Omega) = \mathrm{Re}\frac{\Gamma\Gamma_1}{(\Gamma - i\Omega)[\Gamma_1 - i(\Omega-\Delta)]} = \frac{\Gamma\Gamma_1[\Gamma\Gamma_1 - \Omega(\Omega-\Delta)]}{(\Gamma^2+\Omega^2)[\Gamma_1^2+(\Omega-\Delta)^2]}, \tag{7}$$

$$N_{nm} = (2J_m+1)(\rho_n-\rho_m), \quad N_{n_1m_1} = (2J_{m_1}+1)(\rho_{n_1}-\rho_{m_1}). \tag{8}$$

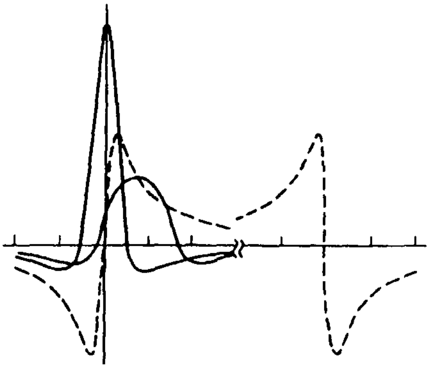

FIG. 2. The function $f(\Omega)$. 1—$\Delta=0$; 2—$\Delta=3\Gamma$; 3—$\Delta\gg\Gamma$. The ordinate scale for curve 3 is larger by a factor of Δ/Γ.

The quantity $\alpha(\Omega)$ is an absorption coefficient. It contains two Lorentzians with central frequencies $\Omega=0$ and $\Omega=\Delta$, corresponding to transitions ω_{mn} and $\omega_{m_1n_1}$. It also contains an interference term whose spectrum is described by the function $f(\Omega)$. The coefficient K is determined by the moments of the four levels and can vary over the range $-1\leqslant K\leqslant 1$.

The interference term is proportional to $N_{n_1m_1}$ and is independent of N_{mn}; the obvious explanation for this circumstance is that this term is of cascade origin. We can show that we have $\int f(\Omega)d\Omega=0$, i.e., that the integral absorption is zero, as it should be for an interference effect. The cases $K>0$ and $K<0$ correspond to amplification and absorption of "interfering oscillations." Finally, we note a remarkable, even unique combination of four Einstein coefficients: $A_{m,n_1}C=\sqrt{A_{m_1n_1}A_{n_1n}A_{m_1m}A_{mn}}$. This combination also stresses the interference nature of the phenomenon.

The behavior of the function $f(\Omega)$ depends strongly on the ratios Δ/Γ and Δ/Γ_1. If $\Delta=0$, then we have

$$f(\Omega)=\Gamma\Gamma_1(\Gamma\Gamma_1-\Omega^2)/(\Gamma^2+\Omega^2)(\Gamma_1^2+\Omega^2), \tag{9}$$

and this function contains a central "peak" of unit amplitude with negative "wings" (curve 1 in Fig. 2). At the minima we have $|f(\Omega)|\leqslant 1/8$. At the point $\Omega=0$ we have

$$\alpha(\Omega)=\frac{\lambda^2}{4\pi}\{N_{nm}A_{nm}\Gamma^{-1}+N_{n_1m_1}A_{n_1m_1}\Gamma_1^{-1}[1+KC/\Gamma]\}. \tag{10}$$

In other words, all three terms are identical in order of magnitude.

With increasing $|\Delta|$, in the interval $|\Delta|\sim\Gamma,\Gamma_1$, the function $f(\Omega)$ becomes wider (curve 2 in Fig. 2). At sufficiently large values of $|\Delta|/\Gamma$ and $|\Delta|/\Gamma_1$, a minimum appears at the middle of the doublet ($\Omega\approx\Delta/2$), while near the points $\Omega=0$ and $\Omega=\Delta$ structure of a dispersion-curve type arises (curve 3 in Fig. 2):

$$f(\Omega)\approx\frac{\Gamma_1}{\Delta}\frac{\Gamma\Omega}{\Gamma^2+\Omega^2}, \quad |\Omega|\sim\Gamma,$$

$$f(\Omega)\approx-\frac{\Gamma}{\Delta}\frac{\Gamma_1(\Omega-\Delta)}{\Gamma_1^2+(\Omega-\Delta)^2},\quad |\Omega-\Delta|\sim\Gamma_1. \tag{11}$$

At the points $\Omega=\pm\Gamma$ and $\Omega-\Delta=\pm\Gamma_1$ we find $|f(\Omega)|\approx\Gamma_1/2|\Delta|$, $\Gamma/2|\Delta|$. At the center of the doublet, at $\Omega=\Delta/2$, on the other hand, we have $f(\Omega)\approx 4\Gamma\Gamma_1/\Delta^2$, i.e., a small quantity of second order. The $\alpha(\Omega)$ behavior at $|\Omega|\gg\Delta$ is interesting:

$$\alpha(\Omega)=\frac{\lambda^2}{4\pi\Omega^2}\{N_{nm}A_{mn}\Gamma+N_{n_1m_1}A_{m_1n_1}(\Gamma_1-KC)\}. \tag{12}$$

The splitting thus plays no role in the remote wings of the doublet (the doublet is "perceived" as a single line from distant frequencies), and the relative contribution of the interference term is on the order of one [as in the case $\Omega=0$; cf. Eq. (10)]. According to relation (12), the absorption coefficient $\alpha(\Omega)$ may be negative under the condition

$$K>0,\quad (KC/\Gamma_1-1)N_{n_1m_1}A_{m_1n_1}\Gamma_1>N_{nm}A_{mn}\Gamma. \tag{13}$$

If $K<0$ and $\Delta=0$, the condition

$$(|K|C/\Gamma-1)N_{n_1m_1}A_{m_1n_1}\Gamma>N_{nm}A_{mn}\Gamma_1 \tag{14}$$

means an amplification at the center of the line ($\Omega=0$). Consequently, a polarization cascade can lead to field amplification without a population inversion.

Amplification without a population inversion is characteristic of nonlinear interference effects:[3,6] Since the contribution of nonlinear interference effects to the absorption, integrated over ω, is zero, this contribution must be negative in certain frequency intervals. Amplification without inversion was predicted in Ref. 8 (see also Refs. 3 and 6) and was observed in Ref. 9; it has recently attracted increased interest. All the papers cited, however, have dealt with the amplification of a weak (probe) field during the simultaneous application of intense laser light or microwave radiation to an atom. In the case at hand, in contrast, there is no external radiation, and the interference results from an interaction with vacuum zero modes, i.e., an interaction "with itself" in a sense. It is in this sense that a spontaneous polarization cascade can be regarded as a very simple interference effect.

It is easy to see that analogous phenomena occur near the doublet ω_{m_1m}, ω_{n_1n}. To describe these phenomena, it is sufficient to make the interchange $m\leftrightarrow n_1$ in all of relations (1)–(13) and to understand Γ and Γ_1 as the half-widths of the n_1-n and m_1-m transitions. The overall absorption is given by the sum of expression (5) and its analog with the specified changes.

I wish to thank E. V. Podivilov, V. A. Sorokin, A. M. Tumaĭkin, A. M. Shalagin, and D. A. Shapiro for a discussion of these questions.

This study was supported by the Russian Universities program and the International Science Foundation.

[1] I. I. Sobel'man, *Introduction to the Theory of Atomic Spectra* [in Russian] (Nauka, Moscow, 1977).
[2] M. P. Chaĭka, *Interference of Degenerate Atomic States* [in Russian] (Izd. LGU, Leningrad, 1975).
[3] S. G. Rautian, G. I. Smirnov, and A. M. Shalagin, *Nonlinear Resonances in Atomic and Molecular Spectra* [in Russian] (Nauka, Novosibirsk, 1979).

[4]L. A. Vaĭnshteĭn, I. I. Sobel'man, and E.A. Yukov, *Excitation of Atoms and Spectral-Line Broadening* [in Russian] (Nauka, Moscow, 1979).
[5]E. B. Aleksandrov, G. I. Khvostenko, and M. P. Chaĭka, *Interference of Atomic States* [in Russian] (Nauka, Moscow, 1991).
[6]S. G. Rautian and A. M. Shalagin, *Kinetic Problems of Non-Linear Spectroscopy* (North-Holland, Amsterdam, 1991).
[7]S. G. Rautian, JETP Lett. **60**, 481 (1994).
[8]S. G. Rautian and I. I. Sobel'man, Zh. Éksp. Teor. Fiz. **41**, 456 (1961) [Sov. Phys. JETP **14**, 328 (1961)].
[9]A. M. Bonch-Bruevich, V. A. Khodovoĭ, and N. A. Chigir', Zh. Éksp. Teor. Fiz. **67**, 2069 (1974) [Sov. Phys. JETP **40**, 1027 (1974)].

Translated by D. Parsons