EFFECTS OF ATOMIC COLLISIONS ON RADIATION LINE WIDTH

V. A. Klivadenko

Vestnik Moskovskogo Universiteta. Fizika,
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The quantum kinetic equation for the density-matrix elements has been used in calculating the line width given by a gas laser on the basis of atomic collisions in the working medium. A strong-collision model has been used in deriving analytic expressions for the intensities of the amplitude and phase fluctuations in the polarization. The results for strong fields are in qualitative agreement with experiment.

The purpose of this paper is to incorporate the effects of collisions in the laser medium on the width of the spectra of the amplitude and phase fluctuations, particularly in order to relate the line width to the parameters of the medium and the field amplitude. The line width is determined by thermal fluctuations and by polarization ones. A major aspect lies in determining the polarization noise in the working medium, since the thermal fluctuations are defined by the standard Callen-Welton equation. The calculation is based on a quantum kinetic equation for the density-matrix elements in the dipole approximation.

In a two-level model, we can write the kinetic equation in the quasi-classical approximation as four equations for the density-matrix elements f_a, f_b, f_{ab}, f_{ba} [1]:

$$\begin{cases} \left(\frac{\partial}{\partial t}+V\frac{\partial}{\partial R}\right) f_a(R,V,t)=\frac{i}{\hbar}(d_{ac}f_{ba}-d_{ba}f_{ab})E-\gamma_a(f_a-f_a^0)+I_a, \\ \left(\frac{\partial}{\partial t}+V\frac{\partial}{\partial R}\right) f_b(R,V,t)=-\frac{i}{\hbar}(d_{ab}f_{ba}-d_{ba}f_{ab})E-\gamma_b(f_b-f_b^0)+I_b, \\ \left(\frac{\partial}{\partial t}+V\frac{\partial}{\partial R}+i\omega_{ab}+\gamma_{ab}\right) f_{ab}(R,V,t)=\frac{i}{\hbar}(f_b-f_a)(d_{ba}E)+I_{ab}, \\ f_{ba}=f_{ab}^* \end{cases} \qquad (1)$$

where E is the field in the cavity and I_n denotes the collision integral, whose detailed form is dependent on the model. For example, in the strong-collision model [2]

$$I_n=\nu_n f^0(V)\int f_n(R,V',t)dV'-\nu_n f_n(R,V,t), \quad n=a,\ b,\ ab,$$

where ν_n is the effective collision frequency and $f^0(V)$ is a maxwellian distribution.

System (1) is a system of equations linear in f_n for a given field with sources f_a^0 and f_b^0 defined by the pumping, while γ_n are dissipative constants reciprocal to the lifetime and relaxation times of levels a and b. We isolate only the contribution from the polarization fluctuations in the expression for the radiation line width. We seek a solution to (1) over an interval τ such that

$$\nu^{-1},\ \gamma^{-1} \ll \tau \ll \Delta\omega_p^{-1},\ \Delta\omega_c^{-1},$$

where $\Delta\omega_p$ is the width of the phase-fluctuation spectrum and $\Delta\omega_c$ is the cavity bandwidth. The field E may be considered as deterministic in this range. We average (1) and subtract it from the initial one to get the system for the deviations $\delta f_n = f_n - \bar{f}_n$, $\delta D = \delta f_a - \delta f_b$ and $\delta N = \delta f_a + \delta f_b$:

$$\begin{cases} \left(\frac{\partial}{\partial t} + V\frac{\partial}{\partial R} + \gamma + \nu\right)\delta D = \frac{2i}{\hbar}(d_{ba}f_{ab} - d_{ab}f_{ba})E + \nu f^0(V)\int \delta D(R, V', t)dV', \\ \left(\frac{\partial}{\partial t} + V\frac{\partial}{\partial R} + \gamma + \nu\right)\delta N = \nu f^0(V)\int dN(R, V', t)\,dV', \end{cases}$$
$$\begin{cases} \left(\frac{\partial}{\partial t} + V\frac{\partial}{\partial R} + i\omega_{ab} + \gamma_{ab} + \nu_{ab}\right)\delta f_{ab} = -\frac{i}{\hbar}(d_{ab}E)\delta D + \\ \qquad + \nu_{ab}f^0(V)\int \delta f_{ab}(R, V', t)\,dV', \\ \delta f_{ba} = \delta f_{ab}^{*}. \end{cases} \tag{2}$$

Here to simplify the subsequent calculations we have put $\nu_a = \nu_b \equiv \nu$, $\gamma_a = \gamma_b \equiv \gamma$.

We consider the single-mode lasing state and represent E in the following form [3]: $E = e_0 E_0 \exp(-i\omega_0 t + ik_0 R)$. We introduce the slowly varying functions $\delta \tilde{f}_{ba}$ for the functions $\delta \tilde{f}_{ab}$, $\delta \tilde{f}_{ba}$:

$$\delta f_{ab} = \delta\tilde{f}_{ab}\exp(-i\omega_0 t + ik_0 R),$$
$$\delta f_{ba} = \delta\tilde{f}_{ba}\exp(i\omega_0 t - ik_0 R).$$

For a homogeneous and isotropic medium, we can assume with good accuracy that δf_{ab} and δf_{ba} are independent of the coordinates and time. After transferring to the Fourier components with respect to $\tau = t - t'$ and $\rho = R - R'$, we get the following system for the deviation correlations from (2):

$$\begin{cases} (\gamma + \nu)(\delta D\delta\tilde{f}_{ab}^{*})_{\omega,k} = \frac{2i}{\hbar}[(d_{ab}e_0)(\delta\tilde{f}_{ab}\delta\tilde{f}_{ab}^{*}) - (d_{ba}e_0)(\delta\tilde{f}_{ba}\delta\tilde{f}_{ab}^{*})]E_0 + \\ \qquad + \nu f^0(V)\int(\delta D\delta\tilde{f}_{ab}^{*})\,dV', \\ (\gamma + \nu)(\delta N\delta\tilde{f}_{ab}^{*})_{\omega,k} = (\delta N\delta\tilde{f}_{ab}^{*})_{\tau=0} + \nu f^0(V)\int(\delta N\delta\tilde{f}_{ab}^{*})\,dV', \\ \Gamma(\delta\tilde{f}_{ab}\delta\tilde{f}_{ab}^{*})_{\omega,k} = \frac{i}{\hbar}(\delta D\delta\tilde{f}_{ab}^{*})(d_{ab}e_0)E_0 + \nu_{ab}f^0(V)\int(\delta\tilde{f}_{ab}\delta\tilde{f}_{ab}')\,dV' + \\ \qquad + (\delta\tilde{f}_{ab}\delta\tilde{f}_{ab}^{*})_{\tau=0}, \\ \Gamma^{*}(\delta\tilde{f}_{ab}^{*}\delta\tilde{f}_{ab}^{*})_{\omega,k} = \frac{i}{\hbar}(\delta D\delta\tilde{f}_{ab}^{*})(d_{ab}e_0)E_0 + \nu_{ab}f^0(V)\int(\delta\tilde{f}_{ab}\delta\tilde{f}_{ab}^{*})\,dV', \end{cases}$$
$$\Gamma = -i(\omega_0 - \omega_{ab} - k_0 V) + (\gamma_{ab} + \nu_{ab}). \tag{3}$$

It is necessary to incorporate the initial conditions for the correlators [4] in (3):

$$(\delta\tilde{f}_{ab}\delta\tilde{f}_{ab}^{*})_{\rho,V,\tau=0} = \frac{(2\pi\hbar)^3}{2N}\delta(\rho)\,\delta(V - V')(f_a(P) + f_b(P)),$$
$$(\delta\tilde{f}_{ab}\delta\tilde{f}_{ab})_{\rho,V,\tau=0} = 0,$$
$$f_a(P) = n\left(\frac{2\pi\hbar}{mkT_i}\right)^{3/2}\exp\left(-\frac{E_a}{kT} - \frac{p^2}{2mkT}\right), \tag{4}$$

where E_a is the energy of level a. The following expressions apply for the intensities of the sources of the amplitude and phase components of the polarization noise [3]

$$\langle \xi_a^2 \rangle = \frac{(4\pi)^2 N^2}{4V} \int \left(\frac{|d_{ab}|^2}{3} (\delta \widetilde{f}_{ab} \delta \widetilde{f}^*_{ab})_{\omega,0} - (e_0 d_{ab})^2 (\delta \widetilde{f}_{ba} \delta \widetilde{f}^*_{ab})_{\omega,0} + \text{c.c} \right) \frac{dP dP'}{(2\pi\hbar)^6},$$
$$\langle \xi_\Phi^2 \rangle = \frac{(4\pi)^2 N^2}{4V} \int \left(\frac{|d_{ab}|^2}{3} (\delta \widetilde{f}_{ab} \delta \widetilde{f}^*_{ab})_{\omega,0} + (e_0 d_{ab})^2 (\delta \widetilde{f}_{ba} \delta \widetilde{f}^*_{ab})_{\omega,0} + \text{c.c} \right) \frac{dP dP'}{(2\pi\hbar)^6}. \qquad (5)$$

The radiation line width $\Delta\omega$ for a single traveling wave is related to the intensity of the polarization noise source by [3]

$$\Delta\omega = \frac{\omega_0^2}{E_0^2} \langle \xi_\Phi^2 \rangle.$$

We solve (3) with (4) and substitute into (5) to get

$$\langle \xi_a^2 \rangle = 2\,\mathrm{Re} \left\{ A^{-1} \int \frac{(\delta \widetilde{f}_{ab} \delta \widetilde{f}^*_{ab})_{\tau=0}\, dV}{\Gamma\,[1 + a_H E_0^2 (\gamma_{ab} + \nu_{ab}) ((A\Gamma^*)^{-1} + (B\Gamma)^{-1})]} \right\},$$
$$\Delta\omega = 2\,\mathrm{Re} \left\{ \frac{\omega_0^2}{E_0^2} A^{-1} \int \frac{(\delta \widetilde{f}_{ab} \delta \widetilde{f}^*_{ab})_{\tau=0} [1 + 2 a_H E_0^2 (\gamma_{ab} + \nu_{ab}) (B\Gamma^*)^{-1}]}{\Gamma\,[1 + a_H E_0^2 (\gamma_{ab} + \nu_{ab}) ((A\Gamma^*)^{-1} + (B\Gamma)^{-2})]} \right\}. \qquad (6)$$

Here

$$A = 1 - \left(\nu_{ab} + 2(\gamma_{ab} + \nu_{ab}) \frac{\nu}{\gamma} a_H E_0^2 \right) \int \frac{f^0(V)}{\Gamma} dV,$$
$$B = 1 - \nu_{ab} \int \frac{f^0(V) dV}{\Gamma}, \quad a_H = \frac{|d_{ab}|^2}{3\hbar^2 (\gamma_{ab} + \nu_{ab})(\gamma + \nu)}.$$

In the immobile atom-approximation, we can put $f^0(V) = \delta(V)$ in (6), and in that case

$$\Delta\omega = \frac{(4\pi)^2 n |d_{ab}|^2 \omega_0^2 (\rho_a^0 + \rho_b^0)}{6VE_0^2} \gamma_{ab} \frac{1 + 2a_H E_0^2}{(\omega_0 - \omega_{ab})^2 + \gamma_{ab}^2 (1 + 2a_H E_0^2)},$$

which completely corresponds to the result obtained in [3]. For a gas laser with an inhomogeneously broadened line, i.e., $\gamma_n \ll \Delta\omega_D$, where $\Delta\omega_D$ is the Doppler broadening, (6) takes a simpler form. If we assume also that $\nu_n \ll \Delta\omega_D$ (weak collisions), we can simplify (6) in two cases.

1. Weak fields ($a_H E_0^2 \ll 1$). From (6) we get

$$\langle \xi_a^2 \rangle = \frac{(4\pi)^2 n |d_{ab}|^2 \sqrt{\pi} (\rho_a^0 + \rho_b^0)}{12V\Delta\omega_D} \left\{ 1 + \frac{\sqrt{\pi}}{\Delta\omega_D} \left(\nu_{ab} + 2(\gamma_{ab} + \nu_{ab}) \frac{\nu}{\gamma} a_H E_0^2 \right) \right\},$$
$$\Delta\omega = \frac{(4\pi)^2 n |d_{ab}|^2 \sqrt{\pi} (\rho_a^0 + \rho_b^0) \omega_0^2}{12V\Delta\omega_D E_0^2} \left\{ 1 + \frac{\sqrt{\pi}\,\nu_{ab}}{\Delta\omega_D} + \left[1 + \frac{4\sqrt{\pi}(\gamma_{ab} + \nu_{ab})\nu}{\Delta\omega_D \gamma} \right] a_H E_0^2 \right\}. \qquad (7)$$

$$\rho_a^0 = \frac{V}{(2\pi\hbar)^3} \int f_a^0 dP. \quad (7)$$

For $\nu_n = 0$, (7) also coincides with the result of [3].

2. Strong fields ($a_H E_0^2 \gg 1$):

$$\langle \xi_a^2 \rangle = \frac{(4\pi)^2 n |d_{ab}|^2 \sqrt{\pi} (\rho_a^0 + \rho_b^0)}{12 V \Delta\omega_D} \left\{ \left(1 + \frac{\sqrt{\pi} (\gamma_{ab} + \nu_{ab}) \nu}{\gamma \Delta\omega_D} \right) \frac{1}{\sqrt{a_H E_0^2}} + \frac{\sqrt{\pi} \nu_{ab}}{\Delta\omega_D} \right\},$$

$$\Delta\omega = \frac{(4\pi)^2 n |d_{ab}|^2 \sqrt{\pi} (\rho_a^0 + \rho_b^0) \omega_0^2}{12 V \Delta\omega_D E_0^2} \left\{ \sqrt{a_H E_0^2} \left(1 + \frac{\sqrt{\pi} (\gamma_{ab} + \nu_{ab}) \nu}{\gamma \Delta\omega_D} \right) + \frac{\sqrt{\pi} \nu_{ab}}{\Delta\omega_D} \right\}.$$

It follows from (8) that the spectra of the amplitude and phase fluctuations become narrower in strong fields as the field amplitude increases. The fluctuation suppression as the mean intensity increases has been observed in [5], where results were given on the fluctuations in the intensity of the third harmonic generated under conditions of three-photon resonance from the 3s-5p transition in sodium vapor. It was found that the relative dispersion in the intensity of the third harmonic was reduced to less than that in the pumping intensity.

Therefore, this result for the spectra of the amplitude and phase fluctuations in the polarization as influenced by the collisions and field amplitude enables one to use a simple two-level model to derive analytic expressions for the line width, which are qualitatively confirmed by experiment.

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Department of General Physics
and Wave Processes