# SPECTRUM OF STATIONARY FLUORESCENCE EXCITATION BY NONMONOCHROMATIC RADIATION DURING THE TRANSITION FROM A SPLIT GROUND STATE

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*The effect of nonmonochromaticity of laser radiation, whose phase and amplitude are modulated by a random purely discontinuous Markovian process, on the fluorescence excitation spectrum has been studied analytically and by means of numerical calculations. It is shown that, for fairly high radiation intensities, the phase and amplitude modulations affect the line profile in essentially different ways. In particular, the amplitude modulation causes line splitting into four components. The latter is most pronounced in the case in which Einstein coefficients are the same for two allowed optical transitions at mean durations of random trains exceeding the radiative decay time of the upper level. A combined effect of the phase–amplitude modulation and collisions on the line profile is examined.*

It has been predicted theoretically[1] that absorption of monochromatic CW radiation resonant with allowed optical transitions by a closed three–level system with closely spaced lower levels results in the formation of a single line profile in the excitation spectrum of stationary fluorescence. Due to interference of polarizations of optical transitions attained through the polarization in the forbidden low–frequency transition, the resonance profile in the fluorescence excitation spectrum (or absorption) is considerably shifted and broadened as the ground state splitting is reduced. Under optimal conditions, the interference shift can amount to over a thousand linewidths of allowed transitions and is limited by collisions, time of flight of atoms through the light beam, and laser nonmonochromaticity. The shift was examined in detail as a function of radiative and collisional parameters of relaxation, splitting $\Delta$, and laser pulse duration. It was noted in particular that the extreme behavior of the shift as $\Delta \to 0$ predetermines the possibilities of a thoroughly analyzing statistical properties of radiation.

The object of this work is to study this possibility and the constraints imposed on the interference shift under fluorescence excitation by radiation whose phase and amplitude are modulated by a purely discontinuous Markovian process. The choice of this process is related to the fact that in a number of situations similar to the one considered here it enables one to derive analytical solution of a problem, nonlinear in the noise intensity, for any depth of modulation.[2–4]

Let the Hamiltonian of interaction between the laser radiation and the system have the form

$$\hat{H}_{\text{int}}(t) = \hbar\,[\hat{V} + \hat{\upsilon}(t)]\exp[-i\omega t - i\alpha(t)]\,, \tag{1}$$

where $\alpha(t)$ and $\hat{\upsilon}(t)$ are the randomly (discontinuously) time–varying phase of radiation and the part of $\hat{H}_{\text{int}}$ given by amplitude fluctuations of the electric field of the light wave and $\omega$ is the optical carrier frequency.

Stationary equations for the density matrix of the system under consideration expressed in terms of the interaction representation which includes the random phase $\alpha(t)$ can be given following Refs. 1–4 in the form

$$2\gamma\rho_0-(A_1-\gamma)\rho_1 + 2(V_1+\upsilon_1)\,\text{Im}\,R_1 = \gamma - \gamma_{\text{av}}(\rho_0 - \ll\rho_0\gg_0)\,;$$

$$\gamma_1\rho_1-2(V_1+\upsilon_1)\text{Im}\,R_1-2(V_2+\upsilon_2)\text{Im}\,R_2 = -\gamma_{\text{av}}(\rho_1-\ll\rho_1\gg_0);$$

$$[\Gamma_1-i(\Omega-\delta_1)]\,R_1-i(V_2+\upsilon_2)\,R_3-i(V_1+\upsilon_1)\,(\rho_0-\rho_1) =$$

$$= -\gamma_{\text{av}}(R_1-\ll R_1\gg_1)\,; \tag{2}$$

$$[\Gamma_2-i(\Omega+\Delta-\delta_2)]\,R_2-i(V_1+\upsilon_1)\,R_3^*-i(V_2+\upsilon_2)\times$$

$$\times(\rho_0+2\rho_1) = i(V_2+\upsilon_2)-\gamma_{\text{av}}(R_2-\ll R_2\gg_1)\,;$$

$$[\Gamma_3+i(\Delta+\delta_3)]\,R_3-i(V_2+\upsilon_2)\,R_1-i(V_1+\upsilon_1)\,R_2^* =$$

$$= -\gamma_{\text{av}}(R_3-\ll R_3\gg_0)\,.$$

Here $\rho_0$ and $\rho_1$ are the populations of the lower ground (0) and upper (1) energy levels, $R_i = R'_i + iR''_i$ ($i$ = 1, 2, and 3) are complex off–diagonal elements of the density (polarization) matrix of the allowed optical transitions $0 \to 1$ ($i$ = 1) and $2 \to 1$ (2) and the forbidden low–frequency transition $0 \to 2$ (3), $\gamma_1 = A_1 + A_2$, $A_1$ and $A_2$ are the first Einstein coefficients for the allowed transitions 1 and 2, and $\Gamma_i$ and $\delta_i$ ($i$ = 1, 2 and 3) are the polarization relaxation constants and collision–induced shifts for the corresponding transitions, $\gamma$ is the collisional mixing rate for the zeroth and second level populations; $\hat{H}_{1,2}$ and $\hat{\upsilon}_{1,2}$ are the real matrix elements of the operators $\hat{V}$ and $\hat{\upsilon}$ for the transitions 1 and 2, $\Omega = \omega - \omega_{10}$ is the laser frequency detuning from the eigenfrequency of the transition 1, $\Delta = \omega_{20}$ is the frequency of the forbidden low–frequency transition 3 or the ground state splitting, $\gamma_{\text{av}}^{-1}$ is the mean

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duration of random trains, the radiation being split up into a sequence of such trains with certain phases and amplitudes, and double angular brackets denote an integral operator related to the averaging of the density matrix elements $\rho(\alpha, \varepsilon)$ over the random phase $\alpha$ and the dimensionless electric field amplitude $\varepsilon$ which is proportional to $\upsilon_{1,2} = \varepsilon\,\upsilon^0_{1,2}$. Assuming uncorrelated train to train variation of $\varepsilon$ we have

$$\ll\rho(\alpha,\varepsilon)\gg_0 = \int\int \rho(\alpha_1,\varepsilon_1) f(\alpha-\alpha_1)\varphi(\alpha_1)\varphi(\varepsilon_1)\mathrm{d}\alpha_1\mathrm{d}\varepsilon_1/\varphi(\alpha); \tag{3}$$

$$\ll\rho(\alpha,\varepsilon)\gg_1 = $$

$$= \int\int \rho(\alpha_1,\varepsilon_1)\, e^{i(\alpha_1-\alpha)} f(\alpha-\alpha_1)\,\varphi(\alpha_1)\,\varphi(\varepsilon_1)\,\mathrm{d}\alpha_1\,\mathrm{d}\varepsilon_1/\varphi(\alpha),$$

where $f(\alpha - \alpha_1)$ is the conditional probability density of the phase jump from $\alpha$ to $\alpha_1$, and $\varphi(\alpha)$ and $\varphi(\varepsilon)$ are the stationary distribution densities for $\alpha$ and $\varepsilon$ at any time section of the process.

Let us average Eqs. (2) over phases after multiplying them by $\varphi(\alpha)$. On account of $\int \varphi(\alpha)\mathrm{d}\alpha = 1$, operators (3) are reduced to the averaging of $\rho(\varepsilon) = \int \rho(\alpha,\varepsilon)\varphi(\alpha)\mathrm{d}\alpha$ over random amplitudes

$$\ll\rho(\alpha,\varepsilon)\gg_0 \to \int \rho(\varepsilon_1)\,\varphi(\varepsilon_1)\,\mathrm{d}\varepsilon_1 \equiv \langle\rho(\varepsilon)\rangle\,,$$

$$\ll\rho(\alpha,\varepsilon)\gg_1 \to (\tilde{\gamma}_{\mathrm{ph}} + i\tilde{\delta}_{\mathrm{ph}})\,\langle\rho(\varepsilon)\rangle\,; \tag{4}$$

$$\tilde{\gamma}_{\mathrm{ph}} + i\tilde{\delta}_{\mathrm{ph}} = \int e^{i\beta} f(\beta)\,\mathrm{d}\beta\,.$$

Here $\tilde{\gamma}_{\mathrm{ph}}$ is the measure of the phase memory in the random modulation process. In particular, for $\tilde{\gamma}_{ph} = 0$ the phases of the adjacent trains are absolutely uncorrelated whereas for $\tilde{\gamma}_{\mathrm{ph}} = 1$ and $\tilde{\delta}_{\mathrm{ph}} = 0$ the phases are invariant from train to train, i.e., no phase modulation is observed.[2]

Using the linearity of Eqs. (2) relative to the field amplitudes and the $\hat{H}_{\mathrm{int}}(t)$ splitting into two parts related to the purely phase $(\hat{V})$ and phase–amplitude $\hat{V}$ modulations the phase averaged elements of the density matrix $\rho(\varepsilon)$ can be represented as $\rho(\varepsilon) = \bar{\rho} + \tilde{\rho}(\varepsilon)$, where the first term is for a purely phase modulation, while the second one is for the phase–amplitude modulation. As a result, problem (2) takes the form

$$\begin{cases} [\,\hat{\bar{A}} + \gamma_{\mathrm{av}}(\hat{I}_8 - \hat{C})]\,\hat{\bar{\rho}} = \hat{\bar{B}}; \\ [\,\hat{\bar{A}} + \hat{A}(\varepsilon) + \gamma_{\mathrm{av}}\hat{I}_8]\,\hat{\tilde{\rho}}(\varepsilon) + \hat{\tilde{A}}(\varepsilon)\,\hat{\bar{\rho}} = \hat{\tilde{B}}(\varepsilon) + \gamma_{\mathrm{av}}\hat{C}\langle\hat{\tilde{\rho}}(\varepsilon)\rangle, \end{cases} \tag{5}$$

$$\hat{\bar{\rho}} = (\bar{\rho}_0,\, \bar{\rho}_1,\, \bar{R}'_1,\, \bar{R}''_1,\, \bar{R}'_2,\, \bar{R}''_2,\, \bar{R}'_3,\, \bar{R}''_3)^{\mathrm{T}}\,,$$

where the column $\hat{\tilde{\rho}}$ is represented in the same way as $\hat{\bar{\rho}}$, $\hat{I}_8$ is the unit matrix, and the $\varepsilon$–independent matrices $\hat{\bar{A}}$ and $\hat{C}$ and the column $\hat{\bar{B}}$ as well as the matrices $\hat{\tilde{A}}(\varepsilon)$ and $\hat{\tilde{B}}(\varepsilon)$ are readily specified by means of Eqs. (2), (4) and (5).

Solution (5) averaged over phase and amplitude fluctuations has the form

$$\hat{\bar{\rho}} = [\hat{\bar{A}} + \gamma_{\mathrm{av}}(\hat{I}_8 - \hat{C})]^{-1}\,\hat{\bar{B}}\,,$$

$$\langle\hat{\tilde{\rho}}(\varepsilon)\rangle = [\hat{I}_8 - \gamma_{\mathrm{av}}\langle\hat{P}^{-1}(\varepsilon)\rangle]^{-1}\,\langle\hat{P}^{-1}(\varepsilon)\,[\hat{\tilde{B}}(\varepsilon) - \hat{\tilde{A}}(\varepsilon)\,\hat{\bar{\rho}}]\rangle\,, \tag{6}$$

$$\hat{P}(\varepsilon) = \hat{\bar{A}} + \hat{\tilde{A}}(\varepsilon) + \gamma_{\mathrm{av}}\hat{I}_8.$$

Thus, an exact algebraic solution of the problem with the purely phase modulation $(\hat{v} = 0)$ as well as of a similar problem on a two–level system[2] exists for arbitrary radiation intensities. The complete solution $\langle\hat{\rho}\rangle = \hat{\bar{\rho}} + \langle\hat{\tilde{\rho}}(\varepsilon)\rangle$ is found after performing quadratures which define an explicit form of the distribution function of the field amplitude fluctuations.

In the case of the purely phase modulation the upper level population $\rho_1$, which is proportional to the fluorescence excitation spectrum, has the form

$$\hat{\bar{\rho}}_1 = [2V_1^2V_2^2 + \gamma\,(V_2^2G_1 + V_1^2G_2 - 2V_1^2V_2^2Q)]/[6V_1^2V_2^2 +$$

$$+ (A_1 + 3\gamma)V_2^2G_1 + (A_2 + 3\gamma)V_1^2G_2 +$$

$$+ (\gamma_1 - 6\gamma)\,V_1^2V_2^2\,Q + \gamma\gamma_1(G_1G_2 - V_1^2V_2^2Q^2)]\,; \tag{7}$$

$$G_{1,2} =$$

$$\bar{\Gamma}_{1,2} + [\bar{\Gamma}_{2,1}\Omega_{1,2}^2 + V_1^2V_2^2D''\,(\bar{\Gamma}_{1,2}D'' \mp 2\Omega_{1,2}D')]/\mathrm{Det}\,,$$

$$Q = D' + [\Omega_1\Omega_2D' + (\Omega_1\bar{\Gamma}_2 - \Omega_2\bar{\Gamma}_1)D'' - V_1^2V_2^2D'D''^2]/\mathrm{Det},$$

$$\bar{\Gamma}_{1,2} = \Gamma_{1,2} + \gamma_c(1 - \tilde{\gamma}_{\mathrm{ph}}) + V_{1,2}^2D',\ \mathrm{Det} = \bar{\Gamma}_1\bar{\Gamma}_2 - V_1^2V_2^2D'^2,$$

$$\Omega_1 = \Omega - \delta_1 + \delta_{\mathrm{ph}} + V_2^2D'',\ \Omega_2 = \Omega + \Delta - \delta_2 + \delta_{\mathrm{ph}} - V_2^2D'',$$

$$D' = \Gamma_3/L\,,\ D'' = (\Delta + \delta_3)/L\,,\ L = \Gamma_3^2 + (\Delta + \delta_3)^2,\ \delta_{\mathrm{ph}} = \gamma_{\mathrm{av}}\tilde{\delta}_{\mathrm{ph}}\,.$$

The form of solution (7) is almost the same as that used in the case of monochromatic radiation.[1] The only difference is in the redefinition of $\bar{\Gamma}_{1,2}$ and $\Omega_{1,2}$ supplemented, respectively, with the summands $\gamma_{\mathrm{av}}(1 - \tilde{\gamma}_{\mathrm{ph}})$ and $\delta_{\mathrm{ph}}$ due to the phase modulation. The role of the phase modulation becomes most obvious in the absence of collisions $(\gamma = \Gamma_3 = \delta_1 = \delta_2 = \delta_3 = 0$ and $\Gamma_1 = \Gamma_2 = \gamma_1/2)$ when Eq. (7) is considerably simplified

$$\bar{\rho}_1 = 2V_1^2 V_2^2 \bar{\Gamma}_1 Z^{-1} / [(\Omega - \Omega_0)^2 + \Gamma^2] ; \quad (8)$$

$$\Omega_0 = -\delta_{\rm ph} - [A_2 V_1^2 \Delta - (A_2 V_1^4 - A_1 V_2^4)/\Delta]/Z ,$$

$$\Gamma^2 = \bar{\Gamma}_1^2 + 6\gamma_{\rm av}(1 - \tilde{\gamma}_{\rm ph}) V_1^2 V_2^2 / Z + V_1^2 V_2^2 \{2(A_1^2 V_2^2 + A_2^2 V_1^2) +$$

$$+ [2\gamma_1(A_1 V_2^4 + A_2 V_1^4) + (\gamma_1^2 + 4 A_1 A_2) V_1^2 V_2^2]/\Delta\} Z^2 ,$$

$$Z = A_1 V_2^2 + A_2 V_1^2 , \quad \bar{\Gamma}_1 = \gamma_1/2 + \gamma_{\rm av}(1 - \tilde{\gamma}_{\rm ph}) .$$

It follows from Eq. (8) that dephasing gives rise to an extra line shift, $\delta_{ph}$, due to the phase memory and extra line broadening associated with the second term in the expression for $\Gamma^2$ and the difference of $\bar{\Gamma}_1$ from $\gamma_1/2$. The latter is maximum in the absence of the phase correlation. Thus, as a matter of principle, the random phase modulation does not eliminate the giant interference shift discussed in Ref. 1. The extra "red" shift $\delta_{ph}$ and broadening detected experimentally will permit quantitative determination of such a characteristic of the random Markovian process as the measure of the mean phase memory.

Let us now examine a more complicated case of the phase and amplitude modulations being combined. In determining $<\tilde{\rho}_1>$ (Eq. (6)) we derived analytical expressions for the matrix elements $\hat{P}^{-1}$, while the amplitude averaging of the matrix $\hat{I}_8 - \gamma_{\rm av}<\hat{P}^{-1}(\varepsilon)>$ and its inversion were performed numerically, using Gaussian probability density distribution $\varphi(\varepsilon) = \exp(-\varepsilon^2)/\sqrt{\pi}$. Figures 1*a–h* illustrate calculated line profiles $<\rho_1(\Omega)> = \bar{\rho}_1(\Omega) + <\tilde{\rho}_1(\Omega)>$, as a function of the mean train duration $\gamma_{\rm av}^{-1}$, amplitude modulation depth $a = \nu_1^0/V_1 = \nu_2^0/V_2$, and the $A_1/A_2$ ratio at a constant average laser power $\sim <(V_1 + \nu_1)^2>$ in the absence of collisions.

An analysis of Figs. 1*c–h* from the standpoint of the effect of the phase–amplitude modulation as compared to the purely phase modulation given by Eq. (8) accurate to three decimal places and Figs. 1*a* and *b* shows that the effect of amplitude modulation for $\alpha \gtrsim 1$ differs greatly for $A_1 = A_2$ and $A_1 \neq A_2$. In particular, the line profile $<\rho_1(\Omega)>$ for $A_1 = A_2$ and specified values of laser power tends to exhibit a distinct splitting into four components, which occurs at $\gamma_{\rm av} < \gamma_1$, while for $A_1 \neq A_2$ the splitting is fairly weak (curve *2*, Fig. 1*e*). In this respect the situation is similar to the one observed in Ref. 1 in which for the same values of the laser power and parameters $A_1$ and $A_2$ the calculated line profiles $\rho_1(\Omega)$ exhibit splitting at $A_1 = A_2$ in the range $\gamma \sim 0.1\,\gamma_1$ in the presence of collisions. No splitting occurs (line narrowing is observed) in the case of $A_1 \neq A_2$. However, collisions do induce line splitting into two components. This can be accounted for by the structure of the denominator in Eq. (7) for $\gamma_{\rm av} = \delta_{\rm ph} = 0$ expressed in terms of the fourth–degree polynomial of $\Omega$. The observed splitting into four components due to the amplitude modulation is to be attributed to the complex composition of the line profiles $<\rho_1(\Omega)>$ and $<\tilde{\rho}_1(\Omega)>$ (Eq. (6)).

Following the behavior of the position of the maximum $\Omega_0$ of the line profile in the case of weak splitting ($A_1 \neq A_2$), we note that the amplitude modulation virtually eliminates the great interference shift $\Omega_0 \simeq 20\,\gamma_1$ for $\gamma_{\rm av} \lesssim 1$, while for higher values of $\gamma_{\rm av}$ $A$ is also in "antiphase" with the phase modulation, resulting in a noticeable negative shift $\Omega_0 = -14\,\gamma_1$ for $\gamma_{\rm av} = 200\,\gamma_1$. Notably, for the modulation depth $a = 1$ $\Omega_0$ decreases due to a major contribution of the purely phase modulation (curves *5* and *6* in Fig. 1*e*) as $\gamma_{\rm av}$ increases from 50 to 200 $\gamma_1$, whereas for $a = 100$, virtually without phase modulation, $\Omega_0$ amounts to 20 $\gamma_1$ (curve *6*, Fig. 1*g*). Our calculations of $\bar{r}_1(\Omega)$ have shown that, under conditions in question for $\gamma_{\rm av} \lesssim \gamma_1$ and $a \lesssim 1$, the line profile $<\rho_1(\Omega)>$ is narrower than $\bar{\rho}_1(\Omega)$.

Figures 1*e*, *b*, and 2 visualize the effect of the laser power on the shape of the fluorescence excitation spectrum for preassigned values of $V_1/V_2$ and $a$. The line splitting due to the amplitude modulation is seen to become much more pronounced as the power level increases.

A combined effect of collisions and phase–amplitude modulation on the line profile $<\rho_1(\Omega)>$ is illustrated in Fig. 3. A comparison between Figs. 3 and 2 where the same values of the line profile and radiation parameters were used but without any collisions, shows essentially non–Lorentzian profile under conditions in which the splitting is barely noticeable. This fact makes it possible to estimate $\gamma \sim \gamma_1$ for which the splitting is completely obscured.

Summarizing the obtained results, it can be asserted that under saturation conditions in the absence of collisions the random phase modulation would give rise to an extra line shift and broadening rather than eliminate the interference shift, and the lines still have a Lorentzian shape. The phase amplitude modulation causes the line profile to split up at fairly high power levels and $\gamma_{\rm av} \lesssim \gamma_1$. The splitting is most pronounced in the case of the same constants of the radiative decay of the upper level into two lower levels. Collisions act to smear the splitting, thereby making the line profile even more complicated.

Thus, the shape of the fluorescence excitation spectrum in a closed three–level system may be a sensitive tool in the analysis of the type of the random light modulation, the presence of the phase memory and collisions, and the properties of the radiative relaxation of the system.

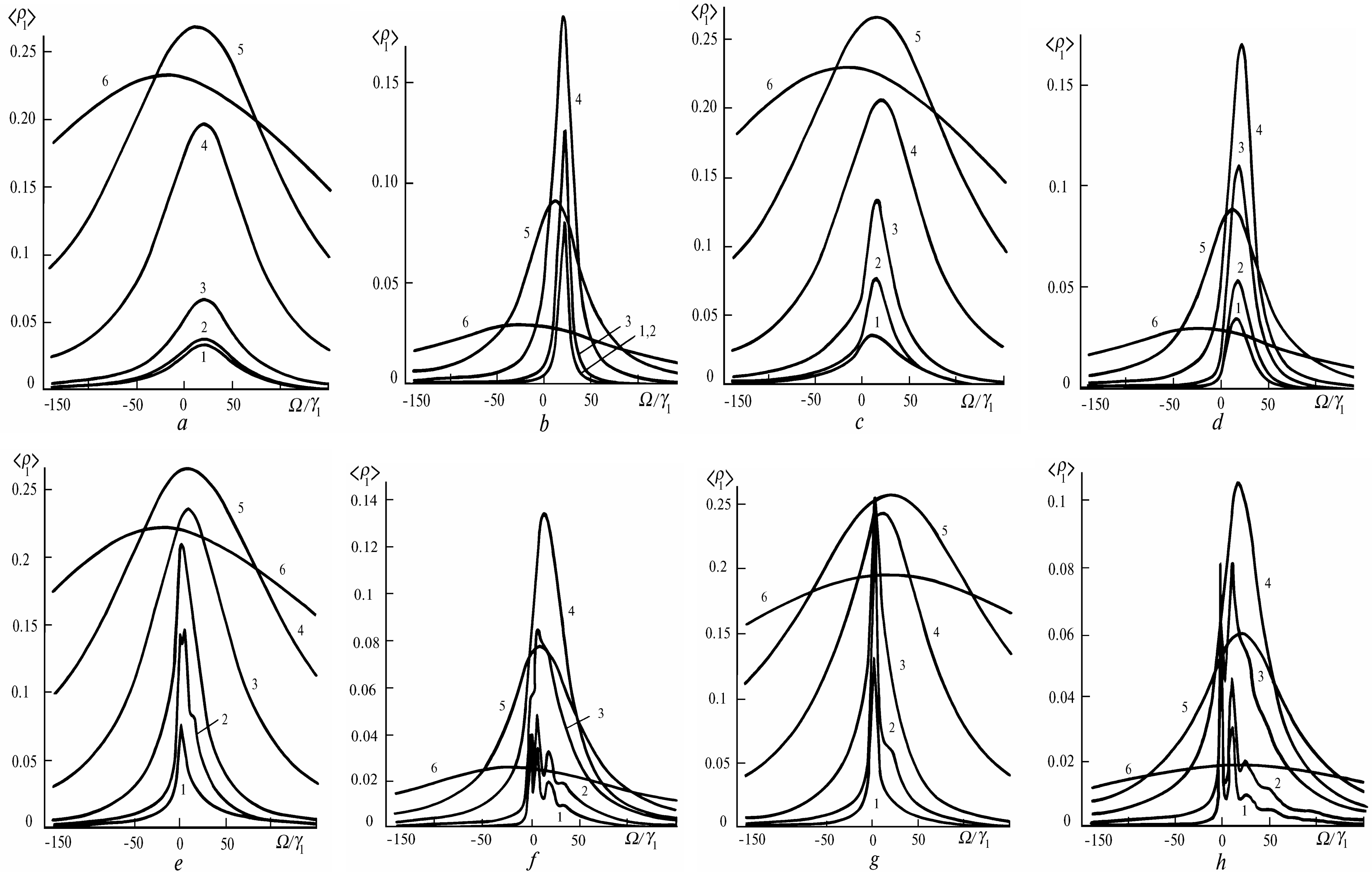


*FIG. 1. Upper level population* $\langle\rho_1\rangle$ *averaged over phase–amplitude fluctuations as a function of frequency detuning* $\Omega$ *for different mean train durations* $\gamma_{av}$, *amplitude modulation depths* a *and the* $A_1/A_2$ *ratio at constant average power. 1)* $\gamma_{av} = 10^{-4}\gamma_1$, *2)* $\gamma_{av} = 0.1\,\gamma_1$, *3)* $\gamma_{av} = \gamma_1$, *4)* $\gamma_{av} = 10\,\gamma_1$, *5)* $\gamma_{av} = 50\,\gamma_1$, *and 6)* $\gamma_{av} = 200\,\gamma_1$. *a and b)* $a = 0.01$, $V_1 = 9.9998\,\gamma_1$, $\nu_1 = 9.9998\cdot10^{-2}\gamma_1$; *c and d)* $a = 0.316$, $V_1 = 9.76\,\gamma_1$, $v_1 = 3.08\,\gamma_1$; *e and h)* $a = 1$, $V_1 = v_2 = 8.17\,\gamma_1$; *g and f)* $a = 100$, $V_1 = 0.141\,\gamma_1$, $\nu_1 = 14.1\,\gamma_1$; *a, b, d, f)* $A_1 = 0.99\,\gamma_1$, $A_2 = 0.01\,\gamma_1$, $\Delta = 2.31\,\gamma_1$; *b, g, e, f)* $A_1 = A_2 = 0.5\,\gamma_1$, $\Delta = 4.11\,\gamma_1$, $V_2 = 0.1V_1$, $\nu_2 = 0.1\nu_1$, $\gamma = \Gamma_3 = \delta_1 = \delta_2 = \delta_3 = 0$, $\Gamma_1 = \Gamma_2 = 0.5\,\gamma_1$, $\tilde{\gamma}_{ph} = 0.4$, *and* $\tilde{\delta}_{ph} = 0.2$.

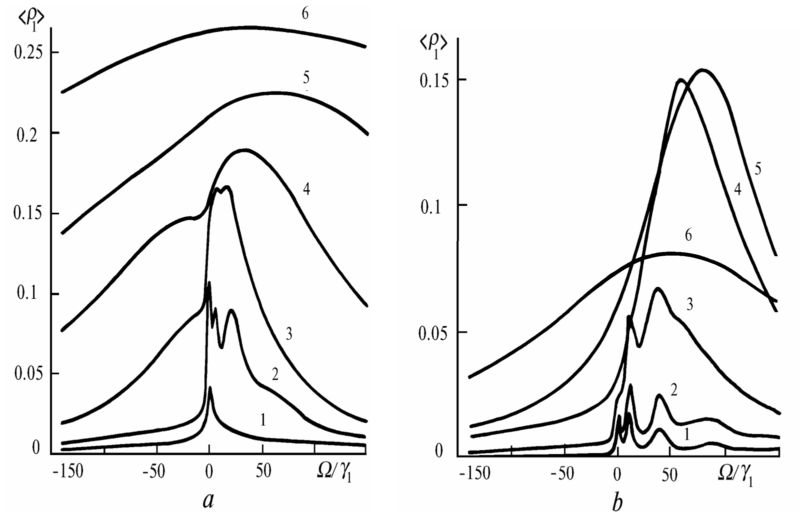


*FIG. 2 Fluorescence excitation spectra for different mean train durations and average power higher than that in Fig. 1.* $V_1 = \nu_1 = 16.33\ \gamma_1$, $V_2/V_1 = \nu_2/\nu_1 = 0.1$. *a)* $A_1 = 0.99\ \gamma_1$, $A_2 = 0.01\ \gamma_1$, *and* $\Delta = 2.31\ \gamma_1$ and *b)* $A_1 = A_2 = 0.5\ \gamma_1$, $\Delta = 4.11\ \gamma_1$. *Other designations being the same as in Fig. 1.*

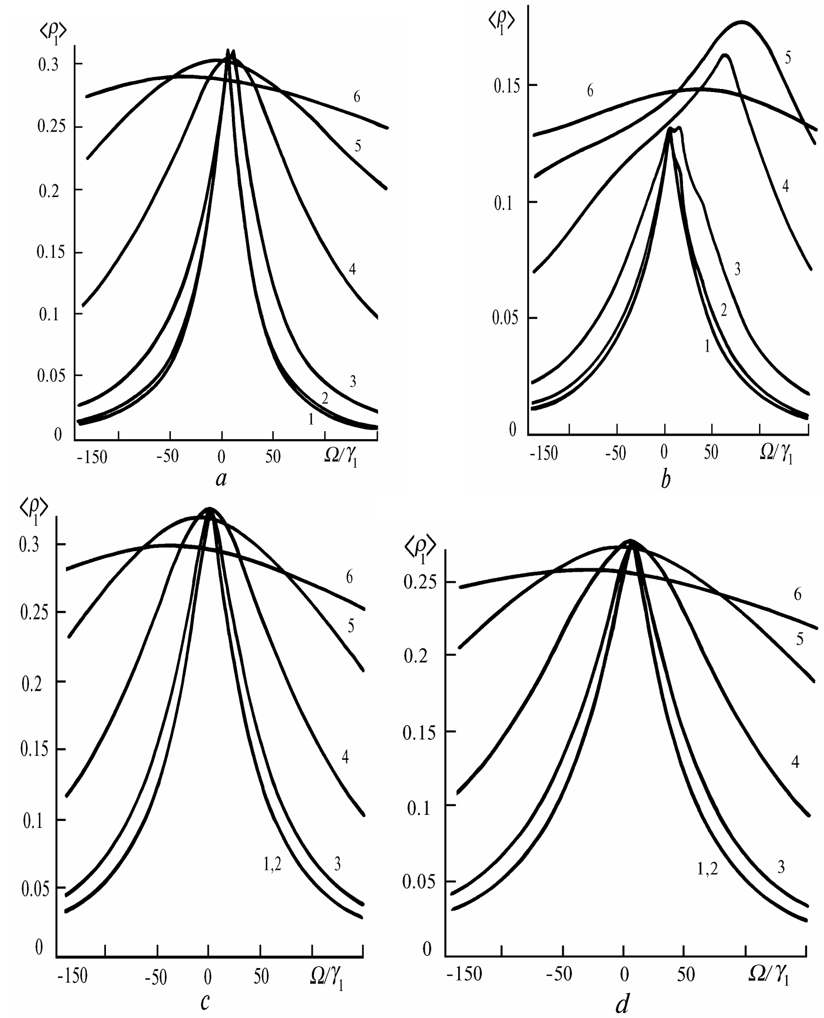


*FIG. 3 The effect of collisions on the shape of the spectrum of fluorescence excitation by the phase– and amplitude–modulated noise radiation for different mean train durations and* $A_1/A_2$ *ratios. a and c)* $\gamma = 0.1\ \gamma_1$, $\Gamma_3 = 0.15\ \gamma_1$, $\Gamma_1 = \Gamma_2 = 0.65\ \gamma_1$, $A_1 = 0.99\ \gamma_1$, *and* $A_2 = 0.01\ \gamma_1$, *b and d)* $\gamma = \gamma_1$, $\Gamma_3 = 1.5\ \gamma_1$, $\Gamma_1 = \Gamma_2 = 2\ \gamma_1$, $A_1 = A_2 = 0.5\ \gamma_1$. *The curve designations and the values of the rest parameters are the same as in Fig. 2.*

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