Resonance fluorescence with excitation of finite bandwidth*

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The problem of resonance fluorescence from a two-level atom that is driven near resonance by a laser beam of finite bandwidth is tackled via the Heisenberg equations of motion, by a simple generalization of our previous treatment. It is found that the two-time intensity correlation function is again factorizable into a product of the mean light intensities at the two times, each of which is slightly modified under broad-band excitation from its value under monochromatic excitation. The spectrum of the fluorescence is affected rather more drastically, and, in general, becomes asymmetric under broad-band excitation.

I. INTRODUCTION

Partly because of the development of the tunable dye laser, and the opportunity it provides for exciting resonant atomic transitions, the theory of resonance fluorescence has recently received a great deal of attention.[1] In most treatments the exciting field has been regarded as strictly monochromatic, and its quantum state has generally been taken to be a coherent state. We have recently obtained solutions for the growth of the fluorescent light intensity, for the spectral distribution, and for the intensity correlation function in the presence of such a monochromatic, coherent exciting field.[2,3] Although the monochromatic approximation is often entirely adequate to describe a laser field, there are circumstances when the bandwidth of the exciting field cannot be neglected, for example when very high resolution measurements are carried out.[4]

In the following we show how various results relating to atomic resonance fluorescence in the presence of a laser beam of finite bandwidth can readily be extracted from our previous treatment,[3] with minor modification. We model the laser field as being in a coherent state, but with a phase that performs a random walk. We recalculate various quantities of interest and subsequently average over the ensemble of phases. When the atom is driven near resonance, we find, as before, that the growth of the fluorescent light intensity and the two-time intensity correlation function are governed by similar integral equations with the same kernel. The time development of the fluorescence is modified somewhat by the finite bandwidth excitation, but the intensity correlation at times t and $t+\tau$ is again reducible to the product of the mean light intensities at times t and $t+\tau$ (see also Agarwal[5]). However, the kernel of the integral equation describing the amplitude correlation function, whose Fourier transform gives the spectral density of the fluorescence, is different from the other kernels, and in general yields an asymmetric spectrum. The effect of a finite bandwidth driving field on the spectrum is therefore much more drastic than on the other features of resonance fluorescence. Although some of the effects of finite bandwidth excitation have very recently been discussed,[6,7] these conclusions appear to be new. They are illustrated graphically for various values of the laser bandwidth.

II. TIME DEVELOPMENT OF THE LIGHT INTENSITY

We consider the resonance fluorescence of a two-level atom, of level spacing $\hbar\omega_0$ located at the origin $\vec{r}=0$, in the presence of the exciting field of a laser whose spectrum is centered at frequency ω_1. Our treatment is based on the solution of the Heisenberg equations of motion, and closely follows the approach that we used previously in Ref. 3. The atom is initially in some arbitrary, known state, and we suppose that the interaction is turned on at time $t=0$. The slowly varying atomic variables[8] $\hat{b}_s(t), \hat{b}_s^\dagger(t)$ are related to the usual atomic lowering and raising operators $\hat{b}(t), \hat{b}^\dagger(t)$ by

$$\hat{b}_s(t) \equiv \hat{b}(t)\, e^{i\omega_0 t}, \quad \hat{b}_s^\dagger(t) \equiv \hat{b}^\dagger(t)\, e^{-i\omega_0 t}, \tag{1}$$

and the slowly varying free-field operator $\hat{\vec{A}}_s^{(+)}(\vec{r},t)$ is related to the positive frequency part of the free-field vector potential $\hat{\vec{A}}_{\text{free}}^{(+)}(\vec{r},t)$ by

$$\hat{\vec{A}}_s^{(+)}(\vec{r},t) \equiv \hat{\vec{A}}_{\text{free}}^{(+)}(\vec{r},t)\, e^{i\omega_0 t}. \tag{2}$$

We shall take the eigenvalue of $\hat{\vec{A}}_s^{(+)}(0,t)$ in the coherent state $|\{v\}\rangle$ of the field to be given by

$$\hat{\vec{A}}_s^{(+)}(0,t)\,|\{v\}\rangle = \vec{V}(t)\,|\{v\}\rangle,$$

with (3)

$$\vec{V}(t) = \vec{\epsilon}\,\mathcal{C} e^{i(\omega_0-\omega_1)t} e^{i\phi(t)},$$

where $\vec{\epsilon}$ is a unit polarization vector, $\mathcal{C}$ is a real amplitude, and $\phi(t)$ a real phase. Whereas the

phase $\phi(t)$ was treated as a constant in Ref. 3, we now suppose that it performs in effect a random walk about zero, and we subsequently average over the phase ensemble, for which we introduce the symbol $\langle\langle\ \rangle\rangle$. As has been shown,[3] the positive frequency part of the fluorescent field produced by the atom at some point $\vec{r}$ in the far-field is proportional to $\hat{b}_s(t)$, through the relation

$$\hat{\vec{E}}^{(+)}(\vec{r},t)=\frac{\omega_0^2}{4\pi\epsilon_0 c^2}\left[\frac{\vec{\mu}}{r}-\frac{(\vec{\mu}\cdot\vec{r})\vec{r}}{r^3}\right]\times\hat{b}_s(t-r/c)\,e^{-i\omega_0(t-r/c)}+\hat{\vec{E}}^{(+)}_{\text{free}}(\vec{r},t)\,, \tag{4}$$

where $\vec{\mu}$ is the transition dipole moment. The mean fluorescent light intensity at points not exposed to the exciting beam is therefore given by

$$\langle\langle\,\hat{\vec{E}}^{(-)}(\vec{r},t+r/c)\cdot\hat{\vec{E}}^{(+)}(\vec{r},t+r/c)\rangle\rangle=\left(\frac{\omega_0^2\mu\sin\psi}{4\pi\epsilon_0 c^2 r}\right)^2\langle\langle\langle\hat{b}_s^\dagger(t)\hat{b}_s(t)\rangle\rangle$$

$$=\left(\frac{\omega_0^2\mu\sin\psi}{4\pi\epsilon_0 c^2 r}\right)^2\langle\langle\,[\langle\hat{R}_3(t)\rangle+\tfrac{1}{2}]\rangle\rangle\,,\qquad(t>0)\,, \tag{5}$$

where ψ is the angle between the $\vec{\mu}$ and $\vec{r}$ vectors. $\langle\langle\,[\langle\hat{R}_3(t)\rangle+\frac{1}{2}]\rangle\rangle$ is therefore proportional to the expected light intensity in the present case.

In Ref. 3 the Heisenberg equations of motion for the atomic and field operators were derived and integrated to yield the following two integral equations:

$$\hat{b}_s(t+\tau)=\hat{b}_s(t)\,e^{(-\beta+i\gamma)\tau}+\frac{2\omega_0}{\hbar}\int_0^\tau dt'\,\hat{R}_3(t+t')\,\vec{\mu}\cdot\hat{\vec{A}}_s^{(+)}(0,t+t')\,e^{(\beta-i\gamma)(t'-\tau)}\,, \tag{6}$$

$$\hat{R}_3(t+\tau)+\tfrac{1}{2}=[\hat{R}_3(t)+\tfrac{1}{2}]\,e^{-2\beta\tau}-\frac{\omega_0}{\hbar}\int_0^\tau dt'\,[\hat{b}_s^\dagger(t+t')\,\vec{\mu}\cdot\hat{\vec{A}}_s^{(+)}(0,t+t')+\text{H.c.}]\,e^{2\beta(t'-\tau)}\,. \tag{7}$$

Here β is half the Einstein A coefficient and γ is the Lamb shift. These two equations can be combined into one if we substitute for $\hat{b}_s(t)$ and $\hat{b}_s^\dagger(t)$ in Eq. (7) from Eq. (6). On taking expectation values, with the assumption that the initial state of the field is the coherent state with eigenvalue given by Eq. (3), and then averaging over the ensemble of phases, we find

$$\langle\langle\,[\langle\hat{R}_3(t)\rangle+\tfrac{1}{2}]\rangle\rangle=\langle\langle\,[\langle\hat{R}_3(0)\rangle+\tfrac{1}{2}]\rangle\rangle\,e^{-2\beta t}-\tfrac{1}{2}\Omega e^{-2\beta t}\int_0^t dt'\,[\langle\langle\langle\hat{b}_s^\dagger(0)\rangle\,e^{i\phi(t')}\rangle\rangle\,e^{\beta(1-i\theta)t'}+\text{c.c.}]$$

$$-\tfrac{1}{2}\Omega^2 e^{-2\beta t}\int_0^t dt'\left(e^{\beta(1-i\theta)t'}\int_0^{t'}dt''e^{\beta(1+i\theta)t''}\langle\langle\langle\hat{R}_3(t'')\rangle\,e^{i[\phi(t')-\phi(t'')]}\rangle\rangle+\text{c.c.}\right). \tag{8}$$

The parameter Ω is the Rabi frequency[9] given by

$$\Omega\equiv 2\vec{\mu}\cdot\vec{\mathcal{E}}\,\omega_0\mathcal{C}/\hbar\,, \tag{9a}$$

and θ is a dimensionless detuning parameter

$$\theta\equiv(\gamma+\omega_1-\omega_0)/\beta\,. \tag{9b}$$

If the ensemble averages under the integrals in Eq. (8) could be factorized, so that

$$\langle\langle\langle\hat{b}_s^\dagger(0)\rangle\,e^{i\phi(t')}\rangle\rangle=\langle\langle\langle\hat{b}_s^\dagger(0)\rangle\,e^{i\phi(0)}\rangle\rangle\langle\langle e^{i[\phi(t')-\phi(0)]}\rangle\rangle\,,\qquad(t'\geq 0)\,, \tag{10}$$

and

$$\langle\langle\langle\hat{R}_3(t'')\rangle\,e^{i[\phi(t')-\phi(t'')]}\rangle\rangle=\langle\langle\langle\hat{R}_3(t'')\rangle\rangle\langle\langle e^{i[\phi(t')-\phi(t'')]}\rangle\rangle\,,\qquad(t'\geq t'')\,, \tag{11}$$

then Eq. (8) would reduce to an integral equation for $\langle\langle\langle\hat{R}_3(t)\rangle\rangle\rangle$ as in Refs. 2 or 3. Fortunately these factorizations are entirely plausible, as $\phi(t')-\phi(0)$ and $\phi(t')-\phi(t'')$ are the changes of the randomly walking phase subsequent to the times 0 and t'', respectively, and the atomic variables are independent of phase changes occurring at subsequent times. As is usual for the random walk problem, we shall take the characteristic function of the phase change to be given by[10]

$$\langle\langle e^{i[\phi(t')-\phi(t'')]x}\rangle\rangle=e^{-\lambda|t'-t''|x^2}\,, \tag{12}$$

where $1/\lambda$ is the diffusion time, or λ is the effective bandwidth of the laser beam.

With the help of Eqs. (10)–(12), Eq. (8) then reduces to the form of a Volterra-type integral equation of the form

$$\langle\langle\langle\hat{R}_3(t)\rangle\rangle\rangle=y(t)+\int_0^t dt'\,K(t-t')\langle\langle\langle\hat{R}_3(t')\rangle\rangle\rangle\,, \tag{13}$$

in which

$$y(t) \equiv -\tfrac{1}{2} + \langle\langle [\langle \hat{R}_3(0)\rangle + \tfrac{1}{2}]\rangle\rangle\, e^{-2\beta t} - \tfrac{1}{2}\Omega\left(\langle\langle \langle \hat{b}_s^\dagger(0)\rangle\, e^{i\phi(0)}\rangle\rangle \frac{e^{-(\beta+\lambda+i\beta\theta)t} - e^{-2\beta t}}{\beta - \lambda - i\beta\theta} + \text{c.c.}\right), \tag{14}$$

$$K(\tau) \equiv \frac{-\Omega^2\{e^{-(\beta+\lambda)\tau}[(1-\lambda/\beta)\cos\beta\theta\tau + \theta\sin\beta\theta\tau] - (1-\lambda/\beta)e^{-2\beta\tau}\}}{\beta[(1-\lambda/\beta)^2+\theta^2]}. \tag{15}$$

These functions are very similar in form to the ones found in Ref. 3 for monochromatic excitation. In particular, the kernel $K(\tau)$ differs only in that some constants β have been replaced by $\beta+\lambda$ and others by $\beta-\lambda$. The solution to Eq. (13) can be written down directly with the help of Laplace transforms, as before,[3] and we find

$$\begin{aligned}\langle\langle [\langle \hat{R}_3(t)\rangle + \tfrac{1}{2}]\rangle\rangle = {} & \frac{\frac{1}{4}\Omega^2(1+\lambda/\beta)}{\frac{1}{2}\Omega^2(1+\lambda/\beta) + (\beta+\lambda)^2 + \beta^2\theta^2} - \frac{1}{2}\sum_{\substack{i=1\\ i\neq j\neq k}}^{3} \frac{(2\beta+p_i)[(\beta+\lambda+p_i)^2+\beta^2\theta^2]e^{p_i t}}{p_i(p_i-p_j)(p_i-p_k)} \\ & + \langle\langle [\langle \hat{R}_3(0)\rangle + \tfrac{1}{2}]\rangle\rangle \sum_{\substack{i=1\\ i\neq j\neq k}}^{3} \frac{[(\beta+\lambda+p_i)^2+\beta^2\theta^2]e^{p_i t}}{(p_i-p_j)(p_i-p_k)} \\ & - \tfrac{1}{2}\Omega\langle\langle \langle \hat{b}_s^\dagger(0)\rangle\, e^{i\phi(0)}\rangle\rangle \sum_{\substack{i=1\\ i\neq j\neq k}}^{3} \frac{(\beta+\lambda+p_i-i\beta\theta)e^{p_i t}}{(p_i-p_j)(p_i-p_k)} \\ & - \tfrac{1}{2}\Omega\langle\langle \langle \hat{b}_s(0)\rangle\, e^{-i\phi(0)}\rangle\rangle \sum_{\substack{i=1\\ i\neq j\neq k}}^{3} \frac{(\beta+\lambda+p_i+i\beta\theta)e^{p_i t}}{(p_i-p_j)(p_i-p_k)},\end{aligned} \tag{16}$$

where p_1, p_2, p_3 are the three roots (assumed to be unequal) of the cubic equation

$$p^3 + (4\beta+2\lambda)p^2 + [(\beta+\lambda)(5\beta+\lambda) + \beta^2\theta^2 + \Omega^2]p + 2\beta(\beta+\lambda)^2 + 2\beta^3\theta^2 + (\beta+\lambda)\Omega^2 = 0. \tag{17}$$

Figure 1 shows the effect of a progressively increasing linewidth on the time development of the fluorescent light intensity, for various values of the ratio λ/β, for an atom that is initially in the lower state. In order to gain some appreciation of the magnitude of this effect, we note that for a typical atomic transition in sodium with $\beta = 6$ MHz, a dye laser linewidth of $\lambda = 1$ MHz corresponds to $\lambda/\beta \sim 17\%$. The effect on the time development of the light intensity is therefore expected to be small in general.

III. INTENSITY CORRELATIONS OF THE FLUORESCENT LIGHT

If a photodetector located at position $\vec{r}$ in the far-field outside the laser beam is used to study the fluorescence emitted by the atom, the joint probability of photoelectric detection at times t and $t+\tau$ is proportional to the expectation of the normally ordered product[11]

$$\Gamma^{(2,2)}(\vec{r}, t, \tau) \equiv \langle\langle \hat{E}_i^{(-)}(\vec{r}, t)\hat{E}_j^{(-)}(\vec{r}, t+\tau) \times \hat{E}_j^{(+)}(\vec{r}, t+\tau)\hat{E}_i^{(+)}(\vec{r}, t)\rangle\rangle, \tag{18}$$

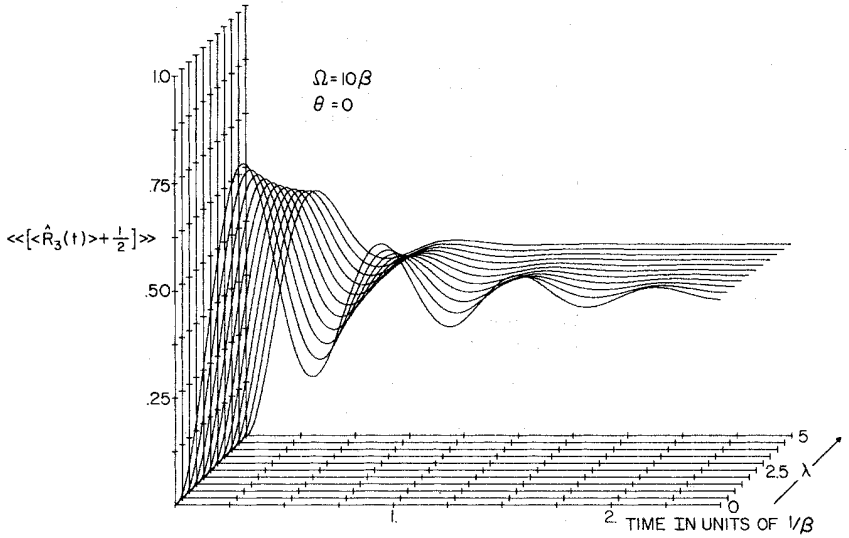

FIG. 1. Time development of the light intensity $\langle\langle[\langle \hat{R}_3(t)\rangle + \frac{1}{2}]\rangle\rangle$ radiated by an atom that is initially in the ground state, for various values of the excitation bandwidth λ, with Rabi frequency $\Omega = 10\beta$, and no detuning ($\theta = 0$).

and with the help of Eq. (4) together with the commutation relation[12]

$$[\hat{b}_s(t), \hat{\vec{E}}^{(+)}_{\text{free}}(\vec{r}, t+\tau)] = 0, \quad \text{for } \tau > r/c\,, \tag{19}$$

this can be written

$$\Gamma^{(2,2)}(\vec{r}, t, \tau) = \left(\frac{\omega_0{}^2 \mu \sin\psi}{4\pi\epsilon_0 c^2 r}\right)^4 \langle\!\langle [\langle \hat{b}_s{}^\dagger(t - r/c)\hat{R}_3(t - r/c + \tau)\hat{b}_s(t - r/c)\rangle + \tfrac{1}{2}\langle \hat{R}_3(t - r/c)\rangle + \tfrac{1}{4}]\rangle\!\rangle\,. \tag{20}$$

In order to calculate $\Gamma^{(2,2)}(\vec{r}, t, \tau)$ we therefore need to determine the average over the phase ensemble of the correlation function

$$\mathcal{K}(t, \tau) \equiv \langle \hat{b}_s{}^\dagger(t)\hat{R}_3(t+\tau)\hat{b}_s(t)\rangle\,. \tag{21}$$

As before,[3] it is convenient to define another function $\mathcal{F}(t, \tau)$ by

$$\mathcal{F}(t, \tau) \equiv \langle \hat{b}_s{}^\dagger(t)\hat{b}_s{}^\dagger(t+\tau)\hat{b}_s(t)\rangle \times e^{i\phi(t+\tau)} e^{i(\omega_0 - \omega_1)(t+\tau)}\,. \tag{22}$$

With the help of the commutation relations (19), with the proviso $\tau \geq 0$, we then have by direct multiplication from Eq. (7),

$$\langle\!\langle \mathcal{K}(t, \tau)\rangle\!\rangle = -\tfrac{1}{2}\langle\!\langle [\langle \hat{R}_3(t)\rangle + \tfrac{1}{2}]\rangle\!\rangle - \tfrac{1}{2}\Omega \int_0^\tau dt' [\langle\!\langle \mathcal{F}(t, t')\rangle\!\rangle + \text{c.c.}] e^{2\beta(t'-\tau)}\,, \tag{23}$$

and from Eq. (6)

$$\langle\!\langle \mathcal{F}(t, \tau)\rangle\!\rangle = \Omega \int_0^\tau dt' \langle\!\langle \mathcal{K}(t, t') e^{i[\phi(t+\tau) - \phi(t+t')]}\rangle\!\rangle \times e^{\beta(1+i\theta)(t'-\tau)}\,. \tag{24}$$

As the phase change during the time interval $t+t'$ to $t+\tau$, which follows time $t+t'$, is expected to be uncorrelated with the atomic variables at earlier times, we again factorize the average of the product with the help of Eq. (12),

$$\begin{aligned}\langle\!\langle \mathcal{K}(t, t') e^{i[\phi(t+\tau) - \phi(t+t')]}\rangle\!\rangle &= \langle\!\langle \mathcal{K}(t, t')\rangle\!\rangle \langle\!\langle e^{i[\phi(t+\tau) - \phi(t+t')]}\rangle\!\rangle \\ &= \langle\!\langle \mathcal{K}(t, t')\rangle\!\rangle e^{-\lambda(\tau - t')}, \quad 0 \leq t' \leq \tau\,,\end{aligned} \tag{25}$$

and then substitute in Eq. (24), to obtain

$$\langle\!\langle \mathcal{F}(t, \tau)\rangle\!\rangle = \Omega \int_0^\tau dt' \langle\!\langle \mathcal{K}(t, t')\rangle\!\rangle e^{-(\beta + \lambda + i\beta\theta)(\tau - t')}\,. \tag{26}$$

If we now write the expression for $\langle\!\langle \mathcal{F}(t, \tau)\rangle\!\rangle$ given by Eq. (26) in Eq. (23), we arrive at the following integral equation for $\langle\!\langle \mathcal{K}(t, \tau)\rangle\!\rangle$, exactly as in Ref. 3,

$$\langle\!\langle \mathcal{K}(t, \tau)\rangle\!\rangle = -\tfrac{1}{2}\langle\!\langle [\langle \hat{R}_3(t)\rangle + \tfrac{1}{2}]\rangle\!\rangle + \int_0^\tau dt' K(\tau - t')\langle\!\langle \mathcal{K}(t, t')\rangle\!\rangle\,, \tag{27}$$

where $K(\tau)$ is the integral kernel that was already encountered in Eqs. (13) and (15). The inhomogeneous term $-\frac{1}{2}\langle\!\langle [\langle \hat{R}_3(t)\rangle + \frac{1}{2}]\rangle\!\rangle$ exceeds the inhomogeneous term $y(t)$ given by Eq. (14) by the factor $\langle\!\langle [\langle \hat{R}_3(t)\rangle + \frac{1}{2}]\rangle\!\rangle$ in the special case in which the atom starts in the lower or ground state at time $t = 0$. We may therefore make use of the previously found solution to the integral equation (13) to write down the solution to Eq. (27), viz.

$$\langle\!\langle \mathcal{K}(t, \tau)\rangle\!\rangle = \langle\!\langle [\langle \hat{R}_3(t)\rangle + \tfrac{1}{2}]\rangle\!\rangle \langle\!\langle \langle \hat{R}_3(\tau)\rangle_G\rangle\!\rangle\,, \tag{28}$$

where the suffix G refers to the fact that the initial atomic state is taken to be the ground state. From Eqs. (19) and (28) we then have, for $\tau > 0$,

$$\Gamma^{(2,2)}(\vec{r}, t + r/c, \tau) = \left(\frac{\omega_0{}^2 \mu \sin\psi}{4\pi\epsilon_0 c^2 r}\right)^4 \langle\!\langle [\langle \hat{R}_3(t)\rangle + \tfrac{1}{2}]\rangle\!\rangle \times \langle\!\langle [\langle \hat{R}_3(\tau)\rangle_G + \tfrac{1}{2}]\rangle\!\rangle\,. \tag{29}$$

Apart from the appearance of the ensemble averages, Eq. (29) is formally identical with the corresponding result found in Ref. 3 for monochromatic excitation, and its interpretation is similar also. The t-dependent factor gives the probability for the emission of a photon at time t following the turn-on of the interaction, after which the atom returns to the lower state, and the τ-dependent factor gives the subsequent emission probability τ seconds later. The effect of the finite bandwidth is merely to modify the light intensity, as expressed by the factors $\langle\!\langle [\langle \hat{R}_3(t)\rangle + \frac{1}{2}]\rangle\!\rangle$ and $\langle\!\langle [\langle \hat{R}_3(\tau)\rangle_G + \frac{1}{2}]\rangle\!\rangle$. Figure 1 therefore shows the effect of broadening the bandwidth of the exciting field on both the t dependence and the τ dependence of the intensity correlation function $\Gamma^{(2,2)}(\vec{r}, t + r/c, \tau)$.

IV. SPECTRAL DENSITY OF THE FLUORESCENCE

From Eq. (4) the two-time amplitude correlation function of the fluorescent light at some point $\vec{r}$ in the far-field of the atom outside the laser beam is given by

$$\langle\!\langle \langle \hat{\vec{E}}^{(-)}(\vec{r}, t) \cdot \hat{\vec{E}}^{(+)}(\vec{r}, t+\tau)\rangle\!\rangle = \left(\frac{\omega_0{}^2 \mu \sin\psi}{4\pi\epsilon_0 c^2 r}\right)^2 e^{-i\omega_0\tau} \times \langle\!\langle \langle \hat{b}_s{}^\dagger(t - r/c)\hat{b}_s(t - r/c + \tau)\rangle\!\rangle\,, \tag{30}$$

and the spectral density in the steady state is just the Fourier transform of $\langle\langle \hat{\vec{E}}^{(-)}(\vec{r},t)\cdot\hat{\vec{E}}^{(+)}(\vec{r},t+\tau)\rangle\rangle$ with respect to τ. We therefore proceed to determine $\langle\langle\langle\hat{b}_s^\dagger(t)\hat{b}_s(t+\tau)\rangle\rangle\rangle$ from the equations of motion (6) and (7), as in Ref. 3.

We define the following correlation functions, which are generalizations of ones introduced by Milonni[13]:

$$g(t,\tau)\equiv\langle\hat{b}_s^\dagger(t)\hat{b}_s(t+\tau)\rangle\, e^{i(\omega_1-\omega_0)\tau}, \tag{31a}$$

$$f(t,\tau)\equiv\langle\hat{b}_s^\dagger(t)\hat{b}_s^\dagger(t+\tau)\rangle\, e^{2i\phi(t+\tau)}e^{i(\omega_0-\omega_1)(2t+\tau)}, \tag{31b}$$

$$h(t,\tau)\equiv\langle\hat{b}_s^\dagger(t)\hat{R}_3(t+\tau)\rangle\, e^{i\phi(t+\tau)}e^{i(\omega_0-\omega_1)t}. \tag{31c}$$

We then find with the help of Eq. (19) from Eqs. (6) and (7), by direct multiplication, with the proviso $\tau\geq 0$,

$$\langle\langle g(t,\tau)\rangle\rangle = \langle\langle[\langle\hat{R}_3(t)\rangle+\tfrac{1}{2}]\rangle\rangle\, e^{-\beta(1-i\theta)\tau} + \Omega\int_0^\tau dt'\,\langle\langle h(t,t')\rangle\rangle\, e^{\beta(1-i\theta)(t'-\tau)}, \tag{32}$$

$$\langle\langle f(t,\tau)\rangle\rangle = \Omega\int_0^\tau dt'\,\langle\langle h(t,t')\, e^{2i[\phi(t+\tau)-\phi(t+t')]}\rangle\rangle \times e^{\beta(1+i\theta)(t'-\tau)}, \tag{33}$$

$$\langle\langle h(t,\tau)\rangle\rangle = -\tfrac{1}{2}\langle\langle\langle\hat{b}_s^\dagger(t)\rangle\, e^{i\phi(t+\tau)}\rangle\rangle\, e^{i(\omega_0-\omega_1)t} - \tfrac{1}{2}\Omega\int_0^\tau dt'\,[\langle\langle f(t,t')\rangle\rangle+\langle\langle g(t,t')\rangle\rangle] \times e^{i[\phi(t+\tau)-\phi(t+t')]}\rangle\rangle\, e^{2\beta(t'-\tau)}. \tag{34}$$

In order to solve these equations for $\langle\langle g(t,\tau)\rangle\rangle$ we first observe that, as the time interval $t+t'$ to $t+\tau$ follows $t+t'(t'\leq\tau)$, we may factorize the correlation functions under the integral in Eqs. (33) and (34) as before, and write

$$\langle\langle h(t,t')\, e^{2i[\phi(t+\tau)-\phi(t+t')]}\rangle\rangle = \langle\langle h(t,t')\rangle\rangle\langle\langle e^{2i[\phi(t+\tau)-\phi(t+t')]}\rangle\rangle = \langle\langle h(t,t')\rangle\rangle\, e^{-4\lambda(\tau-t')}, \quad t'\leq\tau \tag{35}$$

and

$$\langle\langle g(t,t')\, e^{i[\phi(t+\tau)-\phi(t+t')]}\rangle\rangle = \langle\langle g(t,t')\rangle\rangle\langle\langle e^{i[\phi(t+\tau)-\phi(t+t')]}\rangle\rangle = \langle\langle g(t,t')\rangle\rangle\, e^{-\lambda(\tau-t')}, \quad t'\leq\tau,$$

$$\langle\langle f(t,t')\, e^{i[\phi(t+\tau)-\phi(t+t')]}\rangle\rangle = \langle\langle f(t,t')\rangle\rangle\, e^{-\lambda(\tau-t')}, \quad t'\leq\tau, \tag{36}$$

after making use of Eq. (12) for the characteristic function of the phase change. Equations (33) and (34) then simplify to

$$\langle\langle f(t,\tau)\rangle\rangle = \Omega\int_0^\tau dt'\,\langle\langle h(t,t')\rangle\rangle\, e^{-(\beta+4\lambda+i\beta\theta)(\tau-t')} \tag{37}$$

and

$$\langle\langle h(t,\tau)\rangle\rangle = -\tfrac{1}{2}\langle\langle\langle\hat{b}_s^\dagger(t)\rangle\, e^{i\phi(t+\tau)}\rangle\rangle\, e^{i(\omega_0-\omega_1)t} - \tfrac{1}{2}\Omega\int_0^\tau dt'\,[\langle\langle f(t,t')\rangle\rangle+\langle\langle g(t,t')\rangle\rangle] \times e^{-(2\beta+\lambda)(\tau-t')}. \tag{38}$$

The solution for $\langle\langle[\langle\hat{R}_3(t)\rangle+\tfrac{1}{2}]\rangle\rangle$, which is needed in Eq. (32), has already been obtained and is given by Eq. (16). To determine $\langle\langle\langle\hat{b}_s^\dagger(t)\rangle\exp[i\phi(t+\tau)]\rangle\rangle$, which is needed in Eq. (38), we make use of Eq. (6) with $t\to 0$ and $\tau\to t$ and take expectation values. We then find

$$\langle\langle\langle\hat{b}_s^\dagger(t)\rangle\, e^{i\phi(t+\tau)}\rangle\rangle\, e^{i(\omega_0-\omega_1)t} = \langle\langle\langle\hat{b}_s^\dagger(0)\rangle\, e^{i\phi(0)}e^{i[\phi(t+\tau)-\phi(0)]}\rangle\rangle\, e^{-\beta(1+i\theta)t} + \Omega\int_0^t dt'\,\langle\langle\langle\hat{R}_3(t')\rangle\, e^{i[\phi(t+\tau)-\phi(t')]}\rangle\rangle\, e^{\beta(1+i\theta)(t'-t)}, \tag{39}$$

and, with the help of the factorizations (10) and (11), this simplifies to

$$\langle\langle\langle\hat{b}_s^\dagger(t)\rangle\, e^{i\phi(t+\tau)}\rangle\rangle\, e^{i(\omega_0-\omega_1)t} = \langle\langle\langle\hat{b}_s^\dagger(0)\rangle\, e^{i\phi(0)}\rangle\rangle\, e^{-(\beta+\lambda+i\beta\theta)t}e^{-\lambda\tau} + \Omega e^{-\lambda\tau}\int_0^t dt'\,\langle\langle\langle\hat{R}_3(t')\rangle\rangle\, e^{-(\beta+\lambda+i\beta\theta)(t-t')}. \tag{40}$$

with $\langle\langle\langle\hat{R}_3(t')\rangle\rangle\rangle$ given by Eq. (16). Hence every function other than $\langle\langle g(t,\tau)\rangle\rangle$, $\langle\langle f(t,\tau)\rangle\rangle$, $\langle\langle h(t,\tau)\rangle\rangle$ in Eqs. (32), (37), and (38) is known once $\langle\langle\langle\hat{R}_3(t)\rangle\rangle\rangle$ has been determined from Eq. (16), and we merely have to solve these three simultaneous equations as in Ref. 3 in order to obtain the required correlation function $\langle\langle g(t,\tau)\rangle\rangle$.

The equations can be solved by Laplace transform techniques. Alternatively, we may observe that addition of Eqs. (32) and (37), with the substitution for $\langle\langle h(t,\tau)\rangle\rangle$ from Eq. (38), immediately leads to an integral equation for the sum $\langle\langle f(t,\tau)\rangle\rangle+\langle\langle g(t,\tau)\rangle\rangle$, of the form

$$\langle\langle f(t,\tau)\rangle\rangle+\langle\langle g(t,\tau)\rangle\rangle = z(t,\tau)+\int_0^\tau dt'\,M(\tau-t')\,[\langle\langle f(t,t')\rangle\rangle+\langle\langle g(t,t')\rangle\rangle], \tag{41}$$

with

$$z(t,\tau)\equiv\langle\langle[\langle\hat{R}_3(t)\rangle+\tfrac{1}{2}]\rangle\rangle\, e^{-\beta(1-i\theta)\tau}-\tfrac{1}{2}\Omega e^{-(\beta+\lambda+i\beta\theta)t}\left(\frac{e^{-\lambda\tau}-e^{-(\beta+4\lambda+i\beta\theta)\tau}}{\beta+3\lambda+i\beta\theta}+\frac{e^{-\lambda\tau}-e^{-\beta(1-i\theta)\tau}}{\beta-\lambda-i\beta\theta}\right)$$
$$\times\left(\langle\langle\langle\hat{b}_s^{\dagger}(0)\rangle e^{i\phi(0)}\rangle\rangle+\Omega\int_0^t dt'\,\langle\langle\langle\hat{R}_3(t')\rangle\rangle e^{(\beta+\lambda+i\beta\theta)t'}\right), \tag{42}$$

and kernel

$$M(\tau)\equiv-\frac{\Omega^2}{2}\left(\frac{e^{-(\beta+4\lambda+i\beta\theta)\tau}-e^{-(2\beta+\lambda)\tau}}{\beta-3\lambda-i\beta\theta}+\frac{e^{-\beta(1-i\theta)\tau}-e^{-(2\beta+\lambda)\tau}}{\beta+\lambda+i\beta\theta}\right). \tag{43}$$

The solution can therefore be written down as before, and substitution in Eq. (38) yields $\langle\langle h(t,\tau)\rangle\rangle$, from which $\langle\langle g(t,\tau)\rangle\rangle$ follows from Eq. (32). We note that the kernel $M(\tau)$ is complex and differs significantly from the one we encountered in Eq. (15), although they become identical in the limit of monochromatic excitation, when $\lambda=0$. The two-time amplitude correlation of the field therefore reflects some features that are not present in the behavior of the light intensity. We shall see that the complex nature of the kernel is reflected in an asymmetry in the spectrum of the fluorescence.

After straightforward but somewhat tedious manipulation of terms, we arrive at the following general solution for $\langle\langle g(t,\tau)\rangle\rangle$:

$$\langle\langle g(t,\tau)\rangle\rangle=\frac{-\beta\Omega(\beta+3\lambda+i\beta\theta)}{C(-\lambda)}\langle\langle\langle\hat{b}_s^{\dagger}(t)\rangle e^{i\phi(t)}\rangle\rangle e^{i(\omega_0-\omega_1)t}e^{-\lambda\tau}$$
$$+\sum_{\substack{i=1\\ i\neq j\neq k}}^{3}\frac{e^{p_i\tau}}{(p_i+\lambda)(p_i-p_j)(p_i-p_k)}\{\langle\langle[\langle\hat{R}_3(t)\rangle+\tfrac{1}{2}]\rangle\rangle(p_i+\lambda)[(p_i+2\beta+\lambda)(p_i+\beta+4\lambda+i\beta\theta)+\tfrac{1}{2}\Omega^2]$$
$$-\tfrac{1}{2}\Omega(p_i+2\beta+\lambda)(p_i+\beta+4\lambda+i\beta\theta)\langle\langle\langle\hat{b}_s^{\dagger}(t)\rangle e^{i\phi(t)}\rangle\rangle e^{i(\omega_0-\omega_1)t}\},$$
$$(\tau\geq 0) \tag{44}$$

in which the t-dependent terms are given by Eqs. (16) and (40), $C(p)$ is the third-order polynomial defined by

$$C(p)\equiv p^3+p^2[4\beta+5\lambda]+p[5\beta^2+\beta^2\theta^2+\Omega^2+\lambda(14\beta+4\lambda-4i\beta\theta)]$$
$$+2\beta^3+\beta\Omega^2+2\beta^3\theta^2+\lambda(9\beta^2+2\Omega^2+4\lambda\beta+\beta^2\theta^2-8i\beta^2\theta-4i\beta\lambda\theta), \tag{45}$$

and p_1, p_2, p_3 are the three roots (assumed to be unequal) of the cubic equation

$$C(p)=0. \tag{46}$$

It is worth noting that this complex equation again differs from the real cubic Eq. (17) that we encountered in connection with the problem of determining the time development of the light intensity. Although both reduce to the same well-known equation[1-3] when $\lambda=0$, the effect of finite bandwidth excitation is clearly more pronounced on the correlation function $\langle\langle g(t,\tau)\rangle\rangle$ than on the light intensity.

As time $t\to\infty$, the t-dependent terms in Eq. (44) become t-independent, and we find from Eqs. (16) and (40) that they are given by

$$\lim_{t\to\infty}\langle\langle[\langle\hat{R}_3(t)\rangle+\tfrac{1}{2}]\rangle\rangle=\frac{\frac{1}{4}\Omega^2/\beta^2}{\frac{1}{2}\Omega^2/\beta^2+(1+\lambda/\beta)+\theta^2/(1+\lambda/\beta)}, \tag{47}$$

and

$$\lim_{t\to\infty}\langle\langle\langle\hat{b}_s^{\dagger}(t)\rangle e^{i\phi(t+\tau)}\rangle\rangle e^{i(\omega_0-\omega_1)t}=\frac{-\frac{1}{2}(\Omega/\beta)(1+\lambda/\beta-i\theta)e^{-\lambda\tau}}{[\frac{1}{2}(\Omega^2/\beta^2)(1+\lambda/\beta)+(1+\lambda/\beta)^2+\theta^2]}. \tag{48}$$

Hence the atomic correlation function $\langle\langle g(t,\tau)\rangle\rangle$ reaches the steady-state form, as $t\to\infty$,

$$\langle\langle g(\infty,\tau)\rangle\rangle = \frac{\frac{1}{4}\Omega^2/\beta^2}{\frac{1}{2}(\Omega^2/\beta^2)(1+\lambda/\beta)+(1+\lambda/\beta)^2+\theta^2}$$
$$\times\left\{\frac{(\beta+3\lambda+i\beta\theta)(\beta+\lambda-i\beta\theta)e^{-\lambda\tau}}{(\beta+3\lambda+i\beta\theta)(\beta-\lambda-i\beta\theta)+\frac{1}{2}\Omega^2(1+\lambda/\beta)}\right.$$
$$\left.-\tfrac{1}{2}\Omega^2\sum_{\substack{i=1\\ i\neq j\neq k}}^{3}\frac{(p_i+2\beta)-(\lambda/\beta)(p_i-3\beta+\lambda)-(2\lambda/\beta\Omega^2)(p_i+\beta+4\lambda+i\beta\theta)(p_i+2\beta+\lambda)^2}{(p_i+\lambda)(p_i-p_j)(p_i-p_k)}e^{p_i\tau}\right\}$$
$$(\tau\geq 0). \quad (49)$$

If we take the Fourier transform with respect to τ of the correlation function $\langle\langle \hat{\vec{E}}^{(-)}(\vec{r},t)\cdot\hat{\vec{E}}^{(+)}(\vec{r},t+\tau)\rangle\rangle$ in the long-time limit, we arrive at the spectral distribution $\Phi(\vec{r},\omega)$ of the fluorescent light at position $\vec{r}$. Thus from Eqs. (30) and (49), we obtain

$$\Phi(\vec{r},\omega)=\int_0^\infty d\tau\lim_{t\to\infty}\langle\langle\langle\hat{\vec{E}}^{(-)}(\vec{r},t)\cdot\hat{\vec{E}}^{(+)}(\vec{r},t+\tau)\rangle\rangle\rangle e^{i\omega\tau}+\text{c.c.}$$
$$=\left(\frac{\omega_0^2\mu\sin\psi}{4\pi\epsilon_0c^2r}\right)^2\int_0^\infty d\tau\left[\langle\langle g(\infty,\tau)\rangle\rangle e^{i(\omega-\omega_1)\tau}+\text{c.c.}\right]$$
$$=\left(\frac{\omega_0^2\mu\sin\psi}{4\pi\epsilon_0c^2r}\right)^2\left(\frac{\frac{1}{2}\Omega^2/\beta^2}{\frac{1}{2}(\Omega^2/\beta^2)(1+\lambda/\beta)+(1+\lambda/\beta)^2+\theta^2}\right)$$
$$\times\left(\frac{\frac{1}{2}(\beta+3\lambda+i\beta\theta)(\beta+\lambda-i\beta\theta)}{[(\beta+3\lambda+i\beta\theta)(\beta-\lambda-i\beta\theta)+\frac{1}{2}\Omega^2(1+\lambda/\beta)]}\frac{1}{[\lambda+i(\omega_1-\omega)]}\right.$$
$$\left.-\tfrac{1}{4}\Omega^2\sum_{\substack{i=1\\ i\neq j\neq k}}^{3}\frac{(p_i+2\beta)-(\lambda/\beta)(p_i-3\beta+\lambda)-(2\lambda/\beta\Omega^2)(p_i+\beta+4\lambda+i\beta\theta)(p_i+2\beta+\lambda)^2}{(p_i+\lambda)(p_i-p_j)(p_i-p_k)[-p_i+i(\omega_1-\omega)]}+\text{c.c.}\right). \quad (50)$$

The interpretation of this spectral density follows the same lines as for monochromatic excitation. The first term on the right of Eq. (50) corresponds to elastically scattered light, whose bandwidth largely reflects the spectral width λ of the driving field. However, off resonance, when $\theta\neq 0$, even this contribution to the spectral density contains an asymmetric component, that vanishes when $\theta=0$ or $\lambda\to 0$. In the monochromatic limit $\lambda\to 0$ the term becomes proportional to $\delta(\omega-\omega_1)$.

The remaining three terms represent the atomic fluorescence, and they contribute peaks that are centered at frequencies determined by the three roots p_1, p_2, p_3 of the cubic equation (46). These spectral contributions are again asymmetric in frequency off resonance, when $\theta\neq 0$, but become symmetric on resonance.

To illustrate the analytic structure of the spectral distribution (50) in the simplest cases, we consider the strong-field limit $\Omega\gg\beta,\lambda$ on resonance, when $\theta=0$. We readily find that the three roots of the cubic equation (46) are then approximately given by

$$p_1\simeq-(\beta+2\lambda),\quad p_2\simeq-\tfrac{3}{2}(\beta+\lambda)+i\Omega,\quad p_3\simeq-\tfrac{3}{2}(\beta+\lambda)-i\Omega, \quad (51)$$

and the spectral density reduces to

$$\Phi(\vec{r},\omega)\approx\left(\frac{\omega_0^2\mu\sin\psi}{4\pi\epsilon_0c^2r}\right)^2\left(\frac{2\beta(\beta+3\lambda)}{\Omega^2(1+\lambda/\beta)}\frac{1}{[\lambda^2+(\omega-\omega_1)^2]}\right.$$
$$\left.+\frac{\frac{1}{2}(\beta+2\lambda)}{(\beta+2\lambda)^2+(\omega-\omega_1)^2}+\frac{\frac{3}{8}(\beta+\lambda)}{\frac{9}{4}(\beta+\lambda)^2+(\omega-\omega_1+\Omega)^2}+\frac{\frac{3}{8}(\beta+\lambda)}{\frac{9}{4}(\beta+\lambda)^2+(\omega-\omega_1-\Omega)^2}\right). \quad (52)$$

As in the case of monochromatic excitation, there are peaks in the spectrum centered at the driving frequency ω_1, one of which is associated with elastic scattering, and at the Stark-shifted frequencies $\omega_1\pm\Omega$. For the inelastic components, the ratio of the width of the central peak to the side peaks is $2(\beta+2\lambda)/3(\beta+\lambda)$ and the ratio of the height of the central peak to the side peaks is $3(\beta+\lambda)/(\beta+2\lambda)$. These reduce to the well known ratios 2:3 and 3:1, respectively, under monochromatic excitation, but to the ratios 4:3 and 3:2 in the limit of wide band excitation $\lambda\gg\beta$. Similar conclusions were also recently reached by Eberly[6] and Agarwal[7] from other arguments.

Perhaps the most interesting aspects of the spectral distribution (50) relate to its asymmetries off-resonance ($\theta\neq 0$), because these features are not encountered at all under monochromatic excitation,[1,3] and appear never to have been investigated before. For purposes of numerical computation, it is some-

what more convenient to rewrite Eq. (50) in a form that is more analogous to the form that was first given by Mollow[1] for monochromatic excitation. If we replace the sum over the roots by an integral over the p plane, we can transform Eq. (50) and arrive at the expression

$$\Phi(\vec{r},\omega)=\left(\frac{\omega_0^2\mu\sin\psi}{4\pi\epsilon_0 c^2 r}\right)^2\frac{\frac{1}{2}\Omega^2/\beta^2}{(\frac{1}{2}\Omega^2/\beta^2)(1+\lambda/\beta)+(1+\lambda/\beta)^2+\theta^2}$$
$$\times\left[\frac{\lambda}{\lambda^2+(\omega-\omega_1)^2}\right.$$
$$\left.-\tfrac{1}{4}\Omega^2\left(\frac{i(\omega_1-\omega)(1-\lambda/\beta)+2\beta+3\lambda-\lambda^2/\beta-(2\lambda/\beta\Omega^2)[i(\omega_1-\omega)+\beta+4\lambda+i\beta\theta][i(\omega_1-\omega)+2\beta+\lambda]^2}{C[i(\omega_1-\omega)][\lambda+i(\omega_1-\omega)]}+\text{c.c.}\right)\right], \tag{53}$$

where $C(p)$ is the polynomial defined by Eq. (45). This spectral distribution which is symmetric under simultaneous sign change of θ and $\omega_1-\omega$, has been evaluated for several different values of the Rabi frequency Ω, the detuning θ and the excitation bandwidth λ, and the results are shown in Figs. 2–6. Note that in some graphs the central peaks are off scale.

Let us first examine the situation when the exciting field is relatively weak. Figure 2 shows the spectral density for $\Omega=0.1\beta$, for fixed detuning $\theta=-5$, and for gradually increasing bandwidth of the laser field. Under monochromatic excitation, the fluorescence is dominated by elastic scattering; the spectrum is symmetric and is centered at the driving frequency ω_1. However, as the excitation bandwidth λ increases, the elastic contribution rapidly becomes less important than the inelastic one, which tends to become centered at the atomic frequency.

Figures 3 and 4 show the corresponding situation for a strong, exciting field, with $\Omega=10\beta$, on resonance ($\theta=0$) and off resonance ($\theta=-3$), respectively. With monochromatic excitation the Stark splitting leads to the characteristic three-peaked, symmetric spectrum centered at the driving frequency ω_1. As the excitation bandwidth λ increases, the central peak at frequency ω_1 decreases in height and merges into the side peaks. However, off resonance one of the side peaks rapidly disappears, and only the one closer to the atomic resonance frequency ω_0 survives, so that the spectrum quickly becomes markedly asymmetric.

Figure 5 illustrates the effect of increasing de-

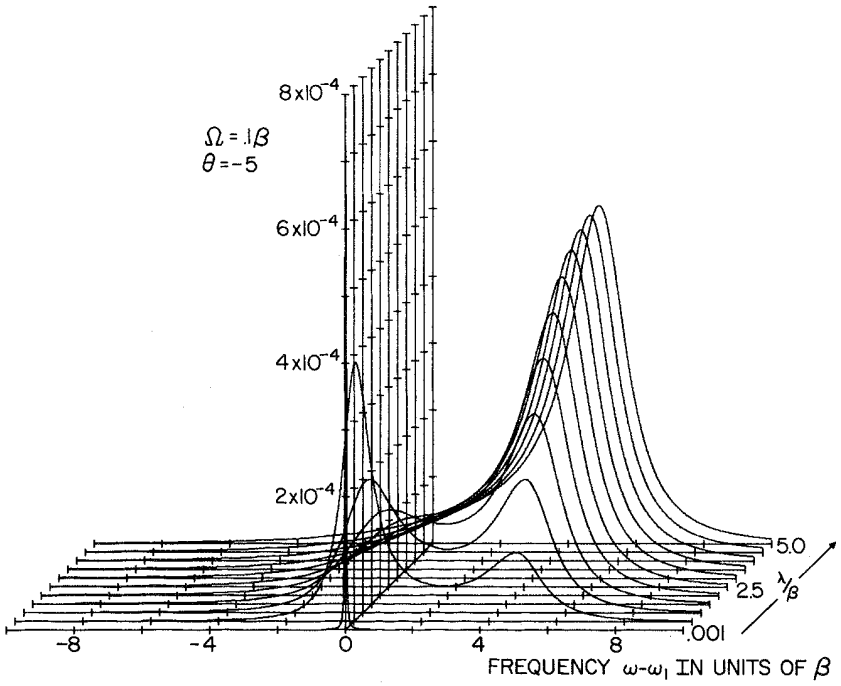

FIG. 2. Spectrum $\Phi(\omega)(4\pi\epsilon_0 c^2 r)^2/(\omega_0^2\mu\sin\psi)^2$ of the fluorescence radiated by a driven atom in the steady state, for various values of the excitation bandwidth λ, with Rabi frequency $\Omega=0.1\beta$, and detuning $\theta=-5$.

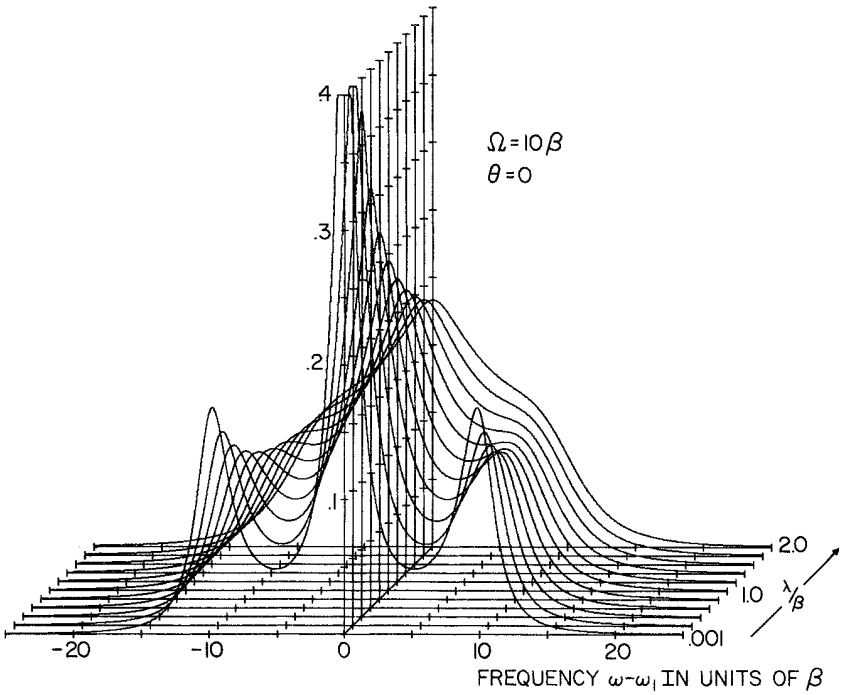

FIG. 3. Spectrum $\Phi(\omega)(4\pi\epsilon_0 c^2 r)^2/(\omega_0^2\mu\sin\psi)^2$ of the fluorescence radiated by a driven atom in the steady state, for various values of the excitation bandwidth λ, with Rabi frequency $\Omega = 10\beta$, and no detuning ($\theta = 0$).

tuning θ on the spectrum, for a fixed, but modest, excitation bandwidth $\lambda = 0.5\beta$. The asymmetry increases with θ, and of the two side peaks produced by Stark splitting, only the one closest to the atomic frequency survives for large detuning. The peak centered at the driving frequency ω_1 shows an interesting behavior, in that it reaches a maximum height for a certain detuning θ, and rapidly declines thereafter.

Somewhat similar effects appear to an exaggerated extent in Fig. 6, in which we have taken the exciting field to be very strong ($\Omega = 100\beta$), with a

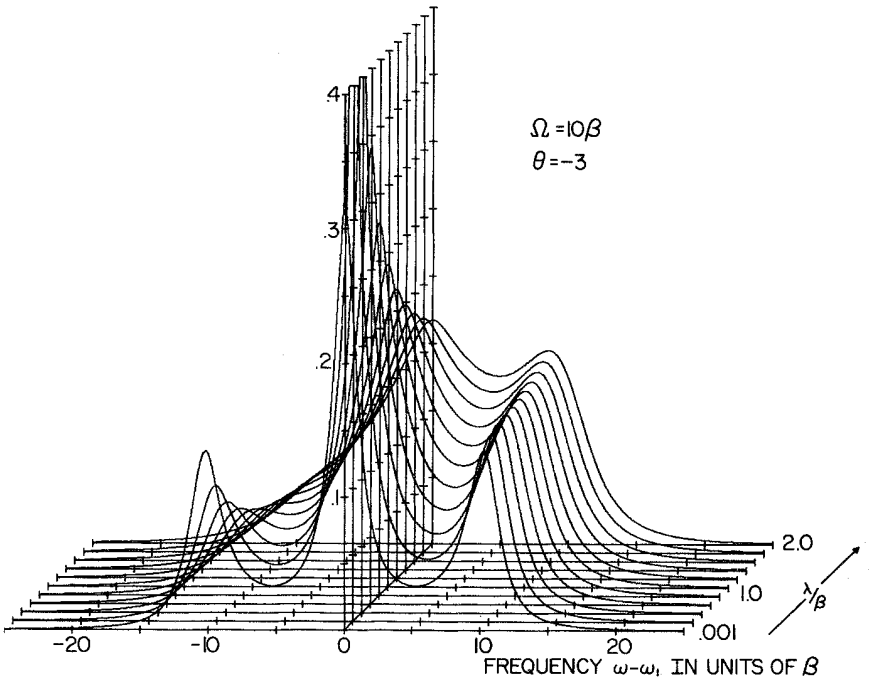

FIG. 4. Spectrum $\Phi(\omega)(4\pi\epsilon_0 c^2 r)^2/(\omega_0^2\mu\sin\psi)^2$ of the fluorescence radiated by a driven atom in the steady state, for various values of the excitation bandwidth λ, with Rabi frequency $\Omega = 10\beta$, and detuning $\theta = -3$.

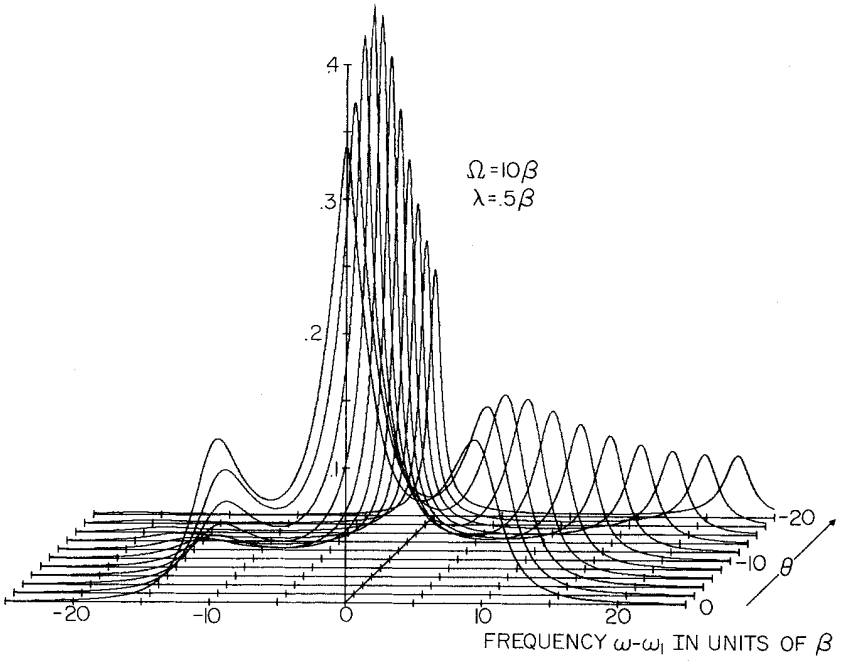

FIG. 5. Spectrum $\Phi(\omega)(4\pi\epsilon_0 c^2 r)^2/(\omega_0^2\mu\sin\psi)^2$ of the fluorescence radiated by a driven atom in the steady state, for various degrees of detuning θ, with Rabi frequency $\Omega = 10\beta$, and excitation bandwidth $\lambda = 0.5\beta$.

fixed excitation bandwidth $\lambda = 10\beta$, and have computed the spectrum up to a detuning of 200β. Under these circumstances not only one of the two side peaks attributable to Stark splitting, but also the peak centered at the driving frequency ω_1, becomes negligible for large detuning, and the peak centered at the atomic frequency clearly dominates. Despite the strength of the excitation field ($\Omega = 100\beta$), for large detuning the ac Stark effect tends to become a small effect with finite bandwidth excitation.

V. SUMMARY

We have shown that the effect of nonmonochromatic excitation in resonance fluorescence is to modify the time development of the fluorescent

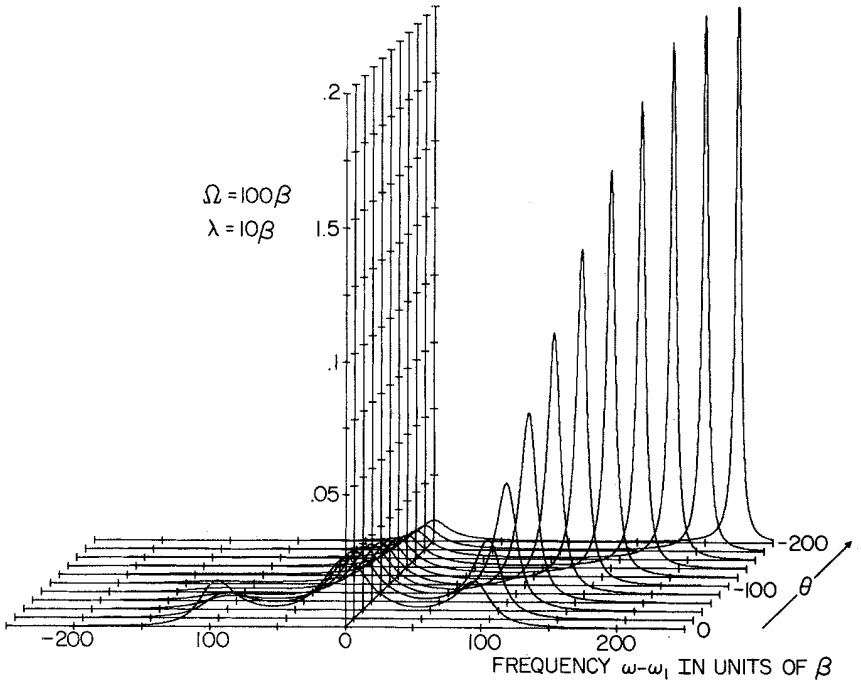

FIG. 6. Spectrum $\Phi(\omega)(4\pi\epsilon_0 c^2 r)^2/(\omega_0^2\mu\sin\psi)^2$ of the fluorescence radiated by a driven atom in the steady state, for various degrees of detuning θ, with Rabi frequency $\Omega = 100\beta$, and excitation bandwidth $\lambda = 10\beta$.

light intensity, its intensity correlation function and its spectral density. But whereas the effect is relatively small in the first two cases, it can become substantial and quite dramatic in the last case. This conclusion is not altogether surprising; the phase fluctuations of the driving field are naturally reflected more in the complex amplitude of the fluorescent field, and therefore in the radiated spectral density, than in its intensity. In particular, with off-resonance excitation the fluorescence spectrum becomes asymmetric, and reflects constructive and destructive interference effects on the two sides of the excitation frequency. These asymmetries are absent under monochromatic excitation, and represent a newly encountered feature of resonance fluorescence. It is possible that they played a role in some of the recently reported observations.[4]

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