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Atomic fluorescence in a mode-hopping laser field

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Abstract. In this paper we apply the recently developed theory of multiplicative stochastic correlation functions with Markovian random jumps to the problem of resonance fluorescence from a two-level atom in a strong laser field. The finite bandwidth of a single-mode laser or the appearance of distinct lines in a multimode laser is considered to be brought about by a random mode-switching process. The two-time dipole correlation functions, which determine the spectral distribution of the fluorescence and the temporal correlations between the emitted photons, are averaged exactly over the stochastics of the driving field. Our explicit expressions only involve single-time averages, which can be easily evaluated for a given probability distribution. These results generalise and modify earlier results in that we obtain closed expressions, rather than implicit differential equations, we can handle the continuous case as well, and we take into account the initial correlations, which always arise when the laser coherence time is finite. We illustrate the dependence on the stochastics of the laser model by comparing the multimode description with the phase-diffusion process and the Lorentz wave, which all give rise to the same laser profile, but to a different fluorescence spectrum.

1. Introduction

The theory of resonance fluorescence from a two-level atom in a strong monochromatic laser field is well established (Mollow 1969, Carmichael and Walls 1976, Kimble and Mandel 1976). Both the frequency distribution and the temporal correlations of the emitted photons were obtained, and even the complete photon statistics was found by Lenstra (1982). Many authors stressed, however, that a reliable comparison with experimental data can only be achieved if the finite linewidth of the laser is taken into account. Phenomena like spectral asymmetry or the appearance of a fluorescence line near the atomic resonance by excitation in the far wing, even in the low-intensity limit, can only be understood from a finite laser linewidth. Therefore, a proper comprehension of atomic fluorescence requires that laser linewidth effects are taken into consideration. The first attempts have been made by Kimble and Mandel (1977), Avan and Cohen-Tannoudji (1977), Agarwal (1978) and Zoller (1978), who represented the laser field by a plane wave with a stochastically fluctuating diffusive phase, which was taken as the Gaussian Wiener-Lévy process. An extension to a non-Gaussian diffusive phase was given by Arnoldus and Nienhuis (1983). The diffusion model can be solved exactly, but it cannot be expected to provide very realistic answers. This is due to the inherent assumption of zero laser coherence time. The treatment of more sophisticated models for a single-mode laser is, however, hampered by the cumbersome stochastics of the field. This also obstructed the development of a reasonable multimode theory.

A finite coherence time of the single-mode field was included by Dixit *et al* (1980), Zoller *et al* (1981), Yeh and Eberly (1981), Jackson and Swain (1982) and Swain (1984). They generalised the Gaussian diffusion model, but at the expense of a factorisation assumption. This was shown to not be exact (Arnoldus and Nienhuis (1986a), since a stochastic initial state cannot in general be replaced by its average. Zoller (1979a, b) and George and Lambropoulos (1979) employed a stochastic representation of a multimode field, and Wódkiewicz *et al* (1984) introduced an alternative model for a two-mode laser. This idea was extended by Deng and Eberly (1984) and Shore (1984) to an N-mode laser field. Apart from the factorisations, which can only be assumed to be reasonable if the coherence time is not too large (van Kampen 1976), the solvability of the models depends strongly on the specific choice of the laser field stochastics. Therefore these models describe only a restricted class of laser profiles.

In this paper we also start from a multimode description of the laser field. The mode distribution can be taken to be continuous or discrete, so that the model can represent a single-mode or a multimode laser. With a proper choice of the mode distribution, we can obtain any laser profile, and the coherence time can be chosen arbitrarily. The stochastics is dealt with exactly, without a decorrelation approximation and in a uniform way. Hence our treatment generalises and improves earlier results in a compact formalism. Furthermore, it will be shown that a Gaussian probability distribution yields field stochastics which greatly resembles the commonly applied Ornstein-Uhlenbeck process.

2. The laser field

The electric field of an intensity-stabilised laser at a given point in space can be represented by

$$\boldsymbol{E}(t)=E_0 \operatorname{Re} \boldsymbol{\varepsilon}_{\mathrm{L}} \exp[-\mathrm{i}(\omega_{\mathrm{L}}t+\phi(t))] \tag{2.1}$$

with ω_{L} the central laser frequency and $\phi(t)$ a real-valued stochastic process. The laser profile, assumed to be stationary, is then given by

$$I_{\mathrm{L}}(\omega)=\frac{1}{\pi}\operatorname{Re}\int_0^\infty \exp(i\Delta\tau)\langle\exp[-\mathrm{i}(\phi(t+\tau)-\phi(t))]\rangle\,\mathrm{d}\tau \tag{2.2}$$

with $\Delta=\omega-\omega_{\mathrm{L}}$. The angular brackets denote an average over the stochastic laser phase. For a fixed value of the phase, e.g. $\phi(t)=\phi_0$, this profile reduces to $I_{\mathrm{L}}(\omega)=\delta(\omega-\omega_{\mathrm{L}})$, and hence a finite laser linewidth is due to the fluctuations in the phase $\phi(t)$. The profile (2.2) is normalised according to

$$\int_{-\infty}^{\infty} I_{\mathrm{L}}(\omega)\,\mathrm{d}\omega=1 \tag{2.3}$$

independent of the stochastics of $\phi(t)$. This shows that the process $\phi(t)$ distributes the power over a finite frequency range around ω_{L}, without affecting the overall strength.

The laser profile is determined by the stochastics of $\phi(t)$. A common choice is the independent-increment process, which is a diffusive Markov process (Doob 1953).

The Gaussian limit of this non-stationary process $\phi(t)$ is the Wiener–Lévy process. The laser profile for a diffusive phase is

$$I_{\mathrm{L}}(\omega)=\frac{1}{\pi}\operatorname{Re}\frac{1}{\lambda-\mathrm{i}\Lambda} \tag{2.4}$$

which is a Lorentzian with a width (HWHM) equal to λ. $\lambda>0$ is an independent parameter of the process. An alternative choice for $\phi(t)$ is a random-jump process, which is known as the Lorentz wave (Kubo 1954, Anderson 1954). This stationary process, which was introduced in quantum optics by Burshtein (1965, 1966), again yields the laser profile (2.4), where λ has now the significance of the phase jump rate.

A fluctuating phase $\phi(t)$ gives essentially a single-mode model, and produces a laser profile that usually approaches a Lorentzian. This is not necessarily a good approximation of a laser field, especially when one describes effects arising from the overlap of the line wing with a resonance. As an example we mention the asymmetry between the three-photon line and the fluorescence line (Arnoldus and Nienhuis 1983).

3. Multimode description

If we introduce the stochastic process $x(t)$ as

$$x(t)=\frac{\mathrm{d}}{\mathrm{d}t}\phi(t) \tag{3.1}$$

then we can write for the electric field

$$\boldsymbol{E}(t)=E_0\operatorname{Re}\boldsymbol{\varepsilon}_{\mathrm{L}}\exp\left(-\mathrm{i}\int_0^t(\omega_{\mathrm{L}}+x(s))\,\mathrm{d}s\right) \tag{3.2}$$

and the spectral distribution can be expressed in $x(t)$ as

$$I_{\mathrm{L}}(\omega)=\frac{1}{\pi}\operatorname{Re}\int_0^\infty\exp(\mathrm{i}\Lambda\tau)\left\langle\exp\left(-\mathrm{i}\int_\rho^\tau x(s)\,\mathrm{d}s\right)\right\rangle\mathrm{d}\tau. \tag{3.3}$$

Instead of prescribing the stochastics of $\phi(t)$, we can equivalently give the stochastics of $x(t)$. However, since $\omega_{\mathrm{L}}+x(t)$ can be looked upon as the instantaneous laser frequency, according to (3.2), this description is a convenient basis for a multimode model. Hence we effectively assume that the laser switches between many modes, and that at any instant only a single mode is occupied. This frequency-hopping model should be distinguished from a chaotic description where different modes are occupied simultaneously. For switching rates that are high compared with radiative transition rates, the results should be similar. Notice that $x(t)$ must be stationary in order that the spectrum be stationary. We recall that for the Wiener–Lévy process $\phi(t)$, as mentioned above, the time derivative $x(t)$ is Gaussian white noise (Fox 1972).

The range of occurring x values can be considered as the occupied laser modes. In this fashion a distribution over a discrete set of x values can be regarded as a genuine multimode model, whereas a continuous distribution around $x=0$ represents a single-mode profile. This notion can be made quite transparent as follows. Let us denote by $P(x)$ the probability distribution of $x(t)$. In the static limit of the process $x(t)$, the value of $x(t)$ is independent of t for each realisation of the process. Then stochastic quantities can only depend parametrically on x, and averaging reduces to trivial weighing with the probability $P(x)$. For instance the laser profile (3.3) becomes

$$I_L(\omega)=\frac{1}{\pi}\operatorname{Re}\int_0^\infty \exp(\mathrm{i}\Lambda\tau)\left(\int P(x)\exp(-\mathrm{i}x\tau)\,\mathrm{d}x\right)\mathrm{d}\tau=P(\Lambda) \tag{3.4}$$

and the spectral distribution around ω_L is identical to the distribution $P(x)$ of x over the laser modes ω_L+x. Since $P(x)$ is arbitrary, we can model any profile $I_L(\omega)$.

During a laser run the instantaneous frequency $\omega_L+x(t)$ will switch between the possible modes. This turns $x(t)$ into a non-trivial stochastic process. We will denote by γ the average mode-switching rate, so γ^{-1} equals the average dwell time in a single mode, which equals the laser coherence time. This frequency-hopping process will cause $I_L(\omega)$ to deviate from $P(\Lambda)$. For instance, if $P(x)$ contains a discrete mode x_0, this will give a delta peak in the static limit (3.4). However, if this mode is occupied for a time γ^{-1} on the average, the peak will have a width of the order of γ.

The process $x(t)$ will be taken as a stationary Markov process with zero average. The complete statistics of $x(t)$ can then be determined by the additional requirement that $x(t)$ is Gaussian, and according to the theorem of Doob this is the Ornstein-Uhlenbeck process (Wax 1954). Then $P(x)$ is the Gaussian

$$P(x)=\frac{1}{\sigma(2\pi)^{1/2}}\exp\left(-\frac{x^2}{2\sigma^2}\right)\qquad \sigma>0 \tag{3.5}$$

with the variance σ^2 arbitrary. The mode correlation becomes

$$\langle x(t_1)x(t_2)\rangle=\sigma^2\exp(-\gamma|t_1-t_2|) \tag{3.6}$$

and the higher-order correlation functions follow from the Gaussian property (van Kampen 1981). For this process the laser profile (3.3) is found to be (Dixit *et al* 1980)

$$I_L(\omega)=\frac{1}{\pi}\operatorname{Re}\frac{1}{\lambda-\mathrm{i}\Lambda}M\left(1,\frac{\lambda-\mathrm{i}\Lambda}{\gamma}+1,\lambda/\gamma\right) \tag{3.7}$$

with $\lambda=\sigma^2/\gamma$ and $M(a,b,z)$ the regular confluent hypergeometric function. We already found that for $\gamma^{-1}\to\infty$ and σ^2 fixed, this profile becomes $P(\Lambda)$. If we take the limit $\gamma^{-1}\to 0$, with λ finite, then equation (3.7) reduces to the Lorentzian (2.4) since $M(1,1,0)=1$. This is the white-noise limit, and $\phi(t)$ is identical to the Wiener-Lévy process. For a finite value of γ, $x(t)$ is the Ornstein-Uhlenbeck process, which gives (3.7) for the spectrum.

The requirement that $x(t)$ should be Gaussian in all higher-order statistics is rather artificial, and will not be satisfied in general. We therefore drop the Gaussian property. We will simply assume that the field can make random transitions between the modes at random instants. This determines the full statistics in terms of the probability distribution $P(x)$ and the coherence time γ^{-1}. The conditional probability distribution for $x=x_2$ at a time τ after an initial value $x=x_1$ is then given by

$$P_\tau(x_2|x_1)=\delta(x_2-x_1)\exp(-\gamma\tau)+P(x_2)[1-\exp(-\gamma\tau)]\qquad \tau\geqslant 0. \tag{3.8}$$

This process is called the Kubo-Anderson process (Kubo 1954, Anderson 1954), or the random-jump process. It is easy to check that the mode correlation is again given by (3.6). Furthermore it can be shown that in the limit $\langle x^2\rangle\to\infty$, $\gamma\to\infty$, $\lambda=\langle x^2\rangle/\gamma$ fixed, this random-jump process reduces to Gaussian white noise for every $P(x)$ (Arnoldus and Nienhuis 1985). In this sense the phase-diffusion model is a special case of the random-jump process.

If we take $P(x)$ as the Gaussian (3.5), then we have a model which resembles the Ornstein-Uhlenbeck process. The stochastics in the static limit $\gamma \to 0$ is again completely determined by the Gaussian $P(x)$. For very fast but finite fluctuations ($\gamma \to \infty$, σ^2 fixed), the atomic response is determined by $\langle x(t) \rangle = 0$, and the laser profile becomes $I_L(\omega) = \delta(\Lambda)$. Finally the limit $\gamma \to \infty$, $\sigma^2 \to \infty$, $\lambda = \sigma^2/\gamma$ fixed, again produces Gaussian white noise. In these three limits the random-jump process with a Gaussian $P(x)$ is identical to the Ornstein-Uhlenbeck process. Only for a finite γ will the higher-order statistics deviate slightly from their Gaussian limit. The random-jump process is, however, much more general, since $P(x)$ is arbitrary. A relationship between the Ornstein-Uhlenbeck process and a different jump process was established by Wódkiewicz (1981).

4. The laser spectrum

The stochastic laser phase turns the equations for the state of the atom and the fluorescence signal into multiplicative stochastic differential equations. With the assumption of a given initial state, these equations have been averaged over the random-jump process by Brissaud and Frisch (1971, 1974) and Shapiro and Loginov (1978). The quantum correlation functions, which depend stochastically on two time arguments, have been obtained by Arnoldus and Nienhuis (1986a). The simplest example of a multiplicative stochastic process is the laser field correlation, which determines the laser profile. The stochastic function

$$g(\tau) = \exp\left(-\mathrm{i} \int_0^\tau x(s)\, \mathrm{d}s\right) \tag{4.1}$$

obeys the equation

$$\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}\tau} g(\tau) = x(\tau) g(\tau) \qquad g(0) = 1. \tag{4.2}$$

According to (3.3) the Laplace transform of $g(\tau)$ determines $I_L(\omega)$. Applying the results of our previous paper (Arnoldus and Nienhuis 1986a), we obtain directly the spectrum

$$I_L(\omega) = \frac{1}{\pi} \mathrm{Re}\left(1 - \gamma \int \mathrm{d}x P(x) \frac{\mathrm{i}}{\Lambda - x + \mathrm{i}\gamma}\right)^{-1} \int \mathrm{d}x\, P(x) \frac{\mathrm{i}}{\Lambda - x + \mathrm{i}\gamma} \tag{4.3}$$

in terms of $P(x)$ and γ.

As an example we consider a two-mode laser with frequencies $\omega_L \pm a$ which are occupied with equal probability. Then $P(x)$ should be taken as

$$P(x) = \tfrac{1}{2}[\delta(x+a) + \delta(x-a)] \tag{4.4}$$

and this process $x(t)$ is known as the random telegraph. Substitution of (4.4) into (4.3) yields

$$I_L(\omega) = \frac{T}{\pi} \frac{(aT)^2}{(\Lambda T)^4 + [1 - 2(aT)^2](\Lambda T)^2 + (aT)^4} \tag{4.5}$$

with $T = \gamma^{-1}$. This result was also found by Wódkiewicz *et al* (1984). If $2^{1/2}a > \gamma$ the spectrum has two maxima at

$$\omega = \omega_L \pm a\left(1 - \frac{\gamma^2}{2a^2}\right)^{1/2} \tag{4.6}$$

and a minimum at $\omega=\omega_L$. For $2^{1/2}a<\gamma$ the two lines overlap and we find a single line at $\omega=\omega_L$. The widths of the lines are of the order of γ, and for $\gamma^{-1}\to\infty$ the spectrum (4.5) reduces to two δ peaks at $\omega=\omega_L\pm a$. We notice that the wings of the lines vanish as Λ^{-4} for $\Lambda\to\pm\infty$, whereas for a Lorentzian we have a Λ^{-2} dependence. If we take the limit $\gamma\to\infty$, $a^2\to\infty$ with $\lambda=a^2/\gamma$ finite, then (4.5) reduces to the Lorentzian (2.4). An example of the spectrum (4.5) is plotted in figure 1.

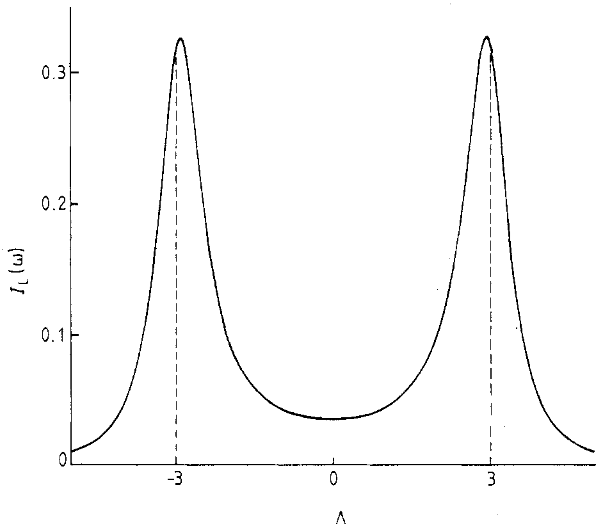

Figure 1. Plot of the laser spectrum $I_L(\omega)$ from equation (4.3) for the case $P(x)=\frac{1}{2}[\delta(x+a)+\delta(x-a)]$ with $a=3$ and $\gamma=1$. The broken lines indicate the two distinct laser modes at $\omega=\omega_L\pm a$. The widths of the lines are of the order of γ, as a result of the finite dwell time γ^{-1} in each mode.

We can also take $P(x)$ to be a continuous distribution. Consider for example a Lorentzian

$$P(x)=\frac{1}{\pi}\,\mathrm{Re}\,\frac{1}{\lambda-\mathrm{i}x}\qquad \lambda>0. \tag{4.7}$$

Then we find

$$\int \mathrm{d}x\,P(x)\frac{\mathrm{i}}{\Lambda-x+\mathrm{i}\gamma}=\frac{1}{\lambda+\gamma-\mathrm{i}\Lambda} \tag{4.8}$$

and $I_L(\omega)$ again becomes the Lorentzian (2.4) for every γ. In this particular situation the finite dwell time γ^{-1} does not affect the laser profile, and we find $I_L(\omega)=P(\Lambda)$ for every γ^{-1}, rather than for $\gamma^{-1}=\infty$ only.

As a third case we take $P(x)$ as the Gaussian (3.5). As pointed out above, the laser profile is then intermediate between a Gaussian and a Lorentzian. If we take σ^2 fixed and let γ approach zero, then $I_L(\omega)$ reduces to $P(\Lambda)$. Conversely, in the limit $\gamma\to\infty$, $I_L(\omega)$ becomes the delta function $\delta(\Lambda)$. Thus for σ^2 constant, the laser profile varies between the Gaussian $P(\Lambda)$ and a delta function, if we vary the magnitude of γ. Alternatively we can keep $\lambda=\sigma^2/\gamma$ constant, while varying γ. In the limit $\gamma\to\infty$ process $x(t)$ becomes Gaussian white noise and $I_L(\omega)$ is the Lorentzian (2.4). For a small γ the laser profile approaches a Gaussian, but for λ fixed this implies σ^2 small.

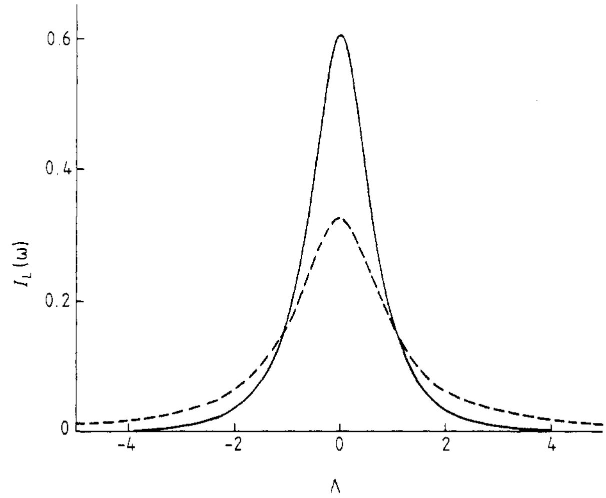

Figure 2. Plot of the laser spectrum $I_L(\omega)$ from equation (4.3) for the case when $P(x)$ is the Gaussian (3.5). In both curves the parameter $\lambda = \sigma^2/\gamma$ is fixed and equal to 1. The broken curve corresponds to $\gamma = 100$, which is nearly the Lorentzian limit. For the full curve we took $\gamma = 1$. The profile then tends to a Gaussian, which can be seen from the disappearance of the line wings. The intensity of the wings is transferred to the line centre, because the profile is normalised according to equation (2.3). In the limit $\gamma \to 0$ this spectrum becomes a delta function.

Hence in the limit $\gamma \to 0$ we again find $I_L(\omega) = \delta(\Lambda)$. This behaviour is illustrated in figure 2.

5. Atomic fluorescence

In this section we formulate the equation of motion for an atom in the stochastic laser field (2.1) and we give expressions for the spectral distribution of the emitted fluorescence and the temporal photon correlations. This general formulation is independent of the stochastics of $\phi(t)$.

We consider an atom with ground state $|g\rangle$, excited state $|e\rangle$ and level separation $\hbar\omega_0$ in the laser field and in a perturber bath. The equation of motion for the atomic density operator $\sigma(t)$ in the rotating frame is given by (Agarwal 1978)

$$\mathrm{i}\frac{\mathrm{d}\sigma}{\mathrm{d}t} = (L_d(\omega_L) + x(t)L_g - \mathrm{i}\Gamma - \mathrm{i}\Phi)\sigma. \tag{5.1}$$

In terms of the bare-state basis operators $P_e = |e\rangle\langle e|$, $P_g = |g\rangle\langle g|$, $d = |e\rangle\langle g|$ and $d^\dagger = |g\rangle\langle e|$ in the atomic Hilbert space, the atom-field Liouvillian $L_d(\omega_L)$ is defined as the commutator

$$L_d(\omega_L)\sigma = -\tfrac{1}{2}[(\omega_L - \omega_0)(P_e - P_g) + \Omega(d + d^\dagger), \sigma] \tag{5.2}$$

which contains the Rabi frequency $\Omega = E_0|\boldsymbol{\mu}_{eg} \cdot \boldsymbol{\varepsilon}_L|/\hbar$ of the dipole coupling. The stochastic part of the evolution is expressed in terms of

$$L_g\sigma = [P_g, \sigma]. \tag{5.3}$$

The operators

$$\Gamma\sigma = \tfrac{1}{2}A(P_e\sigma + \sigma P_e - 2d^\dagger \sigma d) \tag{5.4}$$

$$\Phi\sigma = (\gamma_c + \mathrm{i}\beta)P_e\sigma P_g + (\gamma_c - \mathrm{i}\beta)P_g\sigma P_e \tag{5.5}$$

describe spontaneous decay and collisional damping, with A the Einstein coefficient and γ_c and β the collisional width and shift respectively. Equation (5.1) is a multiplicative process and its stochastic solution can be written as

$$\sigma(t) = Y(t, t_0)\sigma(t_0) \qquad t \geqslant t_0 \tag{5.6}$$

which defines the evolution operator $Y(t, t_0)$. We will always assume that the atom has been in the field for a long time in comparison with A^{-1}, γ^{-1}, γ_c^{-1}, $|\Omega|^{-1}$. Then the stochastic average of (5.6) will have reached a steady state, which will be denoted by

$$\bar{\sigma} = \lim_{t\to\infty} \{\sigma(t)\}. \tag{5.7}$$

Notice, however, that $\sigma(t)$ itself will never reach a constant value, because of the persisting fluctuations in the driving field.

The spectral distribution of the emitted dipole radiation can be written as (Arnoldus and Nienhuis 1985)

$$I(\omega) = \frac{A}{\pi} \lim_{t\to\infty} \mathrm{Re} \int_0^\infty \exp(\mathrm{i}\omega\tau)\{\langle d(t)d^\dagger(t+\tau)\rangle\}\, \mathrm{d}\tau \tag{5.8}$$

with the intensity taken as the number of emitted photons per unit time. The dipole correlation $\langle d(t)d^\dagger(t+\tau)\rangle$ is a stochastic quantity of two times, and just as for $\sigma(t)$, the limit $t \to \infty$ only exists for the average. The number of emitted photons per second becomes

$$I = \int_{-\infty}^{\infty} I(\omega)\, \mathrm{d}\omega = A\langle e|\bar{\sigma}|e\rangle \tag{5.9}$$

which equals A times the population of the excited state, as usual. The time correlation between two emitted photons can be written as (George 1981)

$$I_2(\tau) = A^2 \lim_{t\to\infty} \{\langle d(t)d(t+\tau)d^\dagger(t+\tau)d^\dagger(t)\rangle\} \qquad \tau \geqslant 0. \tag{5.10}$$

$I_2(\tau)$ equals the twofold probability density for a photon emission at time 0, followed by an emission at time τ.

Notice that both I and I_2 are evaluated n the steady-state limit $t \to \infty$, whereas the interaction with the stochastic field has been turned on at some finite time. The density matrix at the initial time t is a stochastic quantity that is correlated with the atomic evolution between t and $t+\tau$. It is this correlation that has been neglected in previous work, as mentioned in § 1.

The stochastic dipole correlations can be transformed to the Schrödinger picture, which yields

$$\langle d(t)d^\dagger(t')\rangle = \exp[-\mathrm{i}\omega_L(t'-t)]\, \mathrm{Tr}\, d^\dagger D(t', t) \tag{5.11}$$

$$\langle d(t)d(t')d^\dagger(t')d^\dagger(t)\rangle = \mathrm{Tr}\, RC(t', t) \tag{5.12}$$

for $t' \geqslant t$. Here we introduced the Liouville operator R, defined as

$$R\sigma = d^\dagger \sigma d \tag{5.13}$$

and the stochastic vectors in Liouville space

$$D(t', t) = \exp[-\mathrm{i}(\phi(t') - \phi(t))] Y(t', t) S\sigma(t) \tag{5.14}$$

$$C(t', t) = Y(t', t) R\sigma(t). \tag{5.15}$$

The Liouville operator S is defined as

$$S\sigma = \sigma d. \tag{5.16}$$

The dipole correlations are now expressed entirely in the evolution operator $Y(t', t)$ for $\sigma(t)$ and in $\sigma(t)$ itself. Hence the equation of motion (5.1) also determines the dipole correlations.

With (5.1) we find that $D(t', t)$ and $C(t', t)$ obey the multiplicative equations

$$\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t'} C(t', t) = (L_d(\omega_{\mathrm{L}}) + x(t') L_g - \mathrm{i}\Gamma - \mathrm{i}\Phi) C(t', t) \tag{5.17}$$

$$\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t'} D(t', t) = [L_d(\omega_{\mathrm{L}}) + x(t')(L_g + 1) - \mathrm{i}\Gamma - \mathrm{i}\Phi] D(t', t) \tag{5.18}$$

and the initial conditions

$$C(t, t) = R\sigma(t) \tag{5.19}$$

$$D(t, t) = S\sigma(t). \tag{5.20}$$

We note that equation (5.17) is identical to the equation for $\sigma(t)$, but that (5.18) is different, due to the appearance of $L_g + 1$. We have to average the solutions of equations (5.17) and 5.18) over the stochastic process $x(t)$. We emphasise that the initial values (5.19) and (5.20) are also stochastic. Therefore the average of $C(t', t)$ is in general not simply the average of $Y(t', t)$, acting on the average of the initial condition, which is $R\bar{\sigma}$. This would be the commonly applied factorisation assumption, which cannot be justified if the coherence time γ^{-1} is not small in comparison with the other time scales in the problem.

6. The stochastic averages

Stochastic equations of the form (5.17), (5.18) with $x(t)$ the random-jump process, have been dealt with in a previous paper (Arnoldus and Nienhuis 1986a). We can therefore skip the mathematics and turn directly to the results. In general the distinct equations (5.17) and (5.18) require distinct resolvents for the averages. In our specific case, however, we can express all solutions in only one resolvent. This is due to the fact that $x(t)$ shifts the laser frequency ω_{L} instantaneously, and that the resolvents depend only parametrically on x. It is therefore sufficient to introduce the inverse operator

$$U(\omega_1, \omega_2) = \frac{\mathrm{i}}{\omega_1 + \mathrm{i}\gamma - L_d(\omega_{\mathrm{L}} + \omega_2) + \mathrm{i}\Gamma + \mathrm{i}\Phi} \tag{6.1}$$

where ω_1 is a Laplace parameter and ω_2 acts as a shift of ω_{L}.

The steady-state density operator $\bar{\sigma}$ is the unique solution of

$$\left(1 - \gamma \int \mathrm{d}x\, P(x) U(0, x)\right)\bar{\sigma} = 0 \qquad \bar{\sigma}^{\dagger} = \bar{\sigma} \qquad \mathrm{Tr}\,\bar{\sigma} = 1. \tag{6.2}$$

The Liouville operator $1-\gamma\int \mathrm{d}x\, P(x)U(0,x)$ is a simple 4×4 matrix with respect to the basis vectors P_e, P_g, d and $d^\dagger$, and can be evaluated immediately for any prescribed $P(x)$. Then $\bar{\sigma}$ is the eigenvector with eigenvalue zero.

The averages of equations (5.17) and (5.18) can also be found. If we substitute the results in (5.11), (5.12), and subsequently in (5.8), (5.10), we obtain the fluorescence spectrum and the (Laplace-transformed) two-photon correlation. The results involve the solution for σ, and are given explicitly by

$$I(\omega)=\frac{A}{\pi}\operatorname{Re}\operatorname{Tr} d^\dagger\gamma\left(1-\gamma\int \mathrm{d}x\, P(x)U(\Lambda-x,x)\right)^{-1} \times\int \mathrm{d}x\, P(x)U(\Lambda-x,x)SU(0,x)\bar{\sigma} \qquad \Lambda=\omega-\omega_{\mathrm{L}} \tag{6.3}$$

$$\tilde{I}_2(\omega)=\int_0^\infty \exp(\mathrm{i}\omega\tau)I_2(\tau)\,\mathrm{d}\tau = A^2\operatorname{Tr} R\gamma\left(1-\gamma\int \mathrm{d}xP(x)U(\omega,x)\right)^{-1}\int \mathrm{d}x\, P(x)U(\omega,x)RU(0,x)\bar{\sigma}. \tag{6.4}$$

The integrals over x, the matrix products and the matrix inversions are analytically very cumbersome. A numerical evaluation of (6.3) and (6.4) is, however, straightforward.

7. The fluorescence spectrum

The most simple example of a multimode laser with discrete modes is a two-mode laser with modes at $\omega=\omega_{\mathrm{L}}\pm a$ and $P(x)$ from (4.4). The laser profile is then given by equation (4.5) and plotted in figure 1. In figure 3 we plot the fluorescence spectrum $I(\omega)$ for this case, as determined by equation (6.3). The two peaks at $\Lambda/A=\pm 3=\pm a/A$ correspond to elastically scattered photons from the laser modes $\omega_{\mathrm{L}}\pm a$. In this example

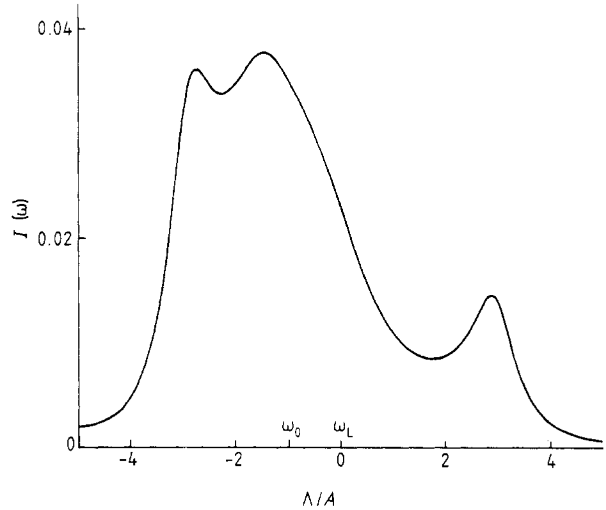

Figure 3. Plot of the fluorescence spectrum $I(\omega)$ from equation (6.3) for the case of a symmetric two-mode excitation. The laser modes are situated at $\omega=\omega_{\mathrm{L}}\pm 3A$ and the inverse coherence time is taken as $\gamma=A$. The atomic resonance equals $\omega_0=\omega_{\mathrm{L}}-A$ and the Rabi frequency is $\Omega=2A$. We neglected collisions.

we took the atomic resonance ω_0 equal to $\omega_L - A$, which corresponds to $\Lambda/A = -1$ in the picture. Therefore the laser mode $\omega_L - a$ is closer to resonance than the mode $\omega_L + a$. This explains why the left peak in the spectrum is stronger than the right peak. The maximum in the middle is situated at ω_0. This is the phase-fluctuations-induced fluorescence, which may be viewed as resulting from the overlap of the laser line wings with the resonance ω_0. Figure 3 illustrates the significant features of the fluorescence spectrum, as they arise in excitation of the atom with a discrete multimode laser.

In the low-intensity limit $\Omega \to 0$ the fluorescence spectrum is completely determined by the laser profile. In this limit every laser photon is scattered independently of other photons, since each photon sees the atom in its ground state. The probability for the appearance of a photon with frequency ω equals $I_L(\omega)$, and the fluorescence spectrum becomes a convolution of the laser profile and $I(\omega)$ evaluated for $P(x) = \delta(x)$. This does not hold anymore for high intensities, as can now easily be shown. We already found that a Lorentzian $P(x)$ gives $I_L(\omega) = P(\Lambda)$, independent of the laser coherence time γ^{-1}. In figure 4 we plotted the fluorescence spectrum $I(\omega)$ for two values of γ, and it appears that there is a distinction. This illustrates explicitly that the fluorescence spectrum is no longer determined by the laser profile alone for high intensities.

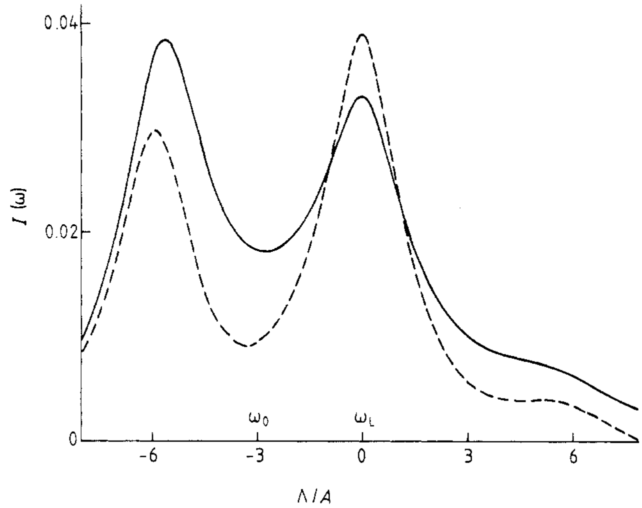

Figure 4. The fluorescence spectrum for $\Omega = 5A$, $\omega_L - \omega_0 = 3A$, $\gamma_c = \frac{1}{2}A$ and $\beta = 0$. The mode distribution $P(x)$ is taken as a Lorentzian with $\lambda = A$ the half width at half maximum. This picture illustrates the dependence of $I(\omega)$ on the laser coherence time γ^{-1}. The full curve corresponds to $\gamma = 5A$ and the broken curve to $\gamma = 0.1A$.

The line at the optical frequency ω_L in figure 4 is termed the Rayleigh line, and it corresponds to elastically scattered laser photons. The line at the left-hand side, near the atomic resonance ω_0, is the fluorescence line. Its shift from ω_0 reflects the dynamical Stark effect, which is a result of the high laser power (Cohen-Tannoudji 1977). We notice that increasing γ enhances the strength of the fluorescence line and diminishes the strength of the Rayleigh line. The limit $\gamma \to \infty$, which is the Gaussian white-noise limit, is given by the broken curve in figure 5. It can be shown in general that the line strengths are independent of γ, provided that γ is much smaller than the line separation (Arnoldus and Nienhuis 1985). If γ becomes comparable with the line distance or

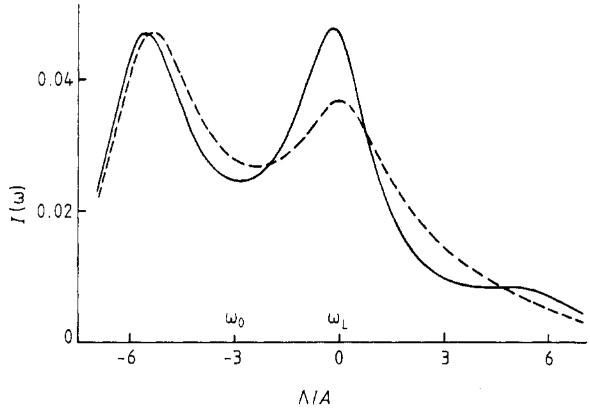

Figure 5. The fluorescence spectrum $I(\omega)$ with the same atomic parameters as in figure 4. The broken curve is the limit $\gamma \to \infty$, which is the phase-diffusion limit. The full curve represents $I(\omega)$, when the laser field is taken as the Lorentz wave.

larger, then the strength of the fluorescence line will always be enhanced by an increasing γ. This general idea is now explicitly verified.

We will now show that a given laser profile and coherence time do not fully determine the fluorescence spectrum. The broken curve in figure 5 gives $I(\omega)$ in the white-noise limit, where the laser spectrum is a Lorentzian and the coherence time is zero. In a previous paper (Arnoldus and Nienhuis 1986b) we employed a different stochastic model for the laser phase. The phase $\phi(t)$ was assumed to perform random jumps at random instants. This laser field, which is known as the Lorentz wave, also has a Lorentzian spectral distribution and a zero coherence time. The full curve in figure 5 gives the fluorescence spectrum for excitation with the Lorentz wave. The total fluorescence (the integrated spectrum) is the same for both cases, but the spectral distribution is still slightly different.

8. Conclusions

The finite bandwidth of a single-mode laser and the presence of discrete modes in a multimode laser are described by a stationary mode distribution $P(x)$. The finite dwell time γ^{-1} of the field in a single mode gives rise to a broadening of a discrete mode of a multimode field and affects the spectral distribution of a single-mode field. The stochastics of the instantaneous laser frequency $\omega_L + x(t)$ is taken as a random-jump process. This is the commonly employed model for a multimode field with distinct modes, and if we take $P(x)$ as a Gaussian then we recover the general model of a single-mode laser. Hence our treatment unifies various models for single-mode and multimode fields. At the same time it is more general since we can prescribe $P(x)$ arbitrarily.

The equation of motion for the atom and the equations for the regressions of correlation functions of the emitted fluorescence attain the form of stochastic differential equations, which we average exactly, including the effect of the initial correlations. This improves earlier treatments of specific cases, where $P(x)$ was taken as the random-telegraph distribution or as a Gaussian, because we do not impose a factorisation assumption for two-time averages. The theory is exemplified with plots of the fluorescence spectrum for various cases, and the significance of a finite coherence time γ^{-1} is discussed.

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