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Effects of an arbitrary laser lineshape on fluorescence radiation, redistributed by collisions

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Abstract. We investigate collisional redistribution in a laser field with arbitrary lineshape. It is shown that the phase diffusion model cannot be correct in general due to its inherent assumption of zero correlation times. We give general expressions for the state of the atom, the dipole correlation function and the steady-state fluorescence spectrum in the presence of collisions and phase fluctuations with finite correlation times. It turns out that collisional effects decouple from the free-atom stochastic problem to a good approximation and that modifications due to the laser bandwidth can be quite different from collisional effects, although they are additive on a microscopic level. We evaluate the fluorescence spectrum explicitly in the limit where the three lines are well separated and we show that the spectrum is determined by simple field correlation functions.

The arbitrary phase fluctuations do not affect the line strengths but only the lineshapes. Collisions, on the other hand, can transfer intensity between the lines, due to the very small collision time. For free atoms and strong driving fields, the fluorescence spectrum becomes a convolution of the normalised laser spectrum with the fluorescence spectrum at monochromatic irradiation, but this does not hold anymore when collisions are present.

1. Introduction

The extensive use of intense laser beams in optical experiments made it necessary to develop the theory of the interaction of single atoms with strong radiation fields outside the perturbation expansion regime and to study the emitted fluorescence radiation in a similar fashion. For a two-level system, irradiated by monochromatic light, Mollow (1969), Carmichael and Walls (1976) and Kimble and Mandel (1976) calculated the state of the atom, the fluorescence intensity, the intensity correlation functions and the spectral distribution of the fluorescence. Hereafter, effort has been made to include the effects of the finite laser linewidth and most commonly this was done with the assumption that the phase of the field fluctuates randomly. If this stochastic phase is taken to be the Wiener-Lévy process (phase diffusion model) the laser line becomes a Lorentzian and the problem can be solved exactly (Kimble and Mandel 1977, Agarwal 1978, Zoller 1978), with a method due to Fox (1972). Although this phase diffusion model is attractive from a theoretical point of view, it turned out that it did not greatly improve the description of experimental results (Armstrong and Eberly 1979, Morellec *et al* 1980). More advanced models for a single-mode laser, with a non-Lorentzian lineshape, were introduced and the theoretical predictions fitted the experimental data better (Dixit *et al* 1980, Georges and Dixit 1981, Zoller *et al* 1981, Yeh and Eberly 1981, Jackson and Swain 1982).

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Another active field of research is the study of collisional redistribution of intense radiation. The characteristics of the fluorescence spectrum of an atom in a buffer gas, driven by an intense monochromatic field, contains information on the collisions in strong fields (Burnett *et al* 1982, Nienhuis 1982). Since realistic laser fields have a finite bandwidth, it is important to study the combined effect of collisions and laser fluctuations on the emitted spectrum. Modifications for illumination with finite bandwidth radiation were made recently by Eberly (1983) for the phase diffusion model and in the impact limit, and by Arnoldus and Nienhuis (1983) with the independent increment process and in the binary collision approximation. In both references, the laser has again a Lorentz profile. It is the aim of this paper to show how more realistic non-Lorentzian laser models can be incorporated in the theory of collisional redistribution and, conversely, how collisions change the free-atom results.

2. Spectral distribution of the laser intensity

The laser field, which is incident upon an active atom, is assumed to be non-monochromatic due to a fluctuating phase $\phi(t)$, considered as a real stochastic process. The electric field at the position of the atom is given by

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

with E_0 the amplitude, $\boldsymbol{\varepsilon}_\mathrm{L}$ the normalised polarisation vector and ω_L the central frequency. The spectrally resolved energy density of the corresponding electromagnetic field is the Fourier transform of the autocorrelation function of the positive frequency part of $\boldsymbol{E}(t)$, so the laser spectrum is proportional to

$$I_L(\omega) = \tfrac{1}{2}\varepsilon_0 E_0^2 \,\mathrm{Re}\, \frac{1}{\pi} \int_0^\infty \exp[\mathrm{i}(\omega - \omega_\mathrm{L})\tau] \{\exp[-\mathrm{i}(\phi(\tau) - \phi(0))]\} \,\mathrm{d}\tau \tag{2.2}$$

provided that the resulting bandwidth is much smaller than ω_L. We denote a stochastic average over the phase fluctuations by $\{\ldots\}$ and assume stationarity in occurring expectation values. Note that for instance the Wiener-Lévy process itself is not stationary.

The total intensity

$$\int_{-\infty}^{\infty} I_\mathrm{L}(\omega) \,\mathrm{d}\omega = \tfrac{1}{2}\varepsilon_0 E_0^2 \tag{2.3}$$

is seen to be independent of phase fluctuations. The stochastic process $\phi(t)$ distributes the intensity over a frequency range around ω_L, but does not change the overall intensity. It is convenient to introduce the normalised laser profile as

$$\tilde{I}_\mathrm{L}(\omega) = I_\mathrm{L}(\omega) / \tfrac{1}{2}\varepsilon_0 E_0^2. \tag{2.4}$$

Now we can invert relation (2.2) to find the correlation function from this normalised spectrum

$$\{\exp[-\mathrm{i}(\phi(\tau) - \phi(0))]\} = \int_{-\infty}^{\infty} \exp[-\mathrm{i}(\omega - \omega_\mathrm{L})\tau] \tilde{I}_\mathrm{L}(\omega) \,\mathrm{d}\omega \qquad \tau \geqslant 0. \tag{2.5}$$

Since the $\dot{\phi}(t)$ process can be assumed to be symmetric in time, this correlation function is real. This again implies that the phase fluctuations only broaden the laser line,

without shifting it and that the spectrum is symmetric around ω_L. In the special case of the phase diffusion model, the correlation function decays exponentially, so the laser profile

$$\tilde{I}_L(\omega) = \mathrm{Re}\frac{1}{\pi}\frac{1}{\lambda - i(\omega - \omega_L)} \tag{2.6}$$

is a Lorentzian with halfwidth at half maximum equal to λ.

3. The equation of motion

The laser field is incident upon a two-level system with internal Hamiltonian H_a, which has eigenstates $|e\rangle$ (excited state) and $|g\rangle$ (ground state) with eigenvalues $\hbar\omega_e$ and $\hbar\omega_g$ respectively. The coupling strength with the external field is determined by the real Rabi frequency Ω, defined by

$$\hbar\Omega = E_0 \boldsymbol{\varepsilon}_L \cdot \langle e|\boldsymbol{\mu}|g\rangle \tag{3.1}$$

with $\boldsymbol{\mu}$ the dipole moment operator. The coupling Hamiltonian in the dipole approximation and the rotating-wave approximation is then

$$H_{ar}(t) = -\tfrac{1}{2}\hbar\Omega\{d\ \exp[-i(\omega_L t + \phi(t))] + \mathrm{HC}\} \tag{3.2}$$

where $d = |e\rangle\langle g|$ is the atomic raising operator. Spontaneous emission into free space is accounted for by a relaxation operator Γ, which acts on a density operator ρ, according to

$$\Gamma\rho = \tfrac{1}{2}A\{P_e\rho + \rho P_e - 2d^\dagger\rho d\} \tag{3.3}$$

with $P_e = dd^\dagger = |e\rangle\langle e|$, the projector on the excited state and A is the Einstein coefficient for spontaneous decay of this transition.

This active atom is surrounded by a gas of N neutral perturbers, which collide elastically with the atom. When T_i is the kinetic energy of the ith perturber and V_i its interaction with the atom, every perturber contributes $H_i = T_i + V_i$ to the Hamiltonian. The equation of motion for the $N+1$ particle density operator $\rho(t)$ then attains the form

$$i\hbar\frac{d\rho}{dt} = \left[H_a + H_{ar}(t) + \sum_{i=1}^{N} H_i, \rho\right] - i\hbar\Gamma\rho \tag{3.4}$$

which is a stochastic differential equation, due to the fluctuating phase $\phi(t)$ in $H_{ar}(t)$.

The time dependence of $H_{ar}(t)$ with $\exp(\pm i\omega_L t)$ and the occurrence of the stochastic phase in the form (3.2) is cumbersome. This can be simplified considerably with the unitary stochastic transformation

$$\begin{aligned}\sigma(t) &= \exp[-i(\omega_L t + \phi(t))L_g]\rho(t) \\ &= \exp[-i(\omega_L t + \phi(t))P_g]\rho(t)\exp[i(\omega_L t + \phi(t))P_g]\end{aligned} \tag{3.5}$$

with

$$L_g\rho = [P_g, \rho] = -\tfrac{1}{2}[P_e - P_g, \rho] \tag{3.6}$$

and $P_g = 1 - P_e = |g\rangle\langle g|$, the projector on the atomic ground state. If we introduce the dressed-atom Hamiltonian

$$H_d = -\tfrac{1}{2}\hbar\{\Delta(P_e - P_g) + \Omega(d + d^\dagger)\} \tag{3.7}$$

with $\Delta = \omega_L - \omega_e + \omega_g$ the detuning from resonance and the Liouville operators L_d and L_i

$$L_d\rho = \hbar^{-1}[H_d, \rho] - i\Gamma\rho \tag{3.8}$$

$$L_i\rho = \hbar^{-1}[H_i, \rho] \tag{3.9}$$

we can transform equation (3.4), which yields

$$i\frac{d\sigma}{dt} = \left(L_d + \dot{\phi}(t)L_g + \sum_{i=1}^{N} L_i\right)\sigma. \tag{3.10}$$

This equation is equivalent with (3.4) since relation (3.5) can be inverted, but in (3.10) the oscillations with the optical frequency ω_L have disappeared and the phase fluctuations show up in a more handsome way.

From (3.6), (3.7) and (3.8) we see that phase fluctuations in the σ picture add up with the detuning $\Delta = \omega_L - \omega_e + \omega_g$ and hence $\dot{\phi}(t)$ can be regarded as a time-dependent shift of the laser frequency ω_L. In a semiclassical picture of perturber motion, the Liouvillian L_i reduces to

$$L_i\rho = -\hbar^{-1} V_i^d(t) L_g\rho \tag{3.11}$$

with $V_i^d(t)$ the difference of the interaction potential for the atom in the upper and lower state (Yeh and Berman 1979), which imposes a combined modification of the detuning from resonance due to the laser linewidth and collisions according to

$$\Delta \to \Delta(t) = \Delta + \dot{\phi}(t) - \hbar^{-1} \sum_{i=1}^{N} V_i^d(t) \tag{3.12}$$

which shows that $V_i^d(t)$ yields an effective shift of the atomic level separation $\omega_e - \omega_g$. The substitution (3.12) exhibits clearly the additive nature of phase fluctuations and collisional perturbations on a stochastic and microscopic level, but it will turn out that this additivity vanishes after averaging over the stochastic process and the velocity distribution of perturber motion.

We introduce the reduced density operator

$$\sigma_0(t) = \mathrm{Tr}_p\, \sigma(t) \tag{3.13}$$

and similar for $\rho_0(t)$, where the trace is taken over the perturber states, which describes the state of the atom, regardless of the state of the perturbers. From transformation (3.5) it follows that $\sigma_0(t)$ and $\rho_0(t)$ have the same diagonal elements, but the σ picture cannot be applied to predict the atomic coherences, averaged over the stochastic process, since the non-diagonal terms contain the stochastic factor $\exp[\pm i(\omega_L t + \phi(t))]$. It was shown by Agarwal (1978) that a different transformation allows one to find these coherences, but then the populations are affected by stochastic factors.

4. Effect of phase fluctuations

On some initial time t_0 we choose the state of the atom and perturbers to have the non-stochastic value $\sigma(t_0)$. Equation (3.10) then determines $\sigma(t)$ for all later times. For $\phi(t) \equiv 0$, the evolution operator is

$$U(t) = \exp\left[-i\left(L_d + \sum_{i=1}^{N} L_i\right)(t - t_0)\right]. \tag{4.1}$$

We introduce the interaction picture by defining

$$\hat{L}_g(t) = U^{-1}(t) L_g U(t) \tag{4.2}$$

and we find as the integral of (3.10)

$$\sigma(t) = U(t)\theta \exp\left(-i \int_{t_0}^{t} \dot{\phi}(s) \hat{L}_g(s)\, ds\right) \sigma(t_0) \tag{4.3}$$

with θ the time-ordering operator. The time-ordered exponential is the additional factor due to phase fluctuations, which becomes unity for monochromatic irradiation. Since $\sigma(t_0)$ is not stochastic, we have to average the time-ordered exponential to find $\{\sigma(t)\}$. Unfortunately this is a very complicated Liouville operator in the $(N+1)$-particle space. In fact, even for free atoms this time-ordered exponential is very cumbersome.

An exception is the phase diffusion model or the independent increment process, for which we can average (4.3) exactly. If we differentiate the result with respect to time, we find a differential equation for the average

$$i \frac{d}{dt} \{\sigma(t)\} = \left(L_d - i W_0 + \sum_{i=1}^{N} L_i\right) \{\sigma(t)\}. \tag{4.4}$$

The effective relaxation operator W_0 is given by (Arnoldus and Nienhuis 1983)

$$W_0 = \lambda L_g^2 \tag{4.5}$$

where λ is the halfwidth of the laser line as in equation (2.6). We firstly note that W_0 is a time-independent operator, acting only on atomic states. Hence it can be absorbed in L_d, whereafter we can treat equation (4.4) in the same way as in the monochromatic case (Nienhuis 1982). A similar way of averaging also applies to correlation functions and spectra, as we demonstrated in a previous paper (Arnoldus and Nienhuis 1983). We now present a treatment of arbitrary phase fluctuations, combined with collisional effects, without choosing a specific model.

Note that W_0 in (4.4) is not proportional to L_g itself, in contrast to the phase fluctuation contribution in the stochastic equation (3.10) and so, on the average, phase fluctuations can no longer be described as an additive modification of the detuning Δ.

5. Finite correlation times

If we expand the time-ordered exponential in the formal solution (4.3) and average over the $\dot{\phi}(t)$ process, we find

$$\{\sigma(t)\} = U(t)\left(1 - i \int_{t_0}^{t} dt_1 \{\dot{\phi}(t_1)\} \hat{L}_g(t_1) - \int_{t_0}^{t} dt_2 \int_{t_0}^{t_2} dt_1 \{\dot{\phi}(t_2)\dot{\phi}(t_1)\} \hat{L}_g(t_2)\hat{L}_g(t_1) + \cdots\right) \sigma(t_0) \tag{5.1}$$

with a non-stochastic arbitrary initial value $\sigma(t_0)$. The first term in large brackets gives the solution for monochromatic driving fields and the second term vanishes due to $\{\dot{\phi}(t)\} = 0$. The lowest-order correction which accounts for phase fluctuations is the third term. In the phase diffusion model we had

$$\{\dot{\phi}(t_2)\dot{\phi}(t_1)\} = 2\lambda\delta(t_1 - t_2) \tag{5.2}$$

so this δ function is situated precisely on the integration limit of the inner integral in (5.1). In order to find out how the t_1 integration should be performed, we consider the $\dot{\phi}(t)$ process as a limit of the Ornstein-Uhlenbeck process, for which we have

$$\langle\dot{\phi}(t_2)\dot{\phi}(t_1)\rangle = \lambda\gamma \exp(-\gamma|t_2 - t_1|) \qquad \gamma > 0. \tag{5.3}$$

The phase diffusion limit is obtained by taking $\gamma \to \infty$, which yields equation (5.2).

The higher-order moments of $\dot{\phi}(t)$ are also δ correlated (Fox 1972) and every term in (5.1) can be evaluated. The resulting expression in large brackets is again a time-ordered integral, and differentiating the result with respect to t yields (4.4) for the average.

The phase diffusion model can be treated exactly, but it provides an approximate description of the field and the agreement with experiment is poor. We will now try to explain why this is understandable. The moments $\langle\dot{\phi}(t_n)\ldots\dot{\phi}(t_1)\rangle$ occurring in (5.1) contain, according to (5.2), δ functions which are positioned arbitrarily close to the limit of an integration interval. If there are other time scales in the integrand, for instance oscillations, the δ functions are always narrower than every oscillation time irrespective of how small. In other words, the $\dot{\phi}(t)$ process is supposed to contain frequencies, which are larger than every other frequency in the problem under study. This is not realistic as we will show by the following example.

If the parameter

$$\Omega' = \Delta(1 + \Omega^2/\Delta^2)^{1/2} \tag{5.4}$$

becomes much larger than other frequencies like the Einstein coefficient, the laser bandwidth and so on, the spectrum of fluorescence consists of three separated lines, the Mollow triplet (Mollow 1969). This justifies the secular approximation (Cohen-Tannoudji 1977), which treats the evolution of every line independently. The same approximation can be made in equation (4.4) for the average $\langle\sigma(t)\rangle$, but also in equation (3.10) for the stochastic $\sigma(t)$. If we average the latter over the phase diffusion process, we do not find the secular limit of $\langle\sigma(t)\rangle$. Hence the secular limit and the stochastic average do not commute. This is due to the fact that in the phase diffusion limit, $\dot{\phi}(t)$ always couples the three lines on the stochastic level, irrespective of how large Ω', the distance between the lines, is. This is reflected in the time behaviour by the δ functions, which are always narrower than Ω'^{-1}. On the other hand, Ω' can become arbitrary large, independent of the $\dot{\phi}(t)$ process. Nevertheless, phase fluctuations with frequency larger than Ω' will always contribute. This is not realistic. We should leave open the possibility of finite correlation times, like $1/\gamma$ in (5.3), which are not necessarily small compared with Ω'^{-1} or the collision time. This is equivalent to the idea that the wings of the laser profile decay faster than a Lorentzian.

6. Binary collisions

The integral (4.3) for $\sigma(t)$ is not easy to deal with, because it involves $(N+1)$-particle operators. We go back to the differential equation (3.10) and consider $L_d + \dot{\phi}(t)L_g$ as a time-dependent atomic Liouvillian, whereafter we can make the binary collision approximation for time-dependent fields (Nienhuis 1983). The reduced atomic density operator $\sigma_0(t)$ is defined in (3.13), and similarly we introduce the density matrix for the atom and a single perturber by

$$\sigma_1(1; t) = \mathrm{Tr}_{p_2 p_3 \ldots p_N}\, \sigma(t). \tag{6.1}$$

Then, a general equation for $\sigma_0(t)$ is given by

$$\begin{aligned} \mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}\sigma_0(t) &= (L_\mathrm{d}+\dot{\phi}(t)L_\mathrm{g})\sigma_0(t) + N\,\mathrm{Tr}_1\, L_1(\rho_\mathrm{p}(1)\sigma_0(t)) \\ &\quad -\mathrm{i}N\,\mathrm{Tr}_1\, L_1\int_{t_0}^{t} U_2(t,t')L_1(\rho_\mathrm{p}(1)\sigma_0(t'))\,\mathrm{d}t' \\ &\quad + N\,\mathrm{Tr}_1\, L_1 U_2(t,t_0)(\sigma_1(1;t_0)-\rho_\mathrm{p}(1)\sigma_0(t_0)) \end{aligned} \tag{6.2}$$

with Tr_1 the trace over the states of perturber 1, $\rho_\mathrm{p}(1)$ the density operator for perturber 1 in thermal equilibrium and the evolution operator U_2 is defined by

$$U_2(t_2,t_1) = \theta\exp\left(-\mathrm{i}\int_{t_1}^{t_2}(L_\mathrm{d}+\dot{\phi}(s)L_\mathrm{g}+L_1)\,\mathrm{d}s\right). \tag{6.3}$$

Note that equation (6.2) only contains $\sigma_1(1;t)$ at time t_0. For free atoms, e.g. in the absence of collisions, the last three terms in equation (6.2) vanish and the solution, written as $\sigma_\mathrm{a}(t)$, is formally

$$\sigma_\mathrm{a}(t) = U_1(t,t_0)\sigma_\mathrm{a}(t_0) \tag{6.4}$$

where $U_1(t_2,t_1)$ follows from $U_2(t_2,t_1)$ by putting $L_1 \equiv 0$. We will call $\sigma_\mathrm{a}(t)$ from (6.4) the free-atom result if the initial condition is chosen as $\sigma_\mathrm{a}(t_0)=\sigma_0(t_0)$. It obviously obeys the differential equation

$$\mathrm{i}\frac{\mathrm{d}\sigma_\mathrm{a}}{\mathrm{d}t} = (L_\mathrm{d}+\dot{\phi}(t)L_\mathrm{g})\sigma_\mathrm{a}. \tag{6.5}$$

The collision time $\tau_\mathrm{c}\sim 10^{-13}$ s is very small and we can safely assume that $\dot{\phi}(t)$ does not change considerably during a collision. The collision integral in (6.2) contains only $\dot{\phi}(t)$ in the evolution operator. During the small collision time τ_c, we can write $\dot{\phi}(s)\,\mathrm{d}s \simeq \phi(s+\tau_\mathrm{c})-\phi(s)$, if the collision occurs at $t=s$, but this is the phase change in the course of the collision, which is negligible. This implies that we can make the substitution

$$U_2(t,t') \to \exp[-\mathrm{i}(L_\mathrm{d}+L_1)(t-t')] \tag{6.6}$$

in the integral in (6.2). Introduction of the Laplace transform

$$\tilde{\sigma}(\omega) = \int_{t_0}^{\infty}\exp[\mathrm{i}\omega(t-t_0)]\sigma(t)\,\mathrm{d}t \tag{6.7}$$

enables us to write equation (6.2) as

$$\begin{aligned} (\omega - L_\mathrm{d} + \mathrm{i}\Phi(\omega))\tilde{\sigma}_0(\omega) &= (\omega - L_\mathrm{d})\tilde{\sigma}_\mathrm{a}(\omega) \\ &\quad + L_\mathrm{g}\int_{t_0}^{\infty}\exp[\mathrm{i}\omega(t-t_0)]\dot{\phi}(t)(\sigma_0(t)-\sigma_\mathrm{a}(t))\,\mathrm{d}t \\ &\quad + N\,\mathrm{Tr}_1\, L_1\tilde{U}_2(\omega,t_0)(\sigma_1(1;t_0)-\rho_\mathrm{p}(1)\sigma_0(t_0)) \end{aligned} \tag{6.8}$$

with $\tilde{U}_2(\omega,t_0)$ the Laplace transform of the operator given by equation (6.3) and the collision operator is defined as usual by

$$\Phi(\omega)\sigma_0 = N\,\mathrm{Tr}_1(\omega - L_\mathrm{d})\frac{\mathrm{i}}{\omega - L_\mathrm{d} - L_1}L_1(\rho_\mathrm{p}(1)\sigma_0). \tag{6.9}$$

In equation (6.7) we used that $\tilde{\sigma}_a(\omega)$, the Laplace transform of (6.4), obeys the relation

$$\tilde{\sigma}_a(\omega) = \frac{i}{\omega - L_d}\sigma_0(t_0) + \frac{1}{\omega - L_d} L_g \int_{t_0}^{\infty} \exp[i\omega(t-t_0)]\dot{\phi}(t)\sigma_a(t)\,dt \tag{6.10}$$

which follows from (6.5).

Solution (6.8) is still stochastic. If we take $\sigma_1(1; t_0) = \rho_p(1)\sigma_0(t_0)$ as the initial condition, the last term disappears. The second term on the right-hand side vanishes if there are no collisions or if $\dot{\phi}(t) \equiv 0$. In general this term will be small on the average if $\dot{\phi}(t)$ is sufficiently random.

More precisely, the atomic density operator $\sigma_0(t)$ deviates from $\sigma_a(t)$ due to the cumulative effect of all collisions in the time interval $[t_0, t]$. The difference can always be written as

$$\sigma_0(t) - \sigma_a(t) = (\sigma_0(t) - \sigma_a(t))_{\dot{\phi}=0}\{1 + \text{corrections}\} \tag{6.11}$$

just as in (5.1), where the corrections are time-ordered integrals, containing products of $\dot{\phi}(t')$ at times in $[t_0, t]$. If we multiply (6.11) with $\dot{\phi}(t)$ and take the average, the first term vanishes, because $\langle\dot{\phi}(t)\rangle = 0$ and the corrections will only contribute significantly if $\dot{\phi}(t)$ correlations survive over the time delay between two collisions. If we indicate this characteristic correlation time by γ^{-1}, just as in the specific case (5.3), we must require that γ is large compared with the collisional linewidth, since this is of the order of the inverse time between two collisions. The parameter γ has also a direct interpretation as can be seen from (5.3). If $|t_2 - t_1| < \gamma^{-1}$ we have $\langle\dot{\phi}(t_1)\dot{\phi}(t_2)\rangle \sim$ constant, and a constant correlation function yields a Gaussian laser profile $\tilde{I}_L(\omega)$. From (2.2) we see that the short-time behaviour of the correlations determines the wings of the spectrum, so if $|\omega - \omega_L| > \gamma$, the profile becomes Gaussian. This shows that $\langle\dot{\phi}(t)(\sigma_0(t) - \sigma_a(t))\rangle$ can be assumed to be small if the collisional width of spectral lines is smaller than the non-Gaussian width of $\tilde{I}_L(\omega)$ and, of course only if $t - t_0 > \gamma^{-1}$.

With this assumption, the explicit solution for the atomic density operator is given by

$$\begin{aligned}\langle\tilde{\sigma}_0(\omega)\rangle &= \frac{1}{\omega - L_d + i\Phi(\omega)}(\omega - L_d)\int_{t_0}^{\infty}\exp[i\omega(t-t_0)]\langle U_1(t, t_0)\sigma_0(t_0)\rangle\,dt \\ &= \frac{1}{\omega - L_d + i\Phi(\omega)}(\omega - L_d)\langle\tilde{\sigma}_a(\omega)\rangle\end{aligned} \tag{6.12}$$

expressed in the free-atom solution $\langle\tilde{\sigma}_a(\omega)\rangle$. The two factors on the right-hand side account for collisions and are independent of phase fluctuations. If $\dot{\phi}(t) \equiv 0$, we have $U_1(t, t_0) = \exp[-iL_d(t - t_0)]$ and equation (6.12) reduces to

$$\langle\tilde{\sigma}_0(\omega)\rangle = \frac{i}{\omega - L_d + i\Phi(\omega)}\sigma_0(t_0) \tag{6.13}$$

which is the correct monochromatic limit (Nienhuis 1982). It can easily be shown that (6.12) also has the correct phase diffusion solution, however with a slightly modified collision operator, due to the neglect of large phase jumps during a collision. We conclude that (6.12) is the correct generalisation of (6.13) for laser fields with arbitrary lineshape.

The long-time limit

$$\langle\bar{\sigma}_0\rangle = \langle\sigma_0(t = \infty)\rangle = \lim_{\omega \to 0} -i\omega\langle\tilde{\sigma}_0(\omega)\rangle \tag{6.14}$$

follows from (6.11) and obeys the equation

$$(L_d - i\Phi(0))\{\bar{\sigma}_0\} = L_d\{\bar{\sigma}_a\}. \tag{6.15}$$

For monochromatic irradiation, the right-hand side of (6.15) is zero.

7. Dipole correlation function and fluorescence spectrum

The spectral distribution of the emitted fluorescence radiation is related to the dipole correlation function

$$\langle d(t)d^+(t')\rangle = \mathrm{Tr}\, d^+ X(t', t)(\rho(t)d) \qquad t' \geq t \tag{7.1}$$

since $d^+(t) \propto E^{(+)}(t)$, the positive-frequency part of the electric-field component. The time dependence of the Heisenberg operators is governed by the time evolution operator for the density operator $\rho(t)$, as it follows from (3.4)

$$\rho(t') = X(t', t)\rho(t). \tag{7.2}$$

With (3.5) we transform to the σ representation.

$$\langle d(t)d^+(t')\rangle = \exp\{-i[\omega_L(t'-t) + \phi(t') - \phi(t)]\}\, \mathrm{Tr}\, d^+ Y(t', t)(\sigma(t)d) \qquad t' \geq t. \tag{7.3}$$

Here $Y(t', t)$ is the propagator of $\sigma(t)$, as given in (4.3). Now we can introduce a Liouville vector in $(N+1)$-particle space by

$$D(t', t) = \exp[-i(\phi(t') - \phi(t))]Y(t', t)(\sigma(t)d) \qquad t' \geq t \tag{7.4}$$

which obeys the differential equation

$$i\frac{d}{dt'} D(t', t) = \left(L_d + \dot{\phi}(t')(L_g + 1) + \sum_{i=1}^{N} L_i\right) D(t', t) \qquad t' \geq t \tag{7.5}$$

and the initial condition

$$D(t, t) = \sigma(t)d. \tag{7.6}$$

Equation (7.5) is very similar to equation (3.10) for $\sigma(t)$, but an important difference is that the initial condition is also stochastic. Even in the long-time limit $D(t', t)$ remains a stochastic function of t' and t, but it will become independent of the initial value $\sigma(t_0)$. In the same fashion as in (3.13), we can define the atomic operator $D_0(t', t)$ by taking the trace over the perturber states. The correlation function can be written as

$$\langle d(t)d^+(t')\rangle = \exp[-i\omega_L(t'-t)]\, \mathrm{Tr}_a\, d^+ D_0(t', t) \qquad t' \geq t \tag{7.7}$$

and $D_0(t', t)$ can be found by applying the binary-collision approximation, just as in (6.7). With

$$\tilde{D}_0(\omega, t) = \int_0^\infty \exp(i\omega\tau) D_0(t+\tau, t)\, d\tau \tag{7.8}$$

we then find

$$\{\tilde{D}_0(\omega, t)\} = \frac{1}{\omega - L_d + i\Phi(\omega)} (\omega - L_d) \int_0^\infty \exp(i\omega\tau)\{U_3(t+\tau, t)(\sigma_0(t)d)\}\, d\tau \tag{7.9}$$

where we neglected the last two terms of (6.8), which can be shown to be small, and $U_3(t_2, t_1)$ follows from $U_1(t_2, t_1)$ by the substitution $L_g \to L_g + 1$. Relation (7.9) also holds without brackets and is t dependent because the correlation function (7.7) depends on two times. In the phase-diffusion limit, the stochastic average in (7.9) factorises according to

$$\{U_3(t+\tau, t)(\sigma_0(t)d)\} = \{U_3(t+\tau, t)\}\{\sigma_0(t)d\} \tag{7.10}$$

and for $\{\sigma_0(t)\}$ we could use the Laplace inverse of (6.12) or the steady-state solution of (6.15). In the case of phase fluctuations with finite correlation times, this factorisation does not hold anymore, but most authors use it as an approximation. In our opinion, this is not correct.

The correlation function $\langle d(t)d^\dagger(t')\rangle$ depends on both t and t' but intrinsically also on t_0, the initial time for the equation of motion for $\sigma_0(t)$. In the long-time limit t, $t' \gg t_0$, this t_0 dependence disappears, but the t and t' dependence survives, due to the phase fluctuations. The stochastic average $\{\langle d(t)d^\dagger(t')\rangle\}$ however, will depend only on τ in this limit and the spectrum of fluorescence, averaged over phase fluctuations will be stationary. Normalised to the number of emitted photons per second, it is given by

$$I(\omega) = \lim_{t\to\infty} A \,\mathrm{Re}\frac{1}{\pi}\int_0^\infty \exp(\mathrm{i}\omega\tau)\{\langle d(t)d^\dagger(t+\tau)\rangle\}\,\mathrm{d}\tau. \tag{7.11}$$

With (7.7) and (7.8) this becomes

$$I(\omega) = \lim_{t\to\infty} A \,\mathrm{Re}\frac{1}{\pi}\,\mathrm{Tr}_a\, d^\dagger\{\tilde{D}_0(\omega, t)\} \tag{7.12}$$

which can be evaluated directly with the result (7.9). Note that the explicit limit $t \to \infty$ is not trivial because of the breakdown of the factorisation (7.10).

The total fluorescence intensity follows from (7.8) and (7.6)

$$\int_{-\infty}^{\infty} I(\omega)\,\mathrm{d}\omega = A \lim_{t\to\infty} \mathrm{Tr}_a\, d^\dagger\{\sigma_0(t)\}d = A\bar{n}_e \tag{7.13}$$

with $\bar{n}_e = \{\langle e|\bar{\sigma}_0|e\rangle\}$, the averaged steady-state population of the excited level, as given by the solution of (6.15). This $\bar{n}_e$ does depend on phase fluctuations in general, so the emitted fluorescence intensity is affected by the laser lineshape.

8. Separated spectral lines

In the previous sections we showed explicitly how the state of the atom and the fluorescence spectrum are modified by the combined effect of collisions and arbitrary phase fluctuations. In the last part of this paper we consider the limit where Ω' from (5.6) is large compared with every other frequency in the problem, except ω_L. In this limit of strong fields or large detunings, the fluorescence spectrum consists of three lines: the fluorescence line F at $\omega_L - \Omega'$, the Rayleigh line R at ω_L and the three-photon line T at $\omega_L + \Omega'$. Photon emissions can be seen as transitions between eigenstates of

the dressed-atom Hamiltonian (3.7) (Cohen-Tannoudji 1977), which are

$$|1\rangle = |g\rangle \cos \tfrac{1}{2}\theta - |e\rangle \sin \tfrac{1}{2}\theta$$
$$|2\rangle = |g\rangle \sin \tfrac{1}{2}\theta + |e\rangle \cos \tfrac{1}{2}\theta \tag{8.1}$$

with θ defined by

$$\tan \theta = \Omega/\Delta \qquad -\pi/2 < \theta < \pi/2. \tag{8.2}$$

The eigenvalue equations are then

$$H_d|1\rangle = \tfrac{1}{2}\hbar\Omega'|1\rangle \qquad H_d|2\rangle = -\tfrac{1}{2}\hbar\Omega'|2\rangle. \tag{8.3}$$

For large Ω', the dominant frequencies in the equation of motion for $\sigma(t)$ are the eigenvalues of $\hbar^{-1}[H_d, \cdot]$ which are $-\Omega'$, Ω' and zero (two-fold degenerate) and the same holds for equations (7.5) for the dipole correlation function $D(t', t)$. The fluorescence spectrum with its three lines follows from the independent evolution in the three subspaces of eigenvectors of $\hbar^{-1}[H_d, \cdot]$ which are $|2\rangle\langle 1|$ (F space), $|1\rangle\langle 2|$ (T space) and the two-dimensional R space, spanned by $|2\rangle\langle 2|$ and $|1\rangle\langle 1|$. In this secular limit, the coupling between these three subspaces is neglected if the coupling is established by frequencies which are small compared with Ω'.

Another time scale in equation (3.10) results from phase fluctuations $\dot{\phi}(t)$. Consider first the phase diffusion model for which we found the exact average (4.4). It is again easy to check that taking the average does not commute with passing to the secular limit. In other words, if we make the secular approximation in the stochastic equation (3.10) and then average over the $\dot{\phi}(t)$ process, we do not find the secular limit of (4.4). It was pointed out in § 4 that this is a consequence of the δ correlations of the $\dot{\phi}(t)$ process. The δ functions are always smaller in time than $1/\Omega'$ and they give a non-vanishing coupling in the limit of arbitrary large Ω'. For realistic fluctuating phases, this will not be correct, so we assume further on that $\dot{\phi}(t)$ correlation times are larger than $1/\Omega'$, which allows us to apply the secular approximation before averaging over the $\dot{\phi}(t)$ process. This requires that the sidebands of the spectrum are situated in the Gaussian wings of the laser profile, whereas the approximation that led to equation (6.12) imposes the constraint that the collisional widths of the lines are smaller than the spectral region where the laser profile is well described by a Lorentzian. Therefore, the next results will be complementary to previous ones (Arnoldus and Nienhuis 1983).

The correspondence between phase fluctuations and collisions was already shown in equation (3.12). We now make the distinction that collisional fluctuations might be arbitrary fast but phase fluctations should have finite time scales. In the phase diffusion model, this time scale is zero and corresponds to the impact limit of the collision process. The bandwidth λ of the laser is additive to the collisional width in the equation of motion for the density matrix $\sigma(t)$. For correlation functions, this is no longer true since then phase fluctuations are proportional to $L_g + 1$ and collisions to L_g, so even in the impact limit and the phase diffusion limit, $\dot{\phi}(t)$ fluctuations and collisions are not additive in general.

Phase fluctuations in the solution (6.12) for the density operator are contained in the atomic propagator $U_1(t, t_0)$ and are proportional to L_g. In the secular limit, L_g becomes $-\cos\theta$ in F space, $\cos\theta$ in T space and zero in R space, so it commutes with L_d and (6.3) reduces to

$$U_1(t_2, t_1) = \exp[-iL_d(t_2 - t_1)] \exp[-i(\phi(t_2) - \phi(t_1))L_g] \tag{8.4}$$

and the same holds for $U_3(t_2, t_1)$, since L_g+1 also commutes with L_d. The operator $-iL_d$ is in this limit (Nienhuis 1982)

$$-iL_d = \begin{cases} \begin{pmatrix} -a & b \\ a & -b \end{pmatrix} & R \text{ space} \\ i\Omega' - \alpha & F \text{ space} \\ -i\Omega' - \alpha & T \text{ space} \end{cases} \tag{8.5}$$

with the positive parameters

$$a = A\cos^4\tfrac{1}{2}\theta \qquad b = A\sin^4\tfrac{1}{2}\theta \qquad \alpha = A - \tfrac{1}{2}(a+b). \tag{8.6}$$

We will now show that the long-time limit of the density matrix $\sigma_0(t)$ is not stochastic anymore. In order to do so, we start with the solution (6.12), but without brackets. In R space, $L_g = 0$, and we simply have

$$(\omega - L_d)\int_{t_0}^{\infty} \exp[i\omega(t-t_0)]U_1(t, t_0)\sigma_0(t_0)\,dt = i\sigma_0(t_0) \qquad \text{in } R \text{ space} \tag{8.7}$$

and (6.12) reduces to the monochromatic result (6.13). The long-time limit should be independent of $\sigma_0(t_0)$, so on t_0 we can choose the coherences of σ_0 to be zero (the F and T components). From (6.12) it then follows that they are zero for all times, just as in the monochromatic case. If the coherences are not zero on t_0, they damp out because of α in (8.5), which gives a decay with $\exp[-\alpha(t-t_0)]$. So the density operator in the long-time limit is simply the monchromatic solution

$$\bar{\sigma}_0 = |1\rangle\frac{p}{p+q}\langle 1| + |2\rangle\frac{q}{p+q}\langle 2| \tag{8.8}$$

with

$$p = a + k(\Omega, \Delta) \qquad q = b + k(\Omega, \Delta) \tag{8.9}$$

and $k(\Omega, \Delta) > 0$ the rate of optical collisions (Nienhuis 1982).

If we transform (8.8) to the bare states $|e\rangle$, $|g\rangle$, we find with (7.13) the total fluorescence intensity to be

$$A\bar{n}_e = A\left(\frac{1}{2} - \frac{A\Delta^2}{A\Delta^2 + (A + 4k(\Omega, \Delta))\Omega'^2}\right). \tag{8.10}$$

It has its maximum for $\Delta = 0$. In the secular approximation Ω must be dominant over A and $k(\Omega, \Delta)$ when $\Delta = 0$, which implies the saturation limit. We notice that collisions enhance the fluorescence but that phase fluctuations have no effect. This was not the case for the phase diffusion field.

9. The line strengths and lineshapes

The fluorescence spectrum follows from (7.12), (7.9), the analogy for $U_3(t_2, t_1)$ of (8.4) and the result that the density operator is independent of time in the steady state. After a slight rearrangement of (7.9) we find explicitly

$$I(\omega) = A\,\mathrm{Re}\frac{1}{\pi}\mathrm{Tr}_a\, d^{\dagger}\left[\left(1 - \frac{i}{\omega - L_d + i\Phi(\omega)}\Phi(\omega)\right)\int_0^{\infty}\exp[i(\omega - L_d)\tau]\right.$$
$$\left.\times\{\exp[-i(\phi(\tau) - \phi(0))(L_g+1)]\}(\bar{\sigma}_0 d)\,d\tau\right]. \tag{9.1}$$

In order to find the line strengths, we have to integrate this expression over ω in every subspace separately. If we close the contour in the upper part of the complex ω plane with half a circle, it can easily be shown that the second term in the large round brackets gives no contribution. The first term gives $2\pi\delta(\tau)$ in the integrand, so we finally find for the strengths

$$S_\beta = A\,\mathrm{Tr}_a\, d^\dagger \mathscr{P}_\beta(\bar{\sigma}_0 d) \qquad \beta = R, F, T \tag{9.2}$$

with $\mathscr{P}_\beta$ the projector on the subspace β. In this derivation we used that the collision operator does not vary much over the width of a spectral line. To be specific, projections of $\Phi(\omega)$ are given by (Nienhuis 1982)

$$\Phi(\omega) = \begin{cases} k(\Omega, \Delta)\begin{pmatrix} 1 & -1 \\ -1 & 1 \end{pmatrix} & R \text{ space} \\ \Phi_f & F \text{ space} \\ \Phi_t = \Phi_f^* & T \text{ space.} \end{cases} \tag{9.3}$$

Evaluation of (9.2) with $\bar{\sigma}_0$ from (8.8) gives

$$S_R = \tfrac{1}{4}A \sin^2\theta \qquad S_F = \frac{Aq}{p+q}\cos^4\tfrac{1}{2}\theta \qquad S_T = \frac{Ap}{p+q}\sin^4\tfrac{1}{2}\theta \tag{9.4}$$

which obey the identity $S_R + S_F + S_T = A\bar{n}_e$, the total fluorescence. We see that the line strengths do not depend on phase fluctuations, in contrast to the phase diffusion model, but they do depend on collisions.

More interesting are the spectral lineshapes. Partial integration of (9.1) gives the alternative expression

$$I(\omega) = A\,\mathrm{Re}\frac{1}{\pi}\mathrm{Tr}_a\, d^\dagger\left[\frac{\mathrm{i}}{\omega - L_d + \mathrm{i}\Phi(\omega)} \times\left(1 + \int_0^\infty \exp[\mathrm{i}(\omega - L_d)\tau]\frac{\mathrm{d}}{\mathrm{d}\tau}\{\exp[-\mathrm{i}(\phi(\tau)-\phi(0))(L_g+1)]\}\,\mathrm{d}\tau\right)(\bar{\sigma}_0 d)\right] \tag{9.5}$$

and it can again be shown that the second term in large round brackets, which now contains all phase fluctuations, does not contribute to the line strengths. If we expand every operator in (9.5) in the eigenstates of $\mathrm{i}(\omega - L_d)$, use the explicit form of $\bar{\sigma}_0$ and the line strengths and perform another partial integration, we find explicitly

$$\begin{aligned} I(\omega) = {} & S_F\,\mathrm{Re}\frac{1}{\pi}\frac{\alpha - \mathrm{i}\Omega' - \mathrm{i}\Lambda}{\alpha + \Phi_f - \mathrm{i}\Omega' - \mathrm{i}\Lambda}\int_0^\infty \exp[(\mathrm{i}\Lambda + \mathrm{i}\Omega' - \alpha)\tau] \\ & \times\{\exp[-\mathrm{i}(1-\cos\theta)(\phi(\tau)-\phi(0))]\}\,\mathrm{d}\tau \\ & + S_T\,\mathrm{Re}\frac{1}{\pi}\frac{\alpha + \mathrm{i}\Omega' - \mathrm{i}\Lambda}{\alpha + \Phi_t + \mathrm{i}\Omega' - \mathrm{i}\Lambda}\int_0^\infty \exp[(\mathrm{i}\Lambda - \mathrm{i}\Omega' - \alpha)\tau] \\ & \times\{\exp[-\mathrm{i}(1+\cos\theta)(\phi(\tau)-\phi(0))]\}\,\mathrm{d}\tau \\ & + S_R\,\mathrm{Re}\frac{1}{\pi}\frac{a+b-\mathrm{i}\Lambda}{(a+b)(p+q)(p+q-\mathrm{i}\Lambda)} \\ & \times\left((a-b)^2\int_0^\infty \exp(\mathrm{i}\Lambda\tau)\{\exp[-\mathrm{i}(\phi(\tau)-\phi(0))]\}\,\mathrm{d}\tau\right. \\ & \left. + 2(aq+bp)\int_0^\infty \exp[(\mathrm{i}\Lambda - a - b)\tau]\{\exp[-\mathrm{i}(\phi(\tau)-\phi(0))]\}\,\mathrm{d}\tau\right) \end{aligned} \tag{9.6}$$

with $\Lambda = \omega - \omega_L$. We notice that only very simple field correlation functions are required to evaluate this expression for a specific laser model. Furthermore we see that the three complicated expressions Re $\pi^{-1}\ldots$ have strength unity, since the total strength of one line is S_F, S_T and S_R, respectively. The lines are not Lorentzians as in the phase diffusion case. Expression (9.6) does not reduce to the phase diffusion limit if we take for $\phi(t)$ the Wiener-Lévy process. This could be expected, since we imposed the condition $\gamma < \Omega'$, but in the Lorentzian limit we have $\gamma = \infty$.

In the monochromatic limit $\phi(t) \equiv \phi(0)$, equation (9.6) reduces to

$$I(\omega) = S_F \,\mathrm{Re}\,\frac{1}{\pi}\,\frac{1}{\alpha + \Phi_f - \mathrm{i}\Omega' - \mathrm{i}\Lambda} + S_T \,\mathrm{Re}\,\frac{1}{\pi}\,\frac{1}{\alpha + \Phi_t + \mathrm{i}\Omega' - \mathrm{i}\Lambda}$$
$$+ S_R\left(\frac{p-q}{p+q}\right)^2 \delta(\Lambda) + S_R\,\frac{4pq}{(p+q)^2}\,\mathrm{Re}\,\frac{1}{\pi}\,\frac{1}{p+q-\mathrm{i}\Lambda}. \tag{9.7}$$

The δ function appears because Λ is not a principle value. In (9.6) and (9.7) the Rayleigh line is subdivided in two parts, but the first part of the R line in (9.6) does not correspond to the first part in (9.7). If we, however, neglect collisions in (9.6) and (9.7), we find that the coherent and incoherent strengths are independent of phase fluctuations. This shows that the collisions couple both parts of the line, but that the amount of intensity transfer depends on phase fluctuations as well. Or, in other words, phase fluctuations couple the two lines only if collisions are present.

10. The strong-field limit

In the secular limit we required Ω' large, so Ω or Δ large. A special case is $\Omega \gg \Delta$, which is the strong-field limit. The total fluorescence is equal to $\frac{1}{2}A$, as follows from (8.10) which corresponds to the saturation limit. The three-line fluorescence spectrum follows from (9.6) in the limit $\Omega \gg \Delta$. We then have $a = b$, $p = q$, $S_F = S_T = \frac{1}{2}S_R = \frac{1}{8}A$, but more important $\cos\theta = \Delta/\Omega' \to 0$, so all field correlation functions reduce to $\{\exp[-\mathrm{i}(\phi(\tau) - \phi(0))]\}$. As shown in equation (2.5) this quantity can be found from the normalised laser spectrum $\tilde{I}_L(\omega)$. With this result, all integrals in (9.6) reduce to the form

$$\int_0^\infty \exp\{[\mathrm{i}(\omega - \omega_L - \omega_1) - x]\tau\}\{\exp[-\mathrm{i}(\phi(\tau) - \phi(0))]\}\,\mathrm{d}\tau$$
$$= \int_{-\infty}^{\infty} \frac{1}{x - \mathrm{i}(\omega - \omega_1 - \omega')}\,\tilde{I}_L(\omega')\,\mathrm{d}\omega' \tag{10.1}$$

a convolution of $[x - \mathrm{i}(\omega - \omega_L - \omega_1)]^{-1}$ with the normalised and shifted laser profile. With (10.1), we can express the fluorescence spectrum in the strong-field limit entirely in the laser profile and terms which are independent of phase fluctuations.

$$I(\omega) = \frac{A}{8}\,\mathrm{Re}\,\frac{1}{\pi}\,\frac{\alpha - \mathrm{i}(\Lambda + \Omega')}{\alpha + \Phi_f - \mathrm{i}(\Lambda + \Omega')}\int \mathrm{d}\omega'\,\tilde{I}_L(\omega')\,\frac{1}{\alpha - \mathrm{i}(\omega - \omega' + \Omega')}$$
$$+ \frac{A}{8}\,\mathrm{Re}\,\frac{1}{\pi}\,\frac{\alpha - \mathrm{i}(\Lambda - \Omega')}{\alpha + \Phi_t - \mathrm{i}(\Lambda - \Omega')}\int \mathrm{d}\omega'\,\tilde{I}_L(\omega')\,\frac{1}{\alpha - \mathrm{i}(\omega - \omega' - \Omega')}$$
$$+ \frac{A}{4}\,\mathrm{Re}\,\frac{1}{\pi}\,\frac{2a - \mathrm{i}\Lambda}{2p - \mathrm{i}\Lambda}\int \mathrm{d}\omega'\,\tilde{I}_L(\omega')\,\frac{1}{2a - \mathrm{i}(\omega - \omega')}. \tag{10.2}$$

Note that α reduces to $3A/4$. Furthermore we see that the coherent part of the Rayleigh line disappears in the strong-field limit, as usual. For free atoms, equation (10.2) reduces to

$$I(\omega)=\frac{A}{2}\,\mathrm{Re}\,\frac{1}{\pi}\int \mathrm{d}\omega'\,\tilde{I}_{\mathrm{L}}(\omega')\left(\frac{1/4}{\alpha-\mathrm{i}(\omega-\omega'+\Omega')}+\frac{1/4}{\alpha-\mathrm{i}(\omega-\omega'-\Omega')}+\frac{1/2}{2a-\mathrm{i}(\omega-\omega')}\right) \tag{10.3}$$

since $\tilde{I}_{\mathrm{L}}(\omega)$ is real.

11. Conclusions

In this paper we investigated the effect of an arbitrary laser profile as compared with collisional redistribution of fluorescence radiation, emitted by a single two-level atom. Phase fluctuations and collisions are competing effects on a stochastic level, but after averaging over the stochastic process $\dot{\phi}(t)$ and over many collisions, the influence of both effects is quite different. In § 5 it was shown why the commonly used phase diffusion model for the laser field cannot be expected to give the correct results, especially for strong driving fields or far from resonance. It was pointed out that we should allow the $\dot{\phi}(t)$ fluctuations to have a finite correlation time, which makes the calculation much more difficult because of the breakdown of the factorisation of the average for the expectation value of a product of two propagators for non-overlapping time intervals.

Subsequently we showed how arbitrary phase fluctuations can be incorporated in the theory of redistribution by collisions in the binary-collision approximation. Explicit expressions for the atomic density operator, averaged over the $\dot{\phi}(t)$ process were given and it was shown (equation (6.10)) how collisional effects decouple from the stochastic problem for one atom. In a similar fashion we derived general expressions for the dipole correlation function and the steady-state fluorescence spectrum. In the limit of separated spectral lines, we showed that the density operator $\sigma_0(t)$ in the long-time limit is independent of phase fluctuations and the same holds for the line strengths. In this limit, it was shown how the fluorescence spectrum can be expressed in simple field correlation functions. For strong driving fields, the spectrum can be expressed entirely in the normalised laser spectrum $\tilde{I}_{\mathrm{L}}(\omega)$ and bandwidth-independent factors. For free atoms the strong-field spectrum is a convolution of the laser spectrum with the fluorescence spectrum at monochromatic irradiance.

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