

## Resonance fluorescence spectrum of an atom near a phase conjugator

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Received 16 October 1990, in final form 28 January 1991

**Abstract.** Resonance fluorescence by an atom near the surface of a phase conjugator is studied. The atom is irradiated by a finite-linewidth laser beam, and the bandwidth is modelled by a randomly fluctuating phase. Expressions for the steady-state atomic density operator and the resonance fluorescence spectrum are obtained. For a small laser linewidth the spectrum contains two delta peaks: the usual peak at the laser frequency and its phase-conjugate image. In the limit of separated lines the spectrum consists of the Mollow triplet and its phase-conjugate replica. In the image triplet the positions of the fluorescence line and the three-photon line are reversed, as compared with the original triplet. Also, the intensity of the image spectrum is distributed differently over the three lines.

### 1. Introduction

Monochromatic laser irradiation of a two-state atom yields a symmetric resonance fluorescence spectrum around the laser frequency (Carmichael and Walls 1976, Kimble and Mandel 1976), as was observed experimentally (Wu *et al* 1975). In the limit of either strong irradiance or large detuning the spectrum splits into three separate lines, which gives the famous Mollow triplet (Mollow 1969). When the finite laser linewidth is taken into account the spectrum becomes asymmetric, such that the intensity tends to centre around the atomic resonance, rather than around the laser frequency (Avan and Cohen-Tannoudji 1977, Kimble and Mandel 1977, Zoller and Ehlotsky 1977, Knight *et al* 1978, Zoller 1978, George 1980, George and Dixit 1981). When the atom is surrounded by a neutral gas, then collisions have a similar effect (Burnett *et al* 1982, Nienhuis 1982). Another influence on the spectral distribution of resonance fluorescence is the presence of a reflecting surface (Lin *et al* 1983, Huang *et al* 1984, Huang and George 1984, Li and Gong 1987). Near a metal or dielectric the spontaneous decay rate changes, which affects the fluorescence spectrum parametrically through a change in the Einstein coefficient for spontaneous emission.

We consider an atom which is located near the surface of a four-wave mixing phase conjugator (PC). The presence of this PC alters atomic lifetimes (Agarwal 1982, Bochove 1987, Hendriks and Nienhuis 1989a), and affects the photon statistics of atomic resonance fluorescence (Hendriks and Nienhuis 1989b). With  $\omega_L$  the laser frequency,  $\omega_0$  the atomic resonance,  $\bar{\omega}$  the pump frequency of the non-linear crystal and  $\omega$  the frequency of a fluorescent photon, the fluorescence spectrum can be expected to depend on the frequency mismatches

$$\Delta = \omega_L - \omega_0 \quad \delta = \omega_L - \bar{\omega} \quad \Lambda = \omega - \omega_L. \quad (1.1)$$

For any linear surface (metal, dielectric, etc) the detuning  $\delta$  does not appear as a parameter. It can therefore be anticipated that the structure of the spectrum for an atom near a PC is essentially different than for an atom near an ordinary dielectric, at least for  $\delta \neq 0$ .

## 2. Equation of motion

Assume that a laser beam with frequency  $\omega_L$  and (complex-valued) amplitude  $E_0$  propagates along the surface of the non-linear crystal. At the position of the atom, the electric field is then

$$E(t)_L = \text{Re } E_0 e^{-i(\omega_L t + \phi(t))} \quad (2.1)$$

where  $\phi(t)$  is a stochastically fluctuating phase which gives rise to a finite laser linewidth. We shall take  $\phi(t)$  to be the independent-increment process (van Kampen 1981), for which the laser profile is a Lorentzian. This model generalizes the more familiar phase-diffusion process (Arnoldus and Nienhuis 1983). In the dipole and rotating-wave approximation the interaction between the atom and the field is

$$H_{ar}(t) = -\frac{1}{2} \hbar \Omega \exp[-i(\omega_L t + \phi(t))] d + \text{HC} \quad (2.2)$$

where  $d = |e\rangle\langle g|$  is the atomic raising operator from the ground state  $|g\rangle$  to the excited state  $|e\rangle$ . The (complex-valued) parameter  $\Omega$  is the Rabi frequency, defined by

$$\Omega = \frac{1}{\hbar} \langle e | E_0 \cdot \mu | g \rangle \quad (2.3)$$

in terms of the atomic dipole operator  $\mu$ . With  $\hbar\omega_e$  and  $\hbar\omega_g$  the energies of the excited state and ground state, respectively, the atomic Hamiltonian  $H_a$  can be written as

$$H_a = \hbar\omega_e P_e + \hbar\omega_g P_g \quad (2.4)$$

in terms of the projectors  $P_e = |e\rangle\langle e|$  and  $P_g = |g\rangle\langle g|$ . Spontaneous decay is taken into account with a Liouville relaxation operator  $\Gamma$ . For an atom near a PC this operator attains the form

$$\Gamma \Pi = \frac{1}{2} A_e \{ P_e \Pi + \Pi P_e - 2d^\dagger \Pi d \} + \frac{1}{2} A_g \{ P_g \Pi + \Pi P_g - 2d \Pi d^\dagger \} \quad (2.5)$$

which defines its action on an arbitrary Hilbert space operator  $\Pi$ . The relaxation constants  $A_e$  and  $A_g$  are the rate constants for transitions from the excited and ground state, respectively. With  $A$  the Einstein coefficient for spontaneous decay in empty space and  $P$  the phase-conjugate amplitude reflectivity for a plane wave,  $A_e$  and  $A_g$  are

$$A_e = A(1 + \frac{1}{2}|P|^2) \quad A_g = \frac{1}{2}A|P|^2. \quad (2.6)$$

Then the equation of motion for the atomic density operator  $\rho(t)$  becomes

$$i \hbar \frac{d\rho}{dt} = [H_a + H_{ar}(t), \rho] - i \hbar \Gamma \rho. \quad (2.7)$$

The time dependence of the Hamiltonian can be simplified considerably with a stochastic transformation of the density operator. With the Liouvillian  $L_g$  defined as

$$L_g \Pi = [P_g, \Pi] \quad (2.8)$$

for arbitrary  $\Pi$ , the transformed density operator  $\sigma(t)$  is given by

$$\sigma(t) = \exp[-i(\omega_L t + \phi(t))L_g]\rho(t). \tag{2.9}$$

From (2.7) we then readily derive the equation of motion for  $\sigma(t)$ :

$$i \frac{d\sigma}{dt} = (L_d + \dot{\phi}(t)L_g - i\Gamma)\sigma. \tag{2.10}$$

Here,  $L_d$  is the dressed-atom Liouvillian which is given by

$$L_d \Pi = \Delta L_g \Pi - \frac{1}{2}[\Omega d + \Omega^* d^\dagger, \Pi] \tag{2.11}$$

for arbitrary  $\Pi$  and in terms of the detuning  $\Delta = \omega_L - \omega_e + \omega_g = \omega_L - \omega_0$ .

### 3. Stochastic average and steady state

The density operator  $\sigma(t)$  is a stochastic process, due to the appearance of  $\dot{\phi}(t)$  in (2.10). A stochastic average over many realizations of  $\phi(t)$  will be indicated by  $\{ \dots \}$ , in order to distinguish from the notation  $\langle \dots \rangle$  which is reserved for quantum averages. As shown in appendix 1, the average density operator  $\{\sigma(t)\}$  obeys the equation of motion

$$i \frac{d}{dt} \{\sigma\} = (L_d - iW_0 - i\Gamma)\{\sigma\}. \tag{3.1}$$

The Liouville operator  $W_0$  is found to be

$$W_0 = \lambda_1 L_g^2 \tag{3.2}$$

where  $\lambda_1$  is a parameter of the independent-increment process, which equals the laser linewidth.

Of particular interest is the steady-state solution  $\bar{\sigma} = \{\sigma(t \rightarrow \infty)\}$  of (3.1). For  $t \rightarrow \infty$  the left-hand side of (3.1) goes to zero, and the equation can be solved immediately for  $\bar{\sigma}$ . The population of the excited state is found to be

$$\bar{n}_e = \langle e | \bar{\sigma} | e \rangle = \frac{\frac{1}{2}|\Omega|^2 \eta + A_g(\Delta^2 + \eta^2)}{|\Omega|^2 \eta + 2\bar{A}(\Delta^2 + \eta^2)} \tag{3.3}$$

where we introduced the abbreviations

$$\bar{A} = \frac{1}{2}(A_e + A_g) \quad \eta = \lambda_1 + \bar{A}. \tag{3.4}$$

The population of the ground state is, of course,  $\bar{n}_g = 1 - \bar{n}_e$ . For the coherence we obtain

$$\bar{\sigma}_{eg} = \langle e | \bar{\sigma} | g \rangle = \frac{\frac{1}{2}\Omega A(i\eta - \Delta)}{|\Omega|^2 \eta + 2\bar{A}(\Delta^2 + \eta^2)}. \tag{3.5}$$

It is interesting to notice that the population of  $|e\rangle$  and the coherence are related by

$$\bar{\sigma}_{eg} = \frac{\Omega}{\Delta + i\eta} (\bar{n}_e - \frac{1}{2}). \tag{3.6}$$

If the intensity reflectivity  $|P|^2$  is much larger than unity and if  $A|P|^2 \gg \lambda_1$ , then  $\bar{n}_e \rightarrow \frac{1}{2}$ ,  $\bar{\sigma}_{eg} \rightarrow 0$ , irrespective of the values of  $\Omega$  and  $\Delta$ .

#### 4. Field correlation function

The atom emits fluorescence radiation, which can be detected by a photon counter in the far field. The positive-frequency part of this field is given by (Arnoldus and George 1991)

$$E(t)^{(+)} = d^{\dagger}(t) - \exp(-2i\bar{\omega}t)P^*d(t) \quad (4.1)$$

where an overall factor and retardation have been suppressed. The time dependence of  $d(t)$  and  $d^{\dagger}(t)$  signifies the Heisenberg picture, and the factor  $\exp(-2i\bar{\omega}t)$  arises due to the fact that the reflected waves at the surface are generated in a four-wave mixing process. The spectral distribution of the fluorescent photons is determined by the two-time field correlation function

$$C(t_1, t_2) = \langle\langle E(t_1)^{(-)}E(t_2)^{(+)} \rangle\rangle \quad (4.2)$$

which involves both a quantum and a stochastic average. When  $C(t_1, t_2)$  would only depend on  $t_1$  and  $t_2$  through  $\tau = t_2 - t_1$ , then the correlation function would be stationary and the fluorescence spectrum would be time independent. Due to the factor  $\exp(-2i\bar{\omega}t)$  in (4.1), this is not necessarily the case, even in the steady state. We now evaluate  $C(t_1, t_2)$  for  $t_1 \rightarrow \infty$ ,  $t_2 \rightarrow \infty$ , but with  $\tau = t_2 - t_1$  kept constant and positive.

Substitution of (4.1) into (4.2) yields

$$C(t_1, t_2) = \langle\langle d^{\dagger}(t_1)d^{\dagger}(t_2) \rangle\rangle + |P|^2 \exp[-2i\bar{\omega}(t_2 - t_1)] \langle\langle d^{\dagger}(t_1)d(t_2) \rangle\rangle \\ - P^* \exp(-2i\bar{\omega}t_2) \langle\langle d(t_1)d(t_2) \rangle\rangle - P \exp(2i\bar{\omega}t_1) \langle\langle d^{\dagger}(t_1)d^{\dagger}(t_2) \rangle\rangle. \quad (4.3)$$

Then we transform the expectation values to the Schrödinger picture, and with (2.9) we go to the  $\sigma$  representation. This leads to

$$C(t_1, t_2) = \exp[-i\omega_L(t_2 - t_1)] \text{Tr} d^{\dagger}\{D_1(t_1, t_2)\} \\ + |P|^2 \exp[i(\omega_L - 2\bar{\omega})(t_2 - t_1)] \text{Tr} d\{D_2(t_1, t_2)\} \\ - P^* \exp(i\omega_L t_1) \exp[i(\omega_L - 2\bar{\omega})t_2] \text{Tr} d\{D_3(t_1, t_2)\} \\ - P \exp(-i\omega_L t_2) \exp[i(2\bar{\omega} - \omega_L)t_1] \text{Tr} d^{\dagger}\{D_4(t_1, t_2)\} \quad (4.4)$$

where we have introduced the stochastic operators

$$D_1(t_1, t_2) = \exp[-i(\phi(t_2) - \phi(t_1))] Y(t_2, t_1) [\sigma(t_1)d] \quad (4.5)$$

$$D_2(t_1, t_2) = \exp[i(\phi(t_2) - \phi(t_1))] Y(t_2, t_1) [\sigma(t_1)d^{\dagger}] \quad (4.6)$$

$$D_3(t_1, t_2) = \exp[i(\phi(t_2) + \phi(t_1))] Y(t_2, t_1) [\sigma(t_1)d] \quad (4.7)$$

$$D_4(t_1, t_2) = \exp[-i(\phi(t_2) + \phi(t_1))] Y(t_2, t_1) [\sigma(t_1)d^{\dagger}]. \quad (4.8)$$

Here,  $Y(t, t')$  is the evolution operator for  $\sigma$ :

$$\sigma(t) = Y(t, t')\sigma(t'). \quad (4.9)$$

With (4.9) and the equation of motion (2.10) for  $\sigma$ , we find from (4.5)-(4.8)

$$i \frac{d}{dt_2} D_{1,4}(t_1, t_2) = (L_d + \dot{\phi}(t_2)(L_g + 1) - i\Gamma) D_{1,4}(t_1, t_2) \quad (4.10)$$

$$i \frac{d}{dt_2} D_{2,3}(t_1, t_2) = (L_d + \dot{\phi}(t_2)(L_g - 1) - i\Gamma) D_{2,3}(t_1, t_2). \quad (4.11)$$

Even though  $D_1$  and  $D_4$  obey the same differential equation, the solutions are different because  $D_1$  and  $D_4$  have different initial values ( $t_2 = t_1$  in (4.5)-(4.8)). Similarly,  $D_2$  and  $D_3$  are different. Equations (4.10) and (4.11) are stochastic differential equations of the type discussed in appendix 1. The solutions for the averages are

$$\{D_1(t_1, t_2)\} = \exp[-i(L_d - iW_{+1} - i\Gamma)\tau]\{\sigma(t_1)d\} \tag{4.12}$$

$$\{D_2(t_1, t_2)\} = \exp[-i(L_d - iW_{-1} - i\Gamma)\tau]\{\sigma(t_1)d^\dagger\} \tag{4.13}$$

$$\{D_3(t_1, t_2)\} = \exp[-i(L_d - iW_{-1} - i\Gamma)\tau]\{\exp(2i\phi(t_1))\sigma(t_1)d\} \tag{4.14}$$

$$\{D_4(t_1, t_2)\} = \exp[-i(L_d - iW_{+1} - i\Gamma)\tau]\{\exp(-2i\phi(t_1))\sigma(t_1)d^\dagger\} \tag{4.15}$$

for  $\tau = t_2 - t_1$  positive. The Liouville operators  $W_{+1}$  and  $W_{-1}$  are given by (A1.5).

The steady-state correlation function follows from (4.12)-(4.15) by taking  $t_1 \rightarrow \infty$  while keeping  $\tau$  fixed. It is shown in appendix 2 that

$$\lim_{t_1 \rightarrow \infty} \{\exp(\pm 2i\phi(t_1))\sigma(t_1)\} = 0. \tag{4.16}$$

Therefore,  $\{D_3(t_1, t_2)\}$  and  $\{D_4(t_1, t_2)\}$  vanish in the steady state, whereas in (4.12) and (4.13) we can simply replace  $\{\sigma(t_1)\}$  by  $\bar{\sigma}$ . We find for the correlation function

$$C(t_1, t_2) = \text{Tr } d^\dagger \exp[-i(\omega_L + L_d - iW_{+1} - i\Gamma)(t_2 - t_1)](\bar{\sigma}d) + |P|^2 \text{Tr } d \exp[-i(2\bar{\omega} - \omega_L + L_d - iW_{-1} - i\Gamma)(t_2 - t_1)](\bar{\sigma}d^\dagger). \tag{4.17}$$

It appears that  $C(t_1, t_2)$  only depends on  $t_1$  and  $t_2$  through  $t_2 - t_1$ , and the correlation function is therefore stationary.

### 5. Intensity

The spectrally unresolved fluorescence intensity is given by

$$I = \langle\langle E(t)^{(-)} E(t)^{(+)} \rangle\rangle \tag{5.1}$$

which becomes with (4.17)

$$I = \bar{n}_e + |P|^2 \bar{n}_g. \tag{5.2}$$

With (3.3) we then find

$$I = \frac{\frac{1}{2}|\Omega|^2 \eta (1 + |P|^2) + A_g (\Delta^2 + \eta^2) (3 + |P|^2)}{|\Omega|^2 \eta + 2\bar{A} (\Delta^2 + \eta^2)}. \tag{5.3}$$

For a very strong laser field we reach the saturation limit

$$I = \frac{1}{2}(1 + |P|^2) \quad |\Omega|^2 \rightarrow \infty \tag{5.4}$$

which is a factor of  $1 + |P|^2$  larger than for an atom in empty space. For a strongly reflecting surface we have

$$I \rightarrow \frac{1}{2}|P|^2 \quad |P|^2 \rightarrow \infty \tag{5.5}$$

and there is no saturation limit. The intensity increases indefinitely with  $|P|^2$ .

## 6. Fluorescence spectrum

Since the two-time correlation function  $C(t_1, t_2)$  is stationary, the spectral distribution of the fluorescence is given by the Wiener-Khintchin relation

$$I(\omega) = \frac{1}{\pi} \operatorname{Re} \int_0^\infty d\tau e^{i\omega\tau} C(0, \tau). \quad (6.1)$$

This spectrum is normalized as

$$\int d\omega I(\omega) = I. \quad (6.2)$$

Combination of (4.17) and (6.1) gives

$$I(\omega) = \frac{1}{\pi} \operatorname{Re} \operatorname{Tr} d^+ \frac{1}{iL_d + W_{+1} + \Gamma - i(\omega - \omega_L)} (\bar{\sigma}d) \\ + \frac{1}{\pi} |P|^2 \operatorname{Re} \operatorname{Tr} d \frac{1}{iL_d + W_{-1} + \Gamma - i(\omega - \omega_L + 2\delta)} (\bar{\sigma}d^+) \quad (6.3)$$

where we have set  $\delta = \omega_L - \bar{\omega}$ . Working out this formal result yields explicitly

$$I(\omega) = \frac{|\Omega|^2}{2\pi} \left(\frac{1}{2} - \bar{n}_e\right) \operatorname{Re} \frac{(\lambda_1 + A - i\Lambda)(\bar{A} + \lambda_2 + i\Delta - i\Lambda)}{(\lambda_1 - i\Lambda)(\eta + i\Delta)D_{+1}(-i\Lambda)} \\ + \frac{1}{\pi} \bar{n}_e \operatorname{Re} \frac{\frac{1}{2}|\Omega|^2 + (2\bar{A} + \lambda_1 - i\Lambda)(\bar{A} + \lambda_2 + i\Delta - i\Lambda)}{D_{+1}(-i\Lambda)} \\ + \frac{|\Omega|^2}{2\pi} |P|^2 \left(\bar{n}_e - \frac{1}{2}\right) \operatorname{Re} \frac{(\lambda_1 - A - 2i\delta - i\Lambda)(\bar{A} + \lambda_2 - i\Delta - 2i\delta - i\Lambda)}{(\lambda_1 - 2i\delta - i\Lambda)(\eta - i\Delta)D_{-1}(-2i\delta - i\Lambda)} \\ + \frac{1}{\pi} |P|^2 (1 - \bar{n}_e) \operatorname{Re} \frac{\frac{1}{2}|\Omega|^2 + (2\bar{A} + \lambda_1 - 2i\delta - i\Lambda)(\bar{A} + \lambda_2 - i\Delta - 2i\delta - i\Lambda)}{D_{-1}(-2i\delta - i\Lambda)} \quad (6.4)$$

in terms of  $\Lambda = \omega - \omega_L$  and the population of the excited state  $\bar{n}_e$  from (3.3). We have introduced the abbreviations

$$D_{+1}(s) = |\Omega|^2(\bar{A} + \frac{1}{2}\lambda_2 + s) + (2\bar{A} + \lambda_1 + s)(\bar{A} - i\Delta + s)(\bar{A} + \lambda_2 + i\Delta + s) \quad (6.5)$$

$$D_{-1}(s) = |\Omega|^2(\bar{A} + \frac{1}{2}\lambda_2 + s) + (2\bar{A} + \lambda_1 + s)(\bar{A} + i\Delta + s)(\bar{A} + \lambda_2 - i\Delta + s) \quad (6.6)$$

and used (3.6),  $\bar{n}_g = 1 - \bar{n}_e$ , and  $A_e - A_g = A$ .

## 7. Small laser linewidth

In order to elucidate the structure of the fluorescence spectrum we consider the limit where the laser bandwidth tends to zero. The halfwidth at half maximum of the Lorentzian laser lineshape equals  $\lambda_1$ . The limit  $\lambda_1 \rightarrow 0$  then also implies  $\lambda_2 \rightarrow 0$ , according to (A1.7). In the first term on the right-hand side of (6.4) we get a factor  $(-i\Lambda)^{-1}$ , which should be interpreted as

$$\frac{1}{-i\Lambda} = P\left(\frac{i}{\Lambda}\right) + \pi\delta(\Lambda) \quad (7.1)$$

where  $P$  stands for principal value. Similarly, the third term gives a  $\delta$  function at  $2\delta + \Lambda = 0$ . When we work out this limit we obtain

$$I(\omega) = |\tilde{\sigma}_{eg}|^2 \{ \delta(\omega - \omega_L) + |P|^2 \delta(\omega + \omega_L - 2\bar{\omega}) \} + \text{smooth background.} \tag{7.2}$$

Apparently, the resonance fluorescence spectrum contains two delta function peaks, located at  $\omega = \omega_L$  and  $\omega = 2\bar{\omega} - \omega_L$ . The line separation is  $2|\bar{\omega} - \omega_L|$ , and the intensity of the line at  $2\bar{\omega} - \omega_L$  is  $|P|^2$  times the intensity of the line at  $\omega_L$ .

### 8. Dressed states

The Liouvillian  $L_d$ , as defined in (2.11), can alternatively be written as

$$L_d \Pi = \hbar^{-1} [H_d, \Pi] \tag{8.1}$$

in terms of the dressed-atom Hamiltonian

$$H_d = -\frac{1}{2} \hbar \{ \Delta (P_e - P_g) + \Omega d + \Omega^* d^\dagger \}. \tag{8.2}$$

Eigenvalues of  $H_d$  are found to be

$$H_d |\pm\rangle = \mp \frac{1}{2} \hbar \Omega' |\pm\rangle \tag{8.3}$$

with

$$\Omega' = \text{sgn}(\Delta) (\Delta^2 + |\Omega|^2)^{1/2}. \tag{8.4}$$

When we introduce the phase  $\psi$  and angle  $\theta$  by

$$\Omega = |\Omega| e^{i\psi} \quad \psi \text{ real} \quad \theta = \tan^{-1} (|\Omega|/\Delta) \tag{8.5}$$

then the dressed states  $|+\rangle$  and  $|-\rangle$  can be parametrized as

$$\begin{aligned} |+\rangle &= |g\rangle e^{-i\psi} \sin \frac{1}{2}\theta + |e\rangle \cos \frac{1}{2}\theta \\ |-\rangle &= |g\rangle \cos \frac{1}{2}\theta - |e\rangle e^{i\psi} \sin \frac{1}{2}\theta. \end{aligned} \tag{8.6}$$

The operator  $L_d$  has four eigenvectors in Liouville space and the eigenvalue equations read

$$L_d |\pm\rangle \langle \pm| = 0 \quad L_d |\pm\rangle \langle \mp| = \mp \Omega' |\pm\rangle \langle \mp|. \tag{8.7}$$

### 9. Spectrum of separated lines

When the value of  $|\Omega'|$  is much larger than any of the relaxation constants  $A_e, A_g, \lambda_1$  and  $\lambda_2$ , then the coupling between eigenvectors of  $L_d$  with different eigenvalues can be neglected in time evolution and time regression. This occurs for either high irradiance ( $|\Omega|$  large) or large detuning  $\Delta$ . For an atom in empty space, this leads to a spectrum of three separate lines, which is the Mollow triplet (Mollow 1969). The fluorescence (F) line appears at  $\omega = \omega_L - \Omega'$ , and for a weak driving field this is approximately the atomic resonance  $\omega_0$ . The Rayleigh (R) line is positioned at  $\omega = \omega_L$ , and the three-photon (T) line appears at  $\omega = \omega_L + \Omega'$ .

In the limit of separated lines, the steady-state density operator  $\bar{\sigma}$  is most conveniently expressed with respect to dressed states. The solution of (3.1), with the left-hand side replaced by zero, is found to be

$$\bar{\sigma} = |+\rangle \bar{n}_+ \langle +| + |-\rangle \bar{n}_- \langle -| \quad (9.1)$$

where the populations of the dressed states are given by

$$\bar{n}_+ = \frac{v}{u+v} \quad \bar{n}_- = \frac{u}{u+v} \quad (9.2)$$

in terms of

$$u = A_e g_-^2 + A_g g_+^2 + 2\lambda_1 g_0^2 \quad v = A_e g_+^2 + A_g g_-^2 + 2\lambda_1 g_0^2. \quad (9.3)$$

The three optical parameters are defined as

$$g_{\pm} = \frac{\Omega' \mp \Delta}{2\Omega'} \quad g_0 = \frac{|\Omega|}{2\Omega'}. \quad (9.4)$$

It then follows that

$$u - v = A\Delta/\Omega' > 0 \quad (9.5)$$

which shows that the population of  $|-\rangle$  is always larger than the population of  $|+\rangle$ .

The resonance fluorescence spectrum  $I(\omega)$ , as given by (6.3), can now be evaluated for the limit of separated lines. First we introduce the parameters

$$w = (4\lambda_1 - \lambda_2) g_0^2 \quad (9.6)$$

$$y_F = \tilde{A}(1 + 2g_0^2) + 2\lambda_1 g_+^2 + \lambda_2 g_+^2 \quad (9.7)$$

$$y_T = \tilde{A}(1 + 2g_0^2) + 2\lambda_1 g_0^2 + \lambda_2 g_-^2 \quad (9.8)$$

where  $y_F$  and  $y_T$  are related by

$$y_T - y_F = \lambda_2 \Delta / \Omega' \geq 0. \quad (9.9)$$

Then the spectrum is found to be

$$\begin{aligned} I(\omega) = & I_{Rc} \operatorname{Re} \frac{1}{\pi} \frac{1}{\lambda_1 - i\Lambda} + I_{Ri} \operatorname{Re} \frac{1}{\pi} \frac{1}{\lambda_1 + u + v - 2w - i\Lambda} + I_F \operatorname{Re} \frac{1}{\pi} \frac{1}{y_F - i(\Lambda + \Omega')} \\ & + I_T \operatorname{Re} \frac{1}{\pi} \frac{1}{y_T - i(\Lambda - \Omega')} + I_{Rc} \operatorname{Re} \frac{1}{\pi} \frac{1}{\lambda_1 - i(\Lambda + 2\delta)} \\ & + I_{Ri} \operatorname{Re} \frac{1}{\pi} \frac{1}{\lambda_1 + u + v - 2w - i(\Lambda + 2\delta)} \\ & + I_F \operatorname{Re} \frac{1}{\pi} \frac{1}{y_F - i(\Lambda + 2\delta - \Omega')} + I_T \operatorname{Re} \frac{1}{\pi} \frac{1}{y_T - i(\Lambda + 2\delta + \Omega')} \end{aligned} \quad (9.10)$$

which is a sum of eight Lorentzians. The factors  $I_\alpha$ , defined below, are the strengths of the lines.

The first line is the coherent part of the ordinary Rayleigh line. This line is located at  $\omega = \omega_L$ , has a width (half-width at half-maximum) equal to the laser linewidth, and a strength

$$I_{Rc} = g_0^2 \frac{(u-v)^2}{(u+v)(u+v-2w)}. \quad (9.11)$$



The second line is the incoherent part of the Rayleigh line, which is also positioned at  $\omega = \omega_L$ , has a width  $\lambda_1 + u + v - 2w$  and a strength

$$I_{Ri} = 2g_0^2 \frac{u(v-w) + v(u-w)}{(u+v)(u+v-2w)} \tag{9.12}$$

The total strength of the R line is

$$I_{Rc} + I_{Ri} = \frac{|\Omega|^2}{4(\Delta^2 + |\Omega|^2)} \tag{9.13}$$

which is independent of any of the relaxation mechanisms and independent of the presence of the PC. The third and the fourth lines are the F and T lines, respectively. Their widths are  $y_F$  and  $y_T$ , and their strengths are given by

$$I_F = g_-^2 \bar{n}_+ \quad I_T = g_+^2 \bar{n}_- \tag{9.14}$$

It follows from (9.9) that the width of the T line is always larger than the width of the F line. With (9.14) it can be shown that

$$I_F \geq I_T \tag{9.15}$$

so that the F line is stronger than the T line. The equal sign holds when  $|P|^2 = 0$ ,  $\lambda_1 = 0$ . Then we also have  $y_F = y_T$ , and the R, F, and T lines form a Mollow triplet which is symmetric around  $\omega_L$ . The triplet is always symmetric for  $\Delta = 0$ . Although the strengths and the widths of the lines in the Mollow triplet are affected by the PC, through the dependence on  $|P|^2$ , the structure of the triplet is essentially the same as for an atom in empty space.

Part of the emitted radiation in the Mollow triplet propagates towards the surface of the PC, where it is reflected as phase-conjugated radiation. A photon with frequency  $\omega$  is reflected as a photon with frequency  $\omega' = 2\bar{\omega} - \omega$  due to the four-wave mixing which is responsible for the reflection. The R, F and T lines are positioned at  $\omega_L$ ,  $\omega_L - \Omega'$  and  $\omega_L + \Omega'$ , respectively. The phase-conjugate image of this triplet can therefore be expected to be again a triplet, but with lines R', F' and T' at  $2\bar{\omega} - \omega_L$ ,  $2\bar{\omega} - \omega_L + \Omega'$  and  $2\bar{\omega} - \omega_L - \Omega'$ , respectively. This appears to be indeed the case, and this reflected triplet is given by the last four Lorentzians in (9.10). Due to the phase conjugation, the reflected sidebands F' and T' lie on different sides of the central R' line, as compared with the positions of F and T with respect to R. For  $\omega_L = \bar{\omega}$  ( $\delta = 0$ ), the R', F' and T' lines are at the same positions as the R, T and F lines, respectively. Furthermore, for the coherent and incoherent parts of the R' line we find

$$I_{R'c} = |P|^2 I_{Rc} \quad I_{R'i} = |P|^2 I_{Ri} \tag{9.16}$$

as is anticipated. For the F' and T' lines, however, we have

$$I_{F'} = |P|^2 g_-^2 \bar{n}_- \quad I_{T'} = |P|^2 g_+^2 \bar{n}_+ \tag{9.17}$$

which is not simply  $|P|^2$  times  $I_F$  and  $I_T$ , respectively. In analogy to (9.15) we obtain

$$I_{F'} \geq I_{T'} \tag{9.18}$$

showing that also the F' line is stronger than the T' line. For  $\Delta = 0$  we have  $I_{F'} = I_{T'}$ .

It is interesting to compare the total strength of the Mollow triplet with the total strength of the reflected triplet, in the case of unit reflectivity ( $|P|^2 = 1$ ). We find for the difference

$$(I_{F'} + I_{R'} + I_{T'}) - (I_F + I_R + I_T) = \frac{A}{u+v} (g_+ - g_-)^2 \tag{9.19}$$

which is non-negative and equal to the population difference  $\bar{n}_g - \bar{n}_e$ . Therefore, the reflected triplet is stronger than the incident triplet, even though  $|P|^2 = 1$ . The strengths are equal only if  $\Delta = 0$ .

Another special case of interest is  $\delta = \Delta = 0$ , which implies  $\omega_L = \omega_0 = \bar{\omega}$ . Then the spectrum becomes

$$I(\omega) = \frac{1}{8\pi} (1 + |P|^2) \operatorname{Re} \left\{ \frac{2}{\lambda_1 + 2(u - w) - i\Lambda} + \frac{1}{y_F - i(\Lambda + \Omega')} + \frac{1}{y_F - i(\Lambda - \Omega')} \right\} \quad (9.20)$$

which is symmetric around  $\omega_L$ . The sum of the intensities of the two sidebands is equal to the intensity of the central line at  $\Lambda = 0$ .

## 10. Conclusions

We have studied the resonance fluorescence spectrum of a two-state atom near the surface of a PC. Stochastic fluctuations in the laser phase have accounted for the finite laser linewidth. This phase has been taken to be the independent-increment process, which has the phase-diffusion process as its Gaussian limit. Equations (3.3) and (3.5) give the stochastically averaged population of the excited state and the atomic coherence, respectively. The field correlation function  $C(t_1, t_2)$  has in general four terms, but in the steady state two contributions vanish identically. This is due to the finite laser bandwidth, as shown in (4.16). In addition,  $C(t_1, t_2)$ , and thereby the fluorescence spectrum, turn out to be stationary in the long-time limit. Without taking into account laser fluctuations this would not be the case.

The fluorescent intensity and its spectral distribution have then been evaluated. It appears that in the limit of a small laser linewidth the spectrum contains delta functions at  $\omega_L$  and  $2\bar{\omega} - \omega_L$ , superimposed on a continuous background. This could be expected, since a wave at frequency  $\omega$  is reflected as a wave at frequency  $2\bar{\omega} - \omega$  by a four-wave mixing PC (Hellwarth 1977). In the limit of separated lines the spectrum consists of six distinct lines. Three of these form the usual Mollow triplet, and the additional three lines are the reflections of this triplet at the surface of the PC. Phase conjugation is seen to have the effect of interchanging the positions of the fluorescence line and the three-photon line, with respect to the central Rayleigh line. In addition, phase conjugation has the effect of redistributing the intensities in the sidebands, such that the strength of the fluorescence line is enhanced at the expense of the three-photon line, provided that the atom is excited off-resonance.

## Acknowledgment

This research was supported in part by the Office of Naval Research.

## Appendix 1

Consider a stochastic operator of the form

$$\zeta_n(t) = \exp(-in \phi(t)) \sigma(t) \quad (\text{A1.1})$$

where  $\sigma(t)$  is related to the atomic density operator according to (2.9). Taking the time derivative of (A1.1) yields

$$i \frac{d}{dt} \zeta_n(t) = (L_d + \dot{\phi}(t)(L_g + n) - i\Gamma)\zeta_n(t) \tag{A1.2}$$

where we have used (2.10). Now we take  $\phi(t)$  to be the independent increment process, for which the solution of (A.12) for the average is (Arnoldus and Nienhuis 1983)

$$\langle \zeta_n(t) \rangle = \exp[-i(L_d - iW_n - i\Gamma)(t - t_0)] \langle \zeta_n(t_0) \rangle. \tag{A1.3}$$

Due to the fact that the process  $\dot{\phi}$  is delta correlated, the time regression for  $t > t_0$  factors from the initial condition at  $t = t_0$ . The Liouville operator  $W_n$  is found to be

$$W_n = \int_{-\infty}^{\infty} d\phi w(\phi) \{1 - \exp[-i\phi(L_g + n)]\} \tag{A1.4}$$

in terms of the jump-rate probability distribution  $w(\phi)$ . The result of (A1.3) and (A1.4) is independent of the form of  $L_g$ . When we take  $L_g$  as defined in (2.8), we obtain explicitly

$$W_n \Pi = \lambda_n \{P_e \Pi P_e + P_g \Pi P_g\} + \lambda_{n-1} P_e \Pi P_g + \lambda_{n+1} P_g \Pi P_e \tag{A1.5}$$

for arbitrary  $\Pi$ . The positive parameters  $\lambda_n$  are defined as

$$\lambda_n = \int_{-\infty}^{\infty} d\phi w(\phi) (1 - \cos(n\phi)) \tag{A1.6}$$

where we have assumed  $w(-\phi) = w(\phi)$ . Obviously,  $\lambda_{-n} = \lambda_n$  and  $\lambda_0 = 0$ . The parameters which determine the fluorescence spectrum are  $\lambda_1$  and  $\lambda_2$ , which are restricted by

$$0 \leq \lambda_2 \leq 4\lambda_1 \tag{A1.7}$$

as follows from (A1.6). In the Gaussian limit (phase-diffusion model) we have  $\lambda_2 = 4\lambda_1$ .

### Appendix 2

In this section we evaluate the limit  $t \rightarrow \infty$  for the solution (A1.3). In terms of the Laplace transform

$$\langle \tilde{\zeta}_n(s) \rangle = \int_{t_0}^{\infty} dt \exp[-s(t - t_0)] \langle \zeta_n(t) \rangle \tag{A2.1}$$

the solution (A1.3) reads

$$\langle \tilde{\zeta}_n(s) \rangle = \frac{1}{s + iL_d + W_n + \Gamma} \langle \zeta_n(t_0) \rangle \tag{A2.2}$$

where the initial value  $\langle \zeta_n(t_0) \rangle$  is arbitrary. The operator  $s + iL_d + W_n + \Gamma$  can be represented by a  $4 \times 4$  matrix, and its inverse is easily calculated. Then we find the long-time solution from the identity

$$\lim_{t \rightarrow \infty} \langle \zeta_n(t) \rangle = \lim_{s \rightarrow 0} s \langle \tilde{\zeta}_n(s) \rangle \tag{A2.3}$$

with result

$$\lim_{t \rightarrow \infty} \langle \zeta_n(t) \rangle = \delta_{n0} \bar{\sigma} \text{Tr} \langle \zeta_n(t_0) \rangle. \tag{A2.4}$$

Here,  $\bar{\sigma}$  is the steady-state density operator from section 3. It is remarkable that the long-time solution is only non-zero for  $n=0$ .

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