

## RESONANCE FLUORESCENCE SPECTRUM OF TWO ATOMS, COHERENTLY DRIVEN BY A STRONG RESONANT LASER FIELD

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In Lehmburg's approach, we consider the resonance fluorescence spectrum of two radiatively interacting atoms. In the strong field limit we have obtained analytical solutions for the spectrum of the symmetric and antisymmetric modes without decoupling approximation. Our solutions are valid for all values of the distance  $r_{12}$  separating the atoms. The spectrum of the symmetric modes contains additional sidebands in  $2\Omega$  ( $\Omega$  is the Rabi frequency) with amplitude dependent on  $(a/\Omega)^2$ , where  $a$  is a parameter dependent on  $r_{12}$ . The antisymmetric part of the spectrum has no additional sidebands in  $2\Omega$ . For small distances  $r_{12}$  ( $a = 1$ ) our results for the symmetric modes are identical with those of Agarwal et al. apart from the so-called scaling factor. For large distances  $r_{12}$  ( $a = 0$ ) the spectra of the symmetric and antisymmetric modes are identical with the well-known one-atom spectrum.

### 1. Introduction

Recently, Carmichael [1] and Agarwal et al. [2] have confirmed an earlier suggestion of Senitzky [3] that the resonance fluorescence spectrum of a cooperative system of atoms should exhibit additional sidebands at  $\omega - \omega_0 = \pm 2\Omega$ , where  $\Omega$  is the Rabi frequency. As it has been shown [1,2], these additional peaks should be distinguishable in experiment at extremely large  $\Omega$ , while at moderately large  $\Omega$  a cooperative system of two atoms has the three-peak structure of the resonance fluorescence spectrum well known from the one-atom theory [4-7]. The numerical calculations of Carmichael [1] and Agarwal et al. [2] are based on the solution of the master equation describing the evolution of a cooperative system under the influence of the laser field in terms of the collective variables. Agarwal et al. [2] have also obtained an exact analytical expression for the two atom resonance fluorescence spectrum with a polynomial of the fifth order in the denominator. From this expression, an approximate resonant structure has been extracted proving the existence of additional sidebands as well as some nonresonant terms. In the master equation approach used in refs. [1] and [2], a strong cooperativity of the system is assumed and no possibility exists to discuss a dependence of the spectrum on the distance between the atoms. Mavroyannis, using the Green function approach, has calculated approximately two-atom resonance fluorescence spectra without [8] and with [9] additional sidebands, depending on the approximations used. Her results include a dipole-dipole type of interaction between the atoms. The role of the dipole-dipole interaction in the resonance fluorescence spectrum has also been discussed by Freedhoff [10] in the dressed-atom description. Steudel and Richter [11] have discussed the radiation properties of an incoherently pumped two-atom system within the framework of Lehmburg's [12] master equation. Their discussion includes the dependence of the radiation properties of such a system on the interatomic distance. The master equation approach has been also extensively discussed by Agarwal [13].

In this paper, we derive the resonance fluorescence spectrum of a two-atom system coherently driven by a resonant laser field, for different interatomic distances. To this aim, we have adapted Lehmburg's [12] approach to the case of coherent pumping, and have obtained a closed system of 15 equations for atomic correlation func-

tions which has been solved by means of the Laplace transform method. According to these solutions as well as the quantum-regression theorem we have calculated the resonance fluorescence spectrum of the system dependent on the interatomic distance. Unfortunately, this expression involves a sixth-order polynomial in the denominator, the roots of which cannot be found strictly. In the strong field limit, however, the approximate roots of this polynomial can be found and we give an approximate analytical formula revealing the resonant structure of the spectrum.

## 2. Time-evolution of atomic variables

Within the framework of our model we consider two identical two-level atoms, distant by  $r_{12}$ , in the field of a strong resonance laser beam. We assume electric dipole transitions between the levels of either atom. For simplicity, the laser frequency  $\omega$  is assumed as exactly equal to the atomic transition frequency  $\omega_0$  i.e. detuning is zero. The strong pumping laser beam is moreover assumed to be in the coherent state  $|\alpha\rangle$ . On these assumptions, application of Lehmbert's [12] master equation approach leads to the following equations of motion describing the time evolution of the pseudo-spin operators of an individual atom (we have ignored here level shifts as well as dipole-dipole interaction):

$$\begin{aligned} \dot{S}_1^+ &= -\frac{1}{2}\gamma S_1^+ - i\Omega S_1^z + \gamma_{12} S_2^+ S_1^z, & \dot{S}_1^- &= -\frac{1}{2}\gamma S_1^- + i\Omega S_1^z + \gamma_{12} S_1^z S_2^-, \\ \dot{S}_1^z &= -\gamma(S_1^z + \frac{1}{2}) + \frac{1}{2}i\Omega(S_1^+ - S_1^-) - \frac{1}{2}\gamma_{12}(S_1^+ S_2^- + S_2^+ S_1^-), \end{aligned} \quad (1)$$

where  $S_1^+$  and  $S_1^- = [S_1^+]^\dagger$  are operators raising and lowering the energy of atom 1 and  $S_1^z$  describes its energy.

The equations for the operators of the other atom are of the same form as (1), albeit with the interchange  $1 \leftrightarrow 2$  of the indices. The operators fulfil the well known commutation relations:

$$[S_i^+; S_i^-] = 2S_i^z, \quad [S_i^z; S_i^\pm] = \pm S_i^\pm, \quad i = 1, 2, \quad (2)$$

and the operators of the two atoms are here assumed to commute. In (1),  $\gamma$  is the Einstein coefficient  $A$ , whereas  $\gamma_{12}$ , which is dependent on  $r_{12}$ , describes the radiative interaction between the atoms and has the form [12]:

$$\gamma_{12} = \frac{3}{2}\gamma \left\{ \left[ 1 - (\boldsymbol{\mu} \cdot \mathbf{r}_{12})^2 \right] \frac{\sin k_0 \mathbf{r}_{12}}{k_0 \mathbf{r}_{12}} + \left[ 1 - 3(\boldsymbol{\mu} \cdot \mathbf{r}_{12})^2 \right] \left[ \frac{\cos k_0 \mathbf{r}_{12}}{(k_0 \mathbf{r}_{12})^2} - \frac{\sin k_0 \mathbf{r}_{12}}{(k_0 \mathbf{r}_{12})^3} \right] \right\}, \quad (3)$$

where  $k_0 = \omega_0/c$ .

If the atoms are far remote  $\gamma_{12} \rightarrow 0$ , whereas if they are close to each other  $\gamma_{12} \approx \gamma$ . If  $\gamma_{12} = \gamma$  the equations (1) are equivalent to the master equation used by Carmichael [1] and Agarwal et al. [2].  $\Omega$  is the Rabi frequency describing interaction of an individual atom with the laser beam.

The atomic operators of eq. (1) are slowly varying parts of the full operators:

$$\tilde{S}_1^+(t) = S_1^+(t) e^{i\omega_0 t}, \quad \tilde{S}_1^-(t) = S_1^-(t) e^{-i\omega_0 t}, \quad \tilde{S}_1^z(t) = S_1^z(t). \quad (4)$$

Defining collective operators

$$S^\pm = S_1^\pm + S_2^\pm \quad \text{and} \quad S^z = S_1^z + S_2^z, \quad (5)$$

one can transform equations (1) to the form:

$$\begin{aligned} \dot{S}^+ &= \frac{1}{2}(\gamma_{12} - \gamma)S^+ + i\Omega S^z + \gamma_{12} S^+ S^z, & \dot{S}^- &= \frac{1}{2}(\gamma_{12} - \gamma)S^- - i\Omega S^z + \gamma_{12} S^z S^-, \\ \dot{S}^z &= (\gamma_{12} - \gamma)(S^z + 1) + \frac{1}{2}i\Omega(S^+ - S^-) - \gamma_{12} S^+ S^-. \end{aligned} \quad (6)$$

For  $\gamma_{12} = \gamma$ , these equations simplify considerably and become equivalent [14] to the master equation used in

[1] and [2] to describe the time evolution of the density matrix of such a cooperative system. Both systems of equations (1) and (6) lead to a finite hierarchy of equations for the correlation functions of the system which can be solved by means of the Laplace transform method. In the case of two atoms we have a system of 15 equations. In our calculations we have solved the system generated by equations (1) for individual atoms rather than for collective variables (5). The system under consideration can be transformed into the following independent subsystems: three of dimension 3 and one of dimension 6:

$$dU/d\tau = AU, \quad dV/d\tau = BV, \quad dW/d\tau = CW, \quad dX/d\tau = DX + \alpha, \quad (7)$$

with

$$A = \begin{bmatrix} -\frac{1}{2}(1-a) & 4\beta & a \\ -\beta & -1 & 0 \\ 0 & -2\beta & -\frac{1}{2}(3-a) \end{bmatrix}, \quad B = \begin{bmatrix} -\frac{1}{2}(1-a) & a & 0 \\ 0 & -\frac{1}{2}(3-a) & 2\beta \\ -\beta & -2\beta & -1 \end{bmatrix},$$

$$C = \begin{bmatrix} -\frac{1}{2}(1+a) & a & 0 \\ 0 & -\frac{1}{2}(3+a) & -2\beta \\ -\beta & 2\beta & -1 \end{bmatrix}, \quad (7a)$$

$$D = \begin{bmatrix} -\frac{1}{2}(1+a) & 4\beta & 0 & 0 & a & 0 \\ -\beta & -1 & -a & 0 & 0 & 0 \\ -\beta & -a & -1 & 0 & 2\beta & 4a \\ -\beta & 0 & 0 & -1 & 2\beta & 0 \\ 0 & -2\beta & -2\beta & -2\beta & -\frac{1}{2}(3+a) & 8\beta \\ 0 & 0 & 0 & 0 & -\beta & -2 \end{bmatrix}$$

and

$$\alpha_i = -4\beta\delta_{1i}, \quad U_1 = S_1^- + S_1^+ - S_2^- - S_2^+, \quad U_2 = S_1^+ S_1^- - S_2^+ S_2^-,$$

$$U_3 = S_1^+ S_1^- S_2^- + S_1^+ S_2^+ S_1^- - S_2^+ S_2^- S_1^- - S_1^+ S_2^+ S_2^-,$$

$$V_1 = S_1^- - S_1^+ - S_2^- + S_2^+, \quad V_2 = S_1^+ S_1^- S_2^- - S_1^+ S_2^+ S_1^- - S_2^+ S_2^- S_1^- + S_2^+ S_1^+ S_2^-, \quad V_3 = S_2^+ S_1^- - S_1^+ S_2^-,$$

$$W_1 = S_1^- - S_1^+ + S_2^- - S_2^+, \quad W_2 = S_1^+ S_1^- S_2^- - S_1^+ S_2^+ S_1^- + S_2^+ S_2^- S_1^- - S_1^+ S_2^+ S_2^-, \quad W_3 = S_1^- S_2^- - S_1^+ S_2^+,$$

$$X_1 = S_1^+ + S_2^+ + S_1^- + S_2^-, \quad X_2 = S_1^+ S_1^- + S_2^+ S_2^-, \quad X_3 = S_1^+ S_2^- + S_2^+ S_1^-,$$

$$X_4 = S_1^+ S_2^+ + S_1^- S_2^-, \quad X_5 = S_1^+ S_1^- S_2^- + S_1^+ S_2^+ S_1^- + S_2^+ S_2^- S_1^- + S_1^+ S_2^+ S_2^-, \quad X_6 = S_1^+ S_2^+ S_1^- S_2^-.$$

We have introduced the notation

$$\tau = \gamma t, \quad \beta = \Omega/2\gamma, \quad a = \gamma_{12}/\gamma. \quad (8)$$

From (7) it is evident that  $U$  and  $V$  represent antisymmetric combinations of atomic operators while  $W$  and  $X$  are symmetric combinations. Non-zero steady-state solutions arise from the  $X$  quantities only. It is worth noting here that the steady-state solutions as calculated from (6) lead to expectation values of the atomic operators other than in the case  $a = 1$ , considered by Agarwal et al. [2]. This fact is connected with the  $S^2$  conservation breaking in our case. The determinant of the  $D$  matrix becomes zero as  $a = 1$ . This means a reduction of the dimension of

the system from 6 to 5 because of the linear dependence of the variables due to  $S^2$ -conservation. The reduced system is identical with that considered by Agarwal et al. [2]. Also the so-called scaling factor discussed previously [15] appears in the system with  $S^2$  conserved and does not appear in the steady-state solutions of (6), which have the form ( $a \neq 1$ ):

$$\begin{aligned} \langle X_1 \rangle_s &= -\frac{32\beta^3 + 4\beta(a+1)}{M}, & \langle X_2 \rangle_s &= \frac{32\beta^4 + 4\beta^2}{M}, & \langle X_3 \rangle_s &= \frac{4\beta^2}{M}, & \langle X_4 \rangle_s &= \frac{4\beta^2(a+1)}{M}, \\ \langle X_5 \rangle_s &= -\frac{16\beta^3}{M}, & \langle X_6 \rangle_s &= \frac{8\beta^4}{M}, \end{aligned} \quad (9)$$

where

$$M = 32\beta^4 + 8\beta^2 + \frac{1}{2}(1+a)^2.$$

Because the square of the total spin of the system can be expressed by the  $X$  operators as follows:

$$S^2 = 2 - X_2 + X_3 + 2X_6,$$

its steady-state expectation value, according to (9), is

$$\langle S^2 \rangle_s = 2 - 16\beta^4/M. \quad (9a)$$

The system reaches this steady-state value even for very small deviations of the parameter  $a$  from unity i.e. in the majority of real situations.

### 3. Resonance fluorescence spectrum

Using the Laplace transform method to solve the equations (6) and applying the quantum-regression theorem, we calculated the resonance fluorescence spectrum of the system under discussion which is the transform of a suitable combination of atomic correlation functions [12]:

$$P_R(z) = w_R \int_0^\infty d\tau e^{-z\tau} \lim_{t \rightarrow \infty} \sum_{\alpha, \beta=1}^2 \exp(ikR \cdot r_{\alpha\beta}) \langle S_\alpha^+(t) S_\beta^-(t+\tau) \rangle, \quad (10)$$

where

$$w_R = (3\gamma/8\pi) \sin^2 \Theta,$$

with  $\Theta$  the angle between the observation direction unit vector  $R$  and the atomic transition dipole moment  $\mu$ ,  $k = \omega/c$ , and  $r_{12}$  the vector connecting the two atoms.

The resonance fluorescence spectrum can easily be obtained from (10) by having recourse to the definition:

$$P_R(\omega) = \text{Re} P_R \left( i \frac{\omega - \omega_0}{\gamma} \right), \quad (11)$$

where  $\omega_0$  is the atomic transition frequency.

The result of our calculations for (10) is:

$$P_R(z) = \frac{1}{2} w_R [(1 - \cos kR \cdot r_{12}) P_-(z) + (1 + \cos kR \cdot r_{12}) P_+(z)], \quad (12)$$

with

$$P_-(z) = \frac{(z+1)(2z+3-a) \langle X_2 - X_3 - X_4 \rangle_s - 2\beta(2z+3-2a) \langle X_5 \rangle_s - 4a(z+1) \langle X_6 \rangle_s}{(z+1)(2z+1-a)(2z+3-a) + 8\beta^2(2z+3-2a)}$$

$$+ \frac{[(z+1)(2z+3-a) + 8\beta^2] \langle X_2 - X_3 + X_4 \rangle_s + 2a\beta \langle X_5 \rangle_s - 4a(z+1) \langle X_6 \rangle_s}{(z+1)(2z+1-a)(2z+3-a) + 8\beta^2(2z+1)}, \tag{13}$$

$$P_+(z) = \frac{[(z+1)(2z+3+a) + 8\beta^2] \langle X_2 + X_3 - X_4 \rangle_s - 2a\beta \langle X_5 \rangle_s + 4a(z+1) \langle X_6 \rangle_s}{(z+1)(2z+1+a)(2z+3+a) + 8\beta^2(2z+1)} + \frac{N(z)}{M(z)}, \tag{14}$$

where

$$N(z) = (z+1-a) \{ [4\beta^2(z+1)^2 - 2a\beta^2(z+2) + \frac{1}{4}(z+2)(z+1+a)[(z+1)(2z+3+a) + 16\beta^2]] \times [\langle X_2 + X_3 + X_4 \rangle_s - (2\beta/z) \langle X_1 \rangle_s] + \frac{1}{2}\beta \langle X_5 \rangle_s [(z+1)(z+2)(2z+3+a) - a(z+1-a)(z+2) + 16\beta^2(2z+3-a)] + a[(z+2)(z+1+a) - 16\beta^2] [\langle X_6 \rangle_s - \beta \langle X_5 \rangle_s] \} + 8a\beta^2 z [\langle X_6 \rangle_s - \beta \langle X_5 \rangle_s], \tag{15}$$

$$M(z) = (z+1)(z+2)(z+1-a)(z+1+a)[z^2 + (2+a)z + \frac{1}{4}(1+a)(3+a)] + 2\beta^2(z+1-a)[2(z+1+a)(2z+1+a)(2z+3-a) + 2a^2(2z+1+a) - a(z+2)(5z+2a+4) + (z+1)(z+2)(2z+3+a)] + 32\beta^4 [2z^2 + (5-2a)z + (1-a)(3+a)]. \tag{16}$$

In (15), the quantities  $\langle X_1 \rangle_s, \langle X_2 \rangle_s, \dots$  are given by (9).

$P_-(z)$  represents the antisymmetric part of the spectrum, and  $P_+(z)$  the symmetric part. The antisymmetric part of the spectrum does not appear in the case of strong cooperativity when the atoms are very close together but the symmetric part has its maximum. If we put  $a = 1$  in (14) and replace the steady-state solutions (9) by the appropriate steady-state solutions for a strongly cooperative system, our spectrum goes over into the spectrum of Agarwal et al. [2].

In the strong field limit  $\beta \gg 1$  the approximate roots of the denominators in (13) and (14) can be found and the analytical formula describing the resonance structure of the spectrum can be obtained. Our calculations, up to terms of the order  $1/\beta^2$  give the following formulas for the anti-symmetric and symmetric parts of the spectrum (incoherent part):

$$P_-(\omega) = \text{Re} P_- \left( i \frac{\omega - \omega_0}{\gamma} \right) = n \left\{ \left( 1 - \frac{a^2 - 2a - 2}{16\beta^2} \right) \frac{1}{\nu^2 + \frac{1}{4}} + \left( 1 - \frac{a+1}{8\beta^2} \right) \left[ \frac{\frac{3}{4}}{(\nu - 2\beta)^2 + \frac{9}{16}} + \beta \rightarrow -\beta \right] + \left[ -\frac{5}{8\beta} \frac{(\nu - 2\beta)}{(\nu - 2\beta)^2 + \frac{9}{16}} + \beta \rightarrow -\beta \right] + \frac{a^2}{16\beta^2} \left[ \frac{\frac{1}{2}(\frac{5}{2} - a)}{(\nu - 2\beta)^2 + \frac{1}{4}(\frac{5}{2} - a)^2} + \beta \rightarrow -\beta \right] \right\}, \tag{17}$$

$$P_+(\omega) = \text{Re} P_+ \left( i \frac{\omega - \omega_0}{\gamma} \right) = n \left\{ \left( 1 - \frac{a^2 + 2a - 2}{16\beta^2} \right) \frac{1}{\nu^2 + \frac{1}{4}} + \left( 1 - \frac{2a^2 - a + 1}{8\beta^2} \right) \left[ \frac{\frac{3}{4}}{(\nu - 2\beta)^2 + \frac{9}{16}} + \beta \rightarrow -\beta \right] + \left[ -\frac{5}{8\beta} \frac{(\nu - 2\beta)}{(\nu - 2\beta)^2 + \frac{9}{16}} + \beta \rightarrow -\beta \right] + \frac{a^2}{16\beta^2} \left[ \frac{\frac{1}{2}(\frac{5}{2} + a)}{(\nu - 2\beta)^2 + \frac{1}{4}(\frac{5}{2} + a)^2} + \beta \rightarrow -\beta \right] + \frac{a^2}{16\beta^2} \left[ \frac{(\frac{3}{2} + a)}{(\nu - 4\beta)^2 + (\frac{3}{2} + a)^2} + \beta \rightarrow -\beta \right] + \frac{a^2}{32\beta^2 \Delta} \left[ \frac{6a^2 - 5a - \frac{1}{4} + (7-6a)\Delta}{\nu^2 + \frac{1}{16}(5-2a+4\Delta)^2} - \frac{6a^2 - 5a - \frac{1}{4} - (7-6a)\Delta}{\nu^2 + \frac{1}{16}(5-2a-4\Delta)^2} \right] \right\}, \tag{18}$$

where, for simplicity, we have used the notation:

$$n = \frac{1}{4}(1 - 1/4\beta^2), \quad \nu = (\omega - \omega_0)/\gamma, \quad \Delta = \frac{1}{4}(12a^2 - 4a + 1)^{1/2}. \tag{19}$$

#### 4. Discussion and conclusions

The formulas (17) and (18) together with (12) give the resonant structure as well as the dependence on interatomic distance of the resonance fluorescence spectrum of two atoms coherently driven by a strong resonant laser field. Because  $a = \gamma_{12}(r_{12})/\gamma$  and the dependence of  $\gamma_{12}(r_{12})$  on  $r_{12}$  can be given explicitly [12], our formulas describe the complete dependence of the spectrum on the interatomic distance  $r_{12}$  as well as the angular dependence for both symmetric and antisymmetric modes. These formulas hold for the whole range of  $r_{12}$ . If the atoms are far apart,  $a \rightarrow 0$  and our spectrum reproduces the well known three-peak spectrum of one atom [4–7] except for a factor 2 standing for two atoms. In the case of very short interatomic distances,  $a \approx 1$  and the spectrum goes over into the one obtained by Agarwal et al. [2], apart from the factor  $n$  which has to be replaced by  $\frac{1}{3}(1 - 1/3\beta^2)$  in connection with the so-called scaling factor [15]. As already mentioned above, this scaling factor appears only in the strictly cooperative case when  $a = 1$  and even small deviations of  $a$  from unity lead to our factor  $n$ . This fact is due to  $S^2$  conservation breaking and possible transitions between triplet and singlet states. Only the symmetric part of the spectrum has the additional sideband displaced from the central peak at double Rabi frequency the amplitude of which is proportional to  $(a/\beta)^2$  and tends to zero as the interatomic distance increases. This result is in agreement with Mavroyannis [9]. Both the antisymmetric and symmetric parts have nonresonant terms which are independent of  $a$  and exist even in the one-atom case. There are also additional contributions to the central line. Also, the widths of some lines depend on the degree of cooperativity  $a$  and change considerably with the change in  $a$ , as it is evident from (17) and (18).

For weak laser fields formulas (17) and (18) fail to hold, in contrast to the formulas (13) and (14) which are valid for any strength of the field and with the use of which the spectrum can easily be plotted with the help of a computer. Although the solutions (13)–(16) are rather complicated, they present the advantage of being exact solutions, obtained without any decorrelation of the correlation functions in question.

#### References

- [1] H.J. Carmichael, Phys. Rev. Lett. 43 (1979) 1106.
- [2] G.S. Agarwal, R. Saxena, L.M. Narducci, D.H. Feng and R. Gilmore, Phys. Rev. A21 (1980) 257.
- [3] I.R. Senitzky, Phys. Rev. Lett. 40 (1978) 1334.
- [4] B.R. Mollow, Phys. Rev. 188 (1969) 1969.
- [5] H.J. Carmichael and D.F. Walls, J. Phys. B9 (1976) 1199.
- [6] H.J. Kimble and L. Mandel, Phys. Rev. A13 (1976) 2123.
- [7] C. Cohen-Tannoudji and S. Reynaud, J. Phys. B10 (1977) 345.
- [8] C. Mavroyannis, Phys. Rev. A18 (1978) 185.
- [9] C. Mavroyannis, Optics Comm. 33 (1980) 42.
- [10] H.S. Freedhoff, Phys. Rev. A19 (1979) 1132.
- [11] H. Steudel and Th. Richter, Ann. Physik, Leipzig 35 (1978) 122.
- [12] R.H. Lehmburg, Phys. Rev. A2 (1970) 883.
- [13] G.S. Agarwal, Springer Tracts in Modern Physics, Vol. 70 (Springer, Berlin, 1974).
- [14] H.J. Carmichael, preprint.
- [15] A.S. Jahangir Amin and J.G. Cordes, Phys. Rev. A18 (1978) 1298.