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The effects of detuning on the quantum theory of an inhomogeneously broadened laser

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Abstract. The quantum theory of a laser developed by Scully and Lamb is reformulated to include the effects of atomic motion and detuning. The equations are solved to fourth order in the atom-field coupling constant. An approximate treatment of the mode structure is shown to reproduce the results of Lamb's semi-classical theory in the Doppler limit. In addition the photon distribution may be studied as a function of the detuning. The width of the photon distribution is found to increase monotonically with increasing detuning.

1. Introduction

In a previous paper (Riska and Stenholm 1970) we generalized the quantum theory for a laser developed by Scully and Lamb (1967) to the case where the active atoms are moving. The treatment was, however, limited to the case of resonance between the lasing cavity mode and the atomic transition sustaining the oscillations. We found that the atomic motion broadened the photon distribution as compared with the case of stationary atoms. In both our paper and that of Scully and Lamb the atoms are supposed to see only an average electromagnetic field. This assumption is more reasonable in the case of moving atoms, when an excited atom travels over several wavelengths of the electromagnetic field during its lifetime. In a letter (Riska and Stenholm 1969) we have shown that taking into account the spatial structure of the cavity mode in the quantum theory of Scully and Lamb we obtain an intensity exactly equal to the one given by semi-classical theory without atomic motion (Lamb 1964).

The effects of the atomic velocity distribution are most easily seen experimentally when the laser is detuned off resonance. An atom moving through the standing wave field sees two Doppler-shifted frequency components. When one of these is in resonance with the atomic transition a strong interaction takes place between the field and the atom. Atoms with Doppler shifts that compensate the detuning saturate strongly. The effect is called hole burning in the population inversion (Bennett 1962). This paper includes the effects of atomic motion and detuning in a model by Scully and Lamb (1967). The exact expressions are given but in order to obtain analytical results we perform a perturbation expansion in the field intensity. A certain neglect of detailed mode structure is introduced and the expression for the intensity given by Lamb (1964) is obtained. In addition we obtain the photon distribution as a function of detuning.

In § 2 of the present paper we describe the laser model and in § 3 we obtain the equation of motion for the density matrix of the radiation field coupled to the atoms. The photon distribution and the intensity are calculated in § 4 and the results are discussed in § 5.

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2. The model

The quantum theory of the laser, developed by Scully and Lamb (1967), is based on the same ideas as the semi-classical theory of Lamb (1964). The laser is assumed to consist of a highly selective optical cavity of length L . The pumping mechanism is described as injection of active two-level atoms in the upper level $|a\rangle$ of the pair $|a\rangle, |b\rangle$ at random times with an average rate r_a . The intensity of the electromagnetic field in the laser cavity is assumed large enough to permit the atoms to interact with the field independently of each other until they decay to lower levels $|c\rangle$ and $|d\rangle$ with rates γ_a and γ_b respectively. The loss mechanism is taken into account in a similar way by atoms introduced with a rate r_β in the lower level of two very broad levels $|\alpha\rangle$ and $|\beta\rangle$, which rapidly decay to levels $|\gamma\rangle$ and $|\delta\rangle$ with rates γ_α and γ_β . Because of the rapid decay of the non-resonant states, the loss mechanism can be treated as a linear process.

We now generalize this model to the case of moving atoms. We assume that, at random times, atoms in the state $|a\rangle$ are injected into the cavity at the point \mathbf{z} with the velocity v at a rate† $\lambda_a(\mathbf{z}, v)$. We treat the cavity as one-dimensional, neglecting the transverse variation of the electromagnetic mode (Lamb 1964). We assume the injection rate $\lambda_a(\mathbf{z}, v)$ to vary only slightly over the cavity length and neglect its \mathbf{z} -dependence. Assuming a Maxwellian velocity distribution for the injected atoms, we can then write $\lambda_a(\mathbf{z}, v)$ as

$$\lambda_a(\mathbf{z}, v) = \frac{r_a}{L} W(v) = \left(\frac{r_a}{Lu\sqrt{\pi}} \right) \exp\left(\frac{-v^2}{u^2} \right) \quad (1)$$

where u is the width of the velocity distribution and r_a is the total injection rate.

3. The equation of motion for the density matrix

The Hamiltonian $\hbar H$ of the interaction between an injected atom with a level difference $\hbar\omega = E_a - E_b$ and a cavity mode of frequency Ω is (in the dipole approximation)

$$H = \Omega a^+ a + (E_b/\hbar) + \frac{1}{2}\omega(1 + \sigma^z) + g\sqrt{2} \sin\{Kz(t)\}(a^+ \sigma + a \sigma^+). \quad (2)$$

Here a^+ and a are the photon creation and annihilation operators and σ^+ and σ are the raising and lowering operators between the states $|a\rangle, |b\rangle$. The population inversion operator σ^z is defined as the commutator of σ^+ and σ . The factor $\sqrt{2}$ is introduced in order to have the same coupling constant g as Scully and Lamb (see Riska and Stenholm 1969). The factor $\sin\{Kz(t)\}$ arises because of the standing-wave structure of the cavity mode; the wave number K is defined as Ω/c and $z(t)$ gives the instantaneous position of the atom in the cavity at time t . The atom injected at the point z_0 at time t_0 with velocity v is at the later time t at (neglecting the effects of atomic collisions)

$$z(t) = z_0 + v(t - t_0). \quad (3)$$

The density matrix of the coupled system of the field and the atom injected at t_0 has the elements

$$\rho_{\alpha n, \beta n'} = \langle \alpha n | \rho | \beta n' \rangle \quad (4)$$

where $|\alpha, n\rangle$ is a state with n photons and the atom in the state $|\alpha\rangle$, being one of:

† This means that we consider an ensemble of atoms introduced at \mathbf{z} with a distribution of velocities—cf. Riska and Stenholm (1970).

$|a\rangle$, $|b\rangle$, $|c\rangle$ or $|d\rangle$. In order to determine the photon distribution we have to obtain the equation of motion for the diagonal elements ρ_{nn} , where the atomic variables of (4) have been traced away. The equations are calculated by the method of Scully and Lamb (1967) with the decay treated in a Wigner-Weisskopf approximation. We obtain

$$\dot{\rho}_{an,an} = -\gamma_a \rho_{an,an} - iV(t)(\rho_{bn+1,an} - \rho_{an,bn+1}) \quad (5a)$$

$$\dot{\rho}_{an,bn+1} = -(\gamma_{ab} + i\Delta)\rho_{an,bn+1} - iV(t)(\rho_{bn+1,bn+1} - \rho_{an,an}) \quad (5b)$$

$$\rho_{bn+1,an} = (\rho_{an,bn+1})^* \quad (5c)$$

$$\dot{\rho}_{bn+1,bn+1} = -\gamma_b \rho_{bn+1,bn+1} - iV(t)(\rho_{an,bn+1} - \rho_{bn+1,an}) \quad (5d)$$

with

$$\gamma_{ab} = \frac{1}{2}(\gamma_a + \gamma_b)$$

$$\Delta = \omega - \Omega$$

and

$$V(t) = g\sqrt{2(n+1)^{1/2}} \sin[K\{z_0 + v(t-t_0)\}]. \quad (6)$$

In addition we obtain

$$\rho_{cn,cn}(t_0 + T) = \gamma_a \int_{t_0}^{t_0+T} dt' \rho_{an,an}(t') \quad (7a)$$

$$\rho_{dn+1,dn+1}(t_0 + T) = \gamma_b \int_{t_0}^{t_0+T} dt' \rho_{bn+1,bn+1}(t'). \quad (7b)$$

The initial condition is $\rho_{an,an}(t_0) = \rho_{nn}(t_0)$ and all other matrix elements equal to zero at t_0 .

In order to construct a coarse-grained time derivative for the density matrix of the radiation field we have to determine the change in ρ_{nn} due to the injection of one active atom. From equations (5) it follows that the elements $\rho_{an,an}$ and $\rho_{bn,bn}$ go to zero exponentially with time because of spontaneous decay† to the lower levels $|c\rangle$ and $|d\rangle$, and consequently we can write

$$\delta\rho_{nn} = \rho_{cn,cn}(t_0 + T) + \rho_{dn,dn}(t_0 + T) - \rho_{nn}(t_0) \quad (8)$$

provided $T \gg \gamma_{ab}^{-1}$. Even if equations (5) can be solved exactly by matrix methods and the expressions (7) in principle are known after one integration, the result has not been obtained as an analytical expression for $\delta\rho_{nn}$ because of the complicated structure of $\rho_{an,an}$ and $\rho_{bn,bn}$. We therefore resort to a perturbation expansion of the solution.

The general term of the iterative solution of equation (5) may be written down conveniently in matrix notation (see Appendix 1), but for the calculation of the terms, diagonal in the field, to fourth order, the straightforward method used by Lamb (1964) for solving a similar system of equations is more practical.

The matrix elements diagonal in n are to zeroth, second and fourth order:

$$\rho_{an,an}^{(0)}(t) = \rho_{nn}(t_0) \exp\{-\gamma_a(t-t_0)\} \quad (9a)$$

$$\rho_{bn+1,bn+1}^{(0)}(t) = 0 \quad (9b)$$

† The exact solution of equations (5) will contain a factor $\exp(-\gamma_{ab}t)$ —see Appendix 1.

$$\begin{aligned} \rho_{an,an}^{(2)}(t) &= - \int_{t_0}^t dt_2 \int_{t_0}^{t_2} dt_1 V(t_2)V(t_1)\rho_{an,an}^{(0)}(t_1) \\ &\quad \times \exp\{(\gamma_{ab} - i\Delta)(t_1 - t_2) + \gamma_a(t_2 - t) + \text{c.c.}\} \end{aligned} \quad (9c)$$

$$\begin{aligned} \rho_{bn+1,bn+1}^{(2)}(t) &= \int_{t_0}^t dt_2 \int_{t_0}^{t_2} dt_1 V(t_2)V(t_1)\rho_{an,an}^{(0)}(t_1) \\ &\quad \times \exp\{(\gamma_{ab} - i\Delta)(t_1 - t_2) + \gamma_b(t_2 - t)\} + \text{c.c.} \end{aligned} \quad (9d)$$

$$\begin{aligned} \rho_{an,an}^{(4)}(t) &= - \int_{t_0}^t dt_2 \int_{t_0}^{t_2} dt_1 V(t_2)V(t_1)\{\rho_{an,an}^{(2)}(t_1) - \rho_{bn+1,bn+1}^{(2)}(t_1)\} \\ &\quad \times \exp\{(\gamma_{ab} - i\Delta)(t_1 - t_2) + \gamma_a(t_2 - t)\} + \text{c.c.} \end{aligned} \quad (9e)$$

$$\begin{aligned} \rho_{bn+1,bn+1}^{(4)}(t) &= \int_{t_0}^t dt_2 \int_{t_0}^{t_2} dt_1 V(t_2)V(t_1)\{\rho_{an,an}^{(2)}(t_1) - \rho_{bn+1,bn+1}^{(2)}(t_1)\} \\ &\quad \times \exp\{(\gamma_{ab} - i\Delta)(t_1 - t_2) + \gamma_b(t_2 - t)\} + \text{c.c.} \end{aligned} \quad (9f)$$

From the structure of equations (9) we see that in order to include saturation effects the fourth-order terms cannot be neglected. We can now use equations (7), (8) and (9) to write the change in the density matrix as

$$\begin{aligned} \delta\rho_{nn} &= \gamma_a \int_0^T d\tau \rho_{nn}(t_0) \exp(-\gamma_a\tau) + \gamma_a \int_0^T d\tau \rho_{an,an}^{(2)}(\tau) + \gamma_a \int_0^T d\tau \rho_{an,an}^{(4)}(\tau) \\ &\quad + \gamma_b \int_0^T d\tau \rho_{bn,bn}^{(2)}(\tau) + \gamma_b \int_0^T d\tau \rho_{bn,bn}^{(4)}(\tau) - \rho_{nn}(t_0). \end{aligned} \quad (10)$$

As the density matrix elements go to zero exponentially with time we may replace the upper limit of integration T by infinity. Performing the integrations in equation (10) we split $V(t)$ into its exponential terms

$$V(t) = \frac{g\sqrt{2}}{2i} (n+1)^{1/2} \exp[iK\{z_0 + v(t-t_0)\}] + \text{c.c.} \quad (11)$$

All integrals are then reduced to the form

$$\int_0^\infty d\tau_n \int_0^{\tau_n} d\tau_{n-1} \dots \int_0^{\tau_2} d\tau_1 \exp\left(\sum_{m=1}^n \alpha_m \tau_m\right)$$

which equals (assuming convergence at the upper limit—see Appendix 2)

$$(-1)^n (\alpha_n)^{-1} (\alpha_n + \alpha_{n-1})^{-1} \dots \left(\sum_{m=1}^n \alpha_m\right)^{-1}.$$

In order to find the macroscopic change in ρ_{nn} due to the atoms introduced during a time Δt we have to calculate

$$\Delta\rho_{nn} = \Delta t \int_{-\infty}^\infty dv \int_0^L dz \lambda_a(z, v) \delta\rho_{nn}. \quad (12)$$

Dividing (12) by Δt we get a coarse-grained time derivative of $\rho_{nn}(t)$.

In calculating the fourth-order terms from equations (9e) and (9f), we replace the second-order terms by their spatial averages over the cavity (i.e. we neglect

terms containing the factors $\exp(\pm i2Kz_0)$ which should be a good approximation for moving atoms. The approximation is not unavoidable (see Appendix 3) but facilitates the analytical work and turns out to agree with earlier results in the Doppler limit. By performing the t and z_0 integrations we obtain

$$\begin{aligned} \frac{d\rho_{nn}(t)}{dt} = & -g^2(n+1) \frac{\gamma_{ab}r_a}{\gamma_a} \left[\int dv W(v) \left\{ \frac{1}{\gamma_{ab}^2 + (Kv + \Delta)^2} + \frac{1}{\gamma_{ab}^2 + (Kv - \Delta)^2} \right\} \right. \\ & \left. - 2g^2(n+1) \frac{\gamma_{ab}^2}{\gamma_a\gamma_b} \int dv W(v) \left\{ \frac{1}{\gamma_{ab}^2 + (Kv + \Delta)^2} + \frac{1}{\gamma_{ab}^2 + (Kv - \Delta)^2} \right\}^2 \right] \\ & \times \rho_{nn}(t) + g^2n \frac{\gamma_{ab}r_a}{\gamma_a} \left[\int dv W(v) \left\{ \frac{1}{\gamma_{ab}^2 + (Kv + \Delta)^2} + \frac{1}{\gamma_{ab}^2 + (Kv - \Delta)^2} \right\} \right. \\ & \left. - 2g^2n \frac{\gamma_{ab}^2}{\gamma_a\gamma_b} \int dv W(v) \left\{ \frac{1}{\gamma_{ab}^2 + (Kv + \Delta)^2} + \frac{1}{\gamma_{ab}^2 + (Kv - \Delta)^2} \right\}^2 \right] \rho_{n-1, n-1}(t). \end{aligned} \quad (13)$$

The velocity integrals can be performed approximately in the Doppler limit $Ku \gg \gamma_a, \gamma_b$ (Appendix 4). By adding linear loss terms as in our previous paper (Riska and Stenholm 1970), we get the equation of motion for ρ_{nn}

$$\begin{aligned} \frac{d\rho_{nn}}{dt} = & -A(n+1) \exp\left(\frac{-\Delta^2}{K^2u^2}\right) \left[1 - \frac{(n+1)B}{4A} \{1 + \gamma_{ab}^2(\gamma_{ab}^2 + \Delta^2)^{-1}\} \right] \rho_{n,n} \\ & + An \exp\left(\frac{-\Delta^2}{K^2u^2}\right) \left[1 - \frac{nB}{4A} \{1 + \gamma_{ab}^2(\gamma_{ab}^2 + \Delta^2)^{-1}\} \right] \rho_{n-1, n-1} \\ & - Cn\rho_{nn} + C(n+1)\rho_{n+1, n+1} \end{aligned} \quad (14)$$

with A, B and C defined in accordance with Riska and Stenholm (1970) as

$$A = \frac{2\sqrt{\pi}g^2r_a}{\gamma_aKu}, \quad B = \frac{4g^2}{\gamma_a\gamma_b} A, \quad C = \frac{4g^2r_b}{\gamma_b(\gamma_a + \gamma_b)}. \quad (15)$$

For the case of resonance ($\Delta = 0$) equation (14) may be compared with the equation derived earlier (Riska and Stenholm 1970)

$$\begin{aligned} \frac{d\rho_{nn}}{dt} = & -A(n+1) \left\{ 1 + \frac{B}{A}(n+1) \right\}^{-1/2} \rho_{nn} + An \left\{ 1 + \frac{B}{A}n \right\}^{-1/2} \rho_{n-1, n-1} - Cn\rho_{nn} \\ & + C(n+1)\rho_{n+1, n+1}. \end{aligned} \quad (16)$$

Expanding the square roots in (16) to first order in n we find the equation (14) with $\Delta = 0$.

4. Steady-state conditions

The steady-state solution $\dot{\rho}_{nn} = 0$ of equation (14) is obtained when

$$\rho_{nn} = \frac{A}{C} \exp\left(\frac{-\Delta^2}{K^2u^2}\right) \left[1 - \frac{nB}{4A} \{1 + \gamma_{ab}^2(\gamma_{ab}^2 + \Delta^2)^{-1}\} \right] \rho_{n-1, n-1} \quad (17)$$

from which we easily find the solution

$$\rho_{nn} = \left(\frac{A}{C}\right)^n \exp\left(\frac{-n\Delta^2}{K^2u^2}\right) \rho_{00} \prod_{\nu=0}^n \left[1 - \frac{\nu B}{4A} \{1 + \gamma_{ab}^2(\gamma_{ab}^2 + \Delta^2)^{-1}\}\right] \quad (18)$$

where ρ_{00} is the normalization constant.

The photon distribution is a monotonically decreasing function of n as long as the amplification factor A is below the value

$$A = \exp\left(\frac{\Delta^2}{K^2u^2}\right) C \quad (19)$$

which is the threshold condition for the laser with detuning. This agrees with the semi-classical result (Lamb 1964) and reduces to our earlier result $A = C$ when $\Delta = 0$. The value of the amplification A needed to reach the threshold (19) increases exponentially with detuning. When $A > \exp(\Delta^2/K^2u^2)C$, equation (18) implies that ρ_{nn} increases with increasing n up to a peak value \bar{n} , after which it decreases towards zero. According to (18) ρ_{nn} becomes negative for very large values of n , which is physically impossible. This is a result of our approximation which includes only fourth-order terms. The photon distribution (18) should not be used for $n \gg \bar{n}$ (the same situation has also been noted by Scully and Lamb 1967, § 4). The value \bar{n} can be found from the condition

$$1 = \frac{A}{C} \exp\left(\frac{-\Delta^2}{K^2u^2}\right) \left[1 - \frac{\bar{n} B}{4A} \{1 + \gamma_{ab}^2(\gamma_{ab}^2 + \Delta^2)^{-1}\}\right] \quad (20)$$

which gives

$$\bar{n} = 4 \frac{A}{B} \left\{1 - \frac{C}{A} \exp\left(\frac{\Delta^2}{K^2u^2}\right)\right\} / \{1 + \gamma_{ab}^2(\gamma_{ab}^2 + \Delta^2)^{-1}\}. \quad (21)$$

Setting $\Delta = 0$ and remembering that (21) is valid only when $A \simeq C$, we obtain agreement to lowest order in $\{(A/C) - 1\}$ with the result derived by Riska and Stenholm (1970)

$$\bar{n} = \frac{A}{B} \left\{\left(\frac{A}{C}\right)^2 - 1\right\} \simeq 2 \frac{A}{B} \left\{\left(\frac{A}{C}\right) - 1\right\} + \dots \quad (22)$$

As long as the laser is not too far above threshold, the result (21) is useful. It also agrees completely with the result derived by Lamb (1964) in the semi-classical theory. The dimensionless intensity parameter $(B/A)\bar{n}$ has been proved to equal the dimensionless intensity parameter I in the semi-classical theory (Riska and Stenholm 1969).

In figure 1 we plot the photon distribution for four values of the detuning. Increasing the detuning from $\Delta = 0$ first raises the peak of the distribution to higher photon numbers, i.e. larger intensities. A further increase in detuning leads to a drop in average photon number and the curve for $\Delta = 4\gamma_{ab}$ shows the approach to the black-body distribution which prevails when the laser is detuned so far that it stops oscillating. The linewidth is a monotonically increasing function of the detuning. An approximate expression for the width of the photon distribution can be obtained in the following way (as in Riska and Stenholm 1970): from (18) we have

$$\begin{aligned} \rho_{\bar{n}+k, \bar{n}+k} &= \left(\frac{A}{C}\right)^k \exp\left(\frac{-k\Delta}{K^2u^2}\right) \rho_{\bar{n}\bar{n}} \\ &\times \prod_{\nu=1}^k \left[1 - \frac{(\bar{n}+\nu) B}{4A} \{1 + \gamma_{ab}^2(\gamma_{ab}^2 + \Delta^2)^{-1}\}\right]. \end{aligned} \quad (23)$$

The halfwidth is obtained if one sets $\rho_{\bar{n}+k, \bar{n}+k} = \frac{1}{2}\rho_{\bar{n}, \bar{n}}$, which with (21) leads to

$$\frac{1}{2} = \prod_{v=1}^k \left[1 - \frac{v}{4} \frac{A}{C} \exp\left(\frac{-\Delta^2}{K^2 u^2}\right) \frac{B}{A} \{1 + \gamma_{ab}^2 (\gamma_{ab}^2 + \Delta^2)^{-1}\} \right]. \quad (24)$$

Assuming the last term to be small we expand and use (21) to obtain

$$k^2 = \frac{\bar{n}}{(A/C) \exp(-\Delta^2/K^2 u^2) - 1}. \quad (25)$$

In figure 2 we plot the linewidth given by equation (25) as a function of detuning for $A/C = 1.2$ and compare it with the values obtained from the photon distributions in figure 1. We see that equation (25) shows the correct trend of the linewidth even if the actual values are too low.†

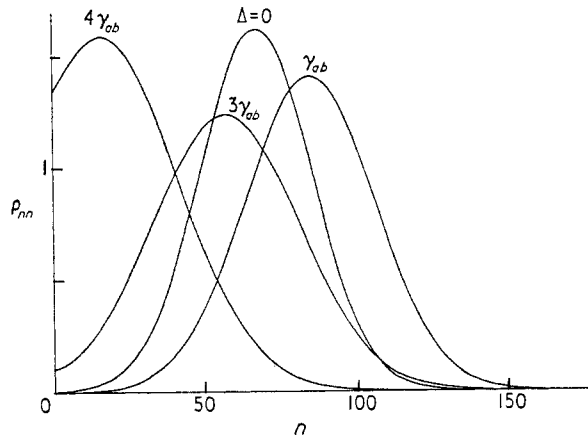


Figure 1. The photon distribution of equation (18) for four values of the detuning $\Delta = 0, \gamma_{ab}, 3\gamma_{ab}, 4\gamma_{ab}$. The parameter Ku is chosen to be $10\gamma_{ab}$ (the Doppler limit). The amplification factor $A/C = 1.2$ and the saturation parameter $B/A = 0.005$.

In figure 3 we compare the approximate photon distribution at resonance as given by equation (18) with the non-perturbative result obtained earlier (Riska and Stenholm 1970). At $A/C = 1.2$ we see that the perturbation approach gives an intensity, approximately 30% too small and a too narrow line. This is understandable as the perturbation theory neglects all processes where an injected atom interacts more than four times with the radiation field before its decay. For smaller values of A/C the agreement is better.

5. Discussion

The quantum theory of an inhomogeneously broadened laser is generalized to the case of a laser with detuning. In order to enable us to perform the calculations analytically we use a fourth-order perturbation expansion in the atom-radiation coupling constant. The standing-wave structure of the cavity mode is included into the formulation of the theory, but in the actual calculation of the nonlinear terms the mode structure is taken into account approximately only. This had been found to be a good

† We take this opportunity to correct an error in Riska and Stenholm (1970)—equation (59) should read: $\sigma^2/\sigma_{sL}^2 = 3/2 > 1$.

approximation for the case of resonance between the atoms and the cavity. The expression obtained for the electromagnetic field intensity agrees with the result given by Lamb (1964) in the Doppler limit. This may be seen as a justification for our approximation method, and our approach makes it possible to consider the exact perturbation expression if need be. This will certainly be the case when $\gamma_{ab} \simeq Ku$. The exact fourth-order terms with the mode structure included are given in Appendix 3.

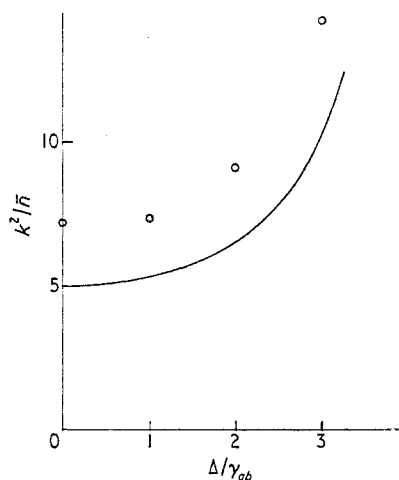


Figure 2. The approximate values of the width k^2/\bar{n} (full curve, calculated from equation (25)) are compared with those found from the photon distribution (points taken from the distributions in figure 1) for varying detuning. $A/C = 1.2$ and $B/A = 0.005$.

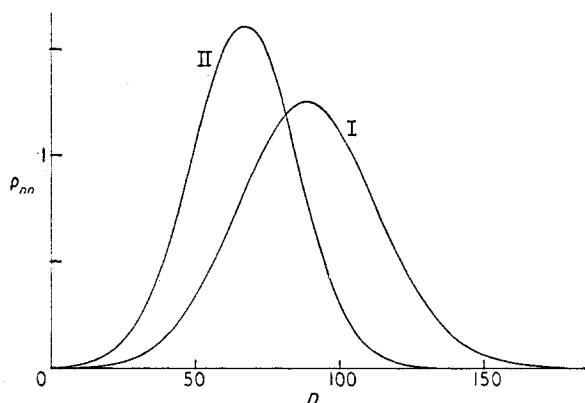


Figure 3. The exact photon distribution at resonance I compared with the approximate one II given in this paper (equation (18)) when we set $\Delta = 0$. The parameters $A/C = 1.2$ and $B/A = 0.005$.

Corrections to the expression (21) may readily be obtained from these. No specific quantum effects are to be expected in the intensity, and in the Doppler limit $Ku \gg \gamma_{ab}$ the quantitative changes introduced are hardly experimentally observable. In addition to the semi-classical expression for the intensity, the quantum theory gives the photon distribution in the laser. The width of this is found to increase with increasing detuning, finally to lose its meaning at the values of the detuning where the oscillations stop and the distribution goes over into a black-body one.

Finally our formulation enables one to obtain the results for any intensity from a straightforward evaluation of the solution to equations (5). This can be done numerically in the 4×4 matrix formulation of Appendix 1. We have, however, not been interested in obtaining the most general solution of the problem, but we have rather wanted to show the connections between the quantum theory and the semi-classical theory and to elucidate their differences. More detailed numerical work may preferably be performed in connection with the analysis of actual measurements.

Acknowledgments

One of us (D.O.R.) would like to thank the Oscar Öflund Foundation for financial support during the time of this work.

Appendix 1

The equations (5) can be written in matrix form as

$$\dot{\rho} = \{P + Q(t)\}\rho \quad (\text{A1.1})$$

where we have defined

$$\rho = \begin{pmatrix} \rho_{an,an} \\ \rho_{an,bn+1} \\ \rho_{bn+1,an} \\ \rho_{bn+1,bn+1} \end{pmatrix} \quad P = \begin{pmatrix} -\gamma_a & 0 & 0 & 0 \\ 0 & -(\gamma_{ab} + i\Delta) & 0 & 0 \\ 0 & 0 & -(\gamma_{ab} - i\Delta) & 0 \\ 0 & 0 & 0 & -\gamma_b \end{pmatrix}$$

$$Q(t) = \begin{pmatrix} 0 & iV(t) & -iV(t) & 0 \\ iV(t) & 0 & 0 & -iV(t) \\ -iV(t) & 0 & 0 & iV(t) \\ 0 & -iV(t) & iV(t) & 0 \end{pmatrix}. \quad (\text{A1.2})$$

The solution of (A1.1) is

$$\rho = \exp\left[\int_{t_0}^t dt' \{P + Q(t')\}\right] \rho(t_0). \quad (\text{A1.3})$$

Because of the complicated structure of equation (A1.3) it proves to be advantageous to use a perturbation expansion of the solution. For this we introduce an 'interaction representation' by writing $\rho = \exp\{P(t-t_0)\}\rho^I$, which in the usual way gives

$$\rho^I(t) = \rho^I(t_0) + \int_{t_0}^t dt_1 Q^I(t_1)\rho^I(t_1) \quad (\text{A1.4})$$

with

$$Q^I(t) = \exp\{-P(t-t_0)\}Q(t)\exp\{P(t-t_0)\}. \quad (\text{A1.5})$$

Iterating the equation (A1.4) and going back to $\rho(t)$ we find

$$\begin{aligned} \rho(t) &= \exp\{P(t-t_0)\}\rho(t_0) + \int_{t_0}^t dt_1 \exp\{P(t-t_1)\}Q(t_1)\exp\{P(t_1-t_0)\}\rho(t_0) \\ &+ \int_{t_0}^t dt_1 \int_{t_0}^{t_1} dt_2 \exp\{P(t-t_1)\}Q(t_1)\exp\{P(t_1-t_2)\}Q(t_2) \\ &\times \exp\{P(t_2-t_0)\}\rho(t_0) + \dots \end{aligned} \quad (\text{A1.6})$$

The zeroth order describes spontaneous decay without interaction with the laser field, the first-order term describes emission of a photon before decay and so forth. In this paper we include terms up to fourth order.

Appendix 2

The calculations of § 3 lead to integrals of the type

$$I = \int_0^\infty d\tau_n \int_0^{\tau_n} d\tau_{n-1} \dots \int_0^{\tau_2} d\tau_1 \exp\left(\sum_{m=1}^n \alpha_m \tau_m\right) \quad (\text{A2.1})$$

where $n = 2$ and 4 . By the substitution

$$\begin{aligned} x_{n-1} &= \tau_{n-1} \\ x_n &= \tau_n - \tau_{n-1} \end{aligned} \quad (\text{A2.2})$$

we write (A2.1) in the form

$$I = \left\{ \int_0^\infty dx_n \exp(\alpha_n x_n) \right\} \left[\int_0^\infty dx_{n-1} \int_0^{x_{n-1}} d\tau_{n-2} \dots \int_0^{\tau_2} d\tau_1 \right. \\ \left. \times \exp\left(\sum_{m=1}^{n-2} \alpha_m \tau_m \right) \exp\{(\alpha_n + \alpha_{n-1})x_{n-1}\} \right]. \quad (\text{A2.3})$$

Repeating this process we can write the integral

$$I = \left\{ \int_0^\infty dx_n \exp(\alpha_n x_n) \right\} \left[\int_0^\infty dx_{n-1} \exp\{(\alpha_n + \alpha_{n-1})x_{n-1}\} \dots \right. \\ \left. \times \left[\int_0^\infty dx_1 \exp\left\{ \left(\sum_{m=1}^n \alpha_m \right) x_1 \right\} \right] \right]. \quad (\text{A2.4})$$

Provided that for all $k \leq n$, $\text{Re} \sum_{m=k}^n \alpha_m < 0$ we get the result

$$I = (-1)^n (\alpha_n)^{-1} (\alpha_n + \alpha_{n-1})^{-1} \dots \left(\sum_{m=1}^n \alpha_m \right)^{-1}. \quad (\text{A2.5})$$

Appendix 3

The calculations in the text neglect the detailed mode structure in $\rho^{(2)}$ when introducing this into the calculation of $\rho^{(4)}$. An exact calculation of the term needed in $\Delta\rho_{nn}$ gives

$$\int_0^\infty d\tau \frac{1}{L} \int_0^L dz \rho_{an,an}^{(4)} = \frac{g^4(n+1)^2}{16\gamma_a^2} \left[\frac{8\gamma_{ab}^3}{\gamma_a\gamma_b} \left\{ \frac{1}{\gamma_{ab}^2 + (Kv + \Delta)^2} + \frac{1}{\gamma_{ab}^2 + (Kv - \Delta)^2} \right\}^2 \right. \\ - \frac{8Kv\gamma_{ab}(Kv + \Delta) - 2\gamma_b\{\gamma_{ab}^2 - (Kv + \Delta)^2\}}{(\gamma_b^2 + 4K^2v^2)\{\gamma_{ab}^2 + (Kv + \Delta)^2\}^2} \\ - \frac{8Kv\gamma_{ab}(Kv - \Delta) - 2\gamma_b\{\gamma_{ab}^2 - (Kv - \Delta)^2\}}{(\gamma_b^2 + 4K^2v^2)\{\gamma_{ab}^2 + (Kv - \Delta)^2\}^2} \\ - 2 \frac{8(Kv)^2\gamma_{ab} - 2\gamma_b\{\gamma_{ab}^2 - (K^2v^2 - \Delta^2)\}}{(\gamma_b^2 + 4K^2v^2)\{\gamma_{ab}^2 + (Kv + \Delta)^2\}\{\gamma_{ab}^2 + (Kv - \Delta)^2\}} \\ - \frac{8Kv\gamma_{ab}(Kv + \Delta) - 2\gamma_a\{\gamma_{ab}^2 - (Kv + \Delta)^2\}}{(\gamma_a^2 + 4K^2v^2)\{\gamma_{ab}^2 + (Kv + \Delta)^2\}^2} \\ - \frac{8Kv\gamma_{ab}(Kv - \Delta) - 2\gamma_a\{\gamma_{ab}^2 - (Kv - \Delta)^2\}}{(\gamma_a^2 + 4K^2v^2)\{\gamma_{ab}^2 + (Kv - \Delta)^2\}^2} \\ \left. - 2 \frac{8(Kv)^2\gamma_{ab} - 2\gamma_a\{\gamma_{ab}^2 - (K^2v^2 - \Delta^2)\}}{(\gamma_a^2 + 4K^2v^2)\{\gamma_{ab}^2 + (Kv + \Delta)^2\}\{\gamma_{ab}^2 + (Kv - \Delta)^2\}} \right]. \quad (\text{A3.1})$$

Our approximation eliminates all terms of (A3.1) but the first one in braces. This leads to considerable simplification and is shown to be equivalent to the results by Lamb (1964) and Riska and Stenholm (1970) in the Doppler limit, $Kv \gg \gamma$. Corrections may easily be calculated from (A3.1).

Appendix 4

In the Doppler limit $Ku \gg \gamma$ the velocity integrals in equation (13) are easily calculated:

$$\begin{aligned} \frac{1}{u\sqrt{\pi}} \int_{-\infty}^{+\infty} dv \frac{\exp(-v^2/u^2)}{\gamma_{ab}^2 + (Kv + \Delta)^2} &\simeq \frac{\exp(-\Delta^2/K^2u^2)}{Ku\sqrt{\pi}} \int_{-\infty}^{+\infty} dx \frac{1}{\gamma_{ab}^2 + x^2} \\ &= \frac{\sqrt{\pi}}{\gamma_{ab}Ku} \exp\left(-\frac{\Delta^2}{K^2u^2}\right) \end{aligned} \quad (\text{A4.1})$$

and

$$\begin{aligned} \frac{1}{u\sqrt{\pi}} \int_{-\infty}^{+\infty} dv \frac{\exp(-v^2/u^2)}{\{\gamma_{ab}^2 + (Kv + \Delta)^2\}^2} &\simeq \frac{\exp(-\Delta^2/K^2u^2)}{Ku\sqrt{\pi}} \int_{-\infty}^{+\infty} \frac{1}{(\gamma_{ab}^2 + x^2)^2} dx \\ &= \frac{1}{2} \frac{\sqrt{\pi}}{\gamma_{ab}^3Ku} \exp\left(\frac{-\Delta^2}{K^2u^2}\right). \end{aligned} \quad (\text{A4.2})$$

Integrals over the product of two Lorentzians can be calculated using the residue theorem:

$$\begin{aligned} \frac{2}{u\sqrt{\pi}} \int_{-\infty}^{+\infty} dv \frac{\exp(-v^2/u^2)}{\{\gamma_{ab}^2 + (Kv + \Delta)^2\}\{\gamma_{ab}^2 + (Kv - \Delta)^2\}} \\ \simeq \frac{2 \exp(-\Delta^2/K^2u^2)}{u\sqrt{\pi}} \int_{-\infty}^{+\infty} \frac{dv}{\{\gamma_{ab}^2 + (Kv + \Delta)^2\}\{\gamma_{ab}^2 + (Kv - \Delta)^2\}} \\ = \frac{\sqrt{\pi}}{Ku\gamma_{ab}^3} \exp\left(\frac{-\Delta^2}{K^2u^2}\right) \frac{\gamma_{ab}^2}{(\gamma_{ab}^2 + \Delta^2)}. \end{aligned} \quad (\text{A4.3})$$

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