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DOUBLE OPTICAL RESONANCE
IN A SYSTEM OF ATOMS

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Many theoretical and experimental studies[2-8] of the cooperative effects in the interaction of atoms and molecules with a laser field and the vacuum of radiation have been carried out since the early work on superradiance by Dicke[1]. In this letter we discuss the collective behaviour of N three-level atoms interacting with two resonant driving fields and an emitted field in the context of double optical resonance (see fig.1). The analytic expressions for the steady-state population distribution on the atomic levels and the collective spectrum of fluorescence are given.

In the cooperative limit $N \to \infty$ the atomic population distribution and fluorescence intensity have a discontinuous behaviour at the critical ratio of the driving field intensities.
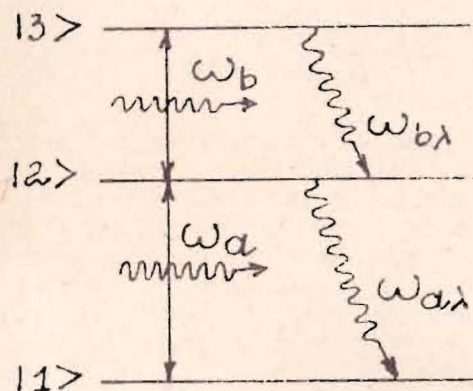
The N three-level atoms are assumed to be concentrated in a region small compared to the wavelength of all the relevant radiation modes. In the boson representation of atoms[9], when each atomic level is compared with boson variable, the master equation in the rotating frame with Markovian electric dipole and rotating wave approximations is of the form[2]

$$\frac{\partial \rho}{\partial t} = - i \Omega \left[(\cos a \, J_{12} + \sin a \, J_{23} + \text{H.C.}), \rho \right] - \gamma_a \, (J_{21} J_{12} \rho - 2 J_{12} \rho J_{21} + \rho J_{21} J_{12}) - \gamma_b \, (J_{32} J_{23} \rho - 2 J_{23} \rho J_{32} + \rho J_{32} J_{23}) = L \rho \, , \quad (1)$$

where ρ is the atomic density matrix, $2\gamma_a$ and $2\gamma_b$ are radiative spontaneous transition probabilities per unit time for a single atom to change from the level $|2\rangle$ to $|1\rangle$ and $|3\rangle$ to $|2\rangle$, respectively. $\Omega = (\Omega_1^2 + \Omega_2^2)^{1/2}$ and $\text{tg}\,a = \Omega_2 / \Omega_1$, where Ω_1 and Ω_2 are the Rabi frequencies for the atomic transitions from the level $|2\rangle$ to $|1\rangle$ and $|3\rangle$ to $|2\rangle$, respectively. $J_{ij} = C_i^{+} C_j$ $(i, j = 1, 2, 3)$,

where C_i and C_i^+ are the annihilation and creation boson operators for the atoms populated on the levels $|i>$. They satisfy the boson commutation relations

$$[C_i, C_j^+] = \delta_{ij}, \quad \text{so} \quad [J_{ij}, J_{i'j'}] = J_{ij'}\delta_{ji} - J_{i'j}\delta_{ij'}.$$

The exact solution of (1) for an arbitrary number of atoms is unknown. In the case of sufficiently intense Ω, so that

$$\Omega \gg N\gamma_a, \quad N\gamma_b, \tag{2}$$

it is possible to develop an approximation scheme that enables us to obtain analytic results.

After performing the canonical transformation

$$C_1 = -\frac{\sin a}{\sqrt{2}} Q_1 + \cos a\, Q_2 + \frac{\sin a}{\sqrt{2}} Q_3,$$

$$C_2 = \frac{Q_1}{\sqrt{2}} + \frac{Q_3}{\sqrt{2}}, \tag{3}$$

$$C_3 = -\frac{\cos a}{\sqrt{2}} Q_1 - \sin a\, Q_2 + \frac{\cos a}{\sqrt{2}} Q_3,$$

one can find that the Liouville operator L appearing in equation (1) splits into two components L_0 and L_1. The component L_0 is slowly varying in time whereas L_1 contains rapidly oscillating terms at frequencies 2Ω and 4Ω. For intense fields, it is reasonable to make the secular approximation, i.e., to retain only the slowly varying part [6]. Corrections to the results obtained in this fashion will be of an order of $(\gamma_a N/\Omega)^2$ or $(\gamma_b N/\Omega)^2$. The new master equation has the structure:

$$\frac{\partial \tilde{\rho}}{\partial t} = -i\Omega [\Delta_3, \tilde{\rho}] - \gamma_a \{[\frac{1}{4}\cos^2 a\, \Delta_3^2 + \frac{1}{2}\sin^2 a\,(R_{12}R_{21} + R_{32}R_{23}) +$$

$$+ \frac{\cos^2 a}{4}(R_{13}R_{31} + R_{31}R_{13})]\tilde{\rho} + [-\frac{1}{4}\cos^2 a\, \Delta_3 \tilde{\rho}\Delta_3 - \frac{1}{2}\sin^2 a\, \cdot$$

$$\cdot (R_{21}\tilde{\rho}R_{12} + R_{23}\tilde{\rho}R_{32}) - \frac{\cos^2 a}{4}(R_{13}\tilde{\rho}R_{31} + R_{31}\tilde{\rho}R_{13})] + \text{H.C.}\} -$$

$$- \gamma_b \{[\frac{1}{4}\sin^2 a\, \Delta_3^2 + \frac{1}{2}\cos^2 a\,(R_{21}R_{12} + R_{23}R_{32}) + \frac{\sin^2 a}{4}(R_{13}R_{31} + R_{31}R_{13})] \cdot$$

$$\cdot \tilde{\rho} + [-\frac{1}{4}\sin^2 a\, \Delta_3 \tilde{\rho}\Delta_3 - \frac{1}{2}\cos^2 a\,(R_{12}\tilde{\rho}R_{21} + R_{32}\tilde{\rho}R_{23}) -$$

$$- \frac{\sin^2 a}{4}(R_{13}\tilde{\rho}R_{31} + R_{31}\tilde{\rho}R_{13})] + \text{H.C.}\}. \tag{4}$$

here $R_{ij} = Q_i^+ Q_j$ $(i, j = 1, 2, 3)$; $\Delta_3 = R_{33} - R_{11}$; $\tilde{\rho} = U\rho U^+$, where U is a unitary operator representing the canonical transformation (3) [11].

The exact stationary solution of equation (4) is given by

$$\tilde{\rho} = \Omega^{-1} \sum_{R=0}^{N} x^{-R} \sum_{M=-R,-R+2,\ldots}^{R} |M,R><R,M|, \tag{5}$$

where

$$x = \frac{\gamma_a \sin^2 a}{\gamma_b \cos^2 a}, \quad \Omega = \frac{N+1-(N+2)x+x^{N+2}}{x^N \cdot (1-x)^2}.$$

$|M,R>$ is an eigenstate of the operators Δ_3, $R_{11} + R_{33}$ and the operator of total number of atoms $N = J_{11} + J_{22} + J_{33} = R_{11} + R_{22} + R_{33}$.

Using the canonical transformation (3) and solution (5), one can write the stationary population distribution for the atoms on the levels $|3>, |2>$ and $|1>$

$$N_3 \equiv <J_{33}>_s = <R_{11}>_s \sin^2 a + <R_{22}>_s \cos^2 a,$$

$$N_2 \equiv <J_{22}>_s = <R_{11}>_s = \frac{N - <R_{22}>_s}{2}, \tag{6}$$

$$N_1 \equiv <J_{11}>_s = <R_{11}>_s \cos^2 a + <R_{22}>_s \sin^2 a.$$

where

$$<R_{22}>_s = \frac{NX - (N+2)X^2 + (N+2)X^{N+2} - NX^{N+3}}{N+1-(2N+3)X + (N+2)X^2 + X^{N+2} - X^{N+3}}. \tag{7}$$

Here we denote by the symbol $<A>_s$ the steady-state value of an operator A.

In the one atom case $N = 1$ the expressions (6-7) reduce to the results of the work [12].

One can see from (6-7) that the atomic population distribution depends only on the ratio of the Rabi frequencies Ω_2/Ω_1. For the case of $X = 1$ (i.e., $\Omega_2/\Omega_1 = (\gamma_a/\gamma_b)^{1/2}$) the expression (6) reduces to $N_1 = N_2 = N_3 = N/3$. Thus, the three atomic levels are equally populated.
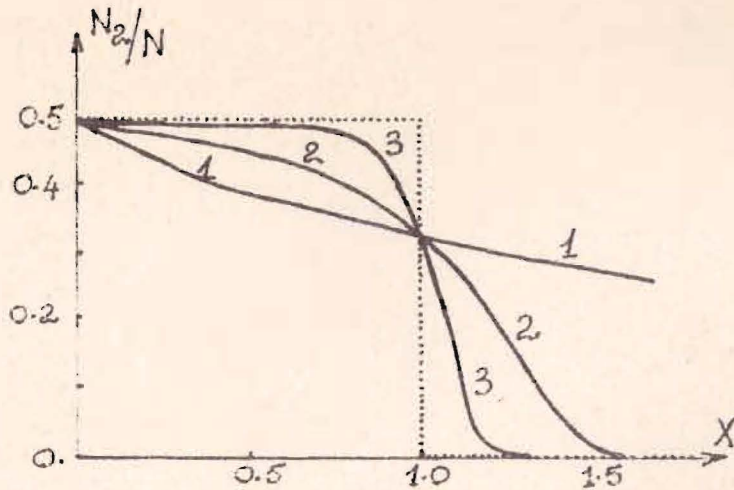
Fig.2. *Steady-state atomic population on the common level (per atom) as a function of* X. *The curves marked 1 to 3 correspond to* N = 1, 5, 50, *respectively. The dotted curve indicates the expected behaviour as* N → ∞.

The detailed behaviour for the population on the common level per one atom N_2/N is shown in fig.2, where N_2/N is plotted as a function of the parameter X. For all finite values of N one observes a smooth variation of N_2/N with X. For the cooperative limit N → ∞ the function N_2/N has a discontinuous behaviour at the critical point X = 1. This result is different from the critical behaviour of resonance fluorescence where the atomic system seems to exhibit a sharp transition reminiscent of a typical second-order phase transition[4]. The critical point X = 1 in the three-level case is invariant and does not depend on N. However, we note that in the limit N → ∞ it is also necessary to increase Ω so that the condition (2) is satisfied. One can also see from fig. 2 that N_2/N is zero (population trapping) not only in the case of $\gamma_b = 0$ (X → ∞), as in the one atom case[10] but in the case of X > 1 and N → ∞ too. Now we consider the collective behaviour of steady-state fluorescence spectrum, for example the fluorescence spectrum from the upper atomic transition. Using expressions (3,5) and the method of factorization by Compagno and Persico[7] one can find the first-order atomic correlation function (which is proportional to the field-amplitude correlation function) in the form

$$G_{32}^{(1)}(\tau) \equiv \lim_{t \to \infty} <J_{32}(t+\tau)J_{23}(t)> =$$

$$= \frac{1}{4}\sin^2 a <\Delta_3^2>_s \ell^{-\Gamma_1 \tau} + \cos^2 a <R_{21}R_{12}>_s \cdot$$

$$\cdot \ell^{-\Gamma_2 \tau}\cos(\Omega\tau) + \frac{1}{2}\sin^2 a <R_{31}R_{13}>_s \cdot \ell^{-\Gamma_3 \tau}\cos(2\Omega\tau),$$

(8)

where

$$<\Delta_3^2>_s = 2<R_{31}R_{13}>_s = \frac{1}{3}N(N+2) - \frac{2}{3}(N+1)<R_{22}>_s + \frac{1}{3}<R_{22}^2>_s,$$

$$<R_{21}R_{12}>_s = \frac{1}{2}(N+2)<R_{22}>_s - \frac{1}{2}<R_{22}^2>_s,$$

$$<R_{22}^2>_s = \frac{NX - 4X^2 - (N+2)X^3 + (N+2)^2 X^{N+2} - 2(N^2+2N-1)X^{N+3} + N^2 X^{N+4}}{(1-X)^2[N+1-(N+2)X+X^{N+2}]}$$

$$\Gamma_1 = \gamma_a + \gamma_b \sin^2 a + (\gamma_a \sin^2 a - \gamma_b \cos^2 a)<R_{22}>_s,$$

$$\Gamma_2 = \frac{\gamma_a}{2}(1+\sin^2 a) + \frac{\gamma_b}{2} - \frac{1}{2}(\gamma_a \sin^2 a - \gamma_b \cos^2 a)(N-2<R_{22}>_s),$$

$$\Gamma_3 = \frac{\gamma_a}{2}(2+\cos^2 a) + \frac{3}{2}\gamma_b \sin^2 a + (\gamma_a \sin^2 a - \gamma_b \cos^2 a)<R_{22}>_s.$$

Expression (8) yields the five-peaked structure of fluorescence spectrum. In the case of X = 1 the intensity of each line in (8) varies as N^2 while the width of each line is the same as in the single-atom spectrum. An analogous behaviour for the triplet of resonance fluorescence in the two-level system was given in[3,6,7]. The picture changes for the case of X ≠ 1. Take for example the case of X > 1 and N >> 1 so that $X^N >> N$. In this case the five-peak intensities of (8) are independent of N. The widths of the three lines located at the frequencies $\omega_{b\lambda} = \omega_b$, $\omega_b \pm 2\Omega$ are analogous with the width of the single-atom spectrum while the width of two lines located at frequencies $\omega_{b\lambda} = \omega_b \pm \Omega$ are proportional to N. In the cooperative limit N → ∞ the fluorescence intensity per atom $G_{32}^{(1)}(0)/N^2$, has a discontinuous behavuour at the critical point X = 1.

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СООБЩЕНИЯ, КРАТКИЕ СООБЩЕНИЯ, ПРЕПРИНТЫ И СБОРНИКИ ТРУДОВ КОНФЕРЕНЦИЙ, ИЗДАВАЕМЫЕ ОБЪЕДИНЕННЫМ ИНСТИТУТОМ ЯДЕРНЫХ ИССЛЕ-ДОВАНИЙ, ЯВЛЯЮТСЯ ОФИЦИАЛЬНЫМИ ПУБЛИКАЦИЯМИ.

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Двойной оптический резонанс в системе атомов

Обсуждено коллективное поведение N трехуровневых атомов, взаимодействующих с двумя резонансными внешними полями и полем излучения в смысле двойного оптического резонанса. Обсуждено также критическое поведение в коллективном пределе.

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Double Optical Resonance in a System of Atoms

The collective behaviour of N three-level atoms interacting with two resonant driving fields and an emitted field in the context of double optical resonance is discussed. The critical behaviour in the cooperative limit is discussed too.