

COOPERATIVE EFFECTS AND BISTABILITY FOR RESONANCE FLUORESCENCE

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On the basis of a simple model we discuss the stationary behaviour of a homogeneously broadened system of atoms driven by a coherent resonant field. We give an analytical description of bistability both for the transmitted and for the fluorescent light. In particular we predict that the spectrum of the fluorescent light undergoes a first-order phase transition for high density of the atomic system: when the intensity of the input field crosses a critical value the spectrum changes *discontinuously* from a single narrow line to three widely separated lines which corresponds to a sudden appearance of a large Dynamical Stark Shift.

A system of two-level atoms continuously driven by a coherent quasi-resonant field exhibits a bistable behaviour. The transmitted light varies discontinuously with an hysteresis cycle as a function of the incident amplitude. This behaviour has been very recently experimentally observed [1] and theoretically predicted [2], keeping fully into account propagation and boundary conditions. However, in this way the analysis turns out to be very involved and only numerical results have been obtained.

Without claiming a quantitative comparison and relation with previous theories and experiments we propose a very simple theoretical model which is very similar in spirit to the one-mode model with distributed losses for laser processes with homogeneous broadening. This model gives a simple and exact analytical description of the bistable behaviour for the transmitted light which is deeply similar to the numerical one, giving explicit expressions for the values of atomic density and input field amplitude at which the discontinuities occur.

In correspondence with the same values our model predicts a bistable behaviour also for the fluorescent light diffused at 90° . Precisely the usual assumption that fluorescence intensity is proportional to the number of atoms (non-cooperative behaviour) is found to be correct only below a critical density of atoms.

Above this critical density two stationary situations appear (bistability) and in one of them fluorescence exhibits a cooperative behaviour and is inversely proportional to the number of atoms.

Furthermore we predict a discontinuous variation also for the spectrum of the fluorescent light. Namely in the non-cooperative stationary state the spectrum is three-peaked (Dynamical Stark Shift, DSS) as predicted by previous one-atom theories[†], whereas in the cooperative stationary state the spectrum is a single narrow line. Hence if one continuously increases the input field one should see an *abrupt* appearance of the DSS when the system jumps from the cooperative to the non cooperative stationary state.

This first-order phase transition for resonance fluorescence intensity and spectrum is just the stationary counterpart of transient cooperative spontaneous emission superfluorescence [6] for a system coherently and continuously excited.

We consider a homogeneously broadened active medium composed of $N \gg 1$ two level atoms of transition frequency ω_0 , contained in a pencil-shaped cavity of length L . A classical external field α of frequency ω_0 (perfect resonance) is injected into the cavity in the longitudinal direction inducing a macroscopic po-

[†] The theoretical prediction of the DSS is due to Mollow [3]. See also [4] and references quoted therein. The first experimental observation of the DSS is due to Schuda [5].

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polarization wave S and a population difference

$$\Delta = \frac{1}{2}(N_1 - N_2), \quad (1)$$

where $N_1(N_2)$ is the population of the lower (upper) level. The polarization radiates in turn an internal field a which reacts back on the atoms. Obviously S and a have the same frequency and the same direction of the injected field. This is all for what concerns the "coherent part" of the atom-field interaction; all other incoherent interactions are taken into account as relaxation processes described by suitable phenomenological damping terms, in which the damping constants $\gamma_\perp, \gamma_\parallel, \kappa$ appear. Hence the dynamics of the system atoms + radiation field is described by the following model equations

$$\dot{S} = 2g(a + \alpha)\Delta - \gamma_\perp S, \quad (2a)$$

$$\dot{\Delta} = -2g(a + \alpha)S - \gamma_\parallel(\Delta - \frac{1}{2}N), \quad (2b)$$

$$\dot{a} = -gS - \kappa a, \quad (2c)$$

where g is the coupling constant

$$g = (\omega_0 \mu^2 / \hbar V)^{1/2}, \quad (3)$$

V being the volume of the cavity and μ the modulus of the atomic dipole moment; γ_\perp and γ_\parallel are the transversal and longitudinal atomic relaxation rates; κ is the inverse lifetime of the photons in the cavity, given by

$$\kappa = C(1 - R)/L, \quad (4)$$

R being the reflectivity coefficient of the mirrors.

We stress that putting $\alpha = 0$ and replacing $-\frac{1}{2}N$ by a positive inversion our model equations (2) reduce to the one-mode laser model in a ring laser cavity in the semi-classical approximation [7].

Further eqs. (2) with $\alpha = 0$ and $R = 0$ (cavity without mirrors) reduce to the one-mode model for superfluorescence (see ref. [6]), again in the semi-classical approximation. Hence eqs. (2) are a generalization of these physical situations to include the effect of an injected signal [8].

In our model the quantity α is proportional to the incident field amplitude E_i , whereas the total field $a + \alpha$ is proportional to the transmitted field E_t . Neglecting the internal field a , i.e. putting $a = 0$, eqs. (2a, b) become a closed system of *linear* equations for S, Δ which coincide with the equations for resonance

fluorescence (in the case of exact resonance) studied in refs. [3-5]. In these works resonance fluorescence is described as the interaction of the external radiation field with a *single* atom, assuming that the N -atom dynamics can be simply inferred from the one-atom dynamics. Hence atomic cooperation is neglected. On the other hand, the internal field a which describes radiation reaction is the very origin of atomic cooperation.

Let us introduce the Rabi frequencies [9] ϵ_i and ϵ_t in the incident field and in the transmitted field respectively:

$$\epsilon_i = 2g\alpha = (2E_i \mu)/\hbar,$$

$$\epsilon_t = 2g(a_{st} + \alpha) = (2E_t \mu)/\hbar, \quad (5)$$

where we have used eq. (3) and the simple correspondence $\hbar\omega_0\alpha^2 = E_i^2 V$; a_{st} is the stationary value of the internal field. Correspondingly, let us consider the saturation parameters y and x of the incident field and of the total or transmitted field, respectively:

$$y = \epsilon_i / \sqrt{\gamma_\perp \gamma_\parallel}, \quad x = \epsilon_t / \sqrt{\gamma_\perp \gamma_\parallel}. \quad (6)$$

In the stationary situation eqs. (2a, b) give

$$S_{st} = \frac{1}{2}N \sqrt{\frac{\gamma_\parallel}{\gamma_\perp}} \frac{x}{1 + x^2}, \quad \Delta_{st} = \frac{1}{2}N \frac{1}{1 + x^2}, \quad (7a, b)$$

$$N_{2,st} = \frac{1}{2}N - \Delta_{st} = \frac{1}{2}N \frac{x^2}{1 + x^2}. \quad (7c)$$

Note that if one neglects the reaction field a_{st} (one-atom theory [3-5]) one has by definition $x = y$. In such a case $S_{st}, N_{2,st}$ depend on N in an extensive way. In general, however, $a_{st} < 0$ so that $x < y$, because in an absorbing system the reaction field is opposite in phase to the incident field. In fact, substituting eq. (7a) into eq. (2c) with $\dot{a} = 0$ one gets the equation for x which fixes the stationary solutions:

$$y = \frac{2cx}{1 + x^2} + x. \quad (8)$$

In eq. (8) we have put

$$c = \frac{1}{2}\gamma_R/\gamma_\perp = \frac{1}{2}\alpha_{abs}L/(1 - R), \quad (9)$$

with

$$\gamma_R = g^2 N / \kappa = \gamma_0 \rho L \lambda_0^2 / 8\pi(1 - R), \quad (10)$$

where α_{abs} is the absorption coefficient, γ_0 is the natural linewidth, λ_0 the wavelength and $\rho = N/V$ the atomic density. We notice that for $R = 0$, γ_R is the inverse of the time duration τ_R of the hyperbolic secant pulse in pure superfluorescence [6]. Hence c is the ratio between a cooperative and a non-cooperative decay rate. The term with c in eq. (8) takes atomic cooperation and radiation reaction into account; in fact, putting $c = 0$ in eq. (8) one gets the one-atom solution $x = y$.

Inverting eq. (8) one obtains $x \propto E_t$ as a function of $y \propto E_i$, which can be a multivalued function. Note that in general x depends on N via c , so that S_{st} , $N_{2,\text{st}}$ are no longer extensive quantities.

As shown in fig. 1 the curve (8) has a qualitatively different shape according to whether $c < 4$ or $c > 4$. In fact for $c < 4$, y is a monotonic function of x , so that eq. (8) has only one solution for all values of y . Hence for $c < 4$, $x \propto E_t$ is a continuous single valued function of $y \propto E_i$. The same holds for the fluorescence intensity I_f , which is proportional to the population of the upper level $N_{2,\text{st}}$.

On the contrary above the threshold

$$c = 4 \quad (11)$$

the function (8) has one maximum x_M and one minimum x_m given by

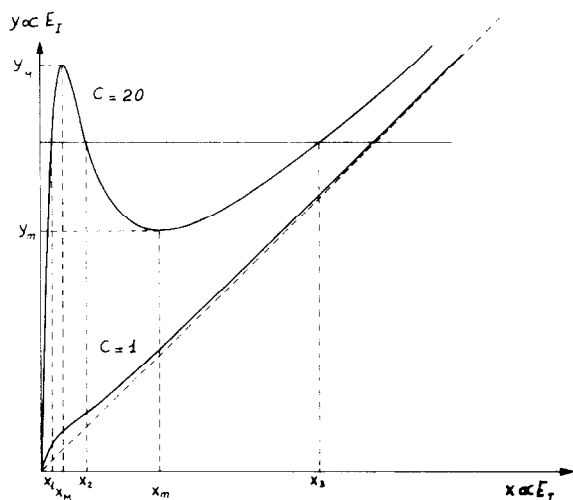


Fig. 1. Plot of the function $y = [2cx/(1+x^2)] + x$ for $c = 1$ and $c = 20$. For $c \geq 4$ one has $x_M = 1$, $y_M = c$, $x_m = \sqrt{2c}$, $y_m = \sqrt{8c}$. Points x_1 and x_3 are stable, whereas points x_2 are unstable.

$$x_M = \left\{ c \frac{2c+1}{1+\sqrt{c^2-4c}} \right\}^{1/2}, \quad (12a)$$

$$x_m = (c-1+\sqrt{c^2-4c})^{1/2}. \quad (12b)$$

Hence x is a *multivalued* function of y , since there is an interval of values of y in correspondence of which one finds *three* stationary solutions. We stress that exchanging ordinates with abscissae in fig. 1 for $c > 4$ one obtains a graph which closely resembles the numerical plot of E_t versus E_i given in ref. [2] and the experimental plot of ref. [1].

For the sake of definiteness, let us consider the situation $c \gg 4$ in which the maximum and the minimum are well pronounced and separated (in fact one has $x_M \simeq 1$, $y_M \simeq c$ and $x_m \simeq \sqrt{2c}$, $y_m \simeq \sqrt{8c}$). In such a case one has one or three stationary solutions according to the value of y :

a) For $y < \sqrt{8c}$ one has one stationary solution x_1 approximately given by:

$$x_1 = y/2c \ll 1. \quad (13)$$

b) For $\sqrt{8c} \leq y \leq c$ one has three stationary solutions $x_1 \leq x_2 \leq x_3$. The linear stability analysis shows that points x_1 and x_3 are stable, whereas x_2 is unstable; x_1 and x_3 are approximately given by

$$x_1 = y/(c + \sqrt{c^2 - y^2}),$$

$$x_3 = \frac{1}{2}y \left(1 + \sqrt{1 - \frac{8c}{y^2}} \right). \quad (14)$$

c) For $y > c$ one has one stationary solution $x_3 \simeq y \gg 1$ which practically coincides with the one-atom solution.

Point x_3 can be considered as the "one-atom stationary solution", whereas point x_1 arises from atomic cooperation so that it can be properly termed "cooperative stationary state". Hence in case (b) the system exhibits bistability, which leads to a first-order phase transition with an hysteresis cycle for $x \propto E_t$ obtained by continuously varying the incident field E_i . This implies via eq. (7) a hysteresis cycle for all other physical quantities and in particular for the fluorescence intensity $I_f \propto N_{2,\text{st}}$. The behaviour of E_t and I_f as a function of E_i is shown in figs. 2a, b. Note that the discontinuity of E_t is of a factor $c \gg 1$ at both transition points, and the discontinuity of I_f is at least of a factor 2. The non-cooperative region in which I_f is

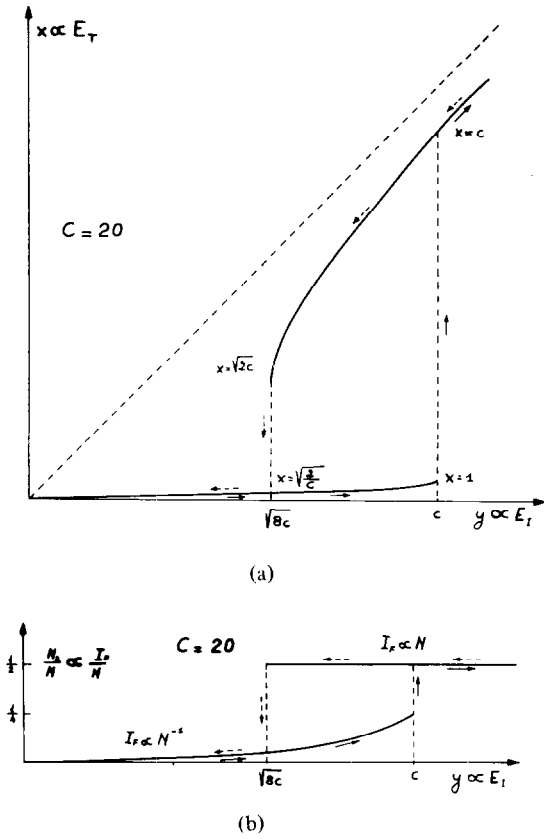


Fig. 2. Hysteresis cycles of (a) the transmitted field and (b) the total fluorescent intensity. Full (dotted) line arrows indicate the variation obtained increasing (decreasing) the input field E_I . The upper (lower) part of the plots corresponds to the stationary solution $x_3(x_1)$.

constant corresponds to complete saturation ($x_3 \gg 1$). In this region eq. (7c) shows that $I_f \propto N$. On the contrary in the cooperative region (lower part of the plots) eqs. (7c) and (14) show that I_f scales as N^{-1} .

The description of the spectrum of the fluorescent light requires the analysis of the approach to the stationary state. Detailed informations of the approach of the system to the stationary states x_1, x_3 can be easily obtained when the photon decay time κ^{-1} is the shortest characteristic time in play, (i.e. $\gamma_{\perp}\gamma_{\parallel}, \epsilon_i, \gamma_R \ll \kappa$). In such a situation, one can adiabatically eliminate the field from eqs. (2a, b, c) obtaining the following system of equations for S, Δ :[‡]

$$\dot{\Delta} = \frac{2\gamma_R}{N} S^2 - \epsilon_i S - \gamma_{\parallel}(\Delta - \frac{1}{2}N),$$

$$\dot{S} = -\frac{2\gamma_R}{N} S\Delta + \epsilon_i \Delta - \gamma_{\perp} S. \quad (15)$$

Linearizing eqs. (15) around the equilibrium points one finds that for $c \gg 1$, x_3 corresponds to a *focus* in the $S - \Delta$ plane, x_1 corresponds to a *nodus* and x_2 is an unstable saddle point. The oscillatory to x_3 is ruled by two exponentials $\exp\{\lambda_{3\pm}t\}$ with

$$\lambda_{3\pm} = -\frac{1}{2}(\gamma_{\perp} + \gamma_{\parallel}) \pm i\epsilon_i; \quad y \gg c \gg 1. \quad (16)$$

The monotonic approach to x_1 is ruled by the exponential $\exp(\lambda_1 t)$ with

$$\lambda_1 = -2\gamma_{\parallel} \left(1 - \frac{c}{c + \sqrt{c^2 - y^2}} \right), \quad 1 \ll y \leq c. \quad (17)$$

We notice that the damping constant λ_1 varies from γ_{\parallel} for $y \ll c$ to 0 for $y = c$, whereas the real part of λ_3 is constant and for $\gamma_{\parallel} = 2\gamma_{\perp}$ coincides with the width of the sidebands in the DSS. Furthermore, on the basis of the regression theorem it is reasonable to assume that an oscillatory character of the approach to the stationary state is necessary to have oscillations of the correlation functions in the stationary state, which in turn are the very origin of the DSS. Hence the imaginary part of λ_3 is the cause of the DSS which in fact appears when the shift ϵ_i becomes larger than the linewidth which is roughly given by the real part of λ_3 . On the contrary we do not expect any DSS when the system is in the cooperative stationary state x_1 because the approach to x_1 is monotonic[☆].

The condition $y > c$ for the appearance of the DSS in a dense system ($c \gg 1$) becomes intuitive in the case $\gamma_{\perp} \simeq \gamma_{\parallel} \simeq \gamma$. In this situation we have from eqs. (6), (9) that $c \simeq \gamma_R/\gamma \gg 1$ and $y/c \simeq \epsilon_i/\gamma_R$. Hence if the cooperative damping γ_R is much larger than the one-atom damping the one-atom threshold condition $\epsilon_i > \gamma$ has to be replaced by $\epsilon_i > \gamma_R \gg \gamma$. This substitution of the threshold condition is rather expected; what is less obvious is the discontinuous appearance of the DSS. Precisely one-atom theories [3–5] pre-

[‡] We recall that the adiabatic approximation becomes exact (i.e. is no longer an approximation) in the stationary state.

[☆] Clearly the rigorous proof of these and the following considerations on the spectrum of the fluorescent light can be given only in the framework of a fully quantum-mechanical calculation of correlation functions.

dict that the two sidebands smoothly merge from a singleline spectrum when ϵ_i becomes larger than γ . On the contrary we predict that if cooperative effects are relevant ($\gamma_R \gg \gamma$) the DSS appears abruptly increasing the incident field; in fact even if $\epsilon_i > \gamma$ but $\epsilon_i < \gamma_R$ we get a single peaked spectrum whose linewidth is ruled by λ_1 and becomes very narrow as ϵ_i approaches γ_R from below ($\nu \rightarrow c$)^{*}. When ϵ_i becomes larger than γ_R this narrow line breaks suddenly into three well separated lines. In fact now the linewidth and frequency shift are ruled by the real and imaginary parts of λ_3 and in the present situation $\epsilon_i > \gamma_R \gg \gamma$. Roughly speaking the appearance of the DSS for increasing field is a second-order phase transition for low density ($\gamma_R < \gamma$) whereas it is a first-order phase transition if cooperative effects are relevant ($\gamma_R > \gamma$).

In conclusion we suggest that this phase transition

^{*} This line narrowing can be connected with a critical slowing down ($\lambda_1 \rightarrow 0$), similar to the one which occurs in tunnel diodes (see [10]).

should be observed in a homogeneously broadened system looking at 90° to the incident beam, just under the same experimental conditions in which one observes bistability for the transmitted light.

References

- [1] H.M. Gibbs, S.L. McCall and T.N.C. Venkatesan, Phys. Rev. Lett. 36 (1976) 1135.
- [2] S.L. McCall, Phys. Rev. A9 (1974) 1515.
- [3] B.R. Mollow, Phys. Rev. 188 (1969) 1969.
- [4] H.J. Carmichael and D.F. Walls, University of Waikato preprint.
- [5] F. Schuda, C.R. Stroud and M. Hercher, J. Phys. B7 (1974) 198.
- [6] R. Bonifacio and L.A. Lugiato, Phys. Rev. A11 (1975) 1507; 12 (1975) 587.
- [7] H. Haken, Handbuch der Physik vol. XXV/2 c (Springer Verlag, Berlin, 1970).
- [8] V. De Giorgio and M.O. Scully, Phys. Rev. A2 (1970) 1170.
- [9] I.I. Rabi, Phys. Rev. 51 (1937) 652.
- [10] R. Landauer and J.W.F. Woo, in: Synergetics, ed. by H. Haken (Teubner, Stuttgart, 1973).