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LETTER TO THE EDITOR

Photon statistics and absorption spectrum in cooperative resonance fluorescence

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Abstract. Analytic results for the absorption spectrum in cooperative resonance fluorescence are presented. An initial broad-band absorption spectrum on the cooperative branch develops a sharp spike as the instability point is approached. The usual one-atom absorption spectrum is regained on the non-cooperative branch.

The photon statistics of the scattered light are studied through a calculation of the second-order correlation function $g^{(2)}(0)$. On the cooperative branch the value of $g^{(2)}(0)$ is unity reflecting the coherent scattering from the cooperative atomic state. On the non-cooperative branch the value of $g^{(2)}(0)$ jumps to two, reflecting the incoherent scattering from independently scattering atoms.

The phenomena of optical bistability and cooperative resonance fluorescence provides another example of cooperative behaviour in non-equilibrium systems. For two recent reviews of this rapidly expanding subject see Prigogine and Nicolis (1977) and Haken (1977). The feature of hysteresis and optical bistability in the output of a driven Fabry–Perot cavity with Na vapour has been observed recently by Gibbs *et al* (1976). This behaviour was predicted by a numerical analysis (McCall 1974). Subsequent analytic results for the hysteresis in the transmitted intensity have been obtained by Bonifacio and Lugiato (1976, 1978) using optical Bloch equations. A quantum mechanical analysis has enabled analytic expressions for the spectrum of the fluorescent light to be obtained in a point system (Carmichael and Walls 1977, to be referred to as I) and in a cavity (Agarwal *et al* 1978).

In this Letter we continue the analysis of cooperative behaviour in the interaction of a single-mode laser field with a collection of N two-level atoms confined to a small volume of dimensions less than the wavelength of the incident light. The results of a cavity calculation may be obtained from our results by replacing the number of atoms N by a cooperativity factor $2c$ (see I).

We shall be concerned with two main features: (a) the absorption spectrum of the cooperative atomic system when probed with a weak field; and (b) the photon statistics of the fluorescent light as exhibited by a calculation of the second-order correlation function.

We derive the optical Bloch equations for the collective atomic operators from the master equation (equation (1) in I). The steady-state solution to these equations is obtained by adopting a scheme of factorisation of operators for different atoms (as in I

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and Agarwal *et al* 1978). The results of the present analysis are equivalent to a mean field theory and their validity await justification by a full Fokker–Planck treatment. The linearised equations about the steady state have the form

$$\begin{aligned}\langle \dot{S}_+(t)\rangle &= -\Gamma_1\langle S_+(t)\rangle - \frac{\mathrm{i}\gamma}{\sqrt{2}} X\langle S_z(t)\rangle \\ \langle \dot{S}_z(t)\rangle &= -\gamma\langle S_z(t)\rangle - \frac{\mathrm{i}\gamma}{2\sqrt{2}} X\left(1-\frac{N-1}{1+X^2}\right)(\langle S_+(t)\rangle - \langle S_-(t)\rangle) \\ \langle \dot{S}_-(t)\rangle &= -\Gamma_1\langle S_-(t)\rangle + \frac{\mathrm{i}\gamma}{\sqrt{2}} X\langle S_z(t)\rangle\end{aligned} \tag{1}$$

where S_+, S_- and S_z are the collective atomic operators. γ is the natural linewidth of the atom, Ω is the Rabi frequency and

$$\Gamma_1 = \frac{\gamma}{2}\left(1+\frac{N-1}{1+X^2}\right). \tag{2}$$

The dimensionless quantity X is a measure of the net driving field

$$X = \frac{2\Omega - \gamma(1-N^{-1})\langle S_y\rangle_{\mathrm{ss}}}{\gamma/\sqrt{2}}. \tag{3}$$

The relation between the dimensionless quantity $Y = 2\sqrt{2}\Omega/\gamma$, a measure of the incident laser field, and X is given by

$$X^3 - YX^2 + NX - Y = 0 \tag{4}$$

exhibiting the hysteresis and bistability.

In the presence of an additional weak probe field of amplitude $\mathscr{E}'$ and of frequency ν the absorption lineshape may be calculated through the quantity (cf Mollow 1972 and references therein)

$$g_{\mathrm{a}}(\Delta\nu) = 4\mathscr{E}'^2 \int_{-\infty}^{\infty} \mathrm{d}t\, \mathrm{e}^{\mathrm{i}\Delta\nu t} g_{\mathrm{a}}(\tau) \tag{5}$$

where

$$\Delta\nu = \nu - \omega_0$$

$$g_{\mathrm{a}}(\tau) = \langle [S_-(t), S_+(t+\tau)]\rangle$$

where this atomic correlation function is due to the presence of the driving field alone.

We have solved equations (1) by Laplace transform methods (cf Hassan and Bullough 1975). The two-time correlation functions are then calculated via the quantum regression theorem (Lax 1967). In a similar fashion to the emission spectrum the absorption spectrum exhibits a discontinuous behaviour between the cooperative and non-cooperative branches. The threshold for the instabilities of these branches is given by

$$\left(1-\frac{N-1}{1+X^2}\right)\left(1-\frac{N-1}{1+X^2}-8X^2\right)=0. \tag{6}$$

We shall give explicit expressions for the absorption spectrum on these two branches.

For values of X below the threshold (the cooperative branch of the hysteresis cycle)

$$g_a(\Delta\nu)=\frac{2\mathscr{E}'^2N}{\pi(1+X^2)}\left(\frac{\Gamma_1}{\Delta\nu^2+\Gamma_1^2}-\frac{-\gamma X^2+\gamma-\Gamma_2-\Delta}{2\Delta}\frac{\Gamma_2+\Delta}{\Delta\nu^2+(\Gamma_2+\Delta)^2}\right.$$
$$\left.+\frac{\gamma X^2+\gamma-\Gamma_2+\Delta}{2\Delta}\frac{\Gamma_2-\Delta}{\Delta\nu^2+(\Gamma_2-\Delta)^2}\right) \tag{7}$$

where

$$\Gamma_2=\gamma-\frac{\gamma}{4}\left(1-\frac{N-1}{1+X^2}\right)$$
$$\Delta=\left[\left(\frac{\gamma}{4}\right)^2\left(1-\frac{N-1}{1+X^2}\right)^2-\frac{\gamma^2}{2}X^2\left(1-\frac{N-1}{1+X^2}\right)\right]^{1/2}.$$

For X above threshold (the non-cooperative branch) we have

$$g_a(\Delta\nu)=\frac{2\mathscr{E}'^2N}{\pi(1+X^2)}\left[\frac{\Gamma_1}{\Delta\nu^2+\Gamma_1^2}-\left(\frac{\Gamma_2}{2}+\frac{(-\gamma X^2+\gamma-\Gamma_2)(\Delta\nu-\Delta')}{2\Delta'}\right)\frac{1}{(\Delta\nu-\Delta')^2+\Gamma_2^2}\right.$$
$$\left.-\left(\frac{\Gamma_2}{2}-\frac{(-\gamma X^2+\gamma-\Gamma_2)(\Delta\nu+\Delta')}{2\Delta'}\right)\frac{1}{(\Delta\nu+\Delta')^2+\Gamma_2^2}\right] \tag{8}$$

where $\Delta'^2=-\Delta^2$. The behaviour of the absorption spectrum is shown in figure 1.

On the cooperative branch far from the instability point the absorption spectrum is a broad Lorentzian of width $N\gamma/2$ centred at $\nu=\omega_0$ the resonant laser frequency (figure 1(a)). This broad bandwidth is a feature of the collective behaviour of the N atoms. As the laser field (Y) is increased a small spike appears around $\nu=\omega_0$ (figure 1(b)). This spike grows and narrows until at the instability point $X=1$ it has zero width (figure 1(c)). We note that similar features were exhibited in the spectrum of the scattered light on the lower branch (Carmichael and Walls 1977, Agarwal *et al* 1978).

For values of $X>1$ the lower branch becomes unstable and the system makes a discontinuous transition to the upper branch ($X\sim N$). An absorption spectrum of the form shown in figure 1(d) suddenly appears. This is the absorption spectrum of a single atom as calculated by Mollow (1972). The single-atom absorption spectrum has recently been observed by Wu *et al* (1977). For values of $|\Delta\nu|<\Delta'$ ($\approx 2\Omega$) the absorption lineshape becomes negative, that is acquires two peaks which exhibit amplification.

The absorption function becomes zero at $\nu=\omega_0\pm 2\Omega$ and changes sign in the region $|\Delta\nu|<2\Omega$. As we now decrease the laser field the two amplified peaks of the absorption spectrum move closer (figure 1(e)) then completely merge into a single amplified peak (figure 1(f)). At the instability point ($X\sim\sqrt{n}$) this single peak narrows to zero and the cooperative branch becomes unstable, the system returning to the stable cooperative branch.

To reveal further the nature of the interaction of the atoms with the light field we consider the photon statistics of the scattered light. Information on the photon statistics is obtained through a calculation of the normalised second-order correlation

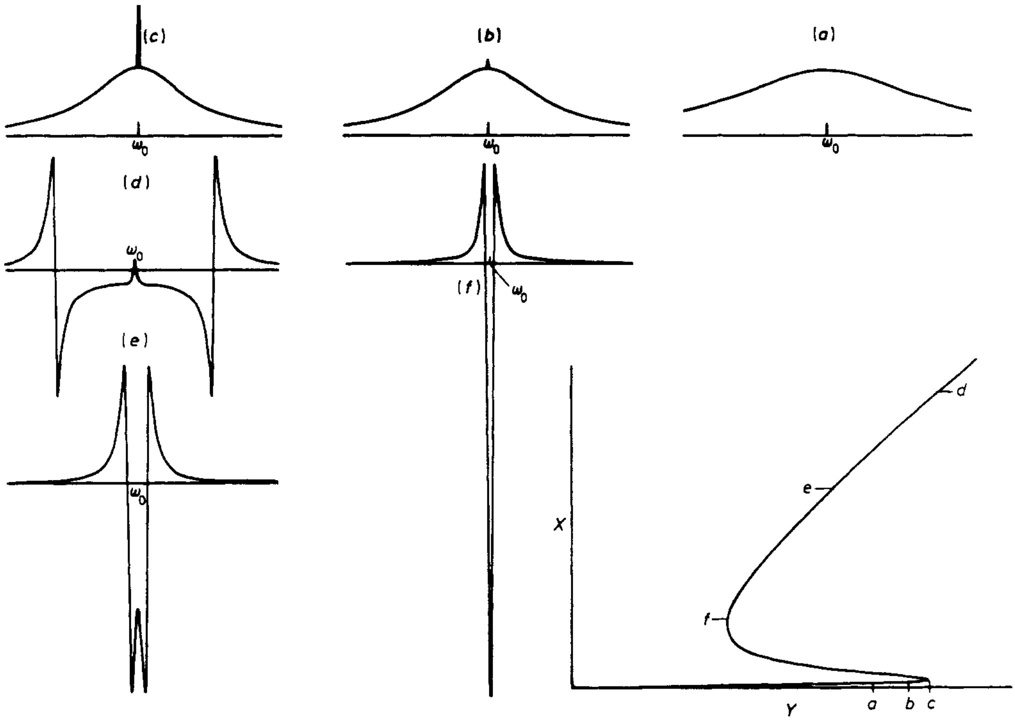

Figure 1. Absorption spectrum $g_a(\Delta\nu)$ plotted against $\Delta\nu$ throughout a hysteresis cycle for $N=80$. Each spectrum is normalised for unit integrated intensity and plotted in the frequency range $\omega_0-40\gamma$ to $\omega_0+40\gamma$.

function (Glauber 1963)

$$g^{(2)}(\tau)=\frac{G^{(2)}(\tau)}{|G^{(1)}(0)|^2} \tag{9}$$

where

$$G^{(2)}(\tau)=\langle E^{(-)}(t)E^{(-)}(t+\tau)E^{(+)}(t+\tau)E^{(+)}(t)\rangle.$$

$G^{(2)}(\tau)$ may be calculated via the atomic operators using the relation of equation (6) in I. We first consider $g^{(2)}(0)$. Again we adopt the scheme of factorisation of operators for different atoms to facilitate the factorisation of higher moments†. Under this approximation and using the steady-state solutions for the atomic expectation values obtained in I we obtain

$$G^{(2)}(0)=\frac{I_0^2X^4}{(1+X^2)^2}\left[\tfrac{1}{4}N(N-1)(N-2)(N-3)\left(\frac{1}{1+X^2}\right)^2 + N(N-1)(N-2)\left(\frac{1}{1+X^2}\right)+\tfrac{1}{2}N(N-1)\right]. \tag{10}$$

† Equivalently we may adopt the assumption that the atoms are in an atomic coherent state to facilitate the factorisation of higher moments.

From I we have

$$|G^{(1)}(0)|^2 = I_0^2\left(\frac{N^2}{2}\frac{X^2}{(1+X^2)^2}+\frac{NX^4}{2(1+X^2)^2}\right)^2. \tag{11}$$

The behaviour of $g^{(2)}(0)$ as a function of the driving field Y is illustrated in figure 2.

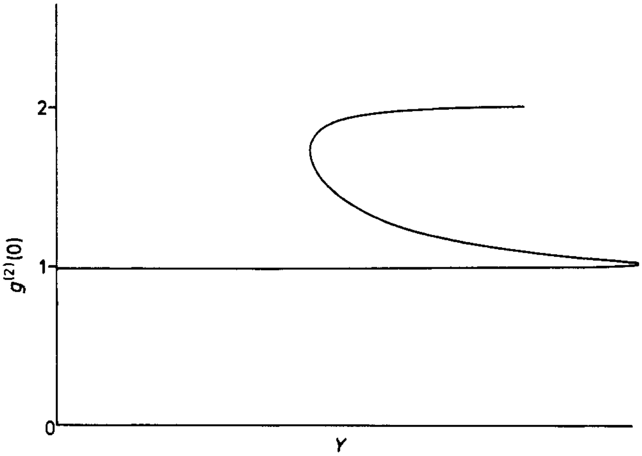

Figure 2. Second-order correlation function $g^{(2)}(0)$ plotted against Y throughout a hysteresis cycle for $N=80$.

On the cooperative branch we find for $N \gg 1$, $g^2(0)=1$. This is the value of $g^2(0)$ characteristic of coherent light and corresponds to the dominance of the coherent scattering from the collective atomic system in this region. The feature of photon antibunching, a property of the fluorescent light from a single atom (Carmichael and Walls 1976, Cohen-Tannoudji 1976, Kimble *et al* 1977), is essentially lost here.

A calculation of $g^{(2)}(\tau)$ on the cooperative branch yields

$$g^{(2)}(\tau) = 1 + \mathrm{O}\left(\frac{1}{N}\right) \tag{12}$$

again characteristic of coherent scattering.

The value of $g^{(2)}(0)$ remains at one as Y increases along the cooperative branch. At the instability point the value of $g^{(2)}(0)$ makes a sudden transition to two as the system goes over to the non-cooperative branch. This value of $g^{(2)}(0)=2$ is characteristic of chaotic light sources. A calculation of $g^{(2)}(\tau)$ on the non-cooperative branch yields

$$g^{(2)}(\tau) = 1 + |g^{(1)}(\tau)|^2 + \mathrm{O}\left(\frac{1}{N}\right). \tag{13}$$

On this branch the incoherent scattering from N independently scattering atoms dominates over the coherent scattering and the above expression reproduces the result of the central limit theorem (of Carmichael and Walls 1976). Thus the behaviour of the second-order correlation function clearly demonstrates the regime of cooperative and non-cooperative behaviour of atoms.

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