# Two-level atom saturated by a fluctuating resonant laser beam. Calculation of the fluorescence spectrum

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**Abstract.** The behaviour of a two-level atom saturated by a resonant fluctuating laser beam is studied. A classical description of the light wave is used, corresponding to a laser well above threshold. Two different approximations are introduced for treating the effects of slow and fast fluctuations; slow fluctuations are followed adiabatically by the atom and fast fluctuations appear as a relaxation mechanism for the population difference and for the optical dipole moment obeying the motional-narrowing condition. The perturbation of the spectral distribution of the fluorescence light is determined quantitatively and the great sensitivity of this spectrum to higher order correlation functions of the light wave is exhibited.

# 1. Introduction

Laser light sources now give the possibility of easily saturating atomic transitions and these saturation effects must be described correctly to obtain a quantitative understanding of the characteristics of the fluorescence light (spectral distribution, polarization,  $\ldots$ ).

When the spectral width  $\Delta v$  of the light wave is very large, which is the case in thermal light sources and some multimode lasers (operating in a great number of closely spaced free-running modes), a perturbative treatment of the interaction between atoms and photons is possible. The correlation time  $1/\Delta v$  of the light wave is so short that there is at most one interaction process during this correlation time. Simple *rate equations* can be derived which describe the coupled evolution of  $\sigma_e$  and  $\sigma_g$  (atomic density matrices in the upper and lower states e and g) under the effect of such uncorrelated one-photon processes (Barrat and Cohen-Tannoudji 1961, Ducloy 1973, 1974; see also Cohen-Tannoudji 1962, 1975).

A great amount of theoretical work has been devoted to the opposite case ( $\Delta v = 0$ ) of a monochromatic resonant wave which has an infinitely long correlation time. Bloch-type equations are then more appropriate for describing the coherent nutation of atoms between e and g. Such equations have been used extensively in the field of quantum optics (see for example Sargent et al 1974 and references therein). Another example is the problem of the spectral distribution of the fluorescence light emitted by a two-level atom resonantly driven by an intense monochromatic laser beam; this has received a lot of attention (Mollow 1969, Oliver et al 1971, Carmichael and Walls 1975;

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see also references given in Cohen-Tannoudji 1976, Swain 1975, Renaud *et al* 1976, Carmichael and Walls 1976, Kimble and Mandel 1976) in connection with the recent experimental observation of such a spectrum (Schuda *et al* 1974, Walther 1975, Wu *et al* 1975).

In this paper we consider the intermediate situation of a quasi-monochromatic intense laser beam. The spectral width  $\Delta v$  is assumed to be sufficiently small so that several nutations between e and g can occur during the correlation time  $1/\Delta v$ , excluding any perturbative approach. More precisely, if  $\omega_1$  is the mean Rabi nutation frequency, we take

$$\omega_1 \gg \Delta v. \tag{1.1}$$

However  $\Delta v$  is also assumed to be large compared with the natural width  $\Gamma$ , i.e.

$$\Delta v \gtrsim \Gamma \tag{1.2}$$

so that the laser light does not appear monochromatic for the atom. Because of (1.1) and (1.2), neither rate equations nor Bloch-type equations can be used.

It is clear from (1.1) and (1.2) that atoms are now sensitive to higher order correlation functions of the laser beam. Since such a field is generally not Gaussian, knowledge of  $\Delta v$  is not sufficient to characterize these higher order correlation functions (Glauber 1964) and some model is necessary for describing the fluctuations of the light wave.

We consider, in this paper, a classical fluctuating wave corresponding to the well known properties of a single-mode laser well above threshold: a very well defined amplitude undergoing small fluctuations and a phase  $\phi(t)$  which, in addition to fast fluctuations, slowly diffuses in the complex plane (Haken 1970). We show how conditions (1.1) and (1.2) can be used for treating the effects of slow and fast fluctuations separately. The general idea is that slow fluctuations are sufficiently slow to be followed adiabatically by the atoms, whereas fast fluctuations are sufficiently fast to be considered as a relaxation process obeying the motional-narrowing condition (Abragam 1961).

Such an approach is applied to the computation of the correlation function of a two-level-atom dipole moment which gives, by a Fourier transform, the spectral distribution of the fluorescence light. The perturbation of the well known three-peak structure of this spectrum is evaluated quantitatively and related to the correlation functions of fast and slow fluctuations. A comparison between the spectra obtained with two light beams with the same intensity and spectral width but with quite different statistical properties (laser beam well above threshold and Gaussian beam) illustrates the great sensitivity of such spectra to higher order correlation functions.

After a presentation of the general method which is used ( $\S$ 2), a quantitative description of the fluctuating laser beam is given in  $\S$ 3. The results are then applied in  $\S$ 4 to the determination of the spectral distribution of the fluorescence light.

# 2. General method

### 2.1. Description of the light beam

A light beam emitted by a single-mode laser well above threshold has a very well defined amplitude |E(t)|, undergoing only very small fluctuations, and a phase  $\phi(t)$  which, in addition to short time fluctuations with correlation time  $\tau_c$ , slowly diffuses in the complex plane with a characteristic time  $\tau_d \gg \tau_c$  (see figure 1). We will describe



Figure 1. Time variations of the amplitude |E(t)| and phase  $\phi(t)$  of the electric field of the laser wave, showing fast and slow fluctuations characterized by the correlation times  $\tau_c$  and  $\tau_d$ .

classically such a light beam by the fluctuating electric field

$$\operatorname{Re}\left(E_{0} + e(t)\right)\exp\left[-\mathrm{i}(\omega t + \phi(t))\right]$$
(2.1)

where  $E_0$  is the mean amplitude of the field, e(t) is the fluctuating part of the amplitude and  $\phi(t)$  is the fluctuating phase. Due to these fluctuations, the light beam is no longer monochromatic and has a spectral width  $\Delta v$  around its mean pulsation  $\omega$ .

## 2.2. Geometrical representation of the problem

We are interested in the behaviour of two-level atoms irradiated by such a light beam which is supposed to be resonant ( $\omega$  coincides with the energy splitting  $\omega_0$  between the two atomic levels e and g; we take  $\hbar = c = 1$ ). The evolution of the atomic density matrix  $\sigma$  is given by

$$i\frac{d}{dt}\sigma = [H,\sigma]$$
(2.2)

where H is the total Hamiltonian represented in the basis  $\{|e\rangle, |g\rangle\}$  by the matrix

$$H = \begin{pmatrix} \frac{1}{2}\omega_0 & \frac{1}{2}d(E_0 + e(t))\exp[-i(\omega_0 t + \phi(t))] \\ \frac{1}{2}d(E_0 + e(t))\exp[i(\omega_0 t + \phi(t))] & -\frac{1}{2}\omega_0 \end{pmatrix}.$$
 (2.3)

The rotating-wave approximation (RWA) has been used, d is the dipole matrix element between e and g.

It is well known that such a problem can be formulated in terms of a fictitious spin  $\frac{1}{2}$ , **S**, having two levels  $|+\rangle$  and  $|-\rangle$  (corresponding to  $|e\rangle$  and  $|g\rangle$ ), and precessing around a longitudinal static magnetic field  $\mathbf{B}_0$  of amplitude  $-\omega_0/\gamma$  ( $\gamma =$  gyromagnetic ratio of  $\mathbf{S}$ ) and a transverse magnetic field  $\mathbf{B}_1$ , of amplitude  $-d(E_0 + e(t))/\gamma$ , and rotating with the phase  $\omega_0 t + \phi(t)$ .

In addition to the laboratory frame  $0xyz(\Sigma)$ , it will be convenient to introduce two reference frames: the coherent frame  $0x'y'z'(\Sigma')$  rotating with angular velocity  $\omega_0$ around  $B_0$  and the instantaneous frame  $0x''y''z''(\Sigma'')$  which follows  $B_1$ , i.e. which rotates around  $B_0$  with the phase  $\omega_0 t + \phi(t)$  (see figure 2). Applying the rotation  $\exp[i(\omega_0 t + \phi(t))S_z]$  to equation (2.2), one easily finds that in  $\Sigma''$  the fictitious spin 'sees' three fields (see figure 2): a large *constant* field  $B_1$  parallel to 0x'' with an



Figure 2. Coherent frame  $(\Sigma')$  and instantaneous frame  $(\Sigma'')$  deduced from the laboratory frame  $(\Sigma)$  by rotations around 0z with angles respectively equal to  $\omega_0 t$  and  $\omega_0 t + \phi(t)$ . In the fictitious spin  $\frac{1}{2}$  representation,  $B_1$  is associated in  $\Sigma''$  with the mean amplitude  $E_0$  of E(t),  $b_{\parallel}$  with the amplitude fluctuations,  $b_{\perp}$  is the Larmor field associated with the phase variation  $\dot{\phi}(t')$ .

amplitude  $-dE_0/\gamma$ , and two small *fluctuating* fields  $\boldsymbol{b}_{\parallel}$  and  $\boldsymbol{b}_{\perp}$  respectively, parallel and perpendicular to  $\boldsymbol{B}_1$  (along 0x'' and 0z'') with amplitudes  $-de(t)/\gamma$  and  $\dot{\phi}(t)/\gamma$  (the longitudinal field  $\boldsymbol{B}_0$  disappears in  $\Sigma'$ ; the rotation  $e^{i\phi(t)S_z}$  which carries from  $\Sigma'$  to  $\Sigma''$ introduces the 'Larmor' field  $\boldsymbol{b}_{\perp}$ ).

#### 2.3. Evolution of the system. Statistical averages and separation of fast and slow fluctuations

Let  $S_p$ ,  $S'_p$ ,  $S''_p$  be the spherical components of the fictitious spin in  $\Sigma$ ,  $\Sigma'$  and  $\Sigma''$  respectively  $(p = +, z, -; S_{\pm} = \mp \sqrt{\frac{1}{2}}(S_x \pm iS_y))$ . To study the evolution of the spin in  $\Sigma$  or  $\Sigma'$  (which are both non-fluctuating frames), we will first determine its motion in the instantaneous frame  $\Sigma''$ , and then return from  $\Sigma''$  to  $\Sigma'$  by using

$$S'_{p}(t) = e^{-ip\phi(t)}S''_{p}(t).$$
(2.4)

In  $\Sigma''$ , **S** precesses around the three fields  $B_1$ ,  $b_{\perp}$  and  $b_{\perp}$ , so that the transformation from  $S''_q(0)$  to  $S''_p(t)$  is equivalent to a 'rotation'  $\{R_{pq}(t)\}$ :

$$S_{p}''(t) = \sum_{q} R_{pq}(t) S_{q}''(0).$$
(2.5)

The elements  $R_{pq}(t)$  depend on  $B_1$  and on the whole behaviour of  $b_{\parallel}(t')$  and  $b_{\perp}(t')$ , i.e. of e(t') and  $\dot{\phi}(t')$ , between 0 and t. From (2.4) and (2.5) one easily derives the following relation between  $S'_p(t)$  and  $S'_q(0)$ :

$$S'_{p}(t) = \sum_{q} R_{pq}(t) e^{-ip\phi(t)} e^{iq\phi(0)} S'_{q}(0)$$
  
=  $\sum_{q} R_{pq}(t) \exp[-ip(\phi(t) - \phi(0))] \exp[i(q - p)\phi(0)] S'_{q}(0)$  (2.6)

which clearly exhibits the initial phase  $\phi(0)$ , the phase diffusion  $\phi(t) - \phi(0)$  during 0-tand the total precession in  $\Sigma''$  described by  $R_{pq}(t)$ . Expression (2.6) must now be averaged over all possible realizations of the random functions e(t) and  $\phi(t)$  in the interval 0-t. We will see in the next section that, for a laser well above threshold,  $\phi(t)$  undergoes one-dimensional Brownian motion. The phase diffusion  $\phi(t) - \phi(0)$  and the velocities  $\phi(t')$  for t' > 0 (which are implicit in  $R_{pq}(t)$ ) are therefore non-correlated with the initial phase  $\phi(0)$  and the average of expression (2.6) over  $\phi(0)$  reduces to the average of exp[ $i(q - p)\phi(0)$ ]. Since  $\phi(0)$  is uniformly distributed between 0 and  $2\pi$ , one immediately gets

$$\exp[\mathbf{i}(q-p)\phi(0)] = \delta_{pq} \tag{2.7}$$

so that expression (2.6) becomes

$$S'_{p}(t) = \overline{R_{pp}(t) \exp[-ip(\phi(t) - \phi(0))]} S'_{p}(0).$$
(2.8)

The main problem which remains is to determine whether or not  $R_{pp}(t)$  and  $\exp[-ip(\phi(t) - \phi(0))]$  are independent random variables.

The phase diffusion  $\phi(t) - \phi(0)$  which can be written as

$$\phi(t) - \phi(0) = \int_0^t \phi(t') \, \mathrm{d}t'$$
(2.9)

depends, as  $R_{pp}(t)$ , on the whole behaviour of  $\phi(t')$  between 0 and t, so that, at first sight, it does not seem possible to average independently the two terms  $R_{pp}(t)$  and  $\exp[-ip(\phi(t) - \phi(0))]$  appearing in (2.8). It can be shown however that, as a consequence of condition (1.1) and for a laser well above threshold,  $R_{pp}(t)$  and  $\phi(t) - \phi(0)$  are independent variables. Let us just give here a simple interpretation of this result. Some of the statistical properties of  $\phi(t)$  which are used in this discussion will be established in §3 where a quantitative description of phase fluctuations is presented.

The general idea is to separate slow and fast fluctuations in  $\phi(t)$  (see figure 1). This can be done by averaging  $\phi(t)$  over a time interval  $\theta$  which is small compared to  $\tau_d$  but large compared to  $\tau_c$ . More precisely we introduce

$$\phi_{\text{slow}}(t) = \int_{-\infty}^{+\infty} g(\tau) \,\phi(t-\tau) \,\mathrm{d}\tau \tag{2.10}$$

where the averaging function  $g(\tau)$  satisfies  $\int_{-\infty}^{+\infty} g(\tau) = 1$  and has a width  $\theta$ , and

$$\phi_{\text{fast}}(t) = \phi(t) - \phi_{\text{slow}}(t). \tag{2.11}$$

Now one easily understands that the phase diffusion  $\phi(t) - \phi(0)$  is mainly correlated with the slow phase fluctuations  $\dot{\phi}_{slow}(t')$  (with  $0 \le t' \le t$ ), since for  $t \gg \tau_c$ 

$$\phi(t) - \phi(0) \simeq \int_0^t \phi_{slow}(t') dt'.$$
 (2.12)

In fact, it is possible to show that  $\phi(t) - \phi(0)$  and  $\dot{\phi}_{fast}(t')$  are, to a very good approximation, independent variables (see demonstration at the end of §3). It follows that, if we show that  $R_{pp}(t)$  only depends on  $\dot{\phi}_{fast}(t)$ ,  $R_{pp}(t)$  is not correlated with the exponential of expression (2.8) and the two quantities can be averaged independently.

In  $\Sigma''$  associated with  $\phi_{slow}(t)$  is a field

$$b_{\perp \, \text{slow}} = (1/\gamma) \phi_{\, \text{slow}} \tag{2.13}$$

which has a very small amplitude (the corresponding Larmor frequency being of the order of  $1/\tau_d$ ) and very slow variations with characteristic times of the order of  $\tau_d$ .

Let us first consider in  $\Sigma''$  the motion of the spin around  $B_1$  and  $b_{\perp slow}$ . If  $B_1$  is very large, more precisely if

$$\omega_1 \gg 1/\tau_d \tag{2.14}$$

where

$$\omega_1 = \gamma \boldsymbol{B}_1 = d\boldsymbol{E}_0 \tag{2.15}$$

is the Rabi nutation frequency associated with the average electric field  $E_0$ , the resultant of  $B_1$  and  $b_{\perp slow}$  makes an angle with  $B_1$  which always remains small and which fluctuates slowly with a characteristic time  $\tau_d$ . It follows that the spin precesses several times around  $B_1 + b_{\perp slow}$  before  $b_{\perp slow}$  changes appreciably. The slow phase fluctuations are therefore so slow that they are followed adiabatically by the spin which is not affected by them. To summarize, if  $\omega_1 \gg 1/\tau_d$ ,  $R_{pp}(t)$  does not depend on the slow fluctuations and is therefore not correlated with  $\exp[-ip(\phi(t) - \phi(0))]$ .

Finally, the average appearing in (2.8) can be done in two steps: average of  $\exp[-ip(\phi(t) - \phi(0))]$  over the slow phase diffusion, average of  $R_{pp}(t)$  over fast fluctuations, which is equivalent to computing in  $\Sigma''$  the relaxation produced by the fluctuating fields  $b_{\parallel}(t)$  and  $b_{\perp \text{ fast}}(t)$ . We will assume that the fast fluctuations are so fast that they satisfy the motional-narrowing condition (Abragam 1961), so that the corresponding relaxation can be evaluated perturbatively. The condition of validity of such an approximation is

$$\gamma^2 \boldsymbol{b}_{\parallel}^2 \tau_{\rm c}^2 = d^2 [e(t)]^2 \tau_{\rm c}^2 \ll 1 \tag{2.16}$$

$$\gamma^2 \boldsymbol{b}_{\perp \, \text{fast}}^2 \, \tau_c^2 = [\phi_{\text{fast}}]^2 \, \tau_c^2 \simeq [\phi(t)]^2 \, \tau_c^2 \ll 1.$$
(2.17)

Let us finally mention that it is easy to include the effects of spontaneous emission (which gives a natural width  $\Gamma$  to the excited state) just by adding the corresponding damping coefficients in the evolution of the components of the spin. Such a procedure is justified by the ultra-short correlation time of the vacuum fluctuations of the electromagnetic field (which are at the origin of spontaneous emission).

We now need a more precise model of the light beam in order to check the conditions of validity ((2.14), (2.16) and (2.17)) of the theoretical method described above and to get statistical information on  $\phi(t)$  and e(t) which is necessary for computing  $\overline{R_{pp}(t)}$  and  $\overline{\exp[-ip(\phi(t) - \phi(0))]}$ .

For the sake of simplicity, from now on we will ignore amplitude fluctuations, assuming that they have been greatly reduced by some stabilizing device. It would however be quite easy to include their effect, which as seen above is equivalent to the relaxation produced by the fluctuating field  $\boldsymbol{b}_{\parallel}(t)$ .

## 3. Quantitative description of phase fluctuations

#### 3.1. Analogy with Brownian motion

For the evolution of the electric field from a single-mode laser, we will use the following simple picture which results from several quantum theories of lasers (see for example Haken 1970, Sargent *et al* 1974).

The motion of the point representing, in the complex plane, the electric field of the mode  $\omega_0$  has the same characteristics as the classical motion of a fictitious particle



Figure 3. A symmetric vertical plane section containing 0z of the potential well in which the point representative of the laser electric field moves. Well above threshold, the equilibrium position corresponds to a non-zero amplitude  $E_0$ .

subjected to the two-dimensional potential represented in figure 3 and, in addition, to damping and fluctuating forces. The shape of the potential well of figure 3 has a very simple physical meaning. Due to the amplifying atomic medium, the amplitude of the electric field tends to increase, but, above a certain value, the non-linearities of the medium introduce saturations which are at the origin of the minimum of the potential well. The damping and fluctuating forces are a consequence of the coupling of the oscillator to various reservoirs (losses in the cavity, thermal noise, spontaneous emission, ...).

The radial oscillations of the representative point correspond to amplitude fluctuations around  $E_0$  and are very small if the curvature of the potential well is sufficiently high (i.e. for a laser well above threshold). On the other hand, due to axial symmetry the tangential motion of the fictitious particle, which corresponds to the phase evolution, is free. It follows that the evolution of the phase  $\phi(t)$  is analogous to one-dimensional Brownian motion on the circle  $\mathscr{C}$  of radius  $E_0$ , described by the Langevin equation

$$\ddot{\phi} + \kappa \dot{\phi} = F(t). \tag{3.1}$$

In this equation the effect of the interaction with noise reservoirs has been split into two contributions:

(i) a damping term with a friction coefficient  $\kappa$ , describing the mean effect of this interaction, and

(ii) an additional fluctuating force F(t), of zero mean value:

$$\overline{F(t)} = 0. \tag{3.2}$$

We assume that F(t) has an extremely short correlation time compared to all other characteristic evolution times of the system, i.e.

$$\overline{F(t_1) F(t_2)} = C \,\delta(t_2 - t_1) \tag{3.3}$$

where C is a constant giving the order of magnitude of F.

Moreover it seems reasonable to assume that F, which results from the interaction of a large number of independent systems (atoms in the cavity,...), has a Gaussian probability distribution. According to the general properties of Gaussian distributions, the statistical behaviour of F is therefore completely determined by (3.2) and (3.3).

## 3.2. Slow phase diffusion; spectral distribution of the incident laser light

The spectrum  $\mathcal{J}(\omega)$  of the emitted light is given by the Fourier transform of the correlation function of the electric field (2.1):

$$\mathscr{J}(\omega) \propto \int e^{-i\omega\tau} \overline{E(t) E^*(t-\tau)} \, \mathrm{d}\tau.$$
(3.4)

If the amplitude fluctuations are very small,  $\mathcal{J}(\omega)$  can be written as

$$\mathscr{J}(\omega) \propto E_0^2 \int \exp[-i(\omega - \omega_0)\tau] \,\overline{\exp[i(\phi(t) - \phi(t - \tau))]} \,d\tau \qquad (3.5)$$

which shows that the spectral width  $\Delta v$  of  $\mathcal{J}(\omega)$  around  $\omega_0$  arises from the phase diffusion  $[\phi(t) - \phi(t - \tau)]$ .

As F(t) is a random Gaussian function, it follows from (3.1) that  $\phi(t)$  is also Gaussian (Chandrasekhar 1943), so that one can easily show from the general properties of Gaussian functions that

$$\overline{\exp[i(\phi(t) - \phi(0))]} = \exp[-\frac{1}{2}(\phi(t) - \phi(0))^2].$$
(3.6)

Finally the phase diffusion  $\overline{(\phi(t) - \phi(0))^2}$  can be computed from the Fourier transform of the Langevin equation (3.1). One finds that for t larger than  $1/\kappa$ ,  $\overline{(\phi(t) - \phi(0))^2}$  increases linearly with t:

$$\overline{(\phi(t) - \phi(0))^2} = (C/\kappa^2)t \tag{3.7}$$

which gives the possibility of defining precisely the slow diffusion time  $\tau_d$  as

$$\tau_{\rm d} = \kappa^2 / C. \tag{3.8}$$

Introducing (3.8) into (3.7), (3.6) and (3.5) gives for  $\mathcal{J}(\omega)$ :

$$\mathcal{J}(\omega) \simeq \frac{1}{(\omega - \omega_0)^2 + (1/2\tau_d)^2}.$$
 (3.9)

The conclusion of this simple calculation is that the slow diffusion time is nothing but the inverse of the spectral width  $\Delta v$  of the laser light:

$$\Delta v = 1/\tau_{\rm d}.\tag{3.10}$$

The condition of validity (2.14) of the adiabatic approximation can therefore be written as

$$\omega_1 \gg \Delta v \tag{3.11}$$

which is precisely what we have supposed in the introduction (see (1.1)).

#### 3.3. Fast phase fluctuations

To define precisely the short correlation time  $\tau_c$  introduced in §2.1 let us consider the correlation function  $\overline{\phi(t)\phi(t-\tau)}$  of the time derivative of  $\phi(t)$ . This correlation function is readily computed from the Fourier transform of the Langevin equation (3.1). One gets

$$\dot{\phi}(t)\phi(t-\tau) = \frac{1}{2}\kappa\,\Delta v\,\mathrm{e}^{-\kappa\tau}.\tag{3.12}$$

This result shows that  $\tau_c$  is nothing but the inverse of the damping coefficient  $\kappa$ :

$$\tau_{\rm c} = 1/\kappa. \tag{3.13}$$

To have an order of magnitude of  $\kappa$ , i.e. of  $\tau_c$ , let us return to the simple model studied by Haken (1970) where the atomic medium consists of motionless two-level atoms with natural width  $\gamma$  contained in a cavity of width  $\xi$  ( $\kappa$  is found in this case to be equal to  $\gamma + \xi$ ). Well above threshold the spectral width  $\Delta v$  of the emerging laser light is much smaller than the atomic and cavity widths  $\gamma$  and  $\xi$ , which shows that  $\tau_d$  is much longer than  $\tau_c$ . Such a result ( $\tau_d \gg \tau_c$ ) certainly holds for other types of lasers where the lasing atoms or molecules have an inhomogeneous width.

From equation (3.12), one also derives

$$\overline{(\phi(0))^2} = \frac{1}{2}\kappa\,\Delta\nu\tag{3.14}$$

which gives the possibility of checking the motional-narrowing condition (2.17). According to (3.13) and (3.14), this condition can be written as

$$\overline{(\phi(0))^2} \tau_{\rm c}^2 = \frac{1}{2} \kappa \,\Delta \nu \,(1/\kappa^2) = \frac{1}{2} (\Delta \nu/\kappa) \ll 1. \tag{3.15}$$

Such a condition is obviously fulfilled since  $\tau_c = 1/\kappa$  is much smaller than  $\tau_d = 1/\Delta v$ .

Finally, the conditions of validity of the general method presented in §2 (separation of fast and slow fluctuations, motional-narrowing condition) are satisfied in the limit  $\omega_1 \gg \Delta v$  expressed by condition (1.1). Since we can add independently the damping terms representing the effect of spontaneous emission (because of the ultra-short correlation time of this process), the method of §2 applies whether  $\Gamma$  is large or small compared to  $\Delta v$ . However let us recall that the case  $\Delta v \ll \Gamma$  is more simply treated by Bloch's equations, since in that case the light perturbation appears monochromatic to the atom. (When  $\Delta v \ll \Gamma$ , the slow fluctuations are negligible during the radiative lifetime  $1/\Gamma$ , and the relaxation rates associated with fast fluctuations, which will be shown later to be of the order of  $\Delta v$  (see (4.9)), can also be neglected in comparison to  $\Gamma$ .) This is why the calculations presented in this paper are essentially useful when  $\Delta v \gtrsim \Gamma$ (condition (1.2)). We will of course check at the end (see §4.4) that the results of these calculations reduce to the ones given by Bloch's equations when  $\Delta v \ll \Gamma$ .

We will end this section with a brief discussion on the problem of the statistical independence of  $\phi(t) - \phi(0)$  and  $\dot{\phi}_{fast}(t'')$  with  $0 \le t'' \le t$ . We shall first calculate  $(\phi(t) - \phi(0))\phi_{fast}(t'')$ . From (2.9), (2.10) and (2.11), one gets

$$\overline{(\phi(t) - \phi(0))\phi_{\text{fast}}(t'')} = \int_0^t dt' \ \overline{\phi(t')\phi_{\text{fast}}(t'')}$$
$$= \int_0^t dt' \ \overline{\phi(t')\phi(t'')} - \int_0^t dt' \ \int_{-\infty}^{+\infty} d\tau \ g(\tau) \ \overline{\phi(t')\phi(t'' - \tau)}.$$
(3.16)

We will suppose that  $t \gtrsim \tau_d$  since, for  $t \ll \tau_d$ , the phase diffusion  $\phi(t) - \phi(0)$  is negligible and the exponential of expression (2.8) is equal to unity, so that the average of (2.8) reduces to  $\overline{R_{pp}(t)}$ . Using (3.12) and the fact that the width  $\theta$  of the averaging function  $g(\tau)$  is large compared to  $\tau_c$  and small compared to  $\tau_d$ , and therefore to t, one

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immediately gets

$$\overline{(\phi(t) - \phi(0))\phi_{\text{fast}}(t'')} \simeq \frac{1}{2}\Delta\nu \left(1 - \int_0^t dt' \int_{-\infty}^{+\infty} d\tau g(\tau) \delta(t' - t'' + \tau)\right)$$
$$= 0 \quad \text{if} \quad \theta < t'' < t - \theta.$$
(3.17)

We have used  $\int_{-\infty}^{+\infty} d\tau g(\tau) = 1$ . So, except when t'' lies in two small regions of width  $\theta$  near the two extremities of the interval 0-t, we have

$$\overline{(\phi(t) - \phi(0))} \,\phi_{\text{fast}}(t'') = 0 = \overline{(\phi(t) - \phi(0))} \,\overline{\phi_{\text{fast}}(t'')}.$$
(3.18)

This demonstration can be generalized to higher order averages of the type

$$(\phi(t) - \phi(0))^{p} \phi_{\text{fast}}(t''_{1}) \dots \phi_{\text{fast}}(t''_{q}) = \int_{0}^{t} dt'_{1} \dots \int_{0}^{t} dt'_{p} \overline{\phi(t'_{1}) \dots \phi(t'_{p}) \phi_{\text{fast}}(t''_{1}) \dots \phi_{\text{fast}}(t''_{q})}.$$
(3.19)

Using the properties of random Gaussian functions, the average appearing on the right of (3.19) can be factorized in products of second-order correlation functions. Let us consider a 'mixed' correlation function of the type  $\phi(t'_i) \phi_{fast}(t'_j)$ . The same calculation as above shows that the integral over  $t'_i$  of this correlation function gives 0 (except when  $t''_j$  lies in two small regions of width  $\theta$  near 0 and t). It follows that all the mixed correlation functions give no contribution and that

$$\overline{(\phi(t) - \phi(0))^p \phi_{\text{fast}}(t_1'') \dots \phi_{\text{fast}}(t_q'')} = \overline{(\phi(t) - \phi(0))^p \phi_{\text{fast}}(t_1'') \dots \phi_{\text{fast}}(t_q'')}$$
(3.20)

which proves the statistical independence of  $\phi(t) - \phi(0)$  and  $\phi_{fast}(t'')$ .

## 4. Spectral distribution of the fluorescence light

# 4.1. Expression of the signal

We are interested in the spectral distribution  $L_{\rm F}(\omega)$  of the fluorescence emitted by a two-level atom irradiated by the quasi-monochromatic light described above. In order to get rid of the Doppler effect, the atoms form a beam irradiated at right angles by the laser beam and the fluorescence is detected in the third perpendicular direction.

It is well known that  $L_F(\omega)$  is given by the Fourier transform of the correlation function of the atomic dipole moment (Mollow 1969). As such a dipole is related to the transverse component  $S_{\pm}$  of the fictitious spin, it may easily be shown that

$$L_{\rm F}(\omega) \sim \int_0^T {\rm d}t \int_0^{T'} {\rm d}t' \langle S'_+(t) S'_-(t') \rangle \exp[-i(\omega - \omega_0) (t - t')]. \tag{4.1}$$

In (4.1),  $S'_{\pm}(t')$  are Heisenberg operators in the coherent frame  $\Sigma'$ . The average is taken in the time-independent Heisenberg state of the system. T is the observation time, i.e. the transit time of atoms through the laser beam diameter. T is generally sufficiently large so that most of the time spent by an atom within the laser beam corresponds to a steady-state regime. In such a case  $\langle S'_+(t) S'_-(t') \rangle$  only depends on t - t' and the expression (4.1) of  $L_{\rm F}(\omega)$  may be reduced to

$$L_{\rm F}(\omega)/T \sim \int_0^\infty d\tau \langle S'_+(\tau) S'_-(0) \rangle \exp[-i(\omega - \omega_0)\tau].$$
(4.2)

## 4.2. Computation of the correlation function

The equation written above in (2.6) is valid for Heisenberg operators and may be used to compute the correlation function  $\langle S'_+(\tau) S'_-(0) \rangle$ :

$$\langle S'_{+}(\tau)S'_{-}(0)\rangle = \overline{R_{++}(\tau)\exp[-i(\phi(\tau)-\phi(0))]} \langle S'_{+}(0)S'_{-}(0)\rangle.$$

$$(4.3)$$

We have shown in §§2 and 3 that  $R_{++}(\tau)$  and  $\exp[-i(\phi(\tau) - \phi(0))]$  could be averaged independently. Before doing these two averages, let us evaluate  $\langle S'_{+}(0) S'_{-}(0) \rangle$  which is also equal to  $\frac{1}{2} + \langle S'_{z}(0) \rangle$  (for a spin  $\frac{1}{2}$ ,  $S_{+}S_{-} = \frac{1}{2} + S_{z}$ ). Since we assume a steady state is reached, and  $\omega_{1}$  is assumed to be very large, the atomic transition can be considered to be completely saturated (equalization of the populations of *e* and *g*) so that  $\langle S'_{z}(0) \rangle = 0$  and

$$\langle S'_{+}(0) S'_{-}(0) \rangle = \frac{1}{2}.$$
 (4.4)

Now, from (3.6), (3.7), (3.8) and (3.10) we have

$$\overline{\exp\left[-i(\phi(\tau) - \phi(0))\right]} = \exp\left[-\frac{1}{2}(C/\kappa^2)\tau\right] = \exp\left(-\tau/2\tau_d\right) = \exp\left(-\Delta v\tau/2\right)$$
(4.5)

so that (4.3) may be rewritten as

$$\langle S'_{+}(\tau) S'_{-}(0) \rangle = \frac{1}{2} \exp(-\Delta v \tau/2) R_{++}(\tau).$$
 (4.6)

To evaluate  $\overline{R_{++}}(t)$ , let us take the average of equation (2.5) giving the motion of the spin in the instantaneous frame  $\Sigma''$ :

$$\langle S_p''(\tau) \rangle = \sum_q \overline{R_{pq}}(\tau) \langle S_q''(0) \rangle.$$
(4.7)

As explained in §2, averaging over fast fluctuations is equivalent in the fluctuating frame  $\Sigma''$  to computing the relaxation produced by the fluctuating field  $b_{\perp fast}$ .

It is well known that such a relaxation can be described by two relaxation times  $T_1$  and  $T_2$  (Abragam 1961) giving the damping of the components of S'' respectively parallel and perpendicular to the large and non-fluctuating field  $B_1$  seen by the spin in  $\Sigma''$ . As the motional-narrowing condition is satisfied (see §3.3), the longitudinal relaxation time  $T_1$  associated with the fluctuating field  $b_{\perp \text{ fast}}$  with an amplitude  $\phi_{\text{ fast}}/\gamma \simeq \dot{\phi}(t)/\gamma$  is easily shown to be

$$1/T_1 = \frac{1}{2} \int_{-\infty}^{+\infty} e^{i\omega_1 \tau} \overline{\phi(t)} \overline{\phi(t-\tau)} d\tau.$$
(4.8)

Let us recall that  $\omega_1$  is the Larmor frequency around  $B_1$ . Using the expression (3.12) for the correlation function of  $\dot{\phi}(t)$  finally gives

$$\frac{1}{T_1} = \frac{1}{2} \frac{\kappa^2}{\kappa^2 + \omega_1^2} \,\Delta v. \tag{4.9}$$

Since amplitude fluctuations are neglected, the only fluctuating field appearing in  $\Sigma''$  is  $b_{\perp \text{ fast}}$ , perpendicular to  $B_1$ . It follows that

$$1/T_2 = 1/2T_1. (4.10)$$

This results from the fact that there is no adiabatic contribution to  $T_2$  (the fluctuating field  $b_{\text{fast}}$  has no diagonal elements in the basis  $\{|\pm\rangle_x\}$  of eigenstates of  $S_x$ ,

i.e. in the energy levels of the fictitious spin interacting with  $B_1$ ). It follows that  $1/T_2$  is equal to half the sum of the transition rates from  $|\pm\rangle_x$  which are both equal to  $1/2T_1$ . This proves (4.10).

The previous discussion clearly shows that it would be easy to include the effect of fast amplitude fluctuations associated with the fluctuating field  $\boldsymbol{b}_{\parallel}(t)$ , parallel to  $\boldsymbol{B}_1$ . This would not change the value of  $T_1$  but would add an 'adiabatic' contribution to  $T_2$  (Fourier transform of the correlation function of the amplitude fluctuations  $\boldsymbol{e}(t)$ , taken at frequency 0).

Finally, after averaging over fast fluctuations, equation (4.7) becomes

$$\langle S_x''(\tau) \rangle = e^{-\tau/T_1} \langle S_x''(0) \rangle$$
(4.11)

and

$$\langle (S_y'' \pm \mathrm{i} S_z'')(\tau) \rangle = \mathrm{e}^{\pm \mathrm{i}\omega_1 \tau} \mathrm{e}^{-\tau/T_2} \langle (S_y'' \pm \mathrm{i} S_z'')(0) \rangle.$$

$$(4.12)$$

 $R_{\pm-}(\tau)$  is easily obtained by rewriting equations (4.11) and (4.12) in the basis  $(S''_{\pm} = \pm \sqrt{\frac{1}{2}}(S''_x \pm iS''_y), S''_z)$  and is found to be

$$\overline{R_{++}}(\tau) = \frac{1}{2} (e^{-\tau/T_1} + \cos \omega_1 \tau \ e^{-\tau/T_2}).$$
(4.13)

Inserting (4.13) into (4.6) finally gives for the correlation function:

$$\langle S'_{+}(\tau) S'_{-}(0) \rangle = \frac{1}{4} \{ \exp[-\tau(1/T_{1} + \Delta \nu/2)] + \cos \omega_{1} \tau \exp[-\tau(1/T_{2} + \Delta \nu/2)] \}.$$
(4.14)

*Remark.* It could appear from the previous discussion that amplitude fluctuations cannot be neglected since they are responsible for an adiabatic contribution in  $1/T_2$  (Fourier transform at frequency 0), whereas phase fluctuations appear in a Fourier transform at frequency  $\omega_1$  (see (4.8)). Actually, coming back to figure 3, one sees that the spectrum of amplitude fluctuations is centred not on zero but around a frequency corresponding to the radial oscillation in the potential well. From the simple model of Haken (1970) one can show that such a frequency is much higher than  $\omega_1$ , which considerably reduces the effect of amplitude fluctuations on  $T_2$ .

#### 4.3. Inclusion of the effect of spontaneous emission

Spontaneous emission transfers atoms from e to g with a rate  $\Gamma$  equal to the natural width of e and damps the dipole moment with a rate  $\Gamma/2$ . Such a process is described in  $\Sigma''$  by the following rate equations:

$$\frac{\mathrm{d}}{\mathrm{d}t} \langle S_{z}^{\prime\prime}(t) \rangle = -\Gamma(\frac{1}{2} + \langle S_{z}^{\prime\prime}(t) \rangle)$$

$$\frac{\mathrm{d}}{\mathrm{d}t} \langle S_{\pm}^{\prime\prime}(t) \rangle = -\frac{1}{2}\Gamma \langle S_{\pm}^{\prime\prime}(t) \rangle$$
(4.15)

which can be added independently to the other causes of evolution.

Since  $\omega_1$  is supposed very large compared to  $\Gamma$ , S'' makes several precessions around  $B_1$  before being damped by spontaneous emission. It follows that one can neglect any coupling introduced by spontaneous emission between  $S''_y + iS''_z$ ,  $S''_y - iS''_z$ and  $S''_x$  (which precess around  $B_1$  with different frequencies  $\omega_1, -\omega_1, 0$ ). Keeping only the secular terms in (4.15) gives

$$\frac{\mathrm{d}}{\mathrm{d}t} \langle S_x'' \rangle = -\frac{1}{2} \Gamma \langle S_x'' \rangle$$

$$\frac{\mathrm{d}}{\mathrm{d}t} \langle S_y'' \pm \mathrm{i} S_z'' \rangle = -\frac{3}{4} \Gamma \langle S_y'' \pm \mathrm{i} S_z'' \rangle.$$
(4.16)

The quantum regression theorem (Lax 1968) can now be used for computing the effect of spontaneous emission on two-times correlation functions, and one easily shows from (4.16) that it is sufficient to add two damping factors to equations (4.11) and (4.12):  $e^{-\Gamma \tau/2}$  for the first one,  $e^{-3\Gamma \tau/4}$  for the second. Finally the correlation function  $\langle S'_+(\tau) S'_-(0) \rangle$ , including the effect of spontaneous emission, is given by

$$\langle S'_{+}(\tau) S'_{-}(0) \rangle = \frac{1}{4} \{ \exp[-\tau(1/T_{1} + \frac{1}{2}\Delta\nu + \frac{1}{2}\Gamma)] + \cos\omega_{1}\tau \exp[-\tau(1/T_{2} + \frac{1}{2}\Delta\nu + \frac{3}{4}\Gamma)] \}$$
(4.17)

# 4.4. Shape of the fluorescence spectrum

The Fourier transform of equation (4.17) is readily evaluated and leads to the fluorescence spectrum  $L_F(\omega)$  represented in figure 4.



**Figure 4.** Spectral distribution of the fluorescence light emitted by a two-level atom irradiated by a fluctuating laser wave (well above threshold).  $\Gamma$ , natural width;  $\Delta v$ , spectral width of the laser;  $T_1$  and  $T_2$ , longitudinal and transverse relaxation times associated with the fast phase fluctuations.

One gets three Lorentzian components:

(i) a central peak around  $\omega_0$ , with a halfwidth  $1/T_1 + \frac{1}{2}\Delta \nu + \frac{1}{2}\Gamma$  and a height  $(1/T_1 + \frac{1}{2}\Delta \nu + \frac{1}{2}\Gamma)^{-1}$  and

(ii) two sidebands centred on  $\omega_0 \pm \omega_1$ , with a halfwidth  $1/T_2 + \frac{1}{2}\Delta v + \frac{3}{4}\Gamma$  and a height  $\frac{1}{2}(1/T_2 + \frac{1}{2}\Delta v + \frac{3}{4}\Gamma)^{-1}$ .

For a monochromatic non-fluctuating laser light ( $\Delta v = 1/T_1 = 1/T_2 = 0$ ), these results coincide with the well known conclusion concerning the fluorescence spectrum for a two-level atom resonantly irradiated by an intense monochromatic wave (Mollow 1969). Since  $1/T_1$  is of the order of or smaller than  $\Delta v$  (see (4.9)), the same results holds as long as  $\Delta v \ll \Gamma$ . We therefore check that, when  $\Delta v \ll \Gamma$ , the present treatment leads to the same conclusions as the one based on Bloch's equations. The results obtained above show that the effect of a finite spectral laser width  $\Delta v > \Gamma$  does not simply reduce to a broadening of the three components by an amount  $\Delta v$ . An additional broadening arises from fast fluctuations and affects the central component and the two sidebands differently. Measuring this extra broadening could provide interesting information on the dynamics of fast fluctuations (correlation function of  $\dot{\phi}(t)$  and e(t)).

In the limiting case of a large spectral width ( $\Delta v \gg \Gamma$ ),  $\Gamma$  can be neglected in (4.17). If, in addition,  $\kappa$  is assumed to be large compared to  $\omega_1$ ,  $1/T_1$  coincides with  $\Delta v/2$  (see (4.9)). As  $1/T_2$  is equal to  $1/2T_1$ , it follows that in such a limiting case the central component has a width  $2\Delta v$  larger than the one of the two sidebands which is  $3\Delta v/2$ . The ratio of the heights is  $\frac{2}{3}$  (instead of  $\frac{1}{3}$  in the absence of any fluctuation).

From an experimental point of view, such a study clearly shows that, for a verification of the theoretical predictions concerning resonance fluorescence in intense monochromatic fields, one must avoid not only spatial inhomogeneities of the laser beam, but also reduce the slow and fast temporal variations to an acceptable level.

#### 4.5. Comparison with a Gaussian light beam having the same intensity and spectral width

In order to illustrate the sensitivity of  $L_{\rm F}(\omega)$  to higher order correlation functions, it will be interesting to discuss in a qualitative way the shape which would be obtained for  $L_{\rm F}(\omega)$  with a Gaussian light beam, with the same intensity and spectral width as the laser beam considered above, i.e. the same lowest order correlation function.

In the complex plane, the electric field of the light beam is represented by a point which now moves in a harmonic potential well centred at the origin. The steady-state distribution is Gaussian and symmetric around 0. If one starts at a certain time from a distribution represented by a  $\delta$  function (pure coherent state), the distribution moves toward 0 along a logarithmic spiral with a characteristic time  $\tau_d = 1/\Delta v$ , and its width gradually increases from 0 to the steady-state value with the same characteristic time (Louisell and Marburger 1967). Such a result clearly shows that for a Gaussian field, not only the phase but also the amplitude exhibit (in addition to fast fluctuations) important slow fluctuations characterized by a time  $\tau_d$  which is the inverse of  $\Delta v$ . Instead of a well defined Rabi nutation frequency  $\omega_1$ , there is now a distribution of these frequencies characterized by a large width, of the order of  $(\overline{\omega_1^2})^{1/2}$ , and a correlation time  $\langle S'_+(\tau) S'_-(0) \rangle$ : an average over the initial value of  $\omega_1$  and an average over the slow amplitude fluctuations.

Starting from (4.3):

$$\langle S'_{+}(\tau) S'_{-}(0) \rangle = \frac{1}{2} \overline{R_{++}(\tau) \exp[-i(\phi(\tau) - \phi(0))]}.$$
 (4.18)

Consider first a time interval  $[0, \tau]$  which is short compared with the characteristic time  $1/\Delta v$  of slow fluctuations. In this interval, the amplitude and phase of the electric field can be considered as constant and the correlation function can thus be rewritten as

$$\langle S'_{+}(\tau) S'_{-}(0) \rangle \simeq \frac{1}{2} R_{++}(\tau).$$
 (4.19)

As the relaxation times associated with fast fluctuations are of the order of  $1/\Delta v$ , one can also neglect their effect during the time interval (0,  $\tau \ll 1/\Delta v$ ).  $R_{++}$  is therefore only determined by the precession of S'' around  $B_1$  at the frequency  $\omega_1$ , and



Figure 5. Time dependence of the correlation function of the dipole moment of a two-level atom resonantly driven by an intense Gaussian light beam of spectral width  $\Delta v$ . Two characteristic times appear:  $1/\Delta v$  and  $(\overline{\omega_1^2})^{-1/2}$ , where  $(\overline{\omega_1^2})^{1/2}$  is the mean Rabi nutation frequency.

 $\langle S'_{+}(\tau) S'_{-}(0) \rangle$  is readily found to be

$$\langle S'_{+}(\tau) S'_{-}(0) \rangle = \frac{1}{4} \overline{(1 + \cos \omega_{1} \tau)}.$$
 (4.20)

Averaging  $\cos \omega_1 \tau$  over the initial value of  $\omega_1$  gives for  $\langle S'_+(\tau) S'_-(0) \rangle$  a function which decreases from  $\frac{1}{2}$  (value at t = 0) to  $\frac{1}{4}$  in a very short time interval of the order of  $(\overline{\omega_1^2})^{-1/2}$ .  $\langle S'_+(\tau) S'_-(0) \rangle$  does not change as long as  $(\overline{\omega_1^2})^{-1/2} \leq \tau < 1/\Delta \nu$ ; then one must take into account the relaxation produced by fast fluctuations and the effect of the slow diffusion of phase and amplitude, which both imply a decay of  $\langle S'_+(\tau) S'_-(0) \rangle$  to 0 with a characteristic time of about  $1/\Delta \nu$ .

In conclusion, the correlation function  $\langle S'_+(\tau) S'_-(0) \rangle$  exhibits two decays of equal amplitude but corresponding to quite different time constants  $(\overline{\omega_1^2})^{-1/2}$  and  $1/\Delta \nu$  (see figure 5). Its Fourier transform  $L_F(\omega)$  is therefore made of two peaks of equal area centred at  $\omega = \omega_0$ :

(i) a sharp peak, with a width  $\Delta v$ , and

(ii) a wide peak, with a width  $(\omega_1^2)^{1/2}$ .

The ratio of their heights is  $(\omega_1^2)^{1/2}/\Delta v$ , inverse of the ratio of their widths (see figure 6).



**Figure 6.** Spectral distribution of the fluorescence light emitted by a two-level atom irradiated by an intense Gaussian beam of spectral width  $\Delta v$ . The three-peak structure disappears. A narrow central component of width about  $\Delta v$  is superimposed on a broad background of width about  $(\omega_1^2)^{1/2}$ .

Due to the large dispersion of the amplitude of the light electric field, the threepeak structure in the fluorescence spectrum which appeared when a laser beam of very well defined amplitude was used for the excitation, is now completely washed out in this limiting case.

The disappearance of the three-peak structure clearly shows the sensitivity of the fluorescence spectrum to higher order correlation functions of the light electric field.

So far, we have only considered two-level atoms. It is well known that, when a structure exists in e or g, other interesting signals can be studied, such as for example level-crossing resonances, or double resonance,..., on the total fluorescence light (integrated over frequencies). It seems interesting to investigate the sensitivity of such signals to the fluctuations of the laser beam. As mentioned in the introduction, they cannot be computed from rate equations or Bloch-type equations. We show in a forthcoming paper that the method described in this paper can also be applied to these problems.

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