

ATOMS IN STRONG RESONANT FIELDS
SPECTRAL DISTRIBUTION OF THE FLUORESCENCE LIGHT

Claude Cohen-Tannoudji

Ecole Normale Supérieure and Collège de France, Paris

1). Introduction. Lowest order Q.E.D. predictions

Resonance fluorescence, i.e. absorption and reemission of resonance radiation by free atoms, is a very important process. By looking at the fluorescence light emitted by these atoms, for example by measuring its polarization, its intensity, its spectral distribution or its time dependence, one gets interesting informations on various important atomic parameters such as g factors, fine or hyperfine structures, radiative lifetimes ...

The physical picture which is usually given for such a process ⁽¹⁾ can be visualized by the following lowest order diagram :

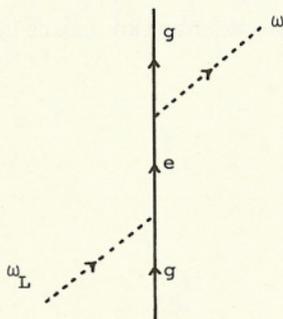


Figure 1 : Lowest order Q.E.D. diagram for resonance fluorescence

One impinging photon, with frequency ω_L , represented by the incoming dotted line is absorbed by the atom which jumps from the ground state g to the excited state e. After a certain amount of time spent in e, the atom falls back to g, spontaneously emitting a photon ω . The scattering amplitude derived from Q.E.D. for such a process obviously contains a $\delta(\omega - \omega_L)$ function which expresses the conservation of energy. Lowest order Q.E.D. therefore predicts that the fluorescence light following a monochromatic excitation must be monochromatic, with the same frequency ω_L as the inci-

dent light. In addition, one finds that the scattering amplitude is the greater, the nearer ω_L is to the atomic frequency ω_0 . More precisely, if Γ is the radiative width of the excited state, the scattering cross section $\sigma(\omega_L)$ varies as $[(\omega_L - \omega_0)^2 + (\Gamma/2)^2]^{-1}$.

Suppose that the incident light beam contains photons with all frequencies ω forming a white continuous spectrum (or at least a spectrum with a width $\Delta \gg \Gamma$). Every photon ω will be elastically scattered with an efficiency given by $\sigma(\omega)$, so that one predicts from the same lowest order treatment that the spectral distribution of the fluorescence light following a broad line excitation has a lorentzian shape, centred at $\omega = \omega_0$, with a half-width $\Gamma/2$.

All these conclusions are clearly no longer valid in very strong resonant fields. We have now at our disposal laser light sources which can easily saturate an atomic transition: the atom can interact several times with the laser light before emitting spontaneously a photon and a lowest order treatment of the fluorescence process is obviously insufficient. How do atoms behave in strong resonant (or quasi-resonant) light beams? What kind of light do they emit? What is the influence of the spectral width of the incident light? Are the higher order correlation functions of the light important? These are examples of questions which arise now in connection with laser experiments.

I have already discussed the problem of optical pumping and level crossing experiments performed with lasers at the 1974 Heidelberg conference on Atomic Physics ⁽²⁾. So, I will rather discuss in the present talk another problem which is the spectral distribution of the fluorescence light emitted by an atomic beam which is irradiated at right angle by a high intensity laser beam (as the 2 beams are perpendicular, there is no Doppler effect). The first experimental observation of such a spectral distribution has been published last year by Schuda, Stroud and Hercher ⁽³⁾. Similar experiments are being performed in other laboratories and will be reported at the present conference in subsequent talks ⁽⁴⁾ ⁽⁵⁾. Concerning the theory of these effects, several calculations have been published, using different methods and approaches ⁽⁶⁾ \leftrightarrow ⁽¹⁷⁾. They don't reach all the same quantitative conclusions. Rather than entering into the details of these calculations, I have thought it would be more interesting in this talk to make a few remarks and comments and to try to give some physical feeling about important parameters. Some new theoretical results will be reported at the end of the paper.

2). What does conservation of energy imply?

The first remark will concern conservation of energy. One could think at first sight that such a principle implies for the fluorescence light following a monochromatic excitation to be always monochromatic, with the same frequency as the

incident light. This is not correct at high intensities. Non linear scattering processes can take place in which N impinging photons (with $N > 1$), having all the same energy ω_L (we take $\hbar = c = 1$), disappear and are replaced by N scattered photons with different energies $\omega_1^s, \omega_2^s \dots \omega_N^s$. Conservation of energy only requires $N\omega_L = \omega_1^s + \omega_2^s + \dots + \omega_N^s$. As an illustration, we have represented on Fig. 2 such non linear scattering processes corresponding to $N = 2$.

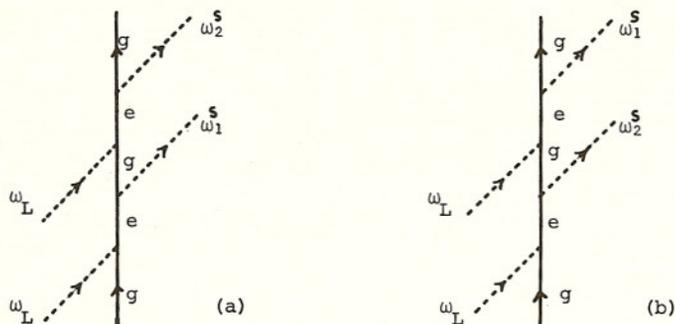


Figure 2 : Example of non linear scattering processes in which 2 impinging photons ω_L, ω_L give rise to 2 scattered photons ω_1^s, ω_2^s with $\omega_1^s + \omega_2^s = 2\omega_L$. The 2 diagrams (a) and (b) differ by the order of emission of the 2 photons ω_1^s and ω_2^s .

I would like also to point out on this example that although $\omega_1^s + \omega_2^s$ is well defined and equal to $2\omega_L$, ω_1^s and ω_2^s are individually spread over finite intervals, which means that inelastic scattering is not monochromatic. Such a finite width of the fluorescence spectrum is due to the energy denominators associated to the intermediate states appearing in diagrams 2a and 2b. When calculating the sum of the 2 scattering amplitudes 2a and 2b, one finds that one of the 2 photons is distributed over an interval of half width $\Gamma/2$ around ω_0 [Γ being the natural width of the excited state e]. Consequently, the second photon is distributed over an interval $\Gamma/2$ around $2\omega_L - \omega_0$.

3). The "dressed atom" approach

It would not be a good idea to consider higher and higher order diagrams for understanding the behaviour of an atom in a strong resonant field. For sufficiently large intensities of this field, the perturbation series would not converge, and the situation would be the more difficult, the nearer ω_L would be to ω_0 . So we are tempted to try another approach.

Why don't we treat to all orders the coupling between the atom and the incoming photons, neglecting spontaneous emission in a first step? Let us call

"dressed atom" the total isolated system which results from the coupling between the atom and the incoming photons. Such a system has stationary states $\psi_\alpha, \psi_\beta \dots$ with energies $E_\alpha, E_\beta \dots$ which can be calculated easily ⁽¹⁸⁾. Then we could treat spontaneous emission by using Fermi's golden rule : the dressed atom jumps from ψ_α to a lower level ψ_β , by spontaneously emitting a photon $\omega = E_\alpha - E_\beta$ with a probability per unit time proportional to $|\langle \psi_\alpha | D | \psi_\beta \rangle|^2$ where D is the atomic electric dipole operator. This process is diagrammatically represented on Fig. 3, where the heavy lines represent the stationary states of the dressed atom.

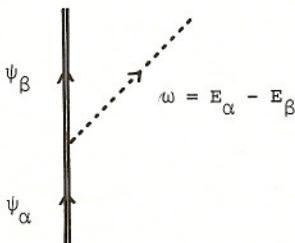


Figure 3 : Spontaneous emission of a photon $\omega = E_\alpha - E_\beta$ by the dressed atom (heavy lines) which jumps from ψ_α to a lower level ψ_β .

4). The difficulty of dealing with cascades

The dressed atom approach is very convenient for finding the number and the mean position of the various components of the fluorescence spectrum which correspond to the Bohr frequencies ($E_\alpha - E_\beta$) of the allowed transitions ($\langle \psi_\alpha | D | \psi_\beta \rangle \neq 0$) of such a system. However, if we want to get more precise informations, concerning for example the widths and the relative amplitudes of these various components, we cannot consider only a single spontaneous emission process as in Fig. 3.

To make this point clear, it will be useful to give some orders of magnitude. An atom, with a thermal velocity $v \sim 10^3 \text{ m.s}^{-1}$, crossing a laser beam of 10^{-3} m. diameter, spends in this light beam a time $T \sim 10^{-6} \text{ s}$, much longer than the radiative lifetime $\tau = \Gamma^{-1}$ of e, which is typically $\tau \sim 10^{-8} \text{ s}$. If the light intensity is large enough, the atomic transition is saturated, and the atom spends half of its time in e, so that an average number of $N = \frac{1}{2} \frac{T}{\tau} \sim 50$ spontaneous emission processes can occur during the interaction time T. It follows that the evolution of the dressed atom is more exactly described by the diagrams of Fig. 4 (where, to simplify, we have supposed N to be only equal to 3).

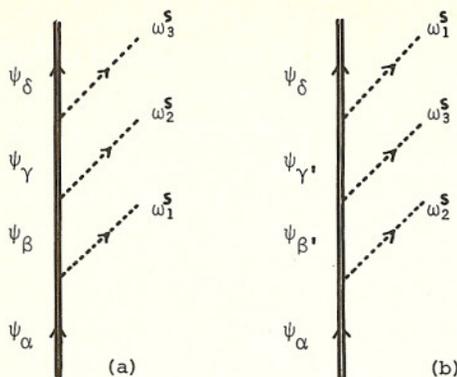


Figure 4 : Sequences of 3 spontaneous emission processes. The 2 sequences (a) and (b) correspond to the same initial and final states ψ_α and ψ_δ , but to a different order of emission of the 3 photons $\omega_1^s, \omega_2^s, \omega_3^s$.

The dressed atom is "cascading" from ψ_α to ψ_β , then from ψ_β to ψ_γ and finally from ψ_γ to ψ_δ , successively emitting photons with frequencies $\omega_1^s, \omega_2^s, \omega_3^s$ close to $E_\alpha - E_\beta, E_\beta - E_\gamma, E_\gamma - E_\delta$ (fig. 4-a). But we can imagine other processes, corresponding to the same initial and final states ψ_α and ψ_δ , to the same frequencies $\omega_1^s, \omega_2^s, \omega_3^s$ of the 3 emitted photons, but to a different order of emission of these 3 photons. $N!$ such possibilities exist, of which only 2 are represented on fig. 4.

The difficulty lies in the fact that, being interested in a precise measurement of the frequencies of the photons, we cannot simultaneously determine the time at which they are emitted (time and frequency are complementary physical quantities) and, consequently, we cannot decide what is the quantum path which is followed by the system. We have $N!$ quantum amplitudes which interfere.

One could at least think that one amplitude is much greater than the others because of the energy denominators associated to the intermediate states. This is not true and comes from the periodical structure of the energy diagram of the dressed atom which is itself due to the quantization of the field mode associated to the laser. For any order of emission of the 3 photons, one can find in general intermediate states ψ_β, ψ_γ , which introduce small energy denominators by approximately matching the energy of the emitted photon (similar difficulties are encountered when one studies the spontaneous emission from a harmonic oscillator (¹⁹)).

The correct way of pursuing the calculation would be to compute, for all values of N , the $N!$ interfering cascading amplitudes, to deduce from them the N -fold probability distribution $\mathcal{P}^{(N)}(\omega_1^s, \omega_2^s \dots \omega_N^s)$ for having N spontaneously emitted photons with frequencies $\omega_1^s, \omega_2^s \dots, \omega_N^s$, finally, after several integrations, to derive from the $\mathcal{P}^{(N)}$ a reduced one photon distribution $\mathcal{J}(\omega)$ giving the probability for any individual photon to have the frequency ω , which is the measured spectral distribution.

5). Why not calculating directly the spectral distribution $\mathcal{J}(\omega)$?

Although such an approach is correct, it seems too ambitious. It gives too many informations which are not useful : we are not measuring the $\mathcal{P}^{(N)}$, but $\mathcal{J}(\omega)$. Would not it be possible to calculate directly $\mathcal{J}(\omega)$ without passing as an intermediate step through the $\mathcal{P}^{(N)}$?

We are thus led to the problem of relating directly $\mathcal{J}(\omega)$ to some simple physical quantities characterizing the radiating atoms. Such a problem has been considered in many references ⁽¹⁰⁾ ⁽²⁰⁾ and I will give here only the results. One finds that the spectral distribution of a given light field is proportional to the Fourier transform (F.T.) of the correlation function (c.f.) of the positive frequency part of the electric field operator. As this electric field is radiated by the atom, it may be related to the atomic electric dipole operator D . So, we find that $\mathcal{J}(\omega)$ is proportional to the F.T. of the c.f. of the atomic dipole moment D . More precisely, let $D_- = d |g\rangle\langle e|$ and $D_+ = d |e\rangle\langle g|$ be the lowering and raising parts of D , d being equal to the matrix element $\langle e|D|g\rangle$ (which is assumed to be real). One finds that :

$$\mathcal{J}(\omega) \sim \int_0^T dt \int_0^T dt' \langle D_+(t) D_-(t') \rangle e^{-i\omega(t-t')} \quad (1)$$

The integrals over t and t' run over the interval of time $[0, T]$ during which the atom radiates (transit time through the laser beam). The operators $D_+(t)$ and $D_-(t')$ are evaluated in the Heisenberg picture, and the average value is taken within the quantum state of the whole system.

6). Spin 1/2 representation of the problem

At this stage of the discussion, and because of lack of time, I will restrict myself to a classical description of the laser field, (but not of the empty modes of the electromagnetic field into which the atom spontaneously emits photons). It would be of course possible to calculate the correlation function written in (1) for the dressed atom introduced above and this has been done ⁽¹³⁾ ⁽¹⁷⁾ (such a calculation is considerably simpler than the computation of the whole set of $\mathcal{P}^{(N)}$). As the number n of impinging photons is very large, we would not make the difference between \sqrt{n} and $\sqrt{n+1}$, and the results would be the same as the ones derived from a classical description of the laser field. Such a classical description will give me the possibility of developing simple geometrical interpretations and fruitful analogies with magnetic resonance experiments.

It is well known that a fictitious spin 1/2 can be associated to any 2 level system, so that our problem can be formulated in the following geometrical

terms ⁽²¹⁾. We have a spin 1/2 \vec{S} , which precesses around a magnetic field \vec{B}_0 parallel to Oz with a Larmor frequency equal to the energy separation ω_0 between e and g (\vec{B}_0 is given by $\omega_0 = -\gamma B_0$, γ being the gyromagnetic ratio of the spin). To D_{\pm} are associated the raising and lowering operators $\mathcal{S}_{\pm} = \mathcal{S}_x \pm i \mathcal{S}_y$, so that we are interested in the c.f. of some transverse components of the spin in the xOy plane.

In this representation, the laser field, of frequency ω_L , is described by an oscillating magnetic field $\vec{B}_1 \cos \omega_L t$ parallel to Ox. We can decompose this oscillating field into 2 left and right circular components, of amplitude $B_1 = B_0/2$, and keep only the one which precesses around \vec{B}_0 in the same sense as the spin \vec{S} . Let $\omega_1 = -\gamma B_1$ be the Larmor frequency associated to B_1 (Rabi nutation frequency). ω_1 characterizes the strength of the coupling between the atom and the laser and must be compared to Γ which measures the strength of spontaneous emission. Neglecting the counter-rotating components of \vec{B}_1 is called "rotating wave approximation" (r.w.a) and amounts to ignore Bloch-Siegert's shifts which are much smaller in optical than in RF range. Note that, when doing r.w.a., we don't exclude "light-shifts" ⁽²²⁾ which may appear for a quasi-resonant irradiation ($\Gamma < |\omega_L - \omega_0| \ll \omega_0$) and which may be much larger than Bloch-Siegert shifts.

It will be convenient to describe the situation in a reference frame OXYZ rotating around Oz = OZ with the "good" component \vec{B}_1 of \vec{B}_1 , so that \vec{B}_1 is static in this reference frame and parallel to OX (fig. 5). Let $\vec{S}(t)$ be the spin operator in this reference frame. The Larmor precession of $\vec{S}(t)$ around OZ is reduced from ω_0 to $\omega_0 - \omega_L$, and we can consider that \vec{S} only "sees" 2 static fields \vec{B}_0 and \vec{B}_1 respectively parallel to OZ and OX and proportional to $\omega_0 - \omega_L$ and ω_1 ($\omega_0 - \omega_L = -\gamma B_0$, $\omega_1 = -\gamma B_1$). Expressed in terms of $S_{\pm}(t) = e^{\mp i \omega_L t} \mathcal{S}_{\pm}(t)$, (1) can be written as :

$$J(\omega) \sim \int_0^T dt \int_0^T dt' \langle S_+(t) S_-(t') \rangle e^{-i(\omega - \omega_L)(t-t')} \quad (2)$$

Finally, we have a simple geometrical representation of the internal energy of the atom (precession of \vec{S} around \vec{B}_0), of its coupling with the laser (precession around \vec{B}_1). The question now is how to describe spontaneous emission in this representation (i.e. the coupling with the empty modes of the electromagnetic field) and how to calculate the c.f. written in (2) ?

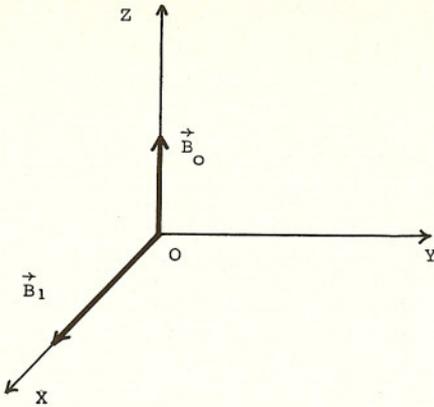


Figure 5 : In the rotating reference frame \vec{S} associated with the 2-level atom precesses around 2 static fields \vec{B}_0 and \vec{B}_1 respectively parallel to OZ and OX and proportional to $\omega_0 - \omega_L$ and ω_1 .

7). Naïve approach based on "Bloch's equations"

Let's first give a naïve approach of the problem, strongly suggested by the analogy with a magnetic resonance experiment, but which, in the present case, is incorrect. Then, in trying to understand where is the mistake, we will get some physical insight into the problem.

It seems reasonable to describe spontaneous emission by some damping terms in the equations of motion of $\langle \vec{S}(t) \rangle$. The transfer of atoms from e to g with a rate Γ can be described by $\langle \dot{S}_Z(t) \rangle = \Gamma (S_0 - \langle S_Z(t) \rangle)$, where $S_0 = -1/2$ (after a time large compared to $\tau = \Gamma^{-1}$, all atoms are in g, so that $\langle S_Z \rangle = -1/2$). As g is not affected by spontaneous emission, $\langle S_{\pm}(t) \rangle$ are damped to zero with a rate $\Gamma/2$

$\langle \dot{S}_{\pm}(t) \rangle = -(\Gamma/2) \langle S_{\pm}(t) \rangle$. Adding these damping terms to the ones which describe the precession around \vec{B}_0 and \vec{B}_1 , one gets the following equations which can be considered as the Bloch's equations of the problem :

$$\begin{cases} \langle \dot{S}_Z(t) \rangle = -i(\omega_1/2) \langle S_+(t) \rangle - \Gamma \langle S_Z(t) \rangle + i(\omega_1/2) \langle S_-(t) \rangle + \Gamma S_0 & (3-a) \\ \langle \dot{S}_{\pm}(t) \rangle = - \left[(\Gamma/2) \pm i(\omega_L - \omega_0) \right] \langle S_{\pm}(t) \rangle \mp i\omega_1 \langle S_Z(t) \rangle & (3-b) \end{cases}$$

What is the solution of these equations for an atom flying through the laser beam ? After a transient regime which starts when the atom enters the laser beam at $t = 0$, and which lasts for a time of the order of $\tau = \Gamma^{-1}$ [damping time of the transient solutions of equations (3)], $\langle \vec{S}(t) \rangle$ gets a stationary value $\langle \vec{S} \rangle_{st}$, independant of t , and corresponding to the steady state solution of (3). This situation lasts during all the transit time T through the laser beam (remember that $T \gg \tau$). After that, the atom leaves the laser beam at time $t = T$, and $\langle \vec{S} \rangle$ damps to zero in a short time, of the order of τ .

At this stage, one is very tempted to consider that the light radiated by the atom corresponds to this evolution of $\langle \vec{S}(t) \rangle$ (we have to return from the ro-

tating to the laboratory reference frame) and, consequently, that its spectrum is given by the squared modulus of the F.T. of $\langle S_+(t) \rangle e^{i\omega_L t}$. If such a conclusion were correct, one would get first an elastic component, at frequency ω_L , representing the contribution of the forced steady state motion $\langle S_+ \rangle_{st} e^{i\omega_L t}$ of the dipole moment driven by the laser field and which, as we have seen above, is the main part of the motion of the dipole. Strictly speaking, this elastic component would have a non zero width $1/T$ (corresponding to the finite transit time T), much smaller however than Γ (as $T \gg \tau$). In addition, one would get a small inelastic component, associated with the 2 small transient regims appearing at the 2 small regions where the atom enters or leaves the laser beam. This would suggest that one can suppress these inelastic components just by eliminating the light coming from these 2 regions.

8). What is missing in this approach ? Importance of the fluctuations

The method we have just outlined is not correct. A mathematical argument for showing it is that, when we calculate the squared modulus of the F.T. of $\langle S_+(t) \rangle$, we find an expression analogous to (2), but where $\langle S_+(t) S_-(t') \rangle$ is replaced by $\langle S_+(t) \rangle \langle S_-(t') \rangle$, and these 2 quantities are not equal.

It is perhaps more interesting to try to understand physically where is the mistake. The important point is that the light emitted by the atom is not radiated by its average dipole moment represented by $\langle S_{\pm}(t) \rangle$, but by its instantaneous dipole moment $S_{\pm}(t)$, and, even though the effect of spontaneous emission on $\langle \vec{S}(t) \rangle$ may be shown to be correctly described by the damping terms of equations (3), such a description is incorrect for $\vec{S}(t)$.

Let's try to visualize the evolution of $\vec{S}(t)$. We can consider the atom as being constantly "shaked" by the "vacuum fluctuations" of the quantized electromagnetic field (²³). These random fluctuations, which have an extremely short correlation time, have a cumulative effect on the atom in the sense that they damp $\langle \vec{S}(t) \rangle$, but we must not forget that they make the instantaneous dipole moment $S_{\pm}(t)$ fluctuate permanently around its mean value. The light which comes out is radiated not only by the mean motion of the dipole, but also by its fluctuations around the mean motion.

When we consider the effect of atoms on the incident electromagnetic wave which drives them, i.e. when we study how they absorb or amplify this wave, the average motion $\langle \vec{S}(t) \rangle$ is very important since it has definite phase relations with the driving field. The fluctuations of $\vec{S}(t)$ act only as a source of noise and can be ignored in a first step. In the problem we are studying here, we cannot ignore the fluctuations since they play an essential role : we are interested in spontaneous emission, not in absorption or induced emission, and the fluctuations of $S_{\pm}(t)$ enti-

rely determine the inelastic part of the fluorescence spectrum as we will show it now.

9). Elastic and inelastic parts of the fluorescence spectrum

Let us write :

$$S_{\pm}(t) = \langle S_{\pm}(t) \rangle + \delta S_{\pm}(t) \quad (4)$$

where $\delta S_{\pm}(t)$ is the deviation from the average value and obviously satisfies :

$$\langle \delta S_{\pm}(t) \rangle = 0 \quad (5)$$

Inserting (4) into (2), and using (5), one gets immediately :

$$\langle S_{+}(t) S_{-}(t') \rangle = \langle S_{+}(t) \rangle \langle S_{-}(t') \rangle + \langle \delta S_{+}(t) \delta S_{-}(t') \rangle \quad (6)$$

One clearly sees from (6) that, in the spectrum of the fluorescence light, there is an elastic component corresponding to the first term of (6) and which is the light radiated by the average motion of the dipole. In addition, we get an inelastic component corresponding to the last term of (6) and which is the light radiated by the fluctuations. The spectrum of this inelastic part is determined by the temporal dependence of these fluctuations, i.e. by their dynamics.

Before studying this problem, let us show how it is possible to derive simple expressions for the total intensity radiated elastically and inelastically, I_{el} and I_{inel} . Integrating (2) over ω , one gets a $\delta(t-t')$ function which gives when using (6) :

$$\begin{aligned} I_{el} &\sim \int_0^T dt |\langle S_{+}(t) \rangle|^2 \\ I_{inel} &\sim \int_0^T dt \langle \delta S_{+}(t) \delta S_{-}(t) \rangle = \int_0^T dt \left[\langle S_{+}(t) S_{-}(t) \rangle - |\langle S_{+}(t) \rangle|^2 \right] \\ &= \int_0^T dt \left[\frac{1}{2} + \langle S_Z(t) \rangle - |\langle S_{+}(t) \rangle|^2 \right] \end{aligned} \quad (7)$$

(We have used the relation $S_{+}S_{-} = \vec{S}^2 - S_Z^2 + S_Z$ and the identities $\vec{S}^2 = 3/4$, $S_Z^2 = 1/4$ valid for a spin $1/2$).

A first remark concerning equations (7) is that, when we are interested in a total intensity (integrated over ω), only a knowledge of $\langle \vec{S}(t) \rangle$ is required. Bloch's equations (2) are sufficient. This justifies the use of such equations (or similar rate equations) for interpreting optical pumping or level crossings experiments where the measured signal is a total intensity integrated over frequencies ⁽²⁾ ⁽²⁷⁾ ⁽²⁸⁾ ⁽²⁹⁾. Interpreting a spectral distribution is more complicated as it requires the knowledge of 2 times averages such as $\langle S_{+}(t) S_{-}(t') \rangle$.

Let's come back to equations (7). As the 2 small transient regims near $t = 0$ and $t = T$ have a very small relative contribution (of the order of τ/T), we can replace in (7), $\langle S_{+}(t) \rangle$ and $\langle S_Z(t) \rangle$ by the steady state solution of

Bloch's equations $\langle S_+ \rangle_{st}$ and $\langle S_Z \rangle_{st}$. This clearly shows that I_{el} and I_{inel} are proportional to T and that the inelastic part of the fluorescence is radiated uniformly throughout the whole period of time spent by the atom in the laser beam, and not only at the beginning or at the end of this period, as suggested by the naive approach described above. The calculation of $\langle \vec{S} \rangle_{st}$ is straightforward and one gets :

$$\frac{I_{el}}{T} \sim \frac{\omega_1^2 \left[\Gamma^2 + 4(\omega_o - \omega_L)^2 \right]}{\left[\Gamma^2 + 4(\omega_o - \omega_L)^2 + 2\omega_1^2 \right]^2} \quad \frac{I_{inel}}{T} \sim \frac{2\omega_1^4}{\left[\Gamma^2 + 4(\omega_o - \omega_L)^2 + 2\omega_1^2 \right]^2} \quad (8)$$

For very low intensities of the light beam ($\omega_1 \ll \Gamma$, $|\omega_L - \omega_o|$), we find that I_{el} varies as ω_1^2 , i.e. as the light intensity I , whereas I_{inel} varies as ω_1^4 , i.e. as I^2 . Most of the light is scattered elastically and we can define a cross section for such a process which is well described by fig. 1. I_{inel} is much smaller and can be considered as due to non linear scattering processes of the type shown in fig. 2.

For very high intensities ($\omega_1 \gg \Gamma$, $|\omega_L - \omega_o|$), we find on the contrary that I_{el} tends to 0. This is due to the fact that the atomic transition is completely saturated : the 2 populations are equalized ($\langle S_Z \rangle_{st} = 0$) and the dipole moment is reduced to 0 ($\langle S_{\pm} \rangle_{st} = 0$). On the other hand, I_{inel} is very large and independant of the light intensity I (this appears clearly on the bracket of the last equation (7) which reduces to $1/2$ as $\langle S_Z \rangle_{st} = \langle S_+ \rangle_{st} = 0$). This means that the atom spends half of its time in e and cannot therefore emit more than $\frac{1}{2} \frac{T}{\tau}$ photons. Increasing the incident light intensity cannot change this number.

One therefore concludes that inelastic scattering, which is due to the fluctuations of S_+ , is predominant in strong resonant fields. If we ignore these fluctuations, we miss all the physics. One can finally try to understand why these fluctuations are so effective at high intensities ($I_{inel} \gg I_{el}$) whereas they have little influence at low intensities ($I_{inel} \ll I_{el}$). I think this is due to the fact that an atom is the more sensitive to the vacuum fluctuations the greater is the probability to find it in the excited state e . Some components of the vacuum fluctuations are resonant for the atom in e as they can induce it to emit spontaneously a photon whereas they can only produce a level shift of g . At low intensities, most of the atoms are in g and are not very sensitive to the vacuum fluctuations whereas at high intensities half of the atoms are in e and fluctuate appreciably.

10). How to study the dynamics of the fluctuations ?

Let's now discuss the temporal dependance of $\langle \delta S_+(t) \delta S_-(t') \rangle$.

Considering the physical discussion given above, it seems that a good idea would be to try to write down an equation of motion for $\vec{S}(t)$ [and not for $\langle \vec{S}(t) \rangle$] including the random character of the force exerted by vacuum fluctuations. These fluctuations have a cumulative effect on $\vec{S}(t)$ which we can try to describe by damping terms analogous to those appearing in (2). In addition, $\vec{S}(t)$ fluctuates around its mean value in a way which can be considered as resulting from the action of a random "Langevin force" $\vec{F}(t)$, having an extremely short correlation time and a zero average value (24). It is clear that some relations must exist between the damping coefficients Γ and the statistical properties of $\vec{F}(t)$ (relations between dissipation and fluctuations) but we will not consider this problem here since, hereafter, we will only use the ultra short memory character of $\vec{F}(t)$. So let's write for example for $S_+(t)$:

$$\dot{S}_+(t) = -\left[(\Gamma/2) + i(\omega_L - \omega_0) \right] S_+(t) - i\omega_1 S_Z(t) + F_+(t) \quad (9)$$

When averaged, (9) reduces to equation (3-b) since $\langle F_+(t) \rangle = 0$.

Consider now the product $S_+(t) S_-(t')$ with $t > t'$, and let's try to understand how it varies with t . When calculating $\frac{d}{dt} S_+(t) S_-(t')$ and using (9) for $dS_+(t)/dt$, the only difficulty which appears comes from the Langevin term $F_+(t) S_-(t')$, since we know very little about $F_+(t)$. But we only need to calculate $d \langle S_+(t) S_-(t') \rangle / dt$, so that we only need to calculate the average $\langle F_+(t) S_-(t') \rangle$. And it is easy to understand that such an average gives 0 since the motion of the dipole at t' , $S_-(t')$, cannot be correlated with the Langevin force $F_+(t)$ at a later time t , as a consequence of the ultra short correlation time of $F_+(t)$. It follows that the rate of the t -variation of the 3 correlation functions $\langle S_i(t) S_-(t') \rangle$ (with $t > t'$, and $i = +, -, Z$) is described by a set of 3 first order differential equations with the same coefficients as the ones appearing in the Bloch's equations giving the rate of variation of $\langle S_i(t) \rangle$ [For $t' > t$, we use the fact that, as $S_+ = (S_-)^+$, $\langle S_+(t) S_-(t') \rangle = \langle S_+(t') S_-(t) \rangle^*$]. This important result is a particular case of the "quantum regression theorem" (25). In the present case, it means that, when the dipole undergoes a fluctuation and is removed from its steady state, the subsequent evolution and the damping of this fluctuation are the same as the transient behaviour of the mean dipole moment starting from a non steady state initial condition.

11). Predicted fluorescence spectrum for an ideal laser light

Once we know how to calculate the dynamics of the fluctuations of $S(t)$, the derivation of $\mathcal{J}(\omega)$ from (2) is simple. We bypass here the corresponding algebra which is straightforward and only give the results for a resonant irradiation ($\omega_L = \omega_0$) and a very high intensity ($\omega_1 \gg \Gamma$). One finds 3 components in the

inelastic spectrum : one central component around $\omega = \omega_L$ with a half-width $\Gamma/2$, and 2 equal sidebands around $\omega = \omega_L \pm \omega_1$, with a half-width $3\Gamma/4$ and a height 3 times smaller than the one of the central component.

Such a structure is simple to understand. The 2 sidebands correspond to the modulation of S_Y due to the transient precession of \vec{S} around \vec{B}_1 at frequency ω_1 (see fig. 5; as we are at resonance, $B_0 = 0$). As the projection of \vec{S} in the plane YOZ perpendicular to \vec{B}_1 is alternatively parallel to OY and OZ, and as the 2 damping coefficients associated to S_Z and S_Y are respectively Γ and $\Gamma/2$ (see equations 2), one understands why, when $\omega_1 \gg \Gamma$, the damping of the precession around \vec{B}_1 is given by $[\Gamma + (\Gamma/2)]/2 = 3\Gamma/4$ and this explains the width $3\Gamma/4$ of the 2 sidebands. The central component is associated with the transient behaviour of S_X , which is not modulated by the precession around \vec{B}_1 and which has a damping coefficient $\Gamma/2$. This explains the position and the width of the central component.

This result has been derived by several authors using either a classical ⁽¹⁰⁾ or a quantum ⁽¹³⁾⁽¹⁷⁾ description of the laser field. Other calculations don't give the same quantitative results ⁽¹²⁾⁽¹⁵⁾⁽¹⁶⁾. I think they are based upon too crude approximations (as the one which neglects the interference between different cascading amplitudes in the dressed atom approach described above).

12). Experimental situation

The experiment of Schuda, Stroud and Hercher ⁽³⁾ has displayed a 3-peak structure. The precision is perhaps not yet sufficient to allow a quantitative comparison between theory and experiment.

Other experiments are presently being made ⁽⁴⁾. The experimental investigations are rather difficult due to several perturbing effects. One is the spatial inhomogeneity of the laser intensity. As the interval travelled by the atom during its radiative lifetime is short compared to the diameter of the laser beam, each part of the illuminated portion of the atomic beam radiates a 3-peak spectrum with a splitting ω_1 corresponding to the local amplitude of the laser field. A too large spreading of this amplitude would wash out the structure. We must not also forget the elastic component which is not completely negligible when ω_1 is not very large compared to Γ . Let's take for example $\omega_1 = 2\Gamma$, in order to have the 3 peaks just well resolved. From (8), one calculates $I_{el}/I_{inel} = 1/8$. But I_{el} is spread over a very small interval (which is the width $\Delta\nu$ of the laser, or $1/T$), whereas I_{inel} is spread over Γ , or even over $\bar{\omega}_1$, if the spreading of ω_1 is sufficiently large to mask the structure. The ratio between the maxima of the elastic and inelastic components is therefore not $1/8$ but $\bar{\omega}_1/8\Delta\nu$, a number which may be much greater than 1. In such a case, one can get the impression that there is only one elastic component emerging

from a broad background. We must have $\omega_1 \gg \Gamma$ in order to have no trouble with the elastic component.

Other possible perturbations of the spectrum calculated above might come from temporal fluctuations of the laser beam. This leads us to the more general problem of the fluorescence light scattered by an atom irradiated by a resonant light which is not an ideal laser light with perfectly well defined phase and amplitude.

13). What happens with a real non ideal laser beam ?

Let's consider a realistic laser light, having a non zero spectral width $\Delta\nu$ and a very large intensity. More precisely, we suppose $\sqrt{\omega_1^2} \gg \Gamma$, $\Delta\nu$ where $\sqrt{\omega_1^2}$ is the mean Rabi nutation frequency associated with the probability distribution of the amplitude of the laser. We don't make any hypothesis concerning the relative magnitude of Γ and $\Delta\nu$.

A first important remark is that the knowledge of $\Delta\nu$ is not sufficient for characterizing the light beam. One can imagine different light beams having all the same spectral width $\Delta\nu$, i.e. the same first order correlation function, but completely different microscopic behaviours, corresponding to different higher order correlation functions (²⁰). One can for example consider a light beam emitted by a laser well above threshold, which has a very well defined amplitude undergoing very small fluctuations, and a phase $\phi(t)$ which, in addition to short time fluctuations, slowly diffuses in the complex plane with a characteristic time $1/\Delta\nu$. At the opposite, we can consider a quasi-monochromatic gaussian field, or a laser just above threshold, for which both phase and amplitude fluctuate appreciably with the same characteristic time $1/\Delta\nu$.

We have done, in collaboration with P. Avan, calculations of the fluorescence spectrum corresponding to different models of laser beams (²⁶). These calculations show that the shape of this spectrum is very sensitive to the microstructure of the light beam. The 3-peak structure described above is only maintained when the fluctuations of the amplitude are sufficiently small. The 3 components are broadened differently in a way which depends not only on the phase diffusion, but also on the short time fluctuations of this phase $\phi(t)$ [more precisely of $d\phi/dt$]. When the fluctuations of the amplitude are too large, only the central component survives, superposed to a broad background having a width of the order of $\sqrt{\omega_1^2}$. This is easy to understand : there is a destructive interference of the various Rabi nutations around \vec{B}_1 , as a consequence of the too large spreading of the possible values of B_1 . To summarize these studies, one can say that they deal with the fluctuations of \vec{S} associated to the fluctuations of the driving field.

We are also investigating the sensitivity of level crossing signals⁽²⁷⁾ to the fluctuations of the laser beam. The only calculations which have been performed up to now suppose, either a pure coherent field⁽²⁸⁾ (2) or a very broad line excitation ($\Delta\nu \gg \Gamma$, $\sqrt{\omega_1^2}$) so that, within the correlation time of the light wave, at most one interaction between the atom and the light can occur⁽²⁾ (29) : in such a case, only the first order correlation function plays a role. It would be interesting to try to fill the gap between these 2 extreme situations.

I would like to conclude with the following remark. The Hanbury-Brown and Twiss experiment has revealed the importance of new experimental methods, such as intensity correlations or photon coincidences, for learning more about light beams⁽²⁰⁾. Perhaps, the behaviour of atoms in strong resonant fields could appear as a new interesting probe for exploring such fields.

References

- (1) W. Heitler - Quantum Theory of Radiation, 3rd Ed. (1954, London, Oxford Un. Press)
- (2) C. Cohen-Tannoudji - Optical Pumping with Lasers. Proceedings of the 4th International Conference on Atomic Physics, in Atomic Physics 4, Plenum Press, p. 589 (1975)
- (3) F. Schuda, C.R. Stroud Jr., M. Hercher - J. Phys. B 7, L 198 (1974)
- (4) H. Walther - Atomic Fluorescence under Monochromatic Excitation. Proceedings of the 2nd International Laser Spectroscopy Conference, present volume
- (5) S. Ezekiel - Private communication
- (6) P.A. Apanasevich - Optics and Spectroscopy, 16, 387 (1964)
- (7) S.M. Bergmann - J. Math. Phys. 8, 159 (1967)
- (8) M.C. Newstein - Phys. Rev. 167, 89 (1968)
- (9) V.A. Morozov - Optics and Spectroscopy 26, 62 (1969)
- (10) B.R. Mollow - Phys. Rev. 188, 1969 (1969)
- (11) M.L. Ter -Mikaelyan and A.O. Melikyan - Soviet Physics JETP, 31, 153 (1970)
- (12) C.R. Stroud Jr. - Phys. Rev. A3, 1044 (1971) and Coherence and Quantum Optics ed. L. Mandel and E. Wolf (New York, London, Plenum Press), p. 537 (1972)
- (13) G. Oliver, E. Ressayre and A. Tallet - Lettere al Nuovo Cimento, 2, 777 (1971)
- (14) R. Gush, and H.P. Gush - Phys. Rev. A6, 129 (1972)
- (15) G.S. Agarwal - Quantum Optics, p. 108, Springer Tracts in Modern Physics (1974)
- (16) M.E. Smithers and H.S. Friedhoff - J. Phys. B7, L 432 (1974)
- (17) H.J. Carmichael and D.F. Walls - J. Phys. B8, L 77 (1975)
- (18) C. Cohen-Tannoudji - Cargese Lectures in Physics, vol. 2, p. 347, edited by M. Lévy, Gordon and Breach, New York, 1968
- (19) S. Haroche - Ann. de Phys. 6, 189 and 327 (1971)
- (20) N. Kroll in Quantum Optics and Electronics, p. 47, Les Houches 1964, edited by C. De Witt, A. Blandin and C. Cohen-Tannoudji, Gordon and Breach, New York (1965)
- (21) R.J. Glauber in Quantum Optics and Electronics, p. 63, Les Houches 1964, same reference as (19)
- (22) C. Cohen-Tannoudji, B. Diu and F. Laloë - Mécanique Quantique, p. 423, Hermann Paris, 1973
- (23) J.P. Barrat and C. Cohen-Tannoudji - J. Phys. Rad. 22, 329 and 443 (1961)
- (24) C. Cohen-Tannoudji - Ann. de Phys. 7, 423 and 469 (1962)
- (25) W. Happer - Rev. Mod. Phys. 44, 169 (1972)
- (26) In the Heisenberg picture, spontaneous emission can be described either by radiation reaction or by vacuum field effects.
- (27) J.R. Senitzky - Phys. Rev. Letters 31, 955 (1973)
- (28) P.W. Milonni, J.R. Ackerhalt and W.A. Smith - ibid. 958 (1973)

- (²⁴) A description of Langevin equation approach to damping phenomena may be found in M. Lax, Brandeis University Summer Institute Lectures, Vol. II, ed. by M. Chretien, E.P. Gross and S. Deser, Gordon and Breach, New York (1968)
 W.H. Louisell - Quantum Statistical Properties of Radiation, John Wiley and Sons, New York (1973)
 See also reference (¹⁵)
- (²⁵) M. Lax - Phys. Rev. 172, 350 (1968)
 M. Lax - Reference (²⁴)
- (²⁶) P. Avan and C. Cohen-Tannoudji - to be published
- (²⁷) For recent level crossing experiments done with single mode lasers and atomic beams, see
 W. Rasmussen, R. Schieder, H. Walther - Opt. Commun. 12, 315 (1974)
 H. Brand, W. Lange, J. Luther, B. Nottbeck and H.W. Schröder - Opt. Commun. 13, 286 (1975)
 See also reference (⁴)
- (²⁸) P. Avan and C. Cohen-Tannoudji - J. de Phys. Lettres, 36, L 85 (1975)
- (²⁹) M. Ducloy - Phys. Rev. A8, 1844 (1973)
 Phys. Rev. A9, 1319 (1974)
- Level crossing experiments done with broad line sources are described in
 M. Ducloy - Ann. de Phys. 8, 403 (1973-74)