

Dressed-atom description of resonance fluorescence and absorption spectra of a multi-level atom in an intense laser beam

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Abstract. A dressed-atom approach to resonance fluorescence in intense laser fields is presented. Simple and general results are derived which include the now well known predictions concerning two-level atoms but are not restricted to such simple cases. The positions of the various components of the fluorescence and absorption spectra are given by the allowed Bohr frequencies of the total system: atom + laser mode (dressed atom). The master equation describing spontaneous emission from the dressed atom is solved in the limit of high intensities. Simple expressions, taking into account the effect of cascades, are derived for the widths of the components. The intensities are related to the populations of the dressed-atom energy levels and to transition rates. Important differences between fluorescence and absorption spectra are explained. The existence of amplifying components in the absorption spectrum is pointed out.

1. Introduction

The problem of the fluorescence spectrum $L_f(\omega)$ of a beam of two-level atoms irradiated at right angles by an intense resonant laser beam has received a lot of attention (several theoretical references prior to 1975 are given in Cohen-Tannoudji 1975, 1976). The interest for this problem has been renewed by the recent experimental observation of such a spectrum on the sodium D_2 line by several independent groups (Schuda *et al* 1974, Walther 1975, Wu *et al* 1975). The first theoretical paper giving a complete calculation of the spectrum, including the non-resonant case, is Mollow (1969) which used a classical description of the laser field. Subsequent full quantum treatments have confirmed these results (Oliver *et al* 1971, Carmichael and Walls 1975, 1976, Hassan and Bullough 1975, Smithers and Freedhoff 1975, Kimble and Mandel 1976, Renaud *et al* 1976). See in particular Mollow (1975) in which the general quantum-mechanical applicability of the c -number applied field is pointed out.

Most of these calculations have been restricted to the simple situation of a two-level atom although some authors have suspected the neglected atomic states to be responsible for some experimentally observed asymmetries which were not theoretically predicted for a two-level atomic system. Furthermore, most of the quantum calculations which have been published, although giving correct results, do not seem to lead to simple general rules for evaluating the positions, the heights and the widths

of the various components of the fluorescence spectrum and which could be easily generalized to multi-level atoms.

In this paper we try to present a dressed-atom approach to resonance fluorescence in intense laser fields from which general results applicable to multi-level atoms and having a simple physical interpretation can be derived. The general idea of this method is to neglect, in the first step, spontaneous emission, and to determine the energy levels of the total isolated system: atom + laser photons (dressed atom). Resonance fluorescence can then be considered as spontaneous emission from such a system. The great advantage of such a method is that the Bohr frequencies of the dressed atom corresponding to allowed electric dipole transitions immediately give the positions of the various components of the fluorescence spectrum (at this stage of the calculation, one only needs to know the energy levels, i.e. to diagonalize an $r \times r$ matrix where r is the number of levels of the multi-level atom). It is of course necessary to go further and the purpose of this paper is to derive simple rules for evaluating the widths and the heights of these various components. For example, cascade effects are very important and must be taken into account (Cohen-Tannoudji 1975, Swain 1975), which means that a careful description of spontaneous emission is required (a simple application of Fermi's golden rule would be insufficient). We start in this paper from the master equation describing spontaneous emission for the dressed-atom density matrix (Cohen-Tannoudji 1976) and we show that in the limit of high laser intensity (Rabi nutation frequency ω_1 large compared to the natural width Γ) such a master equation has a very simple solution which allows a general discussion of the fluorescence spectrum of multi-level atoms.

Let us emphasize that all the calculations presented in this paper can be transposed into a c -number description of the laser field. For example, one can also neglect in the first step the spontaneous emission terms in the equations of motion of the atomic density matrix σ driven by a c -number field. This leads to a Hamiltonian equation of motion of σ which can be solved by diagonalizing the corresponding Hamiltonian (see for example Derbov *et al* 1975). For a two-level system, and with the rotating-wave approximation, such a diagonalization amounts to finding the energy levels of the equivalent fictitious spin $\frac{1}{2}$ in the rotating reference frame. At high intensities, one can then simplify the spontaneous emission terms by keeping only the secular ones in this basis. It is clear, however, that the necessity of such a diagonalization appears in a more natural way in the quantum approach, especially in the multi-level atom case, since it corresponds to the determination of the energy levels of the dressed atom. The physical picture of the dressed atom cascading down its energy diagram shows also how the photons contained in the laser mode are progressively replaced by fluorescence photons.

Another advantage of this approach, in our opinion, is that it leads to extremely simple expressions for the intensities of the various components of the fluorescence spectrum in an intense laser beam. We show in this paper that the total intensity of a given component is proportional to the total number of transitions having the corresponding Bohr frequency and occurring during the time T spent by the atom inside the laser beam, a result which has an obvious physical meaning. It follows that for evaluating the intensities of the various components, it is sufficient to calculate the populations of the dressed-atom energy levels and the transition rates between them. Such calculations which are not limited to a two-level atom and which take into account the steady-state value of the populations as well as their transient evolution when the atom enters the laser beam, allow a simple discussion of various

asymmetries which are expected in the fluorescence spectrum. The corresponding calculations do not appear in a natural way in the classical treatment.

Finally, the dressed-atom approach leads to very simple expressions for the absorption spectrum of a probe light beam by an atom interacting with an intense laser beam (in terms of transition rates and differences in population between the various levels of the dressed atom). This allows a clear understanding of the important differences between the fluorescence and absorption spectra, the first being, for example, a triplet in the case of a two-level atom, whereas the second is a doublet. One also easily predicts that at high intensities the absorption spectrum exhibits an equal number of absorbing and amplifying components.

In §2 we introduce the energy levels of the dressed atom and the matrix elements of the atomic dipole-moment operator. It is then possible to write down in §3 the master equation describing spontaneous emission from such a system. This equation can be simplified in intense fields through the secular approximation which implies that the various components of the fluorescence spectrum are not overlapping. The results of the calculations will therefore be valid only at the lowest order in Γ/ω_1 , where Γ and ω_1 have been introduced previously. As the energy diagram of the dressed atom has a periodic structure, some transfer coefficients between off-diagonal elements corresponding to the same Bohr frequency must be taken into account. The initial state of the dressed atom is defined in §4. The quasi-classical character of the laser field leads to a simple approximate solution of the equations of motion which exhibits a quasi-steady-state regime reached after a transient one and superimposed on a slow general drift down the energy diagram corresponding to the scattering of photons from the laser mode. This solution is used to determine the evolution of the populations and the mean dipole moment. One shows how the transfer coefficients introduced above change the damping times of the various frequency components of the mean dipole moment, which solves the problem of the influence of cascades. These results are finally applied in §§5 and 6 to the determination of the fluorescence and absorption spectra. Simple rules concerning the positions, the widths and the intensities of the various components of these spectra are established. Finally, a summary of the results is presented in §7.

2. Energy levels of the total system: atom + laser

We consider an atomic system resonantly driven by a single-mode laser field. Let e, e', \dots be the sublevels of the upper state; g, g', \dots be the sublevels of the lower one.

In the absence of coupling, the energy levels of the total system (atom + laser mode) are labelled by two quantum numbers: $e, e', \dots, g, g', \dots$ for the atom, n for the number of photons in the laser mode of frequency ω_L ($\hbar = c = 1$). Since the irradiation is quasi-resonant, the levels $|e, n\rangle, |e', n\rangle, \dots, |g, n+1\rangle, |g', n+1\rangle, \dots$ are quasi-degenerate and form an r -dimensional multiplicity \mathcal{E}_n . The unperturbed energy diagram has a periodic structure, two successive multiplicities $\mathcal{E}_n, \mathcal{E}_{n+1}$ being separated by ω_L .

In the so-called rotating-wave approximation (RWA), one keeps only the coupling between states of a given multiplicity. When diagonalizing the coupling Hamiltonian V within the r -dimensional multiplicity \mathcal{E}_n , one gets r perturbed states $|i, n\rangle$. In the following, we will suppose that the energies $n\omega_L + E_i$ of the $|i, n\rangle$ states are known,

as well as the expansion of these states in terms of $|e, n\rangle$, $|e', n\rangle, \dots, |g, n+1\rangle$, $|g', n+1\rangle, \dots$

The $|i, n\rangle$ states may be considered as the energy levels of the atomic system 'dressed' by the photons of the laser mode (dressed atom).

For studying spontaneous emission from such a system, the matrix elements of the atomic dipole-moment operator D between the $|i, n\rangle$ states are needed. Being an odd atomic operator, D cannot change the number n of photons and only connects a sublevel e of the upper state to a sublevel g of the lower state. From the expansion of $|i, n\rangle$, one easily deduces that the non-zero matrix elements of D are between two states $|i, n\rangle$ and $|j, n \pm 1\rangle$ belonging to adjacent multiplicities. We will put

$$\langle i, n | D | j, n \mp 1 \rangle = d_{ij}^{\pm}. \quad (2.1)$$

Note that it should be, in principle, necessary to add an index n to d_{ij}^{\pm} , but (see later §4.1) we only consider states of the laser field corresponding to a dispersion of the possible values of n having a small relative width Δn around the mean value \bar{n} ; as d_{ij} is a slowly varying function of n , one can ignore this variation and d_{ij}^{\pm} is an abbreviated notation for $d_{ij}^{\pm}(\bar{n})$.

As D is Hermitian, it follows that

$$d_{ij}^{\pm} = (d_{ji}^{\mp})^*. \quad (2.2)$$

Remark: Because of the quasi-resonant character of the laser irradiation, the matrix elements of the coupling V between different multiplicities have been neglected (RWA); virtual transitions to other atomic levels have also been ignored. If these approximations are relaxed, one finds that D has other non-zero matrix elements, for example between \mathcal{E}_n and $\mathcal{E}_{n \pm 3} \dots$ (matrix elements between \mathcal{E}_n and $\mathcal{E}_{n \pm 2}$ remain excluded by parity considerations). The new Bohr frequencies appearing in the motion of D describe non-linear processes such as harmonic generation.

3. Master equation describing spontaneous emission

3.1. Strong coupling assumption: secular approximation

The energy splittings $\omega_{ij} = E_i - E_j$ between two states $|i, n\rangle$ and $|j, n\rangle$ of the same multiplicity are supposed to be large compared to the natural width Γ of the excited sublevels e ,

$$|\omega_{ij}| \gg \Gamma. \quad (3.1a)$$

This implies either large detunings between the laser and atomic frequencies or, if two states $|e, n\rangle$ and $|g, n+1\rangle$ are degenerate in the absence of coupling, that the matrix element $\langle e, n | V | g, n+1 \rangle$ is large compared to Γ . As V is a linear combination of the creation and annihilation operators a^\dagger and a of the laser mode, such a matrix element is proportional to $\sqrt{\bar{n} \mp 1}$, i.e. to the amplitude of the laser electric field and is directly related to the Rabi nutation frequency ω_1 between e and g induced by the laser irradiation. At resonance, the laser intensity is therefore supposed to be sufficiently high to saturate the atomic transition.

Condition (3.1a) gives the possibility of neglecting, in the master equation describing spontaneous emission, any coupling between the diagonal elements of σ (populations) and the off-diagonal elements (secular approximation). This leads to a closed

system of equations for the populations which, as shown in the next paragraph, have a very simple physical meaning in terms of transition rates. Condition (3.1a) also means that the central component of the fluorescence spectrum at frequency ω_L (transitions $|i, n\rangle \rightarrow |i, n - 1\rangle$) does not overlap the lateral components at $\omega_L + \omega_{ij}$ (transitions $|i, n\rangle \rightarrow |j, n - 1\rangle$).

To simplify the discussion we will also suppose that the lateral components of the fluorescence spectrum are non-degenerate and not overlapping, i.e. that for any set of indices i, j, k, l (with $i \neq j, k \neq l, (i, j) \neq (k, l)$)

$$|\omega_{ij} - \omega_{kl}| \gg \Gamma. \tag{3.1b}$$

It is therefore possible to neglect any coupling by spontaneous emission between the off-diagonal elements of σ corresponding to the different Bohr frequencies $\omega_L + \omega_{ij}, \omega_L + \omega_{kl}, \dots$ of the lateral components.

Note, however, that the central component at frequency ω_L has an intrinsic r -fold degeneracy since it corresponds to r degenerate transitions $|i, n\rangle \rightarrow |i, n - 1\rangle$ (with $i = 1, 2, \dots, r$).

All the calculations presented in this paper are therefore valid only at the lowest order in $\Gamma/(\omega_{ij}), \Gamma/(\omega_{ij} - \omega_{kl})$.

Condition (3.1b) is more stringent than (3.1a) and we will indicate in the appendix how to generalize the calculations presented in this paper when several lateral components overlap. We will also describe briefly the modifications which must be introduced in the equations when condition (3.1a) is not fulfilled.

3.2. Evolution of the density matrix elements

The equations of evolution of σ are easily derived from the general theory of damping (Louisell 1973, Agarwal 1974, Cohen-Tannoudji 1976). The various elements of the density matrix $\sigma(t)$ will be noted

$$\langle i, n | \sigma(t) | j, n - p \rangle = \sigma_{ijm}^p(t). \tag{3.2}$$

Let us consider first the diagonal elements of σ , i.e. the populations $\sigma_{iin}^0(t)$ (see figure 1(a)). Because of the secular approximation, the coupling between populations and off-diagonal elements can be neglected in the master equation describing spontaneous emission (the error made being of the order of $\Gamma/|\omega_{ij}|$). σ_{iin}^0 decreases because of spontaneous transitions from $|i, n\rangle$ to a lower level $|j, n - 1\rangle$ with a rate given by†

$$\Gamma_{ji} = |\langle j, n - 1 | D | i, n \rangle|^2 = |d_{ij}^-|^2 = |d_{ji}^+|^2 \tag{3.3}$$

(note that in the notation Γ_{ji} , the transition is from i to j); σ_{iin}^0 increases because of spontaneous transitions from upper levels $|l, n + 1\rangle$ to $|i, n\rangle$ with a rate Γ_{il} (as in equation (2.1), the variation with n of the transfer coefficients Γ_{ji} and Γ_{il} is neglected).

† Several well known factors such as 2π , the density of photon states at $\omega_L \dots$ are reincluded in the definition of D , in order to get a simple expression for Γ .

Strictly speaking, D in (3.3) is the component of the vector operator D on one of the three unit vectors ϵ_q ($q = -1, 0, 1$) associated with the three polarizations σ^-, π, σ^+ of the fluorescence photon. The three corresponding values of Γ_{ji} must be added independently. Γ_{ji} can be written as $|d_{ji}^-|^2$ where $d_{ji}^- = \langle j, n | D | i, n + 1 \rangle$.

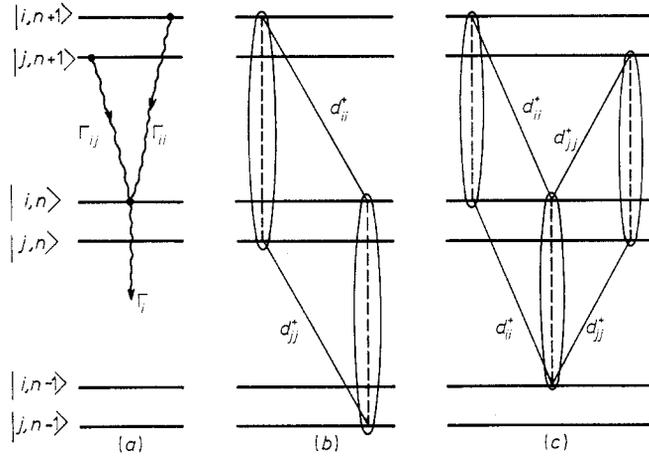


Figure 1. (a) The population of $|i, n\rangle$ decreases because of transitions to lower states (total transition rate Γ_i) and increases because of transitions from upper states (with rates $\Gamma_{ii}, \Gamma_{ij}, \dots$). (b) The 'coherence' between $|i, n\rangle$ and $|j, n-1\rangle$ with $i \neq j$ is damped with a rate $\frac{1}{2}(\Gamma_i + \Gamma_j)$. It is also coupled to the coherence between $|i, n+1\rangle$ and $|j, n\rangle$ corresponding to the same Bohr frequency. The coupling coefficient is proportional to the product $d_{jj}^+(d_{ii}^+)^*$ of matrix elements of D . (c) The coherence between $|i, n\rangle$ and $|i, n-1\rangle$ is coupled to the r coherences between $|j, n+1\rangle$ and $|j, n\rangle$ ($j = 1, 2, \dots, r$).

One gets therefore

$$\dot{\sigma}_{iin}^0(t) = - \sum_j \Gamma_{ji} \sigma_{iin}^0(t) + \sum_i \Gamma_{ii} \sigma_{iin+1}^0(t) = - \Gamma_i \sigma_{iin}^0(t) + \sum_i \Gamma_{ii} \sigma_{iin+1}^0(t) \quad (3.4)$$

where we introduce

$$\Gamma_i = \sum_j \Gamma_{ji} \quad (3.5)$$

(Γ_i is the total spontaneous transition rate from $|i, n\rangle$).

For the evolution of the off-diagonal elements of σ , for example for the elements $\sigma_{ijn}^{\pm 1}(t) = \sigma_{ijn}^{\pm}(t)$ appearing in the expression of $\langle D \rangle = \text{Tr } D\sigma(t)$ we will consider separately the cases for $i \neq j$ and $i = j$.

In the rate of variation of $\sigma_{ijn}^+(t)$ there is first a term proportional to $\sigma_{ijn}^+(t)$, describing the free evolution of $\sigma_{ijn}^+(t)$ at frequency $\omega_L + \omega_{ij}$ (the Lamb shifts of the atomic states are supposed to be reincluded in the atomic Hamiltonian) and the damping of $\sigma_{ijn}^+(t)$ which arises from spontaneous transitions from $|i, n\rangle$ or $|j, n-1\rangle$ to lower levels (the corresponding rate being $\frac{1}{2}(\Gamma_i + \Gamma_j)$). Furthermore, as the energy diagram has a periodic structure there is a coupling by spontaneous emission between $\sigma_{ijn}^+(t)$ and $\sigma_{ijn+1}^+(t)$, the corresponding coupling term being (see also figure 1(b))

$$\langle i, n | D | i, n+1 \rangle \langle i, n+1 | \sigma | j, n \rangle \langle j, n | D | j, n-1 \rangle = d_{ii}^- d_{jj}^+ \sigma_{ijn+1}^+(t) \quad (3.6)$$

(when the polarization is taken into account, $d_{ii}^- d_{jj}^+$ must be replaced by $d_{ii}^- \cdot d_{jj}^+$).

As will be shown later, this coupling is very important for the determination of the widths of the lateral components at $\omega_L + \omega_{ij}$. Similar couplings appear in the master equation describing spontaneous emission from a harmonic oscillator which also has a periodic energy diagram (Cohen-Tannoudji 1976). Ignoring them

would lead to a completely wrong result (dependence of the spectral width of the emitted light on the initial excitation).

Finally one gets for $\dot{\sigma}_{jn}^+(t)$:

$$\dot{\sigma}_{jn}^+(t) = -[i(\omega_L + \omega_{ij}) + \frac{1}{2}(\Gamma_i + \Gamma_j)]\sigma_{jn}^+(t) + d_{ii}^- d_{jj}^+ \sigma_{jn+1}^+(t). \quad (3.7)$$

The case of $\sigma_{jn}^+(t)$ is a little more complicated. There are several matrix elements $\sigma_{jn+1}^+(t)$ which have the same Bohr frequency ω_L as $\sigma_{jn}^+(t)$ and which are coupled to it with the coefficient (see figure 1(c))

$$\langle i, n | D | j, n + 1 \rangle \langle j, n | D | i, n - 1 \rangle = d_{ij}^- d_{ji}^+ = \Gamma_{ij} \quad (3.8)$$

(we have used (2.2) and (3.3)).

Equation (3.7) becomes now

$$\dot{\sigma}_{jn}^+(t) = -i(\omega_L + \Gamma_i)\sigma_{jn}^+(t) + \sum_j \Gamma_{ij}\sigma_{jn+1}^+(t). \quad (3.9)$$

Except for the free evolution term, this equation is exactly the same as equation (3.4) for the populations. More generally, equations (3.7) and (3.9) can be extended respectively to $\sigma_{jn}^p(t)$ and $\sigma_{jn}^s(t)$ provided that the free evolution terms $(\omega_L + \omega_{ij})$ and ω_L are replaced by $(p\omega_L + \omega_{ij})$ and $p\omega_L$.

To summarize the results of this section, let us call $\sigma(n, t)$ an ensemble of matrix elements of σ which are coupled together by the master equation describing spontaneous emission. For each value of n , $\sigma(n, t)$ is actually an ensemble of 1 or r matrix elements, for example $\{\sigma_{jn}^+(t)\}$ (1 element) or $\{\sigma_{in}^+(t) \ i = 1, 2, \dots, r\}$ (r elements). The master equation may be written as

$$\frac{d}{dt}\sigma(n, t) = -A\sigma(n, t) + B\sigma(n + 1, t) \quad (3.10)$$

where A and B are two matrices which are either of rank 1 (see for example (3.7)) or of rank r (see for example (3.4) or (3.9)).

4. Initial conditions and evolution of the system

4.1. Initial density matrix

We suppose that convenient slits define an irradiation zone such that the light intensity experienced by the moving atom rises abruptly from 0 to a constant value and then, after a transit time T , goes back to 0.

The state of the laser field is a quasi-classical one: the normalized distribution $p_0(n)$ of the possible values of the number n of photons in the laser mode has a dispersion Δn around the mean value \bar{n} which is large in absolute value, small in relative value:

$$\Delta n \gg 1 \quad (4.1a)$$

$$\Delta n \ll \bar{n}. \quad (4.1b)$$

Starting from the ground state g , the atom is suddenly coupled to the radiation field at $t = 0$ when it enters the laser beam. The initial state of the total system is therefore

$$\sigma(0) = |g\rangle\langle g| \otimes \sigma_R(0) \quad (4.2)$$

where $\sigma_{\mathbf{R}}(0)$ is the state of the radiation field. Taking the matrix elements of (4.2) in the basis $\{|i, n\rangle\}$ of the energy levels of the dressed atom, and using the fact that $p_0(n)$ varies very slowly with n (4.1a), one easily finds that $\sigma_{ij}^p(t=0)$ may be written as

$$\sigma_{ij}^p(t=0) = \rho_{ij}^p(t=0)p_0(n) \quad (4.3)$$

where the $\rho_{ij}^p(t=0)$ may be calculated from the expansion of $|i, n\rangle$ and from (4.2).

In the condensed notation of (3.10), (4.3) reads

$$\sigma(n, 0) = \rho(0)p_0(n). \quad (4.4)$$

4.2. Approximate resolution of the master equation

As $\sigma(n, t)$ is a slowly varying function of n , the last term of the master equation (3.10) can be rewritten

$$B\sigma(n+1, t) = B\sigma(n, t) + B\frac{\partial}{\partial n}\sigma(n, t) \quad (4.5)$$

so that equation (3.10) becomes

$$\frac{\partial}{\partial t}\sigma(n, t) = -A\sigma(n, t) + B\sigma(n, t) + B\frac{\partial}{\partial n}\sigma(n, t). \quad (4.6)$$

According to (4.4) the order of magnitude of the last term of (4.6) is $(B/\Delta n)\sigma(n, t)$ which is much smaller than $B\sigma(n, t)$ (see (4.1a)). It follows that the last term of (4.6) can be neglected in the first approximation and that the solution of this equation is

$$\sigma(n, t) = p_0(n)\rho(t) \quad (4.7)$$

where $\rho(t)$ is the solution of the equation

$$\frac{\partial \rho}{\partial t} = -(A - B)\rho(t). \quad (4.8)$$

After a transient regime which lasts a time of the order of $1/\Gamma$ ($\sim 1/\text{Re}(A - B)$) the system reaches a steady state. It is now easy to understand the effect of the last term of (4.6): it produces a drift of the distribution $p_0(n)$ towards lower values of n with a velocity of the order of B . Such a decrease in the mean number of photons of the laser mode corresponds of course to the emission of fluorescence photons in all other modes. In actual experiments the incident photons are also permanently emitted by the laser source, so that it would be, in principle, necessary to add some source terms in the master equation. However, the losses introduced by the atomic beam are so small and the transient time so short that one can completely ignore the last term of (4.6) as well as other new source terms and keep (4.7) and (4.8) as the solutions of the equation of motion.

As mentioned in the introduction, equation (4.8) can be shown to be equivalent to the one which would be obtained with a c -number description of the laser field after diagonalizing the undamped part of the Hamiltonian and keeping only the secular spontaneous emission terms.

4.3. Evolution of the populations

As a first application of the previous results, let us consider the evolution of the populations $\sigma_{ii}^0(n, t)$ of the energy levels $|i, n\rangle$. In this case, equation (4.7) reads

$$\sigma_{ii}^0(n, t) = p_0(n)\Pi_i(t) \tag{4.9}$$

where $\Pi_i(t)$ is an abbreviated notation for $\rho_{ii}^0(t)$

$$\rho_{ii}^0(t) = \Pi_i(t). \tag{4.10}$$

Let $\Pi(t)$ be the r -component vector $\{\Pi_i(t)\}$, \mathcal{F} the corresponding $r \times r$ matrix $A - B$ appearing in (3.10).

Equation (4.8) becomes

$$\frac{d\Pi}{dt} = -\mathcal{F}\Pi. \tag{4.11}$$

The elements \mathcal{F}_{ij} of \mathcal{F} are easily computed from (3.4)

$$\mathcal{F}_{ij} = \Gamma_i\delta_{ij} - \Gamma_{ij} \tag{4.12}$$

where Γ_{ij} and Γ_i are given in (3.3) and (3.5).

It appears clearly from (3.5) and (4.12) that the sum of the r lines of \mathcal{F} is zero. It follows that \mathcal{F} has an eigenvalue which is zero and which corresponds to a steady-state regime for the populations.

To summarize, we can describe the evolution of the populations in the following terms. Starting from the initial values $\Pi_i(0)p_0(n)$, these populations reach a steady state $\Pi_i(+\infty)p_0(n)$ after a transient regime described by the $(r - 1)$ non-zero eigenvalues of \mathcal{F} . On a much longer time scale ($t > \Delta n/\Gamma$), an additional variation appears due to the drift of $p_0(n)$ (see §4.2). The transient time T is supposed to be too short to observe this drift. T is usually longer than the time constants of the transient regime although in some cases a few eigenvalues of \mathcal{F} can be sufficiently small that the steady-state regime cannot be reached in the time interval T .

Finally, let us mention that the steady-state solution of (4.12) usually satisfies

$$\Gamma_{ji}\Pi_i(+\infty) = \Gamma_{ij}\Pi_j(+\infty). \tag{4.13}$$

The number of transitions from i to j balances the number of transitions from j to i (detailed-balance condition). A necessary condition for (4.13) is that the Γ satisfy

$$\Gamma_{ij}\Gamma_{jk}\Gamma_{ki} = \Gamma_{ik}\Gamma_{kj}\Gamma_{ji} \tag{4.13'}$$

for all ensembles $\{i, j, k\}$.

4.4. Evolution of the mean dipole moment

In the Schrödinger representation, the raising part of the atomic dipole operator can be written as

$$D^+ = \sum_{i,j,n} |i, n\rangle\langle i, n|D|j, n - 1\rangle\langle j, n - 1| = \sum_{ij} D_{ij}^+ \tag{4.14}$$

where, according to (2.1),

$$D_{ij}^+ = \sum_n d_{ij}^+ |i, n\rangle\langle j, n - 1|. \tag{4.15}$$

It can be shown from (4.7) that the mean value of D_{ij}^+ satisfies

$$\langle D_{ij}^+ \rangle(t) = \text{Tr } \sigma(t) D_{ij}^+ = \sum_n d_{ij}^+ \sigma_{jn}^-(t) = d_{ij}^+ \rho_{ji}^-(t). \quad (4.16)$$

It follows that $\langle D_{ij}^+ \rangle(t)/d_{ij}^+$ and $\rho_{ji}^-(t) = (\rho_{ij}^+(t))^*$ satisfy the same equations of motion.

Let us first suppose that $i \neq j$. According to (3.7) and (4.8), $\rho_{ij}^+(t)$ is only coupled to itself. It follows that

$$\frac{d}{dt} \langle D_{ij}^+ \rangle(t) = i\Omega_{ij} \langle D_{ij}^+ \rangle(t) \quad (4.17)$$

where

$$\Omega_{ij} = \omega_L + \omega_{ij} + iL_{ij} \quad (4.18)$$

$$L_{ij} = \frac{1}{2}(\Gamma_i + \Gamma_j) - d_{ii}^- d_{jj}^+. \quad (4.19)$$

It may be easily shown (from the Schwarz inequality) that $L_{ij} > 0^\dagger$.

Equation (4.17) shows that the component of the dipole moment oscillating at the Bohr frequency $\omega_L + \omega_{ij}$ is damped with a time constant $1/L_{ij}$. Note that this damping constant L_{ij} is not the half sum of the natural width of the two dressed-atom levels connected by D_{ij}^+ . The transfer of coherence by spontaneous emission between two pairs of levels corresponding to the same Bohr frequencies changes the damping constant of $\langle D_{ij}^+ \rangle$. This effect is the consequence on the mean dipole moment of the interferences between different cascade amplitudes of a system having degenerate Bohr frequencies.

When $i = j$ the r mean values $\langle D_{ii}^+ \rangle(t)/d_{ii}^+$ which correspond to the same Bohr frequency ω_L satisfy the coupled equations of motion of the ρ_{ii}^+ . Let us call $\langle \mathcal{D}_c^+ \rangle(t)$ the r -component vector $\{\langle D_{ii}^+ \rangle(t)/d_{ii}^+\}$.

From (3.9) and (4.8), one deduces

$$\frac{d}{dt} \langle \mathcal{D}_c^+ \rangle(t) = i\Omega_c \langle \mathcal{D}_c^+ \rangle(t) \quad (4.20)$$

where Ω_c is the $r \times r$ matrix

$$\Omega_c = \omega_L I + i\mathcal{T} \quad (4.21)$$

I being the unit matrix and \mathcal{T} being defined in (4.12). It follows that there are r independent possible oscillations of the mean dipole moment at the frequency ω_L which are damped with time constants given by the inverse of eigenvalues of \mathcal{T} . Since one of these eigenvalues is 0 (see previous section) there is an undamped oscillation at frequency ω_L corresponding to the elastic component of the fluorescence spectrum.

5. Spectral distribution of the fluorescent light

5.1. Simple derivation of the essential results

From the previous discussion on the evolution of the mean dipole moment one expects the following results for the fluorescence spectrum (they will be more rigorously derived in the next section).

[†] In writing (4.18) and (4.19), we have supposed that d_{ii}^- and d_{jj}^+ are real, which is generally the case. Otherwise it would be necessary to separate the real and imaginary parts of L_{ij} .

(i) The number of components in the fluorescence spectrum is the number of different Bohr frequencies appearing in the evolution of the mean dipole moment $\langle D^+ \rangle(t)$. There is a central component at ω_L and $r(r - 1)$ sidebands at frequencies $\omega_L + \omega_{ij}$ symmetrically distributed with respect to ω_L since $\omega_{ij} = -\omega_{ji}$ (less if there are some forbidden transitions for which $d_{ij}^+ = 0$). Conditions (3.1) mean that these $r(r - 1) + 1$ components are supposed to be well separated.

(ii) The width of the lateral component at frequency $\omega_L + \omega_{ij}$ is the inverse of the (single) damping time constant of $\langle D_{ij}^+ \rangle(t)$, i.e. L_{ij} . The two components at frequency $\omega_L + \omega_{ij}$ and $\omega_L + \omega_{ji} = \omega_L - \omega_{ij}$ have the same width since $L_{ij} = L_{ji}$ (see (4.19)).

(iii) The weight $\mathcal{J}(\omega_L + \omega_{ij})$ of this lateral component at frequency $\omega_L + \omega_{ij}$, i.e. the total area under this component is proportional to the total number of atoms undergoing a $\omega_L + \omega_{ij}$ transition (for example $|i, n\rangle \rightarrow |j, n - 1\rangle$) during the transit time T , i.e. to†

$$\mathcal{J}(\omega_L + \omega_{ij}) = \int_0^T dt \sum_n \Gamma_{ji} \Pi_i(t) p_0(n) = \Gamma_{ji} \int_0^T \Pi_i(t) dt. \tag{5.1}$$

Usually the transit time T is much longer than the time constant associated with the transient regime of the atom entering the laser beam and the steady-state values $\Pi_i(\infty)$ of $\Pi_i(t)$ are not negligible. In such a case (5.1) reduces to

$$\mathcal{J}(\omega_L + \omega_{ij}) = T \Gamma_{ji} \Pi_i(\infty) \tag{5.2}$$

(we have neglected the contribution of the transient regime).

If in addition the detailed-balance condition (4.13) is satisfied, one deduces from (5.2) and (4.13) that

$$\mathcal{J}(\omega_L + \omega_{ij}) = \mathcal{J}(\omega_L - \omega_{ij}). \tag{5.3}$$

The fluorescence spectrum is therefore completely symmetric with respect to ω_L . It results, however, from this discussion that such a symmetry is only approximate (since we have neglected the transient regime) and can disappear if the steady-state contribution is not predominant (some $\Pi_i(\infty)$ being extremely small or one of the transient time constants associated with the eigenvalues of \mathcal{F} being longer than T). We will discuss in forthcoming papers several examples of such asymmetries.

(iv) The central component is a superposition of r components centred on ω_L but having different widths corresponding to the eigenvalues of \mathcal{F} . One of them is extremely narrow (width determined by the transit time, or by the laser linewidth) and corresponds to elastic scattering. The total weight of the central component is proportional to the total number of atoms undergoing an ω_L transition

$$\mathcal{J}(\omega_L) = \int_0^T dt \sum_i \sum_n \Gamma_{ii} \Pi_i(t) p_0(n) = \sum_i \Gamma_{ii} \int_0^T \Pi_i(t) dt. \tag{5.4}$$

† If the fluorescent light is detected with a given polarization $\hat{\epsilon}_D$, the coefficient Γ_{ji} in (5.1) must be replaced by $|\hat{\epsilon}_D \cdot d_{ji}^+|^2$. The fluorescence photons corresponding to a given transition $|i, n\rangle \rightarrow |j, n - 1\rangle$ have a given polarization d_{ji}^+ .

5.2. More rigorous treatment

The spectral distribution of the fluorescent light $L_f(\omega)$ is given by the Fourier transform of the correlation function of the atomic dipole moment. More precisely

$$L_f(\omega) = \frac{1}{\pi} \text{Re } g(\omega) \quad (5.5)$$

where

$$g(\omega) = \int_0^T dt' \int_0^T dt \langle D^+(t)D^-(t') \rangle \exp[-i\omega(t-t')]\theta(t-t'). \quad (5.6)$$

$D^+(t)$ and $D^-(t')$ are the raising and lowering parts of the Heisenberg atomic dipole operator; $\theta(t-t')$ the Heaviside function†.

According to (4.14)

$$\langle D^+(t)D^-(t') \rangle = \sum_{i \neq j} \langle D_{ij}^+(t)D^-(t') \rangle + \sum_i \langle D_{ii}^+(t)D^-(t') \rangle. \quad (5.7)$$

Let us call $g_{ij}(\omega)$ and $g_c(\omega)$ the quantities obtained when replacing in (5.6) $\langle D^+(t)D^-(t') \rangle$ respectively by $\langle D_{ij}^+(t)D^-(t') \rangle$ and $\sum_i \langle D_{ii}^+(t)D^-(t') \rangle$. One gets

$$g(\omega) = \sum_{i \neq j} g_{ij}(\omega) + g_c(\omega) \quad (5.8)$$

$g_{ij}(\omega)$, $g_c(\omega)$ corresponding to the contribution to $L_f(\omega)$ respectively of the $(\omega_L + \omega_{ij})$ transitions and of the ω_L transitions.

The problem is therefore to evaluate the correlation functions $\langle D_{ij}^+(t)D^-(t') \rangle$ of the atomic dipole moment. From the quantum regression theorem (Lax 1968), which has been the basis of Mollow's and most subsequent papers, it is well known that, for $t > t'$, the two times average $\langle D_{ij}^+(t)D^-(t') \rangle$ satisfies the same equation of motion as the one time average $\langle D_{ij}^+(t) \rangle$, so that one can use the results of §4.4. It follows from (4.17) and (4.20) that for $t > t'$

$$\frac{d}{dt} \langle D_{ij}^+(t)D^-(t') \rangle = i\Omega_{ij} \langle D_{ij}^+(t)D^-(t') \rangle \quad (5.9)$$

$$\frac{d}{dt} \langle \mathcal{D}_c^+(t)D^-(t') \rangle = i\Omega_c \langle \mathcal{D}_c^+(t)D^-(t') \rangle \quad (5.10)$$

which can be integrated to give

$$\theta(t-t') \langle D_{ij}^+(t)D^-(t') \rangle = \exp[i\Omega_{ij}(t-t')] \langle D_{ij}^+(t')D^-(t') \rangle \theta(t-t') \quad (5.11)$$

$$\theta(t-t') \langle \mathcal{D}_c^+(t)D^-(t') \rangle = \exp[i\Omega_c(t-t')] \langle \mathcal{D}_c^+(t')D^-(t') \rangle \theta(t-t'). \quad (5.12)$$

Let us first integrate (5.6) over $t-t'$, the upper bound of the integral being extended to $+\infty$ (the finite value T of the real upper bound introduces a small additional broadening of the order of $1/T$ which affects essentially the elastic component).

We will first evaluate $g_{ij}(\omega)$. (5.11) leads to

$$g_{ij}(\omega) = \frac{1}{i(\omega - \Omega_{ij})} \int_0^T dt' \langle D_{ij}^+(t')D^-(t') \rangle. \quad (5.13)$$

† In writing (5.6) we suppose that the fluorescent light is collected during the whole transit time T of the atom through the laser beam. Convenient slits remove the light emitted by the atoms having left the laser beam.

From (4.15) and (4.7)

$$\begin{aligned} \langle D_{ij}^+(t)D^-(t) \rangle &= \sum_n \text{Tr} \sigma(t) d_{ij}^+ |i, n\rangle \langle j, n-1| D^- \\ &= \sum_{n,l} d_{ij}^+ d_{jl}^- \sigma_{ln}^0(t) = \sum_l d_{ij}^+ d_{jl}^- \rho_{li}^0(t). \end{aligned} \tag{5.14}$$

If $l \neq i$ it may be easily shown from (4.8) that

$$\rho_{li}^0(t) = \exp(-i\omega_{li}t) \exp(-L_{li}t) \rho_{li}^0(0) \tag{5.15}$$

($\rho_{li}^0(t)$ is only coupled to itself). The steady-state value of $\rho_{li}^0(t)$ is zero so that

$$\left| \int_0^T \rho_{li}^0(t) dt \right| \sim \left| \int_0^\infty \rho_{li}^0(t) dt \right| \sim |\rho_{li}^0(0)/\omega_{li}| \tag{5.16}$$

(we have used condition (3.1) for neglecting L_{li} in comparison to ω_{li}). The diagonal terms $\rho_{ii}^0(t) = \Pi_i(t)$ have a steady-state value which contributes to (5.14). Even if the steady-state value is negligible, the integral over t' of the transient regime is of the order $\Pi_i(0)/\Gamma_i$ which is much larger than (5.16) according to (3.1). It is therefore possible to neglect all the off-diagonal terms of (5.14) and to write (we use equations (3.3))

$$\int_0^T dt' \langle D_{ij}^+(t')D^-(t') \rangle = \int_0^T dt' d_{ij}^+ d_{ji}^- \Pi_i(t') = \Gamma_{ji} \int_0^T \Pi_i(t) dt. \tag{5.17}$$

This shows that the integral in (5.13) is real. Finally the contribution of D_{ij}^+ to $L_r(\omega)$ is, according to (5.13), (4.18) and (5.5)

$$\frac{2L_{ij}}{(\omega - \omega_L - \omega_{ij})^2 + (L_{ij})^2} \Gamma_{ji} \int_0^T \Pi_i(t) dt \tag{5.18}$$

which confirms the previsions of the earlier sections concerning the positions, the widths and the weights of the lateral components of the fluorescence spectrum.

The same method applies to the central component, the only difference being that Ω_c is now an $r \times r$ matrix. (5.12) leads to

$$\int_0^\infty dt(t-t') \langle \mathcal{D}_c^+(t)D^-(t') \rangle \exp[-i\omega(t-t')] = \frac{1}{i(\omega - \Omega_c)} \langle \mathcal{D}_c^+(t)D^-(t) \rangle. \tag{5.19}$$

Coming back to the definition of \mathcal{D}_c^+

$$D_{ii}^+(t) = d_{ii}^+(\mathcal{D}_c^+(t))_i \tag{5.20}$$

one gets

$$g_c(\omega) = \sum_{ij} d_{ii}^+ \left[\frac{1}{i(\omega - \Omega_c)} \right]_{ij} \int_0^T dt' \left\langle \frac{D_{jj}^+(t')}{d_{jj}^+} D^-(t') \right\rangle. \tag{5.21}$$

The same considerations as above lead to

$$\int_0^T dt' \langle D_{jj}^+(t')D^-(t') \rangle = d_{jj}^+ d_{jj}^- \int_0^T \Pi_j(t') dt'. \tag{5.22}$$

Equation (5.21) simplifies to

$$g_c(\omega) = \sum_{ij} d_{ii}^+ \left[\frac{1}{i(\omega - \Omega_c)} \right]_{ij} d_{jj}^- \int_0^T \Pi_j(t') dt'. \tag{5.23}$$

Inverting the matrix $\omega - \Omega_c$ gives the explicit form of the central component of the fluorescence spectrum. The elastic part of this component which is associated with the eigenvalue ω_L of Ω_c (eigenvalue 0 of \mathcal{F}) can be evaluated from the residue at $\omega = \omega_L$ of expression (5.23). A simple calculation (Reynaud 1977) gives†

$$L_{f,el}(\omega) = \left(\sum_i d_{ii}^+ \Pi_i(+\infty) \right) \left(\sum_j d_{jj}^- \int_0^T \Pi_j(t') dt' \right) \delta(\omega - \omega_L). \quad (5.24)$$

When the steady-state contribution is predominant, (5.24) becomes

$$L_{f,el}(\omega) = T \left(\sum_i d_{ii}^+ \Pi_i(+\infty) \right) \left(\sum_j d_{jj}^- \Pi_j(-\infty) \right) \delta(\omega - \omega_L). \quad (5.25)$$

Integrating (5.21) over ω gives the total weight of the central component

$$\mathcal{J}(\omega_L) = \sum_i \int_0^T dt' \langle D_{ii}^+(t') D^-(t') \rangle = \sum_i \Gamma_{ii} \int_0^T dt' \Pi_i(t'). \quad (5.26)$$

6. Absorption spectrum

In addition to the strong laser light which saturates the atomic transition, we now consider a second tunable monochromatic laser beam irradiating the atomic beam at right angles. We are interested in the absorption spectrum observed on this probe beam which is supposed to have a sufficiently weak intensity not to perturb the dressed-atom evolution. The absorption spectrum is given by the Fourier transform of the correlation function (Mollow 1972)

$$\langle [D^-(t), D^+(t')] \rangle. \quad (6.1)$$

The first term $D^-(t)D^+(t')$ is associated with absorption processes whereas the second one $-D^+(t')D^-(t)$, which has an opposite sign, corresponds to stimulated emission.

The calculation of the absorption spectrum from (6.1) follows the same lines as in §5.2 and we will give only the results. The various components have the same positions and widths as in the fluorescence spectrum. The central component at $\omega = \omega_L$ disappears. The total weight of a lateral component at $\omega_L + \omega_{ij}$ is given by

$$\mathcal{J}_a(\omega_L + \omega_{ij}) = K \sum_n |\langle i, n | D | j, n-1 \rangle|^2 \int_0^T [\sigma_{jn-1}^0(t) - \sigma_{in}^0(t)] dt \quad (6.2)$$

i.e. according to (3.3) and (4.7)

$$\mathcal{J}_a(\omega_L + \omega_{ij}) = K \Gamma_{ji} \int_0^T [\Pi_j(t) - \Pi_i(t)] dt \quad (6.3)$$

where K is a coefficient proportional to the probe light intensity and to the optical depth of the atomic beam.

The interpretation of (6.2) and (6.3) is straightforward (see figure 2). The total absorption at $\omega_L + \omega_{ij}$ is proportional to the difference between the total number of absorption and stimulated emission processes at frequency $\omega_L + \omega_{ij}$ (upward and

† Strictly speaking, the $\delta(\omega - \omega_L)$ function has a finite width ($1/T$).

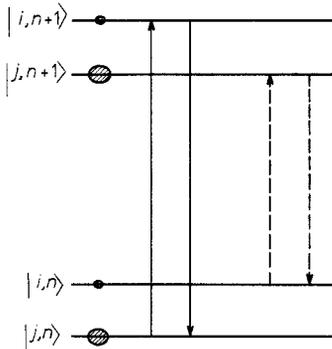


Figure 2. The total absorption at $\omega_L + \omega_{ij}$ is given by the difference between absorption processes from $|j, n\rangle$ to $|i, n + 1\rangle$ (upward full arrow) and stimulated emission from $|i, n + 1\rangle$ to $|j, n\rangle$ (downward full arrow).

If, as supposed on the figure, $\Pi_j > \Pi_i$, absorption is predominant. It follows that for the symmetrical component at $\omega_L + \omega_{ji} = \omega_L - \omega_{ij}$ (dotted arrows), stimulated emission is predominant.

downward full arrows on figure 2) occurring during the transit time T . As the stimulated emission and absorption rates are equal, this explains why the difference of populations $(\Pi_j - \Pi_i)$ between the lower and the upper levels appears in (6.3). This also explains why the central component vanishes in the absorption spectrum. This component is associated with the transitions $|i, n\rangle \rightarrow |i, n - 1\rangle$ ($i = 1, 2, \dots, r$) and according to (4.7) the two levels $|i, n\rangle$ and $|i, n - 1\rangle$ are equally populated (the difference between $p_0(n)$ and $p_0(n - 1)$ is completely negligible). Let us recall, however, that due to the secular approximation, the previous results are only valid to lowest order in $\Gamma/|\omega_{ij}|$. Higher order terms would give a non-zero contribution to the central component. From (6.3) we also deduce:

$$\Gamma_{ij} \mathcal{I}_a(\omega_L + \omega_{ij}) = -\Gamma_{ji} \mathcal{I}_a(\omega_L + \omega_{ji}). \tag{6.4}$$

This shows that one of the two symmetrical components $\omega_L + \omega_{ij}$ and $\omega_L + \omega_{ji}$ is necessarily amplifying whereas the other is absorbing.

When the steady-state contribution is predominant, (6.3) can be replaced by

$$\mathcal{I}_a(\omega_L + \omega_{ij}) = K\Gamma_{ji}[\Pi_j(+\infty) - \Pi_i(+\infty)]T. \tag{6.5}$$

If the detailed-balance condition (4.13) is fulfilled, one easily derives from (6.5)

$$\mathcal{I}_a(\omega_L + \omega_{ij}) + \mathcal{I}_a(\omega_L + \omega_{ji}) = \left(K(\Gamma_{ij} - \Gamma_{ji})^2 \frac{\Pi_i + \Pi_j}{\Gamma_{ij} + \Gamma_{ji}} T \right) > 0 \tag{6.6}$$

which shows that the absorbing component is more intense than the amplifying one.

Finally, the dressed-atom approach leads to very simple results for the absorption spectrum in an intense laser field. It explains why the central component disappears (leading to a different number of components in the fluorescence and absorption spectra). It predicts also an equal number of absorbing and amplifying components. For example, for a two-level atom, the fluorescence spectrum exhibits a triplet whereas the absorption spectrum contains one absorbing and one amplifying component which disappear (at zeroth order in Γ/ω_1) at resonance ($\omega_0 = \omega_L$) since $\Gamma_{12} = \Gamma_{21}$, $\Pi_1 = \Pi_2$.

Similar conclusions concerning the possibility of amplification by a medium which is not inverted but saturated by a strong resonant beam have been derived by various theories (Haroche and Hartman 1972 and references therein, Mollow 1972). The gain is, however, very small either as a consequence of Doppler averaging, or due to the fact that the pump irradiation is supposed to be resonant in which case the absorption of the probe vanishes in the zeroth order in Γ/ω_1 . We predict here that in the off-resonant case and for an atomic beam, the population difference $\Pi_j - \Pi_i$ can be quite important, leading to a significant gain per atom.

Note that we have considered the fluorescence and absorption spectra on the same atomic transitions as the one excited by the strong laser light. Transitions to atomic levels other than e, e', \dots and g, g', \dots exhibit only an r -component structure both in fluorescence and absorption spectra, all components being either absorbing or amplifying (Autler-Townes splitting) (Autler and Townes 1955).

The formalism developed in the present section could also be applied to the determination of the RF absorption spectra (transitions between states of the same multiplicity \mathcal{E}_n). This would allow a quantitative interpretation of double resonance experiments performed with very intense laser beams (in the limit of weak probing RF fields).

7. Conclusion

From the previous discussion, the following procedure can be derived for the computation of the fluorescence spectrum of a multi-level atom irradiated by an intense laser beam.

(i) Determination of the energy diagram of the dressed atom, i.e. diagonalization of the coupling Hamiltonian V in each multiplicity $|e, n\rangle, |e', n\rangle, \dots, |g, n+1\rangle, |g', n+1\rangle, \dots$ which gives the energies E_i and the expansion of the perturbed states $|i, n\rangle$. One gets immediately in this way the position of the various components of the fluorescence spectrum $\omega_L + \omega_{ij} = \omega_L + E_i - E_j$.

(ii) Computation of the matrix elements of the dipole moment in the $\{|i, n\rangle\}$ basis: $d_{ij}^- = \langle i, n-1 | D | j, n \rangle$ (2.1), and of the transition rates Γ_{ij} , Γ_i (from expressions (3.3) and (3.5) $\Gamma_{ji} = |d_{ji}^-|^2$, $\Gamma_i = \sum_j \Gamma_{ji}$). This gives immediately the width of the lateral components (through expression (4.19) $L_{ij} = \frac{1}{2}(\Gamma_i + \Gamma_j) - d_{ii}^- d_{jj}^+$).

(iii) Resolution of the master equation (4.11) for the populations. This gives the weights of the lateral components

$$\mathcal{I}(\omega_L + \omega_{ij}) = \Gamma_{ji} \int_0^T \Pi_i(t) dt$$

of the elastic component

$$\mathcal{I}_{e1} = \left(\sum_i d_{ii}^- \Pi_i(+\infty) \right) \left(\sum_j d_{jj}^+ \int_0^T \Pi_j(t) dt \right)$$

and the total weight of the central component

$$\mathcal{I}(\omega_L) = \sum_i \Gamma_{ii} \int_0^T \Pi_i(t) dt.$$

The comparison between the contributions of the transient and steady-state regimes gives indications about possible asymmetries.

(iv) If one is interested in the detailed shape of the central component, expression (5.23) must be explicitly evaluated.

(v) The absorption spectrum from a weak light probe beam is easily determined from the knowledge of the population differences $\Pi_j - \Pi_i$ and transition rates Γ_{ji} (expression (6.3)).

Let us recall that all previous results are only valid if the splitting between the components is large compared to their width (conditions (3.1)), and if the laser is in a quasi-classical state (conditions (4.1)).

The application of the general method presented in this paper to the particular case of a two-level atom leads to results in quantitative agreement with the now well known predictions concerning such a spectrum at high intensities. Since the dressed-atom approach to the fluorescence spectrum of a two-level system has already been discussed by Cohen-Tannoudji (1976), we will not come back to this problem (see also Reynaud 1977).

Our results will be applied in forthcoming publications to some problems involving the resonant interaction of multi-level atoms with intense laser beams. One of these problems is the modification of Raman scattering at very high intensities. Another one concerns polarization effects in resonance fluorescence (excitation and detection with different polarizations for example). A slight generalization of the present formalism also allows us to study the dynamic Stark splitting of two atomic transitions sharing a common level and simultaneously driven by two intense laser beams.

Let us finally mention the application of this approach to the interpretation of some asymmetries observed in the fluorescence spectrum of sodium. In contradiction with the results of Higgins (1975) using a three-level model, we have found that the neglected hyperfine levels can be responsible for some of the observed asymmetries (Reynaud 1977). This problem requires further study, however, since it has been shown recently (Wu *et al* 1976) that the laser intensity gradient plays an important role.

Appendix. Possible generalizations

We will first discuss the situation where (3.1*b*) is not fulfilled, (3.1*a*) being maintained. More precisely, we consider the possibility of overlapping lateral components remaining however well separated from the central component. Such a situation can occur, for example, when three levels i, j, k of the same multiplicity are equally spaced ($\omega_{ij} = \omega_{jk}$), the spacing being large compared to Γ .

The method for dealing with an s -fold degenerate (or quasi-degenerate) lateral component follows closely the treatment given in §5.2 for the r -fold degenerate central component. One introduces an s -component vector $\mathcal{D}_l^+ = D_{ij}^+/d_{ij}^+$ where (i, j) indicates the s degenerate (or quasi-degenerate) transitions $|i, n\rangle \rightarrow |j, n-1\rangle$, $|k, n\rangle \rightarrow |l, n-1\rangle \dots$ and one can easily, as for (4.20), derive the following matrix equation for $\langle \mathcal{D}_l^+(t) \rangle$

$$\frac{d}{dt} \langle \mathcal{D}_l^+(t) \rangle = i\Omega_l \langle \mathcal{D}_l^+(t) \rangle \quad (\text{A.1})$$

where Ω_l contains the various Bohr frequencies of the components of \mathcal{D}_l^+ and the various couplings between these components. A straightforward calculation following

the same steps as in §5.2 leads from (A.1) to

$$g_l(\omega) = \sum_{\substack{ij,kl \\ |\omega_{ij}-\omega_{kl}|\leq\Gamma}} d_{ij}^+ \left[\frac{1}{i(\omega - \Omega_i)} \right]_{ij,kl} \int_0^T dt' \left\langle \frac{D_{kl}^+(t')}{d_{kl}^+} D^-(t') \right\rangle \quad (\text{A.2})$$

the shape of the degenerate lateral component being given by $2\text{Re } g_l(\omega)$. As (3.1a) is maintained, the last integral of (5.28) can be transformed (see equation (5.17)) into

$$d_{lk}^- \int_0^T \Pi_k(t') dt'$$

which finally gives

$$g_l(\omega) = \sum_{\substack{ij,kl \\ |\omega_{ij}-\omega_{kl}|<\Gamma}} d_{ij}^+ \left[\frac{1}{i(\omega - \Omega_i)} \right]_{ij,kl} (d_{kl}^+)^* \int_0^T \Pi_k(t') dt'. \quad (\text{A.3})$$

Integrating (A.3) over ω , as for (5.26), one gets the total weight of the degenerate lateral component:

$$\mathcal{J}_1 = \sum_{i,j} \Gamma_{ji} \int_0^T \Pi_i(t') dt'. \quad (\text{A.4})$$

Releasing condition (3.1a) leads to a more difficult situation. In the master equation describing spontaneous emission the populations σ_{in}^0 can now be coupled to off-diagonal elements σ_{ijn}^0 between two states of the same multiplicity. Transfers are possible between σ_{in+1}^0 and σ_{ijn}^0 or between σ_{ijn+1}^0 and σ_{iin}^0 . It is still possible to factorize $\sigma(n, t)$ as in (4.7) but the equations obtained for $\rho(t)$ (equation (4.8)) cannot be separated into independent blocks of smaller dimensions. It is in general necessary to keep all the couplings between the various elements of $\rho(t)$. Because of these couplings one must drop the simple physical picture of a system flowing from one energy level to the other with definite transition rates. When (3.1a) is not fulfilled, the dressed-atom approach does not introduce any simplification.

It is actually better in this case to work in the bare-atom basis $\{|e, n\rangle, |e', n\rangle, \dots, |g, n+1\rangle, |g', n+1\rangle, \dots\}$ since spontaneous emission is described by simple equations in this basis. However the difficulties reappear, of course, in the terms describing the coupling with the laser.

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