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Simultaneous saturation of two atomic transitions sharing a common level

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Abstract. A dressed-atom approach, introduced in a previous paper, is generalised for studying several effects which could be observed on an atomic beam interacting with two monochromatic resonant laser beams saturating two atomic transitions sharing a common level, a situation which occurs frequently in stepwise excitation experiments. The problem of the optimisation of the population of the upper state is investigated. Analytical expressions are derived for the positions, the widths and the weights of the various components of the fluorescence and absorption spectra. The results obtained at the limit where one of the two lasers has a much weaker intensity than the other one are interpreted perturbatively by treating to lowest orders the scattering of the weak laser beam by the atom dressed by the intense one.

1. Introduction

When an atomic transition ab is saturated by a high-intensity resonant laser beam, a lot of interesting effects can be observed which have been extensively studied both theoretically and experimentally: for example, a triplet structure of the fluorescence spectrum $F_{ab}(\omega)$ emitted from the same transition ab has been predicted theoretically (Mollow 1969; see also some references to several subsequent works in Cohen-Tannoudji and Reynaud 1977a) and observed experimentally (Schuda *et al* 1974, Walther 1975, Hartig *et al* 1976, Wu *et al* 1975, Grove *et al* 1977). Another interesting effect is the doublet structure appearing in the absorption of a second weak laser beam (figure 1) probing another atomic transition bd or ad sharing a common level with the saturated transition. Such an effect, which was first discovered in the microwave region (Autler and Townes 1955) has been extensively studied recently in the optical region both theoretically (see for example Mollow 1972, Feneuille and Schweighofer 1975) and experimentally (Delsart and Keller 1976a, b, Picqué and Pinard 1976)†.

Up to now, the consequences of the simultaneous saturation of two atomic transitions ab and bc sharing a common level b (figure 2) by two intense laser beams with frequencies ω_L and ω_L' close to the atomic frequencies ω_0 and ω_0', have received

† Although we will consider only atomic beams in the following, we should mention that there are other theoretical and experimental works dealing with vapours and taking into account the Doppler effect (Beterov and Chebotaiev 1974 and references therein, Schabert *et al* 1975, Delsart and Keller 1976a, b).

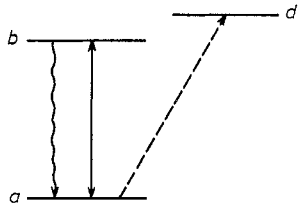

Figure 1. Saturation of an atomic transition ab by a high-intensity resonant laser beam (full arrow). One can observe the fluorescence spectrum emitted from the same transition (wavy arrow) or the absorption spectrum from another transition starting from a or b (for example ad, dotted arrow).

much less attention (see, however, Whitley and Stroud 1976) although such a situation occurs frequently in experiments using a stepwise excitation of atomic states. The first important question is to determine what the intensities of the two lasers should be in order to have the largest population for the upper level c. One can also observe the fluorescence spectra $F_{ab}(\omega)$ and $F_{bc}(\omega)$ emitted from the saturated transitions ab and bc; how the triplet structures observed for a single saturated transition are modified when the second transition is also saturated and how the splittings of the spectral lines are related to the Rabi nutation frequencies ω_1 and ω_1' characterising the coupling of the atom with the two laser beams. Suppose now that one observes the absorption spectrum from another transition sharing a common level with ab or bc, for example ad (see figure 2). It may be asked whether or not the saturation of the transition bc which has no common level with ad has any influence on the Autler–Townes doublet structure of ad associated with the saturation of ab.

Let us finally mention some intriguing questions which arise when one of the two lasers has a much weaker intensity than the other one. It occurs frequently that some answers which seem obvious are actually completely wrong. Consider for example the case where the ω_L' laser saturating bc is much weaker than the other one and let us try to understand $F_{bc}(\omega)$. One knows, of course, that the role of the intense ω_L laser does not reduce to populating b. It also produces a splitting of the absorption spectrum of the weak laser beam probing bc. One could observe such a doublet by measuring the variation of the total fluorescence light $\mathscr{I}_{bc}$ (integral over ω of $F_{bc}(\omega)$) as a function of the frequency ω_L' of the weak laser beam. However our problem here is completely

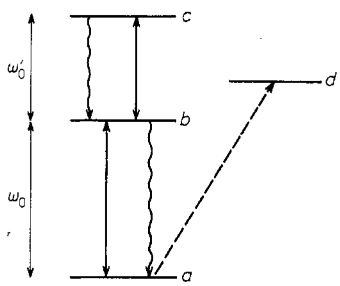

Figure 2. Simultaneous saturation of two atomic transitions ab and bc with frequencies ω_0 and ω_0' by two intense resonant laser beams (full arrows). One can observe the fluorescence spectra emitted from these transitions (wavy arrows) or the absorption spectrum from another transition starting from a, b or c (for example ad, dotted arrow).

different. We are interested in the fluorescence spectrum, i.e. in the variation with ω of $F_{bc}(\omega)$, the laser frequency ω'_L being fixed. A naïve approach based on the idea that b is split in two states would lead to the wrong conclusion that the fluorescence spectrum $F_{bc}(\omega)$ has the same structure as the absorption one.

The case where the ω_L laser is the weaker one is still more delicate. Since the lower level a is not directly coupled to the intense ω'_L laser, one is tempted to predict that, for sufficiently weak intensity of the ω_L laser, the fluorescence spectrum $F_{ab}(\omega)$ is mainly elastic (Rayleigh scattering). Since the ω_L laser is too weak to split the level b, one would also think that the fluorescence spectrum $F_{bc}(\omega)$ has the well known triplet structure of a single saturated transition, the role of the ω_L laser being just to populate b. We will see later on that these conclusions concerning $F_{ab}(\omega)$ and $F_{bc}(\omega)$ are both incorrect.

As a final example of a wrong conclusion which can be derived from too crude an approach to the problem, let us mention the idea that, as soon as the two intensities are sufficient to saturate ab and bc, the three populations are more or less equalised. We will actually show that increasing the intensity of one of the two lasers too much considerably reduces the population of the upper state c (even if the two intensities are sufficient to saturate each transition individually).

In this paper, we present a theoretical treatment of the various effects associated with the simultaneous saturation of two atomic transitions sharing a common level. We will follow closely the dressed-atom approach developed in a previous paper (Cohen-Tannoudji and Reynaud 1977a, to be referred to as I) and dealing with the interaction of an intense laser beam with multi-level atoms.

We will consider an atomic beam irradiated perpendicularly by the two saturating laser beams so that one can eliminate the Doppler effect.

We will suppose that a is the ground state and that, by spontaneous emission, the atom decays from c only to b and from b only to a, so that the a–b–c system can be considered as closed. We will denote the natural widths of c and b by γ' and γ.

To simplify, we will also forget the Zeeman degeneracy of a, b, c. As in recent experiments (Hartig *et al* 1976, Grove *et al* 1977), optical-pumping methods can be used to concentrate atoms in a single Zeeman sublevel of each level a, b, c.

Let us give a few examples of atomic transitions to which the calculations presented in this paper could be applied. One could first consider the two transitions $3s_{1/2}$, $F = 2 \leftrightarrow 3p_{3/2}$, $F = 3$ and $3p_{3/2}$, $F = 3 \leftrightarrow 3d_{5/2}$, $F = 4$ of Na. If the two lasers have right circular polarisation, optical pumping concentrates atoms in the higher m_F sublevels of each state. One is therefore led to a three-level system $a = 3s_{1/2}, F = 2, m_F = 2$; $b = 3p_{3/2}$, $F = 3$, $m_F = 3$; $c = 3d_{5/2}$, $F = 4$, $m_F = 4$, which obviously satisfies the assumptions made previously on spontaneous decay. The wavelengths of these two transitions are $\lambda_0 = 5890$ Å, $\lambda'_0 = 8195$ Å. It would seem easier to use the 3p–4d transition which has a more convenient wavelength ($\lambda'_0 = 5690$ Å) for the presently available dye lasers, but the level $4d_{5/2}$, $F = 4$, $m_F = 4$ decays not only to 3p but also to 4p so that the three-level system a, b, c would not be closed and so would require more elaborate calculations. Another interesting system is Li: the two transitions $2s_{1/2}$, $F = 2 \leftrightarrow 2p_{3/2}$, $F = 3$ and $2p_{3/2}$, $F = 3 \leftrightarrow 3d_{5/2}$, $F = 4$ have convenient wavelengths ($\lambda_0 = 6707$ Å, $\lambda'_0 = 6103$ Å) and one can neglect, during the interaction time, the spontaneous decay from 3d to 3p which has a very low probability.

In §2, the general method developed in I is extended to the present case: the energy diagram of the atom dressed by two resonant laser modes ($\omega_L = \omega_0$ and $\omega'_L = \omega'_0$) is determined and the transition rates between the various energy levels are evaluated.

The resolution of the corresponding master equation allows a quantitative study of the population of the upper state (§3) and of the fluorescence and absorption spectra (§4). In §5, the limiting cases where one of the two lasers has an intensity much weaker than the other are investigated and interpreted from a perturbative approach by treating to lowest orders the scattering of the weak laser beam by the atom dressed by the intense one. Finally (in §6), the results obtained in this paper are compared with those of previous publications.

2. Extension of the dressed-atom approach to the interaction with two laser beams

2.1. Unperturbed multiplicities

In addition to the atomic quantum number, a, b or c, two quantum numbers are now necessary to describe the state of the two lasers. The ket $|l, n, n'\rangle$, with $l = a, b, c$, will represent the state of the whole system corresponding to the atom in level l in the presence of n photons ω_L and n' photons ω'_L, with an unperturbed energy $E_l + n\omega_L + n'\omega'_L$ (we take $\hbar = 1$).

We will suppose that the two atomic frequencies ω_0 and ω'_0 are sufficiently different (a precise condition on $|\omega_0 - \omega'_0|$ will be given later on, see equation (2.3)) so that a given laser beam cannot be simultaneously resonant for these two transitions. In order to simplify the discussion, we will, in the following, restrict ourselves to the case of two resonant excitations,

$$\omega_L = \omega_0 \qquad \omega'_L = \omega'_0 \tag{2.1}$$

although the calculations presented in this paper could be easily generalised to a more general situation (Reynaud 1977).

It will be useful to introduce the three-fold degenerate multiplicities $\mathscr{E}_{n,n'}$ defined by:

$$\mathscr{E}_{n,n'} = \{|a, n+1, n'\rangle, |b, n, n'\rangle, |c, n, n'-1\rangle\}. \tag{2.2}$$

The shape of the unperturbed energy diagram is given in figure 3 where some multiplicities $\mathscr{E}_{n,n'}$ are represented by horizontal lines.

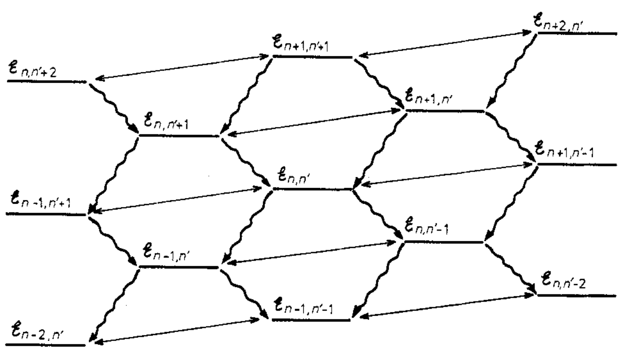

Figure 3. Energy diagram showing some of the (three-fold degenerate) unperturbed multiplicities $\mathscr{E}_{n,n'}$. The thin arrows correspond to neglected non-resonant couplings (which should be taken into account when $\omega_0 = \omega'_0$). The wavy arrows pointing to the left or to the right describe the emission of a fluorescence photon from transitions ab or bc respectively.

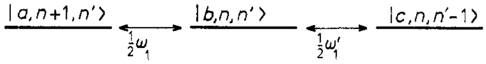

Figure 4. Unperturbed degenerate states of the multiplicity $\mathscr{E}_{n,n'}$. The thin arrows represent the couplings between them, proportional to the Rabi frequencies ω_1 and ω_1'.

2.2. Couplings

The couplings between the three unperturbed states of $\mathscr{E}_{n,n'}$ are represented in figure 4. An atom in a can absorb an ω_L photon and jump in b, which couples the two states $|a, n+1, n'\rangle$ and $|b, n, n'\rangle$ with an amplitude $\frac{1}{2}\omega_1$ proportional to the Rabi nutation frequency ω_1 characterising the coupling of the ω_L laser with the transition ab†. Similarly the two states $|b, n, n'\rangle$ and $|c, n, n'-1\rangle$ are coupled with an amplitude $\frac{1}{2}\omega_1'$ (ω_1 and ω_1', which are proportional to the amplitudes of the two laser waves ω_L and ω_L', will be supposed real and positive).

Strictly speaking, an atom in a can jump to b by absorbing an ω_L' photon. This process corresponds to a coupling between two states $|a, n+1, n'\rangle$ and $|b, n+1, n'-1\rangle$ which belong to two different multiplicities $\mathscr{E}_{n,n'}$ and $\mathscr{E}_{n+1,n'-1}$ (thin lines of figure 3). The amplitude of such a coupling is of the order of ω_1' (interaction with the ω_L' laser) and can be neglected if the energy difference $\omega_0 - \omega_0'$ between the two multiplicities is sufficiently large. Similar couplings, proportional to ω_1, also exist between different multiplicities. We will suppose

$$|\omega_0 - \omega_0'| \gg \omega_1, \omega_1' \tag{2.3}$$

so that one can neglect all these non-resonant couplings‡. We therefore exclude the accidental case of three equidistant levels a, b, c, which would require to take into account the couplings between an infinite number of degenerate multiplicities (thin lines of figure 3).

2.3. Perturbed multiplicities

The diagonalisation of the coupling V inside each multiplicity $\mathscr{E}_{n,n'}$ is straightforward.

One first introduces two linear combinations of $|a, n+1, n'\rangle$ and $|c, n, n'-1\rangle$ which are respectively coupled and uncoupled to $|b, n, n'\rangle$. Putting

$$\Omega_1 = (\omega_1^2 + \omega_1'^2)^{1/2} \tag{2.4}$$

$$\cos\alpha = \omega_1/\Omega_1 \qquad \sin\alpha = \omega_1'/\Omega_1 \tag{2.5}$$

$$|u, n, n'\rangle = \cos\alpha\,|a, n+1, n'\rangle + \sin\alpha\,|c, n, n'-1\rangle \tag{2.6a}$$

$$|v, n, n'\rangle = -\sin\alpha\,|a, n+1, n'\rangle + \cos\alpha\,|c, n, n'-1\rangle \tag{2.6b}$$

† As in I, we will restrict ourselves to quasiclassical states of the two laser fields: the distributions $p_0(n)$ and $p_0'(n')$ of the numbers n and n' of ω_L and ω_L' photons have widths Δn and $\Delta n'$ very large in absolute values, but very small compared to the mean values $\bar{n}$ and $\bar{n}'$. All the energy diagrams represented below are therefore limited to values of n and n' close to $\bar{n}$ and $\bar{n}'$ (within Δn and $\Delta n'$) and they have a periodic structure in this range.

‡ We have also neglected non-resonant couplings between $\mathscr{E}_{n,n'}$ and $\mathscr{E}_{n\pm 2,n'}$ or $\mathscr{E}_{n,n'\pm 2}$ (rotating-wave approximation, which has a condition of validity $\omega_1, \omega_1' \ll \omega_0, \omega_0'$ much broader than (2.3)).

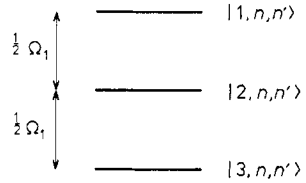

Figure 5. Perturbed states of the multiplicity $\mathscr{E}_{n,n'}$, with a splitting determined by $\Omega_1 = (\omega_1^2 + \omega_1'^2)^{1/2}$.

one easily checks that $|v, n, n'\rangle$ is not coupled to $|b, n, n'\rangle$ while $|u, n, n'\rangle$ is coupled to this state with an amplitude $\frac{1}{2}\Omega_1$. It follows that $\mathscr{E}_{n,n'}$ splits in three perturbed states $|i, n, n'\rangle$ $(i = 1, 2, 3)$

$$
\begin{aligned}
|1, n, n'\rangle &= \frac{1}{\sqrt{2}}(|b, n, n'\rangle + |u, n, n'\rangle) \\
|2, n, n'\rangle &= |v, n, n'\rangle \\
|3, n, n'\rangle &= \frac{1}{\sqrt{2}}(-|b, n, n'\rangle + |u, n, n'\rangle)
\end{aligned}
\tag{2.7}
$$

with energies (measured with respect to the unperturbed energy of $\mathscr{E}_{n,n'}$) respectively equal to:

$$
\begin{aligned}
E_1 &= +\tfrac{1}{2}\Omega_1 \\
E_2 &= 0 \\
E_3 &= -\tfrac{1}{2}\Omega_1
\end{aligned}
\tag{2.8}
$$

(see figure 5).

The equidistance between the three perturbed levels is a consequence of the simplifying assumption (2.1). In the general case, the three energy levels obtained by the diagonalisation of a 3×3 matrix are not equidistant. Let us note finally that, as long as $\omega_L + \omega_L' = \omega_0 + \omega_0'$, the two states $|a, n+1, n'\rangle$ and $|c, n, n'-1\rangle$ remain degenerate, so that the introduction of $|u, n, n'\rangle$ and $|v, n, n'\rangle$ leads to a two-level problem ($|v, n, n'\rangle$ is an eigenstate of the perturbed Hamiltonian).

2.4. Spontaneous transition rates

As a consequence of the assumptions made on the spontaneous decay of the three-level system a–b–c, the only non-zero matrix elements of the atomic dipole moment $\mathscr{D}$ are:

$$
\boldsymbol{d} = \langle a|\mathscr{D}|b\rangle \qquad \boldsymbol{d}' = \langle b|\mathscr{D}|c\rangle. \tag{2.9}
$$

From the expansion (2.7) of the dressed atom states, and from (2.9), one easily derives that $\mathscr{D}$ couples $\mathscr{E}_{n,n'}$ only to adjacent multiplicities ($\mathscr{E}_{n\pm1,n'}$ or $\mathscr{E}_{n,n'\pm1}$). When the number n (n') changes by one unit, this coupling describes the spontaneous emission of one photon with a frequency close to $\omega_L(\omega_L')$ (wavy arrows of figure 3 pointing to the left (right)).

Putting:

$$\begin{aligned} \boldsymbol{d}_{ij} &= \langle i, n-1, n' | \mathscr{D} | j, n, n' \rangle \\ \boldsymbol{d}'_{ij} &= \langle i, n, n'-1 | \mathscr{D} | j, n, n' \rangle \end{aligned} \tag{2.10}$$

one gets:

$$\begin{aligned} \boldsymbol{d}_{ij} &= \langle i, n-1, n' | a, n, n' \rangle \, \boldsymbol{d} \, \langle b, n, n' | j, n, n' \rangle \\ \boldsymbol{d}'_{ij} &= \langle i, n, n'-1 | b, n, n'-1 \rangle \, \boldsymbol{d}' \langle c, n, n'-1 | j, n, n' \rangle \end{aligned} \tag{2.11}$$

which, according to (2.7), gives:

$$\frac{\boldsymbol{d}_{ij}}{\boldsymbol{d}} = \begin{array}{c|ccc} i \backslash j & 1 & 2 & 3 \\ \hline 1 & \frac{1}{2}\cos\alpha & 0 & -\frac{1}{2}\cos\alpha \\ 2 & -\dfrac{\sin\alpha}{\sqrt{2}} & 0 & \dfrac{\sin\alpha}{\sqrt{2}} \\ 3 & \frac{1}{2}\cos\alpha & 0 & -\frac{1}{2}\cos\alpha \end{array}$$

$$\frac{\boldsymbol{d}'_{ij}}{\boldsymbol{d}'} = \begin{array}{c|ccc} i \backslash j & 1 & 2 & 3 \\ \hline 1 & \frac{1}{2}\sin\alpha & \dfrac{\cos\alpha}{\sqrt{2}} & \frac{1}{2}\sin\alpha \\ 2 & 0 & 0 & 0 \\ 3 & -\frac{1}{2}\sin\alpha & -\dfrac{\cos\alpha}{\sqrt{2}} & -\frac{1}{2}\sin\alpha \end{array} \tag{2.12}$$

Since $|2, n, n'\rangle$ does not contain $|b, n, n'\rangle$ (see (2.6) and (2.7))†:

$$\boldsymbol{d}_{i2} = \boldsymbol{d}'_{2j} = 0. \tag{2.13}$$

The transition rates

$$\gamma_{ij} = |\boldsymbol{d}_{ij}|^2 \qquad \gamma'_{ij} = |\boldsymbol{d}'_{ij}|^2 \tag{2.14}$$

between $|j, n, n'\rangle$ and $|i, n-1, n'\rangle$, $|j, n, n'\rangle$ and $|i, n, n'-1\rangle$ are immediately calculated from (2.12). Using

$$\gamma = |\boldsymbol{d}|^2 \qquad \gamma' = |\boldsymbol{d}'|^2 \tag{2.15}$$

† The orthogonality between $|2, n, n'\rangle$ and $|b, n, n'\rangle$, and consequently the property (2.13), remain valid as long as $\omega_L + \omega'_L = \omega_0 + \omega'_0$.

one gets:

$$\frac{\gamma_{ij}}{\gamma} = \begin{array}{c|ccc} i \backslash j & 1 & 2 & 3 \\ \hline 1 & \frac{1}{4}\cos^2\alpha & 0 & \frac{1}{4}\cos^2\alpha \\ 2 & \frac{1}{2}\sin^2\alpha & 0 & \frac{1}{2}\sin^2\alpha \\ 3 & \frac{1}{4}\cos^2\alpha & 0 & \frac{1}{4}\cos^2\alpha \end{array}$$

$$\frac{\gamma'_{ij}}{\gamma'} = \begin{array}{c|ccc} i \backslash j & 1 & 2 & 3 \\ \hline 1 & \frac{1}{4}\sin^2\alpha & \frac{1}{2}\cos^2\alpha & \frac{1}{4}\sin^2\alpha \\ 2 & 0 & 0 & 0 \\ 3 & \frac{1}{4}\sin^2\alpha & \frac{1}{2}\cos^2\alpha & \frac{1}{4}\sin^2\alpha \end{array} \tag{2.16}$$

All possible spontaneous decays from the three perturbed states of $\mathscr{E}_{n,n'}$ to lower multiplicities are represented in figure 6.

2.5. *Evolution of the populations of the dressed-atom states*

We will suppose in the following that:

$$\Omega_1 \gg \gamma, \gamma'. \tag{2.17}$$

This condition is the basis of the secular approximation which, as explained in I, allows a simple resolution of the master equation describing spontaneous emission from the dressed atom.

In particular, this approximation leads to a closed system of equations for the populations $\sigma_{ii}^0(n, n', t)$ of the levels $|i, n, n'\rangle$ which have the simple physical meaning of rate equations: $\sigma_{ii}^0(n, n', t)$ decreases because of spontaneous transitions to lower states (with a total transition rate $\Sigma_l \gamma_{li} + \Sigma_l \gamma'_{li}$) and increases because of spontaneous transitions from the upper states $|j, n+1, n'\rangle$ and $|j, n, n'+1\rangle$ (with transition rates respectively equal to γ_{ij} and γ'_{ij}):

$$\frac{\mathrm{d}}{\mathrm{d}t}\sigma_{ii}^0(n, n', t) = -\sum_l (\gamma_{li} + \gamma'_{li})\sigma_{ii}^0(n, n', t) + \sum_j \gamma_{ij}\sigma_{jj}^0(n+1, n', t) + \sum_j \gamma'_{ij}\sigma_{jj}^0(n, n'+1, t). \tag{2.18}$$

As in I, one can show that, to a very good approximation, the populations can be factorised as:

$$\sigma_{ii}^0(n, n', t) = \pi_i(t)\, p_0(n)\, p'_0(n') \tag{2.19a}$$

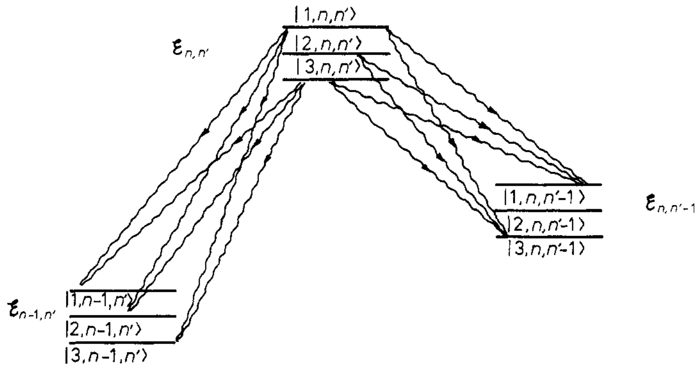

Figure 6. Possible spontaneous decays (wavy arrows) from the three perturbed states of $\mathscr{E}_{n,n'}$ to the lower multiplicities.

where $p_0(n)$ and $p'_0(n')$ are the distributions of the numbers n and n' of ω_L and ω'_L photons and where the $\pi_i(t)$ satisfy the rate equations

$$\dot{\pi}_i = -\Gamma_i \pi_i + \sum_j \Gamma_{ij} \pi_j \tag{2.19b}$$

with:

$$\begin{aligned} \Gamma_{ij} &= \gamma_{ij} + \gamma'_{ij} \\ \Gamma_i &= \sum_l \Gamma_{li}. \end{aligned} \tag{2.20}$$

Equations (2.19) have a steady-state solution which is found to be

$$\begin{aligned} \pi_1 = \pi_3 &= \frac{\gamma' \cos^2 \alpha}{\gamma \sin^2 \alpha + 2\gamma' \cos^2 \alpha} \\ \pi_2 &= \frac{\gamma \sin^2 \alpha}{\gamma \sin^2 \alpha + 2\gamma' \cos^2 \alpha}. \end{aligned} \tag{2.21}$$

One can check that such a solution satisfies the detailed-balance condition†

$$\Gamma_{ij}\pi_j = \Gamma_{ji}\pi_i. \tag{2.22}$$

2.6. Damping of the frequency components of the dipole moment

The various frequencies appearing in the motion of the dipole moment are the various Bohr frequencies corresponding to the allowed transitions represented on figure 6.

The periodic structure of the energy diagram is responsible for some important couplings between different optical coherences (off-diagonal elements of the density matrix σ) evolving at the same frequency (cascade effects).

Let us consider for example the case of the off-diagonal element $\langle 3, n, n' - 1|\sigma|1, n, n'\rangle$ evolving at $\omega'_L + \Omega_1$ (see figure 7). This coherence is first damped with a rate $\frac{1}{2}(\Gamma_1 + \Gamma_3)$ (half sum of the transition rates from the two connected states: wavy arrows of figure 7). But it is also coupled to $\langle 3, n+1, n'-1|\sigma|1, n+1, n'\rangle$ (with a coefficient $\boldsymbol{d}_{33} \cdot \boldsymbol{d}_{11}$) and to $\langle 3, n, n'|\sigma|1, n, n'+1\rangle$ (with a coefficient $\boldsymbol{d}'_{33} \cdot \boldsymbol{d}'_{11}$) (thin arrows of

† The detailed-balance condition is no longer satisfied when the resonance conditions $\omega_L = \omega_0$, $\omega'_L = \omega'_0$ are released.

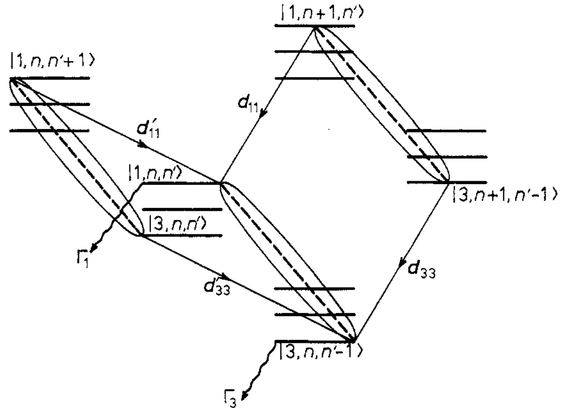

Figure 7. Various terms describing the evolution of the off-diagonal element $\langle 3, n, n' - 1|\sigma|1, n, n'\rangle$. In addition to the damping due to the transition rates Γ_3 and Γ_1 from $|3, n, n' - 1\rangle$ and $|1, n, n'\rangle$ (wavy arrows), this off-diagonal element is coupled (thin arrows) to $\langle 3, n + 1, n' - 1|\sigma|1, n + 1, n'\rangle$ (with a coefficient $\boldsymbol{d}_{11} \cdot \boldsymbol{d}_{33}$) and to $\langle 3, n, n'|\sigma|1, n, n' + 1\rangle$ (with a coefficient $\boldsymbol{d}'_{11} \cdot \boldsymbol{d}'_{33}$).

figure 7). A calculation similar to that of I shows that the $(\omega'_L + \Omega_1)$ frequency component of the dipole moment is damped with a rate L_{13} given by:

$$L_{13} = \tfrac{1}{2}(\Gamma_1 + \Gamma_3) - \boldsymbol{d}_{11} \cdot \boldsymbol{d}_{33} - \boldsymbol{d}'_{11} \cdot \boldsymbol{d}'_{33}. \tag{2.23}$$

More generally, one can show that the components of the dipole moment evolving at $\omega'_L + E_i - E_j$ or $\omega_L + E_i - E_j$ with $i \neq j$ are damped with a rate:

$$L_{ij} = L_{ji} = \tfrac{1}{2}(\Gamma_i + \Gamma_j) - \boldsymbol{d}_{ii}\boldsymbol{d}_{jj} - \boldsymbol{d}'_{ii}\boldsymbol{d}'_{jj}. \tag{2.24}$$

It could appear surprising to obtain such a simple formula for L_{12} or L_{23}: since $E_1 - E_2 = E_2 - E_3$, the off-diagonal elements $\langle 2, n, n' - 1|\sigma|1, n, n'\rangle$ and $\langle 3, n + 1, n' - 1|\sigma|2, n + 1, n'\rangle$ evolve at the same frequency $\omega'_L + \frac{1}{2}\Omega_1$ and it seems necessary to take into account their coupling. However such a coupling vanishes as a consequence of (2.13).

Finally, the ω'_L component of the dipole moment is proportional to

$$\sum_{ni} \boldsymbol{d}'_{ii} \langle i, n, n' - 1|\sigma|i, n, n'\rangle.$$

Taking into account all the couplings by spontaneous emission and using (2.13), one shows after a calculation similar to that of I (see also Reynaud 1977) that this central component (as well as the ω_L component) is damped with a rate L_c:

$$L_c = \Gamma_1 = \Gamma_3. \tag{2.25}$$

3. Optimisation of the stepwise excitation of the upper level c

The purpose of a stepwise excitation is generally to populate appreciably the upper level c of figure 2.

If one can use pulsed lasers, the best thing is of course to make first a π pulse of the ω_L laser which transfers all atoms from the ground state a to b; then a π pulse of the

ω'_L laser brings all these atoms to c (such a sequence should of course take a time short compared to the radiative lifetimes).

We will consider here the situation where both lasers ω_L and ω'_L have been simultaneously applied for a long time (steady-state regime) and we will determine under what conditions the population π_c of c is a maximum.

Such a population, in the presence of the two lasers, is given by:

$$\pi_c = \mathrm{Tr}|c\rangle\langle c|\sigma = \sum_{nn'} \langle c, n, n'|\sigma|c, n, n'\rangle \tag{3.1}$$

where σ is the density matrix of the dressed atom.

The expansion of $|c, n, n' - 1\rangle$ in terms of the perturbed states $|i, n, n'\rangle$ ($i = 1, 2, 3$) can be easily derived from (2.6) and (2.7):

$$|c, n, n' - 1\rangle = \frac{\sin\alpha}{\sqrt{2}}(|1, n, n'\rangle + |3, n, n'\rangle) + \cos\alpha\,|2, n, n'\rangle. \tag{3.2}$$

Inserting (3.2) into (3.1), using the diagonal character of σ in the steady-state regime† and expression (2.19*a*), one gets:

$$\pi_c = \tfrac{1}{2}\sin^2\alpha\,(\pi_1 + \pi_3) + \cos^2\alpha\,\pi_2 \tag{3.3}$$

which, when one uses the steady-state values (2.21) of the π_i, gives:

$$\pi_c = \frac{(\gamma + \gamma')\sin^2\alpha\cos^2\alpha}{\gamma\sin^2\alpha + 2\gamma'\cos^2\alpha}. \tag{3.4}$$

An atom of the atomic beam which, after having reached a steady state, leaves the irradiation zone sharply delimited by convenient slits (the intensities of the two lasers are supposed to be constant in this zone), has a probability π_c that it has been excited into the upper state c. Expression (3.4) is therefore a quantitative measure of the efficiency of the stepwise excitation.

As it appears from (3.4), this efficiency depends only on the two ratios γ/γ' and ω_1/ω'_1 (through the parameter α defined in (2.5)). The maximum of π_c, for a given value of γ/γ', is

$$(\pi_c)_{\max} = \frac{\gamma + \gamma'}{(\sqrt{\gamma} + \sqrt{2\gamma'})^2} \tag{3.5}$$

and corresponds to

$$\frac{\cos^2\alpha}{\sin^2\alpha} = \frac{\omega_1^2}{\omega_1'^2} = \left(\frac{\gamma}{2\gamma'}\right)^{1/2}. \tag{3.6}$$

One easily shows from (3.5) that $(\pi_c)_{\max}$ is always larger than the value $\frac{1}{3}$ which would correspond to an equalisation of the three populations.

When one of the two laser intensities becomes much larger than the other one, i.e. when $\omega_1 \gg \omega'_1$ ($\sin^2\alpha \ll 1$) or $\omega'_1 \gg \omega_1$ ($\cos^2\alpha \ll 1$), the steady-state value of π_c given in (3.4) tends to zero. Such a result only depends on the ratio ω_1/ω'_1 and remains valid even if the smallest Rabi frequency is larger than γ and γ'. We therefore conclude that, even if the two lasers are sufficiently intense to saturate each transition individually,

† The approximate resolution of the master equation for σ, by the method described in I, shows that the off-diagonal element $\langle i, n, n'|\sigma|j, n, n'\rangle$ with $i \neq j$ is damped to zero with the rate L_{ij} given in (2.24).

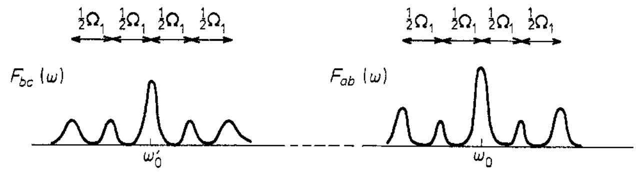

Figure 8. Schematic representation of the fluorescence spectra $F_{ab}(\omega)$ and $F_{bc}(\omega)$. At resonance ($\omega_L = \omega_0$, $\omega_L' = \omega_0'$) each spectrum exhibits five equidistant Lorentzian components with the same splitting $\frac{1}{2}\Omega_1$. Analytical expressions are given in the text for the width and the weight of each of these components.

the stepwise excitation may become completely inefficient if the two laser intensities are too unbalanced ($\omega_1^2/\omega_1'^2$ too far from the optimum value (3.6)). We will return to the physical interpretation of this unexpected result in §5.

4. Fluorescence and absorption spectra

4.1. Fluorescence spectra $F_{ab}(\omega)$ and $F_{bc}(\omega)$

A straightforward extension of the results of I shows that the fluorescence spectrum $F_{ab}(\omega)$ observed on the transition b–a exhibits several components at frequencies $\omega_L + E_i - E_j$, i.e. five components at frequencies ω_L, $\omega_L \pm \frac{1}{2}\Omega_1$, $\omega_L \pm \Omega_1$. A similar result holds for $F_{bc}(\omega)$ which exhibits five components at ω_L', $\omega_L' \pm \frac{1}{2}\Omega_1$, $\omega_L' \pm \Omega_1$ (figure 8). We conclude that, when two transitions sharing a common level are simultaneously saturated by two resonant laser beams, the fluorescence spectra observed from these two transitions have the same quintuplet structure† with a splitting Ω_1, depending on the Rabi nutation frequencies associated with both lasers through $\Omega_1 = (\omega_1^2 + \omega_1'^2)^{1/2}$.

The widths of the lateral components $\omega_L + E_i - E_j$ and $\omega_L' + E_i - E_j$ (with $i \neq j$) are the same and are given by (2.24). Using (2.12), (2.16) and (2.20), one gets:

$$L_{13} = L_{31} = \tfrac{1}{4}\gamma(2 + \cos^2\alpha) + \tfrac{3}{4}\gamma' \sin^2\alpha \tag{4.1}$$

$$L_{12} = L_{21} = L_{23} = L_{32} = \tfrac{1}{4}\gamma + \tfrac{1}{4}\gamma'(1 + \cos^2\alpha). \tag{4.2}$$

Similarly, the two central components at ω_L and ω_L' have the same width L_c given by (2.25) and equal to:

$$L_c = \tfrac{1}{2}\gamma + \tfrac{1}{2}\gamma' \sin^2\alpha. \tag{4.3}$$

It follows that the width of each component of the fluorescence spectra depends on the natural widths γ and γ' of both excited states b and c in a proportion related to ω_1 and ω_1' through the angle $\alpha = \tan^{-1}\omega_1'/\omega_1$. Such a result was expected for $F_{bc}(\omega)$ since this spectrum involves spontaneous transitions between c and b. The fact that the fluorescence emitted on ba depends on the natural width γ' of c seems, at first sight,

† When the resonance conditions (2.1) ($\omega_L = \omega_0$, $\omega_L' = \omega_0'$) are released, the three levels $|i, n\rangle$ (i = 1, 2, 3) are no longer equidistant and the fluorescence spectra exhibit seven unequidistant components. Let us note however that as long as $\omega_L + \omega_L' = \omega_0 + \omega_0'$ (two-photon resonance condition) equations (2.13) remain valid (see footnote, p 2317) so that on each spectrum, two non-symmetric components disappear, and this leads to two asymmetric quintuplets (Reynaud 1977).

more surprising, but it is a consequence of the simultaneous saturation of *ab* and *bc* which mixes all natural widths.

It is also interesting to note that, in our particular case ($\omega_L = \omega_0$, $\omega'_L = \omega'_0$, $\Omega_1 \gg \gamma, \gamma'$), the central components at ω_L and ω'_L have a Lorentzian shape; in particular, we do not find any elastic contribution (which would correspond to an undamped component of the dipole moment). The absence of elastic components is a consequence of the secular approximation ($\Omega_1 \gg \gamma, \gamma'$). These components actually exist but are of higher order in γ/Ω_1 or γ'/Ω_1.

The weights of the various components of $F_{ab}(\omega)$ and $F_{bc}(\omega)$ can be derived as in I. The total number of photons emitted from a given component is equal to the total number of atoms undergoing the corresponding transitions:

$$\begin{aligned} \mathscr{I}(\omega_L + E_i - E_j) &= \gamma_{ji}\pi_i \qquad \text{for } i \neq j \\ \mathscr{I}(\omega'_L + E_i - E_j) &= \gamma'_{ji}\pi_i \end{aligned} \tag{4.4}$$

$$\begin{aligned} \mathscr{I}(\omega_L) &= \sum_i \gamma_{ii}\pi_i \\ \mathscr{I}(\omega'_L) &= \sum_i \gamma'_{ii}\pi_i . \end{aligned} \tag{4.5}$$

It appears clearly from (4.4) and (4.5) that, contrary to what happens for the positions and the widths, the results concerning the weights are not the same for both spectra (since $\gamma_{ji} \neq \gamma'_{ji}$ in general). Using the steady-state values (2.21) of the populations π_i and the expression (2.16) of the γ_{ij} and γ'_{ij}, one gets:

$$\begin{aligned} \mathscr{I}(\omega_L) &= \tfrac{1}{2}\cos^2\alpha\,\mathscr{I} \\ \mathscr{I}(\omega_L \pm \Omega_1) &= \tfrac{1}{4}\cos^2\alpha\,\mathscr{I} \\ \mathscr{I}(\omega_L \pm \tfrac{1}{2}\Omega_1) &= \tfrac{1}{2}\sin^2\alpha\,\mathscr{I} \\ \mathscr{I}(\omega'_L) &= \frac{\gamma'}{2\gamma}\sin^2\alpha\,\mathscr{I} \\ \mathscr{I}(\omega'_L \pm \Omega_1) &= \frac{\gamma'}{4\gamma}\sin^2\alpha\,\mathscr{I} \\ \mathscr{I}(\omega'_L \pm \tfrac{1}{2}\Omega_1) &= \tfrac{1}{2}\sin^2\alpha\,\mathscr{I} \end{aligned} \tag{4.6}$$

where

$$\mathscr{I} = \gamma T \frac{\gamma'\cos^2\alpha}{\gamma\sin^2\alpha + 2\gamma'\cos^2\alpha} \tag{4.7}$$

is the total number of photons emitted on the *ab* transition (T is the transit time of atoms through the laser beam; the transient regime has been neglected).

We first note that $F_{ab}(\omega)$ and $F_{bc}(\omega)$ are symmetric with respect to ω_L and ω'_L. It must, however, be mentioned that such a symmetry completely disappears when the resonance conditions (2.1) ($\omega_L = \omega_0$, $\omega'_L = \omega'_0$) are released.

The expression (4.7) of the total intensity $\mathscr{I}$ emitted from the *ab* transition, as well as the corresponding expression:

$$\begin{aligned} \mathscr{I}' &= \mathscr{I}(\omega'_L) + 2\mathscr{I}(\omega'_L + \tfrac{1}{2}\Omega_1) + 2\mathscr{I}(\omega'_L + \Omega_1) \\ &= \gamma' T \frac{(\gamma + \gamma')\cos^2\alpha\sin^2\alpha}{\gamma\sin^2\alpha + 2\gamma'\cos^2\alpha} \end{aligned} \tag{4.8}$$

of the total intensity emitted from the bc transition, have a simple physical interpretation: let us calculate the reduced population π_b of the unperturbed level b (as we have done it for π_c in §3). One gets

$$\pi_b = \frac{\gamma' \cos^2 \alpha}{\gamma \sin^2 \alpha + 2\gamma' \cos^2 \alpha} \tag{4.9}$$

and one easily checks that

$$\begin{aligned} \mathscr{I} &= \gamma T \pi_b \\ \mathscr{I}' &= \gamma' T \pi_c \end{aligned} \tag{4.10}$$

which means that $\mathscr{I}$ and $\mathscr{I}'$ are respectively proportional to the total number of atoms in b and c, and to the transition rates γ and γ'.

It seems also interesting to study the behaviour of the weights (4.6) when one of the laser fields becomes much weaker than the other one ($\omega_1 \ll \omega'_1$ or $\omega'_1 \ll \omega_1$). This problem will be considered in detail in §5.

4.2. Absorption spectrum of a new transition starting from a, b or c

We consider now a third laser beam having a sufficiently weak intensity and probing a new transition starting from a, b or c. One measures the absorption of this laser light as a function of its frequency.

Let us consider for example the transition ad of figure 2. Since the two intense ω_L and ω'_L lasers are not resonant for the transition ad, the state $|d, n, n'\rangle$ can be considered as an eigenstate of the dressed atom (atom dressed by the ω_L and ω'_L photons). The frequency components appearing in the absorption spectrum $A_{ad}(\omega)$ of the ad transition are given by the Bohr frequencies corresponding to the allowed electric dipole transitions between $|d, n, n'\rangle$ and any dressed-atom state belonging to the multiplicities $\mathscr{E}$ introduced in (2.3). Since $|d, n, n'\rangle$ is only connected to $|a, n, n'\rangle$, which only appears in the expansion of the perturbed states $|i, n-1, n'\rangle$ of $\mathscr{E}_{n-1,n'}$, one immediately deduces that $A_{ad}(\omega)$ exhibits three components at frequencies:

$$\omega''_0 - E_1 = \omega''_0 - \tfrac{1}{2}\Omega_1 \qquad \omega''_0 - E_2 = \omega''_0 \qquad \omega''_0 - E_3 = \omega''_0 + \tfrac{1}{2}\Omega_1 \tag{4.11}$$

(where ω''_0 is the frequency of the ad transition) and corresponding to the three allowed transitions $|i, n-1, n'\rangle \rightarrow |d, n, n'\rangle$ ($i = 1, 2, 3$). It can be easily shown (Reynaud 1977) that the width of the $\omega''_0 - E_i$ component is simply given by $\frac{1}{2}(\Gamma_i + \Gamma_d)$ where Γ_i and Γ_d are the total transition rates from the two connected levels $|i, n-1, n'\rangle$ and $|d, n, n'\rangle$. Finally, the weight $\mathscr{I}_A(\omega''_0 - E_i)$ of each component $\omega''_0 - E_i$ is proportional to the population of the absorbing level $|i, n-1, n'\rangle$ and to the absorption rate $|\langle i, n-1, n'| \mathscr{D} |d, n, n'\rangle|^2$ which leads to:

$$\mathscr{I}_A(\omega''_0 - E_i) \sim \pi_i |\langle i, n-1, n'|a, n, n'\rangle|^2 |\langle a|\mathscr{D}|d\rangle|^2. \tag{4.12}$$

We therefore conclude that the saturation of bc transforms the well known Autler–Townes doublet observed from ad and associated with the saturation of ab into a triplet. Although the ω'_L laser does not act directly on level a, it modifies drastically the absorption spectrum observed on ad: not only the splitting changes from ω_1 to Ω_1, but a third new component appears. Similar results are obtained on the absorption spectra starting from level c. It must however be mentioned that the absorption spectra observed from b, for example on a transition be, remain as a doublet: this is due to the fact that the weight of the transition $|2, n, n'\rangle \rightarrow |e, n, n'\rangle$, which is proportional to $|\langle 2, n, n'|b, n, n'\rangle|^2$, vanishes as a consequence of (2.6) and (2.7) (such a doublet structure remains as long as $\omega_L + \omega'_L = \omega_0 + \omega'_0$, see footnote p 2317). Paradoxically, it is for level b, which is directly coupled to both lasers, that the modifications of the absorption spectra are the smallest.

5. Discussion of two limiting cases ($\omega_1 \ll \omega'_1$ and $\omega'_1 \ll \omega_1$)

5.1. *Limiting case* $\omega_1 \ll \omega'_1$

We consider first the behaviour of the fluorescence spectra $F_{ab}(\omega)$ and $F_{bc}(\omega)$ at the limit where the ω_L laser driving the ab transition has a much weaker intensity than the other one:

$$\omega_1 \ll \omega'_1. \tag{5.1a}$$

The ω'_L laser, however, is supposed to be sufficiently intense so that the secular approximation (2.17) remains valid:

$$\Omega_1 \simeq \omega'_1 \gg \gamma, \gamma'. \tag{5.1b}$$

In such a case, expressions (2.5) may be written as:

$$\cos\alpha \simeq \omega_1/\omega'_1 \qquad \sin\alpha \simeq 1 \tag{5.2}$$

so that the weights of the components of the spectra are, according to (4.6) and (4.7):

$$\begin{aligned}
\mathscr{I}(\omega_L \pm \omega'_1) &= \tfrac{1}{2}\mathscr{I}(\omega_L) = \tfrac{1}{4}\gamma' T(\omega_1/\omega'_1)^4 \\
\mathscr{I}(\omega'_L \pm \omega'_1) &= \tfrac{1}{2}\mathscr{I}(\omega'_L) = \frac{1}{4}\frac{\gamma'^2}{\gamma} T(\omega_1/\omega'_1)^2 \\
\mathscr{I}(\omega_L \pm \tfrac{1}{2}\omega'_1) &= \mathscr{I}(\omega'_L \pm \tfrac{1}{2}\omega'_1) = \tfrac{1}{2}\gamma' T(\omega_1/\omega'_1)^2.
\end{aligned} \tag{5.3}$$

The corresponding shapes of $F_{ab}(\omega)$ and $F_{bc}(\omega)$ are sketched on figure 9. These results do not fit at all the conclusions which would be suggested by a naive approach to the problem (a single elastic Rayleigh component for $F_{ab}(\omega)$, and a triplet for $F_{bc}(\omega)$, see introduction): the spectrum $F_{ab}(\omega)$ contains essentially two components in ω_1^2 at

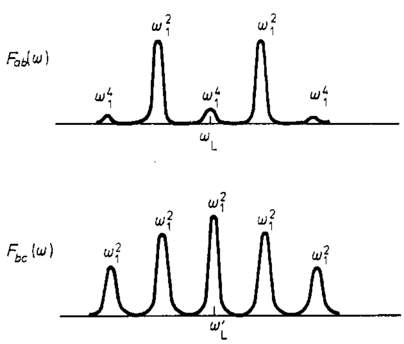

Figure 9. Fluorescence spectra $F_{ab}(\omega)$ and $F_{bc}(\omega)$ at the limit $\omega_1 \ll \omega'_1$. The splitting is now $\frac{1}{2}\omega'_1$. $F_{ab}(\omega)$ exhibits two components in ω_1^2 at $\omega_L \pm \frac{1}{2}\omega'_1$ and three smaller ones in ω_1^4 at ω_L, $\omega_L \pm \omega'_1$. The five components of $F_{bc}(\omega)$ at ω'_L, $\omega'_L \pm \frac{1}{2}\omega'_1$, $\omega'_L \pm \omega'_1$ are all in ω_1^2.

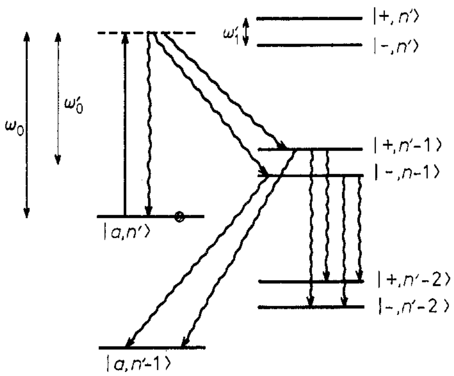

Figure 10. Energy diagram of the atom dressed by the ω_L' photons. From the populated level $|a, n'\rangle$, the dressed atom absorbs one ω_L photon (full arrow) and falls back to $|a, n'\rangle$, $|\pm, n' - 1\rangle$ (wavy arrows). The corresponding Rayleigh and Raman processes take place through two possible intermediate states $|\pm, n'\rangle$. Subsequent decays from the unstable states $|\pm, n' - 1\rangle$ (reached after the Raman process) are indicated by wavy arrows.

$\omega_L \pm \frac{1}{2}\omega_1'$ (three other components in ω_1^4 appear around ω_L, $\omega_L \pm \omega_1'$)†. The spectrum $F_{bc}(\omega)$ contains five components all in ω_1^2.

Let us now show how a correct perturbative treatment of the problem allows a simple interpretation of all these results. Considering that the intense ω_L' laser splits the level b is too crude. The correct approach is first to treat to all orders the system 'atom + intense ω_L' laser interacting together' (atom dressed by the ω_L' photons), which has an infinite number of energy levels. One can then describe perturbatively the scattering of the weak ω_L laser by such a dressed atom.

In figure 10 some energy levels of this dressed atom are represented: the perturbed levels $|\pm, n'\rangle$ of the right column originate from the unperturbed states $|b, n'\rangle$, $|c, n' - 1\rangle, \ldots$ etc. They are given by

$$|\pm, n'\rangle = \frac{1}{\sqrt{2}}(|c, n' - 1\rangle \pm |b, n'\rangle) \tag{5.4}$$

and are separated by ω_1'. The left column contains the states $|a, n'\rangle$ which are not perturbed by the non-resonant coupling with the ω_L' laser.

In the absence of the ω_L laser, the $|a, n'\rangle$ states are the only ones to be stable and therefore populated. Let us now consider the scattering of a single ω_L photon (lowest-order processes, with amplitudes proportional to ω_1). After absorbing one ω_L photon from the $|a, n'\rangle$ state (upwards full arrow on figure 10), the dressed atom can fall back either to $|a, n'\rangle$ (elastic Rayleigh scattering), or to $|\pm, n' - 1\rangle$ (Raman scattering) with the emission of photons at frequencies equal to ω_L, $\omega_L' \mp \frac{1}{2}\omega_1'$ respectively (wavy arrows on figure 10). One must not forget, however, that these three scattering processes can take place through two intermediate states $|\pm, n'\rangle$ so that the corresponding scattering amplitudes result from the interference between two paths.

For example, the total Rayleigh scattering amplitude is, to the lowest order in ω_1, proportional to

$$\omega_1 \sum_{\epsilon = +,-} \frac{\langle an'|D|\epsilon n'\rangle \langle \epsilon n'|D|an'\rangle}{E_{an'} + \omega_L - E_{\epsilon n'} + \frac{1}{2}\mathrm{i}\,\Gamma_{\epsilon n'}} \tag{5.5}$$

† Note that the elastic component remains absent in $F_{ab}(\omega)$: the discussion in §4.1 concerning this component only refers to the secular approximation and to the resonance conditions, which remain valid in the present case (see 5.1b).

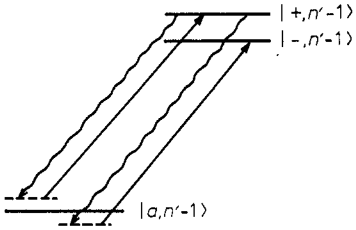

Figure 11. Inverse Rayleigh and Raman processes occurring from level $|+, n'-1\rangle$: emission of a fluorescence photon (wavy arrows) followed by the absorption of one ω_L photon (full arrows), the final state of the dressed atom being $|+, n'-1\rangle$ or $|-, n'-1\rangle$.

where D is the atomic dipole operator (since a, b, c are non-degenerate, we forget all polarisation effects), $E_{an'}$ and $E_{\epsilon n'}$ are the energies of $|a, n'\rangle$ and $|\epsilon, n'\rangle$, $\Gamma_{\epsilon n'}$ the natural width of $|\epsilon, n'\rangle$ which is easily found to be equal to $\frac{1}{2}(\gamma + \gamma')$. From the expression (5.4) for $|\pm, n'\rangle$ and from the resonance conditions (2.1) which imply $E_{an'} + \omega_L - E_{\pm, n'} = \mp\frac{1}{2}\omega_1'$ (see figure 10), one immediately deduces that (5.5) is proportional to

$$\omega_1\left[\left(-\frac{\omega_1'}{2} + i\frac{\gamma + \gamma'}{4}\right)^{-1} + \left(\frac{\omega_1'}{2} + i\frac{\gamma + \gamma'}{4}\right)^{-1}\right] \tag{5.6}$$

which vanishes when we neglect non-secular terms in $(\gamma + \gamma')/\omega_1'$ (secular approximation). We therefore interpret the absence of the elastic Rayleigh component as being due to an interference between two different paths which is completely destructive at resonance ($\omega_L = \omega_0$, $\omega_L' = \omega_0'$) and for high intensities of the ω_L' laser ($\omega_1' \gg \gamma + \gamma'$). Such an interference also exists for the Raman processes of figure 10 but it is constructive, as it can be shown by a calculation very similar to the previous one. This explains the presence in $F_{bc}(\omega)$ of the components $\omega_L' \pm \frac{1}{2}\omega_1'$ with a weight proportional to ω_1^2.

Once the dressed atom is in $|\pm, n'-1\rangle$, it can decay to lower states $|a, n'-1\rangle$ or $|\pm, n'-2\rangle$, spontaneously emitting photons with frequencies $\omega_L \pm \frac{1}{2}\omega_1'$ or ω_L', $\omega_L' \pm \omega_1'$. This explains the three other components in ω_1^2 of $F_{bc}(\omega)$ at ω_L' and $\omega_L' \pm \omega_1'$, as well as the two major components (also in ω_1^2) of $F_{ab}(\omega)$ at $\omega_L \pm \frac{1}{2}\omega_1'$.

It remains for us to explain the three small components of $F_{ab}(\omega)$ at ω_L, $\omega_L \pm \omega_1'$. Since they are in ω_1^4, they imply a second interaction with the ω_L laser (see figure 11). Once the dressed atom is in $|+, n'-1\rangle$ after the Raman process of figure 10 (eventually followed by one or several cascades along the right part of the energy diagram of figure 10), it can emit a spontaneous photon (wavy arrows of figure 11) and then reabsorb a photon of the ω_L laser which puts it back into $|+, n'-1\rangle$ or $|-, n'-1\rangle$ (full arrows of figure 11). These 'inverse' Rayleigh and Raman processes, which can also occur from $|-, n'-1\rangle$, are at the origin of the three components of $F_{ab}(\omega)$ at ω_L, $\omega_L \pm \omega_1'$.

We have therefore been able to understand the main features of the spectra represented in figure 9. By pushing a little further the previous perturbative considerations, it would be possible to derive all the expressions (5.3) for the weights of the various components.

The inefficiency of the stepwise excitation of level c at the limit $\omega_1' \gg \omega_1$, pointed out at the end of §3, can be easily understood from the previous discussion. We see from figure 10 that the levels $|\pm, n'\rangle$ cannot be appreciably populated from $|a, n'\rangle$ by the absorption of an ω_L photon, since this absorption is no longer resonant as a consequence

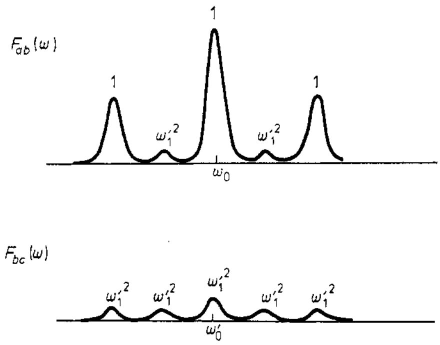

Figure 12. Fluorescence spectra $F_{ab}(\omega)$ and $F_{bc}(\omega)$ at the limit $\omega_1' \ll \omega_1$. The splitting is now $\frac{1}{2}\omega_1$. $F_{ab}(\omega)$ exhibits three large components (to zeroth order in ω_1') at ω_L, $\omega_L \pm \omega_1$ and two smaller ones in $\omega_1'^2$ at $\omega_L \pm \frac{1}{2}\omega_1$. The five components of $F_{bc}(\omega)$ at ω_L', $\omega_L' \pm \frac{1}{2}\omega_1$, $\omega_L' \pm \omega_1$ are all in $\omega_1'^2$.

of the splitting ω_1' appearing between the $|+, n'\rangle$ and $|-, n'\rangle$ levels. Increasing the intensity of one of the two lasers too much suppresses the resonant character of the second laser excitation (let us recall that, in the previous discussion, we have assumed $\omega_L = \omega_0$, $\omega_L' = \omega_0'$. It appears clearly on figure 10 that when $\omega_1' \gg \omega_1$, the efficiency of the stepwise excitation can be improved by tuning the ω_L laser to $\omega_L = \omega_0 \pm \frac{1}{2}\omega_1'$).

5.2. *Limiting case $\omega_1' \ll \omega_1$*

The equivalent of conditions (5.1*a*) (5.1*b*) and (5.2) are now:

$$\omega_1' \ll \omega_1 \tag{5.7a}$$

$$\Omega_1 \simeq \omega_1 \gg \gamma, \gamma' \tag{5.7b}$$

$$\cos\alpha \simeq 1 \qquad \sin\alpha \simeq \omega_1'/\omega_1. \tag{5.8}$$

In this limit, equations (4.6) and (4.7) become:

$$\begin{aligned} \mathscr{I}(\omega_L \pm \omega_1) &= \tfrac{1}{2}\mathscr{I}(\omega_L) = \tfrac{1}{8}\gamma T \\ \mathscr{I}(\omega_L' \pm \omega_1) &= \tfrac{1}{2}\mathscr{I}(\omega_L') = \tfrac{1}{8}\gamma' T\,(\omega_1'/\omega_1)^2 \\ \mathscr{I}(\omega_L \pm \tfrac{1}{2}\omega_1) &= \mathscr{I}(\omega_L' \pm \tfrac{1}{2}\omega_1) = \tfrac{1}{4}\gamma T\,(\omega_1'/\omega_1)^2. \end{aligned} \tag{5.9}$$

The corresponding spectrum $F_{ab}(\omega)$, represented in figure 12, consists mainly of three components at ω_L, $\omega_L \pm \omega_1$ (to zeroth order in ω_1'), as it should be in absence of the ω_L' laser. Two additional components, in $\omega_1'^2$, appear at $\omega_L \pm \frac{1}{2}\omega_1$. The spectrum $F_{bc}(\omega)$ is less obvious. Instead of the doublet suggested by the naive approach (see introduction), one gets five components, all in $\omega_1'^2$.

For interpreting the spectra of figure 12 perturbatively, we consider first the atom dressed by the intense ω_L laser. Some energy levels of this system are represented on figure 13. The left column now contains the levels:

$$|\pm, n\rangle = \frac{1}{\sqrt{2}}(|a, n+1\rangle \pm |b, n\rangle) \tag{5.10}$$

separated by ω_1, while the right column consists of the levels $|c, n\rangle$ not resonantly coupled to the ω_L laser.

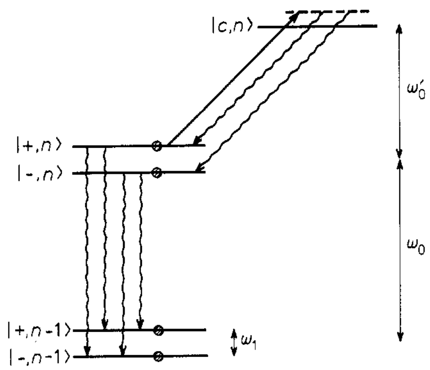

Figure 13. Energy diagram of the atom dressed by the ω_L photons. The cascades between the populated levels $|\pm, n\rangle$ are represented by vertical wavy arrows. From $|+, n\rangle$, the dressed atom can absorb one ω_L' photon (full arrow) and fall back to $|+, n\rangle$ or $|-, n\rangle$ (Rayleigh and Raman processes represented by wavy arrows).

To zeroth order in ω_1', only levels $|\pm, n\rangle$ are populated. They can decay only to the lower states of the left column of figure 13, and the corresponding cascade accounts for the well known triplet ω_L, $\omega_L \pm \omega_1$ of $F_{ab}(\omega)$ (vertical wavy arrows of figure 13).

Suppose now that the dressed atom in $|+, n\rangle$ absorbs an ω_L' photon (leaning full arrow of figure 13), and then falls back to $|+, n\rangle$ or $|-, n\rangle$ (leaning wavy arrows). These Rayleigh and Raman first-order processes, which can also occur from $|-, n\rangle$, explain the presence of the three components ω_L', $\omega_L' \pm \omega_1$ of $F_{bc}(\omega_1)$, with an intensity proportional to $\omega_1'^2$. Because of the natural widths of $|+, n\rangle$ and $|-, n\rangle$ these Rayleigh and Raman components have a finite width.

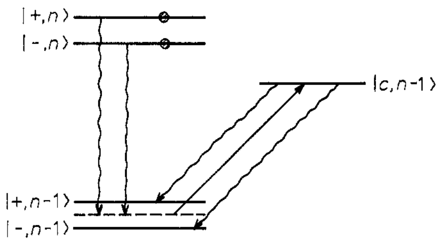

Figure 14. Inverse Raman processes occurring from $|+, n\rangle$ or $|-, n\rangle$: emission of one fluorescence photon (vertical wavy arrows) followed by the absorption of one ω_L' photon (full arrow), the final state of the dressed atom being $|c, n-1\rangle$. Subsequent decays from $|c, n-1\rangle$ are indicated by wavy arrows.

Finally, let us consider the inverse Raman processes represented on figure 14 and occurring from $|+, n\rangle$ or $|-, n\rangle$: emission of a spontaneous photon from $|+, n\rangle$ or $|-, n\rangle$ (vertical wavy arrows) and absorption of a photon of the ω_L' laser (leaning full arrow) which puts the dressed atom into $|c, n-1\rangle$. One immediately understands in this way the last two components of $F_{ab}(\omega)$ at $\omega_L \pm \frac{1}{2}\omega_1$ (which require the absorption of one ω_L' photon and which therefore have an intensity proportional to $\omega_1'^2$)†. From $|c, n-1\rangle$, the dressed atom decays to $|\pm, n-1\rangle$ by emitting

† There are two Raman amplitudes, corresponding to the two intermediate states $|\pm, n-1\rangle$, but a simple calculation shows that they interfere constructively.

spontaneous photons at $\omega_L' \pm \frac{1}{2}\omega_1$ (leaning wavy arrows) which explains the last two components of $F_{bc}(\omega)$.

A discussion similar to the one given at the end of §5.1, explains the decrease of the efficiency of the stepwise excitation of level c at the limit $\omega_1 \gg \omega_1'$: the splitting ω_1 appearing between $|+, n\rangle$ and $|-, n\rangle$ prevents a resonant ω_L' excitation of level $|c, n\rangle$ from the populated levels $|\pm, n\rangle$.

6. Comparison with previous works

It is interesting to compare the results derived above with the ones obtained by Whitley and Stroud (1976) from an integration of the equations of motion of the atomic density matrix driven by two c-number applied laser fields.

Let us first emphasise the complete equivalence between both methods at the high-intensity limit. As mentioned in I, the dressed-atom approach can be exactly transposed into the semiclassical one: in the present case of three atomic levels, there are nine evolution equations for the atomic density matrix elements. In a first step, one could neglect the damping terms and diagonalise the Hamiltonian part of the evolution equations which is exactly equivalent to finding the three dressed-atom states; if γ and γ' are sufficiently small, it would be then a good approximation to keep only the secular part of the damping terms, which is strictly equivalent to the secular approximation used in the dressed-atom approach.

Requiring only the diagonalisation of a 3×3 matrix (instead of a 9×9 one), the dressed-atom approach leads at the high-intensity limit to simpler calculations and to explicit expressions for the positions, widths and weights of the various components†. This allows a quantitative and physical understanding of the influence of the various parameters, more conveniently than from a computer integration of the nine evolution equations.

Of course, these results are only valid to zeroth order in γ/Ω_1, γ'/Ω_1. If one is interested in higher-order corrections or if the various components overlap (which means that γ/Ω_1 or γ'/Ω_1 are not small), the secular approximation cannot be done and it is necessary to solve the whole set of nine equations.

Finally it should be noted that the calculations presented here generalise to some extent those of I where a single laser beam was considered, saturating the transitions between the various sublevels of two atomic states. The method of I has been first applied to the study of the modification of the Raman effect in intense laser fields (Cohen-Tannoudji and Reynaud 1977b) by considering an atom having a single excited and two quasi-degenerate ground-state sublevels. Here two laser beams (instead of one) are saturating a three-level atom (the three levels being distinct and not quasi-degenerate) and it is the presence of two types of photons which requires a generalisation of the formalism of I (compare for example equations (2.18) for the evolution of the populations and (2.23) for the widths of the lateral components with the corresponding equations (3.4) and (4.19) of I).

† For length considerations, we have only studied the resonant case for which the diagonalisation of the 3×3 Hamiltonian is trivial (see equation (2.7)), but obviously, this diagonalisation can be done in the general case. For the same reasons, we have only considered the steady-state contribution to the fluorescence spectra although the transient one could be studied from the results of I (see equation (5.1) of I).

7. Conclusion

To summarise, we have shown that the dressed-atom method can be applied to the problem of the simultaneous saturation of two atomic transitions sharing a common level (the particular case of three equidistant levels, which requires a more elaborate treatment, is not considered in this paper).

We have derived analytical expressions for the positions, the widths and the integrated intensities of the various components of the fluorescence and absorption spectra, in the simple case where the laser excitations are both resonant. New structures, such as quintuplets, triplets (or even septuplets when the resonance conditions are released) have been predicted. The efficiency of the stepwise excitation of the upper level c has been analysed in the steady-state regime.

Finally, the limiting case where one of the two lasers has a much weaker intensity than the other one has been studied in detail. A simple interpretation of the corresponding results has been given by treating perturbatively the scattering of the weak laser by the atom dressed by the intense one.

Acknowledgments

In a recent symposium on resonance fluorescence (MIT, April 1976), where one of us (Cohen-Tannoudji) presented some of the results of this paper, Professor Stroud mentioned to us that he was studying similar problems. We thank him for communicating a preprint of his paper (Whitley and Stroud 1976) before publication.

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