Collisional Effects in Resonance Fluorescence

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1. Introduction

The collisional redistribution of near resonant scattered light has been extensively studied both experimentally [1] and theoretically [2 to 16]. The usual perturbative picture given for such a redistribution is sketched on Fig. 1. In the absence of collisions and at the lowest order in the laser intensity, the fluorescence spectrum is given by the elastic Rayleigh scattering process of Fig. 1a. Collisions are responsible for the appearance of a new fluorescence line around ω_0 which can be interpreted as due to collision induced transitions populating the excited atomic level e from the "virtual" level reached after the absorption of a laser photon (dotted line of Fig. 1b).

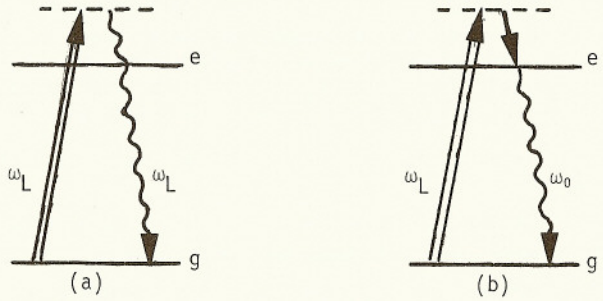

Fig. 1 Perturbative interpretation of Rayleigh scattering (a) and collisional redistribution (b)

Saturation effects associated with an increase of the laser intensity have been investigated by different methods. One of them is based on a non-perturbative solution of the "optical Bloch equations"

$$\frac{d}{dt}\,\sigma_a = -\,i[(H_a - DE_L \cos \omega_L t)\,,\,\sigma_a] - (\mathcal{J}_{rad} + \mathcal{J}_{coll})\,\sigma_a \qquad (1)$$

giving the rate of variation of the atomic density matrix σ_a as a sum of three independent rates: the free atomic evolution and interaction with the laser (first line); radiative damping due to spontaneous emission and

collisional relaxation (second line). The relaxation matrices $\mathcal{T}_{rad}$ and $\mathcal{T}_{coll}$ are taken to be the same as in the absence of laser irradiation [8-9].

Another possible approach, the so-called dressed atom approach, deals with the compound system "atom plus laser photons interacting together" (dressed atom). In the absence of collisions, resonance fluorescence photons can be considered as photons spontaneously emitted by the dressed atom. All features of resonance fluorescence can be quantitatively interpreted in terms of radiative transition rates between the dressed states [18-19]. Collisional effects can be included in such a theoretical frame by adding collision induced transition rates between the dressed states [6-12].

In this paper, we present a survey of the dressed atom approach to collisional redistribution. We introduce the relevant parameters describing the collisional relaxation of the dressed atom (T_1 and T_2 relaxation). We show how the redistribution spectrum and absorption profile may be related to these parameters. We also discuss the validity of the Markov approximation used in writing dressed atom relaxation equations as well as optical Bloch equations. In the so called impact regime, both equations are valid and we make explicit the relation between the collisional redistribution rate w between the dressed states and the dephasing rate γ of the bare atom dipole moment. We show that the dressed atom approach remains valid outside the impact regime which is not the case for optical Bloch equations.

We will ignore in this paper other effects such as quenching, polarization redistribution and velocity changing collisions.

2. The Dressed Atom Approach

The dressed atom energy diagram is sketched on Fig.2, the left part of which gives the uncoupled states labelled by two quantum numbers (e and g for the upper and lower atomic states and n for the number of laser photons).

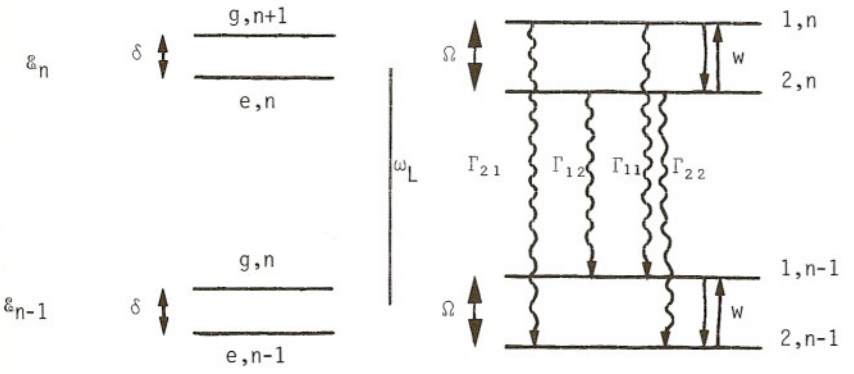

Fig. 2 Energy diagram of the dressed atom. Wavy and full arrows respectively describe radiative and collisional transitions between the dressed states

The two states $|e,n>$ and $|g,n+1>$ are nearly degenerate and form a two dimensional manifold &n (energy difference equal to the detuning δ between the laser and atomic frequencies ω_L and ω_0 and much smaller than the distance ω_L between two adjacent manifolds). The laser atom interaction has a non-zero matrix element between the two states of each manifold which is equal to $\omega_1/2$ (ω_1 , the Rabi frequency, is equal to the product of the dipole moment d by the laser field amplitude E_L). This coupling describes absorption and stimulated emission of laser photons by the atom. The dressed states $|1,n>$ and $|2,n>$ which diagonalize the total Hamiltonian are represented on the right part of Fig. 2. Their splitting is $\Omega = [\omega_1^2 + \delta^2]^{1/2}$. They can be written

$$|1,n> = \cos\Theta\,|e,n> + \sin\Theta\,|g,n+1> \qquad (2a)$$

$$|2,n> = -\sin\Theta\,|e,n> + \cos\Theta\,|g,n+1> \qquad (2b)$$

where $\cos\Theta = [(\Omega-\delta)/2\Omega]^{1/2}$ $\sin\Theta = [(\Omega+\delta)/2\Omega]^{1/2}$. (3)

The coupling of the dressed atom with the empty modes of the electromagnetic field is responsible for a radiative relaxation described by spontaneous transition rates between the dressed states (wavy arrows of Fig.2) and giving rise to three emission lines (fluorescence triplet) at $\omega_L+\Omega$ (transitions from $|1,n>$ to $|2,n-1>$), $\omega_L-\Omega$ ($|2,n> \rightarrow |1,n-1>$) and ω_L ($|i,n> \rightarrow |i,n-1>$ for i=1, 2). This simple description in terms of transition rates is based on a "secular approximation" [18-19] valid when the three lines are well resolved (splitting Ω large compared to the width of the lines).

In the same way, the effect of collisions can be described in this energy diagram as a relaxation mechanism, but the collision induced transitions (full arrows of Fig. 2) now occur inside each manifold (quenching neglected). Such a relaxation produces a population redistribution (T_1 type relaxation) and a coherence damping (T_2 type relaxation) respectively described by

$$\frac{d}{dt}\sigma_{1n,1n} = w\,(\sigma_{2n,2n} - \sigma_{1n,1n}) \qquad (4a)$$

$$\frac{d}{dt}\sigma_{1n,2n} = -(\kappa + i\,\xi)\,\sigma_{1n,2n} \qquad (4b)$$

(σ dressed atom density matrix). To summarize, there are three relevant relaxation parameters w, κ and ξ which have to be derived from the collision S matrix (see some examples of the calculation of w in [6-12]).

The dressed atom picture clearly shows that the emission spectrum has still a triplet structure in the presence of collisions. Only the positions, widths and weights of the three emission lines are modified. We want here to show how these modifications can be related to the relaxation parameters.

Solving the dressed atom relaxation equations for optical coherences gives, in the same way as for collisionless resonance fluorescence [18-19] , the positions and widths of the three lines. One finds a pressure broadening equal to 2 κ for the sidebands and 4 w for the central component (full width at half maximum). The pressure shift increases the splitting Ω by an amount ξ.

The weight (integrated intensity) of a given line can be expressed as the product of the steady state population of the emitting dressed state by the radiative transition rate starting from this state. For example

$$I(\omega_L-\Omega) = \Gamma_{12}\,\pi_2 \qquad I(\omega_L+\Omega) = \Gamma_{21}\,\pi_1 \,. \qquad (5)$$

The steady state populations $\pi_i = \sum_n \sigma_{in,in}$ are deduced from the normalization condition $\pi_1 + \pi_2 = 1$ and from the detailed balance condition

$$(\Gamma_{12} + w)\,\pi_2 = (\Gamma_{21} + w)\,\pi_1 \qquad (6)$$

which expresses that, in the steady state, the total number of radiative and collisional transitions $|1,n> \rightarrow |2,n'>$ balances the total number of transitions $|2,n> \rightarrow |1,n'>$. In the absence of collisions ($w=0$), the detailed balance condition leads to a symmetric spectrum : $I(\omega_L-\Omega) = I(\omega_L+\Omega)$ according to (5). In the presence of collisions ($w\neq 0$) $\Gamma_{12}\,\pi_2 \neq \Gamma_{21}\,\pi_1$ and the spectrum becomes asymmetric [8-9].

3. Conditions of Validity

Both the optical Bloch equations (OBE) and the dressed atom relaxation equations (DARE) are first order differential equations resulting from a Markov approximation. The time derivatives appearing in (1) and (4) actually describe a "coarse grained evolution", averaged over a time interval Δt much longer than the collision time τ_c. This introduces some restrictions on the predictions which can be derived from these equations. We show in this section that these restrictions are less severe for DARE than for OBE.
The conditions of validity of OBE are well known. They can be written

$$\gamma\tau_c,\ \omega_1\tau_c,\ |\delta|\tau_c \ll 1 \qquad (7)$$

where γ is the collisional width, ω_1 the Rabi frequency, δ the detuning. The first condition means that the collisional relaxation time $T_R = \gamma^{-1}$ is much longer than the averaging time Δt ($\tau_c \ll \Delta t \ll T_R$) and is supposed well satisfied. The two last conditions ($\omega_1\tau_c, |\delta|\tau_c \ll 1$) which define the so-called "impact regime" express the fact that the laser atom interaction may be ignored during Δt (and in the interaction representation with respect to the atomic Hamiltonian H_A). This is why the collisional relaxation matrix $\mathcal{T}_{coll}$ is the same as in the absence of laser irradiation.

Conditions (7) may also be interpreted in the frequency domain: the coarse grained time average restricts the frequency range over which the redistribution spectrum is correctly described to an interval Δt^{-1} around the unperturbed atomic frequency ω_0 and conditions (7) just mean that the fluorescence triplet is entirely contained in this interval (Fig.3).

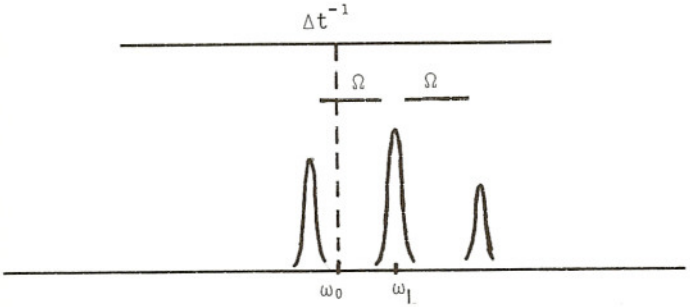

Fig. 3 Frequency range over which OBE describe correctly the redistribution. In the impact regime, the spectrum is entirely contained in this interval

In the dressed atom approach, the collisional relaxation is studied in the dressed atom interaction representation (the laser atom interaction has been first diagonalized). The spectrum derived from DARE is therefore valid in three intervals Δt^{-1} around the three Bohr frequencies ω_L, $\omega_L \pm \Omega$ of the dressed atom (Fig. 4).

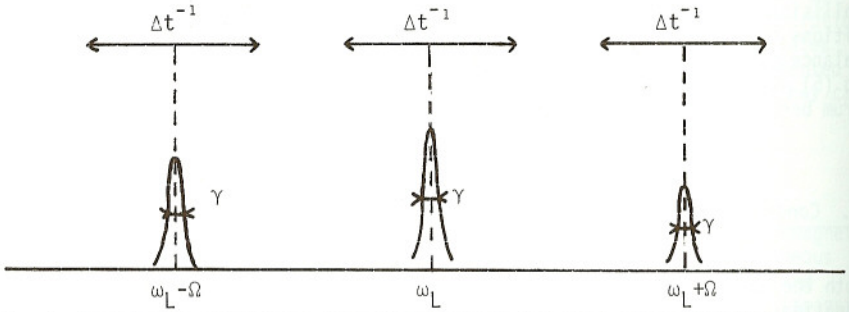

Fig. 4 Frequency intervals over which DARE describe correctly the redistribution

The only condition of validity of DARE is therefore

$$\gamma\tau_c << 1 \qquad (8)$$

which is much less severe than (7) : DARE remain valid outside the impact regime where OBE can no longer be used.

Non-Markovian effects, not contained in DARE (because of the coarse grained average),only appear outside the three intervals represented on Fig.4, i.e. in regions where the redistribution is negligible (the width γ of the three lines is much smaller than Δt^{-1}). It follows that DARE provides a Markovian description of the fluorescence spectrum for all values of ω_1 and δ.

4. Relations Between Optical Bloch Equations and Dressed Atom Relaxation Equations

In the impact regime, both OBE and DARE are valid. Thus, it must be possible to relate w, κ and ξ to the parameters describing the relaxation of the bare atom.

For the bare atom, the effect of a given collision (impact parameter b, relative velocity $\vec{v}$) is just to produce different phase shifts ϕ_e and ϕ_g for the upper and lower states (dephasing collisions, quenching neglected)

$$|e> \rightarrow |e> \exp(-i\phi_e) \qquad |g> \rightarrow |g> \exp(-i\phi_g). \qquad (9)$$

Summing over collisions (i.e. over b and $\vec{v}$) leads to a damping and a shift of the bare dipole moment described by

$$\frac{d}{dt} <e| \sigma_a |g> = -(\gamma + i\eta) <e|\sigma_a|g> \qquad (10)$$

where the two collisional parameters γ and η are given by

$$\gamma = \sum_{\text{coll}} [1 - \cos(\phi_e - \phi_g)] \qquad \eta = \sum_{\text{coll}} \sin(\phi_e - \phi_g) \ . \qquad (11)$$

Suppose now that the dressed atom is before the collision in the state $|1,n> = \cos\Theta\,|e,n> + \sin\Theta\,|g,n+1>$. The impact regime conditions express that the collision time τ_c is so short that one can decouple the atom and laser photons during the collision (i.e. compute the collision S matrix as if the atom

was free). It follows that the state $|\overline{1,n}\rangle$ after the collision can be deduced from the phase shifts ϕ_e and ϕ_g :

$$|\overline{1,n}\rangle = \cos\theta\,|e,n\rangle \exp(-i\phi_e) + \sin\theta\,|g,n+1\rangle \exp(-i\phi_g)\ . \qquad (12)$$

Since ϕ_e and ϕ_g are not equal, $|\overline{1,n}\rangle$ has a nonzero projection on $|2,n\rangle$ which means that the collision has induced a population transfer from $|1,n\rangle$ to $|2,n\rangle$

$$|\langle 2,n|\overline{1,n}\rangle|^2 = 2\cos^2\theta\sin^2\theta\,[1 - \cos(\phi_e - \phi_g)]\ . \qquad (13)$$

Summing (13) over collisions and using (11) leads to the following expression for the redistribution rate w

$$w = 2\gamma\cos^2\theta\sin^2\theta = \gamma\omega_1^2/[2(\omega_1^2+\delta^2)]\ . \qquad (14)$$

Similar calculations give

$$\kappa + i\xi = (\gamma + i\eta)\cos^4\theta + (\gamma - i\eta)\sin^4\theta \qquad (15)$$

$$\kappa = \gamma(\omega_1^2 + 2\delta^2)/[2(\omega_1^2 + \delta^2)] \qquad (16)$$

$$\xi = -\eta\delta/\Omega\ . \qquad (17)$$

Outside the impact regime, the ω_1 and δ dependence of w and κ are no longer given by (14) and (16). They can be described by introducing two "effective parameters" $g(\omega_1,\delta)$ and $h(\omega_1,\delta)$ such that

$$w = g(\omega_1,\delta)\,\omega_1^2/[2(\omega_1^2+\delta^2)] \qquad (18)$$

$$\kappa = h(\omega_1,\delta)\,(\omega_1^2+2\delta^2)/[2(\omega_1^2+\delta^2)] \qquad (19)$$

the impact regime limit of g and h being equal to γ.

The failure of OBE outside the impact regime is due to an incorrect description of the collisional relaxation which is certainly no longer independent of the laser irradiation. Similar situations exist in nuclear magnetic resonance experiments performed with strong radiofrequency fields. It is well known in this case that the relaxation must be described by modified T_1 and T_2 parameters, defined in the rotating frame and depending on the direction and magnitude of the effective field [17]. This is equivalent to the study of the relaxation in the basis of the dressed states.

It must be emphasized that such modified OBE are not obtained by simply replacing in (10) γ by an effective parameter $\gamma(\omega_1,\delta)$. Such a modification would actually lead to expressions (18) and (19) with $g(\omega_1,\delta) = h(\omega_1,\delta) = \gamma(\omega_1,\delta)$ and there is no reason why g and h should be equal. For example, in the perturbative limit ($\omega_1 \ll \delta$), one can show that h remains equal to γ outside the impact regime which is not the case for g [20].

To summarize, the two parameters w and κ (or g and h) are independant outside the impact regime. They are not related to a single parameter as is the case in the impact regime.

5. Absorption Profile

In this section we discuss the absorption profile $A(\delta)$ which is recorded when the net absorption of laser photons is plotted versus the detuning δ. The total numbers of absorbed and reemitted photons are obviously equal so that $A(\delta)$ can be obtained by summing the weights of the three emission lines. These weights are functions of the radiative rates Γ_{ij} and of the collisional redistribution rate w and can therefore be expressed in terms of ω_1,δ,Γ (spontaneous emission rate from the upper level) and g (effective parameter

appearing in the expression (18) of w). One gets in this way

$$A(\delta) = \frac{\Gamma}{2} (\Gamma + 2g)\, \omega_1^2 / [(\Gamma + 2g)\omega_1^2 + 2\Gamma\delta^2] \,. \qquad (20)$$

This expression is nothing but the Karplus Schwinger formula [2] (in the secular approximation) generalized outside the impact regime [6].

In the impact regime, g is a constant (equal to γ) and the absorption profile has a Lorentzian shape. Outside the impact regime, this is no longer true because of the δ dependence of g. Thus, it clearly appears that the calculation of w outside the impact regime is closely related to the standard problem of non-Lorentzian far wing absorption.

The absorption profile $A(\delta)$ only depends on w. This explains why it may be fitted by replacing γ by $g(\omega_1,\delta)$ in OBE. Such a method would lead to a wrong result for the redistribution spectrum which also depends on κ.

6. Conclusion

We have presented in this paper a dressed atom approach to collisional effects in resonance fluorescence which provides an interpretation of collisional redistribution in terms of collision induced transition rates between the dressed states. Such an approach generalizes the perturbative picture of Fig.1 to high intensity and resonant situations.

Furthermore, when compared with the method of optical Bloch equations, the dressed atom approach not only provides a simpler physical unsight but appears to have a larger domain of validity. It gives a correct Markovian description of absorption and of spectral redistribution even outside the impact regime.

Another illustration of the advantages of the dressed atom method may be found in the following paper dealing with the interpretation of resonances between "unpopulated levels" in nonlinear optics.

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