SPONTANEOUS RAMAN EFFECT IN INTENSE LASER FIELDS

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1. Introduction

In this paper, we discuss, from a theoretical point of view, how spontaneous Raman effect is modified at very high laser intensities.

Up to now, intense field effects have been mainly investigated, both theoretically [1] and experimentally [2], in the simple case of 2-level systems. Observation of Raman processes requires systems having at least three levels, one in the upper state, two in the lower state. At first sight, one could think that going from two to three levels complicates very much the algebra, since we have now eight Bloch's equations for the density matrix elements instead of three. In fact, this is not true, and we would like to show in this paper how it is possible to understand the modifications of Raman effect with practically no new calculations, even if the presence of two sublevels in the lower state leads, through optical pumping effects, to results qualitatively different from those of the 2-level case.

Let's first give some notations (Fig. 1). We will call e the upper atomic state, g and g' the two sublevels of the lower state separated by a splitting S, ω_0 and ω'_0 the frequencies of the two transitions e-g and e-g', Γ the natural width of e, equal to the sum of the two spontaneous transition rates γ and γ' from e to g and from e to g'.

We consider a beam of such atoms irradiated at right angles by a single mode laser so that one gets rid of Doppler effect. The laser has a frequency ω_L and a polarization such that it can excite both transitions e-g and e-g'. The coupling of the laser with these two transitions is characterized by the two Rabi frequencies $\omega_1 = \vec{E}_L.\vec{d}_{eg}$, $\omega'_1 = \vec{E}_L.\vec{d}_{eg'}$ equal to the product of the laser electric field $\vec{E}_L$ by the dipole moments $\vec{d}_{eg}$ and $\vec{d}_{eg'}$ associated respectively with e-g and e-g'.

The theoretical problem we are interested in concerns the scattered light emitted perpendicularly to the laser and atomic beams. How does the spectrum of this scattered light change when the laser intensity I_L is progressively increased from very low to very high values ?

We will first briefly recall the well known lowest order results concerning Raman effect [3]. Then, some higher order processes which become important at higher intensities will be discussed. We will also consider the case where the laser is tuned in resonance with one transition, saturating this transition but not the second one. Finally, we will discuss the very high intensity limit where the laser is so intense that it can saturate both

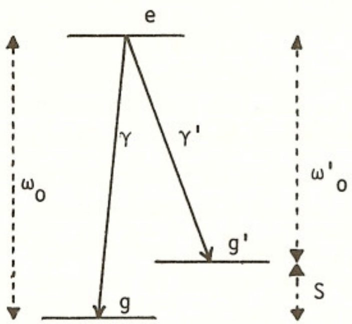

Fig. 1 Energy diagram of the 3-level atomic system

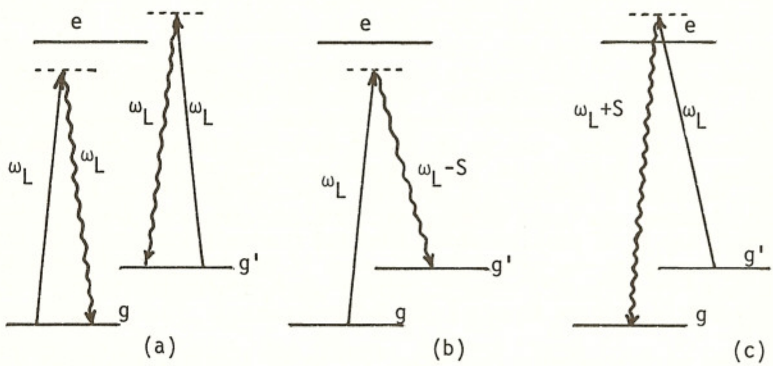

Fig. 2 Lowest order processes corresponding to Rayleigh (a), Raman Stokes (b) and Raman anti Stokes (c) scattering.

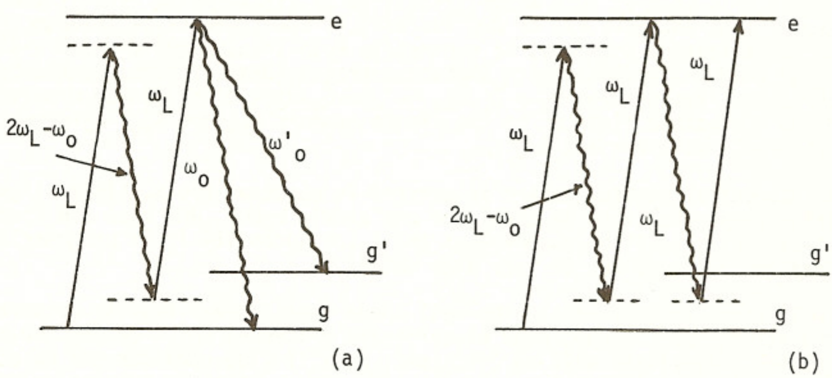

Fig. 3 Some higher order processes

transitions. The emphasis will be put on physical discussions rather than on detailed calculations which can be found in Ref. [4].

2. Lowest order results

Three types of processes can occur at very low intensities (Fig. 2) : Rayleigh type processes (Fig. 2-a) where the atom starts and ends in the same sublevel of the lower state, g or g', absorbing one laser photon ω_L and reemitting one photon which must have exactly the laser frequency because of energy conservation. Raman Stokes processes (Fig. 2-b) where the atom, starting from g, absorbs one ω_L photon and ends in g', emitting a photon with frequency ω_L-S. The symmetric process where the atom starts from g' and ends in g, absorbing ω_L and emitting ω_L+S, is called Raman anti Stokes (Fig. 2-c).

At very low intensities, one expects therefore in the spectrum of the scattered light three delta-functions at ω_L, ω_L-S, ω_L+S, each of them having a weight proportional to the laser intensity I_L. Note also that because of Raman processes, there is an optical pumping of atoms from g to g' or from g' to g. If the interaction time is sufficiently long, a steady state will be reached in which the number of transitions from g to g' balances the number of transitions from g' to g. This means that, in the steady state, the number of photons emitted at ω_L-S and ω_L+S are the same, so that the spectrum is symmetric. Finally, if $\omega'_1 = 0$, i.e. if the laser polarization is such that the transition e-g' cannot be excited, each atom pumped in g' will not be able to leave this state, so that, after a certain time, all atoms will be trapped in g' and the scattered light will vanish.

3. Some higher order processes

If the light intensity is increased, it becomes necessary to consider higher order processes where, instead of interacting with a single laser photon, the atom interacts with two laser photons, three laser photons ... and so on. Some of these higher order processes are sketched on Fig. 3.

For example (Fig. 3-a), the atom can absorb two laser photons (represented by full arrows) and emit two fluorescence photons (represented by wavy arrows). The corresponding amplitude is large when, after the absorption of the second laser photon, the atom reaches the excited state within the natural width Γ of this state. This means that the first fluorescence photon has a frequency $2\omega_L-\omega_0$ (within Γ) and also that the second fluorescence photon has a frequency ω_0 or ω'_0 according as the atom ends in g or in g'. Similar processes occuring from g' lead to fluorescence photons at $2\omega_L-\omega'_0$ and also at ω_0 or ω'_0. We therefore predict four new lines appearing at the four frequencies ω_0, ω'_0, $2\omega_L-\omega_0$, $2\omega_L-\omega'_0$, with an intensity proportional to I_L^2, since two laser photons are involved, and a width of the order of Γ due to the width of the upper level e.

We have also represented on Fig. 3-b an example of a three photon process in which, after having been put in the upper state e as in the two previous examples, the atom emits a fluorescence photon and comes back to e by absorbing a third laser photon ω_L. This can be considered as an "inverse Rayleigh" process from e and gives rise to a new line at ω_L, with an intensity proportional to I_L^3 and a width of the order of Γ.

All these perturbative results which are valid for sufficiently large detunings between the laser and atomic frequencies can be confirmed by a more precise calculation. One finds that there are seven lines in the spectrum of the scattered light. The more intense ones, proportional to I_L, are the Rayleigh (ω_L) and Raman ($\omega_L \pm S$) lines. The Raman lines are not infinitely narrow, as predicted by lowest order theory, but they have a small width of the order of $1/T_p$, where T_p (pumping time between g and g') can be considered as the lifetime of the lower state, much longer however than the lifetime $1/\Gamma$ of the upper state. The Rayleigh line has a structure. One component has a width of the order of $1/T_p$, as the Raman lines. The second component is a $\delta(\omega-\omega_L)$ function corresponding to the coherent scattering by the mean dipole moment driven at ω_L by the laser wave. Then, we have weaker lines, in $I_L{}^2$ or $I_L{}^3$, at ω_0, ω'_0, $2\omega_L-\omega_0$, $2\omega_L-\omega'_0$, ω_L, with a width of the order Γ, corresponding to the higher order processes discussed above. Finally, one must note that the spectrum is symmetric in steady state. We have already discussed this point for the Raman lines. For the weaker lines, this symmetry is partly due to the fact that, in the second order processes, the fluorescence photons always appear by pairs.

If the laser intensity is still increased, it is clear that the perturbative approach used so far breaks down, especially if the laser is tuned in resonance with one of the 2 transitions.

4. Laser in resonance with one transition, saturating this transition but not the second one

Suppose for example that $\omega_L = \omega_0$, so that the laser is in resonance with e-g, and suppose that $\omega_1 >> \Gamma$, so that e-g is saturated. The detuning of the laser with the second transition e-g' is equal to the splitting S between g and g'. We will suppose first that $\omega'_1 << S$, so that the laser is not sufficiently intense for saturating e-g' : e-g is saturated but not e-g'.

In order to understand what happens in that case, the idea is, in a first step, to forget g' and to apply the well known results of the two-level case to the e-g transition which is saturated. Then, in a second step, one can try to understand the perturbation due to g'.

Let's consider first some energy levels of the combined system atom + laser photons, without any coupling, i.e. with $\omega_1 = \omega'_1 = 0$. Since $\omega_L = \omega_0$, the two states $|g, n>$ (atom in g with n photons) and $|e, n-1>$ (atom in e with n-1 photons) are degenerate . The same result holds for $|g, n+1>$ and $|e, n>$ which are at a distance ω_L above. On the other hand, $|g', n>$ is at a distance S above $|g, n>$, $|g', n+1>$ at a distance S above $|g, n+1>$ (Fig. 4-a).

Now, we introduce the coupling ω_1 between $|g, n>$ and $|e, n-1>$, $|g, n+1>$ and $|e, n>$ ..., but we maintain $\omega'_1 = 0$, i.e. we still ignore any coupling between $|g', n>$ and $|e, n-1>$, $|g', n+1>$ and $|e, n>$ ... The two unperturbed states $|g, n>$ and $|e, n-1>$ transform into two perturbed states $|2, n-1>$ and $|3, n-1>$, separated by ω_1 , and similarly $|g, n+1>$ and $|e, n>$ transform into $|2, n>$ and $|3, n>$. The spontaneous transitions between these two doublets, represented by full arrows on Fig. 4-a, give rise, in the scattered light, to a well known triplet, formed by three lines, one at $\omega_L+\omega_1$ (transition $|2, n> \rightarrow |3, n-1>$), one at $\omega_L-\omega_1$ (transition $|3, n> \rightarrow |2, n-1>$), one at ω_L (degenerate transitions $|i, n> \rightarrow |i, n-1>$ with i = 2, 3). The widths are of the order of Γ, since the width of $|e, n>$ is shared between the two perturbed states $|2, n>$ and $|3, n>$. Since the transition is saturated, there is no mean dipole moment, and consequently, no $\delta(\omega-\omega_L)$ function corresponding to a coherent scattering.

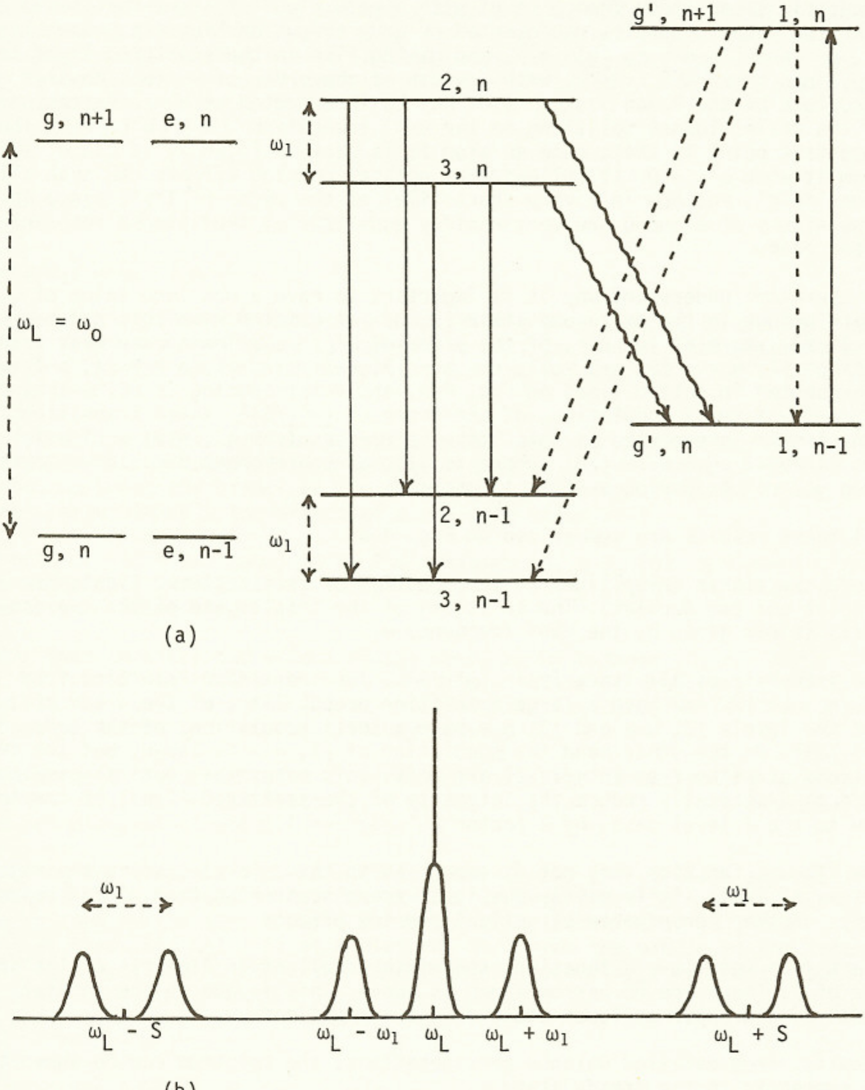

Fig. 4 Raman effect in intense laser fields ($\omega_L = \omega_0$, $\omega_1 \gg \Gamma$, $\omega'_1 \ll S$). Various energy levels of the system atom-laser photons and various transitions between these energy levels (Fig. a) giving rise to the spectrum represented on Fig. b.

Even if $\omega'_1 = 0$, i.e. even if the laser is not coupled to e-g', spontaneous transitions can occur from e to g' with a rate γ'. It follows that there are also two decay channels, represented by wavy arrows on Fig. 4-a, connecting $|2, n>$ and $|3, n>$ to $|g', n>$, and giving rise in the scattered light to two lines at $\omega_L - S \pm \omega_1/2$, with a width of the order of Γ. This doublet structure of the Raman Stokes line may be interpreted as a manifestation of the Autler-Townes splitting on the e-g' transition [5] [6] [7]. The important point is that, once an atom falls back in $|g', n>$, it cannot escape from it when $\omega'_1 = 0$. It follows that optical pumping effects can trap all atoms in g', and now in a very short time, of the order of $1/\gamma'$, since the populations of e and g are very rapidly equalized by the intense resonant laser beam.

We therefore understand why it is important to have a non zero value of ω'_1, which brings in the perturbed state $|1, n>$ associated with $|g', n+1>$ a small admixture of $|e, n>$, of the order of ω'_1/S, allowing new weak transitions $|1, n> \rightarrow |2, n-1>$, $|1, n> \rightarrow |3, n-1>$ and $|1, n> \rightarrow |1, n-1>$ represented in dotted lines on Fig. 4-a, and reintroducing in $|2, n-1>$, $|3, n-1>$ a small population, of the order of $(\omega'_1/S)^2$. These transitions can also be interpreted as anti Stokes processes from g', giving rise to two lines at $\omega_L + S \pm \omega_1/2$. There is also a non-resonant Rayleigh process from g', reintroducing a $\delta(\omega-\omega_L)$ function.

All these results are summarized on Fig. 4-b.

There are always seven lines in the spectrum of the scattered light, one triplet and two doublets. The splitting of the triplet and of the two doublets is now given by the Rabi frequency ω_1.

The intensity of all lines is in $(\omega'_1/S)^2$. The transitions starting from $|2, n>$ or $|3, n>$ have a large transition probability, of the order of 1, but the levels $|2, n>$ and $|3, n>$ have a small population, of the order of $(\omega'_1/S)^2$. On the other hand the population of $|1, n>$ is large, but the transitions starting from this level are weak. This means that optical pumping effects drastically reduce the intensity of the scattered light, in comparison to the 2-level case, by a factor $(\omega'_1/S)^2 \ll 1$.

The $\delta(\omega-\omega_L)$ function does not disappear as in the 2-level case in the high intensity limit. It is mainly due to coherent scattering from g' and represents another consequence of optical pumping effects.

Apart from the $\delta(\omega-\omega_L)$ function, the width of all other lines is of the order of Γ. There are no narrow lines as above. This is due to the finite width of the upper or lower state of the transitions.

Finally, from detailed balance considerations, the spectrum can be shown to be symmetric in the steady state.

5. Very high intensity limit : Laser saturating both transitions

One could think that going to the very high intensity limit, where both ω_1 and ω'_1 are large compared to Γ and S, so that both transitions e-g and e-g' are saturated, can suppress all these optical pumping effects and lead to a saturation of the scattered intensity. We would like now to show that this is not true.

Let's first consider the particular case where S = 0, i.e. where g and g' are degenerate so that $\omega_0 = \omega'_0$ and let's suppose that the laser is in resonance with these 2 transitions, so that $\omega_0 = \omega'_0 = \omega_L$. Since the 2 levels g and g' are degenerate, one can choose any new basis in the lower state by taking any set of 2 orthonormal linear combinations of g and g'. If, for example, one takes

$$|G> = \frac{1}{\Omega_1}\left[\omega_1\ |g> + \omega'_1|g'>\right]$$
$$|G'> = \frac{1}{\Omega_1}\left[\omega'_1|g> - \omega_1\ |g'>\right]$$

where $\Omega_1{}^2 = \omega_1{}^2 + \omega'_1{}^2$,

one can easily show that only the transition e-G is coupled to the laser with a Rabi frequency Ω_1 whereas the second is not. It follows that we are again led to a 2-level system and to optical pumping effects which can trap all atoms in G'.

In fact, S is different from 0, but, in the limit we are considering here, Ω_1 is very large compared to S and Γ, so that we will proceed as follows. First, we treat the effect of the coupling Ω_1, S being neglected. Then, we treat perturbatively the effect of a non-zero value of S.

If both Ω_1 and S are equal to 0, the 3 states $|G, n>$, $|G', n>$ and $|e, n-1>$ are degenerate and similarly $|G, n+1>$, $|G', n+1>$, $|e, n>$ which are at a distance ω_L above (Fig. 5-a).

Let's then consider the effect of the coupling Ω_1 between $|G, n>$ and $|e, n-1>$, $|G, n+1>$ and $|e, n>$. One gets a series of doublets, $|1, n-1>$ and $|3, n-1>$, $|1, n>$ and $|3, n>$ with a splitting Ω_1 . The level $|G', n+1>$ is not perturbed and remains half way between $|1, n>$ and $|3, n>$.

We now treat the effect of S perturbatively. The corresponding operator, $S|g'><g'|$, has in the basis $|1, n>$, $|3, n>$, $|G', n+1>$ both diagonal and off diagonal elements.

The diagonal elements represent first order energy shifts which are small compared to Ω_1 since $S \ll \Omega_1$. One can easily show that $|1, n>$ and $|3, n>$ are shifted by the same amount, different from the shift of $|G', n+1>$. It follows that $|G', n+1>$ is at a distance $S\tau/2$ from the middle of the interval $|1, n>$, $|3, n>$, where the dimensionless parameter τ is easily found to be given by :

$$\tau = \frac{2\omega_1{}^2 - \omega'_1{}^2}{\omega_1{}^2 + \omega'_1{}^2} \quad .$$

If one neglects the off diagonal elements associated with S, the state $|G', n+1>$ remains not coupled to $|1, n-1>$ and $|3, n-1>$ by spontaneous emission, so that one gets only the spontaneous transitions connecting $|1, n>$ and $|3, n>$ to $|1, n-1>$ and $|3, n-1>$, represented by full arrows on Fig. 5-a, and forming a triplet at ω_L, $\omega_L \pm \Omega_1$ and the spontaneous transitions connecting $|1, n>$ and $|3, n>$ to $|G', n>$, represented by wavy arrows on Fig. 5-a and giving rise to two lines at $\omega_L + (\Omega_1/2) - (S\tau/2)$ and $\omega_L - (\Omega_1/2) - (S\tau/2)$.

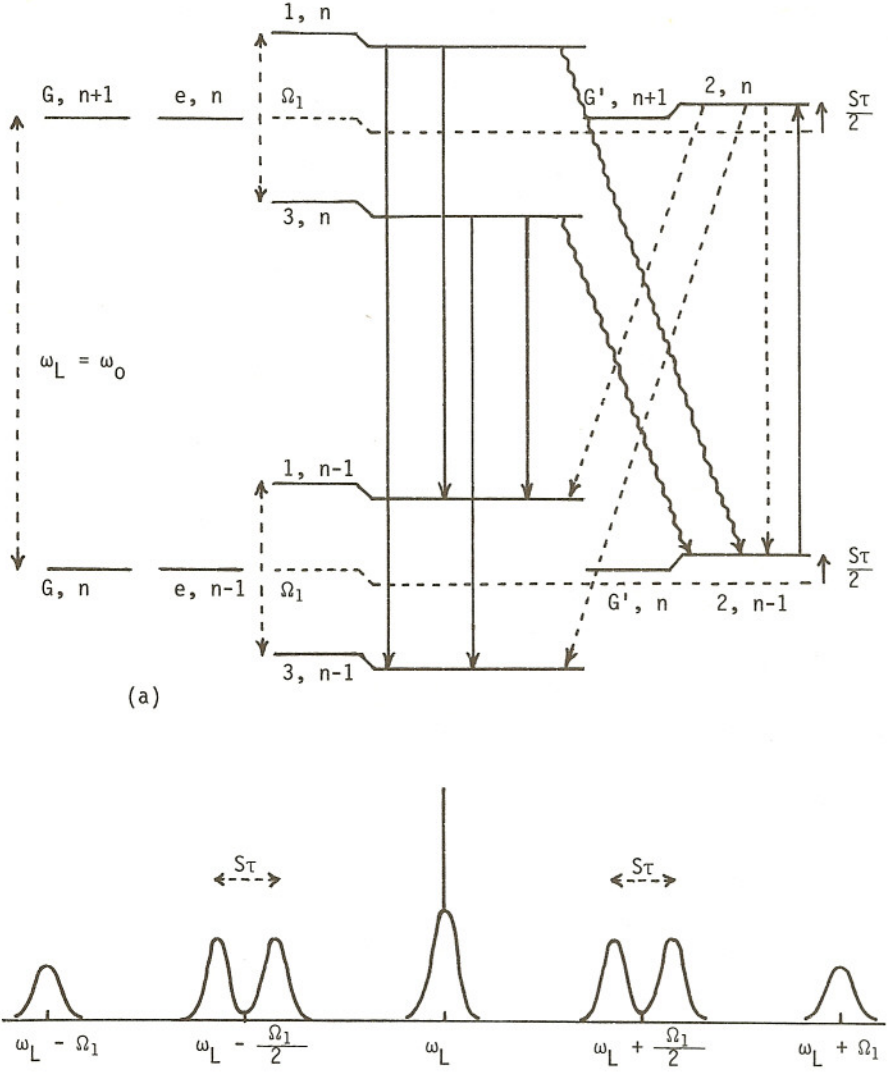

Fig. 5 Raman effect in very intense fields (ω , ω'_1 >> S, Γ). Various energy levels of the system atom-laser photons and various transitions between these energy levels (Fig. a) giving rise to the spectrum represented on Fig. b.

Actually, the off diagonal elements associated with S bring in the perturbed state $|2, n>$ corresponding to $|G', n+1>$ a small admixture of $|e, n>$, allowing spontaneous transitions from $|2, n>$ to $|1, n-1>$ and $|3, n-1>$ (represented in dotted lines on Fig. 5-a) and reintroducing in $|1, n-1>$ and $|3, n-1>$ a small population, of the order of $(S/\Omega_1)^2$. These transitions can also be interpreted as non resonant scattering processes from G', giving rise to two lines at $\omega_L - (\Omega_1/2) + (S\tau/2)$ and $\omega_L + (\Omega_1/2) + (S\tau/2)$ and to a $\delta(\omega-\omega_L)$ function corresponding to coherent scattering.

We therefore arrive to the following conclusions concerning the very high intensity limit where both transitions e-g and e-g' are saturated (Fig. 5-b).

We have always seven lines, but now they form three singlets and two doublets.

The main splittings are determined by Ω_1. S only appears in the splittings of the doublets. This means that we have a complete mixing between the initial Rayleigh, Raman Stokes and Raman anti Stokes lines which can be more precisely followed by a computer calculation of the position of the seven lines for an arbitrary intensity. Fig. 11 of Ref. [4] shows the 3 Rayleigh and Raman lines which first split into a triplet and 2 doublets and which progressively transform into 3 singlets and 2 doublets when the laser intensity is increased.

The most important point concerns perhaps the intensity of all lines which is in $(S/\Omega_1)^2$, i.e. in $1/I_L$. The reason is, as above, that strong transition probabilities correspond to weak populations and vice-versa.

This clearly shows the importance of optical pumping effects.

- First, not only the intensity of the lines does not saturate but it decreases as $1/I_L$.

- Second, we have an accumulation of atoms in G', i.e. in a linear superposition of g and g'.

- Third, the $\delta(\omega-\omega_L)$ function, which is due to coherent scattering from G', does not disappear.

Finally, as above, one can show that the width of all other lines is of the order of Γ and that the spectrum is symmetric in the steady state.

In conclusion, we have shown in this paper how it is possible to understand the modification of Raman effect when the laser intensity is progressively increased from very low to very high values.

We have only considered pure radiative effects. Collision processes in 3-level systems can also lead to very interesting effects. By introducing transfers between the various energy levels, they are responsible for important asymmetries in the spectrum of the scattered light. We will not enter into these problems since they are discussed by CARLSTEN and RAYMER [8].

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