A NEW TYPE OF RESONANCES IN SATURATED ABSORPTION SPECTROSCOPY OF 3-LEVEL SYSTEMS

M. HIMBERT, S. REYNAUD, J. DUPONT-ROC and C. COHEN-TANNOUDJI
Ecole Normale Supérieure and Collège de France, 75231 Paris Cedex 05, France

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We report the experimental observation of new resonances in saturated absorption spectra of a $J = 1$ to $J = 0$ transition of Ne atoms in a static magnetic field. These resonances, which are distinct from the well-known Zeeman and cross-over resonances, result from the modification of stimulated Raman processes by the simultaneous resonant saturation of an optical transition. The light-shifts of the various resonances are also studied.

1. Introduction

Saturated absorption spectroscopy [e.g. 1], with its recent developments [e.g. 2], is now a general and powerful method for investigating atomic structures. Narrow sub-Doppler resonances are observed on the absorption (or dispersion) of a detection beam when atoms are simultaneously interacting with a counterpropagating pump beam.

An interesting situation occurs when optical transitions joining a level b to two sublevels a and a' of a different level are considered. The structure $a - a'$ can be measured by comparing the two sub-Doppler saturation resonances ab and $a'b$. It is well-known that halfway between these two resonances, one observes a so-called "cross-over" resonance corresponding for example to the modification of the absorption on ab due to the saturation of $a'b$. We show in this paper that it is possible to observe two extra resonances which are twice farther from the cross-over than the two resonances ab and $a'b$ and which, to our knowledge, have not yet been predicted or reported.

We interpret these new resonances as related to a modification of the stimulated Raman processes between a and a' (involving one photon from both counter-propagating beams) due to a simultaneous resonant saturation of ab or $a'b$ (by the pump beam). Therefore, these resonances, which we will call from now Raman type resonances, involve at least two pump photons, and require higher intensities of the pump beam than the ab, $a'b$ and cross-over resonances. We should point out that in 2-level systems, if processes involving more than one detection photon are, as in this paper, excluded (weak detection beam), higher order processes (with respect to the pump beam) don't give rise to any new structure (they produce only a power broadening). This clearly distinguishes the Raman type resonances described in this paper, which are specific of 3-level systems and which do not require an intense detection beam, from other higher order resonances which can also be observed on 2-level systems but which involve at least two photons from the detection beam, or use for the pump a standing wave rather than a running one [3].

In this letter we will restrict ourselves to a simple discussion based on the consideration of population holes. It is well known that the dynamical Stark splitting produced by the intense pump beam plays an important role for a quantitative understanding of the width and the depth of the saturation dip [4]. We have performed a complete calculation of saturated absorption of 3-levels systems based on the dressed atom approach and the use of frequency diagrams, which confirms all the conclusions of this paper [5].

2. Experimental set up

Our experiment has been done on the transition $2p_3$ ($J=0$) to $1s_4$ ($J=1$) of ^{20}Ne at 6074 Å (Paschen notations). A weak d.c. discharge ($i = 20$ mA) is maintained in a cell filled with 0.2 Torr of natural Ne. A solenoid produces a static magnetic field B_0 (up to 300 gauss) parallel to the direction of two counterpropagating beams, the pump beam and the detection beam (fig. 1a).

With such a geometry, the excitation and detection polarizations can be only a superposition of σ^+ and σ^-, so that only three levels are involved (fig. 1β): $a(1s_4, m=-1)$, $a'(1s_4, m=+1)$, $b(2p_3, m=0)$. The two Zeeman components of the optical line ab and $a'b$ correspond respectively to the polarizations σ^+ and σ^-. Such a 3-level system presents the following two advantages. First, the Zeeman splitting 2δ between a and a' can be changed by varying B_0 ($2\delta = 4.1$ MHz/gauss). Second, the ratio K/K' of the two Rabi frequencies K and K' characterizing the coupling of the pump laser with ab and $a'b$ can be adjusted by changing the relative amount of σ^+ and σ^- in the pump polarization.

The two laser beams come from the same dye laser and have a frequency ω_L close to the atomic frequency ω_0 in zero magnetic field. The power of the pump beam is of the order of 100 mW, the detection beam being attenuated at least by a factor 200 in order to avoid any saturation effect due to this beam. The pump beam is chopped and the synchronous modulation of detection beam recorded versus ω_L.

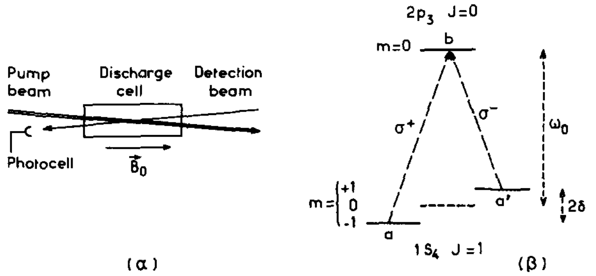

Fig. 1. α) Relative disposition of the static magnetic field B_0 and of the two counterpropagating pump and detection beams. β) Energy level scheme.

3. Saturation resonances

In order to understand the various resonances which can be observed on the absorption of the detection beam, it will be useful to introduce first the frequencies

$$\omega_p(v) = \omega_L(1 + v/c), \quad \omega_d(v) = \omega_L(1 - v/c), \qquad (1)$$

which are the apparent frequencies of the pump and detection beams in the rest frame of an atom of velocity v. For certain values of ω_L only (which actually give the position of saturation resonances) it is possible to find a velocity v such that the corresponding atoms interact resonantly with both beams.

The simplest saturation resonances corresponds to a situation where both beams are simultaneously resonant on the same Zeeman transition, $a'b$ for example (fig. 2α). The saturation of $a'b$ by the σ^- component of the pump beam (double arrow) reduces the absorption of the σ^- component of the detection beam (single arrow). Such a resonance occurs for $\omega_p(v) = \omega_d(v) = \omega_0 - \delta$ which, according to (1) gives $\omega_L = \omega_0 - \delta$, $v=0$. A similar situation appears for $\omega_L = \omega_0 + \delta$, $v=0$ (saturation of ab). Another type of resonance is the cross-over resonance appearing when two different transitions sharing a common level are simultaneously and resonantly excited. For example (fig. 2β), the saturation of ab by the σ^+ component of the pump increases the population of b and, consequently, reduces the absorption on $a'b$ of the σ^- component of the detection beam. The position of this resonance is given by $\omega_p(v) = \omega_0 + \delta$, $\omega_d(v) = \omega_0 - \delta$ i.e., according to (1), $\omega_L = \omega_0$, $v/c = \delta/\omega_L$ [exchanging the polarization of both beams gives the same resonance at $\omega_L = \omega_0$ but with $v/c = -\delta/\omega_L$].

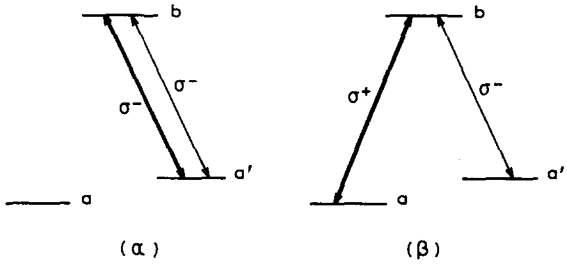

Fig. 2. Resonant processes occuring in the Zeeman saturation resonance $a'b$ (α), and the cross-over one (β). Double arrows: pump photons. Single arrows: detection photons.

The new resonances studied in this work correspond to higher order processes involving at least two pump photons (but only one detection photon). As the previous resonances, they appear for values of ω_L such that two processes can be simultaneously resonant for the same atom‡ for example those of fig. 3α. There is first a stimulated Raman process between a and a' (either from a to a' by absorption of a σ^+ pump photon and stimulated emission of a σ^- detection one, or symmetrically, from a' to a by the reverse processes). On the other hand, we have also a resonant saturation of $a'b$ by the σ^- component of the pump which reduces the population of a' and consequently, modifies the balance between the two symmetric Raman processes $a \to a'$ and $a' \to a$ in favour of the first one, producing an amplification of the σ^- component of the detection beam, or, equivalently, a reduction of its absorption. The two resonance conditions $\omega_p(v) - \omega_d(v) = 2\delta$, $\omega_p(v) = \omega_0 - \delta$ give [according to (1)], $\omega_L = \omega_0 - 2\delta$, $v/c = +\delta/\omega_L$. One would get similarly, for the processes of fig. 3β, $\omega_L = \omega_0 + 2\delta$, $v/c = -\delta/\omega_L$.

If we restrict ourselves to very weak detection beams, and consequently, to processes involving only one detection photon, one can easily show that there are no other resonances than the five previously discussed and occurring at ω_0, $\omega_0 \pm \delta$, $\omega_0 \pm 2\delta$. The single detection photon is necessarily involved, either in an optical resonance, and then the pump photons must be resonant with the same transition (fig. 2α), or with the other one (fig. 2β), or in a resonant Raman process, in which case the second resonant process can be only an optical resonance for the pump (fig. 3). Any higher order process (with respect to the pump) would correspond to additional absorptions and stimulated emissions of pump photons, leading to well known radiative effects (power broadening and light shifts).

‡ This situation reminds the one studied by Woerdman and Schuurmans [6] where one observes cross-resonances between one and two-photon absorption processes in a three level system with a nearly resonant intermediate leve.

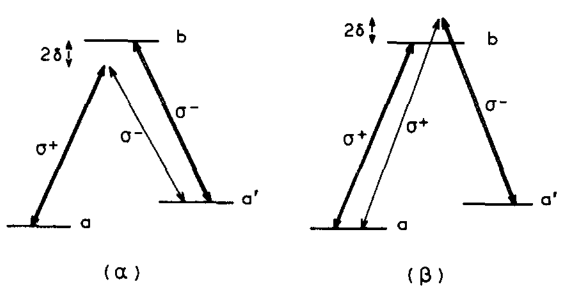

Fig. 3. Resonant processes occuring in the new Raman type resonances: $\omega_0 - 2\delta$ (α), and $\omega_0 + 2\delta$ (β).

4. Experimental results

In order to simplify the spectrum, we have first taken a pure polarization for the detection beam (σ^- for example), so that the resonances $\omega_0 + \delta$ and $\omega_0 + 2\delta$ disappear. It must be pointed out however that the pump beam must contain both polarizations σ^+ and σ^- if one wants the Raman type resonance $\omega_0 - 2\delta$ to be observable (see fig. 3α). The relative amount of σ^+ and σ^- remains a free parameter which is optimized, according to the following considerations.

Let's first show that the ratio of the amplitudes of the two resonances $\omega_0 - 2\delta$ and $\omega_0 - \delta$ is of the order of $(K/\delta)^2$. In both cases (see fig. 2α and 3α), the reduction of the population of a' due to the saturation of $a'b$ by the pump is the same. But the height of the Raman type resonance is proportional, not only to the intensity of the detection beam, as the Zeeman resonance, but also to the intensity K^2 of the σ^+ component of the pump beam divided by the square of the energy defect 2δ of the non resonant intermediate state appearing in the Raman process of fig. 3α. It follows that K must be taken as high as possible.

On the other hand, the width of the various resonances are determined by the power broadening due to the resonant component of the pump, which is the σ^+ component for the cross-over (width K) and the σ^- one for the Zeeman and Raman type resonances (width K'). If one wants the Raman type resonance to be well resolved from the stronger Zeeman resonance, one must therefore take their common width K' small compared to their splitting δ, but not smaller than the homogeneous width γ if one wants to maintain an appreciable saturation of $a'b$ by the σ^- component of the pump. To summarize, the pump polarization must be very close to σ^+ (K as large as possible) and adjusted so that $K' \sim \gamma$.

One can see on fig. 4 some experimental curves obtained in such conditions. They clearly demonstrate the existence of the Raman type resonances. The two narrow Zeeman and Raman type resonances move when the static field B_0 is increased, contrarily to the broad cross-over one. As predicted above, the height of the Raman type resonance decreases when δ is increased.

One can also detect the modulation of the previous signal in phase with a small modulation of the static field around B_0. The cross-over resonance, which is B_0-independent, disappears whereas the Zeeman and Raman type resonances remain as two narrow dispersion shaped curves (see fig. 5). Such a signal has been used for measuring the position of the Zeeman and Raman type resonances versus δ. The experimental points represented on fig. 6 show an important deviation from the linear laws $\omega_0 - \delta$ and $\omega_0 - 2\delta$ predicted above (dotted lines of fig. 6). In particular, at

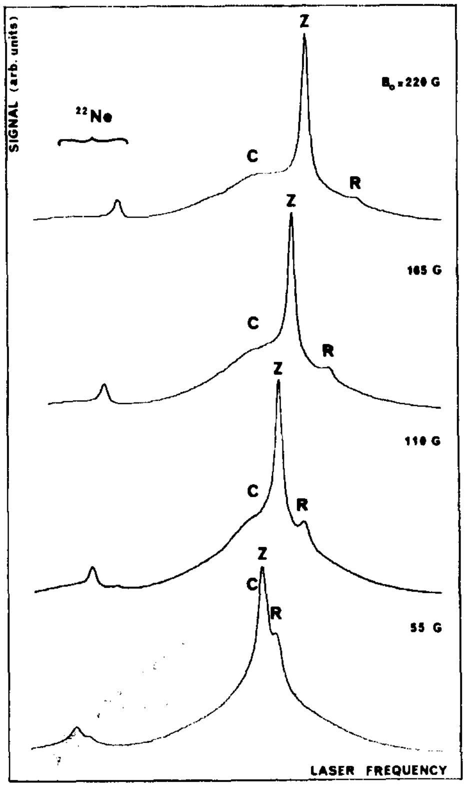

Fig. 4. Saturated absorption signal (arbitrary units) versus laser frequency ω_L for various magnetic fields B_0. C, Z, R respectively indicate the cross-over, Zeeman and Raman type resonances for ^{20}Ne. The Zeeman resonances for ^{22}Ne, appearing on the left side (higher frequencies), fix the frequency scale (isotopic shift equal to 1704 MHz).

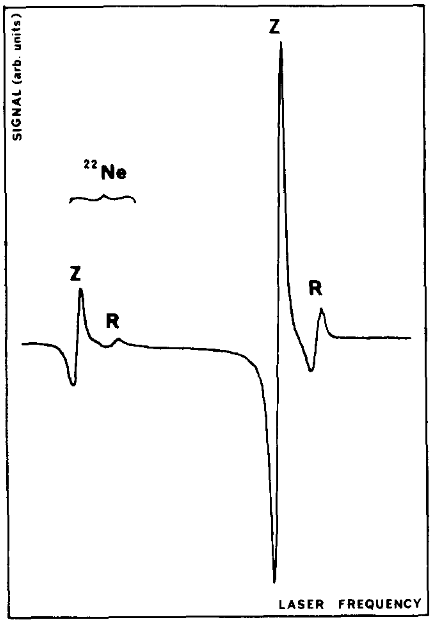

Fig. 5. Modulation of the saturated absorption signal (arbitrary units) in phase with a small modulation of the static field around B_0 = 165 G versus laser frequency ω_L. The cross-over resonance, which is B_0-independant, disappears. The Zeeman (Z) and Raman type (R) resonances of ^{20}Ne and ^{22}Ne remain as dispersion shaped curves.

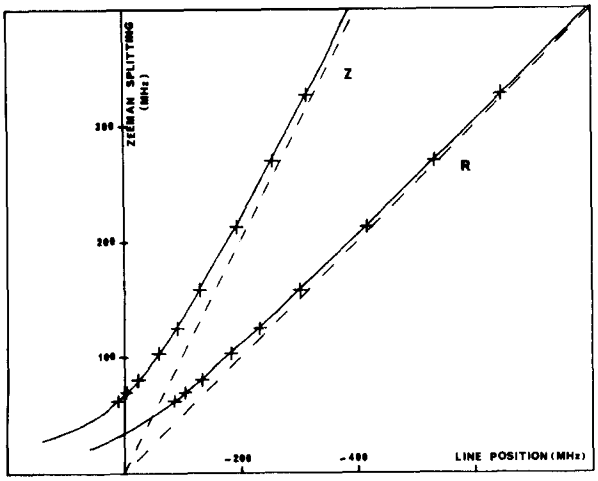

Fig. 6. Positions of the Zeeman (Z) and Raman type (R) resonances of ^{20}Ne (with respect to the atomic frequency in zero-field) for various Zeeman splittings δ. The dotted lines represent the linear laws, $\omega_0 - \delta$ for Z and $\omega_0 - 2\delta$ for R. The full lines give the theoretical light-shifted positions. The crosses are the experimental results.

low magnetic fields, the shift of the line may be as large as the Zeeman effect. These deviations are actually due to light shifts. Coming back to fig. 2α and 3α, one sees that the σ^+ component of the pump beam, which is non resonant on the transition ab (by an amount -2δ) is responsible for an upwards light shift $\epsilon = (K/2)^2/2\delta$ of level b and for a downwards one, $-\epsilon$, of level a (the light shift produced by the much weaker σ^- component of the pump beam is negligible). Introducing these light shifts in the resonance conditions written above, one easily deduces the shifted positions, $\omega_0 - \delta + \epsilon$ for the Zeeman resonance and $\omega_0 - 2\delta + (\epsilon/2)$ for the Raman type one. These theoretical predictions represented by the full lines of fig. 6 are in excellent agreement with the experimental points.

5. Conclusion

We have demonstrated the existence of new resonant structures in saturated absorption spectra of 3-level systems. We have identified the physical processes at the origin of these new resonances and checked the theoretical predictions concerning in particular the light shifts.

These new resonances always exist in saturated absorption spectra of 3-level systems even if the experimental conditions are not, as here, optimized for having them resolved from the main resonances. At high pump intensities, it could be necessary to take them into account for an accurate analysis of the lineshape of the main lines, in particular in ultra high resolution spectroscopy of multilevel systems, or in frequency standards.

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