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CONDITIONS FOR THE OBSERVATION OF THE AUTLER-TOWNES EFFECT
IN A TWO STEP RESONANCE EXPERIMENT

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Résumé. — L'objet de cet article est de déduire théoriquement les conditions d'observation du dédoublement Autler-Townes dans une expérience de résonance à deux étages où l'on étudie les effets simultanés de deux radiations visibles monochromatiques sur un système à trois niveaux. Dans un premier temps, le système à trois niveaux est considéré à repos et les effets de pompage optique vers les autres niveaux sont ignorés ; dans ce cas, le problème admet une solution analytique. Ensuite, les effets de pompage optique sont pris en compte. Finalement on considère un ensemble de systèmes à trois niveaux identiques dans une cellule, les deux faisceaux lumineux se propageant soit dans la même direction, soit dans des directions opposées. Dans ce dernier cas, le problème est pratiquement identique à celui rencontré lorsque le système à trois niveaux est au repos.

Abstract. — The object of this paper is to derive theoretically the conditions for the observation of the Autler-Townes splitting in a two step resonance experiment involving the simultaneous effects of two visible monochromatic radiations in a three-level system. First, the three-level system is considered at rest and optical pumping to other levels is ignored ; in this case, the problem is solved analytically. Following this case, optical pumping effects are taken into account. Finally, one considers a collection of identical three-level systems in a cell, the two light beams traveling either in the same direction or in two opposite directions. In this latter case, the problem is practically identical to the one for a three-level system at rest.

1. Introduction. — Since the pionneering work of Autler and Townes [1], it is well-known that, in the microwave region, molecular absorption lines can be observed to split into two components when a strong R-F field couples one of the two levels involved in the transition with a third one. This splitting was originally interpreted in terms of a dynamic Stark effect, but its origin can be also, and perhaps more deeply, understood by using the concept of dressed atom initially introduced by Cohen-Tannoudji and Haroche [2]. In the visible range, some similar effects have been shown to exist but always in rather complicated experimental situations and therefore, in a more or less indirect way. Among them, the experiments based on cross-saturated absorption techniques (see for example : [7, 8, 9, 10, 11]), and some of the results of this paper have already appeared in the literature under a more or less equivalent form. Here, we pay particular attention to the conditions necessary for the observation of the Autler-Townes splitting by recording the intensity of the fluorescence light emitted spontaneously from the upper (or the intermediate) level.

In section 2, we consider a pure three-level system at rest. The corresponding results can be applied to the atomic beam experiment if optical pumping to other levels can be neglected. In this simple case, the line profile and the conditions for observing the Autler-Townes splitting are derived analytically. In section 3, the system is still at rest but the two excited levels can decay to other levels. The corresponding effects on the line shape are derived for various approximations.

In section 4, these optical pumping effects are
ignored but the atoms are assumed to be in a cell at a given temperature, the two laser beams traveling either in the same direction or in two opposite directions. The line shape is derived analytically both in the weak and in the strong pumping field limit; in the intermediate case, some examples of numerical computer calculations are given.

2. Three-level system at rest. — The three-level system is characterized by a ground state \( |0\rangle \) and two non-degenerate excited states \( |1\rangle \) and \( |2\rangle \), the energies of these states being respectively \( \hbar \omega_0 \), \( \hbar \omega_1 \) and \( \hbar \omega_2 \). Furthermore, it is assumed that the transitions \( 0 \leftrightarrow 1 \) and \( 1 \leftrightarrow 2 \) are allowed while the transition \( 0 \leftrightarrow 2 \) is forbidden. Because of the parity selection rule, this assumption is quite realistic if we consider electric dipole transitions only. The corresponding transition probabilities are denoted \( A_{10} = \gamma_1 \) and \( A_{21} = \gamma_2 \). The system is coupled with two driving fields \( E_1(t) \) and \( E_2(t) \) which are described in a classical way:

\[
E_1(t) = 2^{-1/2} \left\{ \hat{e}_1 e^{-i\omega_1 t} + \hat{e}_1^* e^{i\omega_1 t} \right\} \hat{e}_1, \quad (1a)
\]

\[
E_2(t) = 2^{-1/2} \left\{ \hat{e}_2 e^{-i\omega_2 t} + \hat{e}_2^* e^{i\omega_2 t} \right\} \hat{e}_2, \quad (1b)
\]

\( \hat{e}_1 \) and \( \hat{e}_2 \) being the respective polarization unit vectors of the two fields. \( \Omega_1 \) and \( \Omega_2 \) are assumed to be quite different and quasi-resonant respectively for the transitions \( 0 \leftrightarrow 1 \) and \( 1 \leftrightarrow 2 \). We consider the case \( \omega_2 > \omega_1 \) (see Fig. 1) and we study the stationary part of the population of level 2 as a function of \( \delta_1 = \Omega_1 - \omega_1 + \omega_0 \) and \( \delta_2 = \Omega_2 - \omega_2 + \omega_1 \) for given intensities of the two fields.

Obviously, the main difference with the case originally considered by Autler and Townes [1] is that, in the visible range, damping phenomena due to spontaneous emission cannot be ignored. Therefore, the most convenient way to describe the evolution of the atom under the action of the two driving fields is to use a density operator. After making the usual resonant approximation (rotating wave approximation) in the coupling between the three-level system and the two driving fields, the equations of motion for the density matrix elements can be written (optical Bloch equations):

\[
\dot{\rho}_{00} = \gamma_1 \rho_{11} + i\hbar \left( \sigma_{10}^* - \sigma_{10}^* \right), \quad (2a)
\]

\[
\dot{\rho}_{11} = -\gamma_1 \rho_{11} + \gamma_2 \rho_{22} - i\hbar \left( \sigma_{10}^* + \sigma_{10}^* \right) + i\hbar \left( \sigma_{21}^* - \sigma_{21}^* \right), \quad (2b)
\]

\[
\dot{\rho}_{22} = -\gamma_2 \rho_{22} - i\hbar \left( \sigma_{21}^* - \sigma_{21}^* \right), \quad (2c)
\]

\[
\dot{\sigma}_{10} = \alpha_{10} - i\hbar \sigma_{10} + i\hbar \sigma_{21}^* \sigma_{21}, \quad (2d)
\]

\[
\dot{\sigma}_{21} = \alpha_{21} + i\hbar \sigma_{20} + i\hbar \sigma_{22} - \rho_{11}, \quad (2e)
\]

\[
\dot{\sigma}_{20} = \alpha_{20} + i\hbar \sigma_{20} - i\hbar \sigma_{21}, \quad (2f)
\]

where

\[
r = i\delta_1 - \gamma_1, \quad s = i\delta_2 - \gamma_1 - \gamma_2/2,
\]

\[
t = (\delta_2 + \delta_1) - \gamma_2/2, \quad \alpha_{10} = e^{i\omega_0 t} \rho_{10},
\]

\[
\alpha_{21} = e^{i\omega_1 t} \rho_{21}, \quad \alpha_{20} = e^{i\omega_2 t} \rho_{20},
\]

\[
I_1 = -2^{1/2} \hbar^{-1} \Delta_1 (|D, \hat{e}_1\rangle \langle 0|),
\]

\[
I_2 = -2^{1/2} \hbar^{-1} \Delta_2 (|D, \hat{e}_2\rangle \langle 1|)
\]

\( \hat{D} \) being the atomic electric dipole operator.

For large times, one can find asymptotic values of \( \rho_{00}, \rho_{11}, \rho_{22}, \sigma_{10}, \sigma_{21} \) and \( \sigma_{20} \) which are not time-dependent and which do not depend on the initial conditions. These stationary quantities obey eq. (2) in which all the time-derivatives are equal to zero. By separating the real and the imaginary parts of these linear equations, it appears that the stationary populations \( \bar{\rho}_{00}, \bar{\rho}_{11}, \) and \( \bar{\rho}_{22} \) depend on \( \omega_1^2 = I_1^* I_1 \) and \( \omega_2^2 = I_2^* I_2 \) but neither on \( I_1 I_2^* + I_1^* I_2 \) nor on \( I_1 I_2^* + I_2 I_1^* \). Thus, they are completely phase independent, provided that the coherence time of the two fields is much larger than \( 1/\gamma_1 \) and \( 1/\gamma_2 \). The way to solve such a linear system is straightforward but the final expressions are rather complicated and they will not be reported here. However, if \( | \hat{e}_2 | \) is sufficiently weak in order that \( \omega_2^2 \) is much smaller than \( \gamma_1^2 \) and \( \gamma_2^2 \) and \( \omega_1^2 \) (this is the usual probe field approximation [10], [12]) \( \bar{\rho}_{22} \) is given by

\[
\bar{\rho}_{22} = \frac{w_1^2}{(w_1^2 + 2 w_2^2)} \left\{ \frac{-s}{s + w_1^2} + C.C. \right\} \frac{w_2^2}{\gamma_2} \quad (3)
\]

If \( w_2^2 \) is much larger than \( \gamma_1^2 \) and \( \gamma_2^2 \), i.e. for high saturation for the transition \( 0 \leftrightarrow 1 \), one obtains the
classical result of Autler and Townes [1]: two resonances appear respectively for

$$\delta_2^+ = - \frac{1}{2}(\delta_1 + [\delta_1^2 + 4 w_1^2]^{1/2})$$
and

$$\delta_2^- = - \frac{1}{2}(\delta_1 - [\delta_1^2 + 4 w_1^2]^{1/2}).$$

Let us remark that, if $\delta_1^2$ is much smaller than $w_1^2$, the two resonances have approximately the same intensity, while, if $\delta_1^2$ is much larger than $w_1^2$, $P_{22}(\delta_2^2)$ is larger than $\tilde{P}_{22}(\delta_2^2)$. These characteristics are illustrated on figure 2.

On the contrary, in the limit of a vanishing pump field ($\omega_f \ll \Omega_f, \gamma_f$), only one resonance appears for $\delta_2 = - \delta_1$.

Between these two limits, it is not possible to give a general analytic expression for the position of the extrema of $\tilde{P}_{22}$ except when $\delta_1 = 0$. In this latter case $\delta_2 = 0$ for:

$$\delta_2^+ = \pm \left\{ - \frac{1}{8} \gamma_1^2 - \frac{\gamma_1 \gamma_2}{\gamma_2} w_1^2 + \frac{w_1}{2 \gamma_2} \times \left[ \gamma_1 \gamma_2 \gamma_1^2 (\gamma_1 + \gamma_2) + 4 \gamma_2 \gamma_1 (\gamma_1 + \gamma_2)^2 \right]^{1/2} \right\},$$

where:

$$\bar{P}_{11} = \frac{w_1^2}{(\Omega^* + 2 w_1)} \left\{ 1 - \beta \frac{w_1^2}{w_1^2} \right\},$$

and

$$\beta = \frac{w_1^2}{\gamma_1 \gamma_2 (\Omega^* + 2 w_1^2)} \times \left\{ \frac{s \Omega^* + (s \gamma_2 + s \gamma_1) w_1^2}{s + w_1^2} + C.C. \right\}. \quad (4)$$

which is quite realistic if the transition $0 \leftrightarrow 1$ is a resonance one, $w_{1e}$ corresponds to a saturation parameter for the transition $0 \leftrightarrow 1$, roughly equal to $\frac{\Omega}{\Delta}$, which is a rather small value considering the light power now provided by single-mode tunable lasers.

It must be noticed that the resonance $1 \leftrightarrow 2$ can be also detected by recording the intensity of the fluorescence light spontaneously emitted from level 1. Indeed, $\bar{P}_{11}$, for a given $\delta_1$, exhibits an antiresonant behaviour when $\delta_2$ is tuned. More precisely, in the weak field limit for the transition

$$1 \leftrightarrow 2 \neq (w_2^2 \ll \gamma_1^2, \gamma_2^2, w_1^2),$$

one obtains from eq. (2):

$$\bar{P}_{11} = \frac{w_1^2}{(\Omega^* + 2 w_1)} \left\{ 1 - \beta \frac{w_1^2}{w_1^2} \right\},$$

where:

$$\beta = \frac{w_1^2}{\gamma_1 \gamma_2 (\Omega^* + 2 w_1^2)} \times \left\{ \frac{s \Omega^* + (s \gamma_2 + s \gamma_1) w_1^2}{s + w_1^2} + C.C. \right\}. \quad (4)$$
In the weak field limit for the transition $0 \leftrightarrow 1$, eq. (4) reduces to:

$$\beta = \gamma_2 \bar{\rho}_{22}/\gamma_1 \omega_2^2$$

and then, as could have been foreseen, the antiresonance line has the same profile as the resonance line detected from the upper level.

In the strong field limit ($\omega_1^2 \gg \gamma_1^2, \gamma_2^2$), the antiresonance line appears to split into two components in the same way as the resonance line and with the same splitting.

In the intermediate case, the line profiles are not rigorously identical in the two cases but, in fact, they are very similar. In particular, for $\delta_1 = 0$, the two conditions on $\omega_1^2$ for observing a splitting become identical when $\gamma_2$ is much smaller than $\gamma_1$.

Finally, let us remark that, when $\omega_2^2$ is no longer small with respect to $\omega_1^2$, $\gamma_1^2$ or $\gamma_2^2$, the Autler-Townes splitting can disappear even for high values of $\omega_1^2$. This can be easily understood if one takes into account the supplementary broadening introduced by $\omega_2^2$.

3. Optical pumping effects. — In this section, the three-level system is still considered at rest. However, level 1 can decay to levels different from the ground state and level 2 can decay to new intermediate levels, some of these being allowed to decay to the ground level. It is quite clear that, in this case, the problem is much more complicated, but if the new intermediate levels decay very rapidly to the ground level, all these optical pumping effects can be taken into account by replacing:

i) in eq. (2c), $\gamma_2$ by $\gamma_2 + \Gamma_2$,

ii) in eq. (2b), $\gamma_1$ by $\gamma_1 + \Gamma_1$,

and by adding to the right-hand side of eq. (2a) a term $\rho_T \rho_{22} (\rho < 1)$.

Obviously, there are no longer stationary solutions for eq. (2) but one can write:

$$\rho_{jj} = \sum_{m=1}^{q} r_{jj}^m e^{-\lambda m t}, \quad (j = 0, 1, 2)$$

$$\alpha_{jk} = \sum_{m=1}^{q} a_{jk}^m e^{-\lambda m t};$$

the $\lambda$'s can be obtained by diagonalization of eq. (2) and the coefficients $r_{jj}^m$ and $a_{jk}^m$ are completely determined if one takes into account initial conditions. Unfortunately, such a calculation cannot be done analytically except if $\Gamma_2$ and $\Gamma_1$ are respectively much smaller than $\gamma_2$ and $\gamma_1$. In this latter case, it is quite clear that one of the $\lambda$'s is much smaller than the real part of all the other ones and the corresponding value is very close to:

$$\lambda_0 = (\Gamma_1 \bar{\rho}_{11} + \Gamma_2 (1 - p) \bar{\rho}_{22})$$

where $\bar{\rho}_{11}$ and $\bar{\rho}_{22}$ are the stationary solutions given in eq. (3) and (4).

If one records the intensity of the fluorescence light emitted spontaneously from level 2, the signal is proportional to:

$$\int_0^T \rho_{22}(t) \, dt,$$

$T$ being the time of interaction between atoms and light. With the same approximations as above and for $T$ much larger than $1/(\gamma_1 + \gamma_2)$, this quantity can be approximated by:

$$(\bar{\rho}_{22}/\lambda_0) \left\{ \exp(-\lambda_0 T) - 1 \right\}.$$}

Furthermore, if $\lambda_0 T$ is much smaller than 1, the previous quantity reduces to:

$$\bar{\rho}_{22} T \left\{ 1 - (\lambda_0 T)/2 \right\}.$$}

As it could have been foreseen, the signal is attenuated by optical pumping effects and the resonance is broadened since the attenuating factor contains terms proportional to the initial signal. However, within the considered approximations, the conditions on $\omega_1^2$ for observing a splitting is not actually modified by these optical pumping effects.

4. Velocity effects. — In this section, the optical pumping effects studied in the previous section are ignored but the three-level system is no longer considered to be at rest. In fact, one studies a collection of such three-level systems in a cell at a given temperature and one assumes that the only effects of collisions between these systems is to realize thermal equilibrium. Thus, the number of atoms having a given velocity $v$ along a given axis is:

$$N(v) = N_0 \exp(-v^2/\nu_0^2)$$

where $\nu_0$ is the mean value of the velocity modulus. Now, if we assume that the two light beams can be described as pure traveling waves, propagating either in the same direction or in two opposite directions, the signal obtained by recording the intensity of the fluorescence light emitted spontaneously from the upper level is proportional to:

$$I(\delta_2, \delta_1) = \int_{-\infty}^{\infty} \bar{\rho}_{22}(v, \delta_1, \delta_2) N_0 \exp(-v^2/\nu_0^2) \, dv$$

where $\bar{\rho}_{22}(v, \delta_1, \delta_2)$ is obtained from $\bar{\rho}_{22}(\delta_1, \delta_2)$ given in eq. (3) by replacing $\delta_1$ by

$$\delta_1 - \frac{Q_1}{c} v = \delta_1 - k_1 v$$

and $\delta_2$ by

$$\delta_2 - \frac{Q_2}{c} v = \delta_2 - k_2 v,$$
\( \varepsilon \) being 1 if the two light beams travel in the same direction and \(-1\) if they travel in two opposite directions.

However, under usual experimental conditions \( k_1v_0 \) and \( k_2v_0 \) are much larger than \( w_1, w_2, \gamma_1 \) and \( \gamma_2 \) and therefore, one can replace the rather complicated integral of eq. (6) by the simpler one:

\[
I(\delta_2, \delta_1) = \int_{-\infty}^{+\infty} N_0 \rho_{23}(v, \delta_1, \delta_2) \, dv.
\]

This integral can be evaluated in a straightforward way [10, 11], but the corresponding result is too complicated to be useful to describe the phenomena in simple terms. However, this is not true when \( \delta_1 \) is equal to zero and \( w_1 \) is much smaller than \( \gamma_1 \) and \( \gamma_2 \).

In this case, one obtains

\[
I(\delta_2) = \frac{16 w_1^2 w_2^2}{\gamma_1^2 \left( 4 \delta_2^2 + \left[ \frac{Z + 1}{Z} \gamma_2 \right]^2 \right)^2}.
\]

where \( Z = k_1/k_2 \). Therefore, the width of the resonance is larger when the two light beams travel in the same direction than when they travel in two opposite directions as it has already been noticed for a long time [10, 13]. In the case where \( \gamma_1 \gg \gamma_2 \), this result could have been obtained without integration, since from eq. (3), one can see that \( I(\delta_2) \) is contributed by those atoms having the following velocity:

\[
v = \delta_2/(k_1 + k_2)
\]

and therefore:

\[
I(\delta_2) \propto \frac{w_1^2 w_2^2}{k_1^2 v(\delta_2)^2 + \frac{\gamma_1^2}{4} \delta_2^4 + \left\{ \frac{k_1 + k_2}{2 k_1} \gamma_1 \right\}^2}.
\]

The same type of argument can be used to derive \( I(\delta_2, \delta_1) \), (\( \delta_1 = 0 \)), in the strong field limit (\( w_1^2 \gg \gamma_1^2, \gamma_2^2 \)). In this case, from the results given in section 2, the contribution to \( I(\delta_2) \) comes mainly from atoms with velocity equal to (Autler-Townes conditions):

\[
v_\pm(\delta_2) = \frac{Z}{2 k_1 (1 + Z)} \times
\]

\[
\times \left\{ (Z + 1) \delta_2 \pm \left[ Z^2 \delta_2^2 + 4(1 + Z) w_1^3 \right]^{1/2} \right\}
\]

and, therefore, by putting these values in the intermediate level population term, we get:

\[
I(\delta_2) = I_+ (\delta_2) + I_- (\delta_2)
\]

where:

\[
I_\pm (\delta_2) = \frac{w_1^2}{k_1^2 v_\pm(\delta_2)^2 + 2 w_1^2}.
\]

It appears that \( I_+ \) and \( I_- \) are resonant for \( \delta_2 = -w_1 \) and \( \delta_2 = w_1 \) respectively. We again find the classical result of Autler and Townes (1955), but \( I(\delta_2) \) is not actually split for all values of \( Z \). In order to obtain the corresponding condition on \( Z \), it is sufficient to study the quantity

\[
R(\delta_2 = w_1)/R(\delta_2 = 0)
\]

which is equal to:

\[
\left\{ (Z + 1)^2 + 1 \right\}^3/\left\{ 4(Z + 1) [(Z + 1)^4 + 1] \right\}
\]

for \( Z \) larger than \(-1\) and which is infinite for \( Z \) smaller than \(-1\). Therefore, if \( \omega_{10} \) is larger than \( \omega_{21} \) and if the two light beams travel in opposite directions, the Autler-Townes splitting should be observed with very good contrast in the strong field limit. On the contrary, if the two light beams travel in the same direction, high values of \( Z = k_1/k_2 \) are required for observing the splitting even in the strong field limit for the transition \( 0 \leftrightarrow 1 \). Thus, for \( k_1 \) close to \( k_2 \), one should observe a large power broadening without any splitting. These qualitative results are quite similar to those which have been obtained by Hansch and Toschek [10] in their theory of a three-level gas laser amplifier.

Furthermore, they are in good agreement with those which have been obtained by a numerical calculation of \( I(\delta_2, \delta_1) \) performed on the UNIVAC 1110 of the Centre d'Orsay de l'Université Paris XI. Some of those results for \( \delta_1 = 0 \) are shown on figures 4 and 5.

In the intermediate case (\( w_1^2 \sim \gamma_1^2, \gamma_2^2 \)), the Autler-Townes splitting will be easily observed only if the previous conditions on \( Z \) are fulfilled but this is not sufficient. Moreover, \( w_1^2 \) must be larger than a critical value \( w_{1c}^2 \) which a priori depends on \( Z, \gamma_1, \gamma_2 \). If \( Z \) is smaller than \(-1\) (light beams traveling in opposite directions, \( \omega_{10} > \omega_{21} \)), it appears that \( w_{1c}^2 \) is not strongly dependent on \( Z \) and indeed \( w_{1c}^2 \) is rather close to the corresponding value obtained in section 2 for a three-level system at rest.

![Fig. 4.](image-url)
5. Concluding remarks. — We have shown that a two-step resonance experiment using fluorescence as a detection signal should allow one to observe the Autler-Townes splitting under various conditions for rather low light powers. Evidently, all the situations that we have considered are strongly idealized. In particular, we have assumed that the spectral width of the two driving fields was negligible and we have ignored all the effects connected with level degeneracy and light polarization. However, the spectral width of singlemode tunable lasers is sufficiently small to be ignored in atomic experiments and the extension to degenerate systems is straightforward [10, 14] in the weak limit for the transition $1 \leftrightarrow 2$.

More serious are the difficulties related to power fluctuations (in time) of present tunable lasers. In fact, these difficulties were the same in a recent experiment based on the spectral analysis of the light spontaneously emitted from the upper level of a two-level system in interaction with a strong resonant field [15]. However, the Autler-Townes splitting has been observed in that experiment. Therefore, the observation of the various effects described here should be already possible.

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