OPTICAL PUMPING WITH LASERS

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Optical pumping is a method for transferring to an ensemble of atoms a fraction of the angular momentum carried by a beam of polarized resonance radiation ([1]). The observation of the characteristics of the light absorbed or reemitted by these atoms gives a lot of information about atomic structure, relaxation phenomena, interaction processes between atoms and photons ...

The light sources which have been used so far in optical pumping experiments are ordinary spectral lamps excited by RF discharges. The emitted light has in general a very broad spectral width, of the order of a few gigahertz. Even the brightest lamps have a relatively low intensity : this means that the pumping time, i.e., the mean time between 2 successive absorptions of pumping photons by the same atom, is much longer than the radiative lifetime of the excited state. In other words, the absorption and induced emission processes associated with the light beam are weak compared to spontaneous emission.

The spectacular development of lasers has now changed the situation. We have at our disposal light sources with very interesting characteristics : high intensity which permits one to easily saturate an atomic or molecular transition-tunability over large spectral ranges which gives the possibility of studying more levels-monochromaticity which makes it possible to get rid of the Doppler width for optical lines - pulsed operation which opens the way to a time resolved spectroscopy ...

I would like in this paper to analyse some of the new effects which can be observed in optical pumping experiments and which are

a consequence of the improvement of light sources. I will put emphasis on the theoretical problems and, more particularly, on the modification of the equations describing the optical pumping cycle. Some consequences of these modifications will be illustrated by a few examples, chosen as simple as possible.

More complete and more detailed treatments may be found in references ([2]), ([3]). This paper will be restricted to a simple presentation and discussion of some physical ideas.

OPTICAL PUMPING EQUATIONS

The central problem is to find, and eventually to solve, the equations which describe the evolution of the atomic density matrix. This matrix has the following form :

$$\begin{array}{|c|c|} \hline \sigma_e & \sigma_{eg} \\ \hline \sigma_{ge} & \sigma_g \\ \hline \end{array} \qquad (1)$$

σ_e and σ_g are the density matrices describing the ensemble of atoms inside the excited state e and the ground state g. Their diagonal elements are the populations of e and g sublevels, their off diagonal elements, the "hertzian" coherences between these sublevels. σ_{eg} and $\sigma_{ge} = \sigma_{eg}^{+}$ contain only off diagonal elements connecting a sublevel of g to a sublevel of e; they evolve at optical frequencies and are called for that reason "optical" coherences.

Let us give, as an example, the density matrix corresponding to a $J_g = 0 \leftrightarrow J_e = 1$ transition excited by a light beam having a σ linear polarization, i.e., perpendicular to the axis Oz of quantization. Only sublevels $m = \pm 1$ of the $J_e = 1$ excited state and sublevel $m = 0$ of the $J_g = 0$ ground state have to be considered (we can forget the $m = 0$ excited sublevel; see fig. 1), so that (1) takes the form :

$$\begin{array}{|cc|c|} \hline \sigma_{++} & \sigma_{+-} & \sigma_{+o} \\ \sigma_{-+} & \sigma_{--} & \sigma_{-o} \\ \hline \sigma_{o+} & \sigma_{o-} & \sigma_{oo} \\ \hline \end{array} \qquad (2)$$

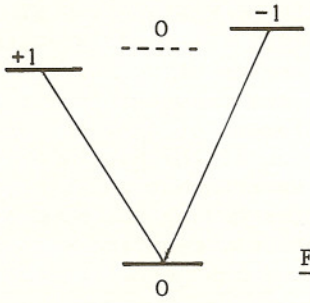

Figure 1

Generally, for an atomic vapour or for a gas, one has to introduce such a density matrix for each velocity group. However, under certain circumstances, it may happen, as we shall see later, that the internal state of an atom and its velocity are uncorrelated. In this case, σ refers to internal variables only. For an atomic beam perpendicular to the light beam, such a problem does not exist as the Doppler effect disappears.

The light beam is supposed to result from the superposition of parallel plane waves having all the same polarization $\vec{u}$, but different (complex) amplitudes $\mathcal{E}_\mu$ and frequencies ω_μ. The matrix describing the interaction between the atom and the light beam is purely non-diagonal and can be written (in the so-called "rotating-wave approximation") :

0	$-\sum_\mu D_{eg}\, \mathcal{E}_\mu\, e^{-i\omega_\mu t}$
$-\sum_\mu D_{ge}\, \mathcal{E}^*_\mu\, e^{i\omega_\mu t}$	0

(3)

We have put $D_{eg} = (\vec{u}.\vec{D})_{eg}$ where $\vec{D}$ is the electric dipole moment operator.

To simplify the discussion, we have treated the light beam as a classical field. Such an approach is not correct for describing the effect of spontaneous emission. This process may be taken into account by just adding to the equations giving the rate of variation of the components of σ the following terms (see references (4)(5)) :

$$\begin{cases} \dot{\sigma}_e = -\Gamma \sigma_e & (4\text{-a}) \\ \dot{\sigma}_{eg} = -\frac{\Gamma}{2} \sigma_{eg} & (4\text{-b}) \\ \dot{\sigma}_g = \mathscr{C}(\sigma_e) & (4\text{-c}) \end{cases}$$

where Γ is the natural width of the excited state (equal to the reciprocal of the radiative lifetime τ of this state). Equations (4-a) and (4-b) give the damping of σ_e and σ_{eg} by spontaneous emission while (4-c) describes the transfer from e to g associated with such a process. For each element of σ_g , the right hand side of (4-c) is a linear combination of the matrix elements of σ_e. Due to the spherical symmetry of spontaneous emission, this coupling appears only between quantities having the same symmetry (populations are coupled only to populations, coherences to coherences). In the example considered above, the last equation (5) can be written :

$$\dot{\sigma}_{oo} = \Gamma(\sigma_{++} + \sigma_{--}) \tag{5}$$

(σ_{oo} is not coupled to σ_{+-} and to σ_{-+}).

Finally the rate of variation of the components of σ, including the effect of the atomic Hamiltonian H_o (free evolution), the coupling with the incident light beam, and the spontaneous emission, is given by (we take $\hbar = 1$) :

$$\begin{cases} \dot{\sigma}_e = -i\left[H_o, \sigma_e\right] - \Gamma\sigma_e + i\sum_\mu \left[D_{eg}\sigma_{ge}\mathcal{E}_\mu e^{-i\omega_\mu t} - \sigma_{eg}D_{ge}\mathcal{E}_\mu^* e^{i\omega_\mu t}\right] & (6\text{-a}) \\ \dot{\sigma}_g = -i\left[H_o, \sigma_g\right] + \mathcal{C}(\sigma_e) + i\sum_\mu \left[D_{ge}\sigma_{eg}\mathcal{E}_\mu^* e^{i\omega_\mu t} - \sigma_{ge}D_{eg}\mathcal{E}_\mu e^{-i\omega_\mu t}\right] & (6\text{-b}) \\ \dot{\sigma}_{eg} = -i\left[H_o, \sigma_{eg}\right] - \frac{\Gamma}{2}\sigma_{eg} + i\sum_\mu \left[D_{eg}\sigma_{gg} - \sigma_{ee}D_{eg}\right]\mathcal{E}_\mu e^{-i\omega_\mu t} & (6\text{-c}) \end{cases}$$

Some algebraic manipulations can be done on equations (6). We can integrate equation (6-c) (and its hermitian conjugate) and insert the expression so obtained for σ_{eg} and σ_{ge} in the right hand side of (6-a) and (6-b). One gets in this way a system of 2 integro-differential equations involving only σ_e and σ_g. This is more easily done in interaction representation with respect to H_o. If we put :

$$\begin{cases} \tilde{\sigma}(t) = e^{iH_o t}\,\sigma(t)\,e^{-iH_o t} & (7\text{-a}) \\ \tilde{D}(t) = e^{iH_o t}\,D\,e^{-iH_o t} & (7\text{-b}) \\ \mathcal{E}(t) = \sum_\mu \mathcal{E}_\mu e^{-i\omega_\mu t} & (7\text{-c}) \end{cases}$$

we get after some simple calculations :

$$\begin{aligned} \dot{\tilde{\sigma}}_g(t) &= e^{iH_o t}\mathcal{C}(\sigma_e)\,e^{-iH_o t} \\ &\quad - \int_0^t dt' \left[\tilde{D}_{ge}(t)\tilde{D}_{eg}(t')e^{-\frac{\Gamma}{2}(t-t')}\mathcal{E}^*(t)\mathcal{E}(t')\tilde{\sigma}_g(t') + \text{hermit.conjug.}\right] \\ &\quad + \int_0^t dt' \left[\tilde{D}_{ge}(t)\tilde{\sigma}_e(t')\tilde{D}_{eg}(t')e^{-\frac{\Gamma}{2}(t-t')}\mathcal{E}^*(t)\mathcal{E}(t') + \text{hermit.conjug.}\right] \end{aligned} \tag{8}$$

plus a similar equation for $\dot{\tilde{\sigma}}_e$. The first integral in the right hand side of (8) involves only $\tilde{\sigma}_g$: it describes how the ground state is affected by the absorption process. The second integral, which involves only $\tilde{\sigma}_e$, describes the transfer from e to g by induced emission.

The problem is now to solve equations (6) or (8) which are equivalent. A first idea would be to get a solution of (6) in the form of a perturbation expansion in the field amplitudes $\mathcal{E}_\mu$. Reference ([6]) gives an example of such a calculation up to 4th order.

These types of calculations are quite laborious. I prefer here to discuss some situations where it is possible to by-pass perturbation treatments and to get non-perturbative solutions of equations (6) or (8). Such a possibility depends of course on the type of light beam which is used, i.e., on the properties of the amplitudes $\mathcal{E}_\mu$. I will consider first the case of an ordinary spectral lamp or of a free-running multimode laser for which it is more convenient to start from (8), and then the case of a single mode laser where equations (6) are simpler.

BROAD-LINE EXCITATION

Figure 2 shows the intensities $|\mathcal{E}_\mu|^2$ of the various waves forming the light beam

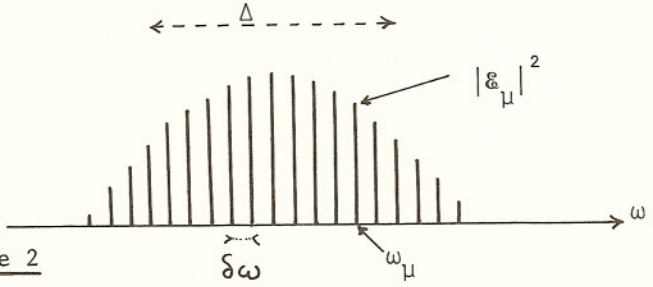

Figure 2

In the case of a spectral lamp, the frequencies ω_μ of these waves form a continuum. If we have a laser beam, we suppose that the laser oscillates on a great number of modes. In both cases, we will assume that the width Δ of the spectral interval covered by the frequencies ω_μ (see fig. 2) is very large compared to the Doppler width $\Delta\nu_D$ and the natural width Γ of the atomic line, and that the spacing $\delta\omega$ between 2 successive modes is small compared to Γ :

$$\begin{cases} \Delta >> \Delta\nu_D,\ \Gamma \\ \delta\omega < \Gamma \end{cases} \qquad (9)$$

In this case (see reference ([2])), the different "Bennett holes" burnt by the various modes in the Doppler profile overlap, and it

is easy to understand that the response of an atom does not depend on its velocity, so that σ refers to internal variables only.

The relative phases of the various modes are assumed to be random : we have a "free-running" multimode laser and not a "phase-locked" one. The instantaneous electric field $\mathcal{E}(t)$ of the light wave (see equation (7-c)) may be considered as a stationary random function. The correlation function $\mathcal{E}(t)\mathcal{E}^*(t-\tau)$ of $\mathcal{E}(t)$ only depends on τ and tends to zero when τ is larger than the correlation time τ_c which is of the order of $1/\Delta$.[19] When the ω_μ's form a discrete set of equidistant frequencies, there is another correlation time $\tau'_c = 1/\delta\omega$, which is much longer.

The strength of the coupling between the atom and the light wave may be characterized by a parameter v which is the product of the atomic electric dipole moment d by an electric field amplitude and which gives an order of magnitude of the evolution frequency of $\tilde{\sigma}$.

$$v = d.\left[\overline{|\mathcal{E}(t)|^2}\right]^{1/2} = d.\left[\sum_\mu |\mathcal{E}_\mu|^2\right]^{1/2} \qquad (10)$$

Let us come back to equation (8), and more precisely to the 2 integrals over t'. $\tilde{D}(t)\tilde{D}(t')e^{-\Gamma(t-t')/2}$ and $\mathcal{E}^*(t)\mathcal{E}(t')$ are the correlation functions of the atomic dipole moment and of the electric field. They have correlation times respectively equal to $1/\Gamma$ and to $1/\Delta$. The product of the 2 correlation functions has a memory which is determined by the shorter correlation time, i.e., by $1/\Delta$ (the other correlation time $\tau'_c = 1/\delta\omega$ of the electric field does not play any role as it is longer than $1/\Gamma$, according to condition (9)). One therefore expects that the contributions of $\tilde{\sigma}_g(t')$ [or $\tilde{\sigma}_e(t')$] with $t-t' \gg 1/\Delta$ will be cut down. This gives the key for a very convenient method for solving equations (8). If $1/v$, which is the characteristic time of evolution of $\tilde{\sigma}$ under the influence of the coupling with the light, is much longer than $1/\Delta$, i.e., if

$$v \ll \Delta \qquad (11)$$

we can replace, to a very good approximation, $\tilde{\sigma}_g(t')$ and $\tilde{\sigma}_e(t')$ by $\tilde{\sigma}_g(t)$ and $\tilde{\sigma}_e(t)$. We transform the system of integro-differential equations (8) into a set of differential equations, or "rate equations" describing the coupled evolutions of σ_g and σ_e. The coefficients γ appearing in these rate equations are given by the integral over t' of the product of the 2 correlation functions of $\tilde{D}$ and $\mathcal{E}$. As soon as $t \gg 1/\Delta$, the result of this integration, i.e., γ, is independent of t : this is due to the stationary character of the random function $\mathcal{E}(t)$. The order of magnitude of γ is found to be :

$$\gamma \sim v\,\frac{v}{\Delta} \qquad (12)$$

So, the evolution frequency of σ under the influence of the coupling with the light is reduced from the expected value, v, by a factor v/Δ much smaller than 1. This result may be compared with the motional narrowing appearing in the theory of relaxation in liquids and gases ([7]) : if the effect of the perturbation is weak during the correlation time, the net effect of this perturbation is reduced even more.

Due to (9), the value of γ does not depend on Γ or $\Delta\nu_D$. It does not depend also on the static magnetic field if we assume that the Zeeman splittings in e and g are weak compared to Δ. γ has in general an imaginary part which describes the so-called light shifts, i.e., the displacements of atomic energy levels produced by the light irradiation ([4]). I will assume here that the distribution of figure 2 is centered on the atomic frequency, in which case γ is real. The angular part of the coefficients appearing in the rate equations giving $\dot{\sigma}_e$ and $\dot{\sigma}_g$ are determined by the polarization $\vec{u}$ of the light and by the values J_e and J_g of the angular momenta. Let us give, as an example, these rate equations for the case considered above ($J_e = 1$, $J_g = 0$, $\vec{u}$ being a linear polarization perpendicular to the axis of quantization Oz). We assume in addition that a magnetic field is applied along Oz and we call ω_e the Larmor frequency in level e.

	Free evolution	Spontaneous emission	Absorption	Stimulated emission
$\dot{\sigma}_{++}$ =		$-\Gamma\sigma_{++}$	$+\gamma\sigma_{oo}$	$-\gamma\,(\sigma_{++}+\sigma_{--}-\sigma_{-+}-\sigma_{+-})/2$
$\dot{\sigma}_{--}$ =		$-\Gamma\sigma_{--}$	$+\gamma\sigma_{oo}$	$-\gamma\,(\sigma_{++}+\sigma_{--}-\sigma_{-+}-\sigma_{+-})/2$
$\dot{\sigma}_{-+}$ =	$2i\omega_e\sigma_{-+}$	$-\Gamma\sigma_{-+}$	$-\gamma\sigma_{oo}$	$+\gamma\,(\sigma_{++}+\sigma_{--}-2\sigma_{-+})/2$
$\dot{\sigma}_{oo}$ =		$+\Gamma(\sigma_{++}+\sigma_{--})$	$-2\gamma\sigma_{oo}$	$+\gamma(\sigma_{++}+\sigma_{--}-\sigma_{-+}-\sigma_{+-})$

(13)

Each vertical column contains the terms corresponding to a given process : free evolution (in this case, the Larmor precession), spontaneous emission, absorption and stimulated emission.

We see from equations (13) that the spontaneous and stimulated emission terms contain both the matrix elements of σ_e and in the first case Γ, the second one γ.

If $\gamma \ll \Gamma$, i.e., if according to (12)

$$v \ll \sqrt{\Gamma\Delta} \tag{14}$$

one can drop the stimulated emission terms which are negligible compared to the spontaneous emission ones (note that we must keep the absorption terms as they involve σ_g which may be much greater than σ_e). Omitting the last column of equations (13), we get the

usual optical pumping equations (see references ([4])) derived for thermal sources (γ is the inverse of the pumping time T_p).

Suppose now that γ is of the order of Γ, or larger than Γ (the condition of validity (11) being conserved) :

$$\sqrt{\Gamma\Delta} \lesssim v \ll \Delta \qquad (15)$$

In this case, we must keep the terms contained in the last column of (13). We will discuss later some of the new effects which come from these terms and which can be observed only if optical pumping is performed with a laser source. Let us already note that these terms are anisotropic, contrary to those of column 2. For example, they couple the Zeeman coherence σ_{-+} to the population σ_{oo}. This is due to the fact that the polarization $\vec{u}$ of the light is a preferential direction introduced by the interaction processes.

We will see later that the physical quantities calculated from equations (13) can be expanded in powers of γ, all powers of γ appearing in these expansions. This shows that the solutions obtained from (13) are non-perturbative.

It is clear however that these solutions do not correspond to a summation of all the terms of the perturbation series because an approximation has been done when replacing t' by t in some terms of equations (8). The physical meaning of this approximation is the following : the correlation time $1/\Delta$ of the light wave is so short that each interaction process may be considered as uncorrelated with the previous ones if (11) is satisfied. In other words, the rate equations (13) take into account the effect of an indefinite number of uncorrelated one-photon processes, but they neglect all the possible interferences between 2 successive interactions.

One can now ask the question of what happens if (11) is not satisfied, i.e., if $v \gtrsim \Delta$. We will come back to this problem after having discussed the case of a monochromatic excitation.

MONOCHROMATIC EXCITATION

We now consider a single mode laser of frequency ω irradiating an atomic beam perpendicular to it, so that there is no Doppler effect.

The correlation time of the perturbation is, in this case, very long and it becomes impossible to neglect the correlations between successive interactions of the atom with the light wave. We cannot consider that the atom undergoes, from time to time and without any phase memory, transitions from g to e or from e to g. We have

now a coherent oscillation between e and g, analogous to the Rabi nutation in magnetic resonance, and which proceeds at a frequency of the order of v [where v = d& is the coupling parameter analogous to (10)] . Furthermore, the optical coherence σ_{eg} becomes significant and oscillates at the same frequency, in quadrature with the populations of e and g. In the previous case, σ_{eg} was negligible as the coherence time $1/\Delta$ was too short for permitting σ_{eg} to build up appreciably. This was the real justification for trying to eliminate the optical coherences from equations (6) and replacing them by equations (8). It is now better to start directly from equations (6) which, since there is only one frequency $\omega_\mu = \omega$, look like Bloch's equations in magnetic resonance. The only difference is that the 2 levels e and g are not simple but have a structure.

A transformation analogous to the transformation to the rotating frame can be performed. If we put :

$$\sigma_{eg} = \rho_{eg}\, e^{-i\omega t} \tag{16}$$

we get a set of differential equations with time independent coefficients, coupling σ_e , σ_g , ρ_{eg} and $\rho_{ge} = \rho_{eg}^{+}$. Let us give these equations for the same transition and for the same polarization $\vec{u}$ as for equations (13) :

	Free evolution	Spontaneous emission	Coupling with the laser
$\dot{\sigma}_{++} =$		$-\Gamma\sigma_{++}$	$-iv(\rho_{o1} - \rho_{1o})$
$\dot{\sigma}_{--} =$		$-\Gamma\sigma_{--}$	$+iv(\rho_{o-1} - \rho_{-1o})$
$\dot{\sigma}_{-+} =$	$2i\omega_e\sigma_{-+}$	$-\Gamma\sigma_{-+}$	$+iv(\rho_{o1} + \rho_{-1o})$
$\dot{\sigma}_{oo} =$		$\Gamma(\sigma_{++}+\sigma_{--})$	$-iv(\rho_{1o}-\rho_{-1o}-\rho_{o1}+\rho_{o-1})$
$\dot{\rho}_{o1} =$	$-i(\omega-\omega_o-\omega_e)\rho_{o1}$	$-\frac{\Gamma}{2}\rho_{o1}$	$-iv(\sigma_{++} - \sigma_{oo} - \sigma_{-+})$
$\dot{\rho}_{o-1} =$	$-i(\omega-\omega_o+\omega_e)\rho_{o-1}$	$-\frac{\Gamma}{2}\rho_{o-1}$	$+iv(\sigma_{--} - \sigma_{oo} - \sigma_{+-})$

(17)

We have already defined the coupling parameter v. ω_0 is the e-g separation in zero magnetic field.

Here again, the solution of (17) corresponds to a summation of the perturbation series as all powers of v appear in the expressions calculated from (17).

Let us note first that it is also possible for a monochromatic excitation to get, in some cases, rate equations coupling only σ_e and σ_g. The correlation function of the atomic dipole moment

$\tilde{D}(t)\tilde{D}(t')\ e^{-\Gamma(t-t')/2}$, which appears in equation (8) and which has now the shortest correlation time (as $\Delta = 0$), cuts the contributions of $\tilde{\sigma}_g(t')$ [or $\tilde{\sigma}_e(t')$] when $t-t' >> 1/\Gamma$. If the nutation frequency ν is small compared to Γ, it is justified to replace, as before, t' by t in these quantities. One can say that spontaneous emission suppresses all phase memory between 2 successive interactions of the atom with the light beam. The rate equations so obtained are however less general than equations (17) which are valid even if $\nu >> \Gamma$.

Finally, we come back to the case of a broad line excitation so intense that $\nu >> \Delta >> \Gamma$. From the previous discussion, we see that the nutation frequency ν is so high that several nutations occur during the correlation time $1/\Delta$ of the light wave. It is therefore no longer possible to introduce rate equations coupling only σ_e and σ_g. Moreover one cannot consider that the light wave appears as a monochromatic excitation for the atom, since the amplitude of this wave fluctuates appreciably within the decay time $1/\Gamma$ of the dipole moment. We are in a difficult intermediate situation which presents some analogies with the problem of thermal relaxation when the motion narrowing condition is not satisfied.

ILLUSTRATION ON A VERY SIMPLE CASE : THE HANLE EFFECT OF A $J_g = 0 \leftrightarrow J_e = 1$ TRANSITION

In order to discuss some of the new effects which appear in optical pumping experiments performed with laser sources, we will consider the simplest possible transition $J_g = 0 \leftrightarrow J_e = 1$, and a resonance which does not require the use of any RF field, the Hanle zero-field level crossing resonance. We will suppose that the light beam, propagating along Oz, is linearly polarized along Ox, and that a magnetic field $\vec{B}_0$ is applied along Oz [situation considered for equations (13) and (17)]. One measures the variation versus B_0 of the total fluorescence light L_f, emitted along Oy with a linear polarization parallel to Ox. One can show that such detection signals are linear combinations of the matrix elements of σ_e. In the present case :

$$L_F \sim \sigma_{++} + \sigma_{--} - \sigma_{-+} - \sigma_{+-} \qquad (18)$$

In some experiments, where g is not the ground state, but the lower level of a pair of excited levels, the observation of the fluorescence light emitted from g, with for example a π-polarization gives a signal I_π proportional to the population σ_{oo} of g

$$I_\pi \sim \sigma_{oo} \qquad (19)$$

[In this case, equations (13) and (17) have to be slightly modified to introduce the rate of preparation of atoms in levels e

and g, the spontaneous decay of g, the spontaneous decay of e to levels other than g ([2]). But this does not modify the physical results .]

We will solve equations (13) or (17) according to the type of light irradiation which is considered. This will give us a quantitative expression for the 2 signals (18) and (19).

Let us first briefly recall what the situation is when an ordinary thermal source is used (broad line excitation with $\gamma \ll \Gamma$). Neglecting the terms in the last column of (13), we readily get the steady state solution of these equations. To lowest order in γ, this solution is :

$$\begin{cases} \sigma_{++} = \sigma_{--} = (N_o - \sigma_{oo})/2 = \gamma N_o/\Gamma & (20\text{-a}) \\ \sigma_{-+} = -\dfrac{\gamma N_o}{\Gamma - 2i\omega_e} & (20\text{-b}) \end{cases}$$

where $N_o = \sigma_{++} + \sigma_{--} + \sigma_{oo}$ is the total number of atoms [N_o is a constant of motion, as can be seen by adding the 2 first equations (13) to the last one] . We see that the Zeeman coherence σ_{-+} exhibits a resonant behaviour when the Larmor frequency ω_e is varied around 0, by sweeping the magnetic field B_o. This is the origin of the Hanle zero field level crossing resonance appearing on the fluorescence light (18) :

$$L_f = \frac{2\gamma N_o}{\Gamma} \left(1 + \frac{\Gamma^2}{\Gamma^2 + 4\omega_e^2} \right) \qquad (21)$$

and which has a Lorentzian shape and a width Γ independent of γ, i.e. of the light intensity. On the other hand, no resonances appear on the populations σ_{++} , σ_{--} , σ_{oo} which are independant of ω_e.

What are the modifications which appear when we use a much more intense broad-line source (for example, a free running multi-mode laser) ? We now have to keep the last column of equations (13). The calculations are a little more difficult, but it remains possible to get analytical expressions for the steady state solution of these equations.

We find for the populations :

$$\sigma_{++} = \sigma_{--} = (N_o - \sigma_{oo})/2 = \frac{\gamma N_o}{\Gamma + 3\gamma} \left[1 - S \frac{\Gamma'^2}{\Gamma'^2 + 4\omega_e^2} \right] \qquad (22)$$

where :

$$S = \frac{\gamma}{\Gamma+4\gamma} \qquad \Gamma' = \left[\frac{\Gamma(\Gamma+\gamma)(\Gamma+4\gamma)}{(\Gamma+3\gamma)}\right]^{1/2} \qquad (23)$$

The populations now exhibit a resonant behaviour near $\omega_e = 0$. The corresponding resonances are called "saturation resonances". They have a Lorentzian shape, a contrast S, and a width Γ' (see figure 3).

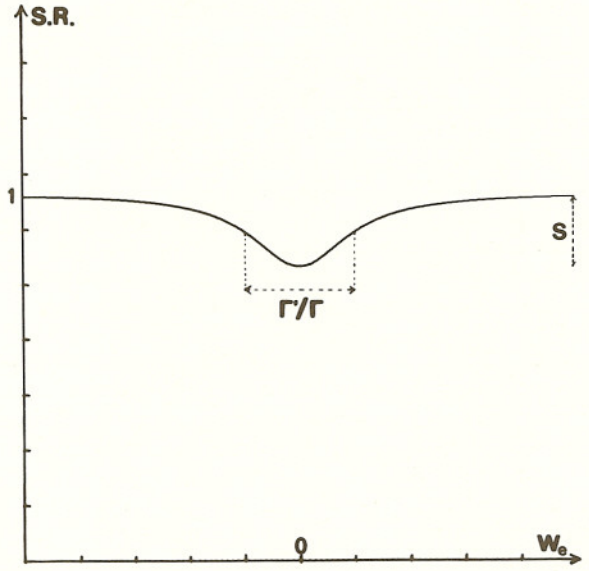

Figure 3 : Saturation resonance S.R. normalized to 1 in high field ($\omega_e >> \Gamma'$). S is the contrast of the resonance, Γ' its width. $W_e = 2\omega_e/\Gamma$ is a normalized Larmor frequency.

The saturation resonance appearing on σ_{oo} may be interpreted in the following way. A first interaction with the laser (absorption process) removes the atom from the ground state and puts it in a coherent superposition of the -1 and +1 sublevels of e (figure 4-a). The combined effect of Larmor precession and spontaneous emission gives rise to the well known resonant behaviour of the Zeeman coherence σ_{-+}. A second interaction with the laser (induced emission process) brings back the atom to the ground state (figure 4-b) and partially confers to the population σ_{oo} of this state the resonant behaviour of σ_{-+}. Such a process cannot occur for spontaneous emission which is an isotropic process and which, on the average, does not couple σ_{-+} to σ_{oo}.

Another way of interpreting the saturation resonance is to take the axis of quantization along the direction Ox of the laser polarization. In zero magnetic field, the energy levels may be taken as the eigenstates of J_x and the polarization of the laser is

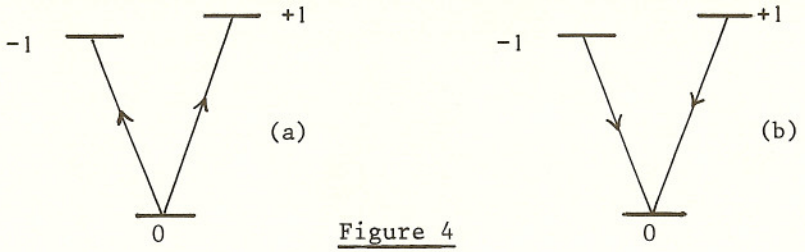

Figure 4

a π-polarization (figure 5). The $0 \leftrightarrow 0$ transition is saturated by the laser and the populations of these 2 sublevels tend to be equalized. The application of a magnetic field along Oz induces transitions between the 3 upper sublevels of figure 5. The corresponding changes in the populations of the m=0 upper sublevel are transferred to σ_{oo} through stimulated emission processes.

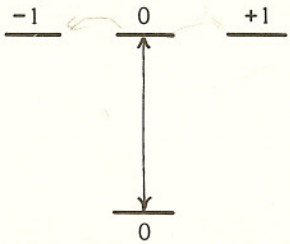

Figure 5

The variations with γ of the contrast S and the width Γ' of the saturation resonance are represented on figures 6 and 7 (C is the dimensionless quantity γ/Γ). If we put $\gamma_0 = \Gamma/4$, the expression (23) of S may be written for $\gamma < \gamma_0$:

$$S = \frac{1}{4} \sum_{p=0}^{\infty} (-1)^p \left(\frac{\gamma}{\gamma_0} \right)^{p+1} \qquad (24)$$

We obtain a perturbation expansion containing all orders of γ and which is not convergent for $\gamma > \gamma_0$. This clearly shows that the solution (22) of equations (13) is non perturbative. Γ', which is equal to Γ for $\gamma = 0$, increases linearly with γ for $\gamma \ll \Gamma$. This may be interpreted as a radiative broadening proportional to the laser intensity. For $\gamma \gg \Gamma$, Γ' increases only as $\sqrt{4\gamma\Gamma/3}$, i.e., as the amplitude of the light wave. This shows that some care must be taken when extracting atomic data from experimental results. Plotting the width Γ' of a saturation (or Hanle) resonance as a function of the laser intensity, and extrapolating linearly to zero light intensity, may lead to wrong results if the majority of experimental points do not fall in the linear range of figure 7 ($C = \gamma/\Gamma \ll 1$).

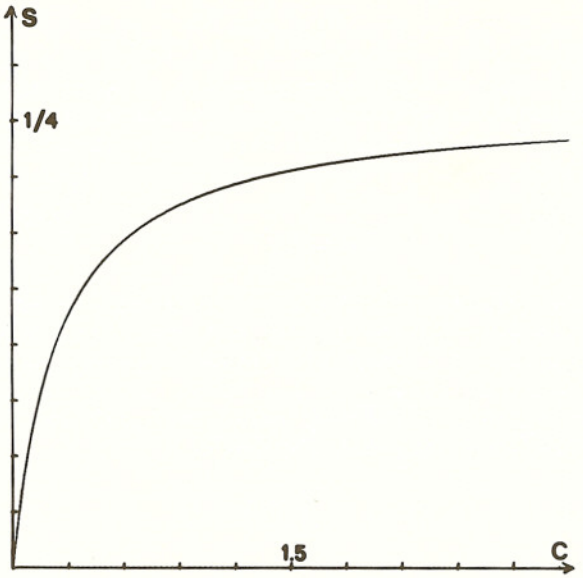

Figure 6 : Variations with $C = \gamma/\Gamma$ of the contrast S of the saturation resonance.

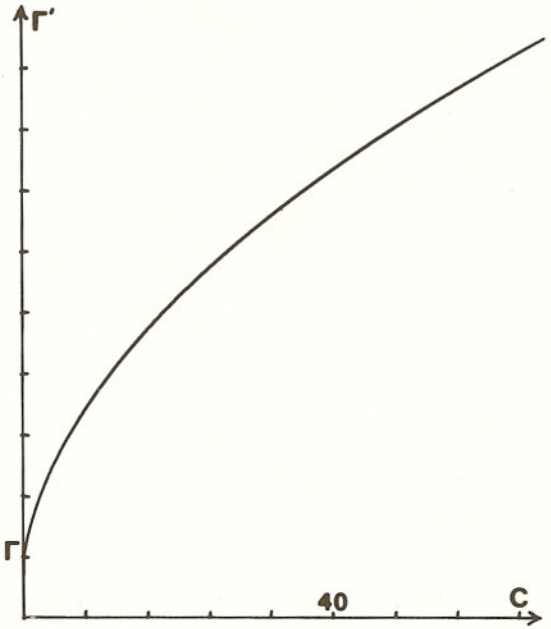

Figure 7 : Variations with $C = \gamma/\Gamma$ of the width Γ' of the saturation resonance (Γ' is also the width of the Hanle resonance). For $C \ll 1$, Γ' increases linearly with C. For $C \gg 1$, Γ' increases as $\sqrt{C}$.

The steady state solution for σ_{-+} may also be calculated from equations (13), and included with (22) in the expression (18) of L_f. We get for L_f :

$$L_f = \frac{2\gamma N_o}{\Gamma+3\gamma} + \frac{2\gamma N_o}{(\Gamma+3\gamma)(\Gamma+4\gamma)} \frac{(\Gamma+2\gamma)}{\Gamma'^2 + 4\omega_e^2} \frac{\Gamma'^2}{} \qquad (25)$$

i.e., the sum of a constant and of a Lorentzian curve having the same width Γ' as the saturation resonance. Figure 8 shows a set of Hanle curves corresponding to different values of the dimensionless parameter $C = \gamma/\Gamma$. One clearly sees the radiative broadening of the resonances. For large values of γ/Γ, the shape of the resonances does not change when γ increases, provided that the scale of the horizontal axis is contracted proportionally to $\sqrt{\gamma}$.

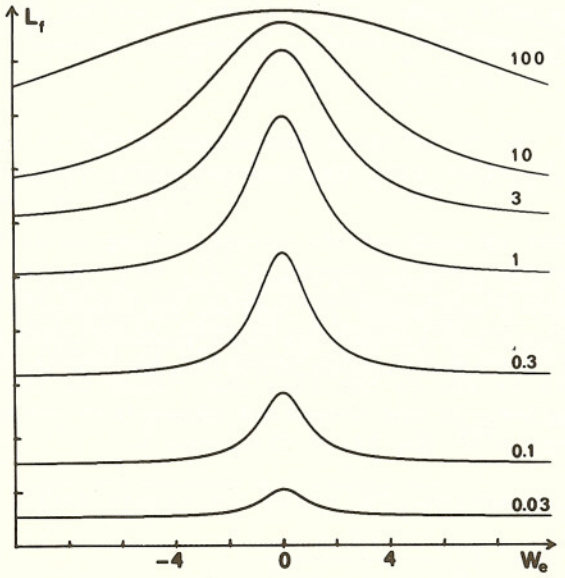

Figure 8 : Set of Hanle resonances detected on L_f . The excitation is broad-line. Each curve corresponds to a value of γ/Γ indicated on the figure. $W_e = 2\omega_e/\Gamma$ is a normalized Larmor frequency.

A detailed experimental verification of all the above results has been done on the $2s_2 \leftrightarrow 2p_1$ transition of Neon ($\lambda = 1.52\ \mu$) ([8]).

Let us summarize these new results which appear when the light source is a free running multimode laser : saturation resonances observable on the populations of the Zeeman sublevels, radiative broadening of these resonances (and also of the Hanle resonances), which is not a simple linear function of the laser intensity.

We consider now the case of an atomic beam irradiated perpendicularly by a single mode laser. We have therefore to use equations (17). We suppose that the illuminated portion of the beam is sufficiently long so that each atom reaches a steady state regime when passing through this zone. As before, $\sigma_{++} + \sigma_{--} + \sigma_{oo} = N_o$ is a constant of motion and represents the total number of atoms in the illuminated zone. To simplify the discussion, we will suppose that $\omega = \omega_0$, i.e., that the laser frequency ω is tuned at the center of the atomic line.

The steady state solution of equations (17) may be found in an analytical form after some simple algebra, and we get for the Hanle signal :

$$L_f = 16\ v^2\ N_o\ (\Gamma^2 + 4v^2)\ /\ D \qquad (26)$$

where :

$$D = 16\omega_e^4 + (8\Gamma^2 + 16\ v^2)\ \omega_e^2 + (\Gamma^2 + 4\ v^2)\ (\Gamma^2 + 16\ v^2) \qquad (27)$$

Figure 9 shows a set of such curves corresponding to different values of the dimensionless parameter $4v^2/\Gamma$. One sees clearly the radiative broadening of the resonance when the laser intensity, i.e., v^2, increases, but the shape is no more Lorentzian and the signal does not tend to a non zero value (as in figure 8) when ω_e is very large. This is due to the fact that, when ω_e increases, the frequencies $\omega_0 \pm \omega_e$ of the 2 optical lines $0 \leftrightarrow +1$ and $0 \leftrightarrow -1$ are out of resonance with the laser frequency ω.

When $v \to 0$, expression (26) takes the simple form :

$$L_f = 16\ v^2\ N_o\ \frac{\Gamma^2}{(4\omega_e^2 + \Gamma^2)^2} \qquad (28)$$

We find the square of a Lorentz curve ([20]) which is easy to understand: a first Lorentz denominator describes, as in the previous case, the decrease of the Zeeman coherence due to the Larmor precession, the second one comes from the Zeeman detuning of the 2 components of the optical line with respect to the laser frequency. Expression (28) may also be obtained from the Born amplitude for the resonant scattering ([9]). The initial state corresponds to the atom in the ground state in the presence of an impinging ω photon. The atom can absorb this photon and jump to one of the 2 excited sublevels

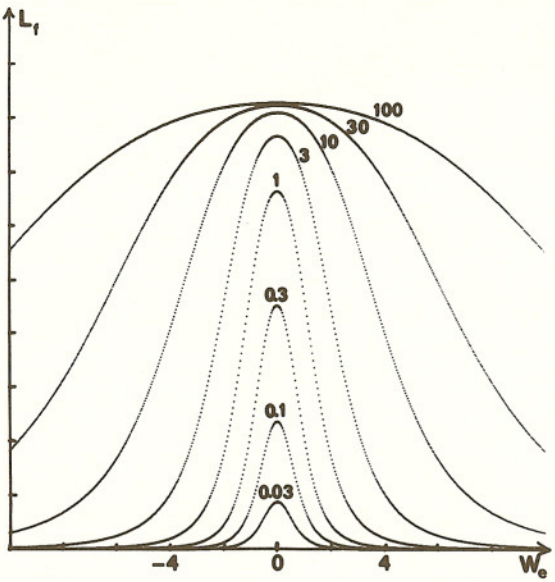

Figure 9 : Set of Hanle resonances detected on L_f. The excitation is monochromatic. Each curve corresponds to a value of $4v^2/\Gamma$ indicated on the figure. $W_e = 2\omega_e/\Gamma$.

± 1 of energies $\omega_0 \pm \omega_e$, and then fall back to the ground state by emitting the fluorescence photon. As there are 2 intermediate states for the scattering process, the scattering amplitude A is the sum of 2 terms which are, respectively, proportional to $1/(\omega-\omega_o-\omega_e + i\frac{\Gamma}{2})$ and to $1/(\omega-\omega_o+\omega_e + i\frac{\Gamma}{2})$. As we assume $\omega = \omega_o$, we get :

$$A = \frac{1}{-\omega_e+i\frac{\Gamma}{2}} + \frac{1}{\omega_e + i\frac{\Gamma}{2}} = -\frac{4i\Gamma}{4\omega_e^2 + \Gamma^2} \qquad (29)$$

The cross section is proportional to $|A|^2$ and has the same ω_e and Γ dependance as expression (28).

For $v^2 >> \Gamma$, the shape of the curve giving L_f does not change any more when v increases, provided that the scale of the horizontal axis is contracted proportionally to v.

Let us also study the variations with the magnetic field of the quantity $\sigma_{-+} + \sigma_{+-}$ (which may be experimentally observed by opposing the σ polarized fluorescence light emitted in 2 directions perpendicular to the magnetic field, and, respectively, parallel and

perpendicular to the laser polarization). One gets from equations (17) :

$$\sigma_{-+} + \sigma_{+-} = -8v^2 N_o \ (\Gamma^2 - 4\omega_e^2 + 4v^2) \ / \ D \qquad (30)$$

where D is given by (27). The shape of such signals is more complicated and is represented in figure 10 for different values of $4v^2/\Gamma$ (in the broad line case, the same signal consists of Lorentz curves tending to zero when $\omega_e \to \infty$). (21)

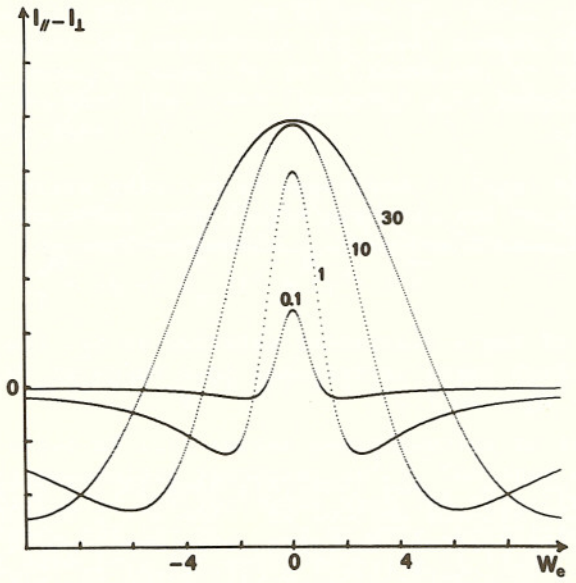

Figure 10 : Set of curves giving the difference between the σ-polarized light emitted in directions perpendicular to the magnetic field and, respectively, parallel and perpendicular to the laser polarization. Each number gives the corresponding value of $4v^2/\Gamma$. $W_e = 2\omega_e/\Gamma$.

We will not consider the variations of σ_{oo}. They result from the combination of different factors : coupling between σ_{-+} and σ_{oo} as in the broad line case, Zeeman detuning of the atomic lines.

To summarize, we see that the essentially new results obtained in the absence of Doppler effect (single mode laser and atomic beam) come from the Zeeman detuning of the atomic lines. The zero-field level crossing resonances have more complicated shapes (non-Lorentzian), but they still have a width which is of the order of Γ at low laser intensity and which increases with the laser intensity.

MORE COMPLICATED SITUATIONS

In the examples studied above, no structure was existing in level g. There was only one Zeeman coherence in level e, and the Hanle effect was only observable in this level.

We would like now to give an idea of what happens when Zeeman coherences and Hanle effects exist in both levels e and g, and, for that purpose, we take the simplest possible example of such a situation, the case of a transition $J_g = 1/2 \leftrightarrow J_e = 1/2$.

We restrict ourselves to a broad-line excitation. The light beam is supposed to be σ^+ polarized and to propagate along Oz, the magnetic field being applied along Ox. The relaxation of the ground state (which was absent in the previous case as $J_g = 0$) is supposed to be produced by the leakage of atoms from the cell through a small hole (the probability per unit time of escaping from the cell is $1/T$). A balance is provided by an entering flux of n_o atoms per unit time, all in the ground state and completely unpolarized. If the collisions with the inner walls of the cell are not disorienting, the relaxation time is simply T.

Relaxation / Larmor precession

$$
\begin{aligned}
\dot{\sigma}^e_{++} &= -\sigma^e_{++}/T + i\omega_e(\sigma^e_{+-}-\sigma^e_{-+})/2 \\
&\quad -\Gamma\sigma^e_{++} + 2\gamma\sigma^g_{--}/3 - 2\gamma\sigma^e_{++}/3 \\
\dot{\sigma}^e_{--} &= -\sigma^e_{--}/T - i\omega_e(\sigma^e_{+-}-\sigma^e_{-+})/2 \\
&\quad -\Gamma\sigma^e_{--} \\
\dot{\sigma}^e_{-+} &= -\sigma^e_{-+}/T - i\omega_e(\sigma^e_{++}-\sigma^e_{--})/2 \\
&\quad -\Gamma\sigma^e_{-+} - \gamma\sigma^e_{-+}/3 \\
\dot{\sigma}^g_{++} &= n_o/2-\sigma^g_{++}/T + i\omega_g(\sigma^g_{+-}-\sigma^g_{-+})/2 \\
&\quad +\Gamma\sigma^e_{++}/3+2\Gamma\sigma^e_{--}/3 \\
\dot{\sigma}^g_{--} &= n_o/2-\sigma^g_{--}/T - i\omega_g(\sigma^g_{+-}-\sigma^g_{-+})/2 \\
&\quad +\Gamma\sigma^e_{--}/3+2\Gamma\sigma^e_{++}/3 - 2\gamma\sigma^g_{--}/3 + 2\gamma\sigma^e_{++}/3 \\
\dot{\sigma}^g_{-+} &= -\sigma^g_{-+}/T - i\omega_g(\sigma^g_{++}-\sigma^g_{--})/2 \\
&\quad -\Gamma\sigma^e_{-+}/3 - \gamma\sigma^g_{-+}/3
\end{aligned}
\qquad (31)
$$

spontaneous emission / absorption / stimulated emission

Equations (31) are the rate equation for the various matrix elements σ^e_{++} , σ^e_{--} , σ^e_{-+}, and σ^g_{++} , σ^g_{--} , σ^g_{-+} of σ_e, and σ_g , and are derived using the same method as above. ω_g is the Larmor precession in g. As before, γ is the reciprocal of the pumping time. We note that the absorption and stimulated emission terms are simpler than in (13), whereas the Larmor precession terms are a little more complicated. This is due to the choice of the axis of quantization Oz which is parallel not to the magnetic field, but to the direction of propagation of the σ^+ polarized laser beam.

In a Hanle experiment performed on a $J = 1/2$ level, one detects components of the atomic orientation $\vec{J}$ perpendicular to the magnetic field $\vec{B}_o$. As $\vec{B}_o$ is along Ox, we are interested here in J^e_z (z component of the orientation of level e), i.e., in $\sigma^e_{++} - \sigma^e_{--}$ (one can for example measure the difference between the σ^+ and σ^- fluorescence light reemitted along Oz). To study these Hanle signals, we have to find the steady state solution of (31). Putting

$$\begin{cases} \Gamma_e = \Gamma + \dfrac{1}{T} \\ \Gamma_g = \dfrac{1}{T} \end{cases} \qquad \begin{cases} \Gamma'_e = \Gamma_e + \dfrac{\gamma}{3} \\ \Gamma'_g = \Gamma_g + \dfrac{\gamma}{3} \end{cases} \tag{32}$$

we get for $\sigma^e_{++} - \sigma^e_{--}$

$$\sigma^e_{++} - \sigma^e_{--} = \frac{n_o T}{2} \; \frac{\Gamma_e \Gamma'_e}{\omega_e^2 + \Gamma_e \Gamma'_e} \; \frac{1}{D} \tag{33}$$

where :

$$D = 1 + \frac{3\Gamma_e}{2\gamma} + \frac{1}{2} \frac{\Gamma_e \Gamma'_e}{\omega_e^2 + \Gamma_e \Gamma'_e} + \frac{1}{2} \frac{\Gamma_e}{\Gamma_g} \; \frac{\Gamma_g \Gamma'_g}{\omega_g^2 + \Gamma_g \Gamma'_g} + \\ + \frac{1}{3} \frac{\Gamma}{\Gamma_g} \; \frac{(\omega_e \omega_g - \Gamma'_e \Gamma'_g) \; \Gamma_e \Gamma_g}{(\omega_e^2 + \Gamma_e \Gamma'_e)(\omega_g^2 + \Gamma_g \Gamma'_g)} \tag{34}$$

Figures 11, 12, 13 show the variations with the magnetic field of the Hanle signal computed from (33). The various curves correspond to different values of the dimensionless parameter $\gamma/3\Gamma$, indicated on the figures. We have supposed that $\Gamma T = 100$, i.e., that the relaxation time T of the ground state is 100 times longer than the radiative lifetime $1/\Gamma$ of the excited state.

Let us first interpret the results at very low intensities ($\gamma \ll \Gamma$). We see on figure 11 that the Hanle signal appears as a superposition of 2 curves. The broad resonance is the Hanle effect of the excited state. It has a width equal to Γ (as $\gamma \ll \Gamma$, the radiative broadening is negligible). The amplitude of this resonance

increases proportionally to γ. The narrow resonance is the Hanle effect of the ground state which has a width determined by $1/T$ (similar resonances have been observed in the ground state of ^{87}Rb atoms optically pumped by a discharge lamp. Relaxation times as long as 1 second may be obtained, so that the width of the resonance may be as low as 10^{-6} gauss) ([10]). For the last curve of figure 11 ($\gamma/3\Gamma = 0.01$), γ is of the order of $1/T$, and the radiative broadening becomes visible. The intensity of the resonance increases as γ^2 and not as γ. This is due to the fact that we detect this resonance indirectly on the fluorescence light (and not on the absorbed light, as this is done usually). We need at least 2 interactions with the pumping beam in order to get the resonance : the first one, to create an atomic orientation in the ground state which gives rise to the Hanle effect of this state, the second one, to transfer this orientation to the excited state from which it is detected on the fluorescence light. This explains why the ground state resonance is so small for the first curve of figure 11 ($\gamma/3\Gamma = 0.001$).

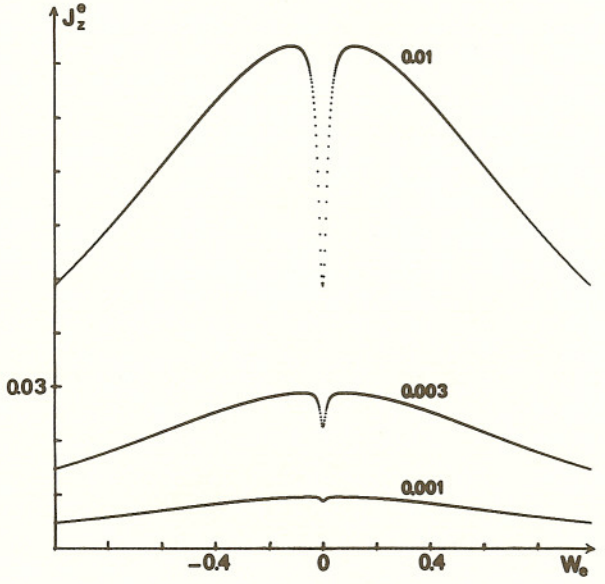

Figure 11 : Set of curves giving the Hanle effect of a $J_g = 1/2 \leftrightarrow J_e = 1/2$ transition. J_z^e is proportional to $\sigma_{++}^e - \sigma_{--}^e$ and is in arbitrary units. Each curve corresponds to a value of $\gamma/3\Gamma$ indicated on the figure. ΓT is equal to 100. To simplify, levels e and g are supposed to have the same Landé factor ($\omega_e = \omega_g$). $W_e = \omega_e/\Gamma$ is a dimensionless Larmor frequency.

When γ increases the width of the ground state resonance increases more rapidly than the width of the other one (figure 12).

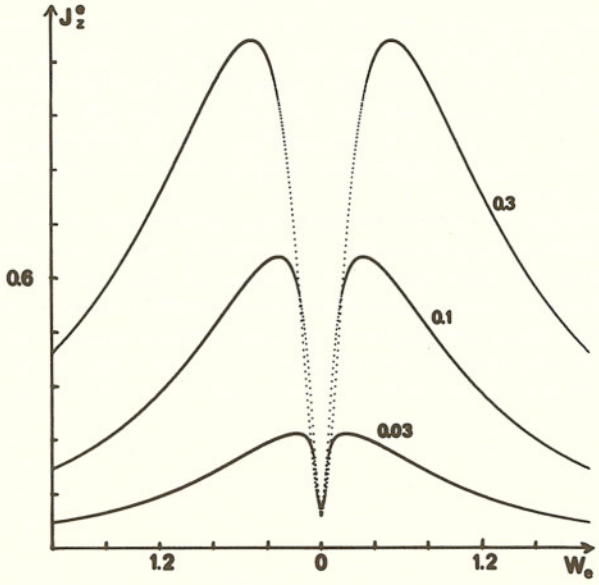

Figure 12 : Same signal as for figure 11 but for higher value of $\gamma/3\Gamma$. The horizontal and vertical scales have been changed.

For very large values of $\gamma/3\Gamma$ (see figure 13), we get the same result as for figures 8, 9, 10. The shape of the curve does not change any more if we contract the horizontal axis proportionally to $\sqrt{\gamma}$. The 2 Hanle effects of both states are completely mixed in a time short compared to $1/\Gamma$ (and of course to T). This mixing does not smooth out the structure apparent on figure 11. We get the superposition of 2 resonances with different widths and opposite signs giving rise to a curve with 2 maxima.

More complicated structures may be observed if the values of the angular momenta J_e and J_g are higher than 1/2. For example, in the case of a $J_e = 1 \leftrightarrow J_g = 2$ transition, and for a σ linearly polarized excitation, one can observe Hanle signals with 3 maxima. As in the previous example, the coupling between the 2 transverse alignments of e and g (perpendicular to the magnetic field) gives rise to a structure similar to that of figures 11, 12, 13. But as $J_g > 1$, there is also in the ground state g a "hexadecapole" moment (hertzian coherence $\sigma^g_{-2,+2}$) which can be induced in this state after 2 interactions with the laser, one absorption and one induced emission processes (see figure 14-a). A third interaction

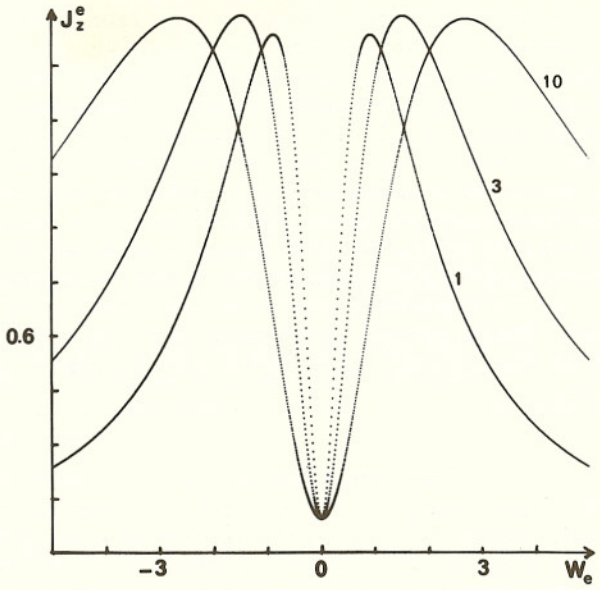

Figure 13 : Same signal as for figures 11 and 12, but for still higher values of $\gamma/3\Gamma$. The horizontal and vertical scales have been changed.

with the laser (absorption) can couple this hexadecapole moment to the transverse alignment of e ($\sigma^e_{-1,+1}$) (see figure 14-b). As the Hanle resonance associated to $\sigma^g_{-2,+2}$ has a smaller width (the resonant denominator is $\Gamma_g^2 + 16\omega_g^2$), the total result of these various couplings is to give for some values of γ a structure with 3 maxima. This effect has been observed on the $3s_2 \leftrightarrow 2p_4$ transition of Ne (λ = 6328 Å) and interpreted quantitatively ([11]). It would be interesting to see if other recent observations ([12]) could be explained in the same way.

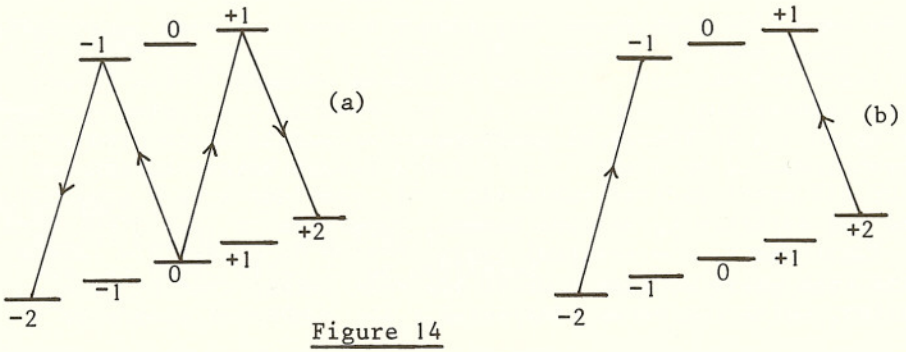

Figure 14

Optical pumping of molecules also provide several examples of Hanle resonances observed in levels having very high angular momentum ([13]). Some efforts are being done to write the optical pumping equations in a basis of quasiclassical states well adapted to the high values of J ([14]).

To summarize the results of this paragraph, we see that one can observe, on the fluorescence light emitted from e, level crossing resonances having a width much smaller than the natural width of e. This is not related to the broad-line or narrow-line character of the pumping light (as it appears already on the results of the previous paragraph). These narrow resonances must be attributed to the other state g of the optical line which has a longer lifetime or a higher J value.

OTHER PROBLEMS

In the previous paragraphs, we have discussed some new effects appearing in optical pumping experiments performed with lasers. This review is far from being complete. Let us just mention a few other interesting problems.

We have restricted ourselves to the study of the intensity of the total fluorescence light, a signal which is proportional to some observables of the excited state (see expression (18) for example). Another possibility would be to study the spectral distribution of this fluorescence light, which gives information on the correlation function of the atomic dipole moment driven by the incident light wave. The lowest order theory predicts that a monochromatic wave is scattered elastically by an atom moving perpendicularly to it (it is the same type of theory which leads to the Born expression (29) for the scattering amplitude). At high laser intensities non linear processes take place which of course conserve the total energy but change the spectrum of the scattered light. Experimental evidence for such effects has just been obtained (see ref. ([15]) and references in).

We have also considered only steady state processes corresponding to stationary light beams. This does not mean of course that short light pulses, such as those delivered by mode locked lasers, are not interesting. In particular, the observation of the quantum beats appearing in the light spontaneously emitted by an atom which has been prepared by a short light pulse in a coherent superposition of different excited sublevels ([16]), provides a very powerful method for determining various Zeeman, hyperfine or fine structures ([17]). Furthermore, the spectral range covered by pulsed dye lasers and the peak power of such sources are much larger than for c.w. operation, which increases considerably the number of atomic or

molecular transitions which can be optically pumped.

Let us finally mention a very interesting possibility, which has recently be demonstrated independently by several groups ([18]), of getting rid of the Doppler effect in 2 photon absorption processes. This possibility rests on the high intensity and monochromaticity of the light delivered by dye lasers. It opens the way to a high resolution study of atomic or molecular transitions connecting 2 levels with the same parity. This would be interesting for example for determining the absolute position of metastable levels.

([1]) Some review articles on optical pumping :
J. BROSSEL, Quantum Optics and Electronics
Les Houches Summer School, 1964, p. 187 (Gordon and Breach, New York, 1964)

C. COHEN-TANNOUDJI and A. KASTLER,
Progress in Optics (North Holland, Amsterdam, 1966), vol 5, p.1

A. KASTLER, Science 158, 214 (1967)

G.W. SERIES, Quantum Optics (edited by S.M. Kay and A. Maitland) p. 395, Academic Press, 1970

W. HAPPER, Rev. Mod. Phys. 44, 169 (1972)

([2]) M. DUCLOY, Phys. Rev. A 8, 1844 (1973)
9, 1319 (1974)

([3]) M. DUMONT, Thesis, University of Paris, 1971
(C.N.R.S., A.O. 5608), chapter III

([4]) J.P. BARRAT and C. COHEN-TANNOUDJI,
J. Phys. Rad. 22, 329 (1961)
22, 443 (1961)

C. COHEN-TANNOUDJI,
Ann. de Phys. 7, 423 (1962)
7, 469 (1962)

([5]) M. DUCLOY and M. DUMONT,
J. Phys. (Paris) 31, 419 (1970)

([6]) M. DUMONT,
J. Phys. (Paris) 33, 971 (1972)

([7]) A. ABRAGAM,
The Principles of Nuclear Magnetism, chapter VIII
Oxford University Press, 1961

([8]) M. DUCLOY,
Optics Commun. 3, 205 (1971)
Ann. de Phys. 1974, to be published

(9) C. COHEN-TANNOUDJI,
Cargese lectures in Physics, vol. 2, edited by M. Lévy (Gordon and Breach, New York, 1968)

(10) J. DUPONT-ROC, S. HAROCHE, and C. COHEN-TANNOUDJI,
Phys. Lett. 28A, 638 (1969)

(11) M. DUCLOY, M.P. GORZA, and B. DECOMPS,
Opt. Commun. 8, 21 (1973)

(12) W. GAWLIK, J. KOWALSKI, R. NEUMANN, F. TRÄGER,
Phys. Lett., to be published

(13) M. BROYER and J.C. LEHMANN,
Phys. Lett. 40A, 43 (1972)

(14) M. DUCLOY, To be published

(15) F. SCHUDA, C.R. STROUD Jr., and M. HERCHER,
J. Phys. B 7, L 198 (1974)

(16) A. CORNEY and G.W. SERIES,
Proc. Phys. Soc. 83, 207 (1964)

(17) S. HAROCHE, J.A. PAISNER and A.L. SCHAWLOW,
Phys. Rev. Lett. 30, 948 (1973)

(18) F. BIRABEN, B. CAGNAC, and G. GRYNBERG,
Phys. Rev. Lett. 32, 643 (1974)

M.D. LEVENSON and N. BLOEMBERGEN,
Phys. Rev. Lett. 32, 645 (1974)

T.W. HÄNSCH, K.C. HARVEY, G. MEISEL, and A.L. SCHAWLOW,
Opt. Commun. 11, 50 (1974)

(19) For coherence functions to represent light fields see:
J. DODD, G.W. SERIES, Proc. Roy.Soc. A 263, 353 (1961).
See also supplementary note published in the above paper reprinted in:
R.A. BERNHEIM,
Optical Pumping, Benjamin, New York 1965.
Later treatment: A. V. DURRANT, B. LANDHEER,
J. Phys. B 4 L 36 and 1200 (1971)

(20) G.W. SERIES,
Proc. Phys. Soc. 89, 1017 (1966)

(21) For recent experimental work done with monochromatic excitation, see: W. RASMUSSEN, R. SCHIEDER, and H. WALTHER,
Abstracts of 4th International Conference on Atomic Physics, Heidelberg 1974, p. 295