LIGHT SHIFTS AND MULTIPLE QUANTUM TRANSITIONS

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1. INTRODUCTION

In optical pumping experiments, atoms are irradiated with polarized resonance light. Kastler [1] has shown how it is then possible to transfer angular momentum from the light beam to the atoms and also to detect *optically* any change of the angular state of the atom resulting from RF transitions between Zeeman sublevels, collisions, etc.

We would like to discuss here how oriented atoms are perturbed when they interact with optical or RF photons. In particular we will show how atomic sublevels in the ground state can be shifted by irradiation with a non-resonant light beam (light-shifts). We will also study a few consequences of the interaction with the RF field (multiple quantum transitions, modification of the atomic Landé factor).

2. INTERACTION WITH OPTICAL PHOTONS. LIGHT SHIFTS

One of the most important results of the quantum theory of the optical pumping cycle [2] * is that atomic sublevels in the ground state are broadened and shifted by irradiation with optical photons. We will not enter here into the details of the theory; we will rather try to give a physical interpretation of these effects.

Let us first recall what happens to an atomic excited state as a consequence of the coupling with the electromagnetic field. There is a finite probability that the atom emits a photon k and falls from the excited state e to the ground state g (fig. 1a).

In such a "real" process, energy is conserved. We have (with $\hbar = c = 1$) $k = k_0 = E_e - E_g$.

As a result of this finite probability, the excited state has a finite "radiative" lifetime τ and, consequently, a natural width $\Gamma = 1/\tau$.

"Virtual" processes illustrated in fig. 1b can also occur where $k \neq k_0$

* A more recent treatment using the resolvent formalism and applicable to all the experiments discussed in this paper is presented in ref. [3].

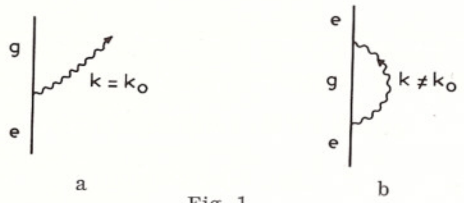


Fig. 1.

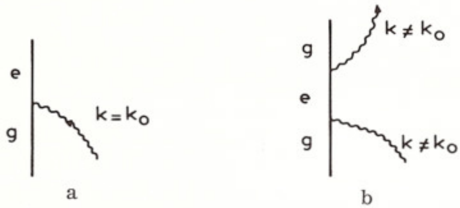


Fig. 2.

(energy is not conserved in the emission process). The emitted photon must then be reabsorbed by the atom after a time less than $1/|k - k_0|$. As a result of this "virtual" process, the wave function of the excited state is contaminated and the energy of this state modified. This is the origin of the "Lamb shift" ΔE of the excited state.

Figs. 2a and 2b illustrate real and virtual processes which are for absorption what figs. 1a and 1b are for spontaneous emission.

If the impinging photon is such that $k = k_0$, it can be "really" absorbed (energy is conserved). It results for the atom a finite probability to leave the ground state, which consequently acquires a finite lifetime T_p, and a width $\Gamma' = 1/T_p$. If $k \neq k_0$ the absorbed photon must be reemitted (fig. 2b). The effect of this "virtual process" is to perturb the wave function and the energy of the ground state. This is the origin of the light shift $\Delta E'$. Γ' and $\Delta E'$ are quantitatively given by

$$\tfrac{1}{2}\Gamma' + \mathrm{i}\Delta E' = \int_0^\infty |R|^2\, u(k)\, \frac{1}{\frac{1}{2}\Gamma - \mathrm{i}(k - k_0)}\, \mathrm{d}k \qquad (2.1)$$

where $u(k)$ is the spectral distribution of the impinging quanta, R the radial part of the matrix element of the electric dipole operator between e and g, Γ the natural width of the excited state. We see from (2.1) that to get Γ' and $\Delta E'$ we must multiply $u(k)$ by the atomic dispersion or absorption curve and integrate k from 0 to ∞ (figs. 3a, b).

The width Δ of $u(k)$ is assumed to be much larger than Γ. One sees that Γ' depends only on resonant photons (whose energy lies between $k_0 + \Gamma$ and $k_0 - \Gamma$) whereas $\Delta E'$ depend on all others. This confirms the physical interpretation given above. It is also clear from (2.1) that Γ' and $\Delta E'$ are proportional to the light intensity.

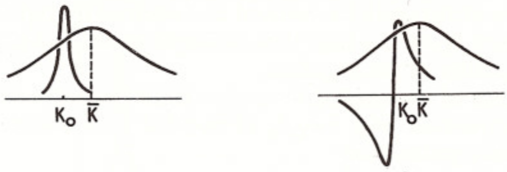


Fig. 3.

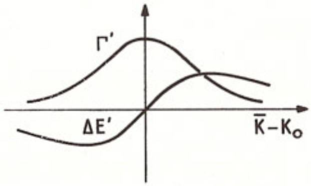


Fig. 4.

Fig. 4 shows how Γ' and $\Delta E'$ vary with $\bar{k} - k_0$ (where $\bar{k}$ is the abscissa of the "center" of $u(k)$).

$\Delta E'$ is maximum for a value of $\bar{k} - k_0$ which is of the order of Δ.

We have so far described the effect *on atoms* of the real and virtual processes of figs. 2a and 2b. The corresponding effects on *photons* are well known. In the real process (fig. 2a) the photon disappears: this is the "absorption" phenomena. In the virtual process 2b, during a very short time $1/(k - k_0)$, the photon is absorbed and does not propagate. This is the basis of "anomalous dispersion".

We have not yet taken into account the structure of the ground state. Let us call μ (and m) the Zeeman sublevels of g (and e). One can show that the effect of light irradiation on the ground state multiplicity is described by an effective Hamiltonian $\mathcal{H}_L$ given by

$$\mathcal{H}_L = (\Delta E' - \tfrac{1}{2}\mathrm{i}\,\Gamma')A \tag{2.2}$$

where $\Delta E'$ and Γ' are defined by (2.1) and A is a Hermitian matrix acting inside the ground state multiplicity

$$\langle \mu | A | \mu' \rangle = A_{\mu\mu'} = \sum_m \langle \mu | \boldsymbol{\varepsilon}^*_{\lambda_0} \cdot \boldsymbol{D} | m \rangle \langle m | \boldsymbol{\varepsilon}_{\lambda_0} \cdot \boldsymbol{D} | \mu' \rangle \tag{2.3}$$

where $\boldsymbol{\varepsilon}_{\lambda_0}$ is the polarization vector of the impinging quanta, $\boldsymbol{D}$ the angular part of the electric dipole operator.

Let $|\alpha\rangle$ and p_α be the eigenstates and eigenvalues of A (p_α is real).

$$A|\alpha\rangle = p_\alpha |\alpha\rangle\,. \tag{2.4}$$

It follows from (2.2) that the state $|\alpha\rangle$ has a width $p_\alpha\,\Gamma'$ and an energy shift $p_\alpha \Delta E'$. Therefore, if the p_α's are not all equal, the substates of the ground state are differently broadened and shifted by light irradiation. This anisotropy results from the fact that the light beam has a given polarization and a given direction of propagation.

In the presence of an applied magnetic field (parallel to Oz), there is also the Zeeman Hamiltonian $\mathcal{H}_Z = \omega_0 I_z$ (I angular momentum of the ground state; ω_0, Larmor pulsation).

$$\mathcal{H}_Z |\mu\rangle = \mu\omega_0 |\mu\rangle \tag{2.5}$$

so that the total Hamiltonian of the ground state is

$$\mathcal{H} = \mathcal{H}_L + \mathcal{H}_Z . \tag{2.6}$$

There are two extreme cases

a) $\mathcal{H}_L \gg \mathcal{H}_Z$.

The eigenstates of $\mathcal{H}$ are the $|\alpha\rangle$ states. If the p_α's are different, the Zeeman degeneracy is removed

b) $\mathcal{H}_Z \gg \mathcal{H}_L$.

$\mathcal{H}_L$ is a perturbation. The state $|\mu\rangle$ has an energy, which differs from $\mu\omega_0$ by the amount $(\Delta E' - \frac{1}{2}\mathrm{i}\Gamma')A_{\mu\mu}$.

We will now describe a few experiments which prove the existence of the $\Delta E'$ terms.

a) The experiment set up [4] is shown in fig. 5. A resonance cell containing ^{199}Hg atoms which have only two Zeeman sublevels in the ground state is placed in a static magnetic field H_0 parallel to Oz and modulated in amplitude by an RF field $H_1 \cos\omega t$. There are two light beams B_1 and B_2. In a first step, B_2 is off. B_1 is a pumping beam, coming from a lamp filled with the isotope ^{204}Hg the wavelength of which coincides with the $F = \frac{1}{2}$ hyperfine component of ^{199}Hg. Using the technique of *transverse* optical pumping in an amplitude modulated field [5] one gets resonance curves which permit to measure with great precision the Zeeman separation ω_0 between the two sublevels of the ground state of ^{199}Hg (curve on the left, fig. 6).

We then apply the B_2 light beam which is choosen in such a way that it produces a light shift as large as possible. B_2 comes from a ^{204}Hg lamp placed in an axial magnetic field H_A. The σ^+ component of this light (fig. 7a)

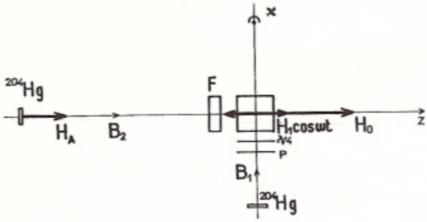


Fig. 5.

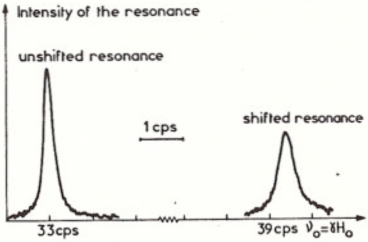


Fig. 6.

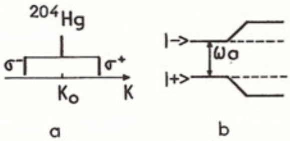


Fig. 7.

affects only the $\mu = -\frac{1}{2}$ sublevel (one easily shows that only $A_{-\frac{1}{2}-\frac{1}{2}}$ is non zero), and, as $\bar{k} - k_0 > 0$, shifts it upwards ($\Delta E'$ and $\bar{k} - k_0$ have the same sign. See fig. 4.)

In the same way, the σ^- component affects only the $+\frac{1}{2}$ sublevel and, as $\bar{k} - k_0 < 0$, shifts it downwards, so that the effect of the two components add (fig. 7b). The curve on the right of fig. 6 represents the magnetic resonance curve in the presence of the B_2 beam. One sees that there is a considerable light shift, much larger than the width of the curves. (In order to avoid any broadening due to *real* transitions, a filter F filled with ^{199}Hg is placed in front of the cell and suppresses all the resonant photons from the B_2 light beam; however this filter is not perfect and this explains why the shifted curve is slightly broader than the original one.)

b) We describe now an experiment performed in zero magnetic field [6]. The experimental set up is the same as in fig. 5 but now $H_0 = H_1 = 0$ (the resonance cell is placed in a triple magnetic shield).

Let us first show that the effect of the B_2 light beam is equivalent to that of a fictitious magnetic field $\boldsymbol{H}_{\mathrm{F}}$. The Hermitian matrix defined in (2.3) is, in the case of ^{199}Hg, a 2×2 matrix. So it can be expanded as

$$A = a_0 1 + \sum_{i=x,y,z} a_i \sigma_i \tag{2.7}$$

where 1 is the unit matrix and σ_i the Pauli matrices, which are proportional to I_x, I_y, I_z. The effective Hamiltonian $\Delta E' A$ ($\Gamma' = 0$) is then a linear combination of I_x, I_y, I_z, and, consequently, has the structure of a Zeeman

Hamiltonian in a "fictitious" magnetic field, $\boldsymbol{H}_F$, whose components are proportional to a_x, a_y, a_z. By symmetry considerations, one easily shows that $\boldsymbol{H}_F$ is parallel to the direction of propagation of B_2.

The experiment is done in the following way. In a first step, B_2 is off. The magnetic dipoles are oriented by the beam B_1 in the Ox direction (fig. 5). B_2 is then suddenly introduced. The dipoles start to precess around H_F, i.e. around Oz and this appears as a modulation of the transmitted B_1 light (fig. 8). As the shift is much larger than the width of the levels many oscillations occur during the lifetime of the ground state.

c) Fig. 9 represents the energy Zeeman diagram of ^{199}Hg in the presence of the B_2 light obtained by measuring the energy difference ω_0' between the two Zeeman sublevels as a function of the static field H_0 [6].

Two cases have been studied:

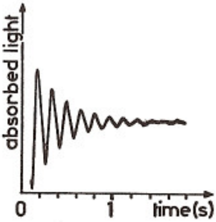
1) $\boldsymbol{H}_0 \parallel \boldsymbol{H}_F$.

In this case $\omega_0' = \omega_0 + \omega_F$ (ω_F is the Larmor pulsation associated with H_F). We get a displaced Zeeman diagram.

2) $\boldsymbol{H}_0 \perp \boldsymbol{H}_F$.

In this case $\omega_0' = \sqrt{\omega_0^2 + \omega_F^2}$. The two levels do not cross any more. (The curves of fig. 9 are theoretical; the points experimental.)

d) The following experiment [7] has been done on ^{201}Hg which has four Zeeman sublevels in the ground state ($I = \frac{3}{2}$). As in the three previous experiments, two light beams are used. The "shifting" B_2 beam is not polarized and propagates along the Oz direction. One can easily show that the



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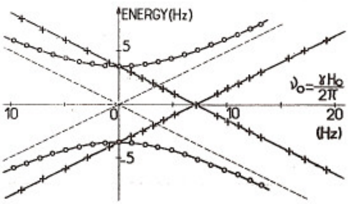

o $H_0 \perp H_F$

× $H_0 \parallel H_F$

Fig. 9.

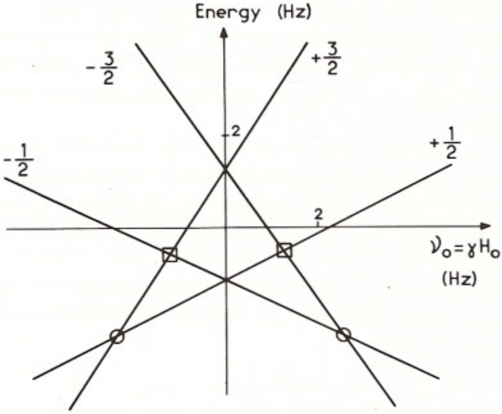

Fig. 10.

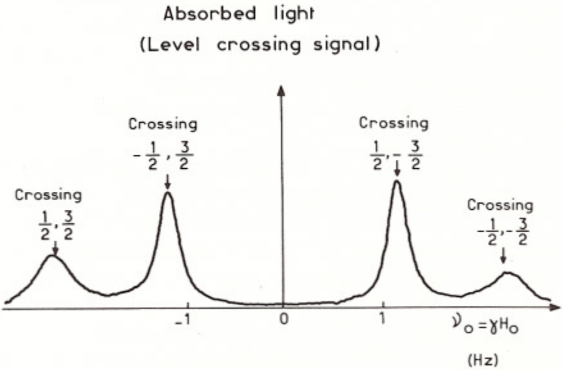

Fig. 11.

corresponding effective Hamiltonian $\mathcal{H}_L$ is $\mathcal{H}_L = \alpha I_z^2$ where α is a constant. If the static field H_0 is parallel to Oz, $\mathcal{H}_Z = \omega_0 I_z$ so that the total Hamiltonian in the ground state is $\mathcal{H} = \alpha I_z^2 + \omega_0 I_z$.

Fig. 10 represents the energy Zeeman diagram associated to $\mathcal{H}$.

There are two level crossings $\Delta\mu = 1$ (circles) and two level crossings $\Delta\mu = 2$ (squares) in this diagram *. They can be detected on the light absorbed on the B_1 light beam, if the direction of propagation of B_1 is perpendicular to Oz. This appears on fig. 11.

* The crossing $\mu = -\frac{1}{2}$, $\mu = +\frac{1}{2}$ ($\Delta\mu = 1$) is not observable for the experimental conditions corresponding to fig. 11.

3. INTERACTION WITH RF PHOTONS

In optical pumping experiments several types of magnetic resonances can be detected on the optical detection signals. We describe now some of these resonances and show how it is possible to explain them on the basis of a unified treatment.

a) *Longitudinal pumping*. Winter's multiple quantum transitions [8].

Suppose we have an atom with two levels in the ground state (for example ^{199}Hg), optically pumped by a σ^+ light beam B propagating along the direction of a static field H_0 (Longitudinal pumping), in the presence of a RF field $H_1 \cos \omega t$, perpendicular to H_0 (fig. 12).

Atoms are pumped to the $+\frac{1}{2}$ level. ω being fixed, we vary H_0, i.e. the energy difference ω_0 between the two levels. We observe several resonant

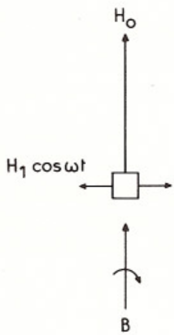

Fig. 12.

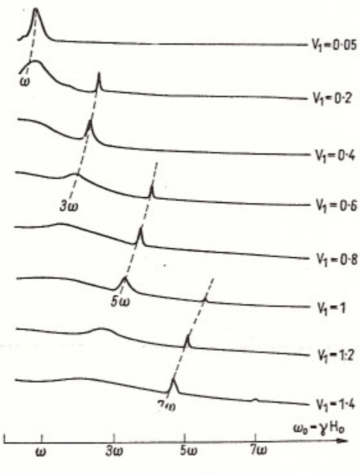

Fig. 13.

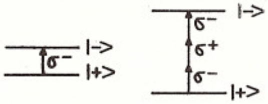

Fig. 14.

variations of the optical signal (for example of the fluorescent light) corresponding to transitions induced by the RF field between the two levels.

These resonances occur for $\omega_0 = (2n+1)\omega$. They are broadened and shifted when H_1 is increased (fig. 13). They correspond to the absorption of an odd number of RF quanta by the atom jumping from $m = +\frac{1}{2}$ to $m = -\frac{1}{2}$. During such a transition, $\Delta E = \omega_0$, $\Delta m = -1$, energy and angular momentum have to be conserved. As the linear RF field can be decomposed into two rotating fields, it contains σ^+ and σ^- RF photons which carry an angular momentum $+1$ and -1 with respect to Oz. So the transition $+\frac{1}{2} \rightarrow -\frac{1}{2}$ may occur by absorption of one σ^- RF quantum when $\omega_0 = \omega$ ($\Delta E = \omega_0$, $\Delta m = -1$), or by absorption of two σ^- and one σ^+ RF quanta when $\omega_0 = 3\omega$ ($\Delta E = 3\omega = \omega_0$, $\Delta m = -2 + 1 = -1$) and so on . . . (fig. 14).

b) *Transverse optical pumping.*

Suppose now that the light beam is perpendicular to H_0 (fig. 15) (transverse pumping). There is no more population differences between the two sublevels and the preceding resonances $\omega_0 = (2n+1)\omega$ are no more detectable.

In fact, we observe on the optical signal *new* resonances which form an "even" spectrum [9]. They occur for $\omega_0 = 2n\omega$. It is impossible to attribute these resonances to the absorption of an even number of RF quanta because the total angular momentum carried by an even number of σ^+ or σ^- quanta cannot be equal to -1 or $+1$, which is required for a transition between the two sublevels $+\frac{1}{2}$ and $-\frac{1}{2}$.

Fig. 16 shows for example the resonance $\omega_0 = 2\omega$ detected on the absorbed light (more exactly on the modulation at 2ω of this absorbed light). Each curve of fig. 16 corresponds to a different value of H_1 measured by the parameter V_1. One sees that the resonances are shifted but *not* broaden-

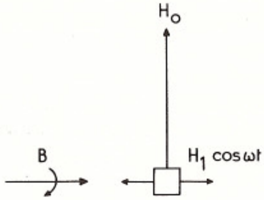

Fig. 15.

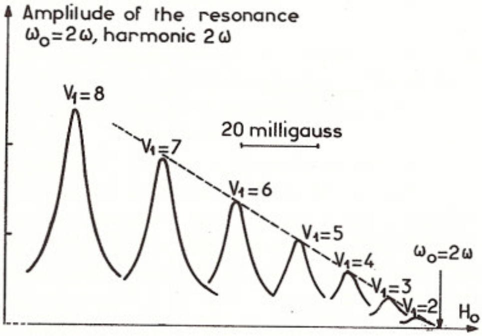

Fig. 16.

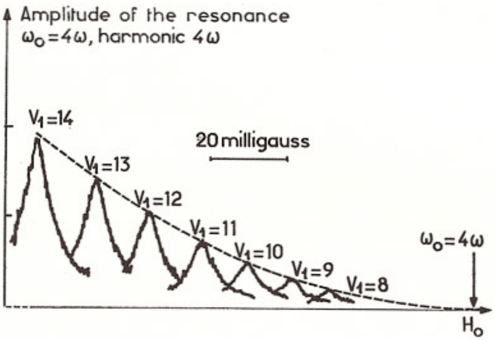

Fig. 17.

ed when H_1 is increased. Fig. 17 shows the same phenomena for the resonance $\omega_0 = 4\omega$.

The resonances observed in transverse optical pumping experiments * seem therefore to be much more difficult to understand in terms of RF quanta than Winter's resonances described in section a.

c) *Unified theoretical treatment for all these resonances* [3,10].

The idea is the following: It is well known that the light scattered by a free atom presents resonant variations when the static field is scanned

* Similar resonances can be observed when the RF field $H_1 \cos \omega t$ is parallel to H_0 instead of being perpendicular (the pumping is always transverse, the light beam is perpendicular to H_0). In this case the resonances occur for $\omega_0 = n\omega$ (full spectrum) [5]. They are not shifted and not broadened when H_1 is increased. As H_1 is parallel to H_0, the RF field contains only π photons which carry no angular momentum with respect to Oz and it is impossible to have any transition from $-\frac{1}{2}$ to $+\frac{1}{2}$ by absorption of such RF photons.

around values corresponding to "crossings" or "anticrossings" of energy levels of this atom *.

In the experiments described in sections a and b, we will consider that the light of the pumping beam is scattered not by the free atom (which we will also call the "bare" atom) but by the system atom plus RF quanta interacting together which we will also call the "dressed" (by RF photons) atom. The energy spectrum of the "dressed" atom is much more complex than the spectrum of the "bare" atom. It contains several crossing and anticrossing points. The light scattered by this "dressed" atom must therefore exhibit resonances when H_0 is scanned around these crossing and anticrossing points. One can explain in this way all the phenomena described in sections a and b.

The Hamiltonian $\mathcal{H}$ of the "dressed" atom can be written as

$$\mathcal{H} = \mathcal{H}_0 + h \tag{3.1}$$

where $\mathcal{H}_0$ represents the sum of the energies of the atom and of the RF photons, h the coupling between them. One has

$$\mathcal{H}_0 = \omega_0 I_z + \omega a^\dagger a \;. \tag{3.2}$$

$\omega_0 I_z$ is the Zeeman Hamiltonian of the atom; $\omega a^\dagger a$ the energy of the RF field oscillating on a given mode ($a^\dagger$ and a are the creation and annihilation operators of a photon of this mode).

In the dipole approximation, h is of the form

$$h = \lambda I_x (a + a^\dagger) \tag{3.3}$$

where λ is a coupling constant, I_x is the component of $\boldsymbol{I}$ on the direction of $\boldsymbol{H}_1$ so that

$$\mathcal{H} = \omega_0 I_z + \omega a^\dagger a + \lambda I_x (a + a^\dagger) \;. \tag{3.4}$$

We suppose $H_1 \ll H_0$ so that h is small compared to $\mathcal{H}_0$ (the number of RF quanta is not too large).

We have plotted on fig. 18 the energy levels of $\mathcal{H}_0$ as a function of ω_0.

$$\mathcal{H}_0 |n, \pm\rangle = (n\omega \pm \tfrac{1}{2}\omega_0) |n, \pm\rangle \;. \tag{3.5}$$

These levels are labelled by two quantum numbers, one for the RF field (n is the number of RF quanta), one for the atom. There are several crossing points for $\omega_0 = n\omega$. As $h \ll \mathcal{H}_0$, the effect of h is important only at these points.

h connects $|-, n\rangle$ to $|+, n-1\rangle$ and $|+, n+1\rangle$ (selection rule: $\Delta m = \pm 1$, $\Delta n = \pm 1$). At an odd crossing of $\mathcal{H}_0$, $\omega_0 = (2p+1)\omega$, the two levels of $\mathcal{H}_0$ which cross, $|-, n\rangle$ and $|+, n-2p-1\rangle$ can be connected by h via $2p$ intermediate states ($|+, n-1\rangle, \ldots |-, n-2p\rangle$). On the other hand, at an even crossing $\omega_0 = 2p\omega$, the two levels which cross, $|-, n\rangle$ and $|+, n-2p\rangle$, cannot be connected by h at any order (in this case, Δm and Δn have not the same parity). This is just a consequence of the conservation of angular

* For a simple discussion of "crossings" and "anticrossings" resonances, see ref. [3] p. 366.

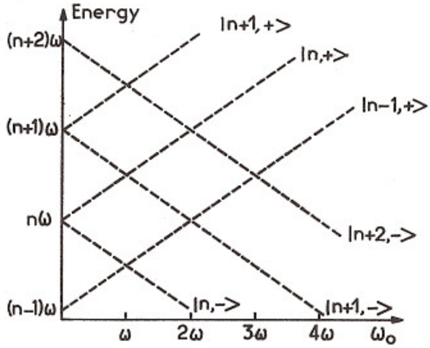

Fig. 18.

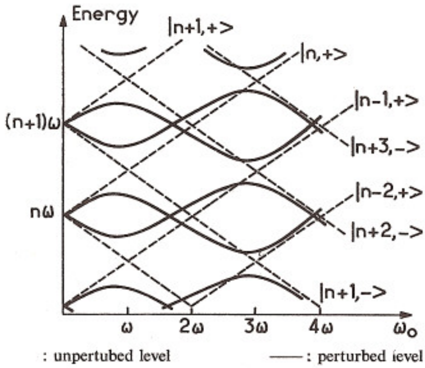

Fig. 19.

momentum. It is now easy to draw the energy diagram of $\mathcal{H}$ (fig. 19). All the odd crossings of $\mathcal{H}_0$ become anticrossings of $\mathcal{H}$ because of the coupling between the two levels of $\mathcal{H}_0$ which cross. The minimum distance between the two branches of the anticrossing $\omega_0 = (2p+1)\omega$ is proportional to $(\lambda\sqrt{n})^{2p+1}$ (order of the coupling), i.e. to $(H_1)^{2p+1}$ (H_1 is proportional to $\sqrt{n}$). On the other hand, all the even crossings of $\mathcal{H}_0$ remain crossings for $\mathcal{H}$ because of the absence of coupling between the two levels of $\mathcal{H}_0$ which cross. In fact, each crossing of $\mathcal{H}$ is shifted with respect to the corresponding crossing of $\mathcal{H}_0$; this is due to the coupling with other levels of $\mathcal{H}_0$ (2nd order perturbation term). The shift is proportional to $(\lambda\sqrt{n})^2$, i.e. to H_1^2. It exists also for each anticrossing of $\mathcal{H}$ (the center of each anticrossing is shifted with respect to the crossing of $\mathcal{H}_0$ from which it comes).

We see now that the resonances described in section a correspond to the anticrossings of $\mathcal{H}$; the resonances described in section b (figs. 16 and 17) to the crossings of $\mathcal{H}$ (one can say also that these last resonances are nothing but the "Franken" [11] effect of the "dressed" atom *). All the characteristics of these resonances (shift, width) can be quantitatively computed from the geometrical features of the crossings and anticrossings (centers, slope of the two levels which cross, minimum distance between the two levels which anticross ...). We will not enter here into the detail of these calculations. Let us just mention that the first resonances (section a) correspond to *real* transitions (induced by h between $|-, n\rangle$ and $|+, n-2p-1\rangle$, which corresponds to the jumping of the atom from $-\frac{1}{2}$ to $+\frac{1}{2}$ with absorption of $2p+1$ quanta); the second resonances (section b) correspond to the interference between two scattering amplitudes [12]; they are observable only because the wave functions of the two levels of $\mathcal{H}$, which cross at $\omega_0 = 2p\omega$, differ from those of $\mathcal{H}_0$ (contamination of the eigenstates of $\mathcal{H}_0$ due to the perturbation h) and do not correspond to a definite value of the number of RF quanta. This can be interpreted as being due to *virtual* absorption and reemission of RF quanta. Finally, one can also understand why these last resonances appear only in transverse pumping experiments: a necessary condition for the observation of the Franken effect is that the *polarization* of the incident light must be "coherent" (linear superposition of σ^+, σ^-, π). This is not realized for longitudinal pumping.

d) *Landé factor of the dressed atom* [13]

So far, we have supposed that H_1 (i.e. $\sqrt{n}$) is sufficiently small for h to be treated as a perturbation with respect to $\mathcal{H}_0$.

We can follow qualitatively what happens in the neighborhood of $\omega_0 = 0$ when H_1 is increased. The distance between the two branches of the first anticrossing $\omega_0 = \omega$ increases more and more. The first crossing $\omega_0 = 2\omega$ shifts more and more towards $\omega_0 = 0$. It follows that the slope of the two levels which cross at $\omega_0 = 0$ gets smaller and smaller and cancels when the shift of the first crossing attains the value 2ω (meeting of the two crossings $\omega_0 = 0$ and $\omega_0 = 2\omega$).

Quantitatively, one can treat $\omega_0 I_z$ as a perturbation with respect to $\omega a^\dagger a + \lambda I_x (a + a^\dagger)$. One finds easily in this way [13], that the slope g_i of the two levels which cross at $\omega_0 = 0$ varies as

$$g_i = g_0 J_0(\omega_1 / \omega) \tag{3.6}$$

where g_0 is the slope of the two levels of $\mathcal{H}_0$ (fig. 18), J_0 the zeroth order Bessel function, $\omega_1 = \gamma H_1$ (γ gyromagnetic ratio). This confirms the preceding qualitative discussion.

But g_i is nothing but the Landé factor of the "dressed" atom. One therefore sees that the coupling between the atom and the RF field modifies considerably the atomic Landé factor. g_i can even be cancelled for all the values of ω_1/ω corresponding to the zeros of J_0.

We have observed this effect experimentally by measuring the Hanle ef-

* More precisely, the "Franken" effect corresponds to a crossing between two sublevels of an excited atomic state. One can easily generalize to a crossing in the ground state by reasoning on the "*holes*" left in the ground state by optical excitation. See for example, ref. [14].

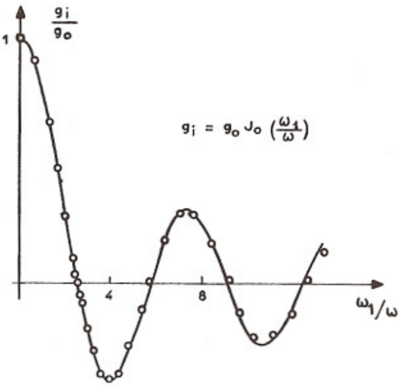

Fig. 20.

fect (zero field crossing experiment) in the presence of the RF field $H_1 \cos \omega t$. The width of the Hanle curve is inversely proportional to the slope of the crossing levels. This leads to a measurement of g_i. Fig. 20 shows the variation of g_i as a function of ω_1/ω. The results are in good agreement with the theoretical prediction (full curve of fig. 20).

This effect (modification of g) can also be interpreted as being due to virtual absorption and reemission of RF quanta by the atom (connection between eigenstates of $\mathcal{H}$ and $\mathcal{H}_0$). Finally we see that virtual absorption and reemission of RF photons can produce effects (light shifts and modification of g) which are parallel to two basic effects of quantum electrodynamics: Lamb-shift and anomalous magnetic moment of electron, that one can interpret in terms of virtual emissions and reabsorptions of photons.

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