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# The early days of quantum optics in France<sup>1</sup>

## Les débuts de l'optique quantique en France

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### Abstract

We review a few early studies on atom–photon interactions which took place in France after World War II and before the seventies. Several examples are discussed, including double resonance and optical pumping, radiofrequency multiphoton processes, quantum interference effects in radiative processes, master equation description of atom–photon interactions, optical resonators and laser media, etc. We try to point out the continuity which exists between these early studies and more recent investigations in new research fields, such as laser cooling and trapping. Some general conclusions about science policy will be finally drawn from such a historical review.

### Résumé

Nous présentons quelques-unes des études initiales relatives aux interactions atome–photons qui ont été réalisées en France après la Seconde Guerre mondiale et avant les années 1970. Nous donnons plusieurs exemples, tels que la double résonance et le pompage optique, les processus multiphotoniques dans le domaine des radiofréquences, les interférences quantiques dans les processus radiatifs, la description en termes d'une équation maîtresse des interactions atome–photon, des résonateurs optiques, des milieux laser, etc. Nous essayons de souligner la continuité qui existe entre ces travaux précoces et les recherches ultérieures menées dans des domaines nouveaux, tels que le refroidissement et le piégeage des atomes par laser. Enfin, quelques considérations générales sur la politique de la recherche scientifique se dégagent de cette revue historique.

### 1. Introduction

Quantum optics can be considered as a quantum theory of light and of its interactions with matter. To be more specific, let me focus on atom–photon interactions. This is a very broad field, so broad that we need a few guiding ideas to give an underlying frame to the discussion. Here are a few.

Basic conservations laws: they explain the exchanges of energy, linear and angular momentum between atoms and photons and they are at the root of important methods such as optical pumping and, more recently, laser cooling and trapping.

Higher-order effects: they appear when the field intensity is so high that the first-order perturbative treatment is no longer sufficient. This covers the whole field of multiphoton processes and nonlinear optics.

Linear superpositions of states: they are at the heart of quantum mechanics and they give rise to a wealth of quantum interference effects observable on the light emitted or absorbed by atoms. Linear superpositions of product states of two systems give rise also to entangled states which are so important in the new field of quantum information.

Master equations: they provide a quantum description of the evolution of the system. They can take different forms, depending on the coherent properties of the field: rate equations, optical Bloch

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equations. More recent developments include stochastic wave functions and Monte-Carlo simulations.

Laser development raised also several interesting questions, such as the design of good optical resonators or the identification of efficient laser media.

I have been asked to focus on the early contributions of France to quantum optics before the seventies, most often reported in short letters published in the *Comptes rendus de l'Académie des sciences*. This is not an easy job because Science is above all international and it may be artificial and even somewhat pretentious to focus on a specific scientific community. Nevertheless, I will try to do it and to point out the connections and the continuity between these early studies and more recent developments of the field such as cavity QED, laser cooling and trapping or matter waves. By looking at the evolution of a scientific community in a given place and for a long period of time, I think also that one can draw a few conclusions on science policy.

## 2. Conservation of angular momentum

### 2.1. Polarization selection rules

Let us consider the three simple states of polarization,  $\sigma_+$ -polarization corresponding to a right circular polarization around the  $z$ -axis,  $\sigma_-$ -polarization corresponding to a left circular polarization around the  $z$ -axis and  $\pi$ -polarization corresponding to a linear polarization parallel to the  $z$ -axis.

The polarization selection rules express that these three polarizations induce transitions between Zeeman sublevels  $m$  and  $m'$  such that

- $\sigma_+$ -polarization  $\leftrightarrow \Delta m = m' - m = +1$
- $\sigma_-$ -polarization  $\leftrightarrow \Delta m = m' - m = -1$
- $\pi$ -polarization  $\leftrightarrow \Delta m = m' - m = 0$ .

The interpretation of these selection rules is straightforward. They express that the angular momentum of the absorbed photon along the  $z$ -axis, which is equal to  $+\hbar$  for  $\sigma_+$  light,  $-\hbar$  for  $\sigma_-$  light, 0 for  $\pi$  light, is transferred to the atom when the atom absorbs the photon, so that the atomic magnetic quantum number  $m$  changes by  $+1$ ,  $-1$  or 0. This connection between polarization selection rules and conservation of angular momentum was pointed out by A. Rubinowicz, whose work inspired very much A. Kastler at the beginning of his investigation of resonance fluorescence. In fact, the basic idea is that, by exciting atoms with polarized light, one can prepare them in well-defined Zeeman sublevels. In other words, one can polarize atoms by light. The second important point is that, by detecting the light emitted or absorbed by an atom with a given polarization, one can determine the Zeeman sublevel occupied by the atom before the emission or absorption process. In other words, one can monitor the angular momentum of atoms by light.

Fig. 1. (a) Zeeman components of a transition  $0 \leftrightarrow 1$ . The excitation is  $\pi$ -polarized and one observes the  $\sigma_-$ -polarized reemitted light. (b) Variations of the  $\sigma_-$ -polarized reemitted light versus the magnetic field which splits the three excited Zeeman sublevels. The various curves correspond to different values of the amplitude of the RF field which induces resonant transitions between the excited Zeeman sublevels. The amplitude of the RF field increases by a factor 7 between the smallest curve and the largest one.

### 2.2. Double resonance

The idea of double resonance was suggested by A. Kastler and J. Brossel in 1949 [1]. Let us consider the simple case of a transition connecting a lower state with angular momentum 0 to an excited state with angular momentum 1 with three Zeeman sublevels  $-1$ ,  $0$ ,  $+1$  (Fig. 1a). If one excites such an atom with  $\pi$ -polarized light, one excites only the sublevel 0 of the excited state. Then, during the time spent by the atom in the excited state, one can perform magnetic resonance and induce resonant transitions between this sublevel 0 of the excited state and the other sublevels  $-1$  and  $+1$ . These

transitions are easily detected by looking at the light emitted with a  $\sigma_+$ - or a  $\sigma_-$ -polarization, or with a so-called  $\sigma$ -polarization, that is to say, a linear polarization perpendicular to the magnetic field (Fig. 1a). Figure 1b gives an example of experimental results which have been obtained by J. Brossel and F. Bitter on the even isotopes of mercury [2]. It represents the magnetic resonance curves in the excited state of the mercury atom detected by observing the  $\sigma$ -polarized fluorescence intensity as a function of the static magnetic field which produces the Zeeman splitting between the sublevels. These curves allow one to measure the magnetic moment of the excited state. By extrapolating the width of the curves at the limit of zero intensity of the RF field, one can also measure the natural width of the excited state.

### 2.3. Optical pumping

The double resonance method allowing the investigation of excited atomic states has been extended by A. Kastler to ground states, giving birth to the so-called optical pumping method [3]. To explain optical pumping, let us consider the simple case of a transition connecting a ground state with an angular momentum  $1/2$  to an excited state with an angular momentum  $1/2$ , so that there are two ground-state Zeeman sublevels  $g_{-1/2}$  and  $g_{+1/2}$ , and two excited Zeeman sublevels  $e_{-1/2}$  and  $e_{+1/2}$ . If one excites such an atom with  $\sigma_+$ -polarized light, one excites only the transition  $g_{-1/2}$  to  $e_{+1/2}$ , because this is the only transition with  $\Delta m = +1$ . Once the atom has been excited in  $e_{+1/2}$ , it can fall back by spontaneous emission either in  $g_{-1/2}$ , in which case it can repeat the same cycle, or in  $g_{+1/2}$  by emission of a  $\pi$ -polarized photon. In the last case., the atom remains trapped in  $g_{+1/2}$  because there is no  $\sigma_+$  transition starting from  $g_{+1/2}$ . This gives rise to what is called an optical pumping cycle transferring atoms from  $g_{-1/2}$  to  $g_{+1/2}$  through  $e_{+1/2}$ . How can one detect such a transfer? It clearly appears in Figure 2 that atoms absorb  $\sigma_+$  light only if they are in  $g_{-1/2}$ . If they are in  $g_{+1/2}$ , they cannot absorb  $\sigma_+$  light because there is no  $\sigma_+$  transition starting from  $g_{+1/2}$ . This means that any transfer from  $g_{+1/2}$  to  $g_{-1/2}$  which would be induced by a resonant RF field or by a relaxation process can be detected by monitoring the amount of light absorbed with  $\sigma_+$ -polarization.

Fig. 2. Principle of optical pumping for a  $1/2 \leftrightarrow 1/2$  transition. Atoms are transferred from  $g_{-1/2}$  to  $g_{+1/2}$  by an “optical pumping cycle”, which consists of an absorption of a  $\sigma_+$ -polarized photon followed by the spontaneous emission of a  $\pi$ -polarized photon.

### 2.4. Interesting features of these optical methods

A first interesting feature of these optical methods is that they provide a very efficient scheme for polarizing atoms at room temperatures and in low magnetic fields. Secondly, they have a very high sensitivity. The absorption of a single RF photon is detected by the subsequent optical absorption or emission of an optical photon and it is much easier to detect an optical photon than a RF photon because it has a much higher energy. It thus becomes possible to study very dilute systems where interactions are weak. The third significant point is that the Doppler effect of the RF transitions induced between Zeeman sublevels is negligible. In a sense, this can be considered as a Doppler free spectroscopy. At last, these optical methods allow one to study and to investigate non-equilibrium situations. Atoms are removed from their thermodynamic equilibrium and, by observing the temporal variations of the absorbed or emitted light, one can study how the system returns to equilibrium using what can be called a pump-probe spectroscopy.

### 2.5. More recent developments

Optical pumping methods still find interesting applications. For example, gaseous samples of optically pumped  $^3\text{He}$  atoms are used for studying the scattering of proton beams by polarized targets or for obtaining magnetic resonance images of the cavities of the human lung [4]. Another example is the field of spin polarized quantum systems where one studies the modifications of the transport properties of a spin polarized gas due to the postulate of symmetrisation which forbids certain collision channels [5].

### 3. Radiofrequency multiphoton resonances

The sensitivity of optical methods has allowed physicists in the beginning of the fifties to observe RF multiphoton resonances between Zeeman sublevels. This section is devoted to the description of a few of these experiments.

Fig. 3. The radiofrequency field  $\vec{b}$  has a component  $\vec{b}_{//}$ , parallel to the static field  $\vec{B}_0$ , which is associated with  $\pi$ -polarized photons with an angular momentum  $J_z = 0$ , and a component  $\vec{b}_{\perp}$ , perpendicular to  $\vec{B}_0$ , which is associated with  $\sigma$ -polarized photons with an angular momentum  $J_z = +\hbar$  or  $-\hbar$ .

#### 3. 1. Conservation Laws for a multiphoton process

**RF multiphoton resonances correspond** to transitions between two Zeeman sublevels  $m$  and  $m'$  by absorption of a number  $n$  of photons with  $n > 1$ . These transitions can be easily understood in terms of two conservation laws. The first one is the conservation of total energy,  $E_{m'} - E_m = n \hbar \omega$ , where  $\omega$  is the frequency of the RF field. The second one is the conservation of the total angular momentum  $(m' - m) \hbar = \Sigma$  angular momenta of the absorbed photons. In a given experiment, if one takes the static field  $\vec{B}_0$  aligned along the  $z$ -axis, one has to consider two components for the radiofrequency field  $\vec{b}$ :  $\vec{b}_{//}$  along  $\vec{B}_0$  and  $\vec{b}_{\perp}$  perpendicular to  $\vec{B}_0$  (Fig. 3). The  $\vec{b}_{//}$  component corresponds to the  $\pi$ -polarized photons with an angular momentum  $J_z = 0$  and the  $\vec{b}_{\perp}$  component to  $\sigma$ -polarized photons which have an angular momentum  $J_z = +\hbar$  or  $-\hbar$ .

#### 3.2. Two-photon transitions between unequally spaced Zeeman sublevels

Let us now consider the Zeeman hyperfine sublevels of an atom in a non-zero magnetic field. Because of the Back-Goudsmit decoupling, the Zeeman sublevels are generally not equidistant. Figure 4 pictures three such sublevels  $m_F$ ,  $m_F + 1$ ,  $m_F + 2$ . The two-photon transition between  $m_F$  and  $m_F + 2$  corresponds to a resonant process where the atom goes from  $m_F$  to  $m_F + 2$  by absorbing two  $\sigma_+$ -polarized RF photons. The conservation of total energy and angular momentum is clearly fulfilled if  $E(m_F + 2) - E(m_F) = 2 \hbar \nu_{RF}$ . It is clear also that such a higher-order process goes through a non-resonant intermediate state  $m_F + 1$ . Finally, Fig. 4 shows that  $\nu_{RF}$  is located between the resonant frequencies corresponding to a single  $\sigma_+$  photon transition between  $m_F$  and  $m_F + 1$  and between  $m_F + 1$  and  $m_F + 2$ . In fact, in the experiment, one keeps the frequency of the RF field fixed and the static field is scanned. In these conditions, the two-photon transition  $m_F \rightarrow m_F + 2$  appears between the two single-photon transitions  $m_F \rightarrow m_F + 1$  and  $m_F + 1 \rightarrow m_F + 2$ .

Fig. 4. Two-photon transition between unequally spaced Zeeman sublevels.

Fig. 5. Spectrum of multiphoton RF resonances observed in the ground state of optically pumped sodium atoms. The curves I, II, III correspond to increasing powers of the RF field. Resonances A, B, C, D correspond to single photon transitions; resonances a, b, c to two-photon transitions; resonances  $\alpha$ ,  $\beta$  to three-photon transitions (figure taken from reference [7]).

#### 3.3. Experimental observation with sodium atoms

Multiphoton resonances have been experimentally observed by A. Kastler, B. Cagnac, and J. Brossel in 1953 [6–8]. Let us consider, for example, the  $F = 2$  hyperfine levels of sodium with five Zeeman sublevels  $m_F = +2, +1, 0, -1, -2$ . The five Zeeman sublevels are not equidistant, thus giving rise to four single-photon transitions at four different values of the magnetic field (curve I of Fig. 5 corresponding to a weak RF power): resonance A corresponding to a transition between  $-2$  and  $-1$ , resonance B corresponding to a transition between  $-1$  and  $0$ , resonance C corresponding to a transition between  $0$  and  $+1$ , and resonance D corresponding to a transition between  $+1$  and  $+2$ . If the RF power of the field is increased (curve II of Fig. 5), three two-photon transitions  $\Delta m_F = 2$  appear: resonance a between  $-2$  and  $0$ , b between  $-1$  and  $+1$ , c between  $0$  and  $+2$ . At last, if the RF power is further

increased (curve III of Fig. 5), two three-photon transitions  $\alpha$  and  $\beta$  also appear: resonance  $\alpha$  between  $-2$  and  $+1$  and  $\beta$  between  $-1$  and  $2$ .

### 3.4. Multiphoton resonances at subharmonics of the Larmor frequency

Multiphoton resonances can also take place between two adjacent Zeeman sublevels  $m_F$  and  $m_F + 1$  with the absorption of one, two, three, four or more RF photons as represented in Fig 6. In that case, it clearly appears that the Larmor frequency which gives the energy splitting between two Zeeman sublevels is equal to an integer multiple of the frequency  $\nu_{RF}$  of the RF field (Fig. 6).

Fig. 6. Multiphoton RF resonances between two Zeeman sublevels  $m_F$  and  $m_F + 1$ . For a  $n$ -photon resonance, the frequency  $\nu_{RF}$  of the RF field is a subharmonic  $\nu_L/n$  of the Larmor frequency  $\nu_L$  which determines the splitting between the two Zeeman sublevels.

These resonances have been also observed in the ground state of sodium atoms. Figure 7 gives an example of such multiphoton resonances obtained by J. Margerie, J. Brossel, and J. Winter in 1955 [9,10]. As in Fig. 5, three curves are represented for three increasing values of the RF field. For low values, one can see only two-photon transitions whereas, for higher values of the RF amplitude, one can also see three and four-photon transitions. Note that the conservation of angular momentum requires that one of the two photons absorbed in the resonance  $\nu_L = 2 \nu_{RF}$  must be  $\pi$ -polarized, whereas the other one is  $\sigma_+$ -polarized. Similar arguments apply to the resonance  $\nu_L = 4 \nu_{RF}$  which must involve  $\pi$ -polarized RF photons as well as  $\sigma_{\pm}$  ones. Coming back to Fig. 3, we see that the RF field must have both parallel and perpendicular components with respect to the static field. Note finally that Fig. 7 clearly shows that these multiphoton resonances are broadened and shifted when the RF power is increased. The theory of this power broadening and power shift has been extensively developed by J. Winter [10].

Fig. 7. Spectrum of multiphoton RF resonances  $m_F \rightarrow m_F + 1$  observed in the ground state of optically pumped sodium atoms. The curves I, II, III correspond to increasing powers of the RF field (figure taken from ref. [19]).

### 3.5. Earlier works on two-photon processes

The first theoretical treatment of two-photon processes was given by M. Goppert Mayer in 1931. It seems also that V. Hughes and L. Grabner observed experimentally in 1950 two-photon processes while studying the RF spectrum of RbF molecules using a molecular beam electric resonance method [II]. They observed an unpredicted group of lines appearing at one half the frequency of another line group. One possible explanation for this unpredicted group of lines was the existence of two-photon transitions, but this was not clearly demonstrated.

### 3.6. Subsequent works on higher-order effects

The development of laser sources has made it possible to extend the observation of multiphoton processes to other frequency ranges, in particular to the optical range. I have selected some examples without attempting to be exhaustive.

Optical multiphoton processes involving multiphoton ionization and harmonic generation have been extensively studied by the Saclay group in France, in particular by C. Manus and G. Mainfray [12]. Another example that is worth mentioning is the new field of nonlinear optics which gave rise to a lot of investigations in several laboratories. Several French physicists were offered the opportunity to make a doctoral or postdoctoral stay in the laboratory of N. Bloembergen in Harvard: J. Ducuing, G. Bret, C. Flytzanis, N. Ostrowski, and P. Lallemand. I will mention also the work developed at the CNET ("Centre national d'études des télécommunications") involving molecular engineering for nonlinear optical materials. A third example of subsequent work involving multiphoton processes is Doppler free spectroscopy. Consider a two-photon transition where the two absorbed photons are propagating in opposite directions. The transfer of linear momentum from the field to the atom is then

equal to zero because the momenta of the two absorbed photons cancel out. As a consequence, there is no Doppler effect and no recoil. This new Doppler-free method has been investigated by several groups: the group of V. Chebotev in the Soviet Union [13], of B. Cagnac, G. Grynberg, and F. Biraben in France [14], of N. Bloembergen, M. Levenson [15], and of T. Hänsch [16] in the United States.

Finally, I will mention the possibility of using high-intensity laser pulses for producing laser inertial confinement fusion. In 1964, A. Kastler published a “Note aux: Comptes rendus de l’Académie des sciences” [17] where he mentioned the high temperature which could be obtained by concentrating the light from a high-power laser source on a target. This note stimulated other groups, in particular a French group at the CEA (“Commissariat à l’énergie atomique”) who investigated, in close collaboration with an industrial company (CGE-Marcoussis), the nuclear reactions of fusion produced by a nanosecond laser pulse [18]. This was achieved one year after the first demonstration of this effect in Soviet Union by N. G. Basov’s group with picosecond laser pulses [19]. Laser-induced fusion has now become a very broad field which is outside the scope of this paper.

#### **4. Quantum interference effects in radiative processes**

Later on, the development of optical methods of radiofrequency spectroscopy made it possible to observe new interesting quantum interference effects between different emission or absorption amplitudes in radiative processes.

##### *4.1 Coherent multiple scattering*

We describe first an experiment which played an important role in the investigation of these problems. In 1966, M.-A. Guiochon-Bouchiat, J.-E. Blamont, and J. Brossel observed a very intriguing effect represented in Fig. 8 [20,21]. This figure shows double resonance curves observed on mercury atoms, for increasing values of the temperature of the reservoir of atoms, i.e. for increasing values of the vapour density. The surprising result is that the width of the curves decreases when the number of atoms increases. In such a situation, one would normally expect a broadening of these curves: when the density increases, the number of collisions increases, producing a broadening of the levels. In fact, the narrowing of the curves was rapidly understood as being due to the following effect. When a mercury atom undergoes magnetic resonance in the excited state, it is put in a linear superposition of the three excited Zeeman sublevels. This atom emits then a photon which can be reabsorbed by another atom if the vapour pressure is high enough. It turns out that this second atom is excited in a linear superposition of its three excited Zeeman sublevels, which keeps a certain memory of the relative phases of the linear superposition in which the first atom was prepared before emitting his photon. In other words, the “Zeeman coherence” in the excited state which determines the width of the magnetic resonance curve is partially conserved in a multiple scattering process and thus appears to have a longer lifetime.

Fig. 8. Three sets of double resonance curves analogous to the set shown in Fig. 1b. These three sets are taken at different temperatures of the reservoir of atoms, increasing from the left to the right and indicated at the top of the figure. One clearly sees that the width of the curves decreases when the vapour pressure increases (refs. [20,21]).

##### *4.2. Linear superpositions of excited Zeeman sublevels*

The above-described experiment demonstrates the importance of linear superpositions of excited Zeeman sublevels, and the first question which can be asked is how they can be prepared. It is clear that a first possibility is the action of a RF field which transforms the state of an atom initially prepared in a well-defined Zeeman sublevel into a linear superposition including the other sublevels with which the initial state is coupled by the RF field. Another possibility for producing linear superpositions of excited Zeeman sublevels is to excite the atom with a light whose polarization is a linear superposition

of  $\sigma_+$ ,  $\sigma_-$ , and  $\pi$  polarizations, so that, starting from a well-defined ground-state sublevel, several excited Zeeman sublevels can be coherently excited by the incoming light field.

Another important question which can be asked is how do these linear superpositions of states show up in the emission process. The answer is that the light emitted by an atom in a linear superposition of excited Zeeman sublevels is not the same as the light emitted by a statistical mixture of these sublevels. Different quantum interference effects between different scattering amplitudes can be observed. Here are a few examples among others of such physical effects (for a review of the field, see, for example, ref. [22] and references therein).

- Coherent multiple scattering, which has been described in § 4.1.
- Hanle effect [23].
- Light beats such as those observed by the group of G. Series, revealing that the light emitted by an atom undergoing magnetic resonance in the excited state is modulated at the frequency of the RF field [24]. It looks like as if the various emission amplitudes were beating giving rise to modulations of the emitted light.
- Quantum beats effects resulting from a sudden excitation of the atom by a pulse of light. The pulse of light prepares the atom in a superposition of two excited Zeeman sublevels. The light subsequently emitted by the atom exhibits modulations resulting from an interference between the two emission amplitudes starting from the two excited Zeeman sublevels [25].
- Level crossing resonances which appear in the light emitted by the atom near the value of the field where two excited Zeeman sublevels cross [26].
- Detection of the first photon emitted in a radiative cascade which puts the atom in a certain linear superposition of the sublevels of the intermediate atomic state of the cascade.

#### 4.3. Master equation/or the atomic density matrix

To interpret the various quantum interference effects described above, a description of the atomic state in terms of the populations of the various energy sublevels is no longer sufficient. It is then necessary to use a density matrix description. The diagonal elements  $\sigma_{MM}$  of the density matrix  $\sigma$  give the populations of the energy sublevels, but  $\sigma$  has also off-diagonal elements, such as  $\sigma_{MM'}$ . These off-diagonal elements are very important because they describe the interference effects between the emission amplitudes starting from  $M$  and  $M'$ . It turned out subsequently that off-diagonal elements of the ground-state density matrix are also important for describing the quantum interference effects between different absorption amplitudes starting from two different ground-state sublevels.

It was therefore very important to establish the equation of the evolution of  $\sigma$ , the so-called master equation. Such master equations were derived first for an excited state [27], and then for the whole optical pumping cycle [28]. This work can be now considered as the precursor of the master equation description of modern quantum optics.

One should also mention that the Bloch equations of Nuclear Magnetic Resonance (see for example [29]) were a great source of inspiration for describing the evolution of an ensemble of atoms coupled with optical fields.

## 5. Light broadening and light shifts of ground-state sublevels

### 5.1. Theoretical predictions

One of the most important predictions of the master equation describing the evolution of the atomic density matrix during the optical pumping cycle is the fact that Zeeman atomic sublevels in the ground state are broadened and shifted by light. On the one hand, the light broadening  $\Gamma_{M'}$  of atomic sublevel  $g_M$  can be interpreted in terms of real absorptions of photons from the sublevel  $g_M$ , which shorten the lifetime of  $g_M$ . On the other hand, the light shift of  $g_M$ , denoted  $\Delta_{M'}$ , can be interpreted in terms of



virtual absorptions or reemissions of photons. The theory shows that  $\Gamma_{M'}$  and  $\Delta_{M'}$  are proportional to the light intensity and vary with the detuning  $\delta$  of the light excitation as Lorentz absorption and dispersion curves, respectively. There is a certain analogy between  $\Gamma_{M'}$  and  $\Delta_{M'}$ , on the one hand, and the natural width  $\Gamma_M$  and the Lamb shift  $\Delta_M$  of an excited sublevel  $e_M$ , on the other hand.  $\Gamma_{M'}$  and  $\Delta_{M'}$  can be considered as radiative corrections due to absorption and stimulated emission processes in the same way as  $\Gamma_M$  and  $\Delta_M$  can be interpreted in terms of radiative corrections due to the coupling with the empty modes of the radiation field. Another analogy can also be drawn between  $\Gamma_{M'}$  and  $\Delta_{M'}$ , on the one hand, and the imaginary and real part of the index of refraction of a light beam passing through the vapour, on the other hand.

Fig. 9. First experimental observation of light shifts on the transition  $1/2 \rightarrow 1/2$  of  $^{199}\text{Hg}$  (from ref. [30]). (a). For a  $\sigma_+$  excitation, only sublevel  $g_{-1/2}$  is light shifted, whereas for a  $\sigma_-$  excitation, only sublevel  $g_{+1/2}$  is light shifted. (b) The splitting between the two Zeeman sublevels is increased by a (blue detuned)  $\sigma_+$  excitation, whereas it is decreased by a blue detuned  $\sigma_-$  excitation. (c) Magnetic resonance signal vs magnetic field. The central curve (circles) is taken without the light-shifting beam. When this beam is introduced, either  $\sigma_+$ -polarized (crosses) or  $\sigma_-$ -polarized (triangles), the magnetic resonance curve is light shifted in opposite directions.

### 5.2 Experimental observation

Light shift and light broadening of ground-state sublevels have been observed in optical pumping experiments performed on the odd isotopes of mercury [30,31] (Fig. 9). These experiments done in 1961 were not using laser sources since these sources were not available at that time. In fact, light shifts could be easily observed because, in general, the two ground-state sublevels undergo different light shifts depending on the light polarization. Consequently, the splitting between two Zeeman sublevels is modified by the light irradiation. And, since the lifetime of the atomic ground state is very long, giving rise to very narrow magnetic resonance curves, it is possible to detect such a modification of the splitting between two Zeeman sublevels by a shift of the magnetic resonance curves. In fact, the first experimental observation of light shifts gave evidence of light shifts which were only of the order of one Hertz. These studies also demonstrated that the broadening  $\Gamma_{M'}$  and the light shift  $\Delta_{M'}$  of the ground state were varying with the detuning  $\delta$  of the light beam with respect to the atomic transition frequency as absorption and dispersion curves, respectively [31].

#### 3.5.3. Continuity with more recent works on radiative forces

About twenty years later, light shifts and light broadening of ground-state sublevels were found to play an important role in the physics of radiative forces [32]. There are in fact two great categories of radiative forces: dissipative forces, which are proportional to  $\Gamma$  and which are due to real absorption and spontaneous emission of photons; and reactive, or dipole forces, which are proportional to  $\Delta'$ , and which are due to virtual absorption and stimulated emission processes. A laser trap which can be achieved at the focus of a red detuned laser beam is a good illustrative example. Since the light shift  $\Delta'$  due to the laser beam is negative for red detuning and maximum in absolute value at the focus of the laser beam, a potential well is formed where sufficiently cold atoms can be trapped. Another example is the atomic mirror formed by a blue detuned evanescent wave produced at the surface of a piece of glass. For blue detuning, the light shift  $\Delta'$  is positive and decreases with the distance to the surface, producing a potential barrier upon which atoms can bounce if they are cold enough. Finally, one can mention one important laser cooling mechanism, called “Sisyphus cooling”, which is due to spatially modulated light shifts and optical pumping rates in a laser standing wave. A situation is achieved where a moving atom runs up potential hills more frequently than down, losing its kinetic energy (Fig. 10). This provides a very efficient cooling mechanism which allows one to reach the microkelvin range [33].

Fig. 10. Sisyphus cooling. In a laser standing wave, the intensity and the polarization of the laser electric field are spatially modulated, giving rise to correlated spatial modulations of the light shifts of two ground-state

sublevels and of the optical pumping rates between them. As a result of these correlations, a moving atom can run up potential hills more frequently than down.

#### *5.4. Other physical effects connected with off-diagonal elements of the ground-state density matrix*

The existence of Zeeman or hyperfine coherences in the atomic ground state manifests itself by several new physical effects which have been studied in detail. Let us mention a few of them.

- Zero-field-level crossing resonances which are the equivalent for the ground state of the Hanle effect in the excited state [34,35]. However, because the lifetime of the ground state is much longer than the lifetime of the excited state, these resonances are extremely narrow, with a width which can be as small as  $10^{-6}$  Gauss. Such resonances have been used to build magnetometers with a sensitivity of  $5 \cdot 10^{-10}$  Gauss [35].
- Modulation of the absorption of a crossed beam [31], which is the extension to atomic ground states of the light beats and quantum beats observed on the light emitted from excited states.
- One can also mention the discovery of coherent population trapping [36], which consists of the optical pumping of atoms in a linear superposition of two atomic states which no longer absorbs light (the so-called “dark states”). More recently, coherent population trapping has found new applications such as the achievement of subrecoil cooling by velocity selective dark states [37], self-induced transparency [38], and dramatic decrease of the light speed in an atomic medium. [39].

### **6. New investigations stimulated by the development of lasers in the sixties**

#### *6.1. Good optical resonators*

When lasers started to be built in the early 1960s, some physicists soon realized that the confocal Fabry-Perot, developed by P. Connes a few years before [40,41], was very useful. Such a scheme uses a cavity formed by two spherical mirrors having equal radii of curvature and coincident foci. It turned out to be a very convenient optical resonator and it played an important role in the development of laser sources.

Another example of development which occurred in the 1960s in France was the pulse compression scheme and the Gires–Tournois interferometer [42].

#### *6.2. Atoms inside a Fabry–Perot*

During the same period, in 1962, A. Kastler published a paper on the modification of the emission properties of atoms inside a Fabry–Perot [43]. He showed how, instead of having an isotropic diagram of emission, the emission of atoms should be concentrated in narrow rings corresponding to the modes of the Fabry–Perot. This paper can be considered as a precursor of cavity QED.

#### *6.3. Laser action in semiconductors*

Finally, let us mention another interesting development dealing with the possibility of having laser action in semiconductors. This question was first raised by Pierre Aigrain as early as 1958 in an unpublished lecture at a conference on solid-state physics in electronics and telecommunications in Brussels. A few years later, Bernard and Durauffourg published a paper where they gave the condition for laser action, a condition which is now known as “Bernard–Durauffourg condition”, connecting the differences between the quasi-Fermi energies of electrons (holes) in the conduction (valence) band to the energy of the emitted photons [44].

#### *6.4. More recent developments*

Laser sources introduced a spectacular improvement for spectroscopy and they stimulated the development of several new research fields. We mention here a few examples where French groups were very active.

- Investigation of forbidden atomic transitions and use of these transitions for getting evidence for parity violation in atomic physics. This research field was pioneered by M.-A. and C. Bouchiat in France [45].
- Laser spectroscopy of series of radioactive isotopes which can bring interesting information on nuclear structures. P. Jacquinot and S. Liberman, from the “Laboratoire Aimé-Cotton” in Orsay, were engaged in international collaborations at CERN (Geneva) for these problems [46].
- Development of the method of saturated absorption for high-resolution spectroscopy by C. Bordé [47].
- Spectroscopy of highly excited Rydberg states and investigation, by the group of S. Haroche, of the radiative properties of these states [48].
- Investigation, by the group of M. Ducloy, of atom–surface interactions by laser spectroscopy [49].

## 7. Interaction atom–single mode laser field

When lasers appeared in the sixties, one important theoretical problem which was raised was the description of an atom interacting with a single-mode coherent field. Two models were then developed in connection with this problem.

### 7.1. Jaynes–cummings model

In this model [50], a two-level atom is put in a single-mode cavity and interacts with a single-mode electromagnetic field. Formally, we have a spin  $\frac{1}{2}$  coupled with a one-dimensional harmonic oscillator. This model has proved to be extremely useful a few years later when experiments started to be achieved in what is called now cavity QED.

### 7.2. Dressed-atom approach. Atoms dressed by RF photons

Another model developed in the 1960s was the so-called dressed-atom approach where a two-level atom is interacting with a single-mode field in free space [51]. In this model, one considers a fictitious single-mode cavity containing  $N$  photons in a volume  $V$  (at the end of the calculation, one lets  $N$  and  $V$  tend to infinity with the ratio  $N/V$  maintained constant). This approach has provided a synthetic description and a better understanding of the various physical effects which can be observed in RF spectroscopy, in particular the shift and the broadening of multiphoton RF resonances. New effects were also predicted by this approach and observed experimentally, in particular the fact that the magnetic moment (or equivalently the  $g$ -factor) of an atom can be modified and even completely cancelled when this atom interacts with a high-frequency RF field [52]. This modification of the  $g$ -factor is somewhat analogous to the electron spin anomaly, which is actually a spontaneous radiative correction. An interesting question was then raised: is it possible to understand the spin anomaly  $g - 2$  as a stimulated radiative correction due to vacuum fluctuations, which could be obtained by summing the effects, calculated here for the coupling between a magnetic moment and a RF field, over all modes of the electromagnetic field (with an energy  $\hbar\omega/2$  per mode)? The difficulty is that, in this way, one gets the wrong sign for  $g - 2$ ! Understanding the origin of this minus sign stimulated several investigations and it turned out that the interpretation of  $g - 2$  requires one to consider, not only the effects of vacuum fluctuations, but also those of radiation reaction [53].

### 7.3. Subsequent developments. Atoms dressed by optical photons

A few years later, in the 1970s, the dressed-atom approach was extended to the optical domain where spontaneous emission plays an important role. This approach provided a very simple interpretation of many physical effects [51].

- Simple interpretation of the “Mollow resonance fluorescence triplet” in intense laser field in terms of transitions between dressed states.

- Simple interpretation of photon correlations and photon antibunching in single-atom resonance fluorescence in terms of a radiative cascade of the dressed atom.
- Simple calculation of the so-called “delay function” or “waiting time distribution” giving the distribution of the time intervals between two successive spontaneous jumps. This delay function has been extremely useful for developing Monte-Carlo simulations of the quantum evolution and for giving simple accounts of intermittent fluorescence and coherent population trapping.
- Interpretation of dipole forces in terms of spatial gradients of the dressed-state energies when the atom is moving in a spatially inhomogeneous laser field.

Let us finally point out that photon correlations have been extremely important for studying and achieving entangled states of photon pairs emitted in an atomic radiative cascade. In this respect, one can mention the important work done by A. Aspect in France for testing Bell’s inequalities [54–55].

## Conclusion

I have tried in this lecture to review a few early developments of quantum optics which took place in France before the 1970s. It seems clear that this research field has known a rather good start in this country and one can try to understand why. A first reason was probably the existence of a longstanding tradition in optics in France, going back to Fresnel and continuing with Fabry, Perot, and many others. A second important point was the role played by a few strong personalities who were able to attract good students and to build creative schools around them. This was the case of A. Kastler and J. Brossel at the “École normale supérieure” in Paris, of A. Abragam in Saclay, of P. Jacquinot in Bellevue and Orsay. I should also mention the great efforts made in France after World War II for introducing young generations to modern physics. I still remember the very stimulating lectures on quantum mechanics which were given every week by A. Messiah in Saclay. Les Houches summer school, which started in 1951, was also a unique place where every year about fifty young graduate or PhD students were spending two months following lectures given by the best physicists of the world. Finally, this period was a golden period for the expansion of research and it was relatively easy at that time to get long-term supports from the national research institutions like the “Centre national de la recherche scientifique”. Unfortunately, this combination of favourable circumstances is not always easy to achieve. More and more often, politicians now prefer to put the priority on specific short-term programs with the hope that scientific solutions will be rapidly found for the problems of health, energy, environment that we have to face. I think that it is not the good way. The history of science clearly shows that all important applications in physics such as lasers, transistors, magnetic resonance imaging, were not planned in advance. They came out after a high-quality basic research essentially motivated by the desire to understand the physical mechanisms. Research is a long-term investment for which perseverance and high quality standards are essential. I hope that an historical article like this one will help us to convey this message.

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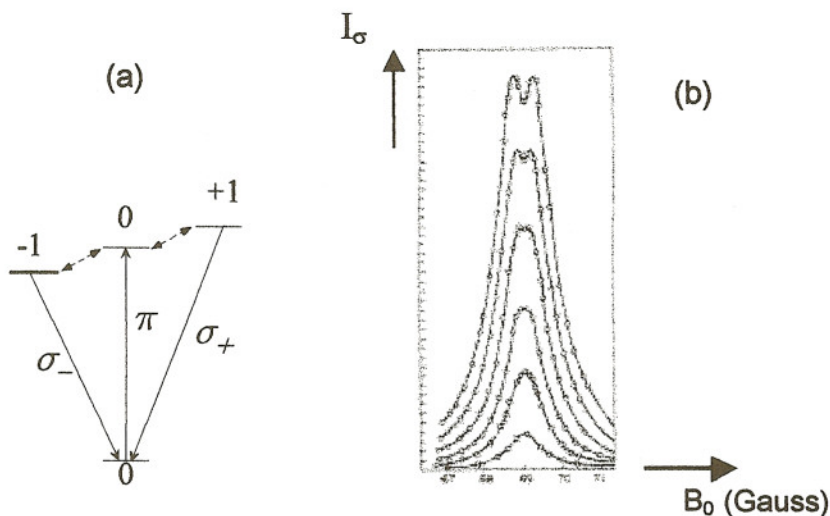


Fig. 1. (a) : Zeeman components of a transition  $0 \leftrightarrow 1$ . The excitation is  $\pi$ -polarized and one observes the  $\sigma$ -polarized reemitted light. (b) : Variations of the  $\sigma$ -polarized reemitted light versus the magnetic field which splits the 3 excited Zeeman sublevels. The various curves correspond to different values of the amplitude of the RF field which induces resonant transitions between the excited Zeeman sublevels. The amplitude of the RF field increases by a factor 7 between the smallest curve and the largest one.

## 2.2. Double resonance

The idea of double resonance was suggested by A. Kastler and J. Brossel in 1949 [1]. Let us consider the simple case of a transition connecting a lower state with angular momentum 0 to an excited state with angular momentum 1 with three Zeeman sublevels  $-1, 0, +1$  (Fig.1-a). If one excites such an atom with  $\pi$ -polarized light, one excites only the sublevel 0 of the excited state. Then, during the time spent by the atom in the excited state, one can perform magnetic resonance and induce resonant transitions between this sublevel 0 of the excited state and the other sublevels  $-1$  and  $+1$ . These transitions are easily detected by looking at the light emitted with a  $\sigma_+$  or a  $\sigma_-$  polarization, or with a so called  $\sigma$ -polarization, that is to say, a linear polarization perpendicular to the magnetic field (Fig.1-a). Figure 1-b gives an example of experimental results which have been obtained by J. Brossel and F. Bitter on the even isotopes of mercury [2]. It represents the magnetic resonance curves in the excited state of the mercury atom detected by observing the  $\sigma$ -polarized fluorescence intensity as a function of the static magnetic field which produces the Zeeman splitting between the sublevels. These curves allow one to measure the magnetic moment of the excited state. By extrapolating the width of the curves at the limit of zero intensity of the RF field, one can also measure the natural width of the excited state.

## 2.3. Optical pumping

The double resonance method allowing the investigation of excited atomic states has been extended by A. Kastler to ground states, giving birth to the so called optical pumping method [3]. To explain optical pumping, let us consider the simple case of a transition connecting a ground state with an angular momentum  $\frac{1}{2}$  to an excited state with an angular momentum  $\frac{1}{2}$ , so that there are two ground state Zeeman sublevels  $g_{-\frac{1}{2}}$  and  $g_{+\frac{1}{2}}$  and two excited Zeeman sublevels  $e_{-\frac{1}{2}}$  and  $e_{+\frac{1}{2}}$ . If one excites such an atom with  $\sigma_+$ -polarized light, one excites only the transition  $g_{-\frac{1}{2}}$  to  $e_{+\frac{1}{2}}$  because this is the only transition with  $\Delta m = +1$ . Once the atom has been excited in  $e_{+\frac{1}{2}}$ , it can fall back by



spontaneous emission either in  $g_{-1/2}$ , in which case it can repeat the same cycle, or in  $g_{+1/2}$  by emission of a  $\pi$ -polarized photon. In the last case, the atom remains trapped in  $g_{+1/2}$  because there is no  $\sigma_+$  transition starting from  $g_{+1/2}$ . This gives rise to what is called an optical pumping cycle transferring atoms from  $g_{-1/2}$  to  $g_{+1/2}$  through  $e_{+1/2}$ . How can one detect such a transfer? It clearly appears in figure 2 that atoms absorb  $\sigma_+$  light only if they are in  $g_{-1/2}$ . If they are in  $g_{+1/2}$ , they cannot absorb  $\sigma_+$  light because there is no  $\sigma_+$  transition starting from  $g_{+1/2}$ . This means that any transfer from  $g_{+1/2}$  to  $g_{-1/2}$  which would be induced by a resonant RF field or by a relaxation process can be detected by monitoring the amount of light absorbed with  $\sigma_+$ -polarization.

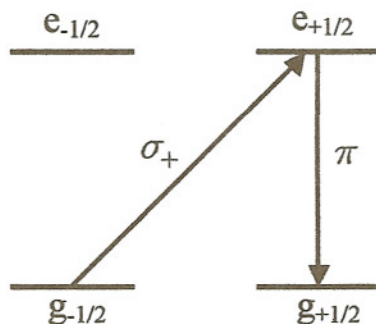


Fig. 2. Principle of optical pumping for a  $1/2 \leftrightarrow 1/2$  transition. Atoms are transferred from  $g_{-1/2}$  to  $g_{+1/2}$  by an "optical pumping cycle" which consists of an absorption of a  $\sigma_+$ -polarized photon followed by the spontaneous emission of a  $\pi$ -polarized photon

#### 2.4. Interesting features of these optical methods

A first interesting feature of these optical methods is that they provide a very efficient scheme for polarizing atoms at room temperatures and in low magnetic fields. Secondly, they have a very high sensitivity. The absorption of a single RF photon is detected by the subsequent optical absorption or emission of an optical photon and it is much easier to detect an optical photon than a RF photon because it has a much higher energy. It thus becomes possible to study very dilute systems where interactions are weak. The third significant point is that the Doppler effect of the RF transitions induced between Zeeman sublevels is negligible. In a sense, this can be considered as a Doppler free spectroscopy. At last, these optical methods allow one to study and to investigate non-equilibrium situations. Atoms are removed from their thermodynamic equilibrium and by observing the temporal variations of the absorbed or emitted light, one can study how the system returns to equilibrium using what can be called a pump-probe spectroscopy.

#### 2.5. More recent developments

Optical pumping methods still find interesting applications. For example, gaseous samples of optically pumped  $\text{He}_3$  atoms are used for studying the scattering of proton beams by polarized targets or for obtaining magnetic resonance images of the cavities of the human lung [4]. Another example is the field of spin polarized quantum systems where one studies the modifications of the transport properties of a spin polarized gas due to the postulate of symmetrisation which forbids certain collision channels [5].



### 3. Radiofrequency multiphoton resonances

The sensitivity of optical methods has allowed physicists in the beginning of the fifties to observe RF multiphoton resonances between Zeeman sublevels. This section is devoted to the description of a few of these experiments

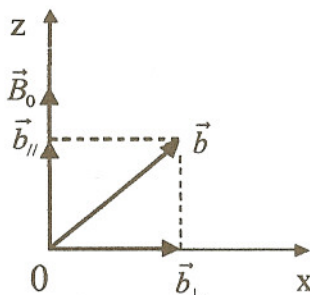


Fig. 3. The radiofrequency field  $\vec{b}$  has a component,  $\vec{b}_{\parallel}$ , parallel to the static field  $\vec{B}_0$ , which is associated with  $\pi$ -polarized photons with an angular momentum  $J_z = 0$ , and a component,  $\vec{b}_{\perp}$ , perpendicular to  $\vec{B}_0$ , which is associated with  $\sigma$ -polarized photons with an angular momentum  $J_z = +\hbar$  or  $-\hbar$ .

#### 3.1. Conservation laws for a multiphoton process

RF multiphoton resonances correspond to transitions between two Zeeman sublevels  $m$  and  $m'$  by absorption of a number  $n$  of photons with  $n > 1$ . These transitions can be easily understood in terms of two conservation laws. The first one is the conservation of total energy,  $E_{m'} - E_m = n\hbar\omega$ , where  $\omega$  is the frequency of the RF field. The second one is the conservation of the total angular momentum  $(m' - m)\hbar = \sum$  angular momenta of the absorbed photons. In a given experiment, if one takes the static field  $\vec{B}_0$  aligned along the  $z$ -axis, one has to consider two components for the radiofrequency field  $\vec{b}$ :  $\vec{b}_{\parallel}$  along  $\vec{B}_0$  and  $\vec{b}_{\perp}$  perpendicular to  $\vec{B}_0$  (Fig.3). The  $\vec{b}_{\parallel}$  component corresponds to the  $\pi$ -polarized photons with an angular momentum  $J_z = 0$  and the  $\vec{b}_{\perp}$  component to  $\sigma$ -polarized photons which have an angular momentum  $J_z = +\hbar$  or  $-\hbar$ .

#### 3.2. Two-photon transitions between unequally spaced Zeeman sublevels

Let us now consider the Zeeman hyperfine sublevels of an atom in a non-zero magnetic field. Because of the Back-Goudsmit decoupling, the Zeeman sublevels are generally not equidistant. Figure 4 pictures three such sublevels  $m_F$ ,  $m_F + 1$ ,  $m_F + 2$ . The two-photon

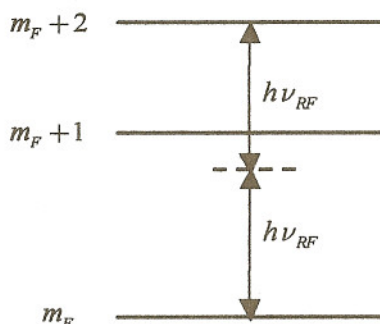


Fig. 4. Two-photon transition between unequally spaced Zeeman sublevels

transition between  $m_F$  and  $m_F + 2$  corresponds to a resonant process where the atom goes from  $m_F$  to  $m_F + 2$  by absorbing two  $\sigma_+$ -polarized RF photons. The conservation of total energy and angular momentum is clearly fulfilled if  $E(m_F + 2) - E(m_F) = 2 h \nu_{RF}$ . It is clear also that such a higher order process goes through a non resonant intermediate state  $m_F + 1$ . Finally, Fig. 4 shows that  $\nu_{RF}$  is located between the resonant frequencies corresponding to a single  $\sigma_+$  photon transition between  $m_F$  and  $m_F + 1$  and between  $m_F + 1$  and  $m_F + 2$ . In fact, in the experiment, one keeps the frequency of the RF field fixed and the static field is scanned. In these conditions, the two-photon transition  $m_F \rightarrow m_F + 2$  appears between the two single-photon transitions  $m_F \rightarrow m_F + 1$  and  $m_F + 1 \rightarrow m_F + 2$ .

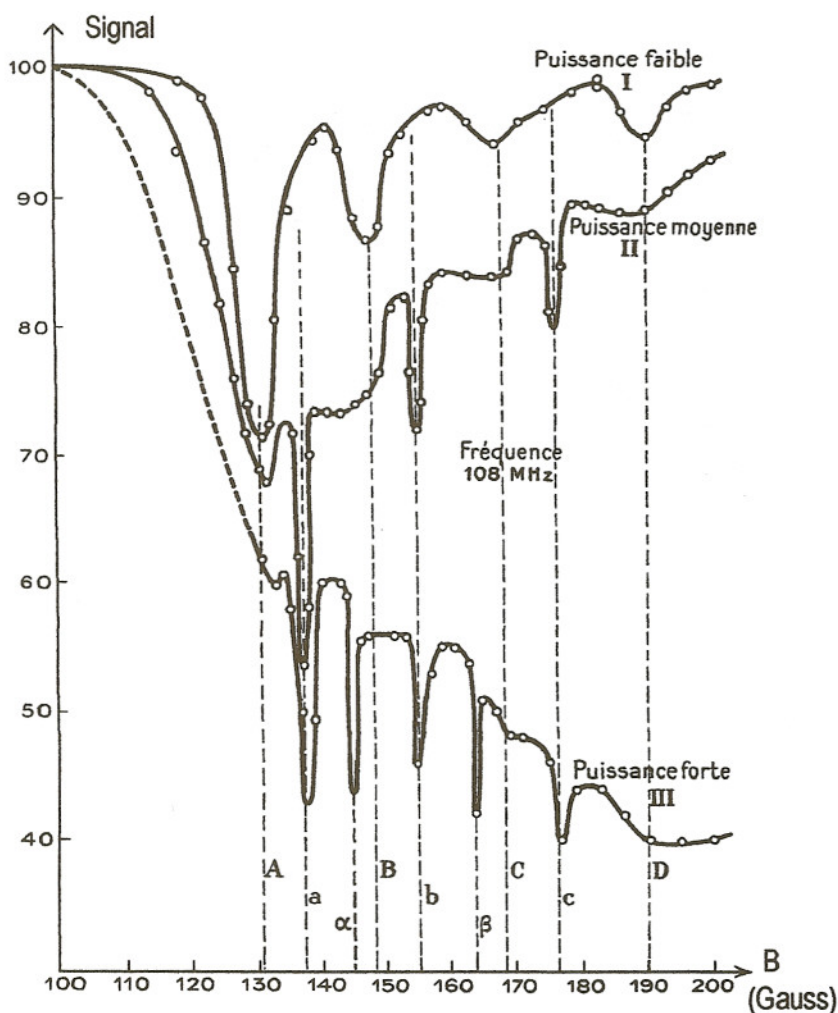


Fig. 5. Spectrum of multiphoton RF resonances observed in the ground state of optically pumped sodium atoms. The curves I, II, III correspond to increasing powers of the RF field. Resonances A, B, C, D correspond to single photon transitions; resonances a, b, c to two-photon transitions; resonances  $\alpha$ ,  $\beta$  to three-photon transitions (Figure taken from Ref. [7]).

### 3.3. Experimental observation with sodium atoms

Multiphoton resonances have been experimentally observed by A. Kastler, B. Cagnac and J. Brosel in 1953 [6-8]. Let us consider, for example, the  $F=2$  hyperfine levels of sodium with five Zeeman sublevels  $m_F = +2, -1, 0, -1, -2$ . The five Zeeman sublevels are not equidistant thus giving rise to four single-photon transitions at four different values of the magnetic field (curve I of Fig.5 corresponding to a weak RF power): resonance A corresponding to a transition between  $-2$  and  $-1$ , resonance B corresponding to a transition between  $-1$  and  $0$ , resonance C corresponding to a transition between  $0$  and  $+1$  and resonance D corresponding to a transition between  $+1$  and  $+2$ . If the RF power of the field is increased (curve II of Fig.5), three two-photon transitions  $\Delta m_F = 2$  appear : resonance a between  $-2$  and  $0$ , b between  $-1$  and  $+1$ , c between  $0$  and  $+2$ . At last, if the RF power is further increased (curve III of Fig.5), two three-photon transitions  $\alpha$  and  $\beta$  also appear : resonance  $\alpha$  between  $-2$  and  $+1$  and  $\beta$  between  $-1$  and  $2$ .

### 3.4. Multiphoton resonances at subharmonics of the Larmor frequency

Multiphoton resonances can also take place between two adjacent Zeeman sublevels  $m_F$  and  $m_F + 1$  with the absorption of one, two, three, four or more RF photons as represented in figure 6. In that case, it clearly appears that the Larmor frequency which gives the energy splitting between two Zeeman sublevels is equal to an integer multiple of the frequency  $\nu_{RF}$  of the RF field .

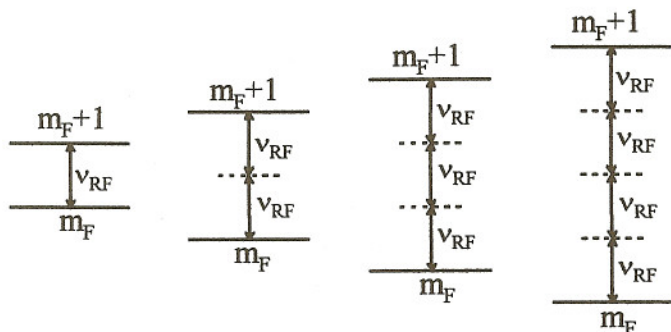


Fig. 6. Multiphoton RF resonances between 2 Zeeman sublevels  $m_F$  and  $m_F+1$ . For a  $n$ -photon resonance, the frequency  $\nu_{RF}$  of the RF field is a subharmonic  $\nu_L/n$  of the Larmor frequency  $\nu_L$  which determines the splitting between the 2 Zeeman sublevels.



These resonances have been also observed in the ground state of sodium atoms. Figure 7 gives an example of such multiphoton resonances obtained by J. Margerie, J. Brossel and J. Winter in 1955 [9,10]. As in figure 5, three curves are represented for three increasing values of the RF field. For low values, one can see only two-photon transitions whereas, for higher values of the RF amplitude, one can also see three and four-photon transitions. Note that the conservation of angular momentum requires that one of the two photons absorbed in the resonance  $\nu_L = 2\nu_{RF}$  must be  $\pi$ -polarized whereas the other one is  $\sigma_+$ -polarized. Similar arguments apply to the resonance  $\nu_L = 4\nu_{RF}$  which must involve  $\pi$ -polarized RF photons as well as  $\sigma_{\pm}$  ones. Coming back to Fig. 3, we see that the RF field must have both parallel and perpendicular components with respect to the static field. Note finally that Fig.7 clearly shows that these multiphoton resonances are broadened and shifted when the RF power is increased. The theory of this power broadening and power shift has been extensively developed by J. Winter [10].

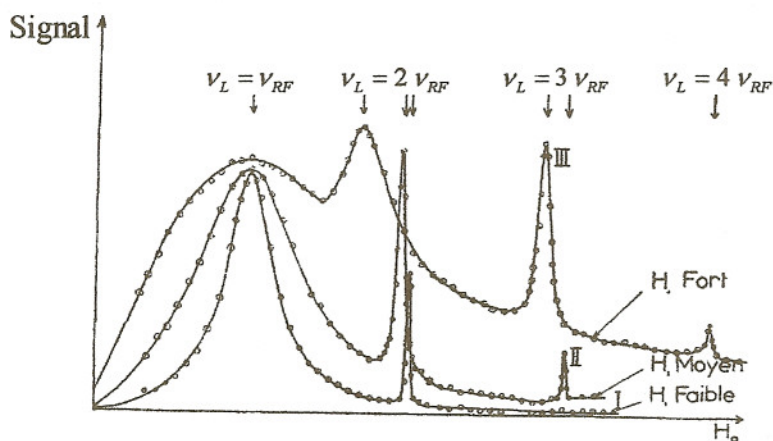


Fig. 7. Spectrum of multiphoton RF resonances  $m_F \rightarrow m_F+1$  observed in the ground state of optically pumped sodium atoms. The curves I, II, III correspond to increasing powers of the RF field (Figure taken from Ref.[9]).

### 3.5. Earlier works on two-photon processes

The first theoretical treatment of two-photon processes was given by M. Goppert Mayer in 1931. It seems also that V. Hughes and L. Grabner observed experimentally in 1950 two-photon processes while studying the RF spectrum of RbF molecules using a molecular beam electric resonance method [11]. They observed an unpredicted group of lines appearing at one half the frequency of another line group. One possible explanation for this unpredicted group of lines was the existence of two-photon transitions, but this was not clearly demonstrated.

### 3.6. Subsequent works on higher order effects

The development of laser sources has made it possible to extend the observation of multiphoton processes to other frequency ranges, in particular to the optical range. I have selected some examples without attempting to be exhaustive.

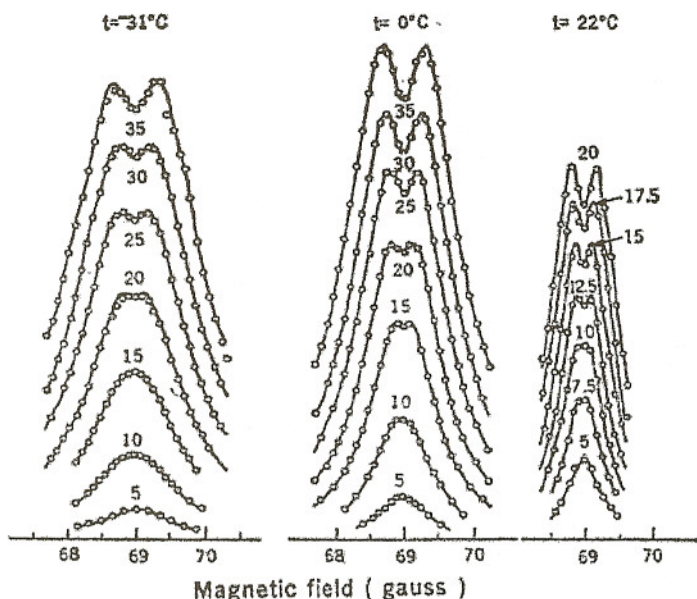


Fig. 8. Three sets of double resonance curves analogous to the set shown in Fig. 1b. These three sets are taken at different temperatures of the reservoir of atoms, increasing from the left to the right and indicated at the top of the figure. One clearly sees that the width of the curves decreases when the vapour pressure increases (Refs [20,21]).

the linear superposition in which the first atom was prepared before emitting his photon. In other words the "Zeeman coherence" in the excited state which determines the width of the magnetic resonance curve is partially conserved in a multiple scattering process and thus appears to have a longer lifetime.

#### 4.2. Linear superpositions of excited Zeeman sublevels

The above described experiment demonstrates the importance of linear superpositions of excited Zeeman sublevels and the first question which can be asked is how they can be prepared. It is clear that a first possibility is the action of a RF field which transforms the state of an atom initially prepared in a well defined Zeeman sublevel into a linear superposition including the other sublevels to which the initial state is coupled by the RF field. Another possibility for producing linear superpositions of excited Zeeman sublevels is to excite the atom with a light whose polarization is a linear superposition of  $\sigma_+$ ,  $\sigma_-$  and  $\pi$  polarizations, so that, starting from a well defined ground state sublevel, several excited Zeeman sublevels can be coherently excited by the incoming light field.

Another important question which can be asked is how do these linear superpositions of states show up in the emission process. The answer is that the light emitted by an atom in a linear superposition of excited Zeeman sublevels is not the same as the light emitted by a statistical mixture of these sublevels. Different quantum interference effects between different scattering amplitudes can be observed. Here are a few examples among others of such physical effects (for a review of the field, see for example Ref.[22] and references therein):

- Coherent multiple scattering, which has been described in § 4.1.



of an excited sublevel  $e_M$ , on the other hand,  $\Gamma_M'$  and  $\Delta_M'$  can be considered as radiative corrections due to absorption and stimulated emission processes in the same way as  $\Gamma_M$  and  $\Delta_M$  can be interpreted in terms of radiative corrections due to the coupling with the empty modes of the radiation field. Another analogy can also be drawn between  $\Gamma_M'$  and  $\Delta_M'$  on the one hand and the imaginary and real part of the index of refraction of a light beam passing through the vapour, on the other hand.

## 5.2 Experimental observation

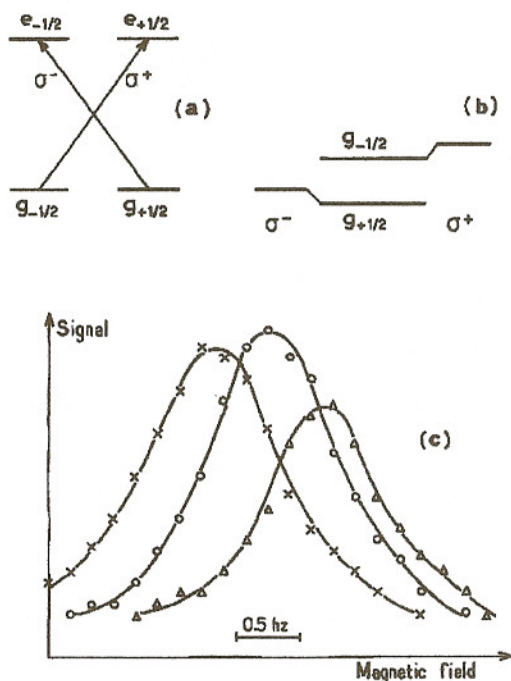


Fig. 9. First experimental observation of light shifts on the transition  $\frac{1}{2} \rightarrow \frac{1}{2}$  of  $^{199}\text{Hg}$  (from Ref. [30]). (a): For a  $\sigma_+$  excitation, only sublevel  $g_{-1/2}$  is light shifted, whereas for a  $\sigma_-$  excitation, only sublevel  $g_{+1/2}$  is light shifted. (b): The splitting between the 2 Zeeman sublevels is increased by a (blue detuned)  $\sigma_+$  excitation, whereas it is decreased by a blue detuned  $\sigma_-$  excitation. (c): Magnetic resonance signal vs magnetic field. The central curve (circles) is taken without the light-shifting beam. When this beam is introduced, either  $\sigma_+$ , - polarized (crosses) or  $\sigma_-$  - polarized (triangles), the magnetic resonance curve is light shifted in opposite directions.

Light shift and light broadening of ground state sublevels have been observed in optical pumping experiments performed on the odd isotopes of mercury [30,31]. These experiments done in 1961 were not using laser sources since these sources were not available at that time. In fact, light shifts could be easily observed because, in general, the two ground state sublevels undergo different light shifts depending on the light polarization. Consequently, the splitting between two Zeeman sublevels is modified by the light irradiation. And, since the lifetime of the atomic ground state is very long, giving rise to very narrow magnetic resonance curves, it is possible to detect such a modification of the splitting between two Zeeman sublevels by a shift of the magnetic resonance curves. In fact, the first experimental observation of light shifts gave evidence of light shifts which were only of the order of one Hertz. These studies also demonstrated that the broadening  $\Gamma_M'$  and the light shift  $\Delta_M'$  of the ground state were varying with the detuning  $\delta$  of the light beam with respect to the atomic transition frequency as absorption and dispersion curves, respectively [31].

### 5.3. Continuity with more recent works on radiative forces

About twenty years later, light shifts and light broadening of ground state sublevels were found to play an important role in the physics of radiative forces [32]. There are in fact two great categories of radiative forces : dissipative forces, which are proportional to  $\Gamma$  and which are due to real absorption and spontaneous emission of photons; and reactive, or dipole forces, which are proportional to  $\Delta'$ , and which are due to virtual absorption and stimulated emission processes. A laser trap which can be achieved at the focus of a red detuned laser beam is a good illustrative example. Since the light shift  $\Delta'$  due to the laser beam is negative for red detuning and maximum in absolute value at the focus of the laser beam, a potential well is formed where sufficiently cold atoms can be trapped. Another example is the atomic mirror formed by a blue detuned evanescent wave produced at the surface of a piece of glass. For blue detuning, the light shift  $\Delta'$  is positive and decreases with the distance to the surface producing a potential barrier upon which atoms can bounce if they are cold enough. Finally, one can mention one important laser cooling mechanism, called "Sisyphus cooling", which is due to spatially modulated light shifts and optical pumping rates in a laser standing wave. A situation is achieved where a moving atom runs up potential hills more frequently than down, losing its kinetic energy (Fig.10). This provides a very efficient cooling mechanism which allows one to reach the microKelvin range [33].

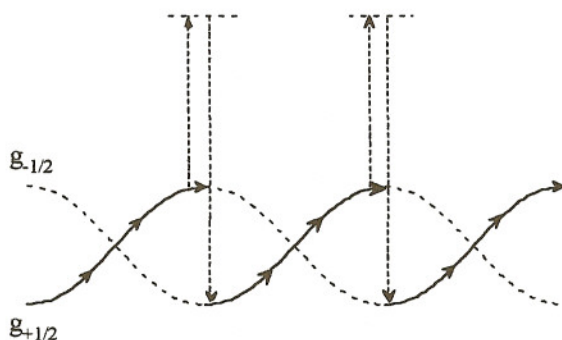


Fig. 10. Sisyphus cooling. In a laser standing wave, the intensity and the polarization of the laser electric field are spatially modulated, giving rise to correlated spatial modulations of the light shifts of two ground state sublevels and of the optical pumping rates between them. As a result of these correlations, a moving atom can run up potential hills more frequently than down.

### 5.4. Other physical effects connected with off diagonal elements of the ground state density matrix

The existence of Zeeman or hyperfine coherences in the atomic ground state manifests itself by several new physical effects which have been studied in detail. Let us mention a few of them:

- Zero field level crossing resonances which are the equivalent for the ground state of the Hanle effect in the excited state [34,35]. However, because the lifetime of the ground state is much longer than the lifetime of the excited state, these resonances are extremely narrow, with a width which can be as small as  $10^{-6}$  Gauss. Such resonances have been used to build magnetometers with a sensitivity of  $5 \times 10^{-10}$  Gauss [35].