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Optical methods for studying Hertzian resonances

Nobel Lecture, December 12, 1966

During my first year of studies at the École Normale Supérieure in Paris, our teacher, Eugène Bloch, introduced us to quantum physics, which at that time was little taught in France. Like he, I was of Alsatian extraction and knew German. He strongly advised me to read Sommerfeld's admirable book *Atom-bau und Spektrallinien*¹. In the course of this reading, I became particularly interested in the application of the principle of conservation of angular momentum during interactions between electromagnetic radiation and atoms, an application which had led A. Rubinowicz² to the interpretation of the selection rules for the azimuthal quantum number and polarization in the Zeeman effect. In the hypothesis of light quanta, this principle attributed to the photons a momentum $+\hbar$ or $-\hbar$ according to whether the light was polarized circularly to the right (σ^+) or to the left (σ^-), natural light being a mixture of the two kinds of photons.

In 1931, W. Hanle and R. Bär³ independently discovered an interesting characteristic of Raman spectra. The study of the polarization of Raman lines at right angles to the incident beam made it possible to classify the Raman lines of a molecule into two categories: « depolarized » lines with a depolarization factor of $6/7$ and « polarized » lines, whose polarization was generally appreciable. Placzek's theory had attributed the former to periodic molecular motions which modify the symmetry elements the molecule possesses at rest, among which are included rotational Raman lines, and the latter to totally symmetric vibrations which maintain the symmetry elements of the molecule at rest.

Hanle and Bär illuminated the medium with circularly polarized incident light and observed that, under these conditions, the Raman lines scattered longitudinally had the same circular polarization as the incident light in the case of totally symmetric vibrations, but that the direction of circular polarization was reversed for lines not totally symmetrical. In a note⁴, I pointed out that for rotational lines this curious result was an immediate consequence of the principle of conservation of angular momentum applied to light scattering.

At about the same time, Jean Cabannes⁵ explained the Hanle and Bär result by the classical polarizability theory, but these publications had been preceded

by an article of Raman and Bhagavantam⁶ who saw proof of the existence of photon spin in the experimental results cited.

At the time, another experiment seemed to me appropriate for demonstrating the possible existence of a transverse component of the momentum of photons: the study of linearly polarized light originating from a rotating atomic oscillator and viewed edge on. This case arises for the S components of the transverse Zeeman effect, which correspond to the S⁺ and S⁻ components of the longitudinal effect. The experiment⁷ that I performed during the Easter vacation of 1931 at the Physics Laboratory of the École Normale Supérieure in Paris, with the aid of Felix Esclangon, was a failure: there is no transverse component of angular momentum in light. Here again, I had been preceded by R. Frisch⁸, who had reached similar conclusions.

These initial attempts caused me to examine more systematically the consequences of the principle of conservation of angular momentum in light scattering and in fluorescence⁹. I realized that the optical excitation of atoms in steps^{10,11} constituted a particularly interesting field of application since, in this case, the operator is free to polarize the different monochromatic radiations whose absorption raises the atom through the successive steps of increasing energy. My thesis consisted in applying this method to the mercury atom¹². It enabled me to check out the various predictions. It constituted a first attempt to obtain, by suitable polarization of the exciting radiation, a selective excitation of definite magnetic sublevels. The very fact that the fluorescence intensity resulting from a step excitation is of nonnegligible order of magnitude relative to the emission intensity resulting from a single excitation showed me, in addition, that the population obtained in the course of a stationary irradiation in the first excited state may become a nonnegligible fraction of the population of the ground state despite the weak intensity of the monochromatic light sources available at that time.

After the development of methods of Hertzian resonance of the ground state of isolated atoms by I. Rabi and his students¹³ and after the first and famous application by Lamb and Retherford¹⁴ of these methods to the states $n = 2$ of the hydrogen atom, the American physicist Francis Bitter attracted attention to the interest inherent in extending the techniques of radio-frequency spectroscopy to the excited states of atoms; but the method he proposed for doing this¹⁵ proved to be inexact¹⁶. My former student Jean Brossel was then working under the direction of Bitter at M. I.T. After an exchange of correspondence, we collectively concluded that the following very simple technique should lead to the desired objective:

The study of optical resonance, for example, that of the mercury atom (ref. 11, Chapter V), had shown that, in the presence of a magnetic field H_0 , excitation with polarized light, or simply with a light beam directed in space, made it possible to obtain a selective excitation of the Zeeman sublevels of the excited state and that this selection still took place in a zero magnetic field¹⁷. Thus, in the case of the even isotopes of mercury, excitation by the 2537-Å line with polarization π leads solely to the sublevel $m=0$ of the excited state 6^3P_1 , whereas excitation with circular polarization S^+ or S^- leads, respectively, to the sublevels $m=+1$ or $m=-1$ of this state. This selective excitation is reflected by the polarization of the resonance light emitted again when the excited atom is not perturbed during the short lifetime of the excited state ($\sim 10^{-7}$ sec). If, while maintaining a constant magnetic field H_0 which separates the Zeeman sublevels from the excited state, one applies perpendicular to this field a radio-frequency magnetic field, $H_1 \cos \omega t$, whose pulsation ω coincides with the Larmor frequency ω_0 , magnetic resonance transitions are induced between the Zeeman sublevels of the excited state, and these transitions are manifested by a depolarization of the light emitted by optical resonance. (In the past, Fermi and Rasetti¹⁸ had already applied an alternating magnetic field to excited atoms, but under conditions which did not correspond to a resonance phenomenon.) Therefore, the observation of the state of polarization of this light permits the optical detection of the magnetic resonance of excited states. We pointed out in the same note that, when the electron beam has a given direction, as in the experiment of Franck and Hertz¹⁹, the excitation of atoms by electron impact also led to the emission of polarized spectral lines²⁰; this proved that this mode of excitation also insured a selective excitation of the Zeeman sublevels of the excited states (alignment), and therefore that this should permit the optical detection of the radio-frequency resonances of these states through observation of the depolarization of the emission lines originating therefrom.

When Jean Brossel was applying the double-resonance method (it combines a magnetic resonance with an optical resonance) to the study of the 6^3P_1 state of the mercury atom, I showed, in an article in *Journal de Physique* of 1950²¹, that the optical excitation of atoms with circularly polarized light made it possible to transfer the angular momentum carried by the light to the atoms and thus to concentrate them in the ground state, either in the positive m sublevels or in the negative m sublevels (depending upon whether the light is S^+ or S^-) and that it was possible, by this optical pumping, to create an atomic orientation and also, due to the coupling between the electronic

magnetic moment and the nuclear spin, a nuclear orientation. In this manner, it should have been possible to obtain distributions very different from the Boltzmann distribution and thus to create conditions permitting the study of the return to equilibrium, either by relaxation or under the influence of a resonant field.

I must confess that, at that time, I had absolutely no knowledge of the slowness of the relaxation processes in the ground state, processes which take place in collisions with the wall or with the molecules of a foreign gas. Essentially, I had planned experiments on atomic beams in vacuum in order to avoid these relaxation effects. It was only later, in the course of the development of the experiments, that it became apparent that the relaxation processes on the walls are slow or can be substantially slowed down by suitable coatings²², that the oriented ground states, insofar as they are orbital *S* states, are much less sensitive to collisions than the excited *P* states²³, and that, consequently, diamagnetic foreign gases can act as buffer gases. This observation later permitted considerable simplification of the experimental technique by working with vapors in a sealed container. When it was found, on the other hand, that the transverse relaxation times were of the same order of magnitude as the longitudinal times²⁴, this made it possible to obtain very narrow resonance lines and led to metrological applications which had not been suspected at the start²⁵.

In 1951, after finishing his pioneering work at M.I.T. on the excited state of the various mercury isotopes²⁶ and thus acquiring a knowledge of the techniques of Hertzian resonance, Jean Brossel returned to Paris. We then decided to organize a team of young research workers recruited from the students of the Écoles Normales Supérieures in order to develop systematically the optical methods of Hertzian resonance. The young people from this team are those who, in about a dozen theses, made personal and original contributions to the common work which is being honored today. In the meantime, the methods we had advocated and applied were picked up by a large number of foreign laboratories, leading to considerable improvements in the technique, which we, in turn, adopted and which were the source of great advances in the research work of our team.

The studies on the excited and ground states of the atoms occupied a primary place in the work of our team and led to a rich harvest of results: we collected numerous data on relaxation processes; from the position of the resonance lines, we were able to make precise measurements of Landé factors and intervals of fine and hyperfine structure and to deduce from them very

precise values of nuclear magnetic moments. We were led to the discovery of numerous phenomena related to high-order perturbations: multiple quantum transitions, effects of Hertzian coherence, demonstration of Hertzian resonance shifts under the influence of optical irradiation, and profound modification of the properties of an atom by the presence of a radio-frequency field. At the same time, with our techniques, foreign teams were achieving important results: measurement of nuclear quadrupole electric moments of alkali metal atoms, discovery of exchange collisions, displacement of hyperfine resonances by collisions with molecules of a foreign diamagnetic gas, and others.

In the course of the development of our research, we frequently had the satisfaction of seeing our predictions confirmed by experiments, but several times the experimental results were contrary to our predictions, thus creating problems whose solution led to advances that were as interesting as they were unexpected. The first piece of research carried out by our team was an example of this.

Indeed, resuming Brossel's experiments on the 6^3P_1 state of the mercury atom and adding an electric field to the magnetic field, Blamont studied the Stark effect of this state for the various even and odd isotopes²⁷ and discovered a narrowing of the magnetic-resonance curves as the density of mercury vapor increases (Fig.1). The width of the magnetic-resonance curves of an excited state of an atom, extrapolated to zero amplitude of the radio-frequency field H_1 , is indeed, as had been shown by Brossel, inversely proportional to the lifetime of this state, and the study of this width permits the measurement of this lifetime. This is a direct consequence of the uncertainty principle. The narrowing observed by Blamont thus appeared to contradict this principle, but Brossel found the explanation for this paradox: the phenomenon occurs because the transverse quantities (Hertzian coherences) are transmitted from atom to atom by processes of multiple scattering of optical-resonance photons. There is thus an « imprisonment of the Hertzian coherence » in the vapor, and this is manifested by the lengthening of the coherence time and by the narrowing of the resonance curves. Indeed, Mlle. Rollet's work, performed within our team²⁸ had just shown that the increasing depolarization of resonance light as a function of the mercury vapor density was due to this multiple scattering of photons and not to collisions, because the depolarization effect is more rapid in a pure isotope than in the natural isotope mixture (Fig. 2). Mlle. Guiochon proved the accuracy of Brossel's hypothesis²⁹ by showing that only the atoms of the same isotope in the mixture of the resonance cell

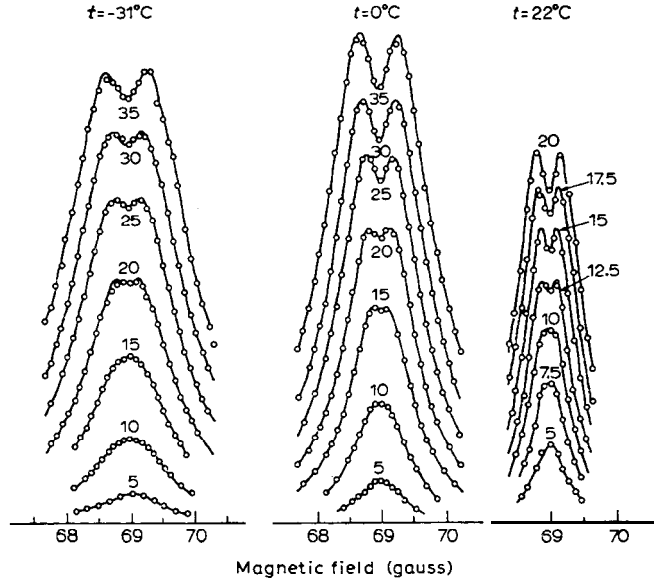


Fig.1. Magnetic resonance curve networks of the 6^3P_1 level of the mercury atom. Each curve corresponds to a constant amplitude of the radio-frequency field H_1 . The numbers indicate the relative values of these amplitudes in arbitrary units. Temperature t of the mercury drop determines the vapor density. (Guiochon, Blamont and Brossel²⁸)

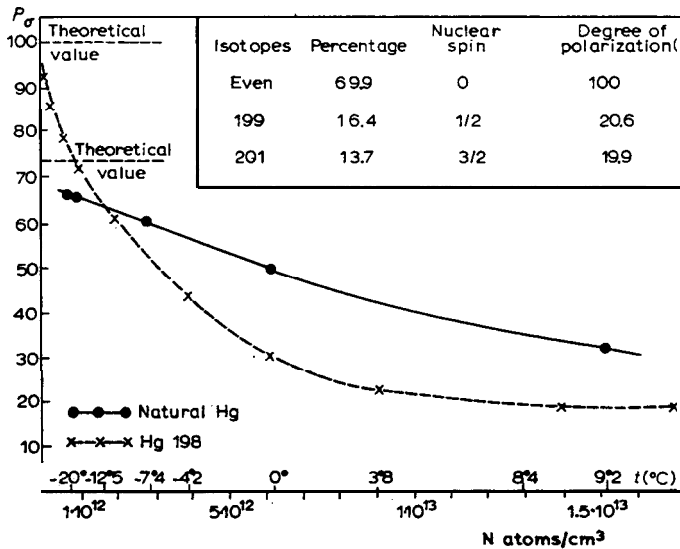


Fig. 2. Degree of polarization of optical resonance light at 2537 Å as a function of vapor density for natural mercury and for the pure ^{198}Hg isotope. (Rollet, Brossel and Kastler²⁹)

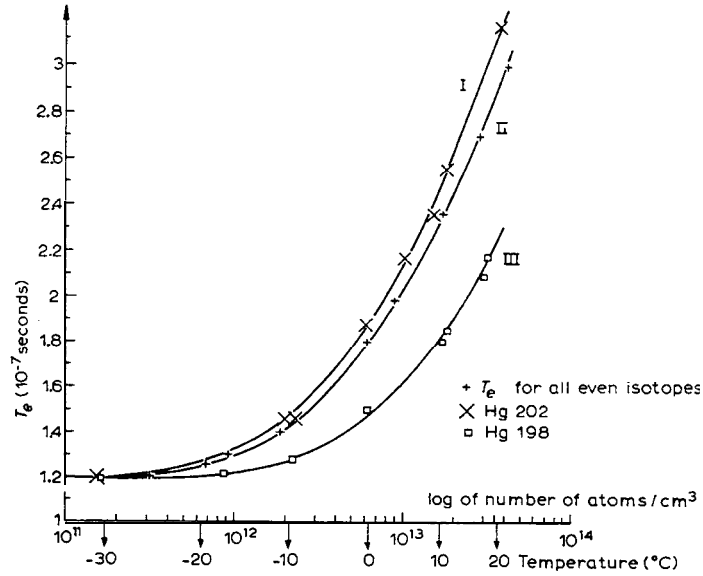


Fig. 3. Coherence time T_e deduced from the width of magnetic resonance curves as a function of the vapor density. Natural mercury in resonance cell. Different light sources: natural mercury, ^{202}Hg and ^{198}Hg . (Guiochon, Blamont and Brossel²⁹)

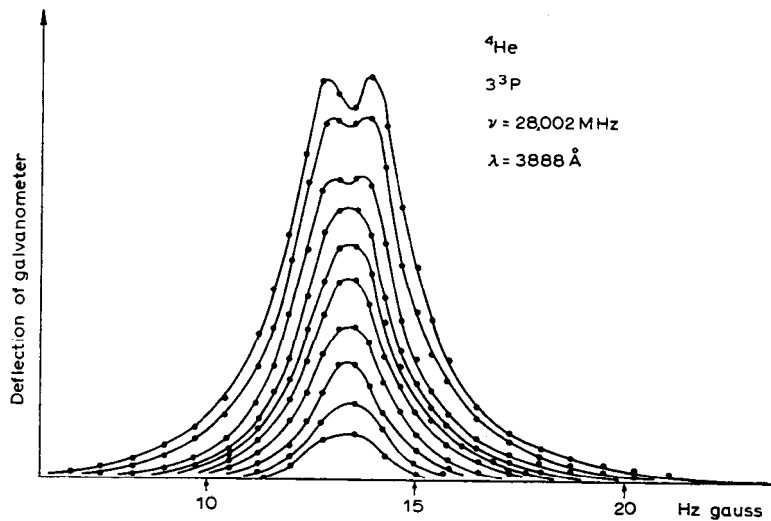


Fig. 4. Network of magnetic resonance curves of the 3^3P level of ^4He atom. (Descoubes³⁵)

produce the narrowing (Fig. 3). In his thesis, Jean-Pierre Barrat³⁰ developed the theory of coherent scattering and verified experimentally all the predictions of this theory. This was the first example of the study of an effect of Hertzian coherence between atomic states, and Barrat showed that these effects can be described in the formalism of the density matrix³¹ which later proved to be very useful and fruitful in the study of other Hertzian coherence effects³².

We charged Pebay-Peyroula with the task of testing the method of excitation by electron impact. In his thesis, he showed the fruitfulness of this technique³³ which later was developed by J. P. Descoubes, who combined it with the level crossing method³⁴; this permitted him to analyze the fine and hyperfine structure of a large number of levels of ^4He and ^3He atoms³⁵ (Fig. 4).

The first tests of the optical pumping technique on a beam of sodium atoms³⁶, from the very first application of a radio-frequency field, led to the discovery of multiple quantum transitions³⁷. Fig. 5 shows the first resonance

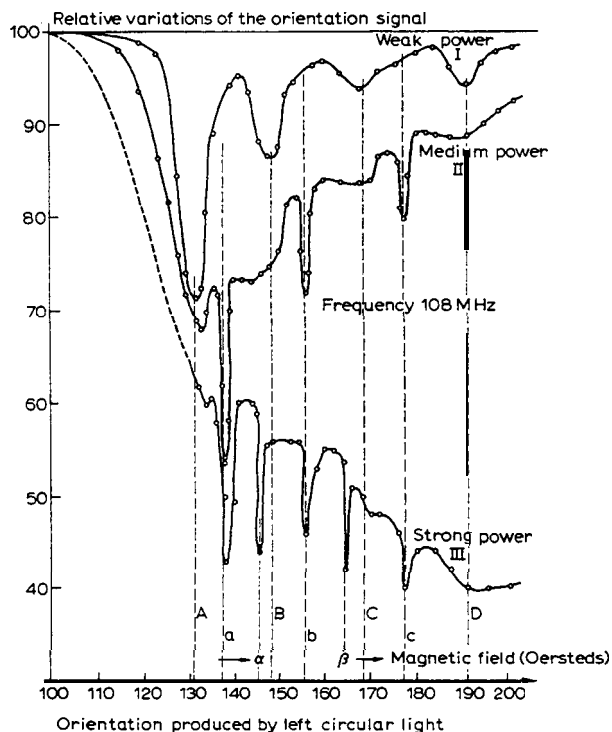


Fig. 5. Magnetic resonance curves of ^{23}Na at a constant frequency of 108 MHz and a variable field H_0 for increasing powers of the radio-frequency field. A, B, C, D, normal resonances $Am=1$; a, b, c, resonances with two quanta $Am=2$; α , β , resonances with three quanta $Am=3$. (Brossel, Cagnac and Kastler³⁷)

curves of the ^{23}Na atom with nuclear spin $I = 3/2$, where, in a field of about 100 gauss, the ordinary Zeeman resonances are already widely separated by the decoupling effect between electronic and nuclear spin. This figure shows the narrow intermediate resonances corresponding to the transition $D m = 2$ induced by the absorption of two radio-frequency quanta.

The study of these multiple quantum transitions was systematically undertaken by J. M. Winter³⁸ who used them to erect a complete theory and was able to predict the existence of a new type of such transitions: in an atomic system possessing only two levels ($m = -\frac{1}{2}$ and $m = +\frac{1}{2}$), multiple quantum transitions are possible when the radiation field contains quanta of different polarization states and when the principle of conservation of energy and the principle of conservation of angular momentum can be satisfied at the same time. In addition, the theory predicts typical radiative broadenings and shifts as a function of the amplitude of the radio-frequency field, and the experiment has verified all predictions one by one.

Fig. 6 shows an example of multiple quantum transitions in the case of the ground state $I = \frac{1}{2}$ of the ^{199}Hg isotope.

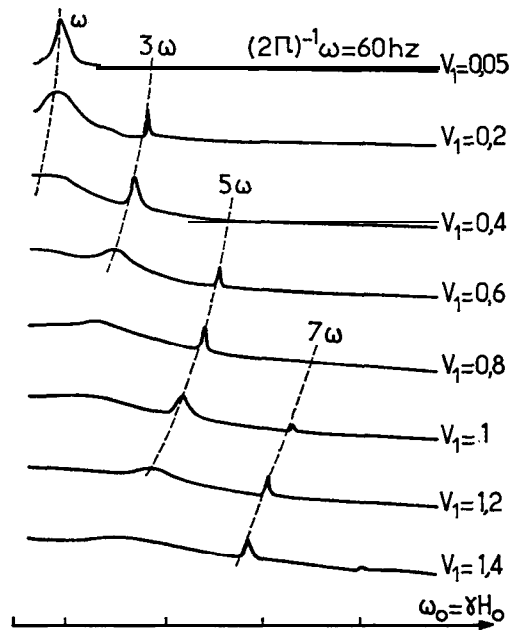


Fig. 6. Resonances with several quanta observed in the ground state. $I = \frac{1}{2}$ of ^{199}Hg ; $\omega_0 = n\omega$ with $n = 1, 3, 5, 7$. From top to bottom, the curves correspond to increasing powers of radio frequency. V_1 indicates the voltage value measured across the terminals of the radio-frequency circuit. (Cohen-Tannoudji and Haroche)

As already noted, the discovery of paraffin coatings²² and the effect of buffer gases²³ considerably facilitated the optical pumping of alkalis in the vapor phase and led to the discovery in the United States of exchange collisions^{39,40}. In our team, Mme. M. A. Bouchiat-Guiochon used paraffin coatings to elucidate the relaxation mechanism on the container wall⁴¹.

After various failures⁴², Bernard Cagnac was the first successfully to obtain nuclear orientation in a low-density vapor (¹⁹⁹Hg and ²⁰¹Hg) by optical pumping and to use it in a study of the nuclear magnetic resonance of these atoms (Fig. 7); with others, he obtained high-precision measurements of nuclear magnetic moments⁴³. Taking advantage of Franzen's elegant method of transients⁴⁴, he was also able to study relaxation processes due to collisions against the walls (Fig. 8), which led to very interesting problems of surface physics⁴⁵.

In similar fashion, J. C. Lehmann was able to orient the nuclei of the odd cadmium isotopes, to observe their magnetic resonance curves, and to measure precisely their nuclear magnetic moments⁴⁶, but he failed in attempts to orient the ⁶⁷Zn nuclei by pumping with the singlet resonance line despite the very high transition probability of this line. This failure caused him to analyze closely the process of nuclear orientation by optical pumping and to show that

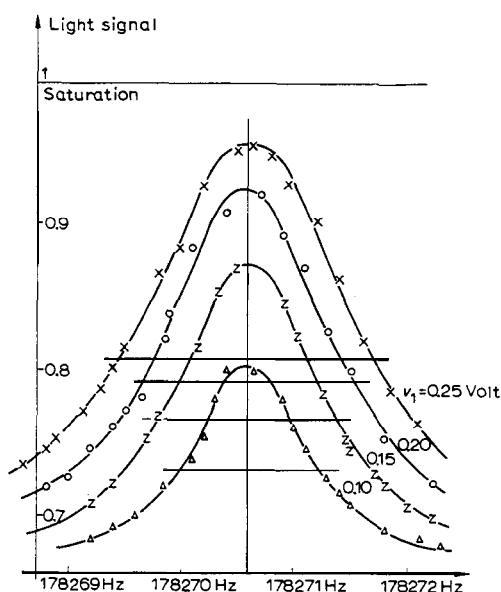


Fig. 7. Network of nuclear magnetic resonance curves of ¹⁹⁹Hg in the ground state. Fixed field H_0 , variable frequency. (Cagnac⁴³)

this orientation does not take place during the light-absorption process, but is produced under the influence of the recoupling of the nuclear and electronic moments in the intermediate stage between absorption and reemission of light⁴⁷. A detailed analysis of the process of nuclear orientation led Lehmann to the development of a method for measuring unresolved hyperfine intervals smaller than the natural width of the levels studied and to a successful application of this method to the odd cadmium isotopes.

We are thus led to a generalization of the Franck-Condon principle: « In a rapid process involving the electronic configuration (spectral transition, disorienting collision, or exchange collision), the position *and orientation* of the atomic nuclei remain unchanged. » The consequences of this principle were verified for disorienting collisions in the excited state by Omont and Faroux⁴⁸, and for exchange collisions in the ground state by Mme. Grossetête⁴⁹.

The cross-beam technique introduced by Hans Dehmelt⁵⁰ proved to be of considerable importance by making the behavior of the transverse macroscopic magnetic moment of a paramagnetic vapor accessible to optical detection (Fig.9). Such a moment which precesses around the constant field H_0 produces a modulation of the absorption of a crossed beam perpendicular to the primary beam and to the field H_0 . This modulation can be readily am-

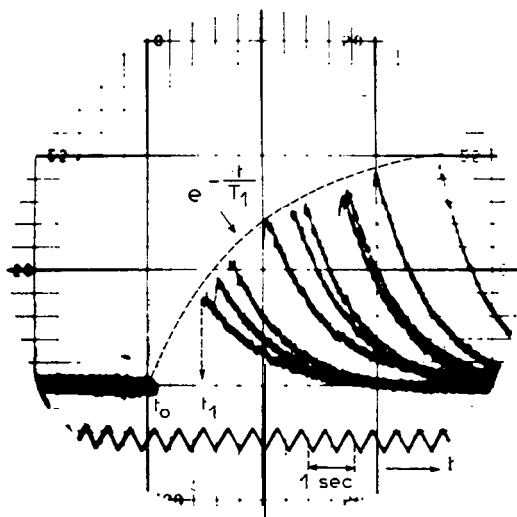


Fig. 8. Transient optical pumping curves of ^{199}Hg photographed on a cathodic oscilloscope screen. After a period of optical pumping, the atoms relax during a dark interval $t_1 - t_0$ and the pumping resumes. This time interval is modified from one curve to another. The dashed curve defines the relaxation exponential and permits measurement of the longitudinal relaxation time T_1 . (Cagnac⁴³)

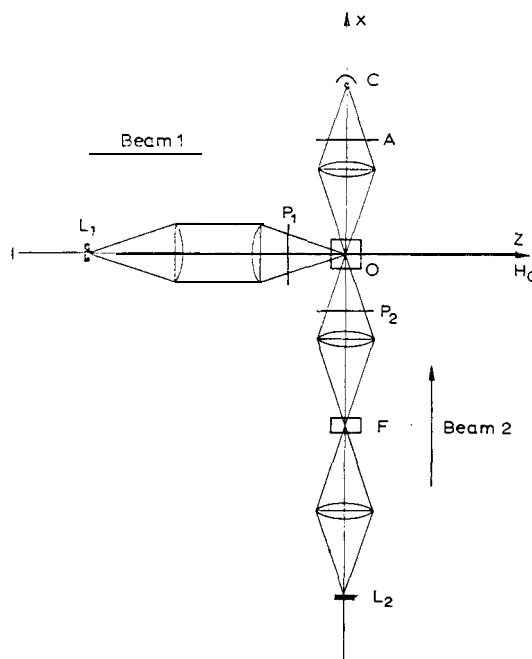


Fig. 9. Diagram of the apparatus used in Dehmelt's cross-beam method. Beam 1, pumping beam in circular light. Beam 2, detection beam whose intensity is modulated by the magnetic resonance; C, photodetector. (Cohen-Tannoudji⁵¹)

plified, and its phase can be precisely determined by synchronous detection techniques.

Using ^{199}Hg as an example, C. Cohen-Tannoudji has shown the advantages inherent in this technique⁵¹, either in studying steady-state resonance effects or in observing transient phenomena. Fig.10 shows how the technique of synchronous detection makes it possible to separate the components of the transverse moment, one of which is in phase and the other out of phase with the alternating field H_1 , producing the resonance. The first of these components varies like a dispersion curve, and the second like an absorption curve. Fig. 11 shows the transient signal obtained when the primary pumping beam and the radio-frequency field are interrupted at the same instant. The exponential decrease of the free precession of the transverse moment is then recorded as a function of time, and from this the transverse relaxation time is directly deduced. The technique of these transients can be combined with 90° and 180° pulse methods. Fig.12 shows the optical signals produced by these pulses.

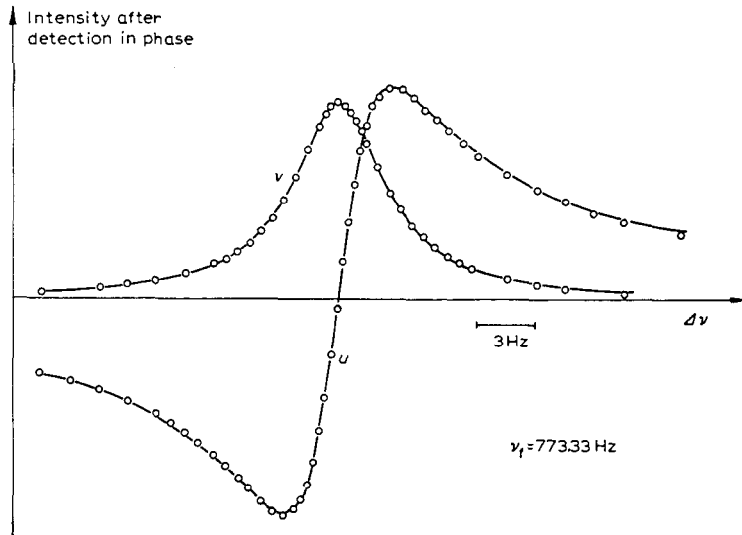


Fig.10. Nuclear magnetic resonance curves of ^{199}Hg observed by the modulation of a cross-beam. Synchronous detection permits the separation of components u and v of the transverse moment: u , component in phase with H_1 ; v , component out of phase with H_1 . (Cohen-Tannoudji⁵¹)

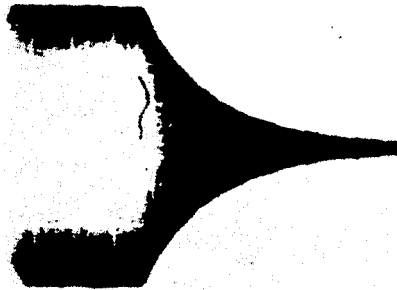


Fig.11. Transient signal of the modulation of the cross-beam: ^{199}Hg . Effect of sudden and simultaneous interruption of the pumping light beam and the radio-frequency field. Free decay of the transverse momentum. Exponential giving transverse relaxation time T_2 (Cohen-Tannoudji⁵¹)

Cohen-Tannoudji used the refinement of these techniques to study the effect of energy shifts caused by a luminous irradiation⁵¹ and predicted by the quantum theory of the optical pumping cycle⁵². He showed the existence of two kinds of such shifts: shifts due to real optical transitions which, due to the oscillation of the atom between the ground state and the excited state, produce

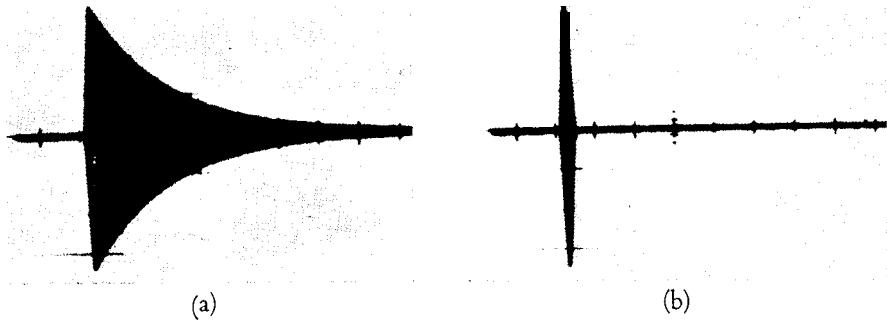


Fig.12. Transient signals of the modulation of cross-beam: ^{199}Hg . (a) Effect of a 90° radio-frequency pulse; (b) effect of a 180° radio-frequency pulse. (Cohen-Tannoudji⁵¹)

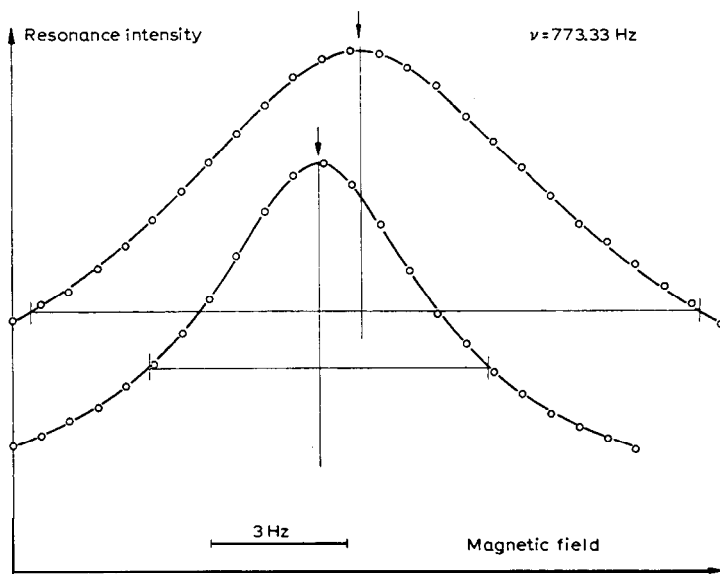


Fig. 13. Nuclear magnetic resonance curves of ^{199}Hg observed by modulation of the cross-beam. For the upper curve, the intensity of the cross beam is five times greater than for the lower curve. The increase of light intensity widens the resonance curve and shifts its center. (Cohen-Tannoudji⁵¹)

a mixture of Larmor precessions of the two states (Fig. 13), and shifts due to virtual transitions produced by radiation which is not absorbed by the atom but which is close to an absorption frequency (Fig. 14). In the latter case, the interaction between the atom and the radiation field is manifested by two complementary effects: the action of the atoms on the light produces a change

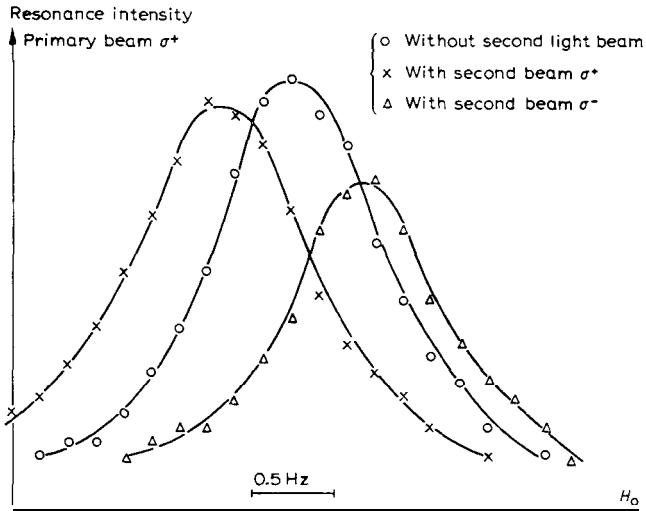


Fig. 14. Effect of virtual transitions. Shifts of the center of the ^{199}Hg nuclear magnetic resonance curve influenced by a second light beam (Cohen-Tannoudji⁵¹)

in the velocity of propagation of the latter described by the abnormal dispersion curve, a phenomenon which has been known for a century; the action of the light on the atom produces a shift of the ground level of the atom. As a function of the deviation of the light frequency from the resonance frequency, the magnitude of the shift also varies like an abnormal dispersion curve (Fig. 15)⁵³.

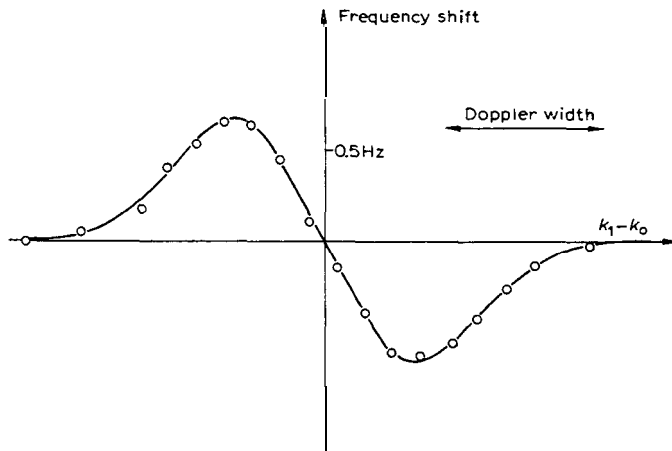


Fig. 15. Magnitude of shift as a function of $k_1 - k_0$, Center of optical absorption line of the atoms; k_1 center of acting radiation. k designs the wave number of the light. (Cohen-Tannoudji⁵¹)

We should note that this latter type of shift was successfully amplified to a considerable extent by Russian physicists using the intense light of a ruby laser whose wavelength is close to that of a transition of the potassium atom⁵⁴.

Cohen-Tannoudji and his students applied themselves to a more extensive study of the interactions related to virtual absorptions and emissions of radio-frequency photons by an atom, interactions which give rise to new resonances whose characteristics are quite distinct from those of resonances corresponding to multiple quantum transitions described above⁵⁵.

Finally, when in the vicinity of zero value of H_0 , an atom is placed in a non-resonant radio-frequency field, one observes a change of the Landé factor of the atom as a function of the intensity of this field⁵⁶, a change illustrated by Fig.16: all of these effects can be understood from a synthetic standpoint by studying the energy diagram of the total system « atom + radio-frequency photons »⁵⁷.

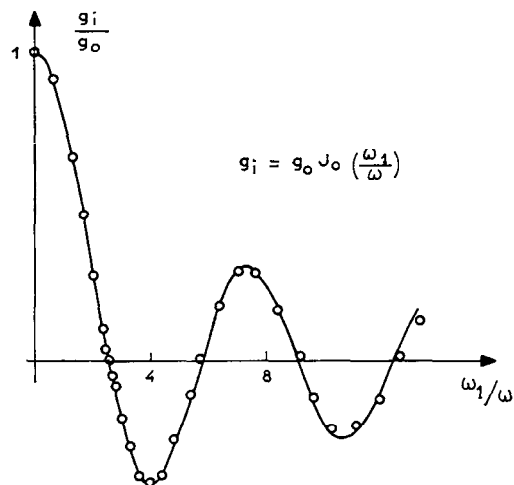


Fig. 16. Modification of the Landé factor of an atom (nuclear Zeeman effect of the ground state of ^{199}Hg) as a function of the intensity of radio-frequency field $H_1 \cos \omega t$ acting upon it. $\omega_1 = \lambda H_1$. (Cohen-Tannoudji and Haroche⁵⁶)

In conclusion, let us note that Jean Margerie⁵⁸ has shown that the optical methods of radio-frequency resonance can be transposed to paramagnetic ions and to F centers in solids and can yield valuable data on the structure of excited levels even in cases where the structure of these levels has not been spectrally resolved.

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