Optical Methods For the Study of Radio-Frequency Resonances

The double resonance method and optical pumping technique have led to advances in the study of nuclear spins and magnetic moments, electron magnetic moments, multiple-quantum transitions, and nuclear hyperfine interactions. Excitation by electron impact has permitted the study of atomic levels not accessible with resonance radiation.

by Alfred Kastler

DURING MY FIRST YEAR as a student at the Ecole Normale Supérieure in Paris, our professor, Eugene Bloch, introduced us to quantum physics, which was seldom taught in France at that time. Like him, I was from Alsace and spoke German. He strongly advised me to read Arnold Sommerfeld's excellent book *Atombau und Spektrallinien*.[1] When reading this book, I was particularly interested in the application of the principle of conservation of angular momentum in interactions between electromagnetic radiation and matter—an application that guided A. Rubinowicz[2] to the interpretation of the selection rules for the angular momentum quantum number and for polarization in the Zeeman effect. On the assumption of light quanta, this principle led to attributing an angular momentum to the photons $+h/2\pi$ or $-h/2\pi$, according to whether the light was circularly polarized to the right (σ^+) or to the left (σ^-), natural light being a mixture of the two types of photon.

In 1931 both Wilhelm Hanle and R. Bär[3] discovered, independently of each other, an interesting peculiarity of Raman spectra: The study of the polarization of Raman lines at right angles to the incident beam had led to a classification of the Raman lines of a molecule into two categories: "depolarized" lines with a depolarization factor of 6/7, and "polarized" lines, the polarization of which was generally considerable. It was Placzek's theory that identified the first category as owing to the periodic molecular movements, which modify the elements of symmetry of the unexcited molecule, among which must be included the Raman rotation lines; it attributed the second to the totally symmetrical oscillations, which maintain the elements of symmetry of the unexcited molecule.

Hanle and Bär illuminated the substance with circularly polarized incident light and showed that under these conditions Raman radiations, longitudinally scattered, had the same circular polarization as the incident light in the case of totally symmetrical oscillations, but that their direction of circular polarization was reversed for oscillations that were not totally symmetrical. In a paper[4] I drew attention to the fact that, for the rotation lines, this curious result was the immediate consequence of the conservation-of-angular-momentum principle applied to the process of light scattering.

At the same time, Jean Cabannes[5] explained the results of Hanle and Bär by the classical theory of polarizability; but before these publications both C. V. Raman and S. Bhagavantam[6] had analyzed the experimental results just cited in terms of the existence of photon spin.

Another experiment then appeared to me to be appropriate for demonstration of the possible existence of a transverse component of the photon angular momentum: It was the study of linearly polarized light, emitted by a rotating atomic oscillator, observed in its own plane. This situation occurs for the σ components of the transverse Zeeman effect, which correspond to the components σ^+ and σ^- of the longitudinal effect. The experiment,[7] which I carried out in 1931 during the Easter holidays at the physics laboratory of the Ecole Normale Supérieure with the help of Felix Esclangon, met with failure: There is no transverse component of angular momentum in light. Here again Otto R. Frisch[8] arrived at an analogous conclusion before me.

These first attempts led me to a more systematic examination of the consequences of the principle of conservation of angular momentum in light scattering and fluorescence.[9] I realized that stepwise optical excitation of atoms[10,11] is particularly interesting because the observer can polarize at will the different monochromatic exciting lines, absorption of which causes the atom to jump the successive steps of the energy scale. My thesis was about the application of this method to the mercury atom.[12] It enabled me to verify the different predictions. It was a first attempt to obtain, by a suitable po-

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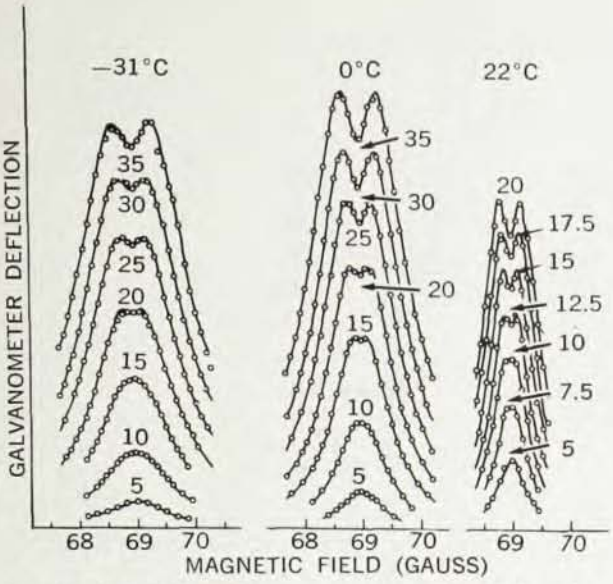
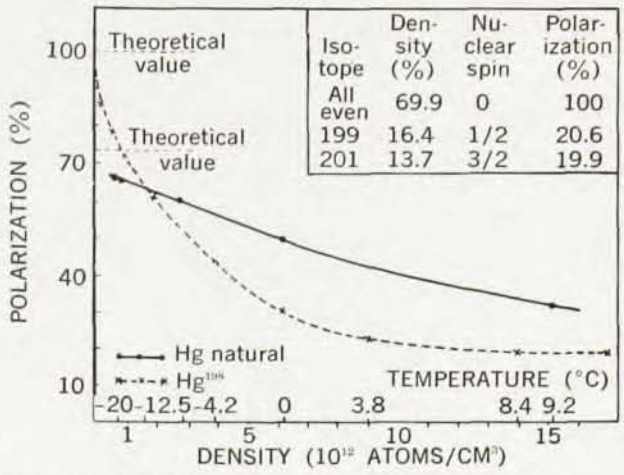


MAGNETIC-RESONANCE curves of the 6^3P_1 level of the mercury atom. Each curve corresponds to a constant amplitude of the radio-frequency field H_1. The numbers indicate the relative values of these amplitudes in arbitrary units. The temperature t of the mercury drop determines the vapor density. (Guiochon, Blamont, Brossel.[25])—FIG. 1



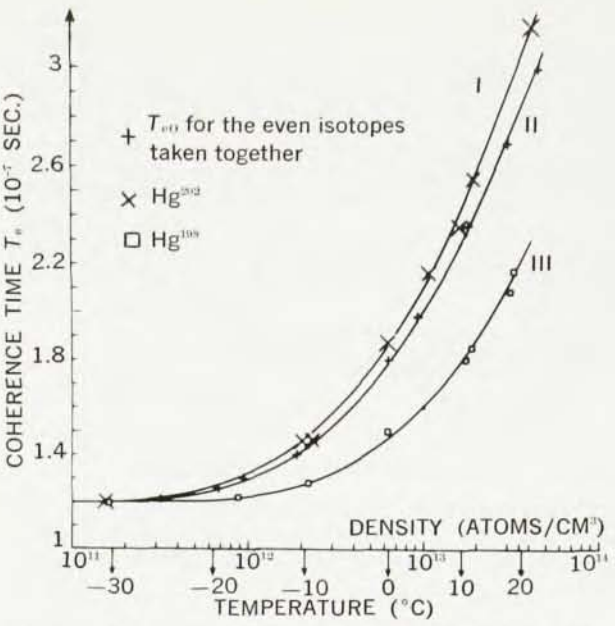
DEGREE OF POLARIZATION of the 253.7 nanometer optical resonance radiation as a function of vapor density for natural mercury and for the pure isotope. (Rollet, Brossel, Kastler.[25])—FIG. 2

larization of the exciting radiations, a selective excitation of definite magnetic sublevels. The fact that the intensity of fluorescence resulting from step-wise excitation is of a magnitude comparable with the intensity of resonance radiation indicated, among other things, that the population in the first excited state obtained during a stationary irradiation can become an appreciable fraction of the population of the ground state, despite the weakness of available monochromatic light sources.

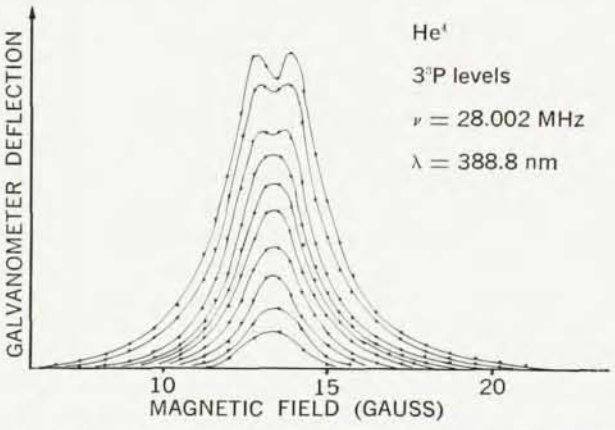
After the development of radio-frequency resonance methods applicable to the study of the ground state of isolated atoms by Isidor I. Rabi and coworkers,[13] and after a famous example of the application of these methods to the $n = 2$ states of the hydrogen atom by Willis E. Lamb and R. Curtis Retherford,[14] American physicist Francis Bitter drew attention to the great interest in extending the techniques of radio-frequency spectroscopy to the study of excited atomic states, but the method he proposed[15] proved to be incorrect.[16] A former student of mine, Jean Brossel, was then working under the direction of Bitter at MIT. After an exchange of correspondence we both concluded that the following simple technique would lead to the desired result.

The study of resonance radiation, in particular in the mercury atom (ref. 11, chap. 5), had shown that, in the presence of a magnetic field H_0, the excitation by polarized light, or simply by light propagating in a given direction, produces a selective excitation of the Zeeman sublevels of the excited state; this is also true in zero magnetic field.[17] Thus in the even isotopes of mercury excitation by plane-polarized π radiation at 253.7 nanometers leads only towards the $m = 0$ sublevel of the excited 6^3P_1 state, whereas excitation by circular polarization σ^+ (or σ^-) leads toward the $m = +1$ (or $m = -1$) Zeeman substates. This selective excitation is reflected in the polarization of resonance radiation, reëmitted in a time of the order of 10^{-7} sec when the excited atom is not perturbed during the lifetime of the excited state. If, therefore, while maintaining a constant magnetic field H_0 that splits the Zeeman sublevels of the excited state, one applies perpendicular to this field a radio-frequency magnetic field $H_1 \cos\omega t$ (the frequency ω of which coincides with the Larmor frequency ω_0 in the field H_0), one causes magnetic-resonance transitions between the Zeeman sublevels of the excited state, and these transitions appear in a depolarization of resonance radiation. (Enrico Fermi and Franco Rasetti[18] had already applied an alternating magnetic field to excited atoms but under conditions that did not correspond to a resonance effect.) And so, the observation of the state of polarization of this light enables optical detection of magnetic resonance in the excited state. In the same paper we drew attention to the fact that excitation of atoms by electron impact, when the electron beam has a well-defined direction (as in the experiment of James Franck and Gustav Hertz[19]), also leads to the emission of polarized spectral lines;[20] this proves that this method of excitation also ensures a selective excitation of the Zeeman sublevels of the excited state (H alignment). Therefore this method should enable optical detection of radio-frequency resonances of all excited states by observing depolarization of the emitted lines.

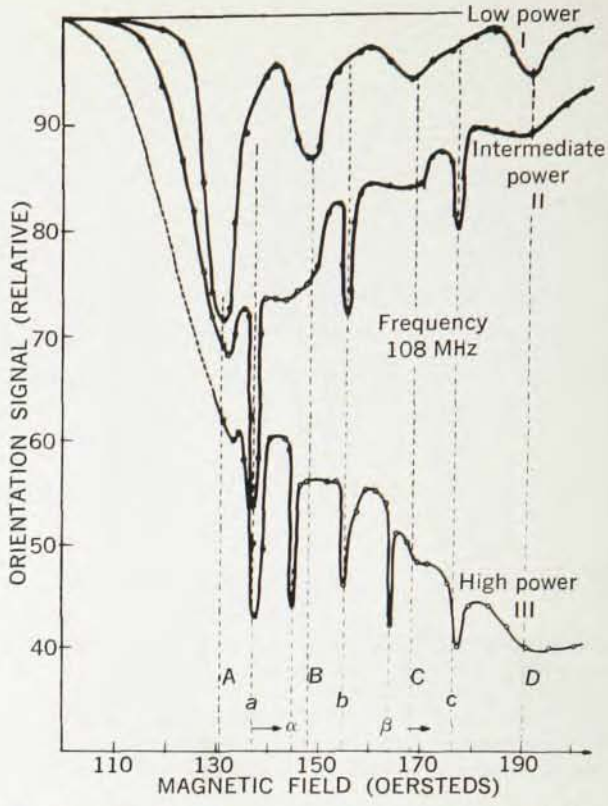
While Brossel put into operation the "double-resonance method" (which uses a magnetic and an optical resonance) to study the 6^3P_1 state of mercury, I showed, in a paper published in the *Journal de Physique* in 1950,[21] that optical excitation by circularly polarized light led to a transfer to the ground state atoms of the angular momentum carried by the light beam. Thus in accumulating these atoms, either in the $+m$ or in the $-m$ sublevels (according to whether the light is σ^+ or σ^-), we were able, by this "optical pumping," to produce an atomic orientation in paramagnetic ground states and also—thanks to the coupling between J, the



COHERENCE TIME T_e deduced from the width of the magnetic resonance curves as a function of vapor density. Natural mercury in the resonance cell. Different light sources: natural mercury, Hg^{202} and Hg^{198}. (Guiochon, Blamont, Brossel.[20]) **—FIG. 3**



MAGNETIC-RESONANCE curves of the 3^3P level of the He^4 atom. Frequency was 28.002 MHz. (Descoubes.[35]) **—FIG. 4**



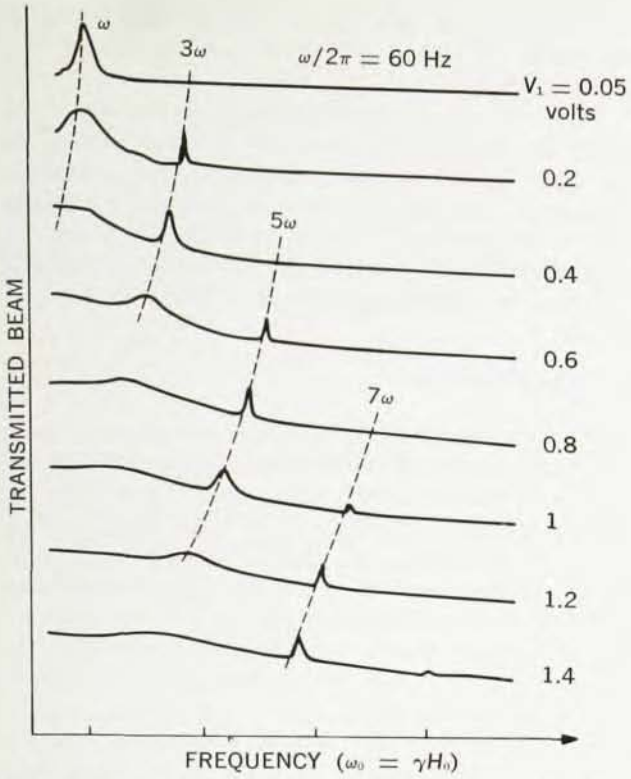
MAGNETIC-RESONANCE curves of Na^{23} at a constant frequency of 108 MHz and variable H_0 field for increasing values of the radio-frequency field: A, B, C, D—normal resonances, $\Delta m = 1$. a, b, c—two-quantum resonances, $\Delta m = 2$. α,β—three-quantum resonances, $\Delta m = 3$. (Brossel, Cagnac, Kastler.[37]) **—FIG. 5**

total angular momentum, and I, the nuclear spin—a nuclear orientation. By this method it should be possible to obtain distributions very different from the Boltzmann distribution, and thus to create conditions allowing the study of the return to equilibrium under the effect of either relaxation or a resonant field.

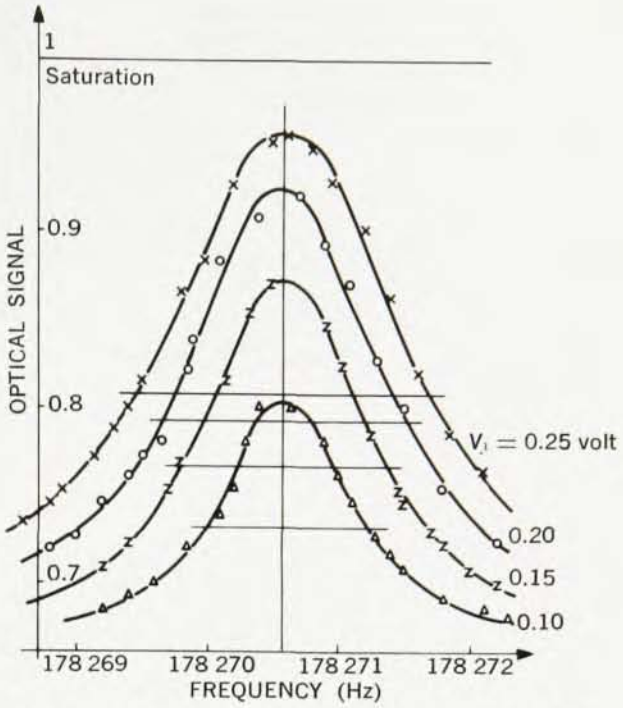
I *must admit* that I was quite unaware of the slowness of the process of relaxation between sublevels in the ground state, which takes place during collisions with the wall or the molecules of a foreign gas. In particular I had planned experiments using atomic beams to avoid these relaxation effects. Later it appeared that the relaxation processes on the walls are slow or can be slowed down considerably by appropriate coatings,[22] and that the oriented ground state, insofar as it is an orbital S state, is much less sensitive to collisions than the excited P states.[23] As a consequence, diamagnetic foreign gases can play the role of buffer gases. Later this fact allowed us to simplify considerably the experimental technique in working on a vapor in a sealed cell. As it was also found that transverse and longitudinal relaxation times are of the same order of magnitude,[24] we have been able to obtain resonance lines of great sharpness that led to measurement techniques unsuspected at the start.[25]

In 1951 Brossel had completed at MIT his pioneering work on the excited state of the various mercury isotopes.[26] He returned to Paris with a good knowledge of the techniques of radio-frequency spectroscopy. We decided to start a research group with the students of the Ecoles Normales Supérieures in order to develop systematically these optical methods of radio-frequency resonance. The young members of this team have provided, in about a dozen theses, personal and original contributions to the work being honored today. Meanwhile, the methods that we had advocated and put into practice had been taken up in many foreign laboratories. In this way technical improvements were introduced which we, in turn, adopted and which became the source of considerable progress in our own team.

Studies on excited and ground states of atoms were carried out simultaneously in our laboratory and led to



MULTIPLE-QUANTUM RESONANCES observed in the ground state $i = 1/2$ of Hg^{199} $\omega_0 = n\omega$ with $n = 1, 3, 5, 7$. Reading from top to bottom the curves correspond to increasing values of the radio-frequency field. V_1 indicates the value of measured potential in volts at the terminals of the radio-frequency circuit. (Cohen-Tannoudji, Haroche.)
—FIG. 6

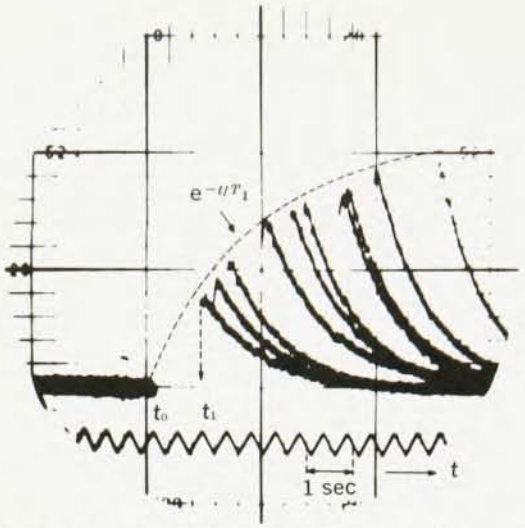


NUCLEAR-MAGNETIC-RESONANCE curves of Hg^{199} in the ground state: constant H_0 field, variable frequency. (Cagnac.[43])
—FIG. 7

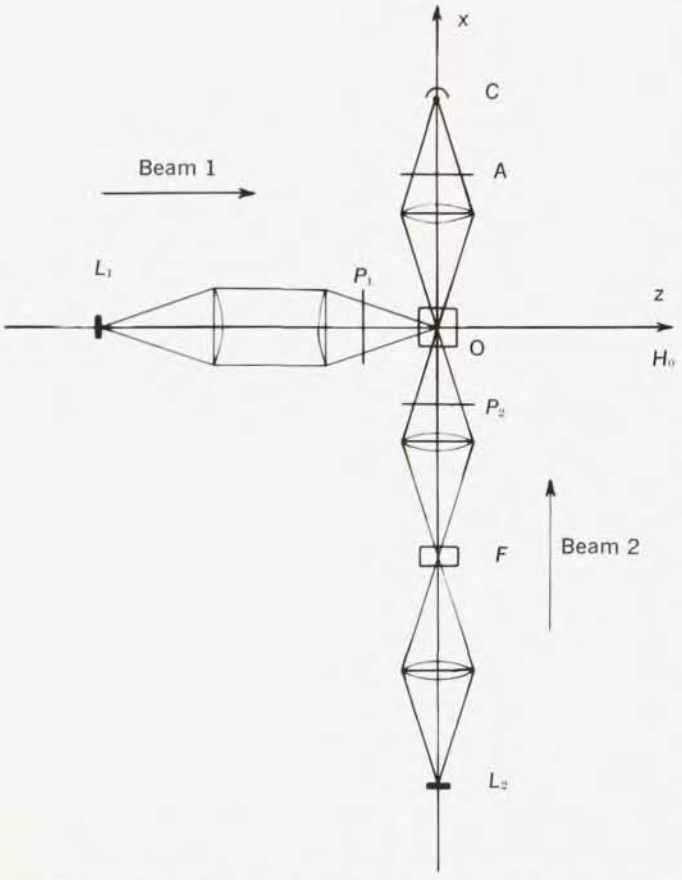
many results: We obtained a great deal of information on the relaxation processes; from the position of the resonance lines we were able to measure with great precision the Landé factors and the fine and hyperfine structure intervals and deduce from them precise values of nuclear magnetic moments. We were led to the discovery of numerous phenomena connected with perturbations of higher order—for instance multiple-quantum transitions—and also effects of radio-frequency coherence, displacements of radio-frequency resonances caused by optical irradiation and the deep alteration of atomic properties by an intense radio-frequency field. By using our techniques, foreign teams succeeded at the same time in obtaining important results: measurement of electric quadrupole moments of alkali-atom nuclei, discovery of exchange collisions, displacement of hyperfine resonances by collisions with molecules of a foreign diamagnetic gas, etc.

During the development of our research we had the satisfaction of seeing our predictions confirmed by experiments; but it often happened that the results of our experiments were quite unexpected and raised most interesting problems. The first work carried out in our team may serve as an example.

Jacques E. Blamont, by continuing Brossel's experiments on the 6^3P_1 state of mercury and by adding an electric field to the magnetic field, studied the Stark effect of this state for various even and odd isotopes[27] and discovered the sharpening of the magnetic-resonance curves when the density of mercury vapor is increased (figure 1). The width of the magnetic-resonance curves of an excited state, extrapolated to zero amplitude of the radio-frequency field H_1, is, in fact, as Brossel had shown, inversely proportional to the lifetime of this state, which can be deduced from it; this is a direct consequence of the uncertainty principle. The sharpening below the uncertainty principle limit as observed by Blamont appeared, therefore, to contradict this principle. Brossel found the explanation of the apparent paradox: The sharpening occurs because the transverse moments (radio-frequency "coherences") are transferred from atom to atom by the process of resonance-radiation imprisonment. There is, therefore, "imprisonment of the radio-frequency coherence" in the vapor, and this is expressed by the lengthening of the "time of coherence" and by the sharpening of the resonance curves. The work of Nelly Rollet[28] had, in fact, shown that the increasing depolarization of resonance radiation as a function of the density of mercury vapor was due to this multiple scattering of resonance radiation and not to collision processes (figure 2). Marie-Anne Guiochon confirmed Brossel's theory[29] by showing that only the atoms of the same isotopic species in the mixture (in the resonance cell) produced the sharpening effect (figure 3). In his thesis, Jean-Pierre Barrat[30] developed the theory of "coherence" of resonance-radiation imprisonment and verified experimentally all his predictions. This was the first example of the effect of "radio-frequency coherence" between atomic states, and Barrat showed for the first time how these effects can be described with the density-matrix[31]



OPTICAL PUMPING transient curves of Hg^{199}, photographed on a cathode-ray screen. After a period of optical pumping, the atoms relax during a darkness interval of $t_1 - t_0$; then the pumping resumes. This interval of time changes from one curve to the other. The curve shown by the dotted line defines the relaxation exponential and allows measurement of the longitudinal-relaxation time T_1. (Cagnac.[43]) —FIG. 8



SCHEMATIC DIAGRAM of the arrangement of Dehmelt's crossed-beam method. Beam 1—circularly polarized pumping beam. Beam 2—detection beam, the transmitted intensity of which is modulated by magnetic resonance. C is the photodetector. (Cohen-Tannoudji.[51]) —FIG. 9

formalism, which later proved helpful and fruitful in the study of other effects of radio-frequency "coherence."[32]

We had asked Jean-Claude Pebay-Peyroula[33] to study the method of excitation by electron impact. In his thesis he showed the value of this technique; following this, Jean-Pierre Descoubes developed it beautifully in conjunction with the level-crossing method.[34] This enabled him to measure[35] the fine and hyperfine structures of a great many levels of He^4 and He^3 (figure 4).

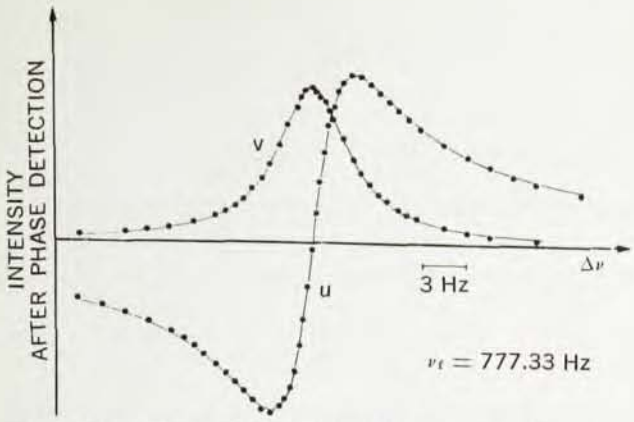
The first attempts at the technique of optical pumping on a beam of sodium atoms[36] had led to the discovery of multiple-quantum transitions.[37] Figure 5 shows the first resonance curves of the Na^{23} atom (nuclear spin $i = 3/2$), where, in a field of about 100 gauss, ordinary Zeeman resonances ($\Delta m = \pm 1$) are already separated by the Back-Goudsmit effect. This figure shows the intermediate sharp resonances that correspond to $\Delta m = 2$ transitions induced by absorption of two radio-frequency quanta.

Study of these multiple-quantum transitions was systematically carried out by Jacques M. Winter,[38] who completed the theory and was able to predict existence of a new type of such transitions: In an atomic system with only two levels ($m = -1/2$ and $m = +1/2$), *multiquantum transitions are possible when the radiation field contains quanta in different states of polarization* so that the principles of conservation of energy and of angular momentum can be satisfied. The theory foresees, among other things, the way in which intensities and positions of the resonances change with the amplitude of the radio-frequency field. All predictions have been confirmed by experiment. Figure 6 shows an example of multiquantum transitions in the case of the ground state $i = 1/2$ of the Hg^{199} isotope.

As we have already mentioned, the discovery of paraffin coatings[22] and of the effect of a buffer gas[23] greatly facilitated the optical pumping of alkali metals in the vapor phase and led to the discovery of exchange collisions made in the US.[39,40] In our team, Bouchiat-Guiochon has studied the mechanism of relaxation on paraffin-coated walls.[41]

After various failures,[42] Bernard Cagnac was the first to succeed in obtaining nuclear orientation in Hg vapor (Hg^{199}, Hg^{201}) by optical pumping, and to use it to study the nuclear magnetic resonance of these atoms (figure 7) and obtain precise measurements of their nuclear magnetic moments.[43] In making use of transients[44] (Franzen's excellent method) he also succeeded in studying the relaxation process owing to collisions against the walls (figure 8). This approach shows that optical pumping is a powerful tool in surface physics.

In an analogous way Jean-Claude Lehmann succeeded in orienting the nuclei of the odd cadmium isotopes. He observed their magnetic-resonance curves and measured their nuclear magnetic moments with great precision,[46] but he failed in his attempts to orient Zn^{67} by pumping with the singlet resonance line despite the very high



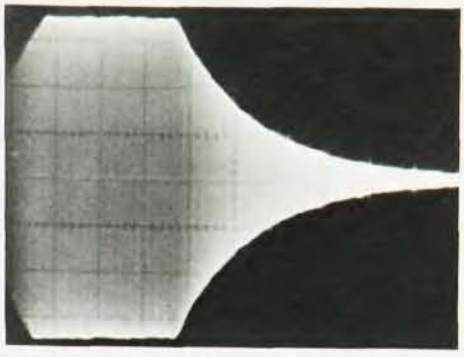
NUCLEAR-MAGNETIC-RESONANCE curves of Hg^{199} observed by crossed-beam modulation. Lock-in detection enables separation of the u and v components of the transverse moment: u, component in phase with H_1; v, component in quadrature (90 deg out of phase) with H_1. Exciting frequency is 773.33 Hz. (Cohen-Tannoudji.[51])

—FIG. 10

transition probability. This setback led him to analyze closely how nuclear orientation is produced by optical pumping. It does not occur during the process of light absorption (electric dipole transition); it does occur because of the recoupling of J and I in the excited state.[47] The detailed analysis of the process of nuclear orientation led Lehmann to describe a method for measuring unresolved hyperfine intervals (smaller than the natural width) and to apply it successfully to odd cadmium isotopes. Thus we are led to a generalization of the Franck-Condon principle: "In a short perturbation which affects the electron configuration (spectral transition, disorienting collision or exchange collision), the position *and the orientation* of the atomic nuclei remain unchanged." The consequences of this principle have been verified for disorienting collisions in the excited state by Alain Omont and Jean-Pierre Faroux,[48] and for exchange collisions in the ground state by Françoise Grossetête.[49]

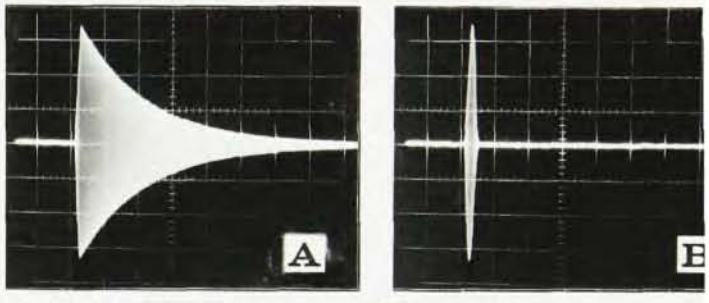
The crossed-beam technique introduced by Hans Dehmelt[50] proved to be of considerable importance in making the evolution of the macroscopic transverse magnetic moment of a paramagnetic vapor accessible to optical detection (figure 9). The existence of a moment that precesses around the constant field H_0 produces a modulation of the absorption of a beam perpendicular to the primary light beam and to the H_0 field. One can easily amplify this modulation and detect its phase by the lock-in detection technique.

Claude Cohen-Tannoudji has shown, in the example of Hg^{199}, all that can be done by this technique[51] to study the stationary effects of resonance or to observe transient phenomena. Figure 10 shows how a lock-in detection technique enables one to separate the components of the transverse moment, of which one is in phase, and the other in quadrature (90 deg out of phase) with the alternating field H_1 that produces the resonance. The first of these components varies as a dispersion curve, the second as an absorption curve.



TRANSIENT SIGNAL from crossed-beam modulation: Hg^{199}. Effect of a sudden and simultaneous interruption in the pumping light beam and the radio-frequency field. Free decrease of the transverse moment. Exponential giving the transverse relaxation time T_2. (Cohen-Tannoudji.[51])

—FIG. 11

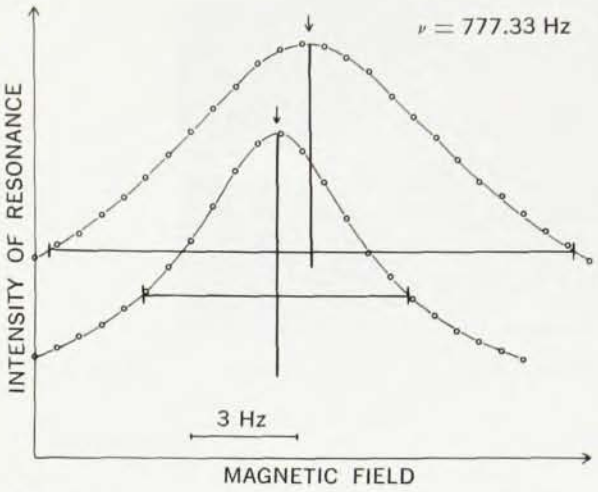


TRANSIENT SIGNALS from crossed-beam modulation: Hg^{199}. (a) Effect of a radio-frequency pulse at 90 deg. (b) Effect of a radio-frequency pulse at 180 deg. (Cohen-Tannoudji.[51])

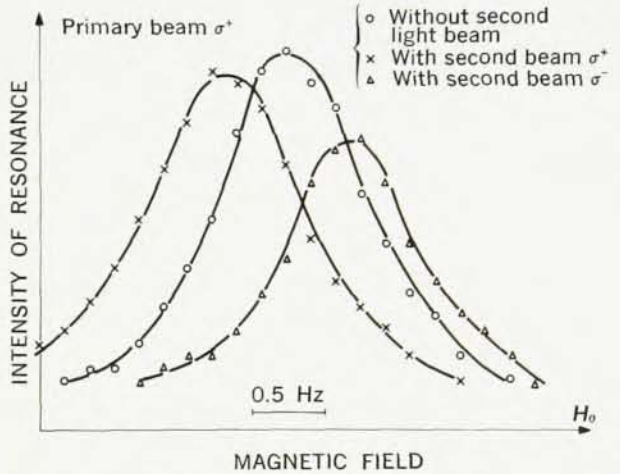
—FIG. 12

Figure 11 shows the transient signal obtained when one interrupts the primary pumping beam and the radio-frequency field simultaneously. Then one observes the exponential decrease of the free precession of the transverse moment as a function of time and deduces from it directly the transverse relaxation time. The technique of these transients can be associated with radio-pulse methods, the pulses being at 90 deg or 180 deg. Figure 12 shows the optical signals produced by these pulses.

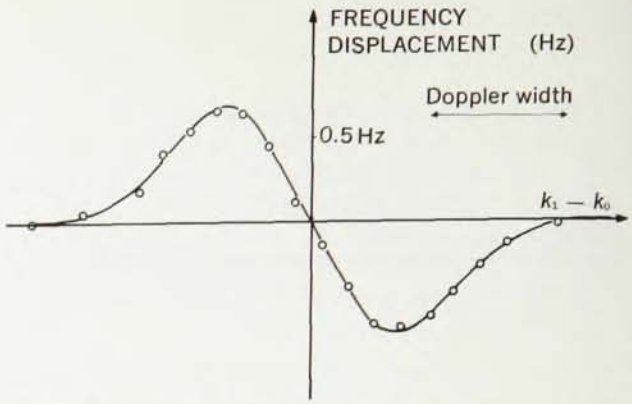
Cohen-Tannoudji used the ingenuity of these techniques to study effects of energy shifts caused by an optical irradiation[51] predicted by the quantum theory of the optical pumping cycle.[52] He showed that there exist two types of such displacement: The first is related to *real* optical transitions, which, by the coming and going of the atom between ground and excited state, produce a mixture of Larmor precessions of the two states (figure 13); the second is related to *virtual* transitions caused by a radiation that is not absorbed by the atom but that is close to an absorption frequency (figure 14). In the latter case, the interaction between atom and radiation field makes itself evident by two complementary effects: (a) The action of the atoms on the light produces an alteration in the speed of light propagation described by the "abnormal dispersion" curve, a phenomenon known for a century; (b) the action of light on the atom produces a displacement of the energy levels of the atom. As a function of the difference be-



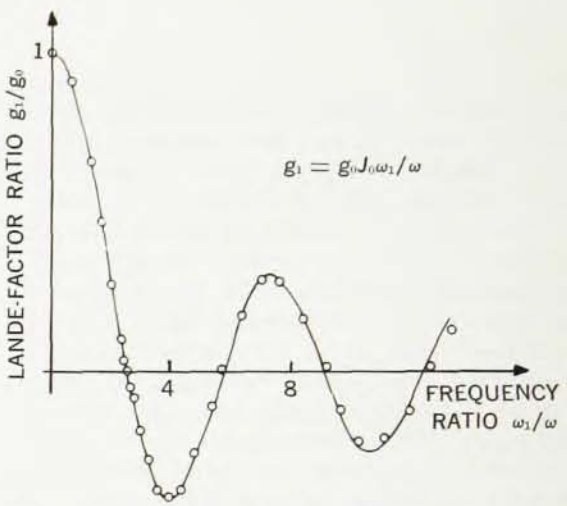
NUCLEAR-MAGNETIC-RESONANCE curves of Hg^{199} with crossed beam modulation. The intensity of the crossing beam in the upper curve is five times stronger than that in the lower curve. The increase in optical intensity raises the resonance curve and displaces its center. Exciting frequency is 773.33 Hz. (Cohen-Tannoudji.[51]) —FIG. 13



EFFECT OF VIRTUAL TRANSITIONS. Displacement of the center of the Hg^{199} nuclear-magnetic-resonance curve under the influence of a second light beam. The primary beam was σ^+. (Cohen-Tannoudji.[51]) —FIG. 14



DISPLACEMENT as a function of $k_1 - k_0$. k_0, center of the optical absorption radiation of the atoms; k_1, center of excitating radiation. (Cohen-Tannoudji.[51]) —FIG. 15



MODIFICATION OF THE LANDE FACTOR of an atom (nuclear Zeeman effect of the ground state of Hg^{199}) as a function of the intensity of a radio-frequency field $H_1 \cos \omega t$ to which this atom is subjected. $\omega_1 = \gamma H_1$. (Cohen-Tannoudji, Haroche.[56]) —FIG. 16

tween the optical frequency and the resonance frequency, the magnitude of this displacement varies just like an abnormal dispersion curve (figure 15).[53]

It must be noted that Russian scientists were able to amplify the latter type of displacement considerably by using the intense light of a ruby laser with a wavelength close to that of a potassium-atom transition.[54]

Cohen-Tannoudji and his students have made a complete and deep study of the interactions related to virtual absorptions and emissions of radio-frequency photons by an atom. These absorptions result in the appearance of new resonances whose characteristics are clearly distinct from those of the multiple-quantum resonances described above.[55]

When one subjects an atom to a nonresonant radio-frequency field in the neighborhood of a zero value for H_0, one observes, as a function of the intensity of this radio-frequency field, a change in the Landé factor of the atom,[56] illustrated by figure 16: All these effects can be understood from a synthetic point of view by studying the energy diagram of the total system "atom plus radio-frequency photons."[57]

Finally, let me mention that Jean Margerie[58] has shown that optical methods of radio-frequency spectroscopy can be applied to paramagnetic ions and F centers in crystals at low temperatures, and they give precise information on the structure of excited levels—even when the structure of these levels is not spectrally resolved.

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