Atomic Lifetimes and Electron Excitation

Controlled-electron spectroscopy permits study of atomic levels not accessible with resonance radiation. With electron-beam excitation, radiation selection rules no longer apply. Polarization data can reveal hyperfine structure, and when the technique is combined with paramagnetic resonance, lifetimes can be obtained from the resonance linewidths.

by H. Henry Stroke

PRECISE STUDIES of atomic lifetimes have intensified during the past few years, undoubtedly with the impetus of the increasing interest of astrophysicists and laser researchers. It had previously been all too common that extensive transition-probability data were not available; one could only rely on calculations. The ensuing progress, however, has not been limited simply to expansion of the more "classical" types of experimentation, which in fact only give the product of the transition probability and the atom density.[1,2] Experiments now measure the lifetime τ directly by some techniques that will be quite familiar to the nuclear physicist. They also measure its Fourier transform, the linewidth $\Delta\omega = 1/\tau$.

Under ordinary optical spectroscopic observation this linewidth is masked by the much larger Doppler linewidth

$$\Delta\omega_D \propto \omega_0 (T/A)^{\frac{1}{2}}$$

where ω_0 is the transition frequency, T the absolute temperature of the radiating atoms, and A their mass number. In the visible, $\omega_0 \approx 10^{15}$ sec^{-1} and typically $\Delta\omega_D = (10^2\text{–}10^3)\Delta\omega$. However, if the linewidth observation is made with radiofrequency photons for which $\omega_0 \approx 10^{10}$ sec^{-1}, then we have $\Delta\omega_D << \Delta\omega$, and the atomic lifetime can be measured. Such observations were achieved first in the optical double-resonance experiments of Francis Bitter and Jean Brossel,[3] illustrated in figure 1. An excellent review of these experiments has been made by Bitter.[4]

Basically these experiments rely on creating among the excited-state magnetic sublevels a population that differs from one that would be obtained in natural excitation. A perturbation of these sublevels is then generally reflected in a change of intensity of polarized radiation.

A May conference on "Electron Excitation of Atomic Vapor," sponsored by the Centre National de la Recherche Scientifique and organized by Jean-Claude Pebay-Peyroula at the University of Grenoble, thus concerned itself with lifetimes of electron-excited states and the polarization of the resulting radiation. The topics discussed included double resonance, level crossing, the Hanle effect, laser excitation, pulsed and modulated excitation, and direct lifetime measurement.

Double-resonance method

Let us consider for illustration the double-resonance experiment in figure 1. For an even isotope of mercury the nuclear spin is zero. First the 253.7-nanometer resonance radiation from the lamp excites the atoms in the cell from the ground state $^1S_0$ to the first optically connected state $^3P_1$. With the polarizer oriented to produce π excitation, we observe that only the $^3P_1$, $m = 0$ sublevel is populated, because of the selection rule $\Delta m = 0$. When the atoms reradiate, the circularly polarized σ components will be absent. However if the excited atoms

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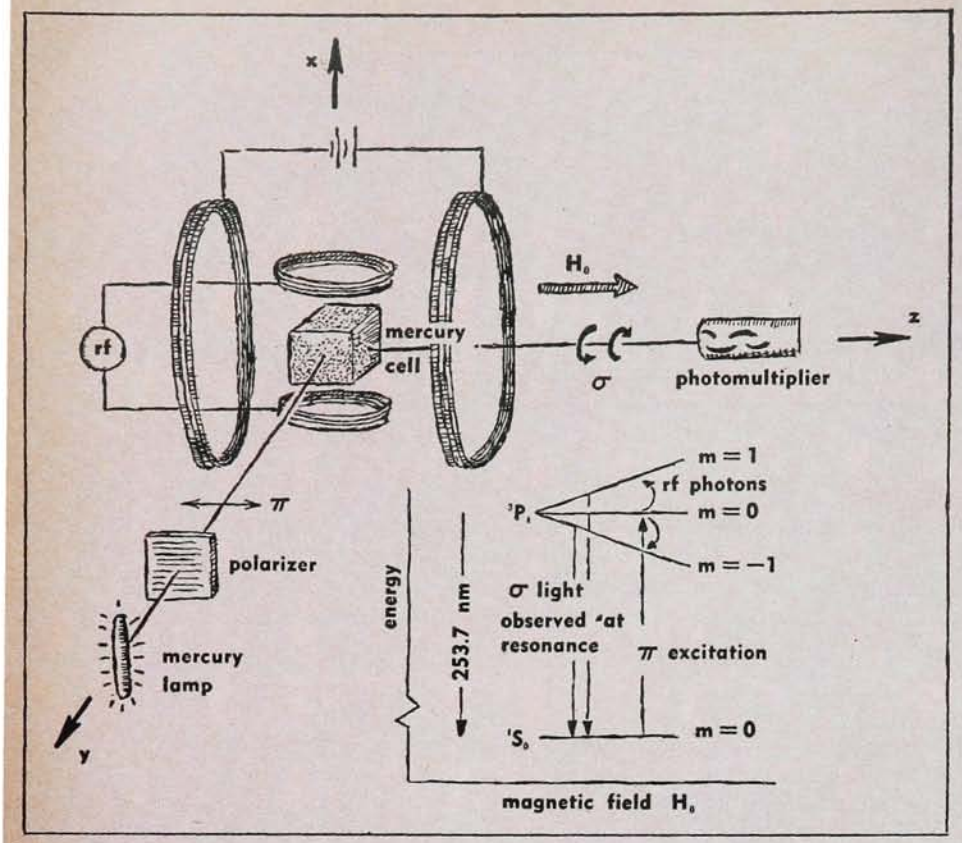


DOUBLE-RESONANCE apparatus of Bitter and Brossel. —Fig. 1

in the cell are subjected to an alternating field, when the frequency is made equal to the Larmor frequency ω_L (the second resonance), atoms undergo transistions from the $m = 0$ to the $m = +1$ and $m = -1$ sublevels. From these sublevels the atoms can reradiate circularly polarized light that is detected by the photomultiplier as an increase in signal. The width of this rf resonance can then give us a "lifetime." We shall discuss later some reasons why this is not the true lifetime of the isolated atom.

This technique and some ingenious variations of it have been very fruitful in the measurement not only of atomic lifetimes but also of electron gyromagnetic ratios (g-values) and hyperfine structure. One limitation, however, is apparent: The only atomic levels that can be studied are those that are reached optically from the ground state (resonance radiation), that is, by strong electric-dipole transitions.

Alignment by electron impact

The restriction of experiments to these relatively few states can fortunately be removed if we cleverly use electrons instead of photons to excite the atoms; for then the radiation selection rules no longer limit us. At the conference, Alfred Kastler, who with Brossel has led for some two decades the most intensive studies in these areas at the Ecole Normale Supérieure in Paris, gave a brief historical survey of controlled-electron spectroscopy. He began with 1914 when James Franck and Heinrich Hertz observed resonance potentials in the inelastic collisions of electrons with atoms in a vapor and deduced positions of the atomic energy levels. In 1926 and 1927 H. W. B. Skinner and E. T. S. Appleyard[5] reported their studies of the excitation of atomic vapor by a collimated beam of electrons. They found that even in the absence of a magnetic field most of the mercury spectral lines radiated after electron impact are polarized such that the maximal electric-field vector is parallel to the electron-beam direction. With the beam directed along a magnetic field the electrons are expected to transfer angular momentum to the atoms in a direction *perpendicular* to the field. Thus at threshold energies the electrons excite the atoms with the selection rule $\Delta m = 0$. We can then readily obtain the threshold polarization

$$P = \frac{I_{//} - I_{\perp}}{I_{//} + I_{\perp}}$$

where $I_{//}$ and $I_{\perp}$ are the radiated light intensities with polarizations parallel and perpendicular to the electron beam observed perpendicular to the field. From the example in figure 2 the threshold polarization can be calculated to be 60%. Furthermore the threshold polarization is expected to be a maximum. The Skinner and Appleyard experiments showed, however, that the threshold polarization was zero but rose to a maximum within a fraction of an electron volt. Although in the ensuing years several theoretical attempts were made to account for the observed threshold polarization, none was entirely satisfactory.

The first part of the Grenoble conference was thus devoted to the studies of the polarization of radiation induced by electron excitation. M. J. Seaton (University College, London) reported on his work with I. C. Percival[6] and D. R. Flower[7] in which they calculated the excitations including fine- and hyperfine-structure effects. He pointed out the possible necessity of relativistic calculation in heavier atoms. In hydrogen the Stark effect must also be considered in accounting for line polarization. The theoretical results still did not give zero polarization at threshold. It was observed, however, that if the experimental polarization values of Skinner and Appleyard are extrapolated from beyond the observed maximum to threshold, reasonable agreement is obtained with theoretical predictions. The onus appears to have been on the early experimental threshold values. The difficulties and progress with the experiments were reviewed by Robert H. McFarland (Lawrence Radiation Laboratory). Higher electron-energy resolution, of the order of 0.2 V, and better signal-to-noise ratio have permitted getting reliable data closer to threshold. Low gas pres-

sures have been found essential in reducing scattered electrons that impair the polarization measurements.

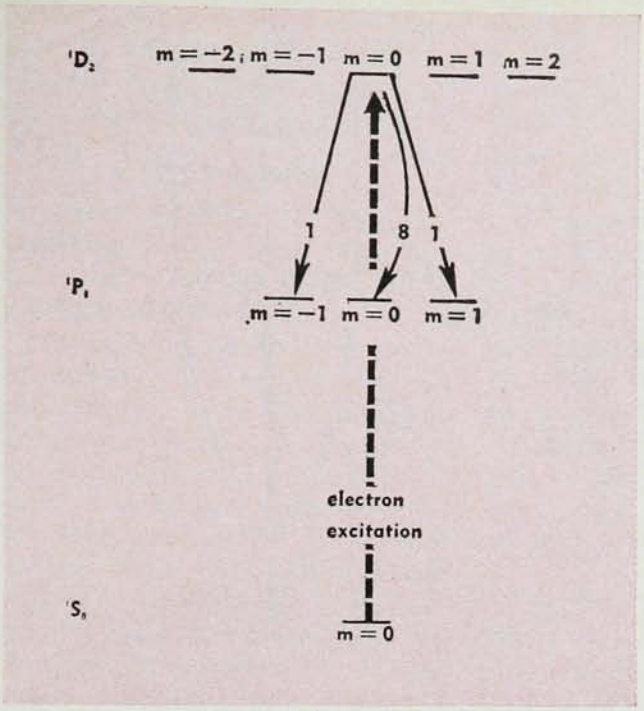
McFarland also emphasized the necessity for adequate spectral resolution in these observations. A monochromator should be used instead of the wider filters common previously. Lack of spectral resolution may have been a widespread source of error as could be indirect excitation processes. The new polarization experiments for the helium $3\,^1P$ and $4\,^1D$ states show indeed that the threshold value is not zero.[8,9] However, another recent experiment with the lithium ($3\,^2D - 2\,^2P$) 610.3-nm line[10] shows that there are still cases in which the threshold polarization unexpectedly approaches zero. The H_α line may present a similar case. Nevertheless Seaton remarked that theory and experiments in many cases are now sufficiently reliable that polarization data can even give a good measure of hyperfine structure.

Crossed electron-atom beams have advantages in avoiding some of the systematic errors. H. Kleinpoppen (Tübingen University) and H. Krüger have used this technique, not only for polarization but also for excitation measurements in hydrogen. They obtain a value of $(0.11 \pm 0.025)\,\pi a_0^2$ for the maximum of the 1s–2s excitation cross section at 11.8 ± 0.3 eV.

F. J. de Heer (Laboratorium voor Massascheiding, Amsterdam) has extended these studies to excitation of atoms with ion impact. Measurements were made with hydrogen- and helium-ion bombardment of helium and argon in the keV region to obtain data on the principal and angular-momentum quantum-number dependence of the gas-atom excitation cross section. The theoretical interpretation emphasizes the influence of electric polarizability on cross section for the several states. De Heer also showed that from the energy dependence of the proton or electron cross section for exciting hydrogen and helium into an optically allowed state, the oscillator strength can be obtained.

Electron-excitation spectroscopy

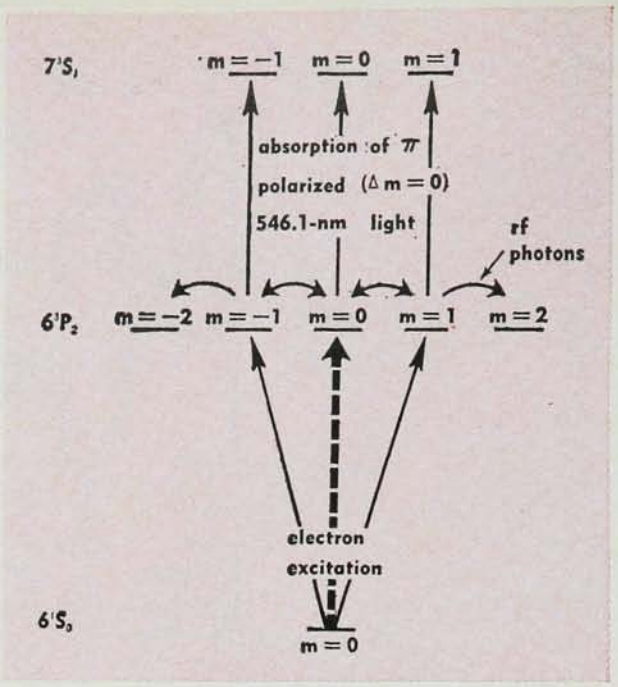
Approximately thirty years after the work of Skinner and Appleyard and after the suggestions by Brossel and Kastler[13a], Hans G. Dehmelt[11] and Willis E. Lamb Jr[12] and, shortly afterward, Pebay-Peyroula, Brossel and Kastler[13b] combined this atomic alignment by electron impact with paramagnetic-resonance techniques. In analogy with the double-resonance experiment the unequal excited-state populations can again serve to detect the radiofrequency resonances by changes in the intensity of polarized light, this time however, either in emission or possibly absorption from a metastable state. We illustrate this in figure 3. Resonance widths again give us lifetime information, and the resonant frequencies the level sructure. Jean-Pierre Barrat (University of Caen) described work done in collaboration with Mireille Barrat and J. Duclos with this method of Dehmelt to measure gyromagnetic ratios in the metastable $^3P_2$ states of zinc and cadmium. They also obtained the excitation cross sections for these states. A further advantage of the electron experiments is noted: Excitation by resonance radiation may require far-ultraviolet light sources that may be difficult to provide whereas electrons can readily reach the same levels. On the other hand we must point out that with electrons many states are excited simultaneously, and one can be plagued by cascading decay processes that may render more difficult the interpretation of lifetime measurements. In this connection rf experiments may be preferable to those that depend on depolarization particularly if the various states have different g factors.



EXCITATION OF MERCURY by electron beam collimated along magnetic field. At threshold energy the polarization of the reradiated light is calculated from the indicated relative intensities observed perpendicular to the field. —Fig. 2

Level crossings

Two further types of closely related experiments are now used extensively in conjunction with both photon and electron excitation for the measurement of atomic level structure and radiation widths. These rely on the Hanle effect[2] and on level crossings.[14] The Hanle effect exhibits depolarization of resonance radiation when the magnetic field is varied from zero to a small value, a few gauss in the case of mercury. Let us again consider figure 1 but without rf coils and with the polarizer turned from the z to the x direction. For $H_0 = 0$ the light measured by the photomultiplier is almost completely linearly polarized in the x direction. If H_0 is slowly increased, the polarization decreases and its direction rotates. It vanishes for a sufficiently large field. The effect can be understood if we consider the excited atoms as classical harmonic oscillators whose axes precess about the magnetic field and whose amplitudes decrease because of atom decay. For relatively small fields the precession angle is small, and the polarization somewhat preserved before radiation. At high fields, with many revolutions of Larmor precession preceeding decay, the preferential polariza-



ELECTRON IMPACT alignment of the mercury $^3P_2$ state. Absorption of the π-polarized 546.1-nanometer radiation by the aligned atoms is decreased when rf resonances are induced, transferring atoms to the nonabsorbing $m = \pm 2$ sublevels. (The $m = \pm 1$ levels are also populated in the excitation due to electron exchange.) —Fig. 3

tion direction is lost. Gregory Breit[2] has shown that the variation of maximal polarization with magnetic field is given by

$$\frac{P(H_0)}{P(0)} = \frac{1}{[1 + (2\,\omega_L\, g\, \tau)^2]^{1/2}}$$

where $\omega_L = eH_0/2mc$. We can also think of the Hanle effect in terms of the Zeeman diagram in figure 1 if we remember that associated with each energy level is a finite radiation width. Therefore a small field H_0, inversely proportional to the lifetime, has to be applied before the degeneracy of the magnetic sublevels is removed. This small-field region is where the polarization signal is found to vary, and the width of the curve once again yields the lifetime τ. Rotation of the polarization direction can also give this result.

The level-crossing technique[14] relies on the property that two excited fine- or hyperfine-structure magnetic sublevels may cross, or become degenerate, at certain magnetic fields. If these two levels, say B and C, differing for example by $\Delta m = 2$, can be excited by the same photon from a lower state A, then with the polarizations of exciting and emitted light given by $\mathbf{e}_1$ and $\mathbf{e}_2$, the observed radiation intensity is

$$I \propto |(A\,|\,\mathbf{e}_2\cdot\mathbf{r}\,|\,B)(B\,|\,\mathbf{e}_1\cdot\mathbf{r}\,|\,A)|^2 + |(A\,|\,\mathbf{e}_2\cdot\mathbf{r}\,|\,C)(C\,|\,\mathbf{e}_1\cdot\mathbf{r}\,|\,A)|^2$$

when the levels do not cross. When B and C cross, the radiation from these two states becomes coherent

$$I \propto |(A\,|\,\mathbf{e}_2\cdot\mathbf{r}\,|\,B)(B\,|\,\mathbf{e}_1\cdot\mathbf{r}\,|\,A) + (A\,|\,\mathbf{e}_2\cdot\mathbf{r}\,|\,C)(C\,|\,\mathbf{e}_1\cdot\mathbf{r}\,|\,A)|^2$$

and a change in intensity is observed. The resonant value of H_0 at which the level crossing occurs, together with a knowledge of the g values can be used to calculate the zero-field level structure. As for the Hanle effect, the width of the resonance, obtained by varying H_0 about the crossing field, gives τ. In fact the Hanle effect can be considered as a level crossing at zero field.

Both the Hanle effect [5, 15] and level-crossing experiments[16] have been successful with collimated-electron excitation. The direction of the electron beam again takes the place of the polarization of the exciting light. Jean-Pierre Descoubes (Ecole Normale Supérieure) and his colleagues have obtained extensive theoretical and experimental results for the lifetimes and fine structure of excited helium levels.

Two experiments have extended these studies to ions. H. Bucka (Technische Universität, West Berlin) uses electrons to ionize a barium atomic beam. After excitation of the ion with its resonance radiation a Hanle-effect experiment permits measurement of the 6 $^2P_{3/2}$ excited-ionic-state lifetime. Since ion density is affected by variations in magnetic field, suitable corrections are required to interpret the results. In the second experiment[17] Alan C. Gallagher (University of Colorado, Boulder) produces the ions of group II elements by introducing traces of them into an argon discharge. The Hanle effect can then be studied, again with the use of the ion resonance radiation. The interesting fact is that the cross sections for the ion and the atom in depolarizing collisions with the noble gas are of the same order of magnitude, 10^{-14} cm^2.

M. Lombardi (University of Grenoble) aligns the excited atomic states in a high-frequency discharge set up by planar electrodes outside the discharge tube. This enables experiments with elements that either attack electrodes or are difficult to vaporize. An added advantage is that there are no electrodes to disorient the atoms. Preliminary Hanle-effect and magnetic-resonance experiments have been done to test the method. D. Kent Anderson (Montana State University, Bozeman) has extended the level-crossing work into the vacuum ultraviolet with both optical and electron excitation.

Laser level studies

With a novel Hanle-effect experiment using laser excitation Bernard Decomps and Michel Dumont (Ecole Normale Supérieure) study excited neon levels. A 5-cm-long discharge tube contained in an axial magnetic field is placed in the optical cavity in series with the much longer (115 cm) laser tube. Fluoresence from the side of the short discharge tube is observed with a monochromator. Because of the Brewster-angle windows on the tubes the light is linearly polarized and suitable for Hanle-effect studies. These are done with both 3390-nm and 632.8-nm laser radiation and yield, for example, a zero-power Hanle width of 8.5 MHz for the $3s_2$ level. The width increases appreciably with laser intensity. This effect is similar to rf broadening in magnetic resonance. Variation of relative fluorescence intensity due to laser irradiation is found to vary with magnetic field and traces out a Doppler curve with a depression at the center. This is interpreted as a "magnetic Lamb dip."

Modulation and pulse techniques

Pebay-Peyroula, who gave a critical appraisal of electron-excitation spectroscopy at Grenoble, summarized gen-

eral principles underlying the experiments in terms of atomic magnetization. Odette Nedelec (Grenoble), who has just completed a detailed theoretical study of Zeeman coherence phenomena in atomic vapor excited by slow electrons,[18] says that all these resonance phenomena (shapes of resonance curves) essentially depend only on polarization of the light emitted at zero field. Thus they do not depend on the type of excitation process or the angular momentum of the level. Two general cases are recognized: (1) the magnetic field H_0 is parallel to the electron motion; symmetry is maintained, and there is no transverse magnetization; (2) H_0 is perpendicular to the electrons, and a transverse magnetization is produced. This condition is referred to as "Hertzian coherence."[19] A macroscopic transverse magnetization is in fact fundamental to the observation of the Hanle effect and of the level crossings. It is also the basis of the modulated and pulsed experiments.

Kastler[20] has suggested the possibility of observing intensity changes in the light beam due to excited-state resonance in a magnetic field when the atoms are excited by light pulses (or modulation) at the Larmor frequency. Under this condition the excited-state spins precess coherently. Such an experiment would be analogous to the work of William E. Bell and Arnold L. Bloom[21] who excited coherently the *ground*-state spins in an optical pumping experiment by modulating the pumping light. Resonance was observed by them in the transmitted light when the modulation frequency equalled the Larmor frequency of the ground-state spin. In relation to many of these experiments it is interesting to note that Nedelec has shown[22] that Hertzian coherence can be transmitted from one atomic state to another by cascade processes.

Two extensive calculations make Kastler's predictions[23] quantitative and thereby again permit extraction of the excited-state lifetime. Recently M. I. Dyakonov extended the theories to arbitrary angular momenta (from those represented in figure 1), first with the assumption of an exciting spectral linewidth much larger than the Zeeman splitting but smaller than the hyperfine structure,[24] and then with the linewidth larger than the hyperfine structure.[25] The latter case is interesting because in addition to finding resonances at ω_L (sometimes $2\omega_L$, $\omega_L/2$), resonances are also expected at the hyperfine transition frequencies in the excited state. This may be a useful result in cases of optically unresolved structure.

Experimental results have been achieved with both modulated light[26] and electron[27] beams. Pebay-Peyroula has pointed out an important advantage of modulated excitation over the usual rf technique: The rf power is not tied to the lifetime of the atom (in the magnetic-resonance experiment the shorter the lifetime, the more power is required) and very short lifetimes can be measured. Also in contradistinction, the resonance width is independent of the rf level. Disadvantages, on the other hand, are more complicated electronics, generally lower signal-to-noise ratios and difficulties above about 50 G with the electron beam that is perpendicular to the magnetic field. There are also possible errors from the "trivial modulation" of the total light signal. Because of this possibility another scheme of Kasler,[21] magnetic field modulation parallel to H_0 (rather than perpendicular as in figure 1), has an advantage. Theoretical and experimental results have also been obtained for this "parametric-resonance" technique.[28]

Tetsuo Hadeishi (Lawrence Radiation Laboratory) has observed[29] modulation of the exponential decay that follows a pulsed coherent electron excitation of the $m = +1$ and $m = -1$ levels in the $7\,^3P_1$ state of cadmium. Moreover he has described the coherent excitation of mercury $^3P_2$ magnetic sublevels observed as light beats in the absorption of resonance radiation from this state.[30] A curious effect was also reported by him: It appears that a change in alignment of xenon $^3P_2$ atoms occurs when what are believed to be aligned xenon $^2P_{3/2}$ ions are subjected to rf resonance. A possible "exchange" type collision between these states has not been explained.

Direct lifetime measurements

Finally several direct methods for measuring lifetimes were presented. Jules Z. Klose (National Bureau of Standards) performed lifetime measurements for noble gases with fast delayed-coincidence techniques.[31] The excited gas atoms are produced by a sharp pulse of low-energy electrons (very near threshold to avoid cascade processes), and the decay rates are measured with a monochromator. Repeated results are accumulated in multichannel analyzers. Measurements have been obtained in the 10-nsec region to better than 10% accuracy. Thomas M. Holzberlein (Principia College, Elsah, Ill.) on the other hand, has constructed a pulse tube of sufficiently high current that the atomic decays can be observed directly on a fast oscilloscope.[32] C. Camhy-Val and Anne-Marie Dumont (Institut d' Astro-

Level Crossings Observable with Electron Excitation*

θ (deg)	$\Delta m = 2$	$\Delta m = 1$
$\theta = 90$	observable	not observable
$0 < \theta < 90$	observable	not observable
$\theta = 0$	not observable	observable

* Θ is the angle between the electron-beam velocity vector and the magnetic-field vector; Δm is the magnetic quantum number difference between the crossing levels.

physique, Paris), working with Evry Schatzman, are attempting to study lifetimes in the range 10–100 nsec by a photon coincidence technique. The measurement of the time delay between two successive photons will give the lifetime.

George M. Lawrence (Princeton University Observatory) described the phase-shift method of lifetime measurements.[33] The excitation of the atoms is modulated and the lifetime is obtained from the phase delay of the radiated light. Particular features of his experiments are modulated 200-V electron-beam excitation and the use of molecules that provide atoms by dissociation. This is expected to be a fast process ($\sim 10^{-12}$ sec) upon electron impact. The use of molecules prevents imprisonment of resonance radiation. The studies are done by varying the modulation frequency over a wide range (0.54–54 MHz) and the data on variation of phase shift with frequency permit corrections to be made for cascade processes. The system resolution is ± 0.2 nsec, with $\pm 15\%$ maximal systematic errors.

We may illustrate here one more recent direct technique by Raymond H. Hughes and coworkers.[34] A fast proton beam (tens or hundreds of keV) traverses a gas in which charge exchange takes place, leaving hydrogen in excited states. The radiated-light intensity is observed as a function of distance (through tens of centimeters) giving the decay curve.

In conclusion we must note that in many of these experiments resonance radiation is involved and imprisonment effects must be taken into account. These are discussed by T. Holstein.[35] More appropriate to phenomena in which decay of polarization is observed, *coherent* multiple scattering of resonant photons must be included in the theory of line shape. This was discussed first by Marie-Anne Guiochon, Jacques E. Blamont and Brossel[36] and treated in detail by Barrat.[37] Extensions to more general cases of irradiating linewidth and Zeeman splittings have been made more recently by Alain Omont, and Dyakonov and V. I. Perel.[38] In the density regions where pressure broadening may still be unimportant, coherent scattering can be thought of as transferring the wavefunction properties from one atom to another, effectively lengthening the lifetime (coherence narrowing). Finally, for the electron experiments Bréchot (Observatoire de Paris, Meudon), commenting on her theoretical work with H. Van Regermorter, pointed out that the broadening of the spectral line by electron impact must also be considered in the interpretation of line breadth. □

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