New techniques of hyperfine spectroscopy

High-resolution atomic spectra can give us detailed information about unstable nuclei, including angular momenta, electric and magnetic multipole moments and variation in charge radii as a function of neutron number.

Olav Redi

The decade of 1965–1975 saw a revolution in optical spectroscopy, culminating in the introduction of lasers into high-resolution spectroscopy. Several laboratories immediately put the dye laser to good use for nuclear physics, by using it to measure hyperfine structures and isotope shifts. These developments have produced new, interesting, and at times unexpected, results. Currently, the main motivation of these studies is to understand the behavior of the nuclear charge distribution as the number of neutrons is varied. The required data on isotope shifts for long chains of stable and radioactive species, which until recently existed only for mercury and thallium, are now becoming available for other elements. Experiments are now extending the isotope-shift measurements by "off-line" techniques to nuclei with half lives greater than about 10 minutes, and by "on-line" techniques with accelerators and isotope separators to nuclei with half-lives in the second and sometimes millisecond ranges. The following article by Hans Schuessler concentrates on on-line techniques applied to nuclei far from the region of stability.

To put the recent innovations in perspective, I will trace the development of some of the experimental techniques for work with radioactive atoms, emphasizing isotope-shift measurements. For the purpose of illustration I will draw on the work with heavy atoms on which we have concentrated at New York University. Pierre Jacquinot and Robert Klapisch have recently published an excellent comprehensive review of this subject.[1]

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Nuclear size effects

We can distinguish several kinds of nuclear effects on atomic spectra

- The monopole part of the Coulomb interaction (shielded by inner electrons), of course, dominates the spectrum.
- Nuclear electric and magnetic multipoles produce effects smaller by five or six orders of magnitude; they give rise to the hyperfine structure of the spectrum.
- The finite size of the nuclear charge distribution and the nuclear motion about the center-of-mass gives rise to differences of the monopole interaction from isotope to isotope on the same order as the hyperfine interactions; that is, they give rise to isotope shifts in the spectra.

Figure 1 illustrates the hyperfine structure and isotope shift in the case of the 5350-Å line in the spectrum of thallium for the two stable isotopes (masses 203 and 205) and two radioactive isotopes (masses 199 and 200). Here the isotopes with odd numbers of nucleons have very similar hyperfine structures, which are shifted with respect to each other by the isotope shift. The magnetic moment of thallium-200, however, is only 0.04 nm (compared to 1.6 nm for the three others) so that the hyperfine structure is unresolved.

Optical spectroscopy is limited in its ability to resolve hyperfine structure, which is often, particularly for light elements, small relative to the line width; the extremely small moment of thallium-200 is an exception. This limitation has been overcome some years ago by measuring the hyperfine structure directly using rf-resonance techniques. For isotope shifts, on the other hand, we have had to await the recent methods discussed below to make possible similar improvements.

The isotope shift consists of a shift of the centroid of the hyperfine levels between isotopes of the same element. As I mentioned, it arises both from the finite size of the nuclear charge distribution, which differs between isotopes, and the fact that the nucleus partakes in the motion of the electron–nucleus system. I will mostly ignore these effects of the nuclear mass, because they are unimportant for heavy elements, and consider the charge-distribution, or volume, effect. To a good approximation, this part of the isotope shift in an atomic transition is proportional to the difference of the mean square nuclear charge radius $\delta\langle r^2\rangle$ between isotopes and the change of the electron density at the nucleus $\Delta|\psi(0)|^2$ during the transition. The electron density at the nucleus arises mostly from the s-electrons and to a much smaller extent from relativistic $p_{1/2}$ electrons. It is important to note that the high-precision techniques of radio-frequency spectroscopy are not applicable to measurements of the isotope shift because two atomic levels for which $\Delta|\psi(0)|^2 \neq 0$ are generally separated by an energy corresponding to the optical or higher-frequency region. (An exception to this is the Lamb-shift transition in hydrogen).

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Before continuing the discussion of the optical spectrum it is worth mentioning two other spectroscopic methods that have been used to measure the nuclear charge distribution:

- x-ray spectroscopy for lines corresponding to inner-shell transitions, and
- x-ray spectroscopy for mu-mesic atoms

The values of $\Delta|\psi(0)|^2$ for inner-shell x-ray transitions are several orders of magnitude larger than those of the optical transitions. The instrumental line widths and broadening due to the short lifetimes of the atomic states involved, however, are such that even under the best conditions the isotope shifts are less than 0.1% of the line width. Hence, though there is no improvement over optical studies in the accuracy of isotope shifts measured with x-rays, there remains the advantage of more reliable extractions of $\delta\langle r^2\rangle$, as we are dealing more nearly with a one-electron problem. The inner-shell x-ray measurements, which have been done only with stable isotopes, can thus be useful in calibrating $\Delta|\psi(0)|^2$ for the optical spectrum.

Because they are heavier than electrons, the orbits of muons are much smaller; muons are also not excluded from the inner orbits of the atom. For a heavy element the probability is nearly one half for the mu meson to be inside the nucleus. Though the isotope shifts are therefore large, these experiments are, like the other x-ray experiments, limited to stable targets and wide lines, with added problems arising from possible nuclear excitations and from separating the contribution of higher moments of the charge distribution from $\delta\langle r^2\rangle$. On the other hand, the spectroscopy of mu mesic atoms determines not only $\delta\langle r^2\rangle$ but also $\langle r^2\rangle$ and is applicable to lighter elements than the electronic x-ray technique.

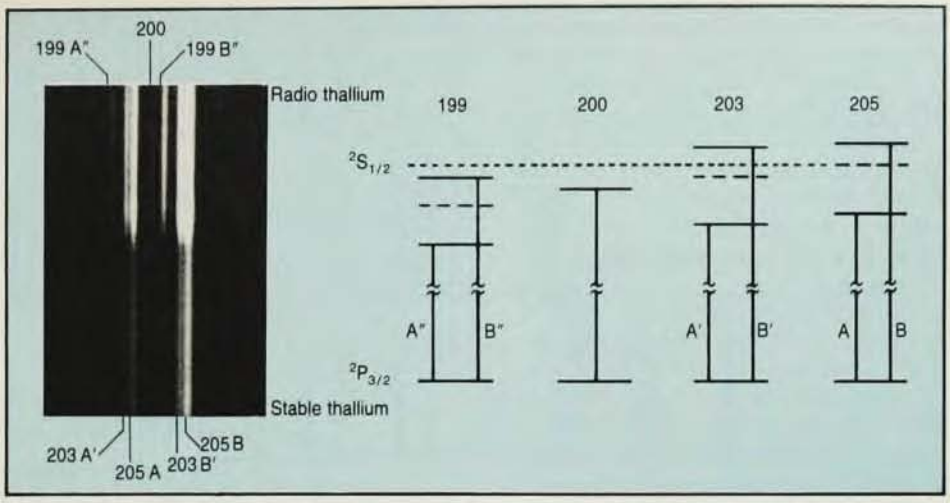
A non-zero electron charge density at the nucleus also gives rise to quite a different nuclear-size effect observed in precision measurements of the hyperfine structure. It is a result of the interaction of the penetrating electron with an extended nuclear magnetization. The minute differences of this interaction from that involving a point nuclear dipole is known as the "hyperfine anomaly," and was first calculated by Aage Bohr and Victor Weisskopf.[2] The new optical methods open up further possibilities for such measurements, which have already been of value in the theory of nuclear magnetism.[3]

Doppler broadening

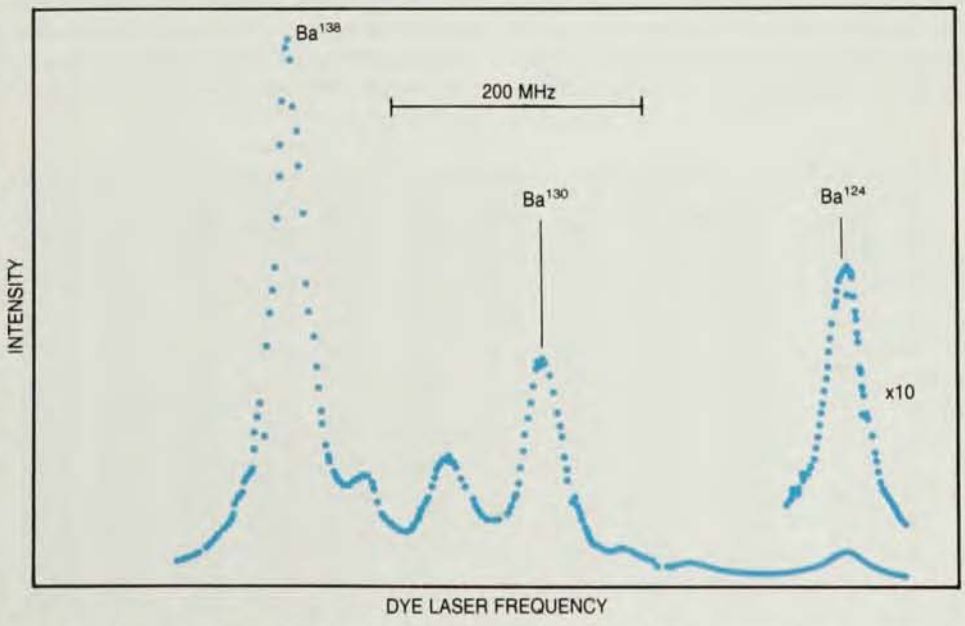
Albert Michelson and others used interferometric measurements, to demonstrate, even before 1900, the existence of structure in atomic lines orders of magnitude finer than "fine structure." Accurate measurements of these nuclear effects in the optical spectrum, which were of course not understood at the time, proved to be difficult, however. The problem is that the hyperfine splittings and isotope shifts are characterized by fractional changes in wavelength that can be as small as on the order of 10^{-6} (that is, a few milliangstroms in the visible part of the spectrum) and therefore spectrographic apparatus of high resolving power (on the order of 10^6) and light sources of narrow line width are required. Spectroscopists made significant progress toward satisfying these requirements, first by the use of Fabry–Perot interferometers; then, in the 1930's, Hermann Schüler introduced the hollow-cathode lamp, and Derek Jackson and Heinrich Kuhn developed the atomic-beam light source or absorber. These sources were designed to minimize the Doppler broadening due to thermal motion of the atoms. The fractional Doppler width of a spectral line for a gas of atoms is

$$\Delta\lambda/\lambda = 7.16\times10^{-7}\ (\text{kelvin})^{-1/2}\sqrt{(T/A)}$$

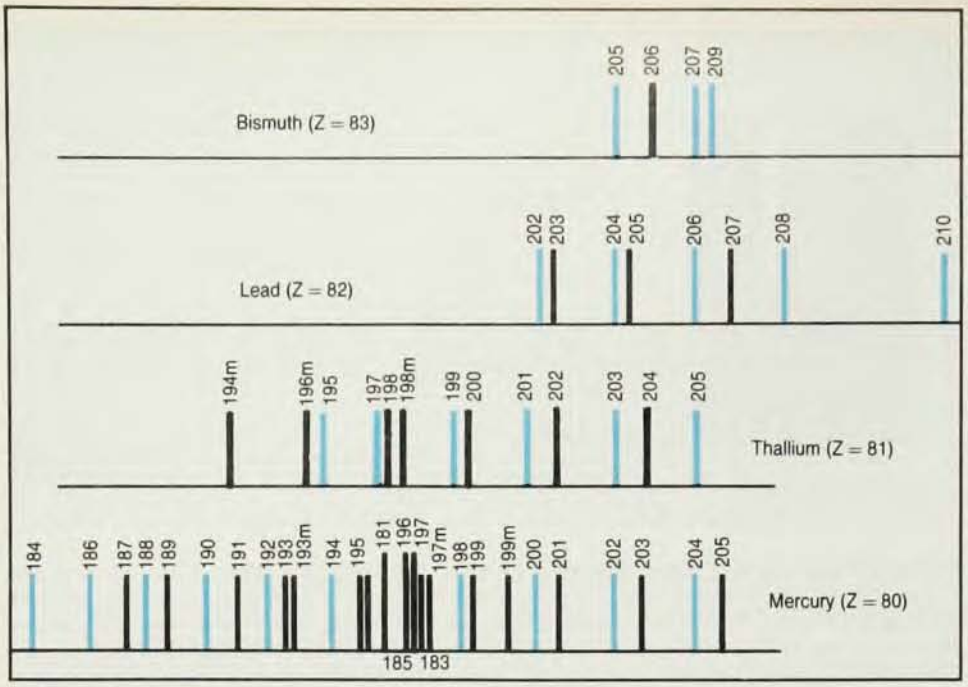
where T is the absolute temperature



Isotope shifts and hyperfine structure in the 5350-Å line of thallium. The photo at left shows the spectra, obtained with a grating spectrograph from a lamp containing radioactive thallium produced in a cyclotron (upper portion of photo) and from a lamp containing stable thallium (lower portion of photo). The level diagrams at right show the transitions involved. The dashed lines indicate the energy of the level in the absence of hyperfine splitting, clearly showing the isotope shift. (From reference 5.) Figure 1



Laser-excited fluorescence spectrum of barium observed in a collimated atomic beam. Barium-124 has a half life of 12 minutes; barium-130 was introduced as a reference, and the remaining peaks are from contamination by natural barium in the beam. A flux of only 500 atoms/sec crossed the laser beam during the measurement, which lasted nearly an hour and consumed 10 picograms of Ba^{124}. The residual Doppler width corresponds to an effective temperature 0.14 K and is comparable to the natural line width due to the finite lifetime of the atomic excited state. (The data were obtained at the Kernforschungszentrum Karlsruhe; see also reference 6.) Figure 2



Relative isotope shifts for mercury, thallium, lead and bismuth. The horizontal scale is a measure of the mean-square charge radius $\langle r^2 \rangle$ of the nuclei. Note the regular spacing of the isotopes with even neutron number and the irregular positions of the odd isotopes; mercury-181, 183 and 185 are an exception to the pattern; the exception arises because of a sudden change in nuclear deformation at these mass numbers.[13] For easier comparisons we have aligned isotopes with $N = 122$ and 124. Figure 3

and A the mass number. In the hollow cathode, Doppler broadening is reduced by cooling the emitting atoms to near liquid-nitrogen temperatures (78 K). The atomic-beam device achieves radically lower effective temperatures (less than 1 K) because one observes the spectra in a direction perpendicular to the beam. These methods to reduce linewidths remained the key to precise measurements of isotope shifts until the advent of laser spectroscopy. Of course, for hyperfine structure, Doppler-free measurements were already made possible in the 1930's with I. I. Rabi's invention of radiofrequency spectroscopy.

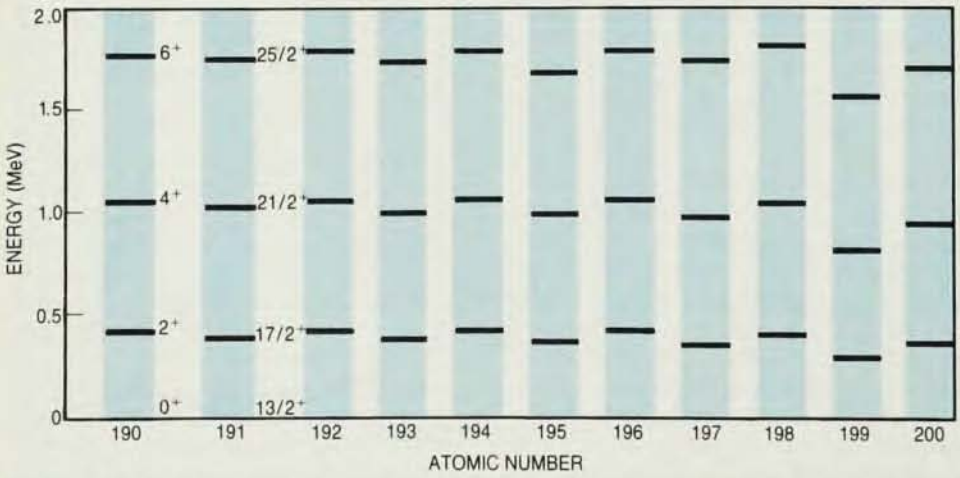
High-resolution optical spectroscopy, with these line-narrowing methods, is not directly applicable to work with small quantities of radioactive material produced in accelerators (typically on the order of 10 nanograms). Fortunately, as illustrated in figure 1, even uncooled lamps yield useful results for heavy elements. This is what made the electrodeless lamp the work-horse for early radioactive atom spectroscopy: It produces intense lines with microscopic amounts of material. The use of such light sources for mercury and thallium by Francis Bitter's group[4] at MIT, begun during the 1950's, made possible the first isotope shift measurement for accelerator produced radioactive atoms. Figure 1, in fact, is an example of a spectrum obtained in these early isotope-shift studies.[5]

For the lighter elements, however, the situation is less favorable. As the mass decreases the Doppler width increases; on the other hand, the electron penetration probability, the nuclear charge and the charge radius decrease. The result of these changes can be seen by comparing the ratio of that part of the isotope shift due to the nuclear charge volume (it is often called the "field shift") to the room-temperature Doppler width for three elements from widely different mass regions: mercury ($Z = 80$), barium ($Z = 56$) and calcium ($Z = 20$). Typical values of the ratio of field shift to Doppler width for these three elements for a change of two neutron numbers are: 5, 0.2 and 0.05. Clearly, Doppler broadening makes it difficult or impossible to measure the isotope shift for the medium and light elements. The recent laser methods, which make it possible to reduce the Doppler width dramatically—or even eliminate it—in work with radioactive atoms, thus play an essential role for these elements.

The recent experiments can be classified into three categories according to the Doppler width reduction technique used:

- Methods that use a thermal beam of radioactive atoms illuminated with a laser beam perpendicular to the atomic velocity. This is based on the earlier idea of achieving a low effective temperature for the atoms by transverse observation, but also includes new, highly sensitive detection techniques, which I will discuss below. An example[6] is shown in figure 2.
- Collinear spectroscopy, in which a fast beam of radioactive atoms is illuminated in a direction collinear with the beam.[7] This technique relies on the fact that atomic velocities in an accelerated and neutralized beam are bunched in a non-thermal distribution; it is described in detail by Schuessler in the following article.
- Nonlinear spectroscopic methods (saturation, two-photon absorption, and other such techniques) performed on atoms in a cell. So far there have been few applications to radioactive nuclei. These techniques were described by Theodor Hänsch in PHYSICS TODAY in May 1977, page 34.

A universal attribute of the spectroscopic experiments that I shall discuss here is that they are applicable to very small samples (less than about 10^{-7}



Nuclear energy levels for the low-lying high-spin states in mercury isotopes with $A = 190$ to $A = 200$ showing the decoupled bands. All the even isotopes have the same spin sequence as mercury-190 and the odd ones have the same spins as for mercury-191. The regularity of the patterns suggests that the odd neutron orbits a core containing an even number of nucleons (whose spins states are $I = 0,2,4...$), adding its orbital plus spin angular momentum of 13/2 to the spin of the core. Figure 4

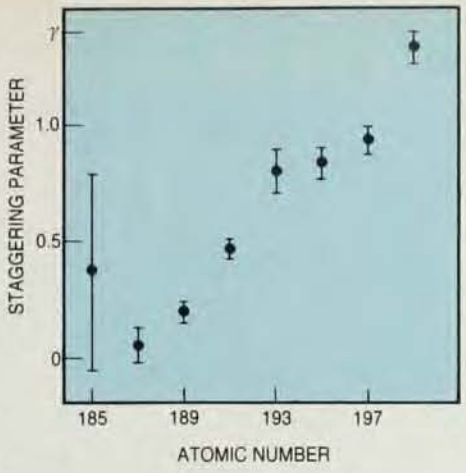
grams). Some of these experiments use only techniques from atomic physics, but with special procedures for handling the small samples to achieve the required sensitivity. In these experiments nuclear radiation from the sample plays no role—except as a possible hazard! In other experiments, on the other hand, nuclear radiations play a crucial role in that they are used for the detection of optical or radiofrequency resonances.

Radiofrequency spectroscopy

Let me first digress somewhat from the main subject of optical methods and review briefly the development of radiofrequency spectroscopy. Spectroscopy of radioactive atoms in the rf range matured considerably earlier than its optical counterpart and to some extent provided an example to follow. Here Rabi's atomic-beam magnetic-resonance technique played an essential role. In its basic form, and through early developments,[8] it offered the possibility of high precision, sensitivity, isotope selectivity, and applicability to a wide range of atoms. Several of these developments remain essential to the present:

- The introduction of the mass spectrometer and of ion counting for the beam detection. This allows one to measure an isotope with an abundance on only 10^{-5}, as well as a number of radioactive isotopes. Jerrold Zacharias introduced this technique in 1948.
- The detection of the radioactive atomic beam through nuclear radiation, a technique pioneered in the early 1950's by E. H. Bellamy and K. F. Smith in Cambridge, Leonard Goodman, Solomon Wexler and William Childs at Argonne, Victor W. Cohen at Brookhaven, William A. Nierenberg and his group at Berkeley, and Donald R. Hamilton's laboratory at Princeton. This technique permits one to do experiments on radioactive isotopes in the presence of at least a million-fold greater number of stable ones.
- The nearly immediate use in the experiment of the radioactive atoms being produced, without separate processing ("on line" spectroscopy). These experiments were initiated in the early 1960's in Princeton. For example, the spectrum of sodium-21, whose half life in 23 seconds, was measured in this manner.[9]

Norman Ramsey describes the earlier period of atomic-beams work, the research prior to the mid-1950's, in his classic book *Molecular Beams*.[10] In the early 1950's Francis Bitter at MIT extended the rf spectroscopy of radioactive atoms to involve the excited states of atoms, and importantly, the optical detection of the resonances through variations in absorption or fluorescence intensity. This new class of experiments included[4] the "double resonance" (optical plus rf) method invented by Bitter and Jean Brossel, Peter Franken's level-crossing technique, and Alfred Kastler's optical pumping. In the optical-pumping experiments one can orient the nuclei by using circularly polarized light; one can then detect rf resonance by observing the angular distribution of the nuclear radiation. Ernst Otten and his colleagues at Heidelberg, and later Mainz, have fruitfully exploited this technique, called nuclear-radiation-detected optical pumping.[11] Hans Kopfermann has described the development of these spectroscopic studies of hyperfine structure and isotope shifts, from the earliest days through 1957, in his book *Nuclear Moments*.[12] Kopfermann inspired a great activity in this field in his laboratory in Heidelberg. Since those early efforts, radiofrequency experiments have reached a high level of sophistication as exemplified by the experiments performed on-line at CERN by the Gothenburg–Uppsala groups of Ingvar Lindgren and Curt Eckstrom. They succeeded in measuring rf resonances of isotopes with half lives as short as 0.38 sec.



Odd-even staggering parameter γ for the nuclear isomers of mercury having spin $^{13}/_{2}$, as a function of mass number. The behavior shown is consistent with the model described in the text and in figure 4. (See also reference 17). Figure 5

Chains of isotopes

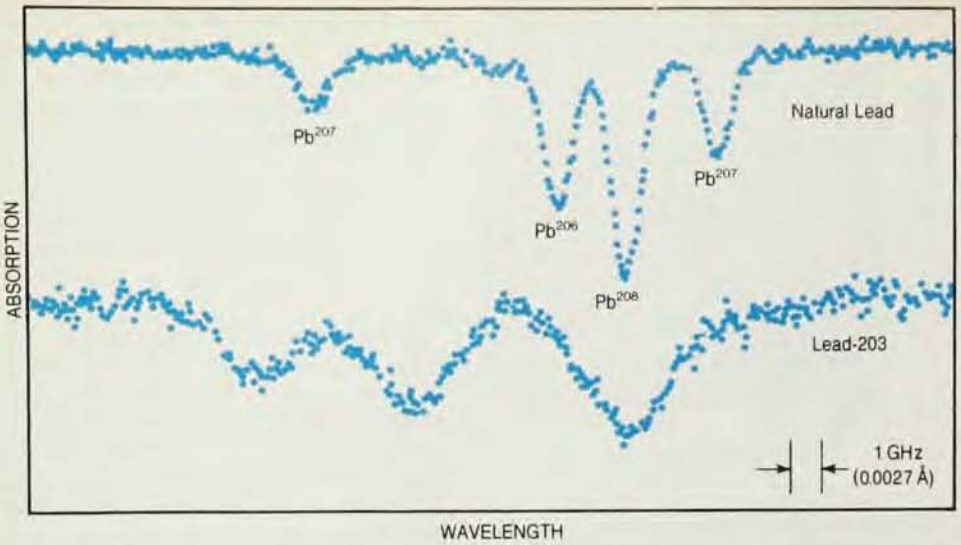
Concurrently with the development of radiofrequency techniques, methods of doing optical spectroscopy with minute quantities of radioactive material were explored in Bitter's laboratory. This effort culminated in the first recording, in 1954, of the optical spectrum of an isotope produced in a cyclotron, mercury-197, whose half-life is 65 hours. This was a significant achievement, because it set the stage for a systematic study of isotope shifts for chains of isotopes. In fact, soon after this first experiment, the group, following a suggestion of Richard Weiner, performed a very interesting measurement of a nuclear *isomer* shift in the optical spectrum for a metastable nuclear state of mercury-197 (24-hour half life). It showed that the change in nuclear radius resulting from an excitation of the nucleus into the isomeric state is comparable to the change induced when a neutron or a proton is added.

These successful experiments provided the impetus for studies of isotope and isomer shifts extending over many mass numbers of mercury, thallium, lead and bismuth. This work was first carried out in Bitter's laboratory under direction of Henry Stroke who later continued this work at his New York University laboratory. Some of the early work was also carried out by Sumner Davis, at the University of California in Berkeley. More recently, Otten has obtained[13] data on very light mercury isotopes, $A \leqslant 193$, and at New York University we have worked with mercury, lead and bismuth. These experiments have yielded the first detailed data for chains of isotopes of neighboring elements; they also make possible isotonic comparisons. Figure 3 shows the isotope shifts that have been obtained.

The most striking feature of the results shown in figure 3 is the lack of regularity displayed by the nuclei with an odd number of neutrons. Specifically, when a neutron is added to a nucleus with an even number of neutrons, the change in the mean-square charge radius is generally less than half of that obtained by adding two neutrons. This effect is called "odd–even staggering" and has been known from work with stable isotopes for a long time. The irregularity of the isotope shifts is usually characterized by the odd–even staggering parameter[14]

$$\gamma = 2\frac{\langle r^2\rangle_{N+1} - \langle r^2\rangle_N}{\langle r^2\rangle_{N+2} - \langle r^2\rangle_N}$$

where N refers to an isotope with an even number of neutrons. This parameter can be interpreted in terms of the relative distortion of a nucleus, N, by an added neutron. Gerald Brown and Roger Barrett calculated[15] this nuclear polarization for different angular momenta of the added neutron and for various nuclear interactions. The comparison of the results with the observed isotope and isomer shifts allowed them to discriminate among different possible interactions. Somewhat earlier, Jean Blaise had suggested a qualitative explanation of the staggering in the light of individual nucleon orbits being filled as N increases. When one takes into account the spins of various nuclei, the results shown in figure 3 lend support to his interpretation. At the same time, however, the



Absorption lines of radioactive lead-203 in a cell (bottom) and a beam of natural lead (top) at 2833 Å. The position of the three hyperfine components of Pb^{203} yield a magnetic-moment value and an isotope shift. These data, resulting from 64 spectrum scans, were obtained at New York University with our high-resolution grating spectrometer and a digital signal-averaging system. (From reference 23.) Figure 6

data also reveal some remarkable consequences of collective effects.

As an example, let us consider the data from figure 3 on the $I = 13/2$ mercury isomers that can illustrate attempts at a unifying explanation of the behavior of optically measured staggering parameters γ and the systematics of nuclear energy levels. Dieter Proetel and his collaborators at Berkeley have found striking regularities in studies of energy levels of neutron-deficient mercury nuclei.[16] As figure 4 shows, these regularities are apparent in a comparison of the lowest energy levels (spin $I = 0,2,4,6$) of even-N nuclei with some of the lowest levels with $I \geqslant 13/2$ of the odd-N nuclei. In this diagram the $I = 13/2$ states (which are not the ground states) are shifted to match the $I = 0$ levels for purpose of comparison. The close resemblance of the energy levels for even-N nuclei and the levels of the odd-N nuclei with the spin–parity sequence $^{13}/_2{}^+$, $^{17}/_2{}^+$, $^{21}/_2{}^+$, $^{25}/_2{}^+$ is exactly what is expected from a simple model in which the odd particle orbits a rotating, deformed core having an even mass number and for which the Coriolis effect is large. The angular momenta of the core and of the particle become aligned and the resulting angular momentum for the system is the sum of the core spin ($I = 0,2,4,6...$) and the angular momentum of the single particle, in this case 13/2. The group of states is referred to as a "decoupled band." Frank Stephens developed this model; Stroke, Proetel and Jürgen Kluge have used[17] it to interpret the regular decrease of γ with mass number, as shown in figure 5. They have found that combining this model with the interpretation of γ in terms of a nuclear-core polarization gives a consistent picture. There is as yet an unexplained jump in both γ and the nuclear energy levels at Hg^{199}. On the whole, odd–even staggering is still very much an open problem in nuclear physics.

New methods in spectroscopy

Let us now turn to a discussion of new spectroscopic methods which have the potential to increase tremendously the usefulness of isotope-shift measurements as a tool for nuclear structure studies. These new methods have already, over the last decade, contributed about one half of the data shown in figure 3, in particular for the short-lived mercury isotopes. They can be categorized as:

- The use of digital control and processing of the signal to improve dispersive spectroscopy with diffraction gratings or interferometers.
- The application to optical spectroscopy of methods directly analogous to those we discussed for rf studies of radioactive atoms to increase sensitivity and selectivity.
- Use of tunable dye lasers to produce great gains in resolution and sensitivity of the experiments done with the preceding methods, as well as to permit radically new types of experiments.

In our own work we used the first of these developments to study the effect of the closed proton shell ($Z = 82$) on the odd-even staggering. This meant an extension of the spectroscopic studies to the isotope shifts in lead. Our approach in developing an off-line technique suitable for the measurement of half-lives as short as a few hours was to improve the sensitivity and precision of the detected spectrum by replacing lamps with absorption cells and using photoelectric detectors (with multiple scanning and digital signal averaging of the weak signals) instead of a photographic plate. From the early days of the spectroscopy of radioactive atoms the problem of rapid loss of the few available atoms to the walls of the discharge lamp severely limited the experiments. In an absorption cell this problem is reduced, so that even 10^{10} atoms have been found sufficient to record a spectrum. Substantial improvement made in the isotope separation of the radioactive samples, done in collaboration with Robert A. Naumann and the Princeton University Cyclotron group, have also played an important role in improving the isotopic purity, crucial for spectroscopic work. Figure 6 shows a recording of an absorption spectrum of Pb^{203}, whose half life is two days. This particularly nice spectrum does not display the difficulty one frequently encounters, that the weak components of radioactive atoms are masked by much stronger ones belonging to stable isotopes.

Radiofrequency methods—atomic-beam experiments and optical pumping—had already provided the ability to work in the presence of overwhelming quantities of stable isotopes. In fact, these techniques, which we discussed earlier, suggest three useful adaptations to optical spectroscopy for radioactive atoms:

- nondispersive measurement
- nonoptical detection
- on-line operation with an accelerator and isotope separator.

I contrast usual dispersive optical spectroscopy and nondispersive measurements of an optical transition frequency: In the latter the frequency of a nearly monochromatic light source is scanned over optical resonances of a sample under study, and one records some parameter related precisely to the scanning frequency. (Alternatively, one may shift the energy levels of the sample by applying magnetic or electric fields while keeping the light source fixed). The most obvious and direct way of detecting the optical resonances is by observing the fluorescence or absorption of light by the sample. This is the "magnetic-scanning" method, which was applied to isotope-shift studies of radioactive mercury in Bitter's laboratory. It is interesting to note that as early as 1929, Marcel Schein reported a number of atomic spectroscopic applications of magnetic scanning.

Nonoptical detection can provide important advantages for optical spectroscopy, as it does for the earlier schemes for detection of resonances without observations on the rf radiation. Thus, the radiation-detected optical-pumping technique of the Heidelberg group, in which angular distribution of nuclear radiation had served as an indication of rf resonance, could now be used for

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detection of optical resonance. In the latter case the light source is tuned through the optical resonance (magnetic scanning). Otten and his collaborators at Mainz have exploited this optical-pumping technique very fruitfully in on-line experiments to measure isotope shifts over the past ten years. Similarly, the radiofrequency transitions in an atomic-beam magnetic-resonance experiment can be replaced by optical transitions in the "C-field" region of the apparatus, as the upper part of figure 7 shows.

Using this method of detection, Richard Marrus and his coworkers at Berkeley, showed[18] in 1965 that the advantages of sensitivity and selectivity of a magnetic-resonance experiment with radioactivity detection, of the sort we discussed earlier, can be made to apply to measurement of the optical spectrum. This was a milestone in optical spectroscopy of radioactive atoms.

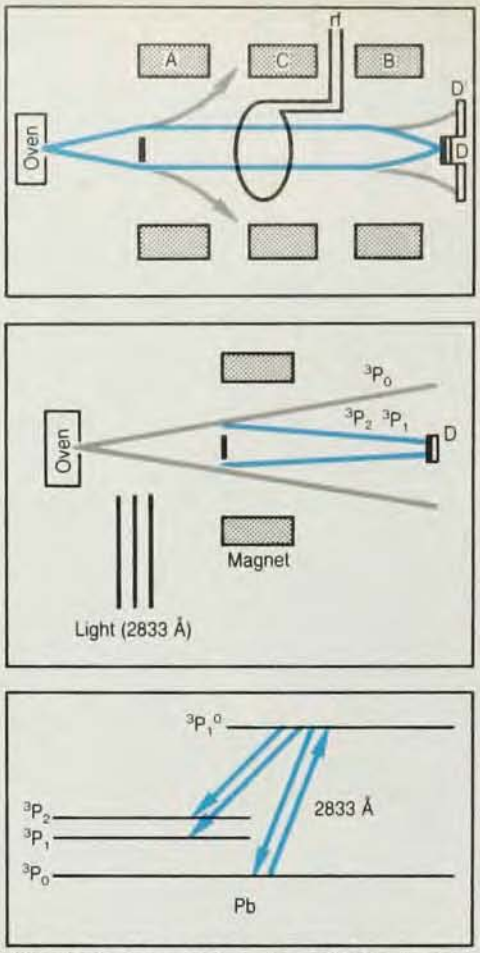
This pioneering work at Berkeley and Mainz played an important role in accelerating the development of optical spectroscopy of short-lived radioactive atoms: It demonstrated both the high sensitivity of nonoptical detection and the feasibility of efficient transfer of short-lived material from an isotope separator to the spectroscopic experiment. My own interest has been the adaptation of Rabi's method to study the isotope shift in very short-lived isotopes of lead. We developed the arrangement shown in the lower part of figure 7 for these experiments.[19]

After the demonstration of continuous operation of a dye laser in 1970, the art of constructing precisely tunable, single-mode devices improved rapidly. With this accelerating development, the dye laser became a very promising spectroscopic device for work with radioactive atoms. The groups at Orsay led by Jaquinot and by Klapisch were the first to apply, in 1975, the tunable dye laser to optical spectroscopy of radioactive atoms. Their experiment combined several recent developments such as nonoptical detection and on-line isotope production, together with the laser—a nearly ideal light source for nondispersive spectroscopy. Specifically, they used the atomic-beam optical-resonance technique together with beam detection, as in the scheme of Zacharias. This combination produced, in a relatively short time, a rich harvest of new results on radioactive alkali atoms ranging from sodium to francium.

An example of the evolution of the method of nuclear-radiation-detected optical pumping with lasers is the recent experiment in which Curtis Bemis and his coworkers measured[20] the isotope shift for a fission isomer with a 1 msec life. Instead of detecting beta or gamma rays, as in the older experiments, they observed fission fragments, replacing the cell that usually contains the sample by helium buffer gas into which the species under study recoils directly from the cyclotron target. This method of transfer of atoms from target to experiment avoids common problems of significant delay, such as atomic diffusion through liquids or solid targets, which can set a lower limit of about 1 sec on the lifetime of isotopes to be studied. Schuessler's article in this issue reviews the results of a number of these on-line experiments.

The high intensity and directionality of the tunable dye lasers has also increased tremendously the potential of nondispersive spectroscopy with optical detection. The experiments have involved measurement of laser-induced resonance fluorescence either from atoms enclosed in a cell or from atoms in a beam. Figure 2 shows an example of resonances obtained with the latter technique.

Gisbert zu Putlitz and his collaborators at Heidelberg have recently done[21] a particularly interesting set of experiments from the point of view of nuclear physics, using laser fluorescence to study calcium isotopes. This set completed the detailed study of variations in the nuclear charge radius for the entire nuclear $f_{7/2}$ shell, going from the magic number $N = 20$ to $N = 28$. Calcium-40 is a doubly magic nucleus (two closed shells); the difference in charge radius between Ca^{40} and Ca^{41} is very small, a result consistent with a recent Hartree–Fock calculation combined with shell model configuration mixing.[22] It is important to note that for these isotope shifts the preponderant origin is the mass effect; the variations in the nuclear charge radius contribute less than 10 percent. This small contribution could, however, be extracted reliably with the available data from mu-mesic isotope shifts.



Atomic-beam experiments. The diagram at top shows a modified version of Rabi's rf-resonance method. The six-pole magnets A and B focus or defocus the beam, depending on the magnetic state of the atoms; region C contains a homogeneous field. When the rf signal is in resonance with an atomic spin-flip transition, the radioactivity detected at the collectors D and D' changes. The lower diagram shows an analogous experiment that detects optical instead of rf resonances. The lead atoms emerging from the oven are in their ground state and have no magnetic moment; they are unaffected by the focussing magnet and do not reach the detector. Transitions at 2833 Å cause some atoms to be "pumped" into the $^3P_2$ and $^3P_1$ metastable states. These have a magnetic moment, and are focussed by the magnet. Thus the detector D records an increase of radioactivity when the light is tuned to the resonance. Figure 7

Future Directions

It is not surprising that the purely optical methods are becoming more widely used because of their simplicity compared with nonoptical detection techniques. In principle, with lasers the optical methods have an advantage in that some 10^8 photons per second can be scattered from a single atom, while radioactive detection gives only one count per atom. However, in the current beam-fluorescence experiments, the atom is illuminated only for a few microsec. Furthermore, purely optical methods can encounter severe background problems, which are particularly critical when there is spectral overlap from overwhelmingly abundant stable isotopes of the element under study.

As I have mentioned above, the potential of laser methods is only beginning to be explored. An extension of the laser work into the uv, where most atomic resonance lines lie, will obviously be important. The ability to measure the hyperfine structure not only in the ground state but for several excited states will open up the possibility for new studies of the hyperfine anomaly. As Josef Speth has pointed out, this can be expected to provide important new tests of the theory of nuclear magnetism.

Charge distributions of ground states of stable nuclei have been the subject of a great number of studies, especially after the first direct measurements, in the early 1950's, of nuclear charge

radii by elastic electron scattering and by x rays from muonic atoms. As compared to the detailed information available on the charge distribution for this small group of nuclei, very little information exists for the rest, which are not stable (a far greater number of nuclei). The optical methods have the potential, which is still to be fully realized, to explore this unknown territory.

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