Laser spectroscopy on-line with nuclear accelerators

Studies of the hyperfine structure and isotope shifts performed immediately after short-lived beta-unstable isotopes are formed in a high-energy collision yield rich data on the properties of nuclei.

Hans Achim Schuessler

During the last few years the study of highly unstable nuclei has made dramatic advances as a result of breakthroughs in experimental technique. As Olav Redi has discussed in the previous article, on page 26, electrons interact with the nucleus so that one can use atomic spectra to probe nuclear structures. The recent experimental developments have both the production of intense and pure beams of mass-separated isotopes, on the one hand, and sensitive and sophisticated techniques for the observation of these nuclei on the other. The basic difficulties in the experiments arise from the very small number of the unstable nuclei that are available even with modern on-line mass separators and from the short lifetimes of the nuclei.

The introduction of the laser in this field has increased the sensitivity of purely optical experiments by several orders of magnitude and has even made possible the detection of single atoms. In suitable cases, coincidence or photon-burst techniques have, in addition, strongly reduced the background noise. It is, of course, the spectral purity, the excellent beam quality and the high power density of lasers that have made possible this dramatic improvement.

At the same time that lasers entered the field, experimenters also developed techniques tailored specifically for measuring spectra "on line," that is, immediately after an unstable isotope has been produced (by an accelerator or reactor) and separated from other isotopes by a mass spectrometer. In this way one can observe spectra of even very short-lived isotopes (lifetimes as short as a millisecond). So far only a handful of true on-line experiments has been performed, but these have usefully extended our information on nuclear structures for long chains of isotopes.

Figure 1 is an example of the models the new data will be used to test. The cover shows a stylized view of the same results. The figure is a contour plot of the calculated total nuclear energy as a function of proton number (for selected neutron-deficient nuclei) and shape, as given by a deformation parameter, \( \beta \), which is positive for prolate (cigar-shaped), and negative for oblate (disk-shaped) nuclei. The deepest valleys are spaced 1 MeV. The calculations, based on a droplet model, were done by I. Ragnarsson of Lund University.
For more than a decade the Isotope Experiment (ISOLDE) at CERN, basing his computations on the droplet model and adding contributions from shell, pairing and Coriolis terms. One can determine the general pattern of nuclear shape and other features of nuclear structure with on-line laser spectroscopy by observing isotope and isomer shifts and the hyperfine structure of suitably chosen atomic states.

**Short-lived isotopes**

The possibilities of on-line research become apparent when one compares the number of stable and unstable isotopes. There are about 190 stable or naturally occurring isotopes. Spectroscopy with these nuclei has thus far provided the foundations and much of the data for the nuclear models we now use. The number of known unstable nuclei is close to 1600, and accurate data on hyperfine structure and isotope shift are available for only a few of these. Nuclear physicists estimate that several thousand beta-unstable nuclei with half lives longer than a millisecond remain to be discovered. It is likely that, as we continue to explore the unknown regions far from beta-stability, we will find new physical phenomena.

The first task is, of course, to produce the isotopes. The techniques differ for neutron-deficient and neutron-rich nuclei, and involve nuclear reactors, heavy-ion accelerators, or high-energy proton beams:

- Neutron-deficient nuclei have been produced by spallation from larger nuclei (induced by protons with energies larger than 100 MeV).
- Neutron deficient nuclei are also produced from fusion-evaporation reactions involving heavy ions accelerated to 5-10 MeV.
- Light, neutron-rich nuclei are produced by fission of heavy targets induced by neutrons (for example, in a reactor) or charged particles.

High-energy proton reactions have so far produced the largest yields of short-lived isotopes. In general, to be useful for on-line spectroscopy, a nuclear reaction must produce the isotope of interest in adequate quantities, and one must be able to bring a fairly pure sample of the isotope to the observation region in a time short compared with the nuclear lifetime. A less obvious requirement is for the isotopic purity of the sample: Usually it is not only the number of nuclei produced but also the background from neighboring isotopes that limits a particular experiment. For more than a decade the Isotope Separator On-Line (ISOLDE) at CERN, has been the leader in the field of on-

**Hyperfine interaction**

Once the isotopes have been made, one wants to observe the structure of their nuclei. Atomic spectroscopy provides one extremely useful probe of nuclear structure: The electromagnetic interaction of the valence electrons with the nucleus splits the fine-structure levels into hyperfine components. Figure 2 shows, as an example, the hyperfine structure of the 2537-Å line in mercury-187. The major part of the interaction arises from the magnetic dipole and electric quadrupole moments of the nucleus. (We will neglect other effects, some of which can lead to observable anomalies in the hyperfine structure, as Redi mentions in his article). The interactions couple the atomic angular momentum \( J \) and the nuclear spin \( I \); the useful parameter to describe the hyperfine levels is thus the total angular momentum \( F \). The measured differences in energy for different values of \( F \), yield a magnetic-dipole factor \( A \), proportional to the nuclear dipole moment and the magnetic field (due to the orbiting electrons) at the nucleus, and an electric quadrupole factor \( B \) proportional to the nuclear quadrupole moment and the electric field gradient at the nucleus. In figure 2, for example, \( \Delta E_4 = 5/2A - 9/4B \) and \( \Delta E_5 = 5/2A - 5/4B \); the magnetic splittings alone, shown in the center, are 3/2A and 5/2A. To obtain the nuclear dipole moment \( \mu_A \) and quadrupole moment \( Q \), from these factors one must calculate the magnetic fields and the gradient of the electric field at the nucleus due to the atomic electrons. Depending on the complexity of the electronic configuration, the calculations can lead to uncertainties of 1-10% for \( \mu_A \) and 10-30% for \( Q \). Because the fields depend chiefly on the electronic structure, they are practically the same for all isotopes of an element. It is therefore common practice to refer measurements obtained for isotopes far from beta-stability to the most abundant stable isotope. One can increase

**Experimental arrangement**

For fluorescence spectroscopy at the ISOLDE facility at CERN. The 600-MeV protons collide with a molten-lead target to produce the short-lived isotopes.
the absolute accuracy by calibrating the hyperfine fields with the known nuclear moments of a stable isotope determined independently and in a few cases by using quadrupole moments obtained from μ-mesic-atom spectra.

One can obtain additional information about the structure of the nuclei by comparing the spectra of different isotopes. The fine-structure levels of two isotopes of the same element are shifted relative to each other because of differences in the electron–nuclear interaction. This shift is most pronounced for s and p electron atomic states for which the active electron spends part of its time inside the nucleus, thus probing the charge distribution of the nucleus. This part of the isotope and isomer shifts, called the field shift, therefore yields information on the nuclear charge distribution, in particular on the difference between isotopes of the mean-square charge radius.

An additional isotope shift arises from the fact that the kinetic energy of the nucleus must be included in calculations of the atomic energy. (This mass shift appears in its simplest form in the Bohr theory of hydrogenic spectra: The mass that appears in the theory is not the electron mass $m_e$, but the reduced mass $m_e/(1 + M_y/M_A)$, where $M_A$ is the mass of the nucleus.) Within each chain of isotopes the heaviest has the highest binding energy and its energy levels lie lowest. In atoms with several electrons there is an additional mass shift, called the “specific mass shift,” due to the influence of correlations in the motion of the electrons on the recoil energy of the nucleus. Both the normal and the specific mass shifts are described by a factor that depends on the wave functions of the electronic states of the transition. The mass shifts dominate the isotope shift in the case of light elements. However, for heavy isotopes the factor $m_e/M_A$ becomes very small, and changes in it become negligible. In the case of mercury, for instance, the mass shift amounts to 1% of the total isotope shift.

The field shift is caused by the change of the finite size and deformation of the nuclear charge distribution when nucleons are added or removed from the nucleus and is given by

$$\delta \nu = \frac{3}{2} \alpha \beta \frac{Z}{2} e^2 \Delta \Psi(0)^2 \delta \langle r^2 \rangle$$

where $\Delta \Psi(0)^2$ is the change of the electron density at the site of the nucleus during the optical transition and $\delta \langle r^2 \rangle$ is the difference in the mean square charge radius between the two isotopes. In the lighter isotopes the nucleus is bound more strongly than in the heavier isotopes, indicating that the effects of the field and mass shifts go in opposite directions.

Like the hyperfine splitting, the field shift (which is also due to the electron–nuclear interaction) can be expressed as a product of a factor depending mainly on the electronic configuration and a factor that reflects the nuclear structure. Although in principle all the radial moments $\langle r^n \rangle$ contribute to the field shift, in fact the contribution of higher moments amounts only to a few percent, and for simplicity we may refer to the nuclear-structure factor as the change in the mean-square charge radius, $\delta \langle r^2 \rangle$. Because the shape of the electronic wave function at the site of the nucleus is almost independent of the principal quantum number, it is possible to compare, or even calibrate, the calculated value of the electronic factor with isotope-shift measurements on x rays from inner-shell transitions or from mesic atoms. One scales the optical and x-ray measurements with the corresponding changes of electronic (or mesic) density at the nucleus. Such x-ray data are available for a few abundant natural isotopes. For these, the nuclear charge distribution is also known from electron-scattering experiments.

One can understand the changes in the nuclear charge radius as arising both from changes of the volume occupied by the charges (considering the nucleus to be a uniformly charged sphere of radius $R$) and from changes in the static and dynamic polarization of the nucleus (which can be characterized by the deformation parameter $\beta$). We can therefore write the change in charge radius, $\delta \langle r^2 \rangle$, when the atomic number changes by $\delta A$ as

$$\delta \langle r^2 \rangle = \frac{3}{2} \alpha \beta \frac{Z}{2} e^2 (\delta A/A) + (3/4\pi) R^2 \delta \beta$$

The factor $\rho$, called the “isotope-shift discrepancy,” is an empirical factor, adjusted to fit the overall trend of the nuclear charge radii in the mass region of interest. The deformation parameter is a measure of the oblateness or prolateness of the nucleus; it is simply the coefficient of the second spherical harmonic $Y_2(\cos \theta)$ in a two-parameter fit to the shape of the nucleus.

**Experimental methods**

A recent review paper discusses both on- and off-line experiments, and Daniel Murnick and Michael Feld have described the many experimental techniques for on-line laser spectroscopy that are now being implemented in several laboratories. In this article, however, I will consider only those methods that have yielded new results and will start and end with experiments with the conceptually simplest approaches in which the short-lived isotopes have been confined to a small volume, such as a resonance cell or quadrupole ion trap.

Mercury has a long chain of isotopes and isomers. The Mainz-ISOLODE group at CERN has carried out measurements on both neutron-rich and neutron-deficient isotopes of mercury far from beta decay.
Our group has also carried out similar measurements on the cadmium sequence with the same apparatus. Unfortunately, for cadmium, unlike mercury, the fluorescence signals from the same isotope are broad enough to overlap; because the element is lighter, the resolution is limited by the Doppler effect.

In a related experiment we measured directly the nuclear spins of the very neutron-deficient metastable mercury isomers 185m, 187m, 189m and 191m. Buy using a short laser pulse to excite selected hyperfine levels coherently, we were able to use a quantum-beat technique that avoids Doppler broadening of the signal. The principle of the experiment is as follows. One tunes the laser frequency to the peak of the hyperfine component of interest and keeps it constant there. The highly unstable atoms are in a resonance cell and also in a magnetic field. Under these circumstances, the total angular momentum $F = I + J$ is known. This method complements Doppler-limited fluorescence spectroscopy in an ideal manner because in a complex hyperfine-structure spectrum each component exhibits a different beat frequency that readily identifies it.

Cell experiments are ideal for elements of low chemical affinity, such as mercury and cadmium. However, for most other elements the atoms tend to stick to the walls of the resonance bulb—or even to react with them—within a time on the order of the diffusion time. This is true, for example, for alkalali atoms. To eliminate wall problems, one does experiments with atomic or ion beams. These methods simultaneously also reduce the Doppler width, a feature that is particularly important in very light elements.

The first successful on-line laser-spectroscopy experiment used an atomic sodium beam and optical pumping by magnetic-state selection. This elegant scheme involves two steps:

1. Intensity optical pumping changes the population of the $F = 1$ and $F = 2$ hyperfine levels. When the laser is tuned to excite exclusively transitions out of the upper ($F = 2$) level, the excited atoms subsequently return to either ground state, and after a few absorption-emission cycles all the atoms accumulate in the lower ($F = 1$) level. Just the opposite happens with the laser tuned to the transitions out of lower ($F = 1$) level, and all atoms are pumped into the upper ($F = 2$) level.

2. Magnetic-state selection is carried out with a six-pole magnet. The magnet focuses atoms with spin polarization $m_s = \frac{1}{2}$ onto a hot wire detector, while atoms with $m_s = -\frac{1}{2}$ are defocused and lost. Figure 5 shows the results obtained with this technique for ten sodium isotopes.

Georg Huber, Pierre Jacquinet, Robert Klapwijk and their groups realized that one can achieve ultrasensitive detection and strong reduction of the background of stable isotopes by using a second small mass separator at the hotwire particle detector. This made it possible to record the Na$^{23}$ signal with about 10$^5$ atoms per second. This method was invented in Orsay, where it was refined and perfected; it was then moved to CERN. In the first experiments at CERN, fragmentation of uranium by 20 GeV protons was used to produce the very neutron-rich sodium isotopes. All the remaining alkalali isotopes were finally measured at the ISOLDE mass separator; where the isotopes are produced by proton-induced spallation or fission reactions. These

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**Isotope shift and quadrupole moments of mercury isotopes.** The upper graph shows the gradual decrease in charge radius as one removes neutrons from the nucleus; the colored dots indicate isomeric states. The isotopes with $A = 185$, 183 and 181 have strongly prolate shapes, and thus have larger charge radii than expected. The change in sign of the quadrupole moments similarly indicates a change in structure at $A = 185$. Figure 6

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**Hyperfine spectra of sodium isotopes** obtained in an on-line experiment involving optical pumping by magnetic-state selection. The curves show the change in the detector output as the laser frequency is scanned through the resonance. Figure 5

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Stability using optical pumping and laser spectroscopy. The pioneering experiments of Ernst Otten, Jürgen Kluge and their group greatly enhanced the interest in such measurements by demonstrating that one could carry out systematic measurements of a model-independent nuclear structure parameter, such as the change in the mean-square charge radius $\langle r^2 \rangle$, for long chains of isotopes, thereby connecting mass regions of very different nuclear structure. The experimental setup for the more recent Doppler-limited fluorescence spectroscopy is shown in figure 3. One first produces radioactive ions by irradiating a molten-lead target with the 600 MeV proton beam of the CERN synchro-cyclotron. The volatile mercury ions pass through a mass separator and are collected on a foil, which is then turned around and inserted into a quartz resonance bulb. Heating the foil evaporates the mercury isotopes as neutral atoms. A pulsed laser beam, whose frequency can be varied around the intercombination line at 2837 Å, is focused through the resonance cell, and one observes the fluorescence of the radioactive isotope at right angles with a photomultiplier. The laser frequency is calibrated by detecting simultaneously the fluorescence from the stable isotopes in a reference cell. This cell is placed in the pole gap of a magnet to shift the Zeeman components into the region of interest. With this setup the Mainz-ISOLDE group investigated a large number of short-lived mercury isotopes and isomers in this way.

The most recent results, for mercury-185, are shown in figure 4, which shows the intensity of the fluorescent light as a function of the laser frequency. The spectrum displays a rich and complicated structure because the atomic hyperfine-structure multiplets of the high-spin isomer and those of the nuclear ground state overlap.

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<table>
<thead>
<tr>
<th>Mass Number A</th>
<th>Quadrupole Moment $Q$ (barn)</th>
<th>Isotope Shift $\Delta$ (MHz)</th>
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<td>181</td>
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**Figure 5**

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**Figure 6**

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**Figure 4**

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**Figure 3**

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The principal ideas are:

- One can exploit the velocity bunching that results from electrostatic acceleration to reduce the Doppler width to almost the natural line width. The spread of kinetic energies in a beam, \( \Delta E \), remains unchanged under electrostatic acceleration; and because \( \Delta E = (m/c^2)v\Delta \omega \), the product of the Doppler shift \( \Delta \nu \) and the Doppler width \( \Delta \nu \) also remains constant. In their experiments, Otten's group reduced the Doppler width in the ion source by a factor of 400 in this accelerated beam, making the resolution comparable to that of Doppler-free methods.

- Efficiently converting the ion beam into a fast atomic beam through charge-transfer collisions in an alkali-vapor cell, makes possible resonance-absorption experiments with continuous dye lasers on many different elements. The first on-line results were obtained at the TRIGA reactor at Mainz for the various alkali elements.

The Mainz-CERN-Goteborg-Aarhus group at ISOLDE recently performed a series of collinear fast-beam experiments with the apparatus shown on page 25. These experiments yielded the nuclear charge radii for barium-122 to 146 and also for ytterbium-162, 164 and 166. The beam intensity for Ba-146 was as low as 10^5 atoms per second, but the experiments still achieved a spectral resolution of a few MHz.

In still another experiment^2 at the ISOLDE mass separator, Curt Eckstrom, Ingvar Lindgren and their coworkers of the Gothenburg-Uppsala group employed an on-line version of the classic atomic-beam magnetic-resonance method (see figure 7 of Redi's article, on page 32) to measure systematically nuclear spins and moments of nuclides far from beta stability. To date, 58 new spin values and 36 magnetic moments have been determined for isotopes of bromine, rubidium, indium, iodine, cesium, europium, thulium, gold, tantalum and francium. Nuclides produced in yields down to fractions of a second and half lives down to 0.38 seconds have been reached for study.

In cases of extremely low production yields, one can use nuclear-radiation-detected optical pumping or variants of this technique. In such experiments the nuclear spin is polarized by absorption and emission of polarized light by the atom. One can monitor the nuclear polarization by observing the asymmetry of beta decay. The signal is practically free from the usual background interference, because one detects the nuclear polarization and performs the optical pumping in completely different spectral regions.

The Oak Ridge group has recently used such a technique to measure the optical isomer shift of Am-240m, which has a one millisecond lifetime for spontaneous fission. Specifically, the experiment involved:

- Production of the isomer by bombarding a uranium target with 49-MeV lithium ions and thermalization of the recoiling isomers from fission in helium gas at 180 torr.

- Optical pumping of Am-240m with circularly polarized light. Because unpolarized light is emitted in the subsequent spontaneous decay, the atom acquires an excess of angular momentum. This excess is transferred via the hyperfine interaction to the nucleus.

- Detection of the decay products. The spontaneous-fission decay from the oriented nucleus is anisotropic and serves as the signal when the laser is tuned through the optical resonance.

With this technique, Curtis Benis, J. R. Beene and their collaborators verified the expected large deformation of Am-240m, which exhibits about twice the quadrupole deformation of the ground state, Am-240.

Nuclear structure

Having considered the major methods of on-line laser spectroscopy, let us next turn to the results of some of these experiments, such as the observed trends in the variation of nuclear charge radii with neutron number, which demonstrate the richness of the information about nuclear structure obtainable with optical spectroscopy.

One particularly interesting set of results consists of the nuclear spins, moments and charge radii of mercury isotopes and isomers spanning the mass region from \( A = 181 \) to \( A = 206 \)—a long chain, containing 25 isotopes and 8 isomers. Figure 6 shows the change of the nuclear charge radii as obtained...
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from field isotope shifts. Except for small irregularities (such as the "odd-even staggering" discussed by Redi), the change in the nuclear charge radius follows an almost straight line for atomic numbers between 206 and 186. This is attributed to a regular shrinking of the nuclear volume accompanied by a smooth change in shape from spherical to slightly oblate. The jump in the charge radius of the odd-numbered mercury ground states with atomic numbers below 186 is due to a sudden onset of a strong prolate deformation of the nucleus. The same effect also produces the large odd-even staggering in this region. Apparently, for atomic numbers less than 186, the nuclei are separated sharply into two groups of quite different shapes. These nuclear shapes are almost degenerate in energy but do not mix—otherwise some nuclei would be found between the two branches. The situation arises because of an intricate balance in the contribution of the neutron and proton binding energies to the total energy. The protons in mercury are close to the end of a shell and strive toward a spherical shape, while the neutrons in the very light neutron-deficient nuclei are in the middle of a shell and strive toward a prolate shape. This tendency causes a double minimum in the total nuclear energy as a function of deformation. For Hg$^{185}$ the absolute minimum is on the oblate side, in part because the neutrons can all pair up. For the odd-numbered isotopes mercury-181, 183 and 185, without the neutron-pairing energy for the odd neutron, the absolute minimum shifts to the prolate side, and, in fact, these nuclei exhibit a strong prolate deformation. The combined shell and pairing energies are very different, which indicates that the nuclear states representing either oblate or prolate deformation are quite different in composition. This might explain why they do not mix.

In the case of mercury-185, there are even two states with different radii. The isomeric state, Hg$^{185m}$ with $I = \frac{13}{2}$, has a small average charge radius and an oblate shape, while the nuclear ground state, Hg$^{185}$ with $I = \frac{1}{2}$, has a larger average charge radius, indicating a strong prolate deformation. These two isomers are not only shape isomers but also some kind of pairing isomers. The pairing gap for protons is small at the oblate minimum; for neutrons, it is small at the prolate minimum. Because of the readiness of Hg$^{185}$ to change shape, we coined the expression "flip-flop" nucleus for this discovery. I should point out that a shape transition is a rather sensitive matter for a nucleus: energy differences as small as a few hundred keV in the binding energy decide for one nuclear shape or the other, as compared to the total nuclear binding energy for about 1600 MeV for a mercury nucleus.

Another interesting behavior occurs at the "magic" nucleon numbers, which indicate closed shells of nucleons. Several groups have studied the effect of shell closure on the change of the mean-square charge radius for different elements and in the vicinity of the closed neutron shells at $N = 50$ and $N = 82$ (see figure 7). The figure shows the change in the charge radius for the case of $N = 82$ for the chain of cesium isotopes as evaluated from the field isotope shift measurements of the Orsay and Mainz groups. For comparison we have also plotted the change in the reduced standard radius, which represents the regular shrinking of the nuclear volume. A deviation from this line is mainly due to a change in deformation that is either static or dynamic in nature. A characteristic kink occurs in the experimental data at the magic number, demonstrating that starting from a closed shell, the nuclei strive towards deformation as the neutron number is changed in both directions. One can observe an apparent $Z$-dependence in the data obtained from both on- and off-line experiments for the barium, xenon, cadmium and tin isotopes. Tin has a magic proton number and has the smallest deformation effects when the neutron number is changed.

Figure 7 also shows data obtained by ion trap for observing hyperfine spectra. The sketch at left shows the apparatus; the ions are trapped in stable orbits within the cavity, where they can interact with a laser beam and an rf field. The natural oscillations in the trap. The graph at right shows the expected and observed spectra of helium-3 ions in the microwave region.
the Orsay and Mainz groups for rubidium isotopes for the case $N = 50$. Again, the deviation from the regular behavior of the reduced standard radius indicates increasing deformation above and below the closed shell. Below $N = 50$ there is even a reversal in the trend of the change of the charge radius: The radius becomes larger with decreasing neutron number. It is currently an open question whether this reversed trend is due to higher moments in the deformation or to diffuseness of the nucleus.

Figure 8 shows the behavior of the deformation parameter $\beta_{2,1/2}$ for the cesium isotopes $A = 115-145$, as obtained from isotope-shift data. A purely static deformation parameter can also be obtained from the spectroscopic measurements of the quadrupole moment. In the case of cesium, the two deformation parameters agree reasonably well, particularly at large deformations. It thus appears that the deformation becomes more static in character as the nucleus becomes more deformed, which is the expected behavior.

Future developments

On-line hyperfine spectroscopy is still in its infancy but it has already contributed a vast amount of knowledge on nuclear structure in regions away from beta-stability. So far only the optical techniques have had the sensitivity required to measure charge radii of short-lived nuclei. In the future, new experimental techniques should increase both sensitivity and resolution even further. As a particular example, numerical studies carried out at Texas A&M University showed that it is possible to use ion-storage techniques for on-line spectroscopy. We are now constructing the apparatus shown in figure 9 to try to verify these results experimentally. The graph shows a side-band microwave spectrum of trapped helium-3 ions; it is due to the binding of the ions within the potential well formed by the trap. A similar spectrum should be seen at optical frequencies.

With such a trap it becomes possible to cool the stored ions to eliminate the Doppler effect. The incident laser beam is tuned to one of the side bands below the resonance frequency. Ions excited by this radiation will reradiate symmetrically at all side-band frequencies. Because the average energy of the emitted photons is larger than energy of the absorbed photons, the ions must lose kinetic energy. This “laser-cooling” technique has already cooled ions to below 1 Kelvin in experiments performed at NBS and at the University of Heidelberg. We hope to use the technique to obtain practically Doppler-free spectra of short-lived ions. Doppler broadening is particularly troublesome in the spectra of the lighter isotopes.

Several of the established high-precision techniques of laser spectroscopy can be adapted to on-line use. New developments in spectroscopic techniques are also changing the picture. For instance, multiphoton fluorescence and selective multistep photoionization with successive counting of the multiphoton ionization events offer alternatives which a few years ago did not even exist.

References