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LASER SPECTROSCOPY OF UNSTABLE ISOTOPES

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Abstract. High-resolution methods of optical spectroscopy in unstable isotopes are discussed, with emphasis on fast-beam laser spectroscopy. The chief goal is to obtain information about nuclear ground-state properties from hyperfine structure and isotope shifts. The methods have to be suitable for use with minute samples, due to the small amount of radioactive atoms available.


1. Introduction
Laser excitation of fast beams of ions or atoms has become an important technique of resonance spectroscopy. It was initiated by the crossed-beam experiments of H.J. Andrä et al. [1] aiming at a good time resolution. Recent experiments take advantage of the collinear-beam geometry [2-5]. Applications of this method to molecular ions are demonstrated in three contributions to this conference [6,7,8].

In this talk I shall discuss the application of fast-beam laser spectroscopy to unstable isotopes. The method has several outstanding features which include high resolution and sensitivity, and ideal suitability for use with on-line isotope separators. The experiments carried out so far on neutron-rich Rb and Cs isotopes will be compared with other experiments using different methods.

2. Nuclear properties from optical spectra
The history of atomic spectroscopy as a tool of nuclear physics [9] started about 50 years ago when the well-known splitting of spectral lines, called hyperfine structure (hfs) was ascribed to a magnetic coupling between the nucleus and the electron shell. Spins and magnetic moments had to be introduced as basic properties of nuclei and could be evaluated from hfs splittings. Improved resolution led to the discovery of quadrupole interaction, connected with a nuclear electric quadrupole moment, and of the nuclear volume effect in isotope shifts (IS). A limit of resolution was the Doppler width of optical lines (~1 GHz). For that reason nuclear volume shifts were only observed in heavy elements, and the quadrupole interaction could be proved in a few favourable cases of strongly deformed nuclei.

Exhaustive investigation of hfs remained to be carried out by means of rf spectroscopy. The general application of optical spectroscopy to studies of nuclei requires a Doppler-free method which allows natural linewidth resolution (~10 MHz).

2.1 Hyperfine structure
The hfs energy of a state with total angular momentum \( F = I + J \) is given by

\[
W_F = \frac{1}{2} KA + \frac{3}{4} \frac{\langle K(K+1) - I(I+1) \cdot J(J+1) \rangle}{2(I^2-1)J(2J-1)} B
\]

with \( K = F(F+1) - I(I+1) - J(J+1) \).

It contains the nuclear spin \( I \), the magnetic dipole interaction constant

\[
A = \mu_I \langle r_s(0) \rangle \quad \text{and the electric quadrupole interaction constant}
\]

\[
B = e Q_s \left< \frac{\partial^2 V}{\partial z^2} \right>
\]

For evaluating the nuclear quantities \( \mu_I \) and \( Q_s \), one has to know the magnetic hyperfine field \( \mu_I \langle r_s(0) \rangle \) and the electric field gradient \( \left< \frac{\partial^2 V}{\partial z^2} \right> \) produced by the electrons. A calibration is possible, if \( \mu_I \) and \( Q_s \) are known for at least one isotope of an element. Magnetic moments are available in practically all cases from nuclear and atomic magnetic resonance, and an increasing number of spectroscopic quadrupole moments is determined from hfs of mesic atoms [10]. Most quadrupole moments are still

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based on calculations of $<\delta^2\nu/\delta z^2>$ which are accurate within 10% to 30% [11].

2.2 Isotope shift

The IS between the centers of gravity of hfs multiplets are not only an effect of change in nuclear volume (field shift), but also in nuclear mass [12]

$$\delta
V^{AA'} = \delta
V_f^{AA'} + M \frac{A'-A}{AA'}$$

(4)

For isolating the field shift one has to know the mass shift which is not very important in the heaviest elements, but predominates in the light ones. A calculation accounting for the correlation of electron momenta is difficult. Only in simple cases such as $s$-$p$ transitions the "specific" mass shift is of the order of the "normal" one-electron mass shift $M = \nu/1836$.

The field shift is connected with the change of the nuclear rms charge radius $\delta <r^2>_{AA'}$ by

$$\delta
V_f^{AA'} = \frac{2}{3} \pi \frac{Z^2 e^2}{A} \Delta |\Psi(0)|^2 \delta <r^2>_{AA'}$$

(5)

where $\Delta |\Psi(0)|^2$ is the change of electron density at the nucleus in the electronic transition. This non-relativistic equation holds also for the relativistic case, if a correction factor depending on $Z$ and $A$ is introduced. $\Delta |\Psi(0)|^2$ has to be calculated, if a calibration by X-ray isotope shifts or data from muonic atoms or electron scattering is not practicable [11].

Disregarding these complications one can expect that optical IS in a long series of isotopes reveal the general trends as well as local irregularities of changes in nuclear radii. This statement is already a part of an answer to the question:

3. Unstable isotopes - why and how?

The fundamental nuclear properties which manifest themselves in the optical spectra are thoroughly known near the valley of stability. This is the region where nuclear models have been developed and are usually tested. But in the region far from stability information about nuclear ground states is rather scarce.

New unexpected deformation effects were discovered in the first systematic study of IS for very neutron-deficient isotopes of Hg [13,14]. In these experiments, due to the large hfs and IS of the heaviest elements, the Doppler width could be tolerated. But sophisticated methods of $\gamma$-radiation detected optical pumping (RADOP) [13] or fluorescence spectroscopy using a frequency-doubled pulsed dye laser [14] had to ensure sensitivity.

Generally, the application of standard methods to unstable isotopes is limited by the small amount of radioactive atoms available, and by their short half-life. Versatile sources of unstable nuclides are the on-line mass separators connected with spallation or fission targets [15]. They yield isotopically pure samples in the form of ion beams.

As an example, Fig.1 shows the set-up used at Mainz. It has been developed for making available neutron-rich Rb and Cs isotopes from fission of $^{235}$U [16]. The reactor, operated by the nuclear chemistry institute, is used as a source of thermal neutrons. $^{235}$U imbedded in porous graphite is placed in a tungsten oven close to the reactor core and exposed to a flux of $2 \times 10^{11}$ neutrons/cm$^2$ sec. At temperatures of about 1200°C the alkalis diffuse out of the target and evaporate from the tungsten surface as positive ions. Accelerated to 3-10 keV they are guided out of the reactor into a separation magnet, and the isotopically pure beam is available in quantities shown in the lower part of the figure.

Although ISOLDE II at CERN/Geneva is much more efficient (cf. section 5), this yield can be taken as typical for similar facilities. Therefore, optical methods designed for general use with radioactive
nuclides should at least be able to work with $10^8$ atoms per second.

The great advance for sensitivity and resolution of optical spectroscopy was the development of a cw dye laser. This laser is now available in single-mode operation over the entire visible range of the spectrum. The output power is sufficient to saturate the transitions, and the linewidth achieved by frequency stabilization is about 1 MHz. These features allow highly sensitive spectroscopy with natural-linewidth resolution.

Up to now three different methods have been introduced for systematic studies of unstable isotopes. The experiments are still going on and further extensions are obvious.

In 1975, G. Huber et al. at Orsay reported the first experiment on $^{21-25}$Ba [17], using excitation of the D lines in a collimated atomic beam which was analyzed by a sixpole magnet and a mass spectrometer. Two years later it was extended to the neutron-rich isotopes up to $^{31}$Na produced by high-energy proton reactions in a uranium target. Quite recently, the same scheme has been applied to neutron-deficient Cs isotopes from ISOLDE.

A collimated atomic beam is also used in fluorescence experiments on neutron-deficient Ba isotopes at Karlsruhe, first reported by G. Nowicki et al. in 1977 [18].

Finally we applied the collinear laser excitation of a fast atomic beam to neutron-rich Rb and Cs isotopes [19].

Since the Cs and Ba experiments are closely related to each other in their physical meaning, I shall take them as examples for further discussion.

4. Atomic-beam fluorescence spectroscopy

The preparation of a sample has to start from the mass-separated ion beam. A straightforward method is to collect the ions on a target and to reevaporate them as neutral atoms. By forming a collimated beam and crossing it with the laser beam one arrives at a standard arrangement of sub-Doppler spectroscopy [20]. Resonance absorption may be detected by fluorescence.

This is essentially the scheme used by the Karlsruhe group in the experiments on Ba [18]. Instead of working on line with the accelerator this experiment is performed step by step. Thus, for isotopes with half-lives $\geq 1$ h it takes advantage of the intense internal beam of the cyclotron. Samples of about $10^{11}$ to $10^{13}$ atoms of the respective isotope are produced by $(d, xn)$ or $(α, xn)$ reactions in Ba or Xe, followed by off-line mass separation. Silicon disks of ultra-high purity are used as matrix material from which Ba is heated off in the atomic-beam oven.

Hfs and IS are investigated in the transition $6s^2 \, ^1S_0 - 6s6p \, ^1P_1$ ($λ = 533.6$ nm). Its natural linewidth is 19 MHz, and the beam collimation allows for Doppler broadening of about 8 MHz. Since at most some $10^4$ atoms/sec are present in the beam, special care of high detection efficiency and low background has to be taken. The overall photon detection efficiency is about 2%. By careful elimination of scattered laser light, incandescent light from the oven and γ-radiation from the sample, the total background is less than 100 counts/sec. The result is demonstrated in a resonance curve (Fig. 2) obtained within 6 min from a sample of about $10^{11}$ atoms [21].

![Fig.2: Spectrum of $^{126}$Ba. Stable $^{132}$Ba is used as a reference.](image)

Measurements were performed for the radioactive isotopes and isomers $^{135m}$Ba, $^{133}$Ba, $^{133a}$Ba, $^{131}$Ba, $^{128}$Ba, $^{126}$Ba, in addition to remeasurements of all stable isotopes. Results are included in Fig. 15.

5. Optical pumping of atomic beams

A collimated atomic beam, crossed with the laser beam, is also used by the Orsay group in experiments on alkali isotopes [17, 22]. They introduced a new efficient detection scheme based on optical pumping (Fig. 3).

The atomic beam passes through a sixpole magnet,
which focusses atoms in the state $m_J = +1/2$ into a surface ionizer acting as the ion source of a small mass separator, and the atoms transmitted through the apparatus are detected by ion counting. If the laser is tuned to a line connected with the $F=I-1/2$ hfs level of the ground state, part of the atoms are pumped to the $F=I+1/2$ level, and the number of atoms belonging to $m_J = +1/2$ states is increased. A positive signal is observed at the ion detector. Similarly the transitions starting from the $F=I+1/2$ level give rise to a negative signal.

This non-optical detection is characterized by an ultimate efficiency and very low background. Up to 50% of the beam are counted by the detector and, with careful shielding of radioactivity, the background is about 1 count/sec. Thus, compared to optical detection, the signal-to-background ratio is increased by roughly a factor of $10^3$, provided one photon per atom is emitted.

In the most recent experiments on neutron-deficient Cs isotopes $6s^2\frac{2S}{1/2} - 6p^2\frac{2P}{3/2}$ ($\lambda = 852.1$ nm) could be measured for the long chain of isotopes ranging from $^{137}$Cs with magnetic neutron number to $^{121}$Cs, and for several isomeric states [23]. For results see Fig. 8.

The apparatus had been connected to ISOLDE. The mass-separated Cs$^+$ beam was stopped inside a tantalum tube coated with yttrium from which Cs atoms were evaporated by heating at about 900$^\circ$C. About $10^{-5}$ of the ISOLDE yield was transmitted to the interaction region. It was a matter of convenience that this yield is between $10^{10}$ and $10^{11}$ atoms/sec in the region under consideration.

6. Fast-beam laser spectroscopy

Quite a different approach to sensitivity may start from the problem of efficiency in preparing the sample. In view of the fact that unstable isotopes are available at mass separators one might think of a method directly using the fast ion beam.

The solution of this problem was given in a paper by S. L. Kaufman in 1976 [24], and soon after that we performed the pilot experiment on fast beams of the stable Na and Cs isotopes [5].

The basic idea is very simple: Sufficiently large excitation rates are obtained, if the time of interaction between the beam and the laser light is long enough. Therefore, the laser beam has to be superposed collinearly to the output beam of the separator. And, since resonance lines of ions are usually not accessible to cw dye lasers, the ion beam has to be converted into a fast atomic beam. This neutralization is efficiently performed by charge transfer in an alkali-vapour cell.

The striking advantage of this concept is the avoidance of any loss of material between isotope separation and laser excitation. But this advantage is only profitable, if all (or a sufficiently large number of) atoms interact with the light. This is a question of the absorption Doppler width.

Since the spread of kinetic energy in the beam remains unchanged under electrostatic acceleration

$$\delta E = \delta \left( \frac{mv^2}{2} \right) = mv \delta v = mc^2 \frac{2}{v^2} \Delta v_D \delta v_D = \text{const}$$

(6)

the product of the average velocity $v$ and the velocity spread $\delta v$, or likewise the product of the Doppler shift $\Delta v_D$ and the Doppler width $\delta v_D$ are constants of the motion. In other words: The Doppler width is considerably reduced from its original value $\delta v_D^{(0)}$ in the ion source. For the ideal case of a thermal distribution (which is realized in surface-ionization sources)

$$\delta v_D = \frac{1}{2} (kT/e)^{1/2} \frac{1}{\sqrt{2}} \delta v_D^{(0)}$$

(7)

The reduction factor is about 400 for a source temperature of 1500 K and an acceleration voltage of 5kV. It corresponds to a Doppler width of 4 MHz for the blue Cs line ($\lambda = 455.5$ nm), compared to a
natural linewidth of 1.2 MHz. Natural linewidths of the strongest resonance lines are even larger by a factor of 10. It may be concluded that essentially all atoms in the beam are excited and that the resolution is comparable to Doppler-free methods.

It is presumed, of course, that the charge-transfer process doesn't change the velocity distribution. This is actually the consequence of the large cross-section of about $10^{-14} \text{cm}^2$ which exceeds the kinetic cross section by two orders of magnitude. In the resonant case, e.g.

$$\text{Cs}^+ + \text{Cs}(6s) \rightarrow \text{Cs}(6s) + \text{Cs}^+$$

the beam energy remains unchanged, whereas in the non-resonant case, e.g.

$$\text{Cs}^+ + \text{Cs}(6s) \rightarrow \text{Cs}(6p) + \text{Cs}^+ - 1.4 \text{ eV}$$

or in collisions between different partners, the fast-beam kinetic energy is changed by almost exactly the amount of the energy defect $\Delta E$. Conservation laws require that in forward scattering the kinetic energy transferred to the target atom is of the order $(\Delta E)^2/\text{eV}$, which is negligible for beam energies in the range of keV.

In any case, the width of velocity distribution and the angular divergence is not affected by charge transfer. However, the Doppler-shifted sharp lines may be split by the energy-loss spectrum corresponding to different reaction channels.

Fig. 4: Experimental set-up for fast-beam laser spectroscopy

6.1 The experiment

Fig. 4 shows the experimental set-up used with the mass-separated beam of alkali isotopes from nuclear fission (see Fig. 1). The ion beam is deflected to merge with the laser beam and neutralized in the heated charge-exchange cell containing alkali metal at a vapour pressure of $10^{-3}$ Torr. Laser light is absorbed by the atoms whenever the Doppler-shifted frequency is tuned to one of the resonance lines. This is detected by counting fluorescence photons, collected along a path length of 20 cm by a cylindrical lens and guided into the photomultiplier by a light pipe.

Varying the Doppler shift at fixed laser frequency offers a convenient way of scanning the absorption spectra. For this purpose a programmable voltage is applied to the charge-exchange cell, post-accelerating the ions prior to neutralization.

Fig. 5 illustrates the hfs measurement for a Cs isotope with $I=7/2$. The ground and excited state splittings give rise to 6 hfs transitions indicated by the vertical arrows. By scanning the acceleration voltage these transitions are observed at a scale shown in the lower part of the figure.

Measurement of IS additionally requires comparison to a reference isotope. For this purpose a beam of stable $^{133}_{55}$Cs from a separate ion source is run through the apparatus alternatively to the unstable isotope beam. Uncertainties of beam-energy calibration are eliminated by relating all IS to $^{137}_{55}$Cs obtained from the mass separator.

6.2 Measurements

Measurements were performed in the transitions $6s \ 2^1S_{1/2} - 7p \ 2^3P_{1/2} (\lambda = 455.5 \text{ nm})$ of Cs and $5s \ 2^1S_{1/2} - 6p \ 2^3P_{3/2} (\lambda = 420.2 \text{ nm})$ of Rb for the neutron-rich isotopes $^{138-142}_{55}$Cs and $^{89-94}_{37}$Rb. Their half-lives extend from 32 min for $^{138}_{55}$Cs to 1.7 sec.
for $^{142}\text{Cs}$, and similarly for the Rb isotopes between $^{89}\text{Rb}$ and $^{94}\text{Rb}$. The isotopes with magic neutron number, $^{137}\text{Cs}$ and $^{87}\text{Rb}$, are chosen for calibrating the isotope shifts.

A spectrum of $^{139}\text{Cs}$ may serve as a typical example (Fig. 6). Both groups of hfs components are scanned at a fixed laser frequency, simultaneously with the $^{133}\text{Cs}$ reference. The voltage scale is used for calculating relative Doppler shifts by

$$(v_2-v_1)/v_0 = (2eU_1/M_1c^2)^{1/2} - (2eU_2/M_2c^2)^{1/2}$$

from which the hfs parameters and the IS can be evaluated. Atomic masses are known with sufficient accuracy from standard tables [25].

The linewidth corresponds to $10\text{ MHz}$ in a frequency scale. By a computer fit the resolution of excited-state hfs is better than $1\text{ MHz}$, yielding the very small quadrupole interaction constants with an accuracy of about $20\%$. Errors in the large ground-state splittings and in isotope shifts are usually determined by the voltage calibration and amount to several MHz.

The hfs splitting of $^{139}\text{Cs}$ is typical for all odd isotopes in this region which have spins $I=7/2$ and therefore similar magnetic moments.

A different case is $^{140}\text{Cs}$ for which the magnetic and electric hyperfine interaction in the excited state are of the same order, due to the small magnetic moment connected with $I=1$. As shown in Fig. 7 part of the splittings nearly compensate.

A characteristic of all spectra are the satellites of each line, separated from the main peak by 1.4 volt. They are due to the non-resonant branch of charge transfer to the first excited $6p$ state of Cs, giving rise to a loss of kinetic energy equal to the $6s-6p$ energy difference. In this way the distribution of charge-transfer reaction products over final states is directly observed. We have shown that this distribution strongly depends on the reaction partners [5, 26]. In fact, these high-resolution measurements can be used to evaluate branching ratios of charge-transfer reaction channels.

7. Results

The information obtained throughout the described work includes spins, magnetic dipole and electric quadrupole moments as well as differences of nuclear charge radii. Most spins and magnetic moments of Rb and Cs isotopes had previously been measured by C. Ekström et al. [27] using atomic beam resonance on line with ISOLDE, and a few spins were measured by optical pumping (RADOP) [16]. These quantities are accessible in the atomic ground state.

The importance of laser spectroscopy is obvious: Quadrupole interaction can only be observed in excited states with $J\geq 1$, and isotope shift measurements have to involve different atomic states. It would be beyond the scope of this talk to discuss all results in detail, apart from the fact that their analysis is far from being complete. Publications of all three groups comprise reports of first results.

A general review of nuclear radii in the Cs-Ba region can already be given. Fig. 8 contains the results of the different experiments, marked by different symbols. The gross run of $\delta<r^2>$ values near neutron-shell closure is characterized by a sharp increase above the magic neutron number $N=82$. This behaviour is certainly due to an influence of neutrons in the opened shell on the proton distribution. It is also observed for the isotonic stable
reached. Therefore the methods have to compete with each other mainly in sensitivity and suitability for the particular case. A decision for one or the other method will depend on the element and atomic transition under consideration, but some general remarks can be made:

1) The detection of fluorescence is usually limited to a few photons/atom, since optical pumping removes the atoms from the absorbing state. Multiple excitation is possible in some special cases such as $^{1}S_0$ ground states of doubly-even ($I = 0$) isotopes. This gain of detection efficiency has been exploited in the measurements on even Ba isotopes $^{128}$ and $^{126}$ (cf. Fig. 2).

2) On the other hand, optical pumping is the decisive condition for the sophisticated detection scheme used by the Orsay group, which relies on state selection in an inhomogeneous magnetic field. Limited to paramagnetic atoms it is ideally suited to $^2S_{1/2}$ alkali ground states. Furthermore, efficient surface ionization of focussed atoms may hardly work in other cases. A possible alternative might be the detection of atoms by radioactivity [27].

3) Fast-beam spectroscopy is based on the reduction of Doppler width occurring by acceleration. The optimum condition is an ideal matching to the natural linewidth, which is easily fulfilled in the strong transitions of alkali atoms to the first excited p states. In the blue Rb and Cs lines, which have been chosen for convenience of laser action and photon detection, the observed Doppler width (determined by beam divergence) is larger than the natural linewidth of 1.2 MHz by a factor of 10.

Extension of this method to other elements may require gas-discharge ion sources which have a broader energy distribution. This, of course, effects the sensitivity, whereas the resolution needed can be achieved by use of velocity-selecting filters. Another way is to increase the acceleration voltage according to (6) as far as sufficiently stable high-voltage sources are available.

It seems to be evident that fast-beam laser spectroscopy is a sensitive method for rather general use with ions or atoms. The charge-transfer neutralization is by no means limited to presently studied elements. In cases where ground-state resonance lines are not accessible to cw lasers the population of metastable states [30] may help to meet with absorption in the visible. Recent experiments on Xe$^+$ [41] and Rb$^+$ [31] took advantage of the considerable population of metastable states in

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**Fig. 8:** Change of rms charge radii relative to $N = 82$. Symbols: ○ for Ba (Karlsruhe) △ for Cs (Orsay-ISOLDE) with isomers ▼ for Cs (Mainz) Full symbols denote stable isotopes.

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Measurements on Cs range to the very neutron-deficient isotopes, and present data seem to fit the assumption of gradually increasing nuclear deformation [29].

### 8. Discussion

Three different high-resolution techniques of laser spectroscopy have proved successful in systematic studies of unstable nuclides, and first results are quite promising for future work.

The resolution limit of natural linewidth is almost
the discharge ion source. Further effort may tackle the problem of sensitivity by improving the efficiency of light collection and suppressing the background. Useful techniques may be the excitation and observation at different wavelengths where it is possible, and, for longer-lived states, the pulsed excitation and observation of the subsequent decay. A non-optical detection scheme might combine fast-beam laser spectroscopy with the RADOP technique [32] by implantation of optically pumped atoms into a suitable crystal, and observation of the asymmetry in the $\beta$-decay of polarized nuclei.

9. Conclusion
I tried to show that optical spectroscopy has recovered its importance for nuclear research. The information obtained on nuclear ground and isomeric states far from stability is hardly accessible to other methods. Present experiments using high-resolution laser techniques have reached a stage of sensitivity sufficient for investigating long isotopic chains which still provide key information about nuclear structure.

The fast-beam experiments have been performed in team work by J. Bonn, S.L. Kaufman, W. Klempt, H. Lochmann, G. Moruzzi, R. Neugart, E.-W. Otten, L. von Reisky, B. Schinzler, K.P.C. Spath, J. Steinhacker and D. Weskott. I like to thank the Karlsruhe and Orsay teams for providing unpublished material, and their permission to present it here.

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