SOME RECENT DEVELOPMENTS IN LASER SPECTROSCOPY OF UNSTABLE ISOTOPES

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I. Introduction

It is a quarter of a century ago that the first optical spectroscopy experiments were conducted on radioactive isotopes. At that time these were off-line experiments, of course, using the whole catalogue of standard methods which was developed for stable isotopes and included also the high-resolving rf-methods, like atomic beam magnetic resonance, optical double resonance, optical pumping etc.

More than a decade ago the first on-line experiments came up, but now with methods adopted to the special situation which asked for handling minute samples of short-lived material. Several rf-methods played at that time the trick of observing the resonance signal from the destruction of the $B$-decay asymmetry, which is natural insofar as all rf-methods have built in basically some kind of nuclear polarization mechanism. One out of this group, the RADOF method (Radioactive Detection of Optical Pumping) opened us the door to the ISOLODE on-line mass separator and to optical spectroscopy of the Hg-chain. Kluge reported on these experiments at the Cargese meeting. At the same occasion Huber reported on spectroscopic experiments in a chain of sodium isotopes by the Orsay group, the first experiment in the new field of on-line laser spectroscopy which developed rapidly since. Certainly I cannot count all the groups interested in the systematic application of laser spectroscopy to the investigation of hyperfine structure and isotopic shift - at stability as well as off –. Since we are considering nuclei far off stability at this conference I should confine myself to on-line experiments, some of them being in progress, others with results. The choice will be obviously somewhat different from the ones taken in recent presentations of the field at the Nashville and Berkeley meetings. I also like to refer to some recent review articles.

II. Experimental Techniques

Laser spectroscopy is so powerful and effective and it offers so many different possibilities and schemes that in fact most of the groups can afford the luxury of playing their own particular method and melody. Nevertheless, one can distinguish some categories: A) The sample can be prepared either as an A1) atomic vapour imprisoned in a cell, A2) a thermal, collimated atomic beam irradiated by the laser at right angle, A3) a fast beam of atoms or ions (e.g. the mass separated beam itself) excited collinearly by the laser.

B) The signal detection falls roughly into two schemes
B1) optical detection,
B2) particle detection.

B2 again splits into the three classical methods, namely the observation of absorption, emission and dispersion. B2 includes selective ionization and ion counting methods as well as those based on radioactive decay. Since almost any B fits to any A, the choice is obviously great and the question about "the best method" illegitimate, since it offers too many different aspects. But it can be said so far that in those cases in which particle counting methods could be applied the experiments proved to be more sensitive than purely optical ones because of the higher background rejection. I turn now to the discussion of a few individual concepts.

II.1. Polarization Spectroscopy in Resonance Cells

The recent experiment on $^{195}$Au by Kluge, Kremmling and their collaborators is an example for the A1, B1 combination. It started as a continuation of preceding on-line experiments on Hg and Cd, where one observed fluorescence from an atomic vapour excited by a pulsed dye laser (these experiments have been presented at earlier meetings and published elsewhere). The Au-experiment is the first attempt to apply laser techniques to a radioactive isotope of a refractive element. This introduced major complications and changes in the concept (see fig. 1).

For keeping gold in the vapour phase temperatures around 1300°C are required close to the melting point of the quartz vessel which is contained in an evacuated oven. The narrow geometry as well as the bright radiation of the oven makes the observation of the weak fluorescence radiation unfeasible and favours another scheme, the so-called polarization spectroscopy. For this purpose the laser beam is split into two counter-propagating parts, the strong circularly polarized pumping beam and the weak linearly polarized probing beam. As seen from the simplified level scheme in fig. 2 the right handed polarized, pumping beam will saturate the transition between the substates $m_\pi = -1/2$ in the lower and $m_\pi = +1/2$ in the upper state; thus it will equalize the population of these two levels. As a consequence the vapour loses its absorptive as well as dispersive power for right handed light and turns from anisotropic to an anisotropic medium which tilts the plane of polarization of the probing beam like a sugar does. This is observed
by an analyzer at right angles to the polarizer. Since the anomalous dispersion curve has natural line width the method is dopplerfree for counterpropagating beams as seen from the following consideration. An atom with resonance frequency $v_0$ and velocity component $v_2$ along the direction of the pumping beam absorbs light only at frequency

$$v_L = v_0 \left(1 + \frac{v_2}{c}\right)$$  \hspace{2cm} (1a)

Contrary the probing beam acts only on atoms with

$$v_L = v_0 \left(1 - \frac{v_2}{c}\right)$$  \hspace{2cm} (1b)

Simultaneous action is therefore restricted to $v_2=0$ and the signal occurs exactly at the resonance frequency $v_0$. At present the line width is still dominated by the frequency doubled pulsed dye laser (necessary, because the resonance line lies in the far UV) which will be replaced by a more power-

Fig. 1 Experimental set-up for polarization spectroscopy of a radioactive gold vapour

ful one, the band width of which will be narrowed by an external etalon followed by a laser amplifier. In the pilot experiment, reported here, the isotope shift and hyperfine structure from a sample of $^{101}^3$ off-line separated $^{199}Au$ atoms was observed. With the new laser system an important increase in resolution and sensitivity is expected, and an on-line version of the experiment seems feasible. But one should not expect, of course, that for a refractive element the same limits can be reached as for the easily handled volatile elements.

II.2 The Photon Burst Method

The second example is an A2-B1 combination with a special trick to suppress background $^{10}$A). A beam of barium atoms emerging from an oven is collimated and excited at right angles (that is dopplerfree) by a laser beam. The interaction region is imaged by an elliptical mirror with a large solid angle onto a photomultiplier (fig. 3).

Fig. 2 Simplified level scheme for polarization spectroscopy

Photon burst method: Greenless etal Minneapolis

Fig. 3 Schematics of the photon burst method $^{10}$. 

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If an atom is passing the laser beam and the transition in question excited to saturation, it radiates fluorescence quanta at the rate of the reciprocal lifetime of the upper state, that is typically $10^8$ s$^{-1}$.

The first photon detected at the photomultiplier, opens therefore a coincidence gate waiting for the next. This multiplicity filter counts only correlated photons and rejects uncorrelated background events which may stem from laser stray light, photomultiplier noise, radioactivity etc. The length of the gate has to be optimized to the signal to background conditions and, of course, to the duration of the photon burst which is limited by the time of flight through the laser beam or the time it takes to pump optically the atom into another nonabsorbing state. The minimum length is of course the lifetime of the radiative state. Asking for a multiphoton coincidence at low laser power the resolution will fall even below the natural linewidth, since the n-photon absorption and re-emission probability is proportional to the $n^{th}$ power of the absorption profile, under these conditions. Extrapolating from the experience of the pilot experiment on stable barium the method is expected to be very sensitive, in favourable cases down to the flux of approximately 10 atoms/s crossing the laser beam. In collaboration with the proposers from Minnesota State University (Grennless et al. 10a) on-line experiments are being prepared at Argonne by C.N. Davids et al. and Davis et al. (Iowa State University) 10b as well as at Rochester by D.L. Clark et al. 10c.

II.3 The Multiphoton Ionization Method

The next topic to be discussed is an A2-B2 scheme based on the multiphoton ionization method developed at the Institute for Spectroscopy in Moscow by V. Letokhov and his collaborators 11 (fig. 4).

By stepwise excitation the atom is raised to an highly excited, so-called Rydberg state close to the ionization limit. There the valence electron is bound so loosely that it may easily be detached in an electric field, Since the transition matrix element to the Rydberg state is rather small (it decreases roughly with the third power of the main quantum number) the second step requires high spectral power density of the laser. But the authors have shown that, at least with pulsed lasers, the second step and hence the ionization efficiency may be saturated (example Yb). This selective ionization method is of great analytical value; in particular it may be used for on-line detection and spectroscopy of unstable isotopes. Fig. 5 shows a simplified scheme of an experiment under preparation at the on-line mass separator at Gatchina (G. Alkhazov et al.) in collaboration with the Moscow group 12. Radioactive atoms emerging from the target oven are radiated by two simultaneous laser shots at a repetition rate of about 10 KHz. Multiplied with the average time of flight through the interaction zone, a duty cycle close to 1 is achieved. The Rydberg atoms are ionized in the field of the extraction electrode, mass separated, and counted.

II.4. Collinear Laser Spectroscopy

Detection by multiphoton ionization may also be used in collinear laser spectroscopy as proposed by a group of the Laboratoire Aimé Cotton at Orsay 13. Alain Huber and his collaborators are preparing an experiment on these lines 14.

The last example is of type A2-B1, collinear laser spectroscopy with a fluorescence detection. Since the pilot experiments which were performed at Mainz (first on stable alkalies 15), then on fission alkalies 16, 17), on-line mass separated at the TRIGA reactor 18), have been presented at earlier meetings 3, 4), I will confine myself to a description of the experiment we set up at the ISOLDE in CERN. So far it yielded results in the barium chain (122S3/2 146) and quite recently in the ytterbium chain (156S43/2 176). This method has been modelled particularly for on-line applications, trying to avoid draw backs connected with the procedure of transforming the mass separated ion beam into a spectroscopically
useful sample. In the cases discussed so far, the ions have been converted to thermal atoms by stopping and evaporating them, forming either an atomic vapour in a cell or a collimated atomic beam. In the former case, chemical stability is a restrictive condition; in the latter, a large factor is lost by collimation. The original idea behind collinear laser spectroscopy was simply to avoid these problems by using the mass separated beam directly as the spectroscopic sample. Since at typical ion velocities of 107 cm/s the interaction time with the laser in a crossed geometry would go down to about 10-8 s, a collinear superposition of laser and ion beam seemed favourable. Thereby the interaction time and the length is easily increased by a factor of 100, observing fluorescence light from a pathlength of 10-20 cm. This is achieved by means of a cylindrical lens which images the beam onto the entrance slit of a light pipe, that adapts the image to the geometry of the photocathode (see fig. 6).

source temperature of 2000 K and an acceleration voltage 10 kV typical for ISOLDE. Under these conditions the rest Doppler width of the green barium resonance line at 535 nm, for example, would be about 1 MHz, far below the natural line width of 19 MHz.

Kaufman also added the charge exchange cell to the concept, containing an alkali vapour which neutralizes the beam. This is of importance, since most ions do not have resonance lines in the visible. It is presumed, of course, that the charge exchange process does not disturb the velocity distribution. This is actually the consequence of the large cross section of about 10-14 cm2 which exceeds the kinetic cross section by 2 orders of magnitude.

If the charge exchange is nonresonant, however, the beam energy is changed by (almost) exactly the energy defect $\Delta E$. (The energy transferred in forward scattering to the target atom is of the order $(\Delta E)^2$ only which is negligible.)

The actual line width depends, of course, crucially on the performance of the system, namely:

(i) the stability of the acceleration voltage which could be improved to a limit of about 1 V \cite{23}.
(ii) The energy spread of the ion source; in case of surface ionization we found values between 1 and 4 Volts, for a plasma source about 20 Volts.
(iii) The angular emittance of the beams, which were of order 1 mrad and did not contribute significantly.

The determination of the atomic transition frequency and hence of the isotope shift and hyperfine structure requires the precise knowledge of the laser frequency, the acceleration voltage and the atomic mass. Instead of calibrating the two former quantities independently, it is safer and easier to run alternatively stable isotopes
through the apparatus for calibration. For this purpose the ISOLDE magnet is switched periodically from one to the other mass and an appropriate voltage \( V \) \( \leq 5 \) K is fed to the post acceleration stage just in front of the charge exchange cell. This correction voltage generates a Doppler shift such that the resonances of the different isotopes coincide in the lab system, that is for one and the same laser frequency. Scanning of the resonance, finally, is achieved by adding another small voltage stepwise to the charge exchange cell. Thus it is sufficient to run the laser at constant frequency in a stabilized but uncalibrated mode. Fig. 7 shows a set of resonances from three even barium isotopes recorded simultaneously in the manner described above.

The strong ground state transition at 3980 Å with oscillator strength \( f_{\omega} \) is just outside the reach of cw dye lasers, whereas the laserwings convenient intercombination line at 5558 Å is very weak. In this situation it is helpful that charge exchange also populates excited states, for instance the metastable \( ^3P_1 \) state. From there starts a strong absorption line at 7702 Å to a \( ^1S_0 \) state. The re-emission has a branch cascading over the \( ^3P_1 \) state down to the ground state. The photomultiplier picks up only the second, green line of the cascade, blocking the red line by its spectral response; thereby stray light from the laser is suppressed. A similar situation is met in many spectra, especially those of group II elements and noble gases.

Avoiding charge exchange the Marburg group exploited the fact that, in suitable cases ions leave the source to a reasonable fraction already in metastable states from which they may be conveniently excited \( 25a \).

On the same lines a group in Leuven performed an off-line experiment on radioactive \( ^{136} \)Ba \( 25b \). These two groups and one in Oakridge \( 26 \) are preparing collinear on-line experiments with fluorescence detection, whereas the Orsay group \( 13 \) and Huber et al. in Mainz \( 14 \) are developing a particle counting version, as mentioned above.

III. Discussion of Isotope Shifts

The nuclear physics contained in the results concerns spins and moments obtained from the hyperfine structure and charge radii from the isotope shift. Since Ekström is going to discuss spins and moments in the following paper \( 27 \), I will confine myself on isotope shifts and charge radii.

The isotope shift (IS) between the centers of gravity of the HFS multiplets of a spectral line is the sum of the so-called field shift \( \delta \omega_F \) and the mass shift caused by the change of the reduced mass of the electrons. It may be written as

\[
\delta \omega = \delta \omega_F + \frac{K(A'-A)}{A' A}
\]

In order to isolate the field shift it is necessary to know the mass shift, which is not very important in the heaviest elements but predominates in the light ones. For one electron spectra the calculation is trivial (normal mass shift), but it becomes very difficult when accounting for the correlation of all the electron momenta in complex spectra (specific mass shift). At this point it is interesting to note that collective effects in atomic spectra are barely known and understood in contrast to the nuclear case. However, specific mass shifts seem to be small in \( \alpha \) transitions which, on the other hand, display the largest field shifts.

The field shift is connected with the change of the nuclear mean square charge radius \( \delta r^2 \) by

\[
\delta \omega_F = \frac{2}{3} e^2 \delta r^2 \chi(0) |\psi(0)|^2 \delta r^2 \, A' A
\]

where \( |\psi(0)|^2 \) is the change of electron
density at the nucleus in the electronic transition. This non-relativistic equation holds for light elements. For high Z it has to be corrected by having a well known Z- and A-dependent numerical factor in front of it, and by contributions of higher radial moments of the charge distribution reaching about 6% of $\delta r^2$ at Z=80. As to the calculation of $\delta |\Psi(0)|^2$; again quantitatively correct calculations are still missing. The difficulties stem predominantly from the polarization of the inner, closed s-shells by the valence shell. In spite of the increasing importance of the data, theoretical efforts in calculating the atomic parameters have slowed down in the past years, unfortunately. In a number of cases comparison with isotopic shift data from muonic spectra and electron scattering is helpful in resolving or at least diminishing this uncertainty, as will be shown in the paper by Fricke in this session 28).

Fig. 9 displays $\delta r^2$ values as a function of N in the vicinity of the magic neutron number N=82. They are plotted relative to the magic isotope.

The similarity in the gross structure (trend of slopes) and in the finestructure (odd-even staggering) between the different elements is striking. For the long isotopic chains of caesium and barium the interpretation of the gross structure has priority. It is dominated by the kink of the slope at the magic number. In order to understand the nature of this shell effect we have a look at the straight, solid line which represents the expansion of the nuclear radius according to a non shell model, namely to Myer's droplet model 29). The difference of the experimental points to this line increases symmetrically to the distance from the magic number towards larger radii. The effect of the open shells could thus be either diluting or deforming the nuclear charge distribution. Since the former costs too much energy (the increasing mode lies higher than the vibrational one) we try an interpretation of the extra radius in terms of nuclear deformation by using the formula

$$\delta r^2 = \frac{2}{3} R_0^2 \delta R + \frac{3}{4 \pi} R_0^2 \delta \rho^2$$  \(6\)

In this formula the first term represents the variation of $r^2$ with mass number for a homogeneously charged liquid drop of radius $R_0$, corrected by the factor $\delta$ which the droplet model predicts to be 0.5 in an isotopic chain. The second term accounts for the effect of the nuclear deformation parameter $\delta$ which enters via its mean squared value. Hence there is no discrimination between static and dynamic deformation like in the case of the BE2 value.

It is therefore nearby to compare deformation parameters derived from the isotope shift with those from BE2 values as shown in fig. 10.

(On the neutron rich side BE2 values for comparison are lacking.) It seems justified therefore to regard nuclear deformation as being the main shell effect on the mean squared radius. A compilation by Myers and Schmidt comes to the same conclusion 31). In lighter mass regions, however, the BE2 value seems not to exhaust the full collective effect on $r^2$; other multipolarities can contribute as well (compare the following papers by Thibault 2b) and Fricke 28). A general change of the surface thickness may also be considered 22).

The long chains of isotopic shifts, extending far off stability, which are
available now in several regions of the nuclear chart provide us for the first time with the possibility to gain also isotonic shifts, spanning over large distances. Fig. 11 gives the systematics between 37Z258 with the elements rubidium, cadmium, caesium and barium as cornerstones, all of them investigated by on-line laser spectroscopy at ISOLDE.

![Systematics of isotonic shifts for even neutron numbers between N=72 and N=54. Cd is always in the origin.](image)

In preparing these plots one needs the absolute $<r^2>$ value of at least one isotope for each element. This is usually available from electron scattering or mesic spectra, except for caesium where we took for $^{137}$Cs a value interpolated between $^{136}$Xe and $^{138}$Ba. For comparison with the droplet expectation the contribution of deformation has been subtracted with the help of known BE2 values and equation 6. The agreement is surprisingly good.

In the same way one can construct plots of isobaric shifts, fig. 12 showing the example of mass 120. By exchanging 7 neutrons by protons $<r^2>$ increases by 1 fm$^2$ instead of staying constant as expected from the naive liquid drop model with homogeneous-distributed charge. If we exclude again a simple blow up of nuclear matter for reasons of binding energy we have to conclude that, in going from cadmium to caesium, the radius of proton distribution increases on cost of the radius of the neutron distribution and vice versa. This phenomenon is also reproduced in microscopic calculations 34).

The recent ytterbium data have not yet been fully analyzed. Fig. 13 shows some raw data, the IS of the even isotopes in the $^{7702}$ Å line between mass numbers 156 and 176.

![Drawing the droplet line (converted into a frequency scale) along the heavy, well deformed isotopes, one observes that towards the lighter isotopes the experimental curve is gradually bending down from the droplet line. It means that the nuclei shrink faster than predicted. Again this may be explained by a gradually decreasing deformation towards lower neutron numbers. In fact, the shape transition, which occurs in the lighter rare earths as a sharp jump between N=88 and N=90 35) is almost washed out. Also in caesium and barium it is not recognized any more (compare fig. 9 and 10). Obviously, the phase transition phenomenon is restricted to a certain Z interval around Z=60. It would be interesting to define it better by more measurements above Z=56 and below Z=70.](image)
IV. Conclusion

This paper aimed at drawing attention to some typical questions in the field of on-line laser spectroscopy, regarding recent developments in the experimental techniques as well as in the interpretation of their results. Being incomplete, the paper should not be regarded as a review paper and, moreover, the selection of topics is certainly biased by the authors' activities. I therefore like to refer again to other papers in the field and especially to the other speakers in this session.

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DISCUSSION

D. Clark: How many atoms must be present in the cell for the polarization spectroscopy measurements? How many must be in the laser beam to produce a useable signal and is a large fraction of the sample on the walls of the cell?

H.-J. Kluge: 10^{13} atoms of ^{197}Au were present in the cell having a volume of about 30 cm^3. The interaction region (crossing of saturation and probing beams) was in the order of 1 mm^2. The fraction of the atoms in the vapour phase is determined as of the order of 1\% - 10\%.

D.E. Marmick: It is difficult to give a general answer to the question of sensitivity in laser spectroscopy experiments. In a recent review, Feld and I (Ann.Rev.Nucl.Part.Sci. 29 (1979) 411) discussed various techniques of high resolution laser spectroscopy which can be applied or modified for nuclear structure studies. The method of choice depends on many factors - yields, backgrounds, atomic oscillator strengths and structure, etc. The highest sensitivity can be obtained with radiative detection as demonstrated, for example with our 2^n\text{Na} experiment (Phys.Letters, Dec. 1979), where equilibrium concentrations of only a few atoms/cm^2 were present.

E.W. Otten: I agree that until present the highest sensitivities have been reached by radiative detection as proved by many experiments, the old RADOP experiments, yours, and to the extreme by the LINUP experiment on the fission isomer 2^n\text{Am}.

P.G. Hansen: Does the IS calculation by Myers, which you compared with the isotone shifts, include shell deformation effects?

E.W. Otten: The radii derived from different versions of Myer’s droplet model differ only very slightly. The main shell effect on the rms radius seems to be the nuclear deformation induced by the open shells. Here <\delta> exhausts most of the collective contribution to the radius, at least in the Ca-region. Also other multipoles, if strong, may contribute. There is evidence for an octupole contribution in the Ca-region.

G. Schatz: To what extent is the comparison with the droplet model influenced by the uncertainties of the mass effect?

E.W. Otten: The extraction of isotonic and isobaric shifts from IS-data far off stability is particularly endangered by the uncertainties in the evaluation of the IS which introduce an error in the scale of 6\%\text{r}^2. Over a distance of, say, 2N=15 from the reference isotope at stability (from which you take the absolute <\text{r}^2> determined e.g. by muonic spectra) a 20\% scaling error would integrate up to more than the radial shift to the neighbouring isotones, that is lines of <\text{r}^2> for different Z as a function of N would cross somewhere. This has not been observed, however, all known isotopic lines are well ordered. From this self-supporting net of consistent data one may conclude that the evaluation of isotope shifts is so far, indeed, a pessimistic thought.

A. Müller: In the figures which have been shown by Prof. Otten, the droplet model of Myers has been used not including corrections accounting for deformation and shell effects. That is for describing the spherical part of the nucleus and ascribing the remaining change in mean-square charge radii to quadrupole deformation.

K. Elsäßer: The odd-even staggering of radii to the left of N=82 is very characteristic for spherical shell-structure (there are similar effects in the separation energies) whereas the typical increase at the right-hand side corresponds to the deformation which is also observed in the middle-line of the periodic table (the separation energies show a similar change).

E.W. Otten: The regularity of odd-even staggering, repeating in all isotopes of the region (Ce, Cs, Ba), is remarkable, indeed. Rehal and Sorensen (1971) gave an explanation of it in terms of the blocking effect of the unpaired particle diminishing the average deformation. Their calculations, however, scatter a lot from the experimental data. But in their picture of hindered deformation I see a similarity to another odd-even effect, namely the reduced fission width of heavy-odd and odd-odd nuclei with respect to their even neighbours. Regarding your second remark about the deformation, developing of the closed shell, the isotopic shift data far off stability add new information in so far as the path into deformation is studied for the first time as a function of N alone at constant Z.