Status Report and Request for Beam Time

Experiment IS 80:

STUDY OF NUCLEAR MOMENTS AND MEAN SQUARE CHARGE RADI P BY COLLINEAR
FAST-BEAM LASER SPECTROSCOPY

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1. Introduction

In our Status Report of 1982 (FSCC/82-61/M123), we gave a review of the optical spectroscopy on unstable isotopes, the experimental features and the prospects of the collinear laser-fast atomic beam technique at ISOLDE and the physics programme of the present experiment for the near future.

The emphasis of this programme was put on the measurement of nuclear spins, moments and isotopic changes of the mean square charge radii in the rare-earth region, where the Ta-foil target offers the unique possibility of studying neutron-deficient nuclides for the whole sequence of elements between Sm and Yb (62 ≤ Z ≤ 70). These nuclides cover the neutron numbers around the shell closure at N = 82, the transitional region 82 < Z < 90 and the region of strong deformation N > 90. Of particular interest is the sharp onset of strong deformation, localized to the neighbourhood of the semi-magic proton number Z = 64 (Nd, Sm, Gd, Dy) and not observed for the lighter and heavier elements (Ba and Yb).

The corresponding region of nearly-spherical to strongly-deformed nuclei Z > 82, N > 126 is rather unexplored because all these nuclei are unstable and only a few of them are commonly available. As a first step, we have made measurements in the atomic (RaI) and ionic (RaII) spectrum of Ra, yielding the hyperfine structures and isotope shifts of several transitions which have to be analyzed consistently as to describe the atomic parameters in terms of the nuclear observables. This analysis in the heaviest element with a simple two-valence-electron spectrum is also a valuable test of the theory of atomic structure including relativistic corrections.

In addition, we have studied selected nuclides of particular interest, such as $^{182}$Hg and $^{207}$Tl, partly in combination with tests of a modified experimental set-up and in collaboration with other groups. The long-term improvements are directed mainly towards a more sensitive detection scheme including ion counting. We have made further promising tests of the state-selective electron stripping for the application to rare gases (see Appendix). The Orsay and Mainz-GSI groups are aiming at the cw laser excitation to
Rydberg states with subsequent field ionization, whereas the Moscow group (V.S. Letokhov et al.) is studying the direct stepwise ionization with pulsed dye lasers pumped by a copper vapour laser with high repetition rate. These new schemes will probably be less versatile than the present fluorescence detection technique, but lead to a considerably improved sensitivity in a number of cases. Future collaborations are discussed between the groups involved (cf. Proposals submitted in parallel).

2. Details on measurements since October 1982

2.1 Rare-Earth Region

Our first review included the measurements on the even-Z elements Yb (156 ≤ A ≤ 176), Er (150 ≤ A ≤ 170) and Dy (146 ≤ A ≤ 164), as well as the complementary work on Ba (122 ≤ A ≤ 146) of which the neutron-rich isotopes cover the same interesting range of neutron numbers. As proposed in our last beam time request, we have extended these studies to Gd and Eu and complemented the Er data by measurements in the odd-A isotopes.

a) Erbium: The gain in sensitivity, required for the measurements of hyperfine structures and isotope shifts in the odd-A Er isotopes, was achieved by making accessible the strong resonance transition $4f^{12}6s^23^D_6 \rightarrow 4f^{12}6s6p \left(^1P\right)$, $J = 5$ at 4151 Å in the far blue. Although its upper state is strongly mixed, the nuclear moments are extracted from the hyperfine structures by the usual calibration with stable isotopes, and the isotope shifts can be related to the earlier measurements in a clean $s^2$-sp transition by a King plot. During the laser tests with the dye Stilben I and special mirrors, it has turned out that the wavelength range of cw lasers can be extended to the $1S_0 - 1P_1$ resonance line of Yb (3988 Å). This will be helpful not only for sensitive measurements on additional neutron-deficient Yb isotopes, close to $N = 82$, but also for the other rare-earth elements which have corresponding transitions in the same range.

b) Gadolinium: The Gd ground state belongs to a fine structure multiplet of the configuration $4f^75d6s^2$, and many low-lying states are populated in the neutralized beam. Sensitivity tests have led to the choice of the transition $4f^75d6s^29^D_6 \rightarrow 4f^75d6s6p \left(^1P\right)$, $J = 5$ at 4226 Å, for an attempt to identify the neutron-deficient isotopes of Gd in the isobaric mixture of nuclides from the Ta-foil target. (Gd is the least volatile element in
this region and only longer-lived isotopes can be expected to escape from the target. They are difficult to detect by nuclear spectroscopy in the presence of the shorter-lived isobars and their decay products.) We found appreciable yields between $10^6$ and $10^8$ atoms/sec for $^{142-152}$Gd and measured the isotope shifts in this sequence, as well as the narrow hyperfine structures of the odd-$A$ isotopes. The results indicate a behaviour of $\delta <r^2>$ similar to the neighbouring elements, but the final analysis requires further measurements on the stable isotopes for which we need a stable Gd beam from ISOLDE for about 2-3 days. The partially resolved hyperfine structures yield rather accurate magnetic moments, but no quantitative information about the small quadrupole moments.

**c) Europium:** As a prominent odd-$Z$ candidate in this region we have chosen Eu ($Z = 63$) because of its single-proton hole in the $2d_{5/2}$ subshell and its relatively simple electronic structure. Of the three strong resonance transitions $4f^76s^2 \, 8s_{5/2} \rightarrow 4f^76s6p \, 8p_{5/2,7/2,9/2}$ (4662 $\AA$, 4627 $\AA$, 4594 $\AA$) the two latter have the best resolved hyperfine structures including strong quadrupole interaction. Measurements in these two transitions have been performed for the sequence $^{140-153}$Eu. All these isotopes exhibit a stable $5/2^+$ proton configuration for which the magnetic moments and spectroscopic quadrupole moments can be followed in the odd-$A$ isotopes over a wide range of neutron numbers (Fig. 1). The quadrupole moments and $\delta <r^2>$ give a consistent picture of the nuclear deformation, with a minimum for $^{145}$Eu ($N = 82$) and the well-known strong increase between $^{151}$Eu and $^{152,153}$Eu ($N = 88-90$). This step is clearly more pronounced as in the neighbouring even-$Z$ elements (Fig. 2).

2.2 Radium Region

We have studied 18 isotopes in the range $^{208-232}$Ra (Fig. 3). They were produced by spallation of $^{238}$U. Without anticipating a final analysis of the electronic factors by atomic-structure calculations we can already give conclusive results. In the strongly-deformed region ($A > 220$), the spectroscopic quadrupole moments are close to the strong-coupling limit as compared to the intrinsic quadrupole moments known from B(E2) values. The spin sequence and the corresponding magnetic moments support the hypothesis of a static intrinsic octupole deformation gradually decreasing from $^{221}$Ra to $^{229}$Ra / 1, 2/. This is also reflected in an inversion of the odd-even staggering of the
isotope shift for $^{221,223,225}\text{Ra}$, and in the absolute values of the mean square charge radii with respect to the nearly-spherical $^{214}\text{Ra}$ (Fig. 4). The transitional isotopes $^{215-219}\text{Ra}$ are very short-lived $\alpha$-emitters and thus inaccessible to the on-line isotope separation and collinear beam-laser techniques. Below the magic neutron number $N=126$, the moments of the odd-$A$ isotopes $^{211}\text{Ra}$ and $^{213}\text{Ra}$ are well described by $(f_{5/2})^{-1}$ and $(p_{1/2})^{-1}$ neutron configurations of the shell model.

2.3 Selected Nuclides

a) Mercury: In the Hg spectrum, the transition $6s6p\,^3P_2 - 6s7s\,^3S_1$ at 5461 Å is most convenient for collinear laser spectroscopy, whereas the earlier experiments in optical cells used the resonance transition $6s^2\,^1S_0 - 6s6p\,^3P_1$ in the UV at 2537 Å. Preliminary measurements for a number of isotopes were performed in collaboration with the Mainz-GSI group (G. Huber et al.) and combined with a test of the essential parts of the new collinear-beam apparatus for the GSI isotope separator. A first success was the long-awaited isotope shift measurement in $^{182}\text{Hg}$ which shows that this nucleus follows the line of nearly-spherical doubly-even isotopes, whereas the odd-$A$ isotopes in this region are strongly deformed. A future continuation will be concentrated on the more neutron-deficient isotopes, the isomers in $^{183,185}\text{Hg}$, and on the long sequence of $13/2^+$ isomers for which theoretical predictions of the quadrupole moments can be tested with higher precision.

b) Thallium: $^{207}\text{Tl}$ is the only known nucleus with a $s_{1/2}$ single-proton hole ground state in a doubly-magic ($^{208}\text{Pb}$) core. The magnetic moment of such a state should be unaffected by meson-exchange currents and thus provide a sensitive test of the core-polarization corrections to be applied to the Schmidt values. We have found that this nucleus can be produced at ISOLDE in uniquely high yields, because it is the only Tl isotope which is fed in a fast $\alpha$-decay chain from the predominant spallation products of the $^{232}\text{Th}$ target ($^{219}\text{Fr} \rightarrow ^{215}\text{At} \rightarrow ^{211}\text{Bi} \rightarrow ^{207}\text{Tl}$). The yield of more than $10^8$ atoms/s, obtained in a recent target test, was sufficient to measure the hyperfine structure of the transition $6s^26p\,^2P_{3/2} - 6s^27s\,^2S_{1/2}$ (5350 Å). The magnetic moment of $^{207}\text{Tl}$ was extracted by comparison with the stable isotopes. There is a second magnetic moment of considerable interest for theory, namely that of the 1.3 s $11/2^+$ isomer in $^{207}\text{Tl}$. A combination of the
magnetic moments of the $h_{11/2}^-$ proton state in $^{207}$Tl and the $h_{9/2}^-$ proton state in $^{209}$Bi should give a clean information about the influence of the meson-exchange currents on the orbital g-factors (cf. I. Bergström, A. Kerek, C. Ekström: Letter of interest for the SIN-ISOLDE project, PSCC/80-56/134). The $11/2^+$ isomer has to be directly produced, and the yield estimates are about $10^5$ atoms/s. A hyperfine structure measurement with this low-intensity beam is conceivable. However, the yield should be better established for a successful attempt by optical spectroscopy.

Further systematic experiments on Tl — particularly on the very neutron-deficient isotopes — should wait for a higher production rate to be expected with the high-intensity $^3$He beam from the SC.

3. Planned Experiments

We are planning to continue the experiments in the regions of special interest described under 2.1 and 2.2. This work will also include complementary measurements on the isotopic chains that have been studied so far.

3.1 In the rare-earth region the data around the shape transition $N \approx 90$ are rather complete for the even-Z elements between Ba ($Z = 56$) and Yb ($Z = 70$). Of the odd-Z elements, only Eu has been studied systematically. For a further exploration of the even-Z/odd-Z differences in the isotope shift curves, we propose the cases of Ho ($Z = 67$) and Tm ($Z = 69$). These elements lie between Dy, Er and Yb, where the sudden shape transition is dissipating for the even-Z elements. It is not known how the unpaired proton influences this effect. Another important feature will be the behaviour of the odd-even staggering of the isotope shift. This phenomenon is still not understood quantitatively, but we have found interesting irregularities and feel that a more systematic study may provide the key for an explanation.

The Tm isotopes are presently being studied with low resolution by the Moscow-Leningrad collaboration at Gatchina. Therefore, we should wait for their first results. With unresolved hyperfine structures, they may have difficulties in the evaluation of precise isotope shift data. Apart from this, a high-resolution experiment is necessary to measure the nuclear spins and moments. The known spin sequence for the isotopes with $N > 88$ differs remarkably from those of the lighter elements, and the character of their ground states should be determined by a measurement of the magnetic moments.
In particular, the appearance of a $1/2^+$ ground state in the transitional nucleus $^{157}$Tm (N = 88) is difficult to understand. An experiment on Tm should also be considered as a first step to the application of the more sensitive multiphoton ionization technique proposed for this element (cf. parallel Proposal by Letokhov et al.).

A particularity of Ho is the large number of isomeric states — both in the nearly-spherical and strongly-deformed regions. Their moments and isomer shifts should reveal important aspects of their nature. On the other hand, these isomers and the high electronic angular momenta involved in the optical transitions complicate the measurements and their analysis (e.g., the ground state configuration is $4f^{11}6s^24_{15/2}$).

The beam time request is made under the assumption that only one of these two cases will be chosen for an experiment in the near future, depending on the results of the Gatchina experiment and the further analysis of our own data.

Complementary measurements to the ones discussed under 2.1 will be performed particularly on Yb and Gd. As mentioned above, the strong $6s^2\,^1S_0 \rightarrow 6s6p\,^1P_1$ resonance transition of Yb at 3988 Å has been made accessible. This will enable to continue the Yb measurements to more neutron-deficient isotopes, beyond $^{156}$Yb, towards the neutron-shell closure at $^{152}$Yb. The next step in sensitivity will be multiphoton ionization detection (cf. parallel proposal) by which we can hope to include $^{152}$Yb and relate the $\langle r^2 \rangle$ values of the whole isotopic chain to a nearly-spherical nucleus. The measurements on Gd should be repeated in a transition with larger hyperfine structure and less configuration mixing, so as to resolve the quadrupole interaction and to facilitate the analysis of the isotope shifts. This work can partly be done with stable beams which we also need to find a transition with appropriate hyperfine structures.

The following section will mainly deal with the rare-gas element Rn. Here, we would like to mention that corresponding measurements on the neutron-rich Xe isotopes (including the possibility of detection by state-selective collisional re-ionization) belong into the context of the "rare-earth region" (N = 82 - 90). We envisage such an extension to Z = 54 and expect an influence of the neighbouring Z = 50 proton-shell closure on the development of deformation.
3.2 In the Ra region we expect to study the isotopes of Rn produced by spallation from $^{232}$Th or $^{238}$U. The optical transitions convenient for high-resolution spectroscopy are starting from the metastable $J=2$ state of the configuration $6p^57s$, populated predominantly in the charge exchange with Cs. As in the case of Ra, no hyperfine structures or isotope shifts of this element have ever been measured. Therefore, we have to investigate a number of transitions, in order to extract nuclear moments and mean square charge radii by a parametric analysis of the electronic factors in terms of single-electron contributions.

The nuclear spins will be directly accessible, and already the spin sequence of the heavier isotopes should reveal a tendency in the development of octupole deformation. More detailed information will be obtained from the magnetic moments and the isotope shifts including the effect of the octupole degree of freedom on the odd-even staggering. For the light Rn isotopes we expect to reach lower neutron numbers than for Ra, where the collective effects in the isotope shift are very small down to $N=120$. Altogether, the sequence of Rn isotopes ($Z=86$) will link the Ra data ($Z=88$) with those of the proton-magic Pb isotopes ($Z=82$) investigated recently by the Karlsruhe group /3/. Both, Ra and Rn are the even-$Z$ neighbours of Fr, of which the neutron-rich isotopes are being studied in a Orsay-Mainz collaboration under IS 70.

3.3 Ion detection: The initial Rn experiments including the atomic physics part will be performed with the conventional set-up using fluorescence detection. In parallel, we shall develop and test a modified apparatus based on the ion detection scheme described in the Appendix: The laser excitation from the metastable state (-4 eV) is followed by a decay cascade to the ground state (-14 eV), and the large difference in the ionization energies of these two states involves rather different ionization cross-sections for collisions with target-gas molecules. Thus the optical pumping to the ground state can be exploited for a simple, inexpensive and efficient ion detection which is particularly suitable for the noble gases. We expect a considerable gain in sensitivity and no serious disturbance by stable-beam contamination in the heavy-mass region of Rn. Therefore, we are planning a continuation of the Rn measurements in the less abundant isotopes ($<10^5/s$) as a first application of this scheme. We may prefer to start with a test on Xe of which also stable isotopes are available.
4. Beam time request

4.1 Stable beams

As pointed out in our last status report, it is essential for the in-beam laser spectroscopy experiments to have sufficient test time with stable beams. These beams are used not only for technical development, but also for complementary measurements and calibrations. Therefore, we ask for about 20 shifts of stable beams in parallel with the proposed programme. This time should be divided into small portions of about 1 - 3 shifts.

4.2 Radioactive beams

For the experiments described under 3.1 - 3 we request 50 shifts of radioactive beams. This time will be shared approximately as follows:

1. Rare-earth elements from the Ta-foil target
   15 shifts
2. Rn from the ThO₂ (ThC₂, UO₂, UC₂) target -- fluorescence experiment
   20 shifts
3. Rare-gas ion detection on Rn (Xe)
   15 shifts
   50 shifts

The latter request is tentative, and we shall submit a new beam time request after the first successful run.

We are prepared to start with the Rn experiment and come back to the rare-earth programme whenever the Ta target will have additional users. Smaller complementary work to earlier experiments can be incorporated into the runs of other groups and will use only a small part of our beam time allocation. In addition, we would like to continue having some flexibility in studying cases of topical interest (cf. ²⁰⁷Tl experiment under 2.3).

Our beam time request should cover a period of about 1½ years. We are aware of the difficulty to run this programme in parallel with those of the other proposals using essentially the same apparatus. Therefore we shall accept delays in a part of this programme and give some priority to the experiments planned in collaboration with other groups.
References


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Stable Intrinsic Octupole Deformation reflected in the Moments and Charge Radii of Radium Isotopes
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Isotopes Shifts, Changes in the Nuclear Mean Square Charge Radii and Deformations of Radium Isotopes in the Mass Range 208≤A≤232
S. A. Ahmad, W. Klempt, R. Neugart, E.W. Otten, K. Wendt and C. Ekström, to be published
Fig. 1: Plot of magnetic ($\mu$) and quadrupole ($Q_s$) moments of $5/2^+$ ground states in Eu as a function of neutron number. The maximum of $\mu$ and the minimum of $Q_s$ correspond to the neutron-shell closure $N = 82$. 
Fig. 2: Change of mean square charge radii $\delta \langle r^2 \rangle$ in the isotopic chain $^{140-153}\text{Eu}$, with $^{145}\text{Eu}$ taken as reference point. The change in deformation $\delta \langle \beta^2 \rangle^{1/2}$, with respect to $N = 82$, is indicated by the parallel lines, the slope of which is given by the droplet model.
Fig. 3: Positions of the resonances and their centres of gravity (full dots) for the sequence of $^{208-232}$Ra isotopes in the RaI transition at 4827 Å. The nuclear spins are given at the top of the figure.
Fig. 4:
Change in the mean square charge radii $\delta <r^2>$ and development of deformation for the isotopic chain of $^{208-232}$Ra. The crosses give the values expected from the droplet model with deformations from experimental B(E2) values. They do not contain contributions from higher-order moments.
Appendix

A SENSITIVE ION-DETECTION SCHEME
FOR COLLINEAR LASER-FAST ATOMIC BEAM SPECTROSCOPY ON RARE GASES

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The rather general applicability of the collinear laser-fast atomic beam technique is based on the neutralization of ion beams by charge exchange with alkali vapours. Thereby, the beams are prepared in (ground or metastable) atomic states about 4–6 eV below the ionization threshold from where resonance excitation is possible with visible light (1.5–3 eV), corresponding to the spectral range of cw dye lasers /1/. This is particularly important for the rare gases whose first excited states lie about 10 eV above the np$^6$ 1$S_0$ ground states. The metastable J = 2 states of the configuration np$^5$(n+1)s -- designated (n+1)s $[\lambda/2]_2$ -- about -4 eV from the ionization threshold are efficiently populated in charge exchange with caesium. They are connected to the states of the configuration np$^5$(n+1)p by strong transitions in the red.

Generally, the excitation is detected by counting the fluorescence photons. This involves a relatively low detection efficiency and background problems mainly due to stray light, molecular excitation in the alkali vapour and radioactivity. The practical sensitivity limits (typically between 10$^4$ and 10$^6$ atoms/s) depends on the strengths of the transitions and the complexity of the hyperfine structures.

A considerable gain in sensitivity is expected from non-optical detection schemes, for example the ion counting after stepwise laser ionization. This technique is well developed for pulsed lasers with high peak power but limited duty cycle /2,3/. For cw lasers, the problems of power limitation remain to be solved.

We have designed an alternative and much simpler scheme which -- at least for the rare gases -- should reach the same goal. It is based on the selective electron stripping from the metastable atoms as they pass through a molecular gas. The beam, after neutralization in the caesium-vapour cell and removal of the remaining ions, interacts with the laser light and then passes a differentially pumped gas cell after which the re-ionized part is deflected into a Faraday cup or an ion counter.