NEW TECHNIQUES AND RESULTS OF COLLINEAR LASER SPECTROSCOPY
RADON, XENON AND HOLMIUM

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ABSTRACT

The moments and radii of extremely neutron-rich radon and xenon isotopes have been investigated by collinear fast-beam laser spectroscopy. The experiment has become possible owing to the breakthrough in sensitivity achieved by the detection of optical pumping via state-selective collisional ionization. The results are discussed within the systematics of nuclear shapes around $N=136$ and $N=90$. In the latter context measurements on the neutron-deficient holmium isotopes give new details about the development of nuclear deformation.

INTRODUCTION

The success of laser spectroscopy in providing moments and radii of nuclei far from stability has provoked new efforts to gain significant factors in sensitivity. A few promising approaches are based on the collinear fast-beam technique which - owing to its conceptual simplicity - can be applied to nearly any beam from a conventional on-line isotope separator.

The principle is to measure hyperfine structures and isotope shifts in a laser-induced optical transition from a properly prepared state of the ion or neutral atom. Narrow absorption linewidth and Doppler-tuning ensure the desirable resolution and precision. Concerning sensitivity a crucial point is the detection. The universal concept of counting fluorescence photons suffers from scattered laser light, collisional excitation and radioactivity. This is where particle detection schemes come into play: The problems will be solved, if one succeeds to selectively count the ions that have interacted with the laser light. An obvious procedure would be stepwise laser ionization^{1,2} which is efficiently performed by use of pulsed high-power lasers. For continuous-beam conditions, this involves serious duty-cycle losses. On the other hand, cw lasers give low efficiency and optical pumping into levels outside the ionization ladder. Therefore, alternatives have been sought in exploiting the effects of optical pumping.

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(i) The RADOP technique - i.e. radioactive detection of optically induced nuclear polarization - has been adapted to the fast-beam conditions and applied to the lithium isotopes of which the exotic ^7Li deserves special interest³.

(ii) The state selectivity of charge-transfer neutralization has been used to detect very sensitively the laser excitation of Sr^+ and Ba^+ beams^{4,5}.

(iii) For neutral beams the collisional ionization can discriminate between atoms in different states⁶, provided that the cross-sections are sufficiently different.

We report the application of such a scheme to the very neutron-rich isotopes of radon and xenon, whereby we have gained several orders of magnitude in sensitivity compared to fluorescence detection.

ION-DETECTED SPECTROSCOPY ON RARE GASES

It is a common feature of the rare-gas spectra that the ionization energy is more than 10 eV for the ground state and only about 4 eV for the metastable first excited state in which one electron is promoted from the closed np^6 valence shell to the next higher $(\text{n}+1)\text{s}$ shell. This involves a considerable difference in the cross-sections for electron stripping. Fig.1 shows the relevant part of the energy level diagram for the example of radon. A fast beam of metastable atoms in the $J=2$ state of the $6\text{p}^57\text{s}$ configuration - designated $7\text{s}[3/2]_2$ - is prepared in the charge-transfer neutralization of the original ion beam with caesium vapour. Laser excitation to $7\text{p}[3/2]_2$ at 705.5 nm depopulates the metastable level and pumps the

Fig. 1. a) Partial energy-level diagram of radon. The caesium ground-state energy is close to the metastable $7\text{s}[3/2]_2$ level. b) Resonance signal of ^{226}Rn .

atom via $7s[3/2]_1$ into the low-lying 1S_0 ground state. In passing the beam through a gas target, one can ionize predominantly the metastable atoms and thus detect the optical pumping as a flop-out signal on the ion current.

Fig. 2. Schematic view of the experimental setup. The ion detection system is shown in detail.

Fig.2 gives a schematic view of the experimental setup used at ISOLDE. The front part including fluorescence detection is essentially identical with the standard apparatus⁷ that has been used extensively to study nuclear moments and radii. The differentially pumped stripping target has an effective thickness of 10 cm at a pressure of 10^{-3} to 10^{-2} mbar optimized for the individual gas. Ions created in this target are detected by a secondary-electron multiplier whose cathode is formed by a remote-controlled moveable metallic tape. This is indispensable for removing the considerable background from the radioactive decay of the nuclei collected on the detector during the experiment.

Another important source of background consists in the isobaric contaminations of the very weak beams of mass-separated radioactive atoms: Only to some extent the neutralization and ionization reactions are specific for the particular chemical element. For the "cold" plasma ion source of ISOLDE we have found that after careful outgassing the beams of heavy Rn and Xe isotopes are very clean, if all less volatile reaction products are trapped in a water-cooled transfer line between the ThC_2 or UC_2 target and the ion source. Before we discuss some results of the initial experiments on $^{223-226}\text{Rn}$ and $^{141-146}\text{Xe}$, we shall demonstrate the sensitivity for the example of the discovered new isotope, ^{146}Xe .

Fig. 3. Resonance signal of ^{146}Xe from a beam of originally 400 atoms/s.

The resonance of Fig.3 represents the transition $6s[3/2]_2 - 6p[3/2]_2$ for the $I=0$ isotope ^{146}Xe , measured relative to ^{136}Xe thus yielding the isotope shift. The total measuring time over a 2.5 times larger scanning range was 35 min with a beam of about 400 atoms/s from ISOLDE. The signal efficiency is given by a 50% total transmission of the beam line and our apparatus, 50% charge exchange and 10% ionization efficiencies and a 50% flop-out signal on the ion current. These numbers refer to the 2×10^{-3} mbar Cl_2 target used in most of our experiments. A constant background of about 10 counts/s is ascribed to the general radioactivity level of the experimental area and should be removed by more careful shielding. It is obvious that even without significant improvements mass-separated beam intensities well below 100 atoms/s are sufficient for a measurement within reasonable time. This should be compared to the fluorescence detection limit for rare gases which is about 10^6 atoms/s in the far-red transitions that are accessible from the metastable state.

RESULTS ON RADON AND XENON

For radon the present measurements on $^{223-226}\text{Rn}$ continue the earlier work on the abundantly produced isotopes $^{202-212, 218-222}\text{Rn}$ for which preliminary results have been reported^a. The particular interest in this

continuation arises from the theoretical and empirical evidence for strong octupole correlation effects in the Ra-Th region around neutron number $N=136$. The Rn nuclei are probably marking the boundary of this region. Indeed the spins, moments, and the odd-even staggering of the isotope shifts indicate unusual ground-state properties for ^{219}Rn and ^{221}Rn . We shall briefly discuss how this trend develops towards the heavier isotopes. Table 1 gives the spins, the hyperfine interaction parameters A and B in the $7s[3/2]_2$ state, and the deduced magnetic dipole and electric quadrupole moments for the odd-A isotopes between ^{219}Rn and ^{225}Rn . The magnetic moments are slightly recalibrated compared to ref.8, as a recent direct g-factor measurement⁹ on ^{209}Rn gives a reference value for the evaluation according to

$$A = \mu H_e(0)/IJ \quad (1)$$

where $H_e(0)$ is the magnetic hyperfine field.

Table 1: Spins and moments of the odd-A Rn isotopes. The magnetic moments include a 1% error for the hfs anomaly, and the quadrupole moments are not corrected for the Sternheimer effect.

	I	A (MHz)	B (MHz)	$\mu(\mu_N)$	$Q_s(b)$
219	5/2	-564.9(1.2)	-5316(10)	-0.442(5)	0.93
221	7/2	-18.4(6)	2184(5)	-0.020(1)	-0.38
223	7/2	-708.5(3)	-4564(3)	-0.776(8)	0.80
225	7/2	-635.0(8)	-4777(9)	-0.696(8)	0.84

There has been no rigorous theoretical interpretation of the ^{219}Rn and the ^{221}Rn moments, but it seems that the transitional nature of these nuclei, together with a weakened tendency of parity-mixing is responsible for changes in the neutron states from radium^{10,11} to radon. In contrast to ^{221}Ra , the isotone ^{219}Rn with the same spin $I=5/2$ has a magnetic moment close to the prediction of reflection symmetric particle-rotor calculations¹². However, the value of the $I=7/2$ for ^{221}Rn , different from $I=3/2$ for ^{223}Ra , is difficult to interpret in combination with the almost zero magnetic moment and the small negative quadrupole moment.

For the isotopes ^{223}Rn and ^{225}Rn it is surprising to find again the spin $I=7/2$ with moments that are completely different from ^{221}Rn . They can be accounted for by assuming a $7/2^-$ orbital based on the $j_{15/2}$ configuration. The isotones ^{225}Ra and ^{227}Ra , on the contrary, have $I=1/2$ and $I=3/2$ and are interpreted within the reflection-asymmetric scheme.

This qualitative difference is further manifested in the odd-even staggering of the mean square charge radii. An inverted staggering - i.e. larger $\langle r^2 \rangle$ of the odd-N isotopes compared to their even-N neighbours - is observed for $^{220-228}\text{Ra}$, and we have found arguments to interpret this behaviour as an effect of the octupole deformation: The reflection-asymmetric shape is stabilized by polarization due to the odd neutron. It is thus an effect in $\langle \beta_3^2 \rangle$ counteracting the normal staggering which is for instance found in the light Ra and Rn isotopes below $N=126$. In Fig.4 this is plotted in the form of differences

Fig. 4. Brix-Kopfermann plot of $\delta \langle r^2 \rangle$ for Rn (solid lines) and Ra (dashed lines). The upper curves correspond to the addition of neutron pairs and the lower ones for the addition of single neutrons (for symbols see text).

$\delta \langle r^2 \rangle_{A-1,A}$ between odd A and even $A-1$ (open symbols) and between even A and odd $A-1$ (full symbols). The striking similarity of both zig-zag curves disappears at $N=135$ (^{223}Rn and ^{225}Ra), where for radon also the nuclear moments indicate a change in the ground-state structure. As the normal staggering is restored for ^{223}Rn and ^{225}Rn , it appears natural to interpret them by reflection-symmetric configurations and to assume $\beta_3 \neq 0$ only for ^{221}Rn and ^{219}Rn .

The recent xenon measurements include a few neutron-deficient test candidates and the systematically studied isotopes $^{136-146}\text{Xe}$ ($^{136-140}\text{Xe}$ from standard fluorescence spectroscopy). These neutron-rich nuclei with $82 \leq N \leq 92$ belong to the classical region of transition from spherical to deformed nuclear shapes which is crossed by the stability line along the lighter rare-earth elements.

Xe ($Z=54$) is the element closest to the magic proton number $Z=50$ for which isotopes around $N=90$ can be reached, and it remains to be analyzed to what extent these nuclei still develop deformed shapes like the neighbouring Ba ($Z=56$)⁷.

SYSTEMATICS IN THE RARE-EARTH REGION: HOLMIUM

For the rare-earth elements between Nd and Dy - i.e. around $Z=64$ - the onset of deformation occurs in a sharp transition between $N=88$ and 90 . Here the systematics of moments and radii mainly covers the even- Z elements^{13,14} and europium¹⁵⁻¹⁷ of which the two stable isotopes happen to fall on these two neutron numbers (see also the discussion by G.D. Alkhazov¹⁸ about recent results of the Leningrad-Moscow collaboration). The measurements including neutron-deficient isotopes show that the shape transition for $Z=63$ is more pronounced than for all even- Z neighbours. This is likely to be connected with the nearby $Z=64$ subshell closure. From the results on even- Z elements one also concludes that holmium lies at the boundary between the sharp and the smooth onset of deformation. Therefore, measurements on holmium should help to clarify the influence of the odd proton which for most of the isotopes can be found in a high-spin ground state or a low-spin isomer.

We have studied $^{151-165}\text{Ho}$ for which an adequate technique is the conventional photon-detected collinear laser spectroscopy. The experiment was performed in the transition $4f^{14}6s^2 \ ^2F_{15/2} \rightarrow 4f^{14}6s6p(15/2,1)_{17/2}$ of neutral holmium at 410.4 nm. The yields from spallation in a thick Ta-foil target were between 10^6 and 10^{10} atoms/s.

Fig.5 gives the change in the mean square charge radii $\delta\langle r^2 \rangle$ for $^{151-165}\text{Ho}$ in comparison with the curve for $^{140-156}\text{Eu}$ (ref. 16,17), both evaluated from isotope shift measurements. Despite the striking similarity the two curves indicate a remarkable difference in the structure of the $N=89$ isotones: ^{152}Eu is strongly deformed in the $I=3$ ground state, whereas in ^{156}Ho both the $I=4$ ground state and the $I=1$ isomer are on the near-spherical branch. Beyond $N=90$ the initial deformation is smaller and continues to increase towards the heavier isotopes. A particularly interesting feature is the isomerism which gives rise to large shape differences between the $I=5$ ground states and the $I=2$ isomers in ^{158}Ho and ^{160}Ho . The isomer shifts below $N=90$ are small. While the onset of deformation for the holmium ground states occurs suddenly between $N=88$ and 90 , the deformation for the isomeric states increases gradually with the neutron number.

All measured spins are compiled in Table 2. They are readily deduced from the spectra, because the number of

Fig. 5. Plot of $\delta\langle r^2 \rangle_{82, N}$ for Eu and Ho. Lines of constant deformation are calculated from the droplet model.

Table 2: Spins of the ground states (I) and isomeric states (I^m) of Ho.

A	151	152	153	154	155	156	157
I	11/2	9	11/2	8	5/2	4	7/2
I^m	1/2	2	1/2	2	-	1	-

A	158	159	160	161	162	163	165
I	5	7/2	5	7/2	1	7/2	7/2
I^m	2	-	2	-	-	-	-

strong hyperfine structure components ($\Delta F=+1$), observed in the $J=15/2 \rightarrow J=17/2$ transition, is equal to $(2I+1)$ for $I < 8$. In a few cases the determined spin differs from earlier assignments. As an example we give the hyperfine structure of ^{156}Ho ($N=89$) in Fig.5 where the ground state spin is obviously $I=4$ and the three small components of the $I=1$ isomer are marked by arrows.

Fig. 6. Hyperfine spectrum for ^{156}Ho and ^{156m}Ho .

The interpretation of the proton configurations is straightforward for the odd-A isotopes whose moments are consistent with spherical shell model assignments of $\pi h_{11/2}$ and $\pi s_{1/2}$ for ^{151}Ho and ^{153}Ho , $\pi d_{5/2}$ for ^{155}Ho and the rather pure $\pi[532\ 7/2]$ Nilsson configuration¹⁹ for the deformed $^{157-165}\text{Ho}$.

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