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HEAVY IONS AS PROBES OF NUCLEI FAR FROM STABILITY

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I. Introduction

Nuclei located far from stability provide us with an opportunity for studying nuclear matter existing under unusual conditions. In these regions of instability, radioactive decay becomes the predominant technique by which one can obtain structure information. We have been involved in the investigation of nuclear properties of nuclei close to the proton drip line. In our explorations we have utilized heavy-ion fusion, followed by particle evaporation, to produce the extremely neutron-deficient nuclei of interest. Their properties were then studied by using on-line isotope separators at Oak Ridge (UNISOR) and Berkeley (OASIS), the Oak Ridge National Laboratory velocity filter, and a fast helium-gas-jet transport system at Lawrence Berkeley Laboratory 88-Inch Cyclotron.

In our studies, single-particle states near the 82-neutron shell, populated in the β decay of short-lived nuclides, have been examined and their excitation energies determined. Numerous new isotopes, isomers, and β -delayed-proton and α -particle emitters have been discovered. This contribution will discuss our particle-decay investigations. These decay modes provide us with a convenient means of discovering new isotopes whose identification opens the way for further, more extensive explorations. Also, particle-decay energies in many instances can be used to determine mass differences between parent and daughter ground states. Such measurements are therefore used to test mass formulae and to obtain estimates of masses for proton rich nuclei.

II. The α Decay of Lead Isotopes

Most isotopes with $A > 140$ are unstable toward α -particle emission. With the exception of naturally occurring ^{147}Sm , however, α decay was not observed for elements below bismuth until 40 years ago, because the rate for α decay is a very sensitive exponential function of the decay energy. (The energy available for decay

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increases rapidly with mass, so that in the region above lead α decay becomes a dominant decay mode.) Neutron deficient nuclides with $Z < 83$ do undergo α -particle emission if they are sufficiently proton rich. With the increasing availability of heavy-ion accelerators during the past two decades, the number of known α -active nuclides below bismuth has steadily increased.

Figure 1 shows the existing situation in the region from neodymium to lead. Isotopes and their α -decay energies are displayed graphically as a function of neutron and proton numbers. For clarity, even- Z nuclides are indicated by bars while odd- Z nuclei are represented by dots. It is seen that α -decay energies increase both with increasing Z (and A) and with decreasing N (as one gets further away from the valley of stability). One notes in the rare earth region that α decay has been detected for many of the isotopes with neutron numbers of 84 and slightly higher. This is due to the extra enhancement in decay energies that comes about as a result of the $N = 82$ shell. Because of the shell's stability α -decay energies reach a maximum for $N = 84$ nuclei; this enhancement is washed out for isotopes with $N > 87$. The stability of the 82-neutron configuration reduces the α -decay energies of $N = 83$ and $N = 82$ nuclides by ~ 1.5 and 3.0 MeV, respectively. While the energies begin to increase once again for nuclei with $N < 81$ the α -decay branching ratios are too small to compete with β decay and no α emission has been observed in rare earth isotopes with $N < 84$.

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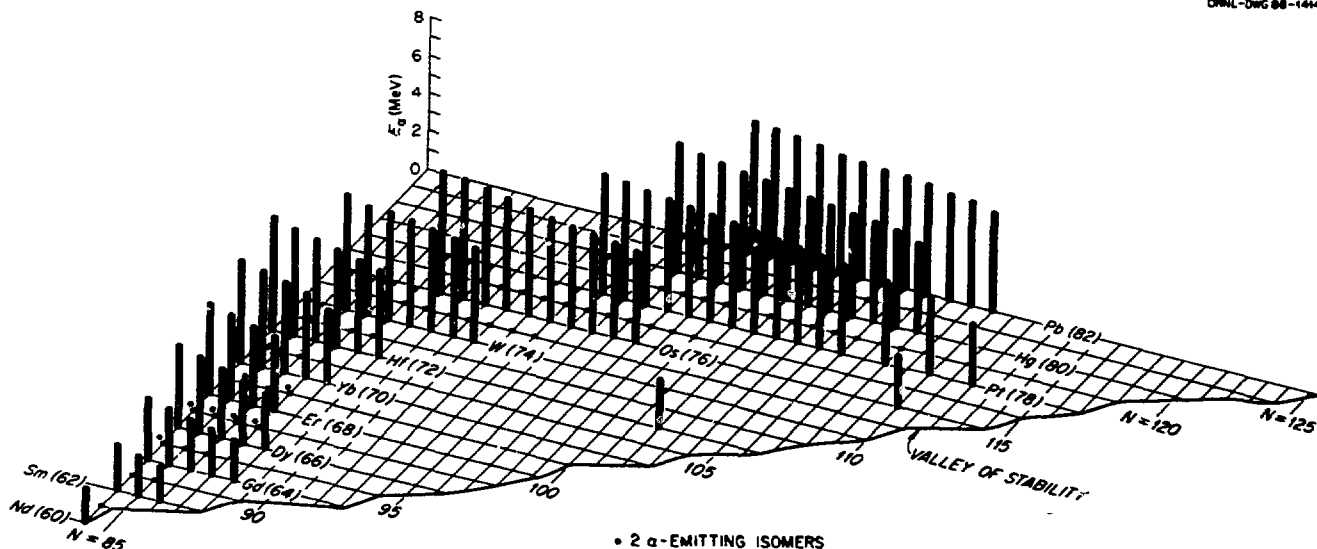


Fig. 1. Known α -emitters in the mass region from neodymium to lead.

During the past ten years we have investigated the α -decay properties of some of the Hg, Tl, and Pb nuclides displayed in Fig. 1. Recently, with the use of a rapid gas-jet-transport system at the LBL 88-Inch Cyclotron we identified the new isotopes $^{182}\text{Pb}/1/$ and $^{181}\text{Pb}/2/$ in ^{40}Ca bombardments of ^{147}Sm and ^{144}Sm , respectively. The α -particle spectrum accumulated in 222-MeV ^{40}Ca irradiations of ^{147}Sm is shown in Fig. 2. The (6.919 ± 0.015) -MeV peak is assigned to ^{182}Pb on the basis of excitation-function and cross-bombardment data and decay-energy systematics. This 55-ms radio-nuclide is 25 mass units away from the line of stability. Yet, because the ^{182}Pb α -decay chain terminates at ^{146}Gd , our decay-energy measurement has resulted in a rather precise mass excess for ^{182}Pb , i.e., -6823 ± 25 keV. The adopted^{/3/} mass excess for ^{182}Pb is -6874 ± 28 keV.

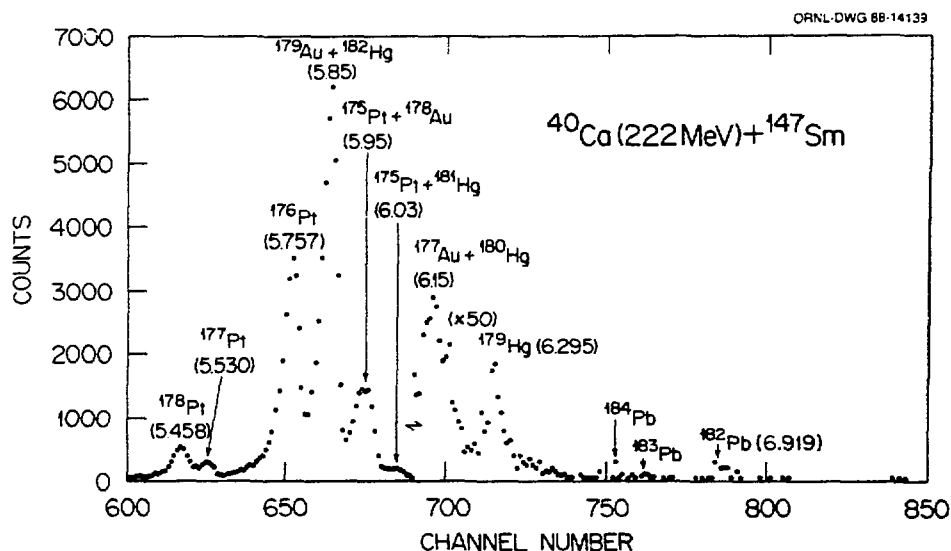


Fig. 2. Alpha-particle spectrum measured during a bombardment of ^{147}Sm with 222-MeV ^{40}Ca ions. Energies shown are in MeV.

In our investigation^{/1/} two incident ^{40}Ca energies were used, i.e., 194 and 222 MeV, to enhance the production of ^{184}Pb [$^{147}\text{Sm}(^{40}\text{Ca},3n)$ product] and ^{182}Pb [$^{147}\text{Sm}(^{40}\text{Ca},5n)$ product], respectively. To obtain more definite data on ^{183}Pb and to provide additional cross-bombardment information we irradiated ^{147}Sm with 212-MeV ^{40}Ca ions. Figure 3 shows the accumulated α -particle spectrum. The 194-MeV spectrum was dominated by ^{184}Pb while the 222-MeV spectrum had only possible traces of ^{184}Pb and ^{183}Pb α activity (see Fig. 2). Figure 3 clearly represents an intermediate situation, namely, the intensity of ^{183}Pb is now somewhat greater than that of ^{184}Pb , and ^{182}Pb is seen with about the same intensity as had been observed at 222 MeV. We assign a total of four α groups to ^{183}Pb . Previously, only two groups have been observed in ^{183}Pb α decay. A mass excess of -7720 ± 310 keV has been adopted^{/3/} for ^{183}Pb ; it is based on a Q_α value deduced

from the highest-energy previously observed $^{4/4}$ α group of 6798 keV. Since we now have seen an α peak of 6874 keV both the adopted Q_α value and mass excess of ^{183}Pb will have to be revised.

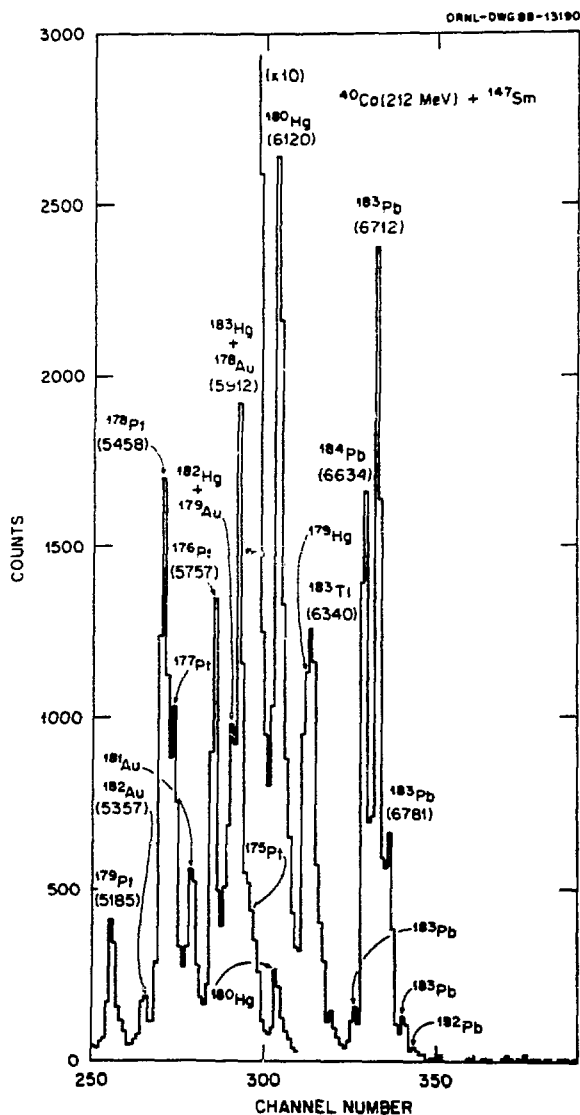


Fig. 3. Alpha-particle spectrum measured in bombardments of ^{147}Sm with 212-MeV ^{40}Ca . Energies shown are in keV.

Because the production of $A < 182$ lead nuclei with ^{147}Sm necessitates unfavorable reactions wherein six or more neutrons have to be evaporated (there is fission competition at each evaporation step) we used ^{144}Sm as the target in our search for ^{181}Pb and ^{180}Pb . Figure 4 shows the spectrum measured at 201 MeV with the ^{144}Sm target. In addition to nuclides with $Z < 80$, one observes ^{184}Pb and ^{183}Pb produced from heavier samarium isotopes present in the target material [^{147}Sm (3.9%), ^{148}Sm (2.2%), ^{149}Sm (2.3%), etc.]. It is possible that ^{183}Pb is also produced following the evaporation of a single neutron from the ^{184}Pb compound system.

A new peak is seen at 7044 ± 15 keV. We assign it to the α decay of ^{181}Pb because its yield as a function of energy and target behaves as one would expect for the $^{144}\text{Sm}(^{40}\text{Ca},3n)$ product. It is observed neither in Figs. 2 and 3 nor in Fig. 5 which shows the spectrum measured at 212 MeV, an energy where the yield of the $^{144}\text{Sm}(^{40}\text{Ca},4n)$ product, ^{180}Pb ; should be maximum.

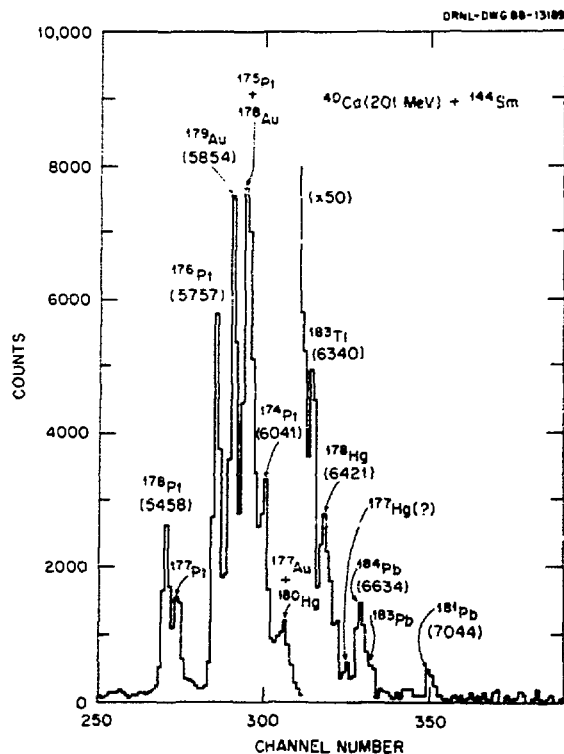


Fig. 4. Alpha-particle spectrum measured in bombardments of ^{144}Sm with 201-MeV ^{40}Ca ions. Energies shown are in keV.

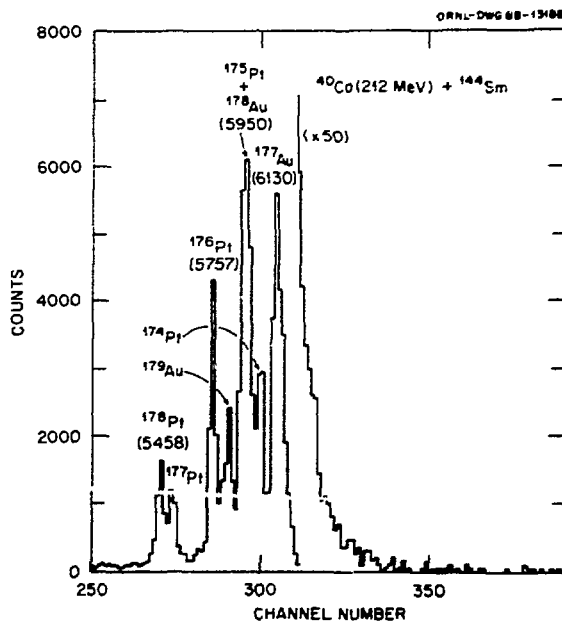


Fig. 5. Alpha-particle spectrum measured in bombardments of ^{144}Sm with 212-MeV ^{40}Ca ions. Energies shown are in keV.

Based on what is known about the α decays of ^{187}Pb , ^{185}Pb , and ^{183}Pb we conclude that the 7044-keV peak is probably only the most intense of several ^{181}Pb α transitions. If one assumes that it proceeds between the ground states of ^{181}Pb and ^{177}Hg then the mass excess of ^{181}Pb can be estimated to be about -3.31 MeV based on the value of (-12950 ± 230) keV adopted^{3/} for ^{177}Hg . Our decay-energy measurement thus leads to the first determination of the mass for ^{181}Pb , a nucleus which lies more than 26 mass units away from the valley of β stability.

There is no indication of ^{180}Pb in Fig. 5; its α -decay energy is expected to be ~ 7.2 MeV. Either its production cross section is below our detection limit or its half-life is too short for an appreciable amount of its activity to survive the minimum transport time of 25 ms through the gas-jet capillary. This transport time

makes it very difficult to observe low-yield radioactivities whose half-lives are much less than 10 ms. Table I summarizes our data on the α decays of the very neutron-deficient lead isotopes and compares them with the results of Schrewe *et al.*^{/4/}

Table I. Half-lives and α -decay energies of lead isotopes.

Isotope	This work			Schrewe <i>et al.</i> ^{/4/}		
	$T_{1/2}$ (ms)	E_{α} (keV)	I_{α} (%)	$T_{1/2}$ (ms)	E_{α} (keV)	I_{α} (%)
¹⁸¹ Pb	50 ⁺⁴⁰ ₋₃₀	7044(15)	100			
¹⁸² Pb	55 ⁺⁴⁰ ₋₃₅	6919(15)	100			
¹⁸³ Pb	300(80)	6579(15)	5.5(20)			
		6712(10)	72(4)		6715(20)	92(4)
		6781(15)	20(4)		6798(25)	8(4)
		6874(15)	2.5(10)			
¹⁸⁴ Pb		6634(10)	100	550(60)	6632(10)	100

III. Reduced widths for the α decay of even-even nuclei

Decay rates for α transitions between ground states of doubly-even nuclei are taken to represent unhindered decays. Their reduced widths are regarded as standards against which other types of α transitions are to be compared. Figure 6 shows reduced widths for s-wave transitions of nuclei with Z from 78 to 100 plotted as a function of neutron number. We utilized Rasmussen's formalism^{/5/} to calculate the width, δ^2 , which is defined as: $\delta^2 = \lambda h/P$, where λ is the decay constant, h is Planck's constant, and P is the penetrability for the α particle to tunnel through a barrier. A rather regular behavior as a function of both neutron and atomic number is observed for these reduced widths. They are largest for nuclei two or four particles beyond a closed shell (with sharp minima occurring at the closed shell), followed by a decrease as one approaches the next closure. These trends can be understood in terms of single-particle models which have shown^{/6/} that the extremely sharp break at $N = 126$ is essentially a shell structure effect. There is also a dip in values at $N = 152$ due to the subshell closure at that neutron number.

As stated above, the reduced widths around $N = 130$ are large; in particular, the available ²¹⁸Ra value (labeled as "Previous Measurement" in Fig. 6) exhausted 75% of the Wigner-sum-rule limit. Suggestions were made that these large widths indicate α clustering on the nuclear surface; the distorting effect of these

clusters could account for reflection asymmetry observed in radium and thorium nuclei near $N = 130$. We remeasured^{/7/} the ^{218}Ra half-life by using the ORNL velocity filter and a novel technique in which reaction products, after being separated from the incident beam, are implanted in a Si(Au) detector and their subsequent α decays are observed in the same detector. We found the half-life of ^{218}Ra to be $25.6 \pm 1.1 \mu\text{s}$ instead of the adopted value of $14 \mu\text{s}$. It yields an α width which in Fig. 6 is indistinguishable from those of ^{216}Rn and ^{220}Th . This smooth trend of α widths from the $N = 130$ region to the well-deformed, prolate, Cm, Cf, and Fm nuclei weakens one argument quoted for the existence of α clusters in the heavy elements. It is the stabilizing effect of the shell closure that produces the sharp decrease in width values from $N = 130$ to $N = 126$.

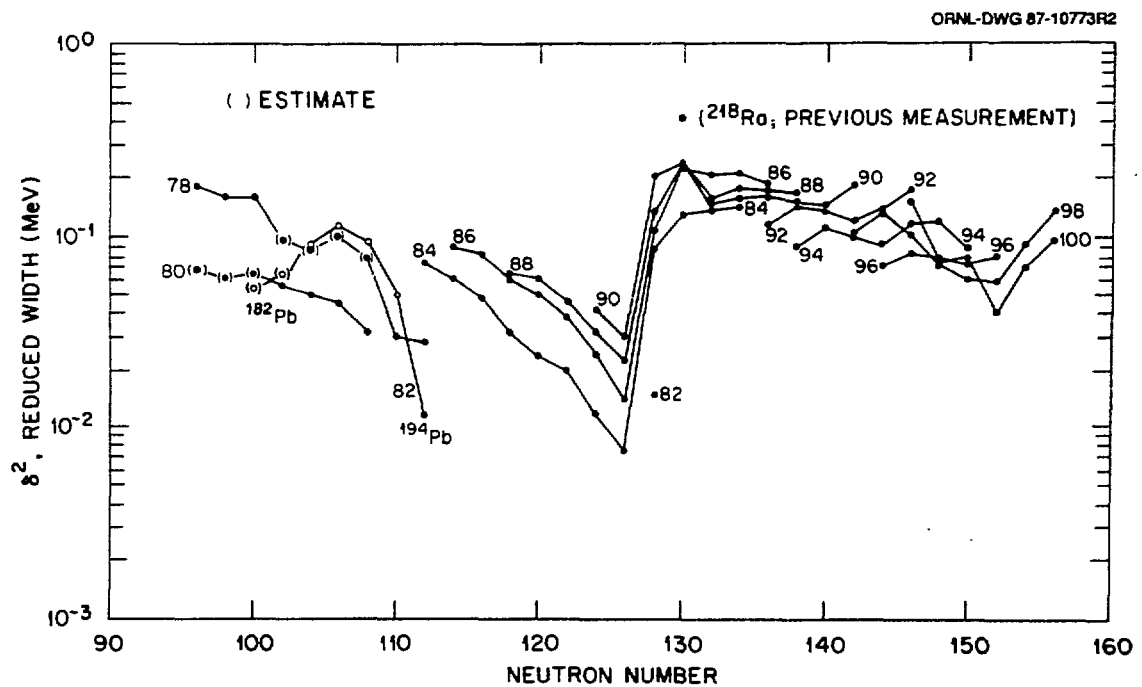


Fig. 6. Reduced widths for s-wave α transitions plotted as a function of N for isotopes with Z from 78 to 100.

However, contrary to an expected shell effect at $Z = 82$, the α -decay rates of $^{186}, ^{188}, ^{190}, ^{192}\text{Pb}$ (open points in Fig. 6 with N from 104 to 110) are less hindered than those of neighboring Hg isotopes. This implies that, midway between $N = 82$ and $N = 126$ the 82-proton shell is not magic.^{/8/} We recently used the UNISOR facility to identify the α decay of ^{194}Pb for the first time and to determine the isotope's α branch.^{/9/} The resultant width for ^{194}Pb (see Fig. 6) is much less than those of nuclei with $N < 110$ and less than the δ^2 for ^{190}Pt ($N = 112$), indicating

that perhaps the $Z = 82$ gap is being restored for $N > 112$. The reduced α widths for ^{190}Hg and ^{192}Hg should be determined to be certain about this proposal; if our suggestion about the $Z = 82$ gap being restored for $N > 112$ is correct, the ^{192}Hg width should be larger than that of ^{194}Pb .

We also show in Fig. 6 widths for ^{182}Pb and ^{184}Pb based on our data and those of Schrewe *et al.*,^{/4/} respectively, assuming assuming α branches of 100%. [From gross β -decay theory the calculated partial half-lives for ^{182}Pb and ^{184}Pb (β^+ + EC) decay yield α branching ratios $> 90\%$.] These δ^2 values are appreciably smaller than the $^{186},^{188},^{190}\text{Pb}$ widths and may indicate that the influence of the $Z = 82$ shell reappears and once again retards the α decay of extremely neutron-deficient Pb isotopes.

IV. Investigation of Rare Earth Nuclei Near the Proton Drip Line

With the use of the OASIS separator facility,^{/10/} we have investigated the decay properties of numerous short-lived neutron-deficient rare earth nuclei with $65 < Z < 71$. Figure 7 shows a portion of the nuclidic chart which encompasses the mass region where these radioactivities are located. Some of the nuclei, on the lighter side of the 82-neutron shell, are at or close to the proton drip line. For many of them β -delayed proton (and in a few instances direct proton) emission becomes a probable mode of decay. Also note, as mentioned earlier, most of the nuclei in Fig. 7 with $N > 84$ are α -particle emitters. We should add that, in a separate experimental program,^{/11,12/} the OASIS separator has been used to study other proton-rich rare earth nuclei, primarily those with $Z < 66$.

A. Delayed Proton Emission For $N = 81$ Precursors

As a result of high level densities, β -delayed-proton decay originating from nuclei with $A > 80$ yields proton spectra that are basically featureless. Near major closed shells, however, pronounced peaks have been observed in some delayed-proton spectra. Are these peaks due to nuclear structure effects or to a fluctuation phenomenon which can be explained by a statistical model approach?

To shed light on this question we investigated spectra arising from an isotonic sequence of six β -delayed-proton precursors with $N = 81$ and $66 < Z < 71$. They are shown in Fig. 8 where one notes that the mean energy values of the spectra from the odd-odd isotopes ^{148}Ho , ^{150}Tm , and ^{152}Lu are higher than those of the spectra from the even- Z nuclei, ^{147}Dy , ^{149}Er , and ^{151}Yb . This effect is explained by the fact that in odd-odd precursors Gamow-Teller β decay occurs without breaking a proton pair so that the proton-emitting nucleus is left in a state of high excitation energy (and high level density). Both sets of $N = 81$ precursors have

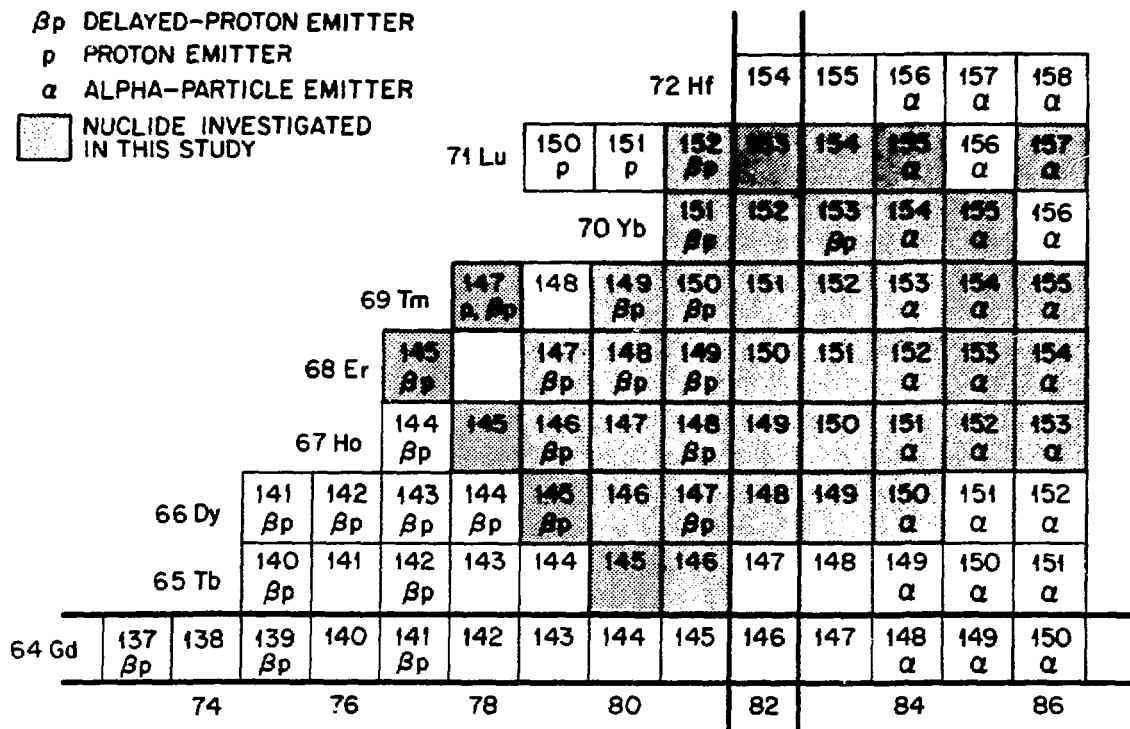


Fig. 7. Portion of nuclidic chart where isotopes investigated in this experimental program are indicated by shaded squares.

β -decaying low- and high-spin isomers, but the final nucleus is very different in the two cases. In the odd-odd case the final nucleus has two nearly degenerate ground states creating parallel paths for the high- and low-spin proton decays [Fig. 9(a)] with energetics that are the same within a few hundred keV. For both paths proton emission occurs from regions of high level density and produces spectra that are statistical in appearance. In contrast, the $N = 82$ final nucleus for an even-odd $N = 81$ precursor [Fig. 9(b)] has a 0^+ ground state, while higher spin states are found only above ~ 1.5 MeV. This leads to different energetics for the low- and high-spin decay branches. The low-spin ($1/2^+$) ground state, associated with proton decay from a region of low level density in the $N = 82$ β -decay daughter, gives rise to a structured spectrum. On the other hand, the high-spin ($11/2^-$) isomer produces a structureless spectrum since angular-momentum and decay-energy considerations make its protons originate from a region of high level density.

The spectra of ^{147}Dy , ^{149}Er , and ^{151}Yb , therefore, feature pronounced peaks superposed on a structureless component which increases in intensity as the β -decay Q window enlarges, i.e., as one proceeds from ^{147}Dy to ^{151}Yb . (The experimental decomposition of the ^{151}Yb spectrum has been discussed^{13/} in detail.) The intense proton peaks from these three $N = 81$ precursors can be seen even more clearly in

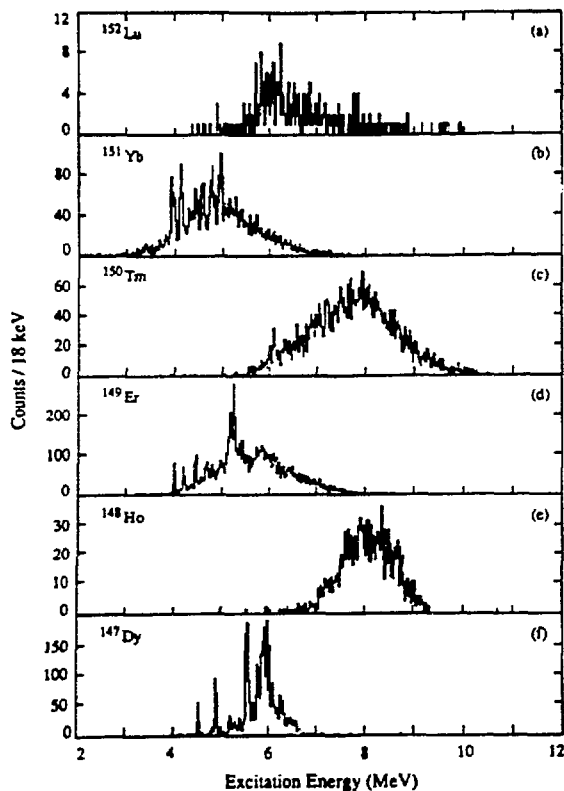


Fig. 8. Beta-delayed proton spectra of $N = 81$ precursors.

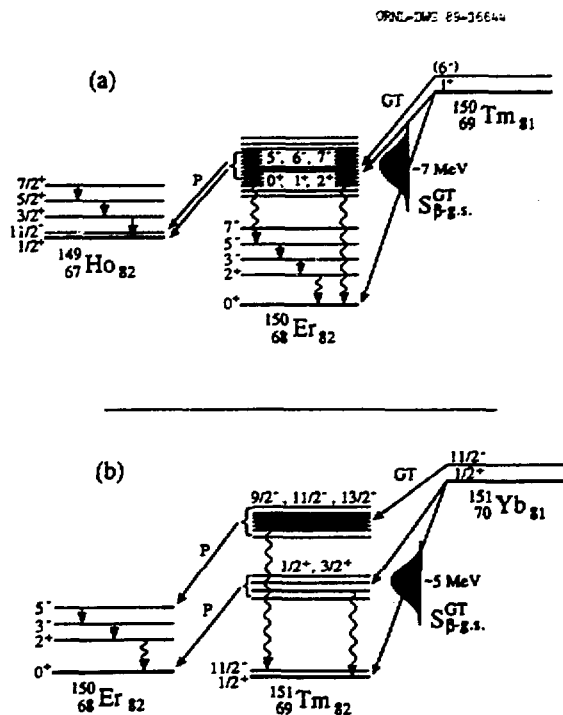


Fig. 9. Schematic representation of β -delayed-proton decay in (a) odd- Z and (b) even- Z $N = 81$ precursors.

Fig. 10. In parts (a) and (b) protons were recorded in coincidence with positrons following the decays of ^{151}Yb and ^{149}Er , respectively. The positron coincidences select preferentially decays from the $s_{1/2}$ ground states of the precursors and suppress decays from the $h_{11/2}$ isomers which give rise to smooth statistical spectra. Because of the lower Q_{EC} available for ^{147}Dy β decay there are very few (β^+ -proton) coincidence events so that in part (c) we show the singles spectrum where the statistical component is very weak. Gamma-ray decay studies, β -strength function measurements, and calculations of state densities and Gamow-Teller strength distributions, have led us to suggest^{/14/} that some of the structure in delayed-proton spectra near $N = 82$ arises from the preequilibrium decay of doorway states populated in β decay.

These peaks disappear from observed spectra almost as soon as the β -decay daughter no longer has a major closed-shell configuration, though there may be weak structures appearing in the spectra of the $N = 79$ precursors ^{145}Dy and ^{147}Er . This and other points are addressed in a recent thesis^{/12/} where the results obtained at the OASIS facility for delayed-proton precursors, ranging from ^{119}Ba to ^{154}Lu , are described and discussed.

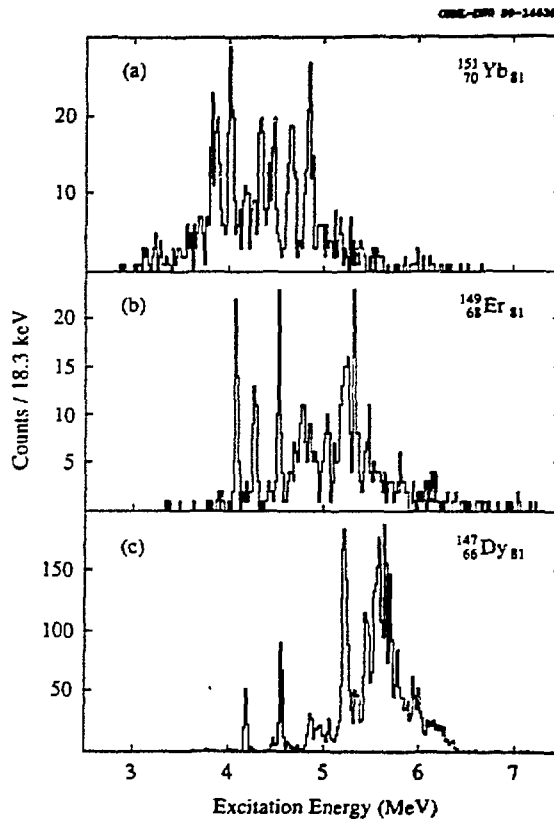


Fig. 10. Beta-delayed-proton spectra of: (a) ^{151}Yb , (b) ^{149}Er , and (c) ^{147}Dy . Parts (a) and (b) are protons in coincidence with positrons; part (c) is a singles spectrum.

B. Alpha-particle Decay of Rare Earth Nuclides

The investigation of α decay in the lanthanides has led to the discovery of many isotopes, the determination of nuclear masses and isomeric excitation energies, and the first observation of the subshell closure at $Z = 64$. We have reinvestigated some of these α emitters and have determined more precisely a number of α branching ratios. Also, we have recently observed^{/15/} fine structure in the α -decay spectrum of ^{153}Tm . These new transitions populate the $d_{3/2}$ (220.4 keV) and $d_{5/2}$ (564.4 keV) single-proton levels in ^{149}Ho . Herein we review briefly our study of the ^{155}Lu and ^{157}Lu α decays.

Figure 11 shows the α spectrum recorded at $A = 155$. Above the intense 5.194-MeV α peak which belongs to ^{155}Yb decay, there are two weak α groups with energies of 5.579 ± 0.005 and 5.648 ± 0.005 MeV. The higher energy group, presumed to follow the α decay of the $h_{11/2}$ ground state ($T_{1/2} = 66$ ms) of ^{155}Lu , has been known^{/16/} since 1965. Recently, Hofmann *et al.*^{/17/} observed α decay ($E_{\alpha} = 5.575 \pm 0.010$ MeV) from a second low-lying level in ^{155}Lu . The data shown in Fig. 11 therefore confirm the existence of this new α -emitting level which we believe is

either the $s_{1/2}$ or $d_{3/2}$ single-proton state. No half-life for it was reported by Hofmann *et al.*;^{/17/} we measure a value of 140 ± 20 ms. Because of the low cross section for the $^{94}\text{Mo}(^{64}\text{Zn},p2n)$ reaction and the fact that ^{155}Lu decays mainly by α -particle emission, we saw no evidence for the isotope's β -decay branch.

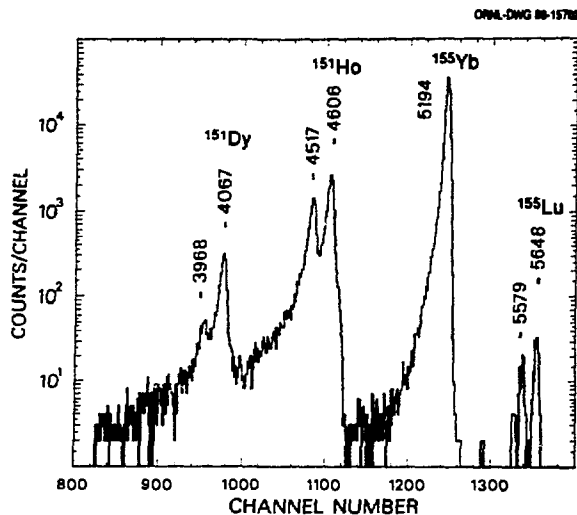


Fig. 11. Alpha-particle spectrum measured at $A = 155$. Energies shown are in keV.

Figure 12 shows the α spectrum that we measured at $A = 157$. In addition to α particles emitted by ^{149}Tb , ^{153}Ho , ^{157}Yb , ^{153}Er , ^{153}Tm , and the known^{/18/} ^{157}Lu ground state (presumably $h_{11/2}$) we observe a new α group, 4.924 ± 0.020 MeV, which we assign to the decay of the previously unreported low-spin ($1/2^+$, $3/2^+$) isomer in ^{157}Lu . Preliminary analysis of our data indicates that the half-life of this isomer is ~ 6 sec rather than the 5.4 ± 0.2 s value adopted^{/18/} for the ground state. Information on the β decay properties of the two ^{157}Lu levels has hitherto not been available.^{/18/} We observe numerous γ rays following ^{157}Lu β decay and are in the process of constructing a scheme for the ^{157}Yb levels that are populated. Based on this decay scheme the α branches and α -decay rates for these two α emitters will be determined. It may then be possible to say if the ^{157}Lu isomer is a $d_{3/2}$ or $s_{1/2}$ proton level.

V. Conclusion

We conclude this paper by illustrating how the investigation of nuclei far from stability yields information to test the predictive capabilities of theoretical models. In the overall program at the OASIS facility forty-two β -delayed-proton precursors have been studied and their half-lives compared^{/12/} with predictions from the gross β -decay theory and Nilsson/RPA β -strength function calculations. Figure 13 shows this comparison between experiment and calculations.

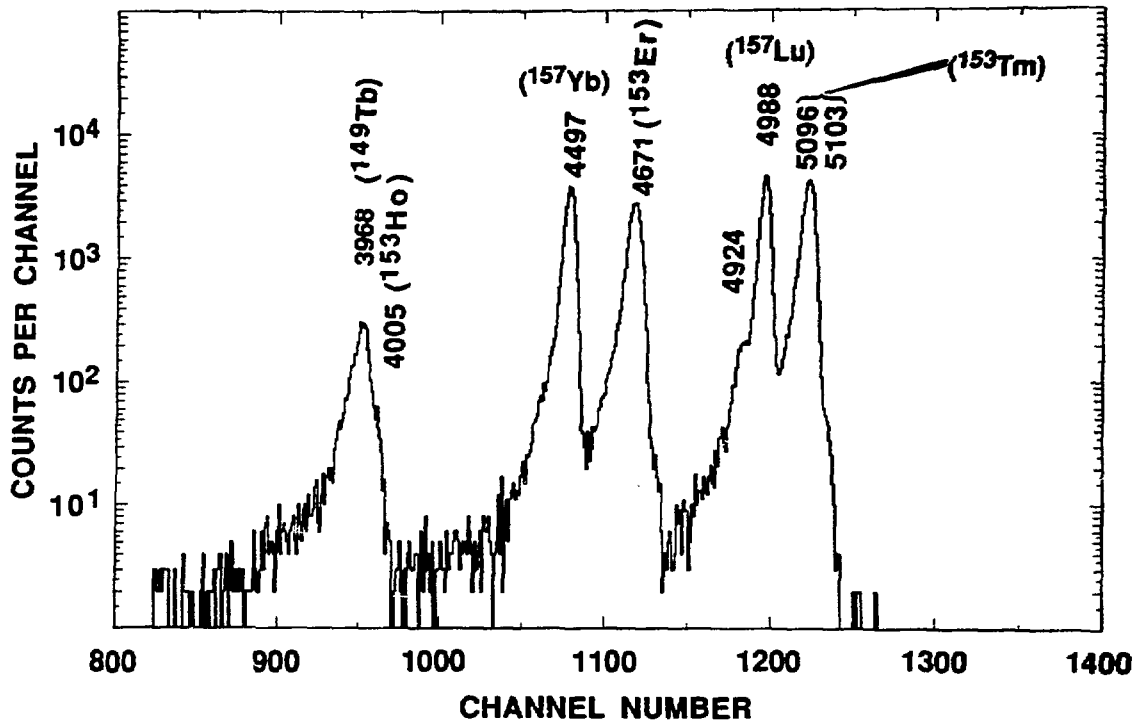


Fig. 12. Alpha-particle spectrum measured at $A \approx 157$. Energies shown are in keV. The 4924-keV α group is assigned to the decay of the previously unobserved low-spin ($1/2^+$, $3/2^+$) isomer in ¹⁵⁷Lu.

Both models predict similar half-lives and show reasonable agreement with measured values. For both calculations Q_{EC} values are taken from Liran and ⁷aldes/¹⁹/ and since the deviations from experiment are similar it is suspected/¹²/ that errors in the predicted Q -values may be the source of these deviations. For example, in the case of the gross β -delayed theory, lowering the Q -value by 5% shortens half-lives by ~ 0.7 , while raising the Q -value by 5% increases half-lives by ~ 1.5 .

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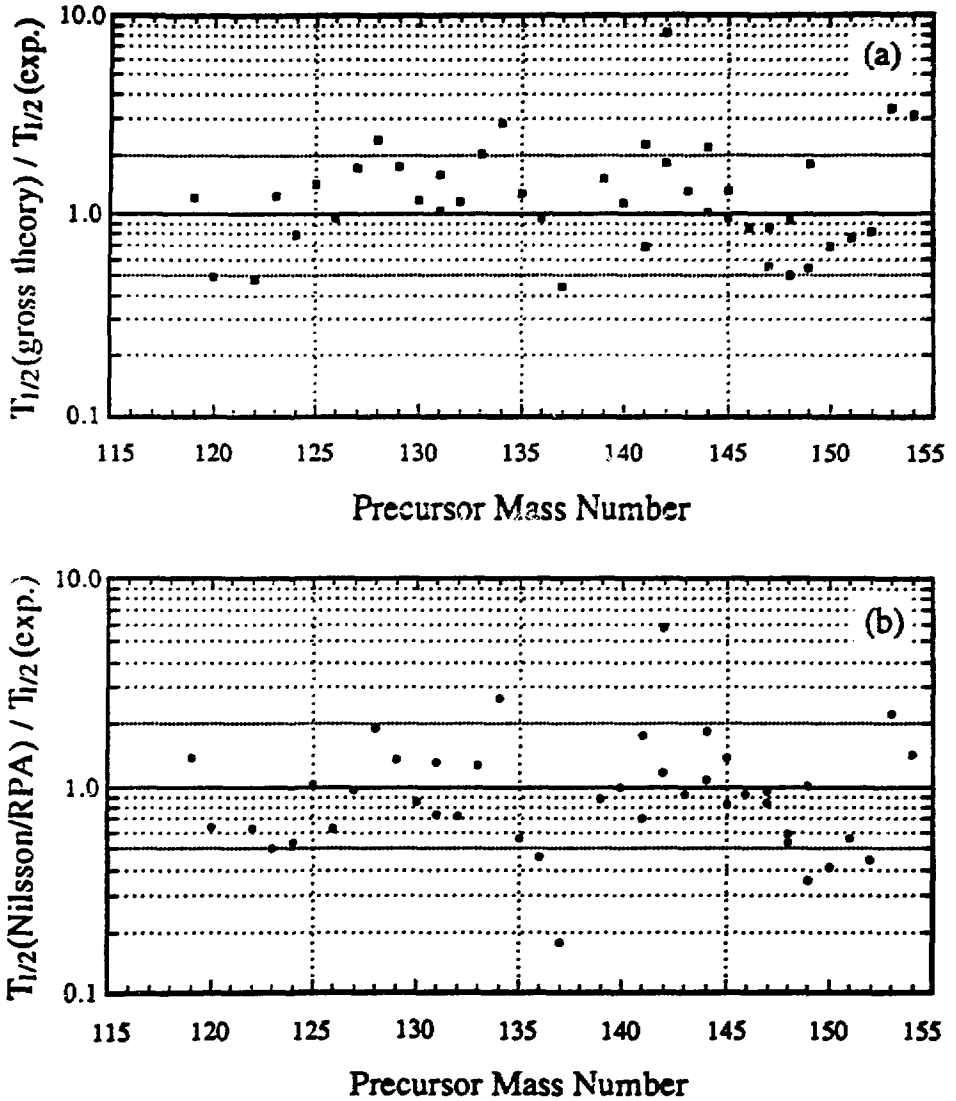


Fig. 13. Comparison of experimental half-lives with values predicted from (a) the gross theory of β decay, and, (b) Nilsson/RPA β -strength function calculations.

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