Exotic $N=82$ Nuclei ^{153}Lu and ^{154}Hf and Filling of the $\pi h_{11/2}$ Subshell

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Microsecond isomers in the very proton-rich $N=82$ nuclei ^{153}Lu and ^{154}Hf have been identified by γ-ray spectroscopy following mass analysis of ^{102}Pd+245 MeV ^{54}Fe reaction products using the Daresbury Recoil Separator. The decays of the isomers, interpreted as $(\pi h_{11/2})^n$ states, are characterized. Examination of reduced $E2$ transition rates between $(\pi h_{11/2})^n$ states in the $N=82$ isotones indicates that the half-filling of the $\pi h_{11/2}$ subshell occurs just below $Z=71$.

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Some ten years ago it was found[1] that the $Z=64$, $N=82$ species ^{146}Gd displays properties resembling those of a doubly magic nucleus. This discovery stimulated spectroscopic studies of many neighboring nuclei which have a few valence nucleons outside the ^{146}Gd core and may be well described in terms of shell-model configurations. Particularly interesting are the proton-rich $N=82$ isotones above ^{146}Gd, where the three nearly degenerate proton subshells $h_{11/2}$, $s_{1/2}$, and $d_{3/2}$ are being filled. The high-j unique-parity $\pi h_{11/2}$ orbitals should be especially important in the formation of high-spin states, and one could expect yrast excitations of the type $(\pi h_{11/2})^n$ to figure prominently in the spectra of $N=82$ isotones with $Z-64$ valence protons. Experiments[2-4] have identified and characterized the decays of related $(\pi h_{11/2})^n$ $E2$ isomers below 3 MeV in the 2–6-valence-proton $N=82$ nuclei ^{148}Dy, ^{149}Ho, ^{150}Er, ^{151}Tm, and ^{152}Yb. In the even-A nuclei the isomeric transitions are between 10^+ and 8^+ $(\pi h_{11/2})^n$ states of seniority $v=2$; in the odd-A nuclei they are $\frac{27}{2}^- \to \frac{23}{2}^-$ transitions between $(\pi h_{11/2})^n$ $v=3$ states. These isomeric decays also populate 3^- octupole, $[\pi(h_{11/2})^{n-1}s_{1/2}]5^-$ and $[\pi(h_{11/2})^{n-1}d_{3/2}]7^-$ $v=2$ excitations in the even-A nuclei, and corresponding $\frac{15}{2}^+$, $\frac{19}{2}^+$, and $\frac{23}{2}^+$ $v=3$ states, formed by coupling with an additional $h_{11/2}$ proton, in the odd-A nuclei.

The results for ^{148}Dy include a complete spectrum[2] of $(\pi h_{11/2})^2$ levels (0^+, 2^+, 4^+, 6^+, 8^+, 10^+), whose energies provide empirical two-body matrix elements for calculating the $(\pi h_{11/2})^n$ spectra in the heavier isotones. Moreover, Lawson[5] has used the ^{148}Dy energies to show that the condition for no seniority mixing is very nearly fulfilled. When seniority is a good quantum number, the reduced $E2$ transition rates between corresponding $(\pi h_{11/2})^n$ states of the same seniority should be proportional to $(6-n)^2$, where n is the $\pi h_{11/2}$ occupation number, and should become zero when $n=6$ at the half-filled subshell.[6,7] The relevant expressions for the $v=2$ and $v=3$ transitions are[8]

$$B(E2;10^+ \to 8^+) = \left(\frac{6-n}{4}\right)^2 \frac{2025}{35\,321\pi}(e_{\text{eff}}\langle r^2\rangle)^2$$

and

$$B(E2;\tfrac{27}{2}^- \to \tfrac{23}{2}^-) = \left(\frac{6-n}{4}\right)^2 \frac{32\,400}{265\,837\pi}(e_{\text{eff}}\langle r^2\rangle)^2 .$$

Using a fixed value for the effective charge $e_{\text{eff}}=1.52e$, and $\langle r^2\rangle=32$ fm^2 for the $\pi h_{11/2}$ orbital, the measured $B(E2)$ values for the five $N=82$ nuclei are very well reproduced[8,9] *by assuming that in every case n equals Z−64, the number of valence protons.* For ^{152}Yb with six valence protons, the experimental $B(E2)$ is not exactly zero as in the ideal picture, but its very small value (0.02 Weisskopf units) demonstrates that the $\pi h_{11/2}$ subshell is close to being half-filled in the 8^+ and 10^+ states.

It is difficult to see why this simple model is so successful, since one would expect that in the relevant states of the $Z>67$ nuclei, the $s_{1/2}$ and $d_{3/2}$ subshells should also be partly occupied by proton pairs, reducing the occupation of $\pi h_{11/2}$. On the other hand, the single-particle energy gap at $Z=64$ is significantly smaller than the gaps at traditional magic numbers, and scattering of proton pairs from $g_{7/2}$ or $d_{5/2}$ across the gap into $h_{11/2}$ may compensate in some measure for the depletion into $s_{1/2}$ and $d_{3/2}$. Calculations[8,10] which take account of these two effects suggest that half-filling of the $\pi h_{11/2}$ subshell might be postponed to $^{153}_{71}$Lu$_{82}$ or $^{154}_{72}$Hf$_{82}$, but there have been no data for these nuclei up to now.

The $N=82$ isotones ^{153}Lu and ^{154}Hf lie very close to the proton drip line. Previous attempts to study these

TABLE I. Production cross sections for the reaction $^{102}Pd+^{54}Fe \rightarrow ^{156}Hf^*$ predicted by the code CASCADE at ^{54}Fe beam energies of 235 and 245 MeV. The right-hand column gives relative experimental yields (in arbitrary units) for the isomeric species identified.

Exit channel	Product	σ(235 MeV) (mb)	σ(245 MeV) (mb)	Isomeric yields[a]
$2pn$	^{153}Yb	73	60	70
$p2n$	^{153}Lu	2	3.5	1.0
pn	^{154}Lu	3	1.2	2.5
$2n$	^{154}Hf	0.2	0.1	0.06

[a]Uncorrected for differences in the separator transmission for different products.

nuclei have been unfruitful mainly because they can be produced only in reactions with very low cross sections. In the present experiments, the sensitivity for detection of weak isomeric decays was considerably enhanced by use of the Daresbury Recoil Mass Separator.[11] The ionization chamber normally located behind the separator focal plane was replaced by an aluminium catcher foil, which was surrounded by four large Ge detectors and a low-energy photon spectrometer. Fusion-evaporation recoils from the reaction $^{102}Pd+^{54}Fe \rightarrow ^{156}Hf^*$ were mass analyzed, and, after passing through the position-sensitive focal-plane detector,[11] were deposited on the catcher foil for γ-ray measurements. Time relationships between signals from the position-sensitive detector and from the γ-ray detectors were used for mass, half-life, and $\gamma\gamma$ coincidence determinations. Since the separator transit time is ~ 1 μs, only long-lived isometric species were observed, but they could be studied under low-background conditions. The measurements were performed over a three-day period using 10–15-particle-nA beams of 240–245-MeV ^{54}Fe ions on a 1-mg/cm^2 ^{102}Pd target.

Production cross sections predicted by the code CASCADE[12] for the reaction products ^{153}Yb, ^{153}Lu, ^{154}Lu, and ^{154}Hf, all of which were expected to have long-lived isomers, are summarized in Table I, which also lists relative yields at the catcher foil for the isomeric species identified in these experiments. In line with the predictions, by far the strongest family of γ rays was observed in the $A=153$ mass window and to be coincident with Yb x rays. The 23 γ rays thus assigned to ^{153}Yb have been placed[13] in the decay scheme of a new 15-μs isomer in that nucleus. A second, much weaker ($<2\%$) γ-ray family, including 130-, 174-, and 217-keV lines, was also observed in the $A=153$ mass window. These γ rays are assigned to the $p2n$ evaporation product ^{153}Lu, because they appeared in coincidence with Lu K x rays [Fig. 1(a)], whereas ^{153}Yb lines were suppressed by more than a factor of 50 in the same spectrum. Other transitions in ^{153}Lu were identified by $\gamma\gamma$ coincidences [e.g., Fig. 1(b)]. The ^{153}Lu γ rays decayed with the half-life $t_{1/2}=15\pm3$ μs, and they are all placed in the isomeric decay scheme

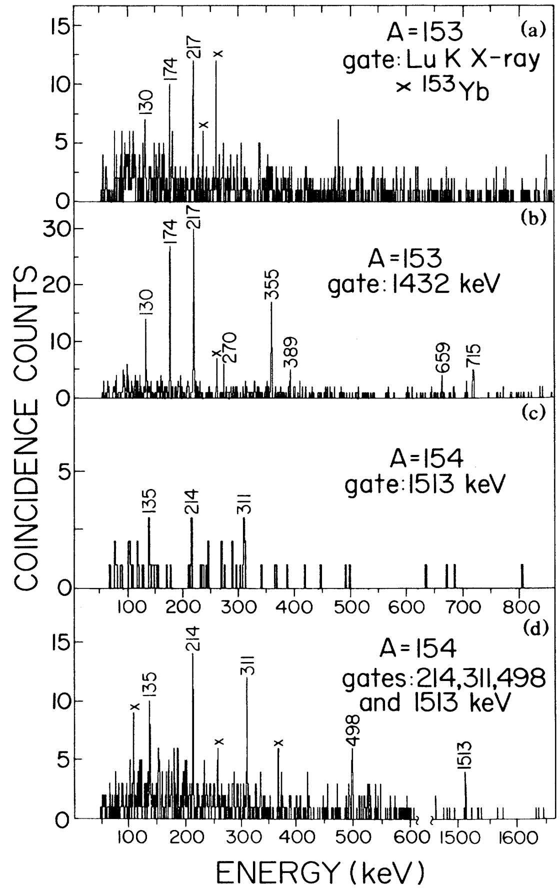

FIG. 1. (a)–(d) Key γ-ray coincidence spectra for the specified mass windows and gating transitions.

shown in Fig. 2 on the basis of the comprehensive $\gamma\gamma$ coincidence results. Spins and parities are assigned mainly by analogy with the similar decay schemes of the shorter-lived $\frac{27}{2}^-$ isomers[3] in ^{149}Ho and ^{151}Tm. The total conversion coefficient of the 130-keV transition, derived from intensity balance, is consistent with $E2$ character, and the measured half-life gives

$$B(E2;130\text{ keV},\,^{153}\text{Lu})=0.45\pm0.09\,e^2\text{fm}^4,$$

the smallest value determined in the $N=82$ series. The $\gamma\gamma$ time distributions for ^{153}Lu indicated a lower isomer with $t_{1/2}>0.1$ μs, that we associate with the $\frac{23}{2}^-$ state.

As Fig. 2 shows, the spectrum of $(\pi h_{11/2})^7$ levels calculated using the empirical $(\pi h_{11/2})^2$ interactions from ^{148}Dy is in very good agreement with experiment, especially for the four topmost levels. The $\frac{21}{2}^-$ level in ^{153}Lu, populated by an unobserved 21-keV transition from the $\frac{23}{2}^-$ level, has no observed counterparts in ^{149}Ho and ^{151}Tm. Theory[5] predicts such a level ~ 26 keV below the $\frac{23}{2}^-$ in each of the three nuclei, but the $\frac{23}{2}^- \rightarrow \frac{21}{2}^-$ $M1$ transition is strongly forbidden. It ap-

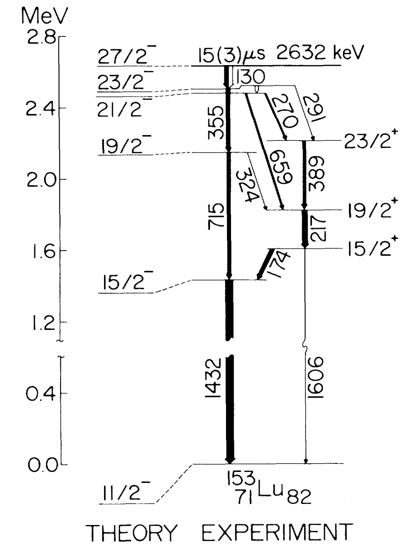

FIG. 2. Isomeric decay scheme for ^{153}Lu. Transition arrow widths are proportional to the measured intensities, with internal conversion contributions unshaded. The calculated $(\pi h_{11/2})^7$ spectrum, with the $\frac{27}{2}^-$ level energy matched to experiment, is shown to the left.

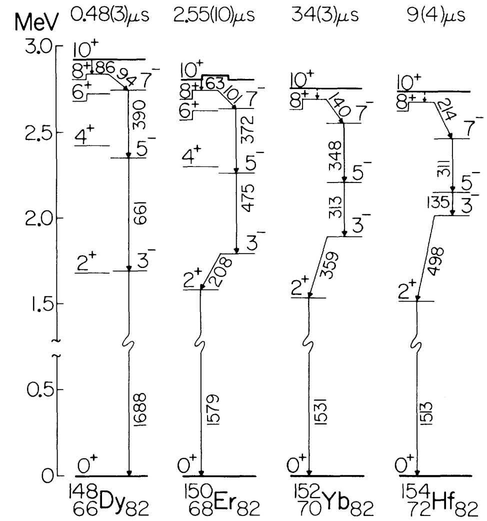

FIG. 3. Energy systematics of even-A $N=82$ isotones, showing the main decay pathways of the 10^+ isomers.

pears that the $\frac{23}{2}^- \rightarrow \frac{19}{2}^-$ $E2$ decay must be extremely retarded, as it is in ^{153}Lu, before the $\frac{23}{2}^- \rightarrow \frac{21}{2}^-$ transition becomes competitive. An interesting note is that the $\frac{27}{2}^-$ 15-μs isomeric state in ^{153}Lu has a positive Q value of 3.3 ± 0.7 MeV (Ref. 14) for proton decay to the ^{152}Yb ground state, but this decay mode must be suppressed by the centrifugal barrier associated with $l=13$ nucleon transfer and by the complicated nucleon rearrangements involved.

The possible $A=154$ products of the reaction were ^{154}Yb, ^{154}Lu, and ^{154}Hf, and of these the $N=84$ nucleus ^{154}Yb has no μs isomers. The most intense $A=154$ γ rays were found to be coincident with Lu x rays, and they are assigned[15] to a new 35-μs isomer in ^{154}Lu. Careful scrutiny of the $A=154$ γ-ray spectrum for a possible ^{154}Hf $2^+ \rightarrow 0^+$ transition in the 1490–1530-keV range suggested by systematics revealed a weak 1513-keV γ ray that decayed with the half-life $t_{1/2}=9 \pm 4$ μs. A gate on the 1513-keV transition [Fig. 1(c)] indicated coincidence peaks (each with 4–6 counts) at 135, 214, and 311 keV, and additional gates on these low-energy lines identified another γ ray of 498 keV in the same nucleus [Fig. 1(d)]. Although the data are statistically poor and provide no Z identification, the firm $A=154$ mass determination, the $N=82$ systematics, and the observed reaction yield (Table I) all favor the assignment of these five γ rays to the decay of a 9-μs isomer in ^{154}Hf. When the transitions are ordered as shown in Fig. 3, the smooth energy-level systematics[2-4] of the even-A $N=82$ isotones are extended to $Z=72$ in a convincing manner. The value of the ^{152}Yb 10^+ half-life shown in Fig. 3 was determined during the present studies. By assuming that the energies of the unobserved $10^+ \rightarrow 8^+$ $E2$ transitions in ^{152}Yb and ^{154}Hf lie within the range 14–70 keV, as systematics indicate, the following results are obtained:

$$B(E2;10^+ \rightarrow 8^+,\ ^{152}\text{Yb}) = 0.9 \pm 0.1\ e^2\text{fm}^4$$

and

$$B(E2;10^+ \rightarrow 8^+,\ ^{154}\text{Hf}) = 2.9 \pm 1.4\ e^2\text{fm}^4 .$$

The faster transition rate for ^{154}Hf is a clear signal that in this nucleus the $\pi h_{11/2}$ subshell is more than half-filled.

For both ^{153}Lu and ^{154}Hf, the $B(E2)$ results are much smaller than the values predicted using the equations and model assumptions specified earlier, and they imply that the $\pi h_{11/2}$ occupation numbers for these nuclei are in fact somewhat less than $Z-64$. The overall results are illustrated in Fig. 4, where the square root of the transition amplitudes, $B(E2)$, for the seven $N=82$ isotones are plotted versus $Z-64$. The square root leaves an ambiguity about the sign of the $E2$ matrix element, which should change at the point of half-filling, but the data strongly indicate that the sign change occurs just before Lu. The fact that the smooth curves through the data points both intersect the zero axis at the same value of $Z-64$ provides reassurance that correct signs have been chosen for the amplitudes. The variation of $E2$ transition amplitude with number of valence protons is not exactly linear, but nearly so, and it is clear that the $\pi h_{11/2}$ subshell is closest to being half-filled in the $Z=71$ isotone ^{153}Lu. It is less easy to specify the magnitude of the $\pi h_{11/2}$ $E2$ effective charge in this region, but it may be

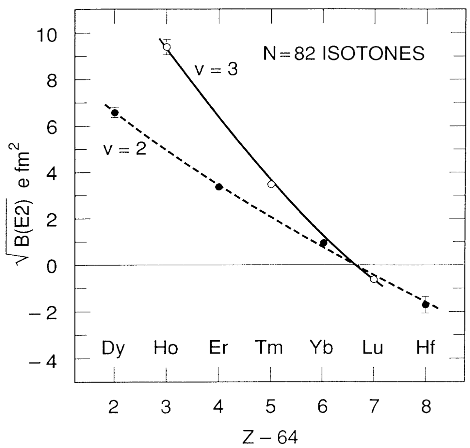

FIG. 4. Measured $E2$ transition amplitudes for $v=2$ and $v=3$ isomeric transitions in $N=82$ isotones as a function of $Z-64$. Where error bars are not shown, they lie within the plotted point.

significantly larger[8] than 1.52e, the value suggested previously.[2]

In summary, experiments using the Daresbury Recoil Mass Separator have identified long-lived $(\pi h_{11/2})^n$ isomers in the previously unknown proton-rich $N=82$ nuclei ^{153}Lu and ^{154}Hf. The results, together with those for lighter $N=82$ isotones, provide an outstanding illustration of the dependence of $E2$ transition rates between j^n states on the subshell occupation, and demonstrate that half-filling of the $\pi h_{11/2}$ subshell in the $N=82$ series occurs just below $Z=71$ ^{153}Lu.

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