Conversion electron spectroscopy at the fragment mass analyzer focal plane: Studies of isomeric decays near the proton drip line

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The decays of seniority isomers in the $N=82$ nuclei ^{150}Er and ^{152}Yb and in their respective $N=81$ isotopes ^{149}Er and ^{151}Yb were studied following mass separation by the Argonne Fragment Mass Analyzer. Conversion electrons were detected with Si p-i-n diodes operated at room temperature. The low-energy isomeric transitions in ^{151,152}Yb have been observed for the first time in the electron spectra. Multipolarity assignments were made for many of the decay γ rays of the four nuclei.

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I. INTRODUCTION

The low-lying yrast states of proton-rich nuclei above ^{146}Gd are well described in terms of shell model configurations involving $s_{1/2}$, $d_{3/2}$, and $h_{11/2}$ valence protons. Long-lived 10^+ and $27/2^-$ isomers with $(\pi h_{11/2})^n$ seniority $\nu=2$ and 3 aligned configurations are known in the $N=82$ isotones up to ^{154}Hf [1–7]. Related yrast isomers involving an additional neutron hole have also been identified in neighboring $N=81$ nuclei [8–10]. Since the cross sections for producing these isomers in fusion evaporation reactions drop sharply close to the proton drip line, the experimental difficulties in studying isomeric decays increase considerably with increasing proton number. Two of the previous investigations [7,10] have taken advantage of the improved sensitivity and selectivity that can be achieved by employing recoil mass spectrometers. In those cases, the evaporation residues were mass analyzed and collected behind the device's focal plane, where γ-ray decays of microsecond isomers were subsequently studied virtually without background.

The basic difficulty remains that measurements of isomeric decay γ rays generally yield no multipolarity information and, hence, nothing about the level spins and parities is known experimentally. Important exceptions are total conversion coefficients for low-energy transitions that are sometimes obtainable from intensity balance arguments. With most transition multipolarities undetermined, the interpretation of spectroscopic data for $N\sim82$ nuclei above $Z=66$ has relied on detailed systematics and on shell model calculations using empirical nucleon-nucleon interactions. An obvious drawback of this approach is that unexpected (and potentially important) changes in spectral features could be missed or misinterpreted. Furthermore, the isomeric transitions are often of very low energy and can escape detection with conventional γ-ray detectors as they are highly converted. The measurement of conversion electrons and γ rays occurring in these isomeric decays is the most direct experimental solution to both problems.

In the present work the Argonne Fragment Mass Analyzer (FMA) [11] was used to separate the isomeric species of interest from the beam and other reaction products. These nuclei were then implanted in a thin catcher foil behind the focal plane where both isomeric decay γ rays and conversion electrons could be detected free from the interference of prompt target radiation and delta electrons. With this arrangement, conversion electrons in the energy range from 20 to 500 keV could be measured with good energy resolution using Si p-i-n diodes. The first test experiment was performed with the reaction ^{94}Mo + 250 MeV ^{58}Ni $\rightarrow$ ^{152}Yb* and resulted in good yields for the 2.55 μs ^{150}Er and 4.8 μs ^{149}Er isomers, the products of $2p$ and $2pn$ evaporation. Isomeric decay schemes for these two nuclei were already well known from γ-ray measurements [3,4,6,9] and the present work has focused on establishing definite multipolarities for all the strong transitions below 500 keV. In the second experiment, isomeric decays were studied in ^{151,152}Yb nuclei produced in the reaction ^{96}Ru + 250 MeV ^{58}Ni$\rightarrow$^{154}Hf*. Although the cross sections of interest are smaller in this case, multipolarity assignments could be made for all yrast transitions in ^{152}Yb (with the exception of the 1531 keV $2^+\rightarrow0^+$ transition) and for one transition in ^{151}Yb. More importantly, in both nuclei, the low-energy transitions depopulating the isomers were identified for the first time from the electron spectra.

II. EXPERIMENTAL PROCEDURES AND RESULTS

A. Nuclei ^{149}Er and ^{150}Er

For the first experiment, a 250 MeV ^{58}Ni beam from the ATLAS accelerator bombarded a target consisting of two 267 μg/cm^2 isotopically enriched self-supporting ^{94}Mo foils stacked together. The evaporation residues were allowed to recoil in an axially symmetric cone about the z axis (i.e., the central axis for the ion optics of the FMA) and products with the same m/q were focused at the same position in the focal plane (i.e., the same x and y focus), while adjacent masses of the same charge state were separated by 6.7 mm. At the focal plane, a parallel plate avalanche counter (PPAC) provided a position and timing signal for the passing ions. Slits in front

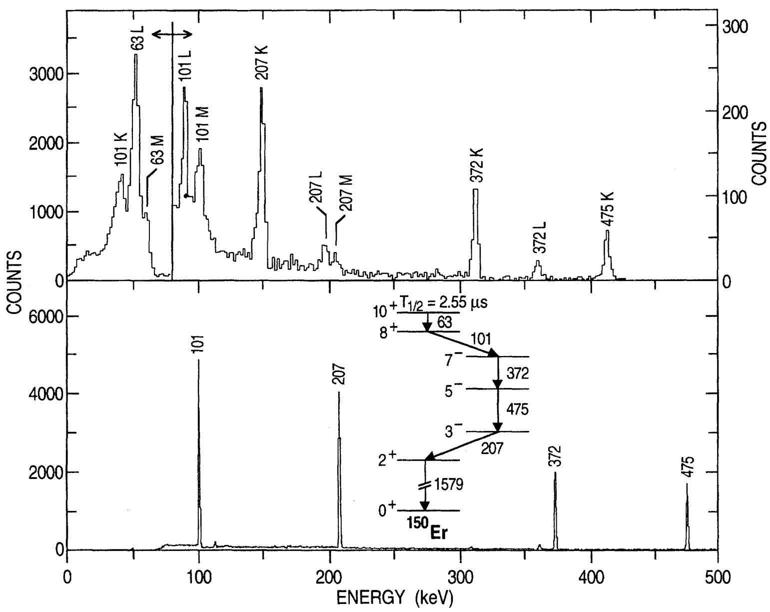

FIG. 1. Mass-gated conversion electron (top) and γ-ray (bottom) spectra up to 500 keV for $A = 150$. As seen from the ^{150}Er level scheme, all peaks can be associated with transitions in that nucleus.

of the PPAC were positioned such that only residues of masses $A=149,150$ with charge state $Q=26^+$ were allowed into the focal plane. These ions were stopped in an Al catcher foil placed approximately 15 cm behind the PPAC, at an angle of $\sim 45°$ with respect to the direction of the recoils. The effective thickness of the foil was 4.6 mg/cm^2. The time of flight for the evaporation residues through the FMA is $\sim 1\,\mu$s and ^{149,150}Er nuclei are the only reaction products of the appropriate mass to have isomeric states of sufficiently long lifetimes to be studied under the very low background conditions behind the FMA focal plane.

One large Ge detector placed approximately 20 cm from the catcher foil was used to detect γ rays. Conversion electrons were measured with two Hamamatsu Si p-i-n diodes (1 cm$\times$1 cm$\times$0.5 mm) operated at room temperature [12]. Relative efficiencies were determined using standard γ-ray sources for the Ge detector. In the case of the electron detectors, sources of ^{109}Cd, ^{141}Ce, ^{203}Hg, and ^{113}Sn provided the necessary calibration points. The efficiency curve for the electrons was found to be constant between 60 and 360 keV. At higher energies the electron detection efficiency drops quickly because of the detector thickness. For example, the efficiency between 360 and 420 keV is reduced by 30%. An event was written to tape whenever a delayed γ ray and/or electron were detected within 30 μs of a timing signal from the PPAC for the passing recoiling ion.

For the results reported here, the p-i-n diodes viewed the back of the catcher foil and covered a solid angle of approximately 10% of 4π. After correcting for the energy loss of the recoiling ions through the PPAC, the thickness of aluminum needed to stop the recoils (on average) was calculated to be 4.2 mg/cm^2, i.e., slightly less than the actual thickness of the catcher foil. This additional thickness has two effects. First, there is an increase in the measured width of electron lines due to straggling; with a 62 keV electron from a thin ^{109}Cd source, the resolution [full width at half maximum (FWHM)] was measured to be 3.5 keV, while 4.4 keV was recorded for K shell electrons of similar energy in the nuclei of interest. Second, the energies of the electrons are slightly reduced due to the energy loss in the catcher foil. For example, with the known K shell binding energy of 57.5 keV for an Er atom [13] and the measured γ-ray transition energy, 167.4 keV, the corresponding 167K conversion line in ^{149}Er should have an energy of 109.9 keV. After correcting the measured electron energy (108.9 keV) for the energy loss in Al, an energy of 110.0 ± 0.7 keV is obtained, which is in good agreement with the expected value.

In the analysis, electron and γ-ray spectra were obtained separately for ions with mass $A=149$ and $A=150$. Since each mass occurs at a different position in the FMA focal plane, a coincidence gate on the PPAC position signal was first applied. An additional coincidence condition on the PPAC ΔE energy loss signal was used as well in order to reduce the random coincidences due to scattered beam. These two requirements were the only ones used to reduce the background. Figure 1 shows the spectra obtained for $A=150$; all γ rays (bottom) and electrons (top) can be associated with the isomeric decay in ^{150}Er. From these spectra the energies and intensities of the various transitions were measured and the K, L, and M conversion coefficients were extracted.

The fitting of the low-energy electron peaks in the 40 – 80 keV range turned out to be a rather delicate task because of the number of transitions occurring within this small energy interval. For ^{149}Er, the K conversion electrons for the 132 keV transition, the L and M electrons of the 55, 63, and 69 keV transitions, and K x rays all occur within 30 keV. The electron spectrum for ^{150}Er was slightly less complicated. By fixing the line shape, position, and width of these lines the actual electron peaks could be extracted from the spectra and areas could be determined reliably. The line shapes and widths used in the fitting procedure were guided by the values determined from the calibration sources.

The K, L, and M internal conversion coefficients measured for the transitions in ^{150}Er are presented in Table I where they are also compared with calculated values [14]. In

TABLE I. Multipolarity assignments based on experimental conversion coefficients for transitions in ^{150}Er, ^{149}Er, ^{152}Yb, and ^{151}Yb. Calculated coefficients for $E1$, $E2$, and $M1$ multipolarities are shown for comparison.

Nucleus	Transition	α_{expt}	$E1$	$E2$	$M1$	Assignment
^{150}Er	63L	15.9(3.5)	0.12	14.0	1.0	$E2$
	63M	3.5(9)	0.034	3.6	0.30	
	101K	0.30(3)	0.30	1.1	2.5	$E1$
	207K	0.031(6)	0.042	0.14	0.28	$E1$
	372K	0.026(7)	0.01	0.030	0.07	$E2$
	475K	0.014(5)	0.0052	0.014	0.035	$E2$
^{149}Er	55L	23(20)	0.05	26	0.47	$E2$
	55M	8.3(7.3)	0.19	6.4	1.7	
	63L	1.2(7)	0.12	14.0	1.0	$M1$
	69L	9.3(3.5)	0.062	9.0	0.60	$E2$
	69M	2.4(9)	0.03	2.0	0.23	
	132K	0.51(15)	0.14	0.57	1.25	$E2$
	132L	0.6(2)	0.0042	0.44	0.038	
	167K	0.066(19)	0.07	0.25	0.60	$E1$
^{152}Yb	140K	0.19(4)	0.14	0.41	1.2	$E1$
	140L	0.018(8)	0.019	0.18	0.44	
	312K	0.040(12)	0.017	0.052	0.13	$E2$
	313L	0.010(4)	0.0022	0.019	0.020	
	348K	0.029(9)	0.013	0.036	0.10	$E2$
	359K	0.0074(23)	0.012	0.032	0.09	$E1$
^{151}Yb	203K	0.068(18)	0.05	0.16	0.42	$E1$

all cases the experimental coefficient could be matched to a theoretical value within 15%. In previous work [3,6], total conversion coefficients for some transitions were determined from γ-ray intensity arguments and multipolarities were assigned on this basis. The results reported here agree with the previous assignments and several new firm multipolarity assignments have been added.

The analysis for transitions in ^{149}Er was somewhat complicated by the fact that ions with mass $A=149$ are not stopped at the center of the catcher foil as are those with $A=150$ of the same charge state, but rather $\sim$1.0 cm off center as estimated by an ion optics calculation. Thus, the $A=149$ ions were further from the p-i-n diodes resulting in a smaller efficiency for detection of electrons. Measurements with a calibration source placed off center by 1.0 cm were performed to determine the value of the correction factor (approximately 20%) to be applied to the efficiency. The measured conversion coefficients for transitions in ^{149}Er are also shown in Table I. In this case also, several firm multipolarity assignments could be made.

B. Nuclei ^{151}Yb and ^{152}Yb

The second experiment used the same beam and energy from ATLAS, but used a single 350 μg/cm^2 ^{96}Ru target to produce ^{151,152}Yb nuclei through the $2pn$ and $2p$ reaction channels. Again, the FMA slits were closed to allow only $A=151,152$ ions with charge state 26^+ into the PPAC. In this experiment the focusing of the FMA was changed. The x focus remained at the center of the PPAC, but the y focus occurred approximately 40 cm further downstream. Ion optics calculations have shown that with this configuration all residues reaching the PPAC come together in one large spot at the y focus, but mass identification is still possible from the dispersion at the x focus. The catcher foil was placed at the y focus for the Yb experiment, in order to eliminate possible geometry effects due to the mass position on the electron detector efficiency.

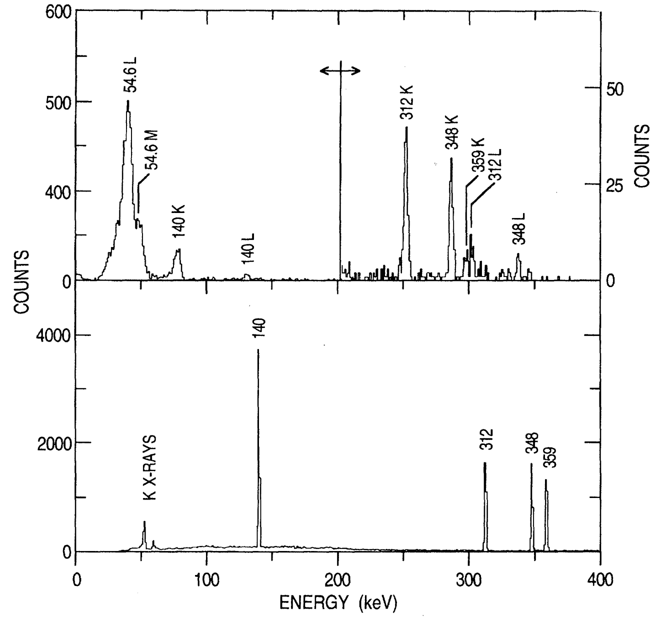

FIG. 2. Mass-gated conversion electron (top) and γ-ray (bottom) spectra for $A=152$. All peaks belong to the ^{152}Yb nucleus. The two lowest-energy peaks in the electron spectrum are the L and M conversion lines for the $10^+\rightarrow8^+$ transition, which had escaped detection in previous γ-ray measurements.

The time window for the event was changed to 100 μs in this experiment because of the longer half-lives involved [3,4,10]. In addition to isomeric decays in ^{151,152}Yb, γ rays and electrons associated with a long-lived state in ^{151}Tm were also registered in this case. Furthermore, with the addition of a second Ge detector, γ-γ and γ-electron coincidence events were also written to tape with the requirement of a time overlap of at most 4 μs. This condition allowed for detection of coincidence events across the known intermediate isomer ($T_{1/2}$ = 2.6 μs) in ^{151}Yb [10].

Mass-gated electron and γ-ray spectra were created for masses $A=151,152$ following an analysis procedure identical to that described above. An additional restriction was placed on the $A=151$ spectra in order to eliminate contaminants from ^{151}Tm by requiring that the time between the detection of the recoil and the subsequent γ-ray or electron decay lie between 5 and 100 μs. All ^{151}Tm events were removed from the spectra as the isomeric lifetime in this nucleus is only 0.47 μs [2]. By fitting all observed γ-ray and electron lines (Fig. 2), conversion coefficients were determined for transitions in ^{151,152}Yb and the results are again presented in Table I.

From the mass-gated electron spectra low-energy electron peaks, which are not associated with any γ ray, were observed in the experiment. This can be seen in the top of Fig.

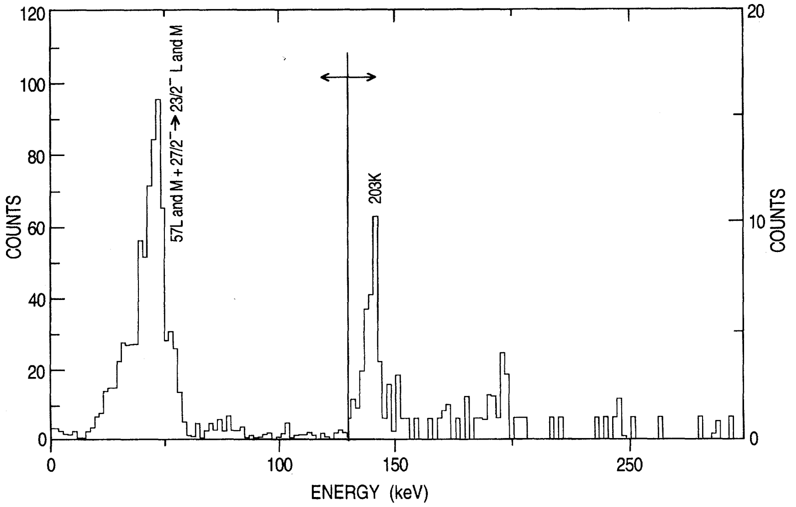

FIG. 3. Mass- and time-gated electron spectrum for $A=151$, performed to isolate conversion lines in the ^{151}Yb nucleus. The low-energy peaks are associated with two 57 keV transitions, one before and one after the known intermediate isomer (see text for details).

2 in the case of ^{152}Yb. These peaks are the L and M conversion lines of the transition deexciting the isomer in ^{152}Yb. The transition in ^{152}Yb was verified to belong to this nucleus through its γ-electron coincidence relationship. To determine the transition energy from the L and M conversion electron peaks, the electron energy loss in the Al catcher foil and the average binding energies of the L and M shells were taken into account. This transition is determined to have an energy of 54.6 $\pm$ 1.0 keV from the measured L and M peaks. With the number of L and M electrons seen in the spectrum, intensity balance arguments exclude the possibility that this transition is of dipole character. The γ ray associated with this transition is too weak to be separable from the large intensity of K_α x rays. However, a slight excess in the ratio of K_α to K_β x rays yields an upper limit to the intensity of this γ ray. With this upper limit on the γ-ray intensity and the sum of the intensities of the L and M electron peaks, a lower limit on the total conversion coefficient for the 54.6 keV transition can be obtained: $\alpha_{tot}(54.6\text{ keV}) > 23$.

This result rules out an $E1$ or $M1$ multipolarity for the 54.6 keV transition, but is consistent with an $E2$ or higher multipole order. This reinforces our assignment of the 54.6 keV transition as the long sought isomeric $10^+ \rightarrow 8^+$ transition in ^{152}Yb. For the low-energy transition (Fig. 3) in ^{151}Yb a similar analysis was conducted. In this case, contributions of a known 57 keV transition corresponding to a decay below the intermediate isomer is a potential problem. By using γ-e coincidence timing conditions, events corresponding to decays before and after the intermediate isomer could be isolated. However, an accurate fit to obtain the electron energies of the L and M conversion electron peaks for the $27/2^- \rightarrow 23/2^-$ transition was not possible due to poor statistics. It is interesting to note that peaks corresponding to the isomeric decay transition (i.e., before the intermediate isomer) and the aforementioned 57 keV L and M conversion electrons (i.e., after the intermediate isomer) occur at nearly the same energy in the electron spectrum. The assignment of 57 ± 2 keV to the $27/2^- \rightarrow 23/2^-$ transition energy reflects the difficulties just discussed.

Before this experiment the Z assignment for the family of γ rays assigned to the isomeric decay in ^{151}Yb [10] was based solely on calculations of production cross section [15]. With the conversion electron spectra measured here and the associated γ rays, it is possible to measure the K shell binding energy. Again, the energy loss of the electron through the aluminum catcher foil was taken into account. This measurement was carried out for the 203.1 keV transition. The corrected energy for the K shell conversion electron of 141.4 $\pm$ 0.7 keV results in an experimental determination of the K shell binding energy of 61.7 $\pm$ 0.7 keV. This value agrees well with the value of 61.3 keV expected for $Z=70$ [13]. This result removes any remaining uncertainty about the Z assignment of the transitions reported in Ref. [9].

III. DISCUSSION

Previous γ-ray measurements for the 2.55 μs 10^+ isomeric decay in ^{150}Er identified the main deexcitation pathway to the ground state through a cascade of six strong transitions [2–4,6]. For three of the six transitions, total conversion coefficients (or useful limits) were extracted from intensity balance requirements and as a result an $E2$ multipolarity had been established for the 63 keV transition while both the 101 and 207 keV transitions were shown to be of $E1$ character [6]. The three remaining transitions of 372, 475, and 1579 keV were assumed to be of $E2$ character, but without experimental verification. From the γ-ray intensity of the transitions seen in the ^{150}Tm$\rightarrow$^{150}Er β decay [17] the ordering of the transitions in the 10^+ isomeric decay could be established to be 63, 101, 372, 475, 207, and 1579 keV. In the present work, the multipolarities of all but the 1579 keV transition have been determined and the previously proposed $10^+ \rightarrow 8^+ \rightarrow 7^- \rightarrow 5^- \rightarrow 3^- \rightarrow 2^+ \rightarrow 0^+$ decay sequence now is firmly established. The seniority-two ($\nu=2$) excitations known below 3 MeV in the four valence proton nucleus ^{150}Er include a complete $(\pi h_{11/2})^n$ spectrum up to 10^+, $(\pi h_{11/2} s_{1/2})5^-$ and $(\pi h_{11/2} d_{3/2})7^-$ states together with some nonyrast members of these multiplets, and a 3^- octupole excitation with a dominant $(\pi h_{11/2} d_{5/2}^{-1})$ particle-hole component. This interpretation is supported by shell model calculations [6,16].

In the original γ-ray study [9] of the 4.8 μs ^{149}Er isomeric decay, total conversion coefficients, and multipolarities for several key low-energy transitions were inferred from intensity balance requirements. The present measurements confirm the previous results, establishing $E2$ character for the 69 and 132 keV transitions, $M1$ multipolarity for the 63 keV transition, and $E1$ character for the 167 keV transition. Also, the 55 keV transition is consistent with the previously assigned $E2$ multipolarity, though the large error bars associated with this measurement do not allow for a unique identification. These multipolarity assignments provide support for the interpretation given in Ref. [8] describing the observed levels as three-quasiparticle states arising from the coupling of $s_{1/2}$, $d_{3/2}$, and $h_{11/2}$ neutron holes with $(\pi h_{11/2})^n$ states.

Gamma-ray studies of the 10^+ isomeric decay in ^{152}Yb [4,5] have identified five intense transitions of 140, 312, 348, 359, and 1531 keV in cascade, but in the 0.7 s β decay of ^{152}Lu to ^{152}Yb only the 312, 359, and 1531 keV γ rays occur strongly [18], establishing these as the bottom three transitions. The $E2$ and $E1$ multipolarities obtained here for the 312 and 359 keV transitions, respectively, show that these are the $5^- \rightarrow 3^-$ and $3^- \rightarrow 2^+$ transitions feeding the state at 1531 keV. These conclusions are supported further by the observation of a weak 1889 keV $3^- \rightarrow 0^+$ γ ray in parallel with the 359 and 1531 keV transitions [4]. This 1889 keV γ ray is the counterpart to the octupole transitions to the ground states found in the isotones ^{146}Gd, ^{148}Dy, and ^{150}Er at respective transition energies of 1579, 1688, and 1786 keV. The 109 keV shift of this state from ^{146}Gd to ^{148}Dy has been attributed [1] to a Pauli interference effect arising from the coupling of the 3^- excitation to the $(\pi h_{11/2})^2 0^+$ component of the ^{148}Dy ground state. Since the ^{150}Er and ^{152}Yb nuclei have two and three such 0^+ pairs, one can expect an upwards shift of 2×109 and 3×109 keV in their 3^- energies with respect to the 1579 keV state in ^{146}Gd. The resulting estimates, 1797 keV in ^{150}Er and 1906 keV in ^{152}Yb, are within 17 keV of the experimental 3^- energies in the two isotones.

As shown in Fig. 4, the ^{152}Yb 10^+ isomer at 2744 keV deexcites via a $10^+ \rightarrow 8^+ \rightarrow 7^- \rightarrow 5^- \rightarrow 3^- \rightarrow 2^+ \rightarrow 0^+$ sequence which is the same as that presented above for ^{150}Er and the six excited states in ^{152}Yb can be associated with similar configurations to those discussed above for ^{150}Er. With the identification of the isomeric transition and the accurate measurement of the 10^+ lifetime (30 ± 1 μs), the $10^+ \rightarrow 8^+$ $E2$ transition probability can be determined precisely for the first time. The result $B(E2;10^+ \rightarrow 8^+, {}^{152}\mathrm{Yb}) = 0.95(4)e^2$ fm^4 corresponds to 0.02 Weisskopf units. This very low transition probability is a consequence of the $h_{11/2}$ proton subshell in ^{152}Yb nearing the point of half-filling, where particle and hole contributions to the $E2$ transition amplitude cancel exactly and the $B(E2)$ becomes zero. In the $N=82$ isotone series, the $\pi h_{11/2}$ subshell becomes half-filled just below the $Z=71$ ^{153}Lu nucleus as has been discussed in detail in Ref. [7]. The observation that the $\pi h_{11/2}$ subshell is half filled at $Z=71$ rather than at $Z=70$ has been

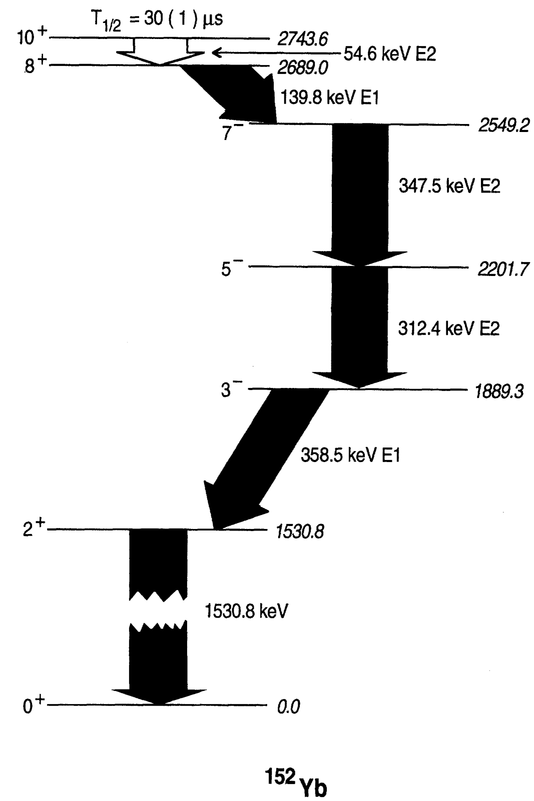

FIG. 4. The low-lying yrast level scheme below the 10^+ isomer in ^{152}Yb. With the exception of the 1531 keV γ ray, all transition multipolarities have now been determined.

attributed to the partial occupation of the $s_{1/2}$ and $d_{3/2}$ proton orbitals which are close in energy to the $h_{11/2}$ orbital above the $Z=64$ shell gap. It is possible that this partial occupation is also responsible for the gradual decrease in energy of the $10^+ \rightarrow 8^+$ isomeric transition from 86 keV in ^{148}Dy to 63 keV in ^{150}Er and to 55 keV in ^{152}Yb ($Z=70$). Within this $(h_{11/2})^n$ seniority scheme, this transition is expected to remain constant in energy [16].

IV. SUMMARY

The decays of seniority isomers in the $N=82$ nuclei ^{150}Er and ^{152}Yb and in their respective $N=81$ isotopes ^{149}Er and ^{151}Yb were studied following mass separation by the Argonne Fragment Mass Analyzer. Under the extremely low background conditions behind the FMA focal plane, conversion electrons and γ rays were detected and many conversion coefficients for the yrast transitions in these nuclei were obtained. The low-energy transitions deexciting the isomers in ^{151,152}Yb have been identified for the first time.

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