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Decay of ^{172}Hf to levels in ^{172}Lu

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The electron-capture decay of ^{172}Hf was investigated. Single and coincidence measurements were made by using germanium x-ray detectors. In addition, a Si(Li) detector was used to study the low-energy portion, $\lesssim 60$ keV, of the γ-ray spectrum. These results were then combined with available conversion-electron data which had been obtained with a magnetic spectrometer. From this information, a decay scheme for ^{172}Hf was constructed. It incorporates 17 transitions into 11 excited levels in ^{172}Lu and differs from previously proposed decay schemes. Several transitions previously attributed to ^{172}Hf decay were not observed in the present study. The electron-capture decay energy was deduced from measured K x-ray intensities and the level scheme to be 350 ± 50 keV.

[RADIOACTIVITY ^{172}Hf, measured E_γ, I_γ, I_x, $\gamma\gamma$ coin; deduced Q_{EC}. ^{172}Lu deduced levels J^π.]

I. INTRODUCTION

The electron-capture decay of 1.87-y ^{172}Hf has been investigated by several groups. Three of the more extensive studies have been the conversion-electron measurements of Harmatz and Handley,[1] and gamma and lifetime determinations of Jastrzebski *et al.*,[2] and the photon measurements of Sen.[3] The decay schemes proposed in Refs. 1 and 3 are in serious disagreement. The goal of the present investigation was to reexamine the decay of ^{172}Hf in an attempt to resolve the differences between Refs. 1 and 3. (Both the Nuclear Data Sheets[4] and the Table of Isotopes[5] have adopted the decay scheme proposed by Sen.[3] It should be noted that in the Nuclear Data Sheets the unpublished data of Ref. 3 have been erroneously referenced to an article[6] dealing with the decay of ^{172}Lu.)

The source used in this investigation had been originally prepared in an effort[7] to observe α-particle emission from ^{172}Hf. It was produced by bombarding natural lutetium oxide (97.41% ^{175}Lu) with 40-MeV protons accelerated in the Oak Ridge isochronous cyclotron. The incident energy was selected to correspond to the expected maximum of the $(p, 4n)$ excitation function. The hafnium activity was chemically separated (see Ref. 7 for the procedure) from the target and electroplated onto a platinum disk. At the time of the present measurements, the only remaining radioactivity in the source was ^{172}Hf in equilibrium with its 6.7-d daughter ^{172}Lu. Low-energy singles and coincidence γ-ray spectra were taken with an intrinsic germanium and a Ge(Li) x-ray detector. The coincidence information was stored on magnetic tapes in a γ-γ-τ list mode. The portion of the γ-ray spectrum below ~60 keV was also studied with a Si(Li) detector.

II. RESULTS AND DISCUSSION

Results of the singles and coincidence measurements are summarized in Tables I and II, respectively. In Table I our data are compared with transition energies deduced from conversion-electron measurements[1] and with photon intensities reported in Refs. 2 and 3. Unless otherwise noted, multipolarities given in Table I are based on the electron intensities of Harmatz and Handley[1] and on our photon data. Total conversion coefficients are taken from the theoretical calculations of Hager and Seltzer[8] and from Dragoun *et al.*[9] by using the computer program developed by the Nuclear Data Project. As can be noted in Table I, a number of transitions reported in the conversion-electron work of Ref. 1 were not observed in the present study. In a

TABLE I. Energies, photon intensities, and multipolarities of transitions in ^{172}Lu.

E_γ (keV) Ref. 1	Present work	I_γ [a] Ref. 2	Ref. 3	Present work	Multipolarity [b]	Total conversion coefficient [c]
	K x rays			950 (50)		
11.8	d			<0.4	$M1$	80.0
12.5	12.41 (20)			~0.1	$M1$ (+$E2$)	300
···	19.94 (10)		0.37 (7)	0.4 (2)		
23.4	d					
24.0	23.98 (5)	193 (50)	225 (26)	180 (10)	$E1$	3.30
34.6	Not seen			<0.1		
41.2	41.13 (10)		2.86 (12)	2.37 (20)	$M1$	8.49
41.9					$M3$	26400
44.15	44.17 (10)		~2.8	2.8 (5)	$E2$	122
48.3	48.17 (20)			~0.8	$M1$ + ~3 % $E2$	7.57
60.65	d			~10 [e]	$M1$ (+$E2$)	2.69
67.4	67.35 (10)	46 (7)	55 (2)	47 (5)	$E1$	0.987
···	68.00 (10)			6.1 (6)	($E1$) [f]	0.963
70.0	69.99 (10)	7.2 (18)	8.4 (4)	7.4 (8)	$M1$ + 2 % $E2$	10.7
73.9	Not seen			<0.1		
81.7	81.75 (5)	35 (4)	50 (2)	40 (2)	$M1$	6.76
91.3	d	1.0 (4)				
114.1	114.06 (10)	20.5 (20)	21.3 (6)	22.6 (25)	$M1$	2.59
116.1	Not seen			<0.3		
119.0	d					
122.95	122.92 (10)	11.5 (30)	11.8 (4)	10.1 (10)	$E2$	1.56
125.8	125.82 (5)	100	100 (2)	100 (5)	$M1$	1.95
···	127.91 (10)	14.5	9.7 (32)	12.9 (13)	($E1$) [f]	0.185
138.1	Not seen	≤0.4		<0.2		
142.4	Not seen	≤0.4		<0.1		
148.8	Not seen			<0.1		
150.4	Not seen			<0.1		
154.75	154.72 (10)	1.3 (2)	0.96 (7)	1.3 (2)	$M1$	1.09
172.2	Not seen			<0.1		
178.5	Not seen			<0.1		
198.9	Not seen			<0.1		
202.5	d					

[a] Relative intensities based on a value of 100 for the 125.82-keV transition.
[b] From conversion-electron data in Ref. 1 and γ intensities from present work, unless otherwise noted.
[c] Total theoretical conversion coefficients from Hager and Seltzer (Ref. 8) and Dragoun *et al.* (Ref. 9).
[d] Obscured by neighboring intense photon peaks.
[e] Calculated from L_1-conversion electron intensity (Ref. 1) and α_{L_1} theoretical conversion coefficient (Ref. 8).
[f] Deduced from absence of conversion lines.

TABLE II. Coincidence results. The letter Y indicates coincidences observed. Transitions that are definitely not in coincidence with one another are indicated by the letter N.

Gate (keV) \ E_γ (keV):	23.98	41.13	44.17	68.00	69.99	81.75	114.06	122.92	125.82	127.91	154.72
23.98		Y	Y	Y	Y	Y	Y	N	Y	N	
41.13	Y				Y	Y			Y		
44.17	Y				Y	Y					
68.00	Y				Y	Y					
69.99	Y	Y	Y	Y					N		
81.75	Y	Y	Y	Y					N		
114.06	Y								N		
122.92	N										
125.82	Y	Y			N	N	N				
127.91	N										
154.72											

few cases, if these transitions exist, they were obscured by photon peaks due to the decay of ^{172}Lu. In the course of analyzing the coincidence data, it was possible to set gates on the 11 γ-ray peaks listed in Table II. Gates were also set on the Lu and Yb K x-ray peaks; all transitions listed in Table II were seen in coincidence with Lu K_{α_1} and K'_{β_2} x rays.

Figure 1 shows a selected portion of the singles spectrum [part (a)] and spectra in coincidence with Lu K_{α_1} [part (b)] and Yb K_{α_2} [part (c)] x rays. As is the case for the well-established 69.99-keV transition of ^{172}Hf, the new 68.00-keV transition (Table I) can be clearly seen in Fig. 1(a) and Fig. 1(b) but not in Fig. 1(c). Note that, as one would expect, the 67.35-keV transition which depopulates[4,5] the 440-μs 109.4-keV level in ^{172}Lu is not seen in Fig. 1(b). The 60.65-keV transition reported by Harmatz and Handley[1] is obscured in Fig. 1(a) by the Lu K'_{β_1} and Yb K'_{β_2} x rays. There is an indication of its presence in Fig. 1(b) where the Yb K'_{β_2} x-ray peak has been eliminated. Supporting evidence was obtained by comparing the ratio of the Lu K'_{β_1} and K'_{β_2} intensities in Fig. 1(b) with the expected[10] value of 3.7. The experimental ratio was 4.4. By using the expected value of 3.7, it was possible to deduce an intensity for the 60.65-keV transition relative to the 69.99-keV photon peak seen in the same coincident spectrum [Fig. 1(b)]. The experimental coincidence ratio $I_\gamma(69.99)/I_\gamma(60.65)$ of ~2.4 agrees with the value of 2.3 ± 0.5 calculated from the proposed decay scheme (Fig. 2) and the photon intensities listed in Table I.

The ^{172}Hf decay scheme is shown in Fig. 2. With the exception of the 19.94-keV transition, it incorporates all of the γ rays observed in this study. Our data support the existence of the 41.9-, 65.8-, 109.4-, and 232.5-keV levels and the placement of their associated radiations.[4,5] The 109.9-keV level proposed earlier[1,2,11] was omitted in Ref. 3. Our coincidence data indicate that the new 68.00-keV transition deexcites this level. The 133.3-keV state reported in Ref. 1 is eliminated because the 67.35-keV transition was not observed in coincidence with the 23.98-keV γ ray in Refs. 3 and 12 where the delayed-gate technique was used. Since the 69.99- and 81.75-keV γ rays were observed by us in coincidence with the 44.17- and 68.00-keV transitions, they cannot deexcite the proposed[3] 135.91- and 147.61-keV states. Instead, our coincidence information

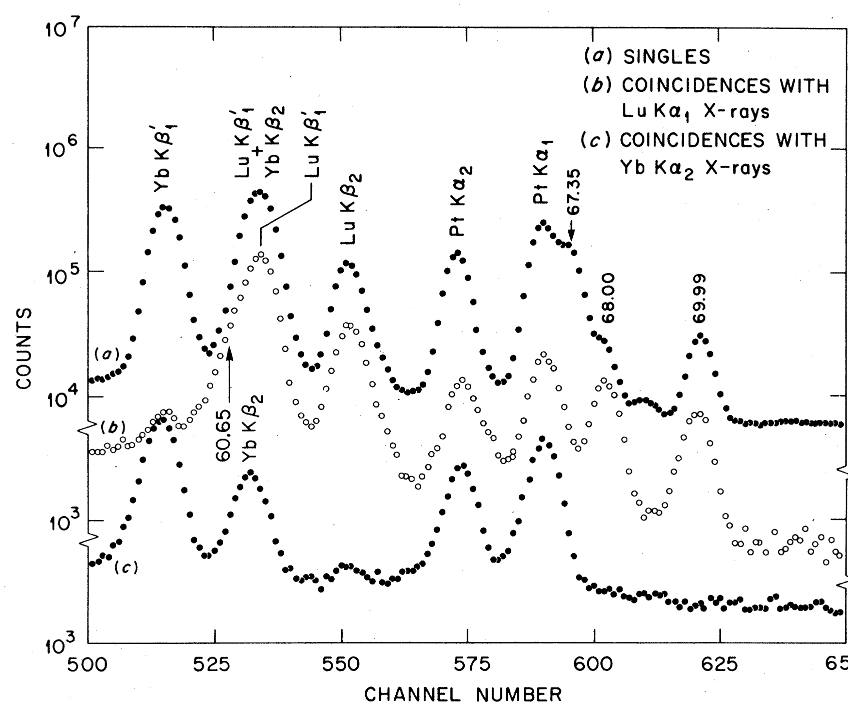

FIG. 1. (a) Selected portion of the singles spectrum; (b) spectrum in coincidence with Lu K_{α_1} x rays; and (c) spectrum in coincidence with Yb K_{α_2} x rays. The new 68.00-keV transition can be seen in parts (a) and (b). The position of the 60.65-keV transition observed in the conversion-electron data of Ref. 1 is indicated in part (b). See the text for a discussion of evidence for its presence in our γ-ray spectral measurements.

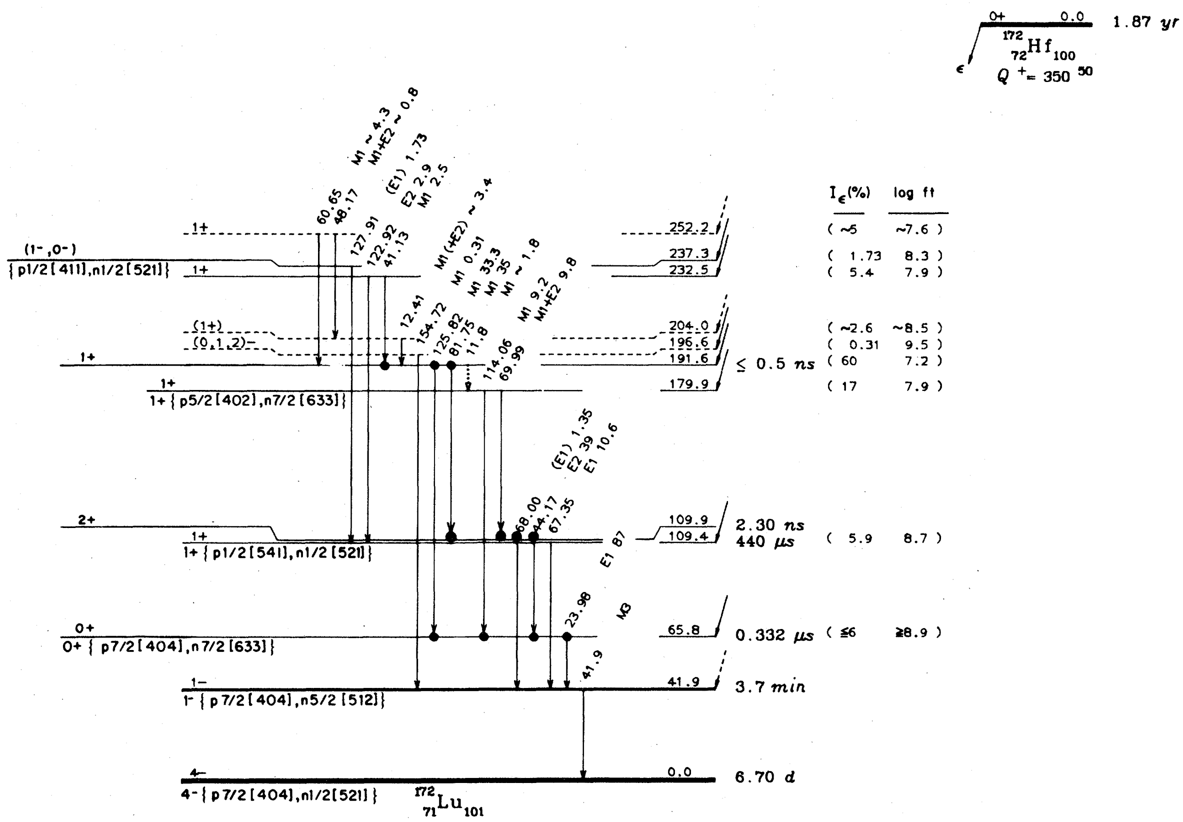

FIG. 2. Decay scheme of ^{172}Hf. Dots indicate observed coincidences. Numbers following γ-ray energies and multipolarities represent total transition intensities per 100 ^{172}Hf electron-capture decays. They are calculated from our photon intensities (Table I) and from theoretical conversion coefficients (Refs. 8 and 9). Half-lives shown for levels are taken from references quoted in Nuclear Data Sheets (Ref. 4).

strongly suggests that they depopulate the 179.9- and 191.6-keV states, as suggested in Ref. 1. Based on the half-life measurements of Jastrzebski *et al.*,[2] the placement[3-5] of the 44.17-keV transition as deexciting the 191.6-keV state is incorrect. The half-lives of the states deexcited by the 125.82- and 44.17-keV γ rays are $\leq$0.5 ns and 2.30 ns, respectively. Thus, the two transitions cannot depopulate the same level as proposed in Ref. 3.

The 264-keV state (Refs. 1 and 3) probably does not exist because the 127.91- and 154.72-keV transitions were not seen in prompt coincidence with any γ ray. These two transitions, therefore, can only populate the 41.9- and 109.4-keV levels. Comparison of the singles spectrum with those in coincidence with K x rays revealed that the ratio $I_\gamma(128)/I_\gamma(155)$ was less in the coincident spectra than in the singles. One postulates then that the level deexcited by the 127.91-keV γ ray has to be above the level deexcited by the 154.72-keV γ ray. We propose that the former transition originates from a 237.3-keV state [a level at 237 keV is seen in a ^{171}Yb(^{3}He,d) reaction study[13]], and the 154.72-keV transition deexcites a 196.6-keV state.

A level at 252 keV was suggested by Harmatz and Handley[1] to be depopulated by γ rays of 60.6, 119.0, and 142.4 keV. In the present study, we observed that the $K'_{\beta_1}/K'_{\beta_2}$ ratios were larger in spectra in coincidence with the 69.99-, 81.75-, 114.06-, and 125.82-keV transitions than in the ones in coincidence with 122.92-, 127.91-, and 154.72-keV γ rays. This supports the placement of the 60.65-keV transition as originating from a level at 252.2 keV. On the basis of energy fits and transition intensities, we suggest that the 48.17-keV γ ray deexcites this level and proceeds to a new level at 204.0 keV, which is then depopulated by the 12.41-keV transition.

The 41.9-keV transition is highly converted and was not observed in our γ-ray spectra. Its total intensity can be calculated from conversion-electron intensities.[1] These intensities may be converted to the relative units in Table I by using theoretical conversion coefficients[8,9] for the 23.98-, 125.82-, and 114.06-keV transitions.

(The expected photon intensity for the 41.9-keV transition was calculated to be 0.03 ± 0.01.) The intensity balance at the 41.9-keV level implies that within the experimental uncertainties, there is no direct capture feeding to it. The absolute photon intensities were then obtained by assuming that any capture branch to that level is negligible. Therefore, the total intensities of the four transitions populating the 41.9-keV level were taken to be 100% of ^{172}Hf electron-capture decays. Capture branches shown in Fig. 2 were then deduced from intensity balance at each level.

The electron-capture decay energy for ^{172}Hf has been estimated[14] from systematics to be 400 keV. We used our K x-ray intensity and selected coincidence intensities to deduce the Q value. The total K x-ray intensity due to K-shell electron conversions and K captures was calculated as a function of Q. In addition, the intensities of the 114.06-, 122.92-, 125.82-, and 127.91-keV γ rays in coincidence with K x-rays were calculated, once again, for various values of the decay energy. The calculated intensities were then compared with the measured numbers for a range of Q values. From this comparison, the value of 350 ± 50 keV was arrived at for the ^{172}Hf decay energy. The logft values shown in Fig. 2 were calculated by using this Q_{EC} and the deduced capture branches.

A spin of 4 has been measured[15] for the ^{172}Lu ground state. Its parity must be negative, based on the logft value[4,5] for the capture decay of ^{172}Lu to the 6^+ level of the ground-state rotational band in ^{172}Yb. The remainder of the spin and parity assignments shown in Fig. 2 for ^{172}Lu levels are based on the ground-state assignment, transition multipolarities, and logft values. Nilsson orbital assignments indicated in Fig. 2 were arrived at as follows.

From systematics,[16] the $\frac{7}{2}$[404] and $\frac{1}{2}$[541] proton orbitals and the $\frac{1}{2}$[521] and $\frac{7}{2}$[633] neutron orbitals are expected to comprise the main components of the ^{172}Lu ground-state structure. The magnetic dipole moment of this level has been measured.[17] Comparisons of calculated[17,18] moments with the experimental value show that the predominant configuration is $4^-\{p\frac{7}{2}[404], n\frac{1}{2}[521]\}$ with possibly a small admixture of $\{p\,\frac{1}{2}[541], n\frac{7}{2}[633]\}$.

Based on the 41.9-keV, $M3$ transition, a $1^-\{p\frac{7}{2}[404], n\frac{5}{2}[512]\}$ configuration has been suggested in previous studies[1,11,12] for the 41.9-keV level.

The 65.8-, 109.0-, and 191.6-keV levels have been proposed[1,11,12] to be the 0^+, 2^+, and 1^+ members of the $K=0$, $\{p\frac{7}{2}[404], n\frac{7}{2}[633]\}$ rotational band. These assignments are consistent with hindrance factors calculated[19] from level half-lives and transition intensities. The Weisskopf hindrance factor of the new 68.00-keV γ ray is

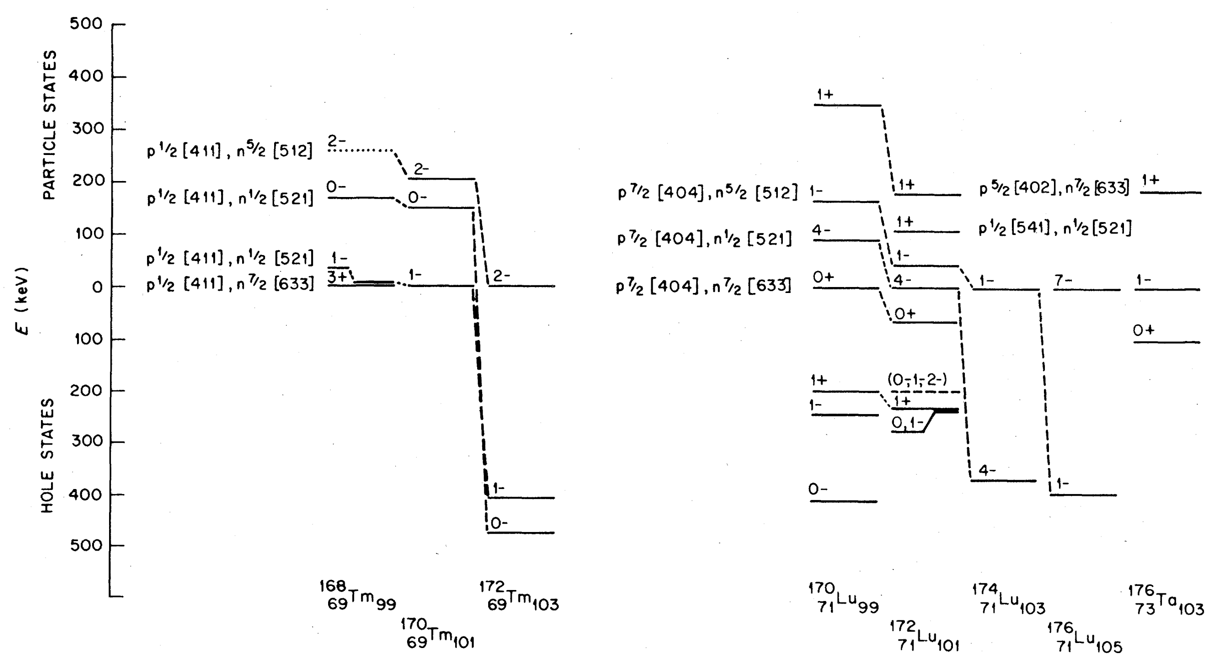

FIG. 3. Nilsson configurations proposed for ^{172}Lu, together with corresponding states in neighboring odd-odd nuclei. Data and assignments for these nuclei are taken from Refs. 20–26 and the present work.

about the same as that of the 23.98-keV transition. Both transitions proceed to the 1^-, 41.9-keV level; this is consistent with the band assignments given above.

The $1^+\{p\,\frac{1}{2}[541], n\,\frac{1}{2}[521]\}$ state is expected at low excitation energies. Harmatz and Handley[1] proposed that the 179.9-keV level has this particular configuration. However, one expects transitions from such a state to the band members of the $0^+\{p\,\frac{7}{2}[404], n\,\frac{7}{2}[633]\}$ state to be forbidden. Experimentally, no hindrances are observed; instead, the 69.99-, and 114.06-keV γ rays are in prompt coincidence with K x rays. We suggest that on the basis of systematics, the main component of the 179.9-keV levels is $1^+\{p\,\frac{5}{2}[402], n\,\frac{7}{2}[633]\}$.

A more likely candidate for the $1^+\{p\,\frac{1}{2}[541], n\,\frac{1}{2}[521]\}$ state is the level at 109.4 keV. A transition between it and the $1^-\{p\,\frac{7}{2}[404], n\,\frac{5}{2}[512]\}$ state should be forbidden. This expected forbiddenness is consistent with the large hindrance factor for the 67.35-keV transition and the resultant long half-life of the 109.4-keV level.

Finally, of the higher lying levels only the 196.6- and 237.3-keV states have negative parities. Possible negative-parity configurations are $\{p\,\frac{1}{2}[411] \pm n\,\frac{1}{2}[521]\}$ with $K=1$ and $K=0$. The existence of the prompt 154.72-keV transition to the $1^-\{p\,\frac{7}{2}[404], n\,\frac{5}{2}[512]\}$ state argues against the $\{p\,\frac{1}{2}[411] \pm n\,\frac{1}{2}[521]\}$ assignment for the 196.6-keV level. However, this same assignment to the 237.3-keV state is consistent with the level's prompt decay, via the 127.91-keV transition, to the $1^+\{p\,\frac{1}{2}[541], n\frac{1}{2}[521]\}$ state at 109.4 keV.

The Nilsson states proposed for ^{172}Lu are shown in Fig. 3, together with the corresponding states in neighboring odd-odd isotopes. Configurations for these nuclei were taken from Refs. 20–26 as follows: ^{170}Lu,[20] ^{174}Lu,[21] ^{176}Lu,[22] ^{176}Ta,[23] ^{168}Tm,[24] ^{170}Tm,[25] and ^{172}Tm.[26] It is seen that that the firm assignments for ^{172}Lu levels generally fit into the overall pattern. On the basis of these trends, it is possible that the $(0^-, 1^-, 2^-)$, 196.6-keV level (Fig. 2) corresponds to the $2^-\{p\,\frac{1}{2}[411], n\frac{5}{2}[512]\}$ state seen in the thulium nuclei. In Ref. 20 the 349.0-keV level was identified as the $1^+\{p\,\frac{1}{2}[541], n\frac{1}{2}[521]\}$ state. However, since it decays to the ground-state band with the $0^+\{p\,\frac{7}{2}[404], n\,\frac{7}{2}[633]\}$ configuration, a more likely assignment (as indicated in Fig. 3) is $1^+\{p\,\frac{5}{2}[402], n\,\frac{7}{2}[633]\}$.

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