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Nuclear Spectroscopy in the Rare Earth Region Near the Proton Drip Line

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ABSTRACT

We have used the isotope separator facility OASIS, on-line at the Lawrence Berkeley Laboratory SuperHILAC, to investigate rare earth nuclei close to the proton drip line. Single-particle states near the 82-neutron shell have been examined and their excitation energies determined. Numerous new isotopes, isomers, and β-delayed proton emitters have been discovered. In addition, the α-decay properties of nuclides with $N > 84$ have been reexamined; this has led to the discovery of several previously unobserved α transitions. The overall experimental program is summarized and some recently obtained results on ^{145}Dy, ^{147}Er, ^{147}Tm, ^{153}Lu, ^{155}Lu, and ^{157}Lu are discussed.

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I. INTRODUCTION AND EXPERIMENTAL METHOD

With the use of the OASIS separator facility,[1] on-line at the Lawrence Berkeley Laboratory SuperHILAC, we have investigated the decay properties of numerous short-lived neutron-deficient rare earth nuclei with $65 \leq Z \leq 71$. Figure 1 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. Note that most of the nuclei in Fig. 1 with $N \geq 84$ are α-particle emitters. 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 and no α-emission has been observed in rare earth isotopes with $N < 84$. Table I lists the investigated radioactivities together with their half-lives and spin assignments. We should add that, in a separate experimental program, the OASIS separator has also been used to study other proton-rich rare earth nuclei, primarily those with $Z \leq 66$ (see Ref. 2).

The isotopes in Table I were produced in fusion reactions induced by bombardments of ^{96}Ru, ^{92}Mo, ^{94}Mo, ^{95}Mo, ^{96}Mo, and ^{93}Nb with ^{64}Zn and ^{58}Ni projectiles. Reaction products recoiling out of the (2 - 3)-mg/cm^2 targets were stopped in a Ta catcher foil located inside a surface ionization source. After diffusing out of the catcher and being ionized, the recoils were accelerated to 50 keV and mass separated. Products of a single mass were then selected by an analyzing slit in the focal plane of the separator, transported ionoptically to a fast-cycling, computer-controlled, tape system, and positioned between an array of detectors (Fig. 2). This consisted of a Si particle ΔE-E telescope and

and a hyperpure Ge detector facing the radioactive layer, with a 1-mm thick plastic scintillator and an n-type Ge detector (relative efficiency of 52%) located on the other side of the tape. In addition, a 24% n-type Ge detector, oriented at 90° with respect to the other two Ge detectors, was placed ~ 4.5 cm from the radioactive source. Coincidences between particles, γ rays, x rays, and positrons were recorded in an event-by-event mode with all events tagged with a time signal for half-life information. Singles γ-ray data were acquired with all three Ge detectors. While counting was in progress a fresh source of radioactivities was collected and then transported to the detector station when the counting interval had terminated. Time intervals were selected on the basis of known or expected half-lives of the evaporation residues of interest.

II. RESULTS AND DISCUSSION

A. Delayed Proton Emission For N = 81 Precursors

As a result of high level densities, β-delayed-proton decay originating from nuclei with $A \geq 80$ 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. 3 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 is explained by the fact that for the 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); the resultant spectra are adequately described[3] by the statistical model. In contrast, the proton spectra of the even-Z precursors exhibit pronounced structure which has been shown[4] to be associated with the decay of the $1/2^+$ ground states. The structureless components of these spectra are associated primarily with the $11/2^-$ isomeric states in the same nuclei (because of angular momentum considerations, these protons originate[4] from levels at high excitation energies). Gamma-ray decay studies, β-strength function measurements, and calculations of state densities and Gamow-Teller strength distributions, have led us to suggest[5] that some of the structure near N = 82 arises from the preequilibrium decay of doorway states populated in β decay and that it can be understood within the framework of a model based on the work by Bloch and Feshbach.[6]

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[7] where the results obtained at the OASIS facility for delayed-proton precursors, ranging from ^{119}Ba to ^{154}Lu, are described and discussed.

B. Single-particle States Near N = 82

The study of low-lying states in nuclei near closed shells is of interest because these levels are relatively pure shell-model states and they provide an opportunity for comparison with calculations based on various models. Nuclei near N = 82 and Z = 64 are particularly interesting because they should provide us with information about the underlying spherical proton structure in isotopes located halfway between the major proton shells of 50 and 82. Much of our experimental effort has been focused on obtaining structure information for nuclei in this mass region. For example, in Ref. 8 we presented experimental systematics of proton states in odd-Z N = 82 nuclei and of neutron states in even-Z N = 81 nuclei, together with results of spherical Hartree-Fock-Bogoliubov calculations. Here we discuss briefly some information that we have garnered from the β decays of ^{147}Tm, ^{145}Dy, ^{147}Er, and ^{153}Lu.

The $h_{11/2}$ ground state of ^{147}Tm has been known[9] for several years to be a direct proton emitter. Cross-section measurements and barrier-penetration calculations indicate that this level should decay mainly by β decay. In 1987 we reported[10] the identification of this β-decay branch through the observation of Er K x rays whose intensity decreased in a manner consistent with the ^{147}Tm half-life of ~ 0.5 s. Further analysis of the data has revealed, in addition to the Er K x rays, an 80.9-keV γ ray that follows the β decay of ^{147}Tm. This transition, together with the Er $K\alpha_1$ peak, can be seen in Fig. 4(a) where γ rays in coincidence with annihilation radiation are displayed. The spectrum was measured during 1.28-s counting cycles designed to emphasize the production of ^{147}Tm *vis-a-vis* isobars with longer half-lives. Indeed the 80.9-keV γ ray is scarcely visible in Fig. 4(b) where data, once again in coincidence with annihilation radiation but accumulated during 4-s cycles, are displayed.

We believe that the 80.9-keV γ ray is the transition that proceeds from the first-excited, $d_{3/2}$ neutron-hole level in ^{147}Er to the $s_{1/2}$ ground state. Our assignment is based on energy systematics of $s_{1/2}$, $d_{3/2}$, and $h_{11/2}$ neutron-hole states in this mass region that were recently updated[11] following our study of the β decay of ^{145}Ho ($h_{11/2}$), the N = 79 isotone just below ^{147}Er. Figure 5 shows these systematics. One notes that the 80.9-keV energy for the $d_{3/2}$ level in ^{147}Er fits well into the overall picture, i.e., this level is at 1.58 keV in ^{141}Sm, and then increases in excitation energy to 45.1 and 66.3 keV in ^{143}Gd and

^{145}Dy, respectively. A discussion of these single-neutron level systematics is presented in Ref. 11.

The radionuclides ^{145}Dy and ^{147}Er are known[12] β-delayed-proton precursors. However, little information is available concerning levels in their β-decay daughters, ^{145}Tb and ^{147}Ho. No γ rays have been assigned to ^{147}Er decay while a total of only four transitions have been observed[13] to follow ^{145}Dy decay. We have investigated the β-decay of the $s_{1/2}$ ground and $h_{11/2}$ isomeric states in ^{145}Dy and ^{147}Er. Here we report on the identification of low-lying single-proton levels in ^{145}Tb and ^{147}Ho based on the observation of three γ rays in cascade, characteristic of odd-Z even-N nuclei in this region, that connect and deexcite the $g_{7/2} \rightarrow d_{5/2} \rightarrow d_{3/2} \rightarrow s_{1/2}$ states.

Figures 6(a) and 6(b) show γ-ray spectra in coincidence with the $g_{7/2} \rightarrow d_{5/2}$ transitions in ^{145}Tb (184.5 keV) and ^{147}Ho (616.7 keV), respectively. The energies of the $d_{5/2} \rightarrow d_{3/2}$ and $d_{3/2} \rightarrow s_{1/2}$ transitions are 145.1 and 108.1 keV in ^{145}Tb [Fig. 6(a)] and 292.0 and 96.1 keV in ^{147}Ho [Fig. 6(b)]. There is also a crossover, 253.1-keV, transition, $d_{5/2} \rightarrow s_{1/2}$, in ^{145}Tb [see Fig. 6(a)]. We show updated single-proton level systematics for N = 80, N = 82, and N = 84 Tb, Ho, and Tm nuclei in Fig. 7. One sees that the new states in ^{145}Tb and ^{147}Ho follow the trend established by the previously available information, i.e., the gap between the $d_{5/2}$ and the $d_{3/2}$ orbitals increases as the atomic number increases. Also, we indicate the $s_{1/2}$ orbital as the ground state in Tb nuclei; the $h_{11/2}$ state, which is isomeric for $Z < 65$, becomes the ground state for the Ho and Tm isotopes (see Ref. 14).

Part of our investigation included a search for the β decay of ^{153}Lu which should populate single-neutron states in the N = 83 nucleus ^{153}Yb. A low production yield was anticipated for the ^{92}Mo(^{64}Zn,p2n) channel and indeed there was no obvious indication of Yb K x rays. Observation of their presence was severely hampered by the much more intense Kα and Kβ x rays of elements with $Z \leqslant 69$. However, ^{153}Yb levels have recently been studied[15] via in-beam γ-ray spectroscopy. In addition to high-lying, high-spin states the authors[15] located the $h_{9/2}$ and $i_{13/2}$ neutron levels at 567 and 1202 keV, respectively. The $h_{9/2}$ state should be the most intensely populated by the decay of the $11/2^-$ ($h_{11/2}$ proton orbital) ^{153}Lu ground state. An examination of our data showed the presence of a weak 566.5-keV γ ray. Gating on the 566.5-keV γ ray revealed Yb K x rays in coincidence. Based on this evidence we conclude that we have identified the β decay of ^{153}Lu.

In addition to the 566.5-keV transition we assign two other γ rays to the decay of ^{153}Lu. All three can be seen in Fig. 8 where we show the spectrum observed in coincidence with photons in the energy range of Yb K x rays. In Fig. 9 we have plotted the energies of $h_{9/2}$ and $i_{13/2}$ neutron states in even-Z nuclei above europium with neutron numbers of 83, 85, and 87. The 566.5- and 1202.0-keV ^{153}Yb levels fit into these systematics. It has been noted[16] that the $h_{9/2}$, $p_{1/2}$, $p_{3/2}$, and $f_{5/2}$ orbitals increase in excitation energy with respect to the $f_{7/2}$ ground states as Z increases up to 64 (gadolinium) and then decrease as Z increases further (see Fig. 9). Also, we note in Fig. 9 a compression of the $h_{9/2}$ level energies as the neutron number becomes larger. The $i_{13/2}$ orbital behaves[16] differently; its energy decreases with increasing Z, reaches a minimum at $Z = 64$, and then increases (Fig. 9). Its behavior with N also differs from that of the $h_{9/2}$ orbital. The evidence is that the $13/2^+$ levels are not pure single-particle states; they are mixed with 3^- octupole core states.

C. 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[14] 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[17] in ^{149}Ho. Herein we review briefly our study of the ^{155}Lu and ^{157}Lu α decays.

Figure 10 shows the α spectrum that we recorded at $A = 155$ during collection and assay cycles that were 1.28 s in duration. 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[18] since 1965. Recently, Hofmann et al.[19] observed α decay (E_α = 5.575 ± 0.010 MeV) from a second low-lying level in ^{155}Lu. Our data shown in Fig. 10 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 in Ref. 19; we measure a $T_{1/2}$ of 140 ± 20 ms. Because of the low cross section for the ^{94}Mo(^{64}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.

Figure 11 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[20] ^{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[20] for the ground state. Information on the β decay properties of the two ^{157}Lu levels has hitherto not been available.[20] 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.

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Table I. Isotopes studied.

Isotope	J^{π}	$T_{1/2}$(s)	Delayed protons
^{145}Er[a]		0.9(3)	Yes[b]
^{145}Ho[a]	$11/2^{-}$	2.4(1)	
^{145}Dy	$11/2^{-}$	15(2)	Yes
^{145}Dy[c]	$1/2^{+}$	8(2)	Yes[b]
^{146}Ho		3.6(3)	Yes[b]
^{146}Dy	0^{+}	29(3)	
^{146}Tb	1^{+}	~8	
^{147}Tm[d]	$11/2^{-}$	~0.5	
^{147}Er	$11/2^{-}$	2.5(2)	Yes
^{147}Er[c]	$1/2^{+}$		Yes[b]
^{147}Ho	$11/2^{-}$	5.8(2)	
^{147}Ho[c]	$(1/2^{+})$		
^{148}Er	0^{+}	4.4(2)	Yes[b]
^{148}Ho	6^{-}	9.7(3)	Yes[b]
^{148}Ho	1^{+}	2.2(11)	Yes[b]
^{149}Tm[a]	$(11/2^{-})$	0.9(2)	Yes[b]
^{149}Er	$11/2^{-}$	9(1)	Yes
^{149}Er	$1/2^{+}$		Yes
^{149}Ho	$11/2^{-}$	21(1)	
^{149}Ho	$1/2^{+}$	54(5)[e]	
^{150}Tm	(6^{-})	2.2(2)[e]	Yes[b]
^{150}Tm	(1^{+})		Yes[b]
^{150}Er	0^{+}	20(1)	
^{151}Yb[c]	$11/2^{-}$	1.6(1)	Yes[b]
^{151}Yb[a]	$1/2^{+}$		Yes[b]
^{151}Tm	$11/2^{-}$	4.3(2)	
^{151}Tm[c]	$1/2^{+}$	~8	
^{152}Lu[a]	(6^{-})	0.7(1)	Yes[b]
^{152}Yb	0^{+}	3.1(2)	
^{152}Tm	(2^{-})		
^{153}Lu	$11/2^{-}$	0.9(2)	
^{153}Yb	$7/2^{-}$	3.9(1)	Yes[b]
^{153}Tm	$11/2^{-}$	1.7(2)	
^{153}Tm	$1/2^{+}$	(2.5)	
^{154}Lu	(7^{+})	1.2(1)	Yes[b]
^{154}Yb[d]	0^{+}	~0.4	
^{155}Lu	$11/2^{-}$	0.066(7)	
^{155}Lu	$(1/2^{+},3/2^{+})$	0.14(2)	
^{155}Yb[d]	$7/2^{-}$	1.7(1)	
^{155}Tm	$11/2^{-}$	21.6(2)	
^{155}Tm[c]	$1/2^{+}$	44(4)	
^{157}Lu[d]	$11/2^{-}$	~5	
^{157}Lu[c,d]	$(1/2^{+},3/2^{+})$	~6	

[a] Isotope discovered in this study.

[b] Delayed-proton emission observed for the first time.

[c] Isomer discovered in this study.

[d] Beta-decay branch of nuclide identified for the first time.

[e] New half-life measured in this investigation.

FIGURE CAPTIONS

Fig. 1. Portion of nuclidic chart where isotopes investigated in this experimental program are indicated by shaded squares.

Fig. 2. Detector arrangement used in nuclear spectroscopic studies at the OASIS on-line facility.

Fig. 3. Beta-delayed proton spectra of $N = 81$ precursors.

Fig. 4. Gamma-ray spectra observed in coincidence with annihilation radiation for $A = 147$ nuclides during 1.28-s [part (a)] and 4-s [part (b)] counting cycles. The 81-keV γ ray in part (a) is assigned to ^{147}Tm β decay. Transitions assigned to ^{147}Dy, ^{147}Ho, and ^{147}Er are labeled by their elemental symbols.

Fig. 5. Systematics of $s_{1/2}$, $d_{3/2}$, and $h_{11/2}$ neutron-hole states in Sm, Gd, Dy, and Er nuclei with $N = 77$, 79, and 81.

Fig. 6. Gamma-ray spectra observed in coincidence with the $g_{7/2} \rightarrow d_{5/2}$ transitions in ^{145}Tb and ^{147}Ho following the β decays of ^{145}Dy [part (a)] and ^{147}Er [part (b)], respectively.

Fig. 7. Systematics of $s_{1/2}$, $d_{3/2}$, $d_{5/2}$, and $g_{7/2}$ proton states in Tb, Ho, and Tm nuclei with $N = 80$, 82, and 84.

Fig. 8. Gamma-ray spectrum measured at $A = 153$ in coincidence with photons in the energy range of Yb, $K\alpha_1$ x rays.

Fig. 9. Systematics of $h_{9/2}$ and $i_{13/2}$ neutron states in Gd, Dy, Er, and Yb nuclei with $N = 83$, 85, and 87.

Fig. 10. Alpha-particle spectrum measured at $A = 155$.

Fig. 11. Alpha-particle spectrum measured at $A = 157$. The 4.924-MeV α group is assigned to the decay of the previously unobserved low-spin ($1/2$, $3/2^+$) isomer in ^{157}Lu.

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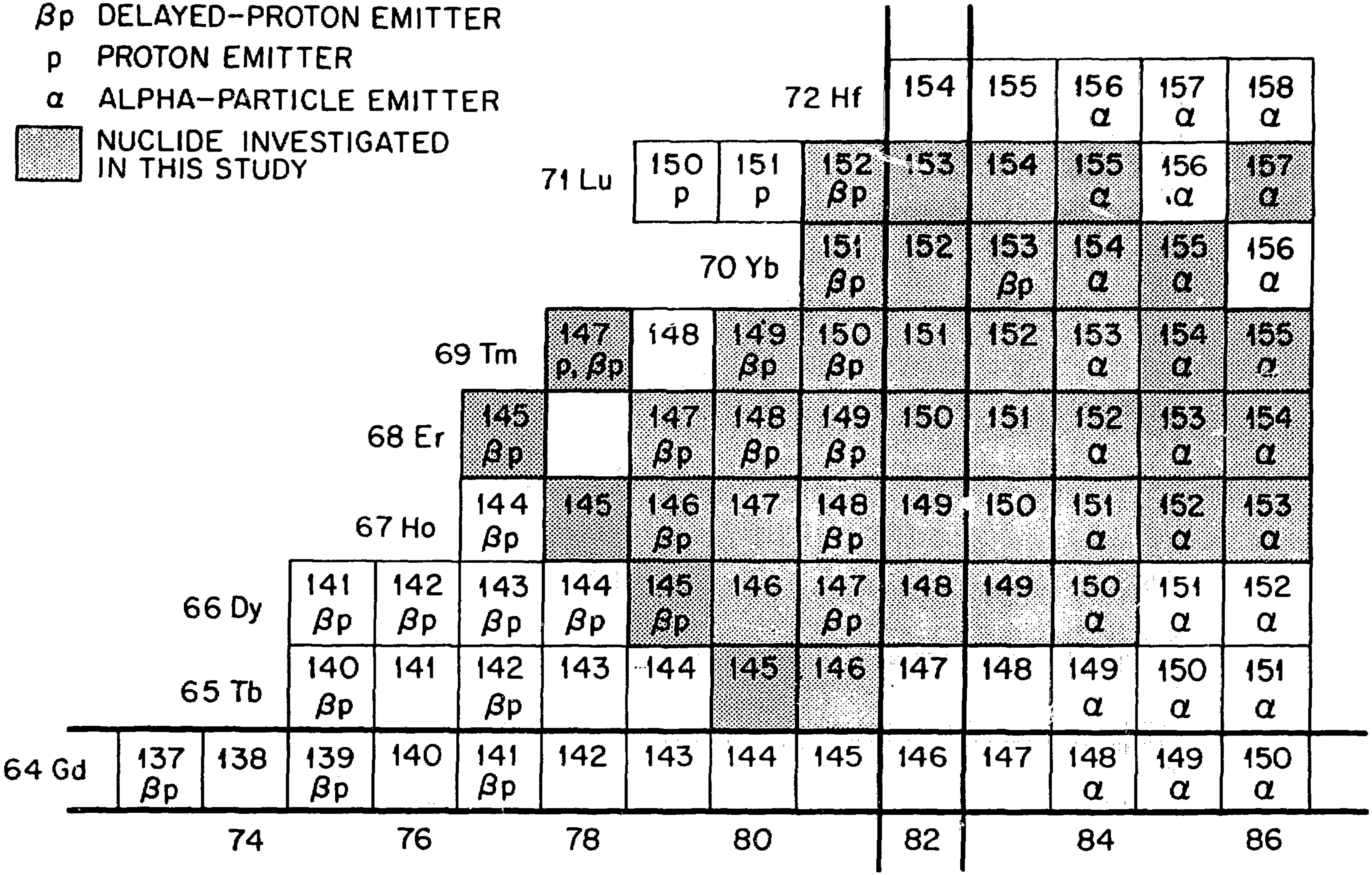
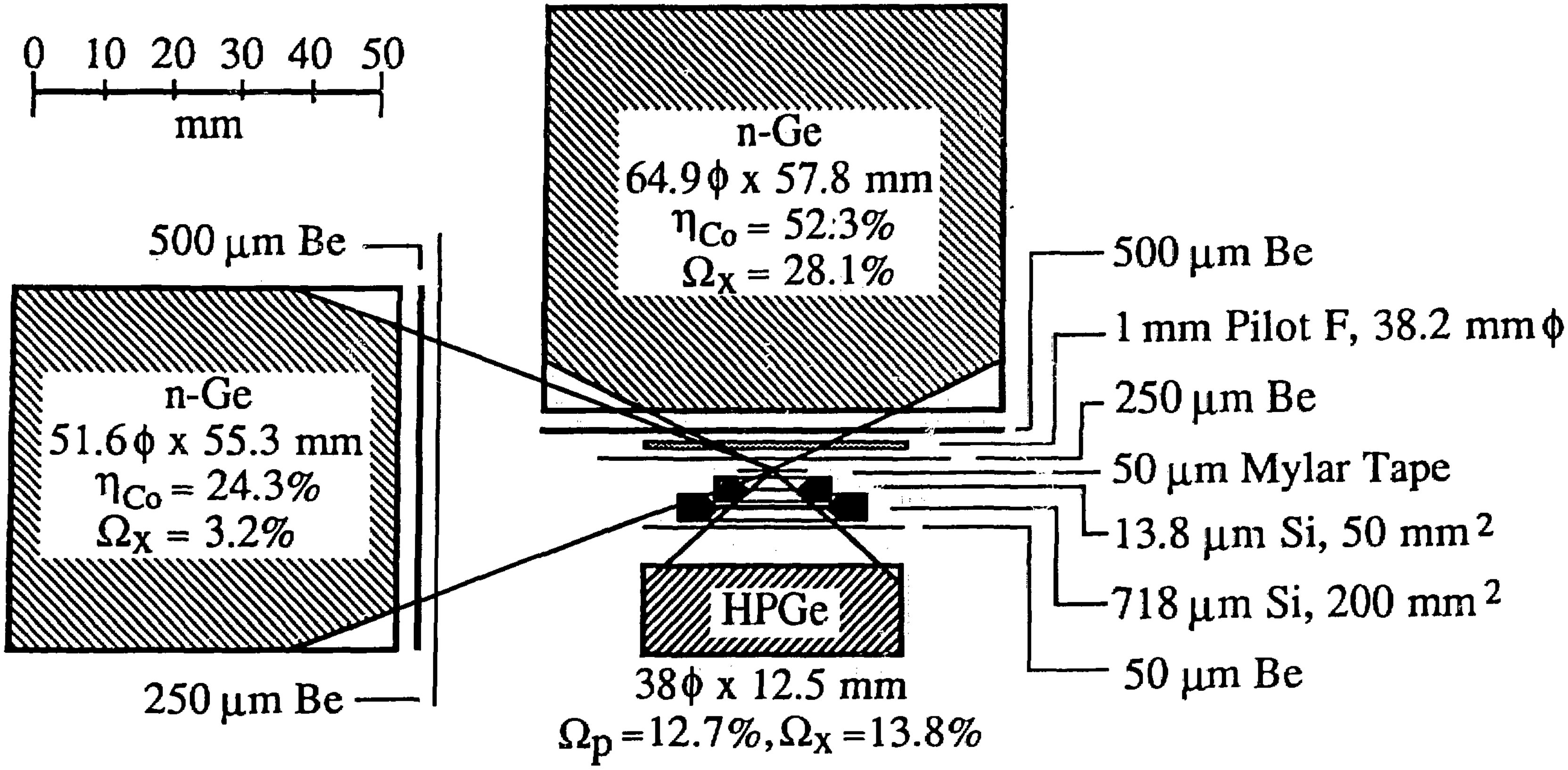


βp DELAYED-PROTON EMITTER
p PROTON EMITTER
α ALPHA-PARTICLE EMITTER
▨ NUCLIDE INVESTIGATED IN THIS STUDY

72 Hf										154	155	156 α	157 α	158 α
71 Lu							150 p	151 p	▨ 152 βp	▨ 153	▨ 154	▨ 155 α	156 α	▨ 157 α
70 Yb									▨ 151 βp	▨ 152	▨ 153 βp	▨ 154 α	▨ 155 α	156 α
69 Tm						▨ 147 p, βp	148	▨ 149 βp	▨ 150 βp	▨ 151	▨ 152	▨ 153 α	▨ 154 α	▨ 155 α
68 Er					▨ 145 βp		▨ 147 βp	▨ 148 βp	▨ 149 βp	▨ 150	▨ 151	▨ 152 α	▨ 153 α	▨ 154 α
67 Ho					144 βp	▨ 145	▨ 146 βp	▨ 147	▨ 148 βp	▨ 149	▨ 150	▨ 151 α	▨ 152 α	▨ 153 α
66 Dy			141 βp	142 βp	143 βp	144 βp	▨ 145 βp	▨ 146	▨ 147 βp	▨ 148	▨ 149	▨ 150 α	151 α	152 α
65 Tb			140 βp	141	142 βp	143	144	▨ 145	▨ 146	147	148	149 α	150 α	151 α
64 Gd	137 βp	138	139 βp	140	141 βp	142	143	144	145	146	147	148 α	149 α	150 α
		74		76		78		80		82		84		86

Fig. 1



0 10 20 30 40 50
mm
n-Ge
64.9φ x 57.8 mm
η_{Co} = 52.3%
Ω_X = 28.1%
500 μm Be
n-Ge
51.6φ x 55.3 mm
η_{Co} = 24.3%
Ω_X = 3.2%
250 μm Be
500 μm Be
1 mm Pilot F, 38.2 mm φ
250 μm Be
50 μm Mylar Tape
13.8 μm Si, 50 mm²
718 μm Si, 200 mm²
50 μm Be
HPGe
38φ x 12.5 mm
Ω_p = 12.7%, Ω_X = 13.8%

Fig. 2

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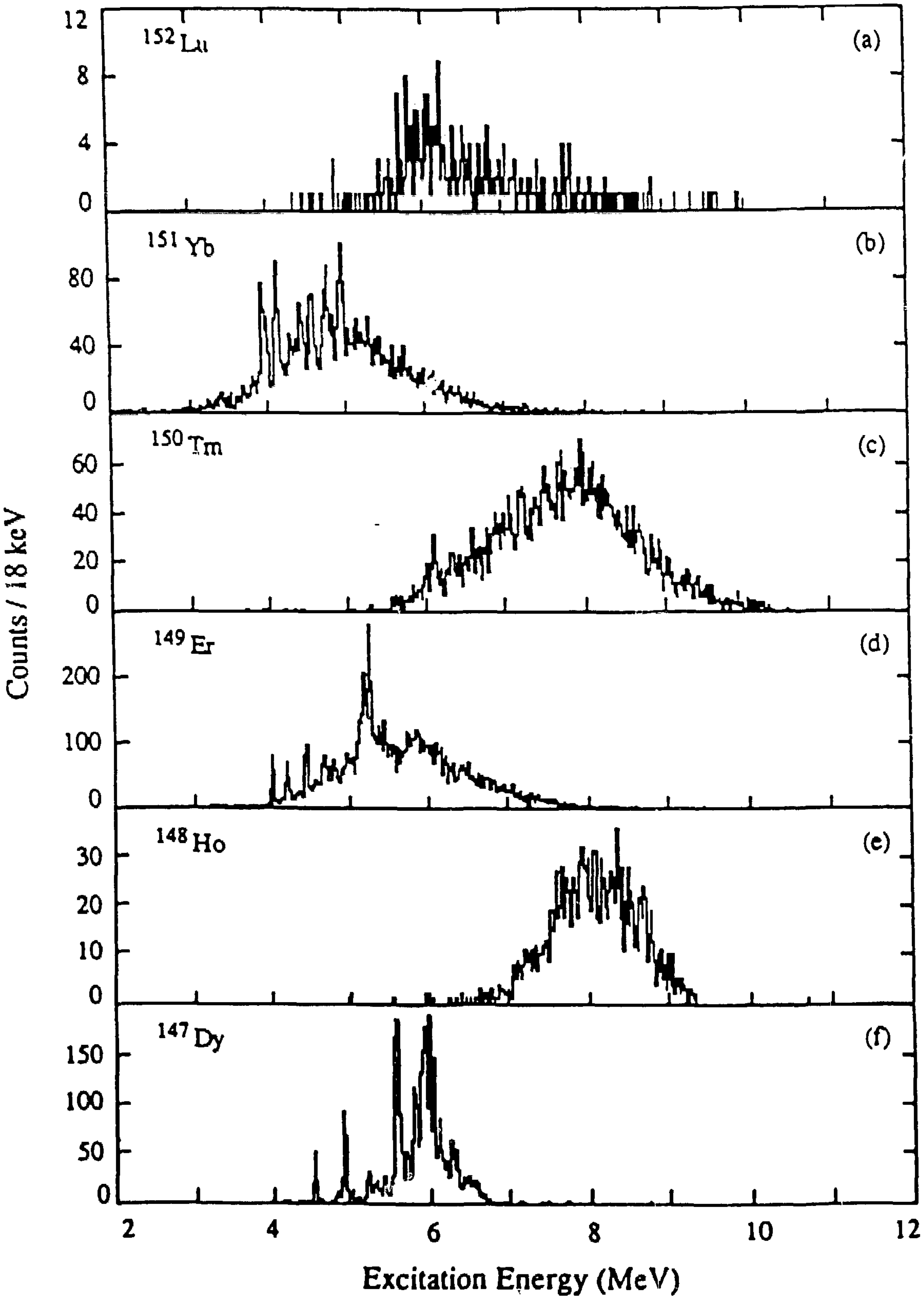

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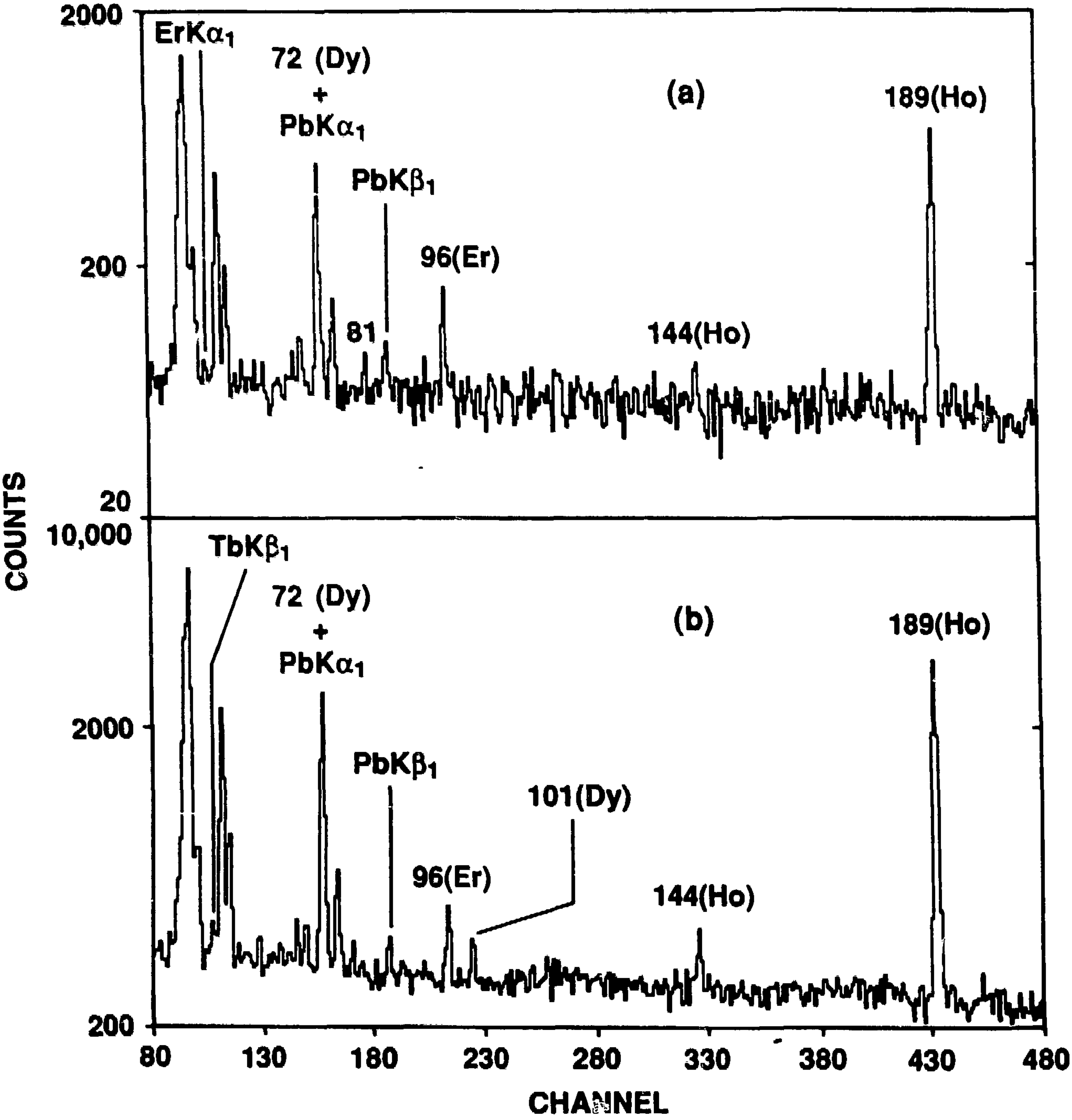

Fig. 4

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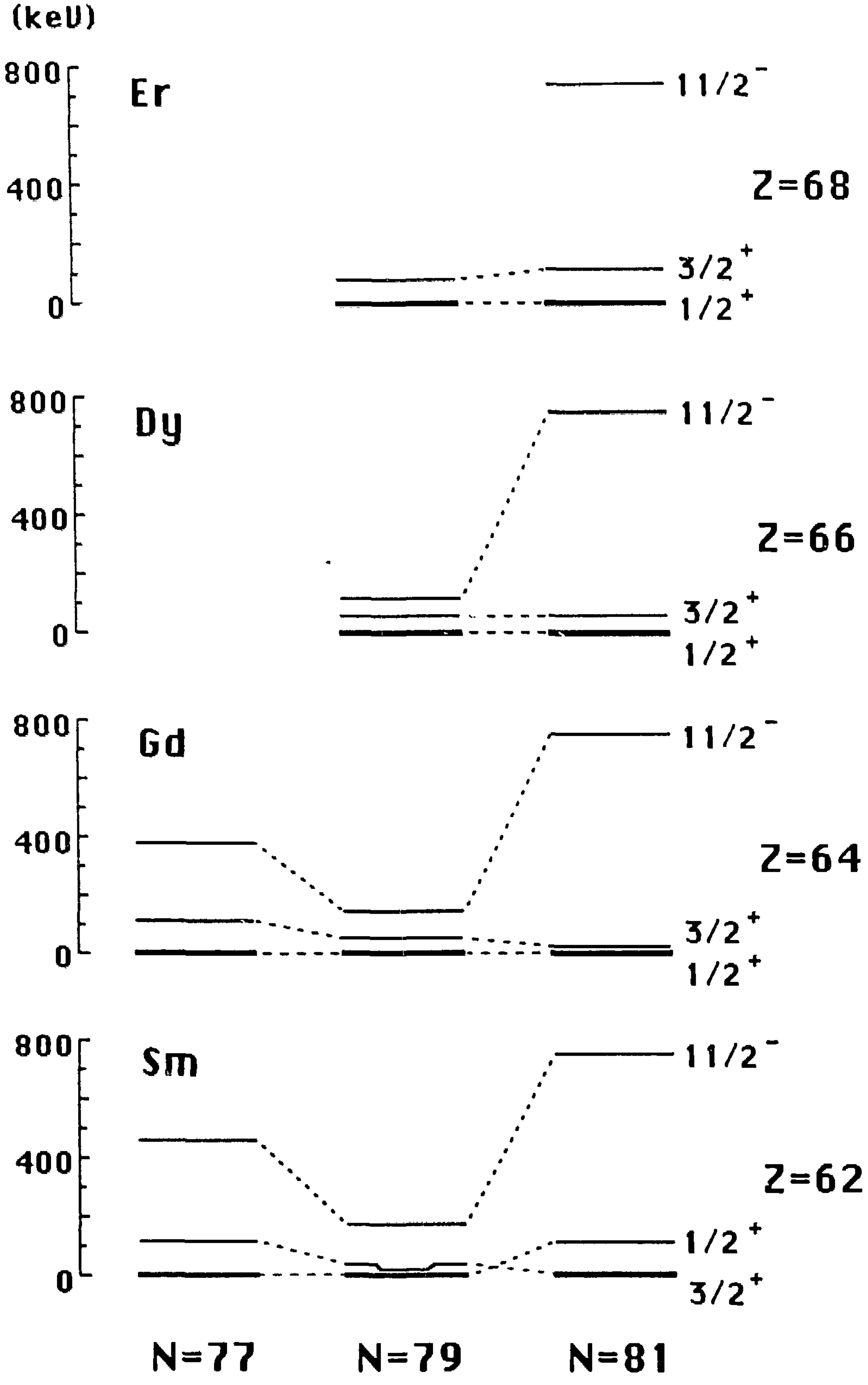

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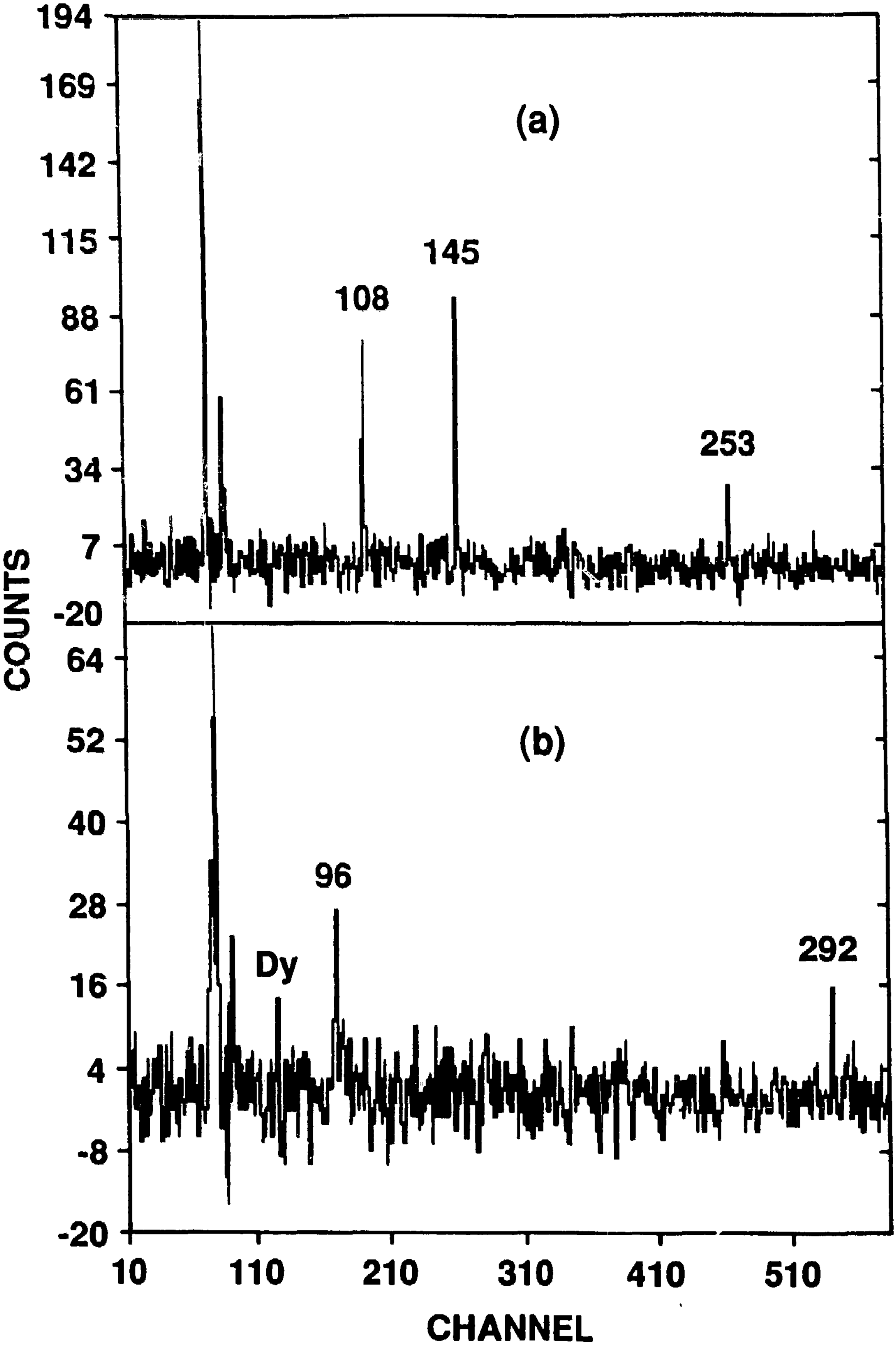

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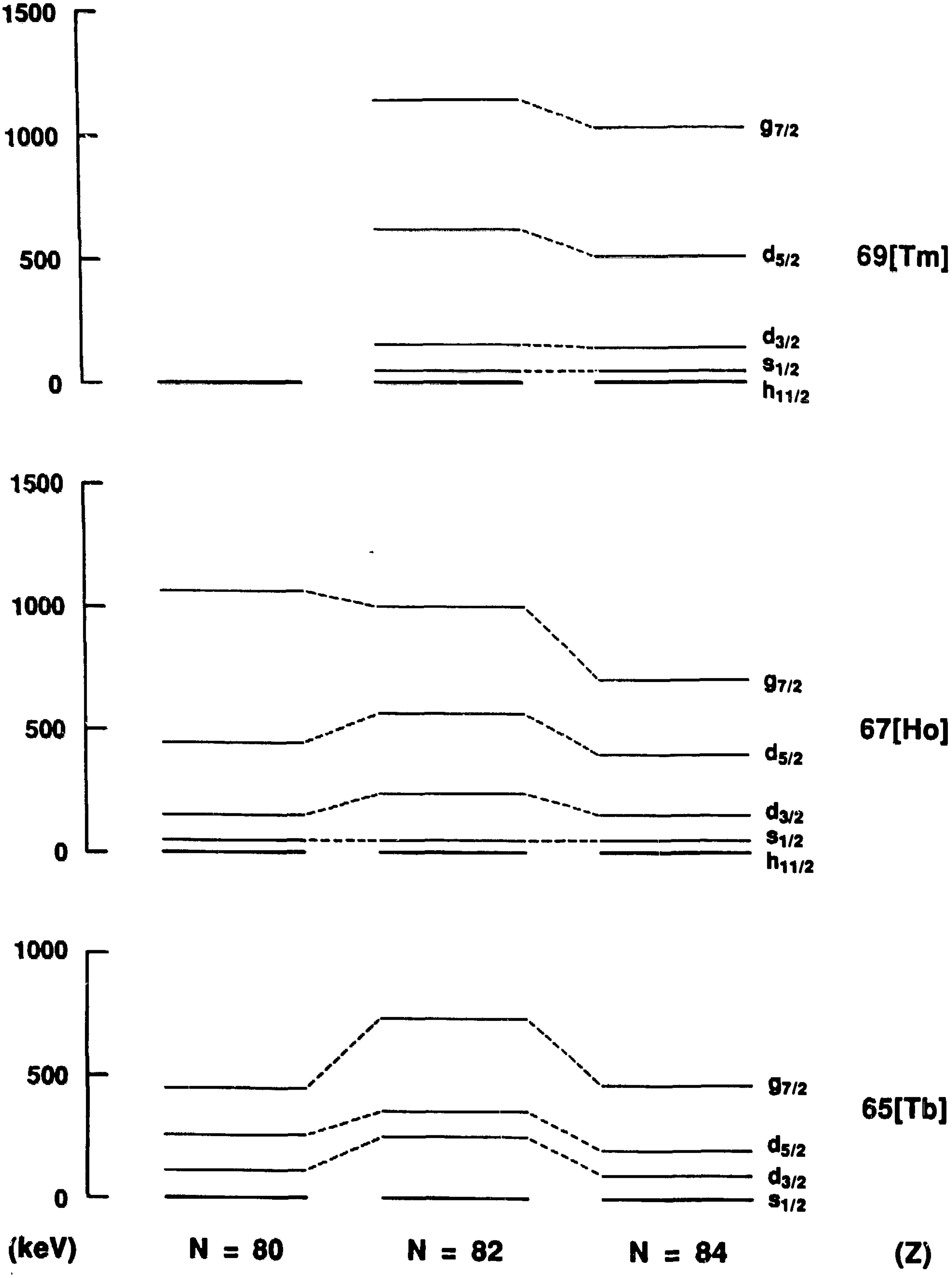
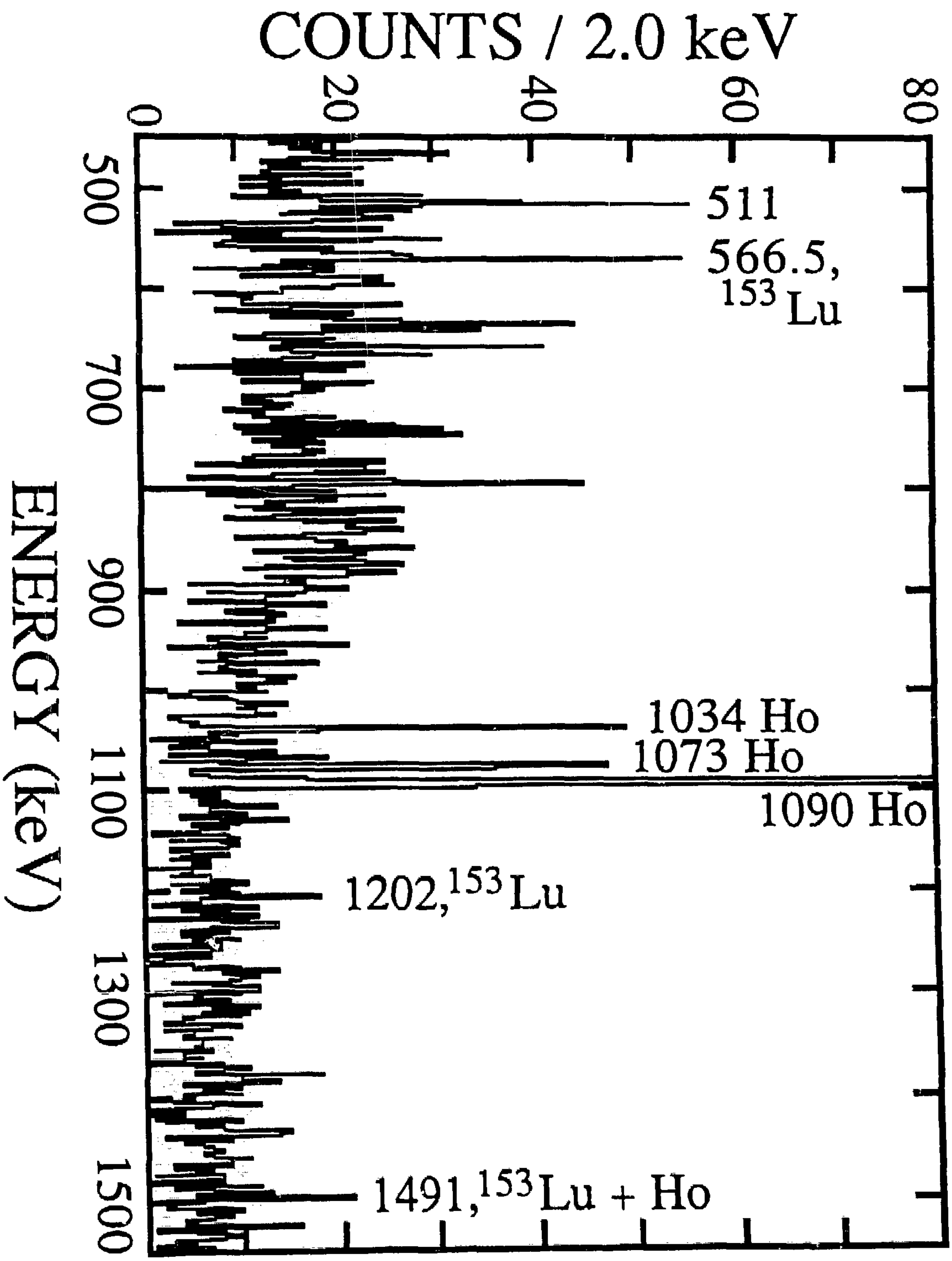


Fig. 7



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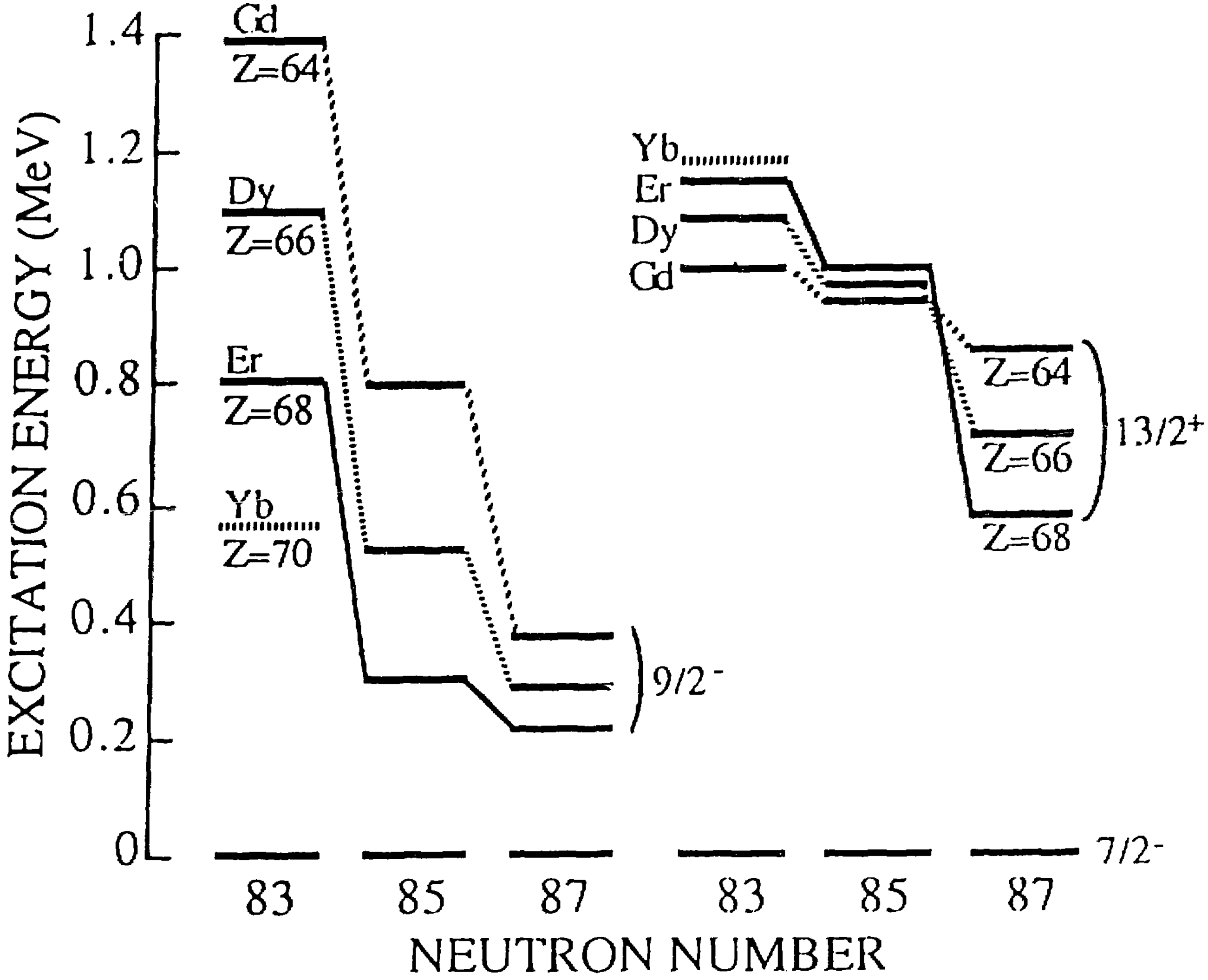
EXCITATION ENERGY (MeV)
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0
Gd
Z=64
Dy
Z=66
Er
Z=68
Yb
Z=70
9/2⁻
Yb
Er
Dy
Gd
Z=64
Z=66
Z=68
13/2⁺
7/2⁻
83
85
87
83
85
87
NEUTRON NUMBER

Fig. 9

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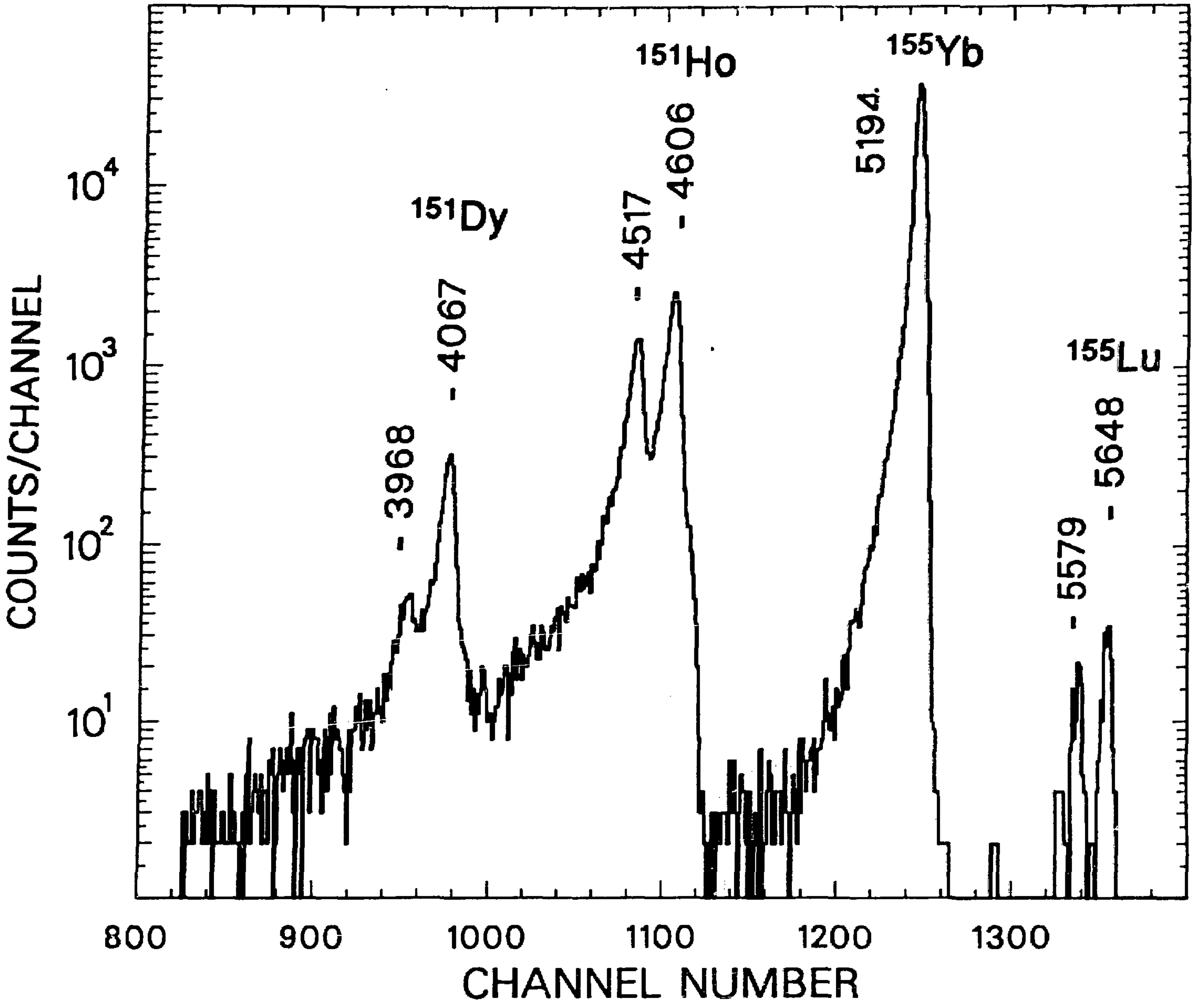

Fig. 10

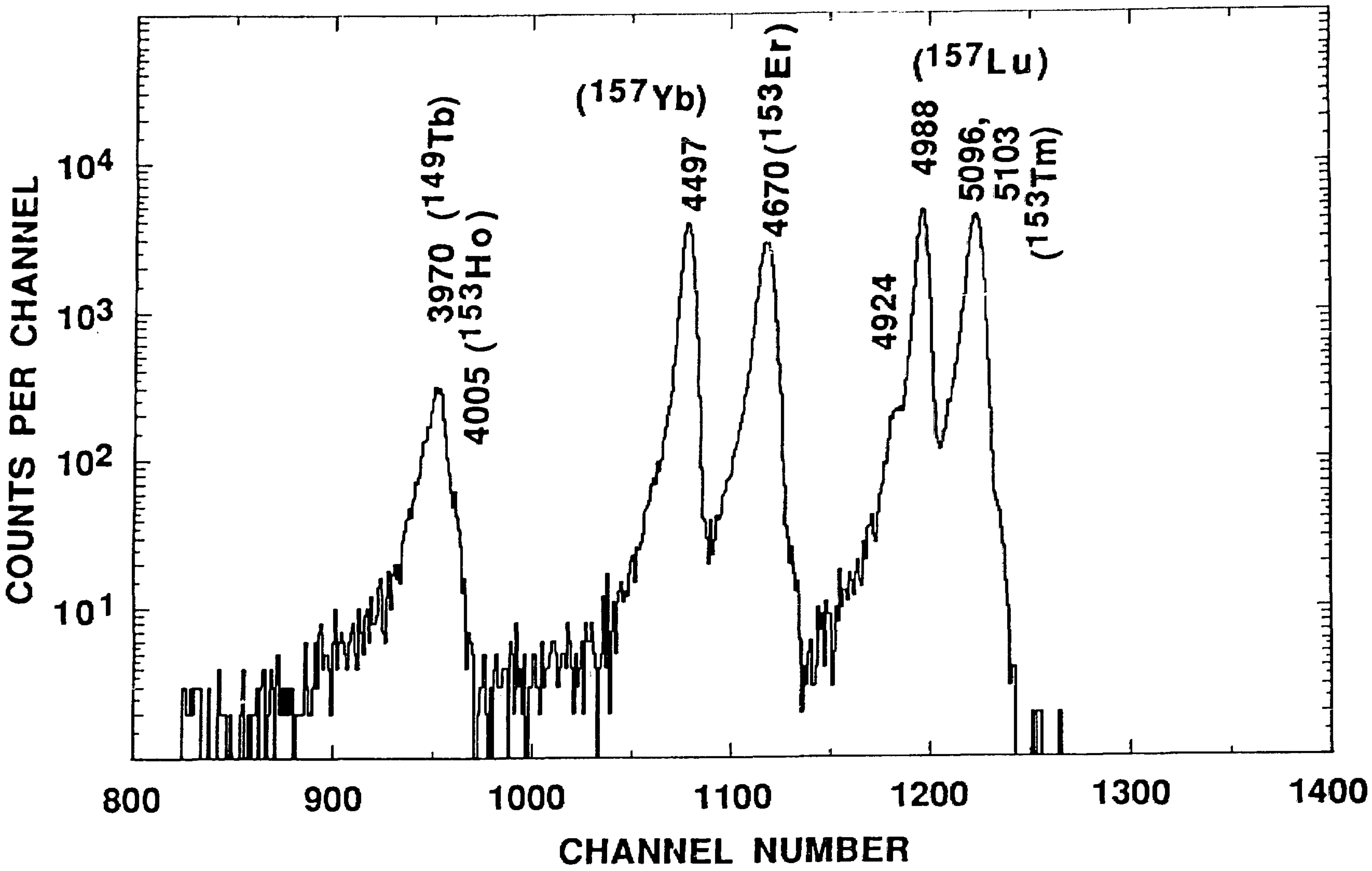
COUNTS PER CHANNEL
10^4
10^3
10^2
10^1
CHANNEL NUMBER
800
900
1000
1100
1200
1300
1400
3970 (149Tb)
4005 (153Ho)
(157Yb)
4497
4670 (153Er)
4924
(157Lu)
4988
5096,
5103
(153Tm)

Fig. 11