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Abstract:

This paper presents a study of the γ rays from the electron capture decay of 3.63 d ^{100}Pd. Singles and coincidence measurements provided an accurate determination of γ-ray intensities and an improved knowledge of the ^{100}Rh level scheme. Experimental evidence supports excitation of levels in ^{100}Rh at 86-, 136- and 154 keV, not previously reported from the decay of ^{100}Pd.

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Introduction

A recent review[1] of $A = 100$ nuclides shows that the γ-ray data from the electron capture decay of ^{100}Pd available in the literature[2-8] are not definitive. Not only the very existence of some of the reported γ rays is uncertain, but also the intensities of even the strong γ-ray transitions differ considerably from one study to another. From the nuclear structure point-of-view, information on the levels of odd-odd ^{100}Rh could provide some insight into the nature of the coupling between unpaired protons and neutrons for the $A = 80 - 100$ mass region. The present study was undertaken to provide better intensities for the γ-ray transitions among the low-lying low-spin levels of ^{100}Rh populated in the decay of ^{100}Pd. Low-spin levels in ^{100}Rh have also been investigated in one $(p, n\gamma)$ reaction study [9].

Aside from its interest to nuclear physicists, ^{100}Pd has potential applications in nuclear medicine. Possible uses of Pd radioisotopes have been mentioned by Lagunas-Solar et al[10] and DeNardo et al[11] in connection with radioimmuno therapy. The ^{100}Pd isotope has been considered for this specific purpose [11] for its desirable half life and appropriate γ-ray energies and intensities. Interestingly enough, the photon emission probabilities presented in reference [11] are significantly different from those determined in the present work; the consequences for absorbed dose estimates are obvious. The γ-ray data presented in this paper should help determine the usefulness of ^{100}Pd for therapeutic purposes. The isotope is easily produced by irradiating natural rhodium metal with a proton beam of moderate energy.

Experimental Arrangements

For the present experiments, metallic foils of > 99% pure rhodium were irradiated with a 45 MeV proton beam produced by the 88-inch cyclotron at the

Lawrence Berkeley Laboratory. A typical irradiation time was 2 h at a beam current of 1μA. The ^{100}Pd is produced by the reaction ^{103}Rh (p, 4n) ^{100}Pd. After irradiation the target was allowed to "cool" for at least 24 hours.

In order to remove contaminant Ru and Rh activities, which are formed through competing reactions, the following chemical procedures were followed. The target foil was removed from the target block after irradiation and the active part cut into small pieces. The pieces were placed on a 0.5 cm deep bed of Bi metal in a pyrex tube. The Rh pieces were covered with an additional 1 cm layer of Bi metal and the mix was heated with a propane torch until it liquefied. The alloy was allowed to "cool" and was then dissolved in 50 ml of concentrated HNO_3. The resulting solution was transferred to a 250 ml beaker and concentrated NaOH was added drop wise until Bi hydrolysis products were observed to precipitate. The mixture was then cooled to room temperature and concentrated HNO_3 added until all the precipitate was dissolved. An 80 ml aliquot of the solution (about 150 ml in total) was placed in a separatory funnel. The Pd fraction was extracted with dimethylglyoxime in chloroform (about 100 ml). The organic phase was washed four times with 0.1 N HNO_3 to yield a Pd fraction of high purity. Aliquots of the organic phase of 5 ml volume were then evaporated to dryness on 0.25 mm thick, 2.5 cm square Al foils to form sources for γ-ray spectroscopy. To immobilize the active deposit, the source foils were doubly coated with acrylic spray.

Singles γ-ray spectra were obtained at both Berkeley and Toronto using 25%-efficient gamma-X intrinsic Ge detectors. For these measurements the source-to-detector was 30 cm. The γ-ray summing was observed to be negligble. Coincidence studies were performed at Toronto using a conventional multi-parameter coincidence spectrometer. One detector was a 25% efficient gamma-X type, the other was a 10% efficient coaxial intrinsic Ge detector. The detectors were aligned at 90° to one another; a 6 mm thick Fe anti–scattering absorber was placed between the active volumes of the two detectors. A conventional coincidence system, with a time resolution of about 100 ns, was used. A total of 15×10^6 coincidence events were stored on magnetic tape for off–line sorting; about 10% of them were accidentals. Gates were set on every γ–ray peak of interest to produce the coincidence spectra. With the system described, the photon emission probability detection threshold was found to be about 0.03% for both singles and coincidence experiments.

Experimental Results

A typical γ–ray singles spectrum is presented in Fig. 1. The energy region below 30 keV was found to contain only Rh and Ru L– and K– X ray lines. Above 160 keV, we observed γ–ray lines produced by the decay of ^{100}Rh. No impurity lines were observed in the energy interval of 30–182 keV of Fig. 1.

The efficiency curve for the Berkeley detector was determined for the energy range 14–1500 keV using standard sources of ^{57}Co, ^{109}Cd, ^{133}Ba, ^{137}Cs, ^{152}Eu, ^{203}Hg and ^{241}Am. The overall uncertainty in the efficiency curve is about 6%. The energies and relative intensities of the γ–ray transitions were obtained with the computer code, SAMPO, and are given in Table 1, together with the results from previous studies [3–6]. Our results agree best with those of Kanbe et al [3] (unpublished report), except for a γ ray they reported at 179.84 keV ($I_\gamma = 0.71$) which was not observed in our spectra. We believe that this γ ray arises from the decay of 4.34 d ^{101m}Rh which was present to a minor degree in our source material, as revealed by the low intensity of the dominant 307 keV γ ray. (The scarcity of such radioimpurities was assured by the "cooling" time and the radiochemical procedures.) There is also no evidence for previously reported γ rays at 51.7 keV [6] and 145.7 keV [7]. A particularly interesting situation arises in connection with a 55.82- keV γ ray which has been reported previously [4,5,7] with intensities of < 1.5 [4] and < 8 [5] . We observe no such γ ray but instead we see in both singles and coincidence spectra, a weak line due to a γ ray of energy 53.52 keV ($I_\gamma = 0.08$).

In Fig. 2 γ–γ coincidence evidence is presented for some of the weak γ rays arising from the decay of ^{100}Pd. Their energies are 53.52, 72.52 and 86.37 keV, defining levels in ^{100}Rh at 86 and 136 keV, which were reported also in a reaction study by Bizetti–Sona et al [9] . The peaks at 74.78 and 84.00 keV are due to random coincidences as indicated by a comparison with several spectra observed in coincidence with gates placed on different regions of the continuum. The γ rays with energies 139.92, 151.88, 154.00 and 158.87 keV were found to be in coincidence with Rh K–X rays and with none of the γ rays. This result establishes a new level at 154 keV in ^{100}Rh and confirms those at 140 and 152 keV suggested earlier [1] .

Figure 3 shows the decay scheme of ^{100}Pd based on the present results. It differs in several respects from that presented by Evans and Naumann [5] . There is no evidence now for the level at 214 keV; no 55.82 keV γ ray was observed by us and the 139.92–keV γ ray was observed in coincidence with Rh K–X rays only. Levels at 140, 152 and 154 keV are populated directly by electron capture. The levels at 86 and 136 keV had been established through the reaction ^{100}Ru (p, nγ) ^{100}Rh [9] , but were not previously observed from the decay of ^{100}Pd. The electron capture intensities and log ft values shown in the decay scheme were deduced from the transition intensity balance at each ^{100}Rh level. The emission probability of the 84.0- keV γ ray was obtained from the ratio of its intensity to that of the 539.6–keV γ ray from 20.8- h ^{100}Rh. The observed ratio, $I_\gamma(84.0\gamma)/I_\gamma(539.6\gamma)$ vs. time, was least squares fitted over a period of 11 d with 4 h counting intervals using the equations for two–component radioactive decay. Using the emission probability of the 539.6–keV γ ray from ^{100}Rh, 78.4(17)%[1], we deduce from the fit, $I_{84\gamma}$(emission prob.) = 53 (5)%. A second determination, made after the two activities had come into equilibrium and were decaying with the 3.63 d half life of ^{100}Pd; produced $I_{84\gamma}$(emission prob.) = 52 (4)% . We adopt, 53 (4)%, which compares well to 61 (7)% measured in reference 4. Our adopted value gives an electron capture branching of 5 ± 5% to the ground state and a log ft > 5.9 consistent with that expected for a first forbidden transition. Our measured Rh K X–ray intensities are: $I(K_\alpha)$ = 206 (13) and $I(K_\beta)$ = 40 (3). These agree well with values of 214 (8) and 40 (2), respectively, deduced from the proposed decay scheme. Such an agreement confirms both the correctness of the decay scheme and the quality of our measured γ–ray emission probabilities.

DeNardo et al [11] in their discussion of the possible use of ^{100}Pd for radioimmunotherapy quote the following photon emission probabilities for the most intense γ rays: 74.7 keV: 35%, 84.0 keV: 40%, 126.0 keV: 20%, 158.7 keV: 7%. From our measurements we deduce the following photon emission probabilities: 49 (4)%, 53 (4)%, 8 (1)%, 1.7 (2)%, respectively.

The cross section, σ, for the ^{103}Rh(p, 4n)^{100}Pd reaction at E_p = 45 MeV was measured as ~ 250 mb[10], using I_γ(emission prob.) = 33% for the 126 –keV γ ray. Our value for $I_\gamma(126)$ = 8.0%, (see Table 1) leads to a surprisingly high and inconsistent (with systematics) value of ~ 1 barn for this cross section. Our experiment was not designed to measure σ, hence the cross section measurement needs to be re–evaluated.

Discussion

The J^π valves for the ground state and the levels at 75 and 159 keV shown in figure 3 are taken from the A = 100 review[1]. The parities of the 33– and 152– keV levels are a consequence of the M1 + E2 and E1 nature [4,9] of the respective ground state transitions. Our results show no direct electron capture to the first excited state at 33 keV, whereas earlier data[1] suggested a significant branching to that state. Absence of direct electron capture to this level is consistent with the assignment $J^{\pi} = 2^{-}$; this assignment was suggested previously by the presence of a possible E3 transition from the 106- keV, (5^{+}) isomeric state in ^{100}Rh [1]. From γ-decay patterns, the spin of the 86- keV level is ≤ 3; however, J = 0 and 3 are ruled out from a comparison of the Weisskopf single particle transition rates for the 53- keV and 86- keV γ rays with the recommended upper limits (300 WU for E2 and 1 WU for M2) by Endt [12]. Using $T_{1/2} < 0.35$ ns [9] for the 86- keV level, the Weisskopf estimates exceed 1400 WU for both the 53- keV and 86- keV γ rays (assuming either an E2 or M2 multipolarity as required by J = 0 or 3). The log ft values of figure 3 suggested J = 0, 1 for the 136-, 140-, 152- and 154- keV levels. But using the criterion of recommended upper limits for transition rates[12], $T_{1/2} < 0.35$ ns [9] for the 136- keV level and 0.97 ns [9] for the 152- keV level, the J = 0 choice for the 136- and 152- keV levels is ruled out since it gives Weisskopf estimates in excess of 4000 for an E2 or M2 62- keV transition and an M2 119- keV transition.

There has not been any theoretical calculation reported for $^{100}_{45}Rh_{55}$, nor are any detailed systematics of odd-odd Rh isotopes available. However, the transitions among known levels in the present level scheme are consistent with a general shell model interpretation [5,9] based on the positive parity states (e.g. 75, 159 keV) arising from the 5 valence protons in the $g_{9/2}$ orbit and 5 valence neutrons distributed in $g_{7/2}$, $d_{5/2}$ and $s_{1/2}$ orbits. The negative parity states (e.g. gs, 33 keV) arise from 4 protons in the $g_{9/2}$ orbit and a proton in the $p_{1/2}$ orbit and 5 neutrons in the above positive parity orbitals. This explains the hindrance of the E1 transitions from the 75–keV and 159–keV levels to the g.s. as well as a 33–keV level of negative parity, since $g_{9/2} \rightarrow p_{1/2}$ is involved in such transitions. The 72–keV transition connecting the 159 and 86 levels is very weak which suggests a negative parity for the 86–keV level.

The β transition from 0^+ (^{100}Pd g.s.) to the 1^+ 159–keV level in ^{100}Rh is most likely a $\pi\, g_{9/2} \rightarrow \nu\, g_{7/2}$ Gamow–Teller transition as found in the β–decay of neighbouring even–even nuclides which decay to odd–odd nuclides. In the case of the ^{100}Pd decay, almost all of the Gamow–Teller strength is in a single transition, as predicted by the independent particle model, rather than distributed over several 1^+ states, which is generally the case in neighbouring nuclides. The experimental value of the Gamow–Teller, B(GT), strength (as in [13], [14]) deduced from the log ft value of the present work is 0.14(4). This value does not agree with the theoretical value of 1.6 to 2.8 [14] using $g_A = 1.26$ [13]. The B(GT) experimental value for ^{100}Pd is much lower than the corresponding values for neighbouring nuclides which range from 1.0 to 2.7.

Conclusion

Through the use of high resolution Ge detectors, the present work has produced a more accurate and consistent set of intensities of γ rays from the decay of ^{100}Pd, together with a knowledge of the emission probabilities of strong γ rays. The detailed γ–γ coincidence data have resulted in an extension and revision of the level scheme for ^{100}Rh. One new level at 154 keV has been added to the ^{100}Rh level scheme and the 214–keV level is removed. We believe that the γ–ray information from the decay of ^{100}Pd is now complete to a lower detection threshold of $\sim$ 0.03%.

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Figure Captions

Figure 1: Singles spectrum of γ rays from ^{100}Pd. All energies are in keV. Peaks marked with B are from room background. Peaks marked with S are from coincident summing of 74- and 84-keV γ rays with Rh K-X rays. The letter U indicates location of the unobserved 179.84-keV γ ray reported in reference [3] (see text). Every fourth data point has been plotted in the figure.

Figure 2: Selected portions of coincidence spectra showing the presence of 53-, 72- and 86-keV γ rays. The peaks at 74.78 and 84.00 keV are due primarily to random coincidences (see text).

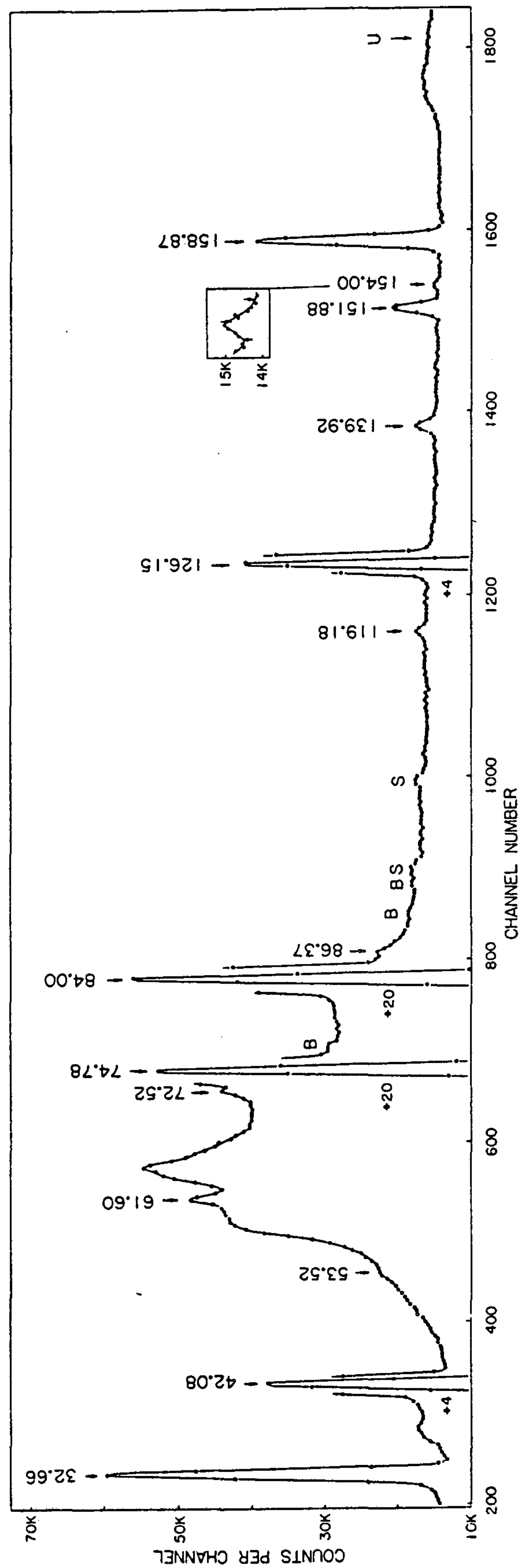
Figure 3: Proposed level scheme of ^{100}Rh from ^{100}Pd ϵ decay. All energies are in keV. Photon intensities are relative to 100 for the 84-keV γ ray. All placements are based on present γ-γ coincidence data. A dot at the beginning of a transition indicates that it was observed in coincidence with another transition. The g.s. transitions from the 139, 152 and 159 keV levels were observed in coincidence with Rh K-X rays only.

Table 1

γ–rays from the electron capture decay of ^{100}Pd

Present work E_γ	I_γ a)	Literature Values I_γ ref. [3]	I_γ ref. [4]	I_γ ref. [5]	I_γ ref. [6]	Multipolarity ref. [4]	Transition intensity d)	Placement
32.66(2)	4.9(3)	8.6(2)	<8	<1.5	1.5(5)	M1+E2 δ=0.15(3)	55(4)	33 → 0
42.08(2)	13.5(8)	17(3)	<10	<1.5	1.5(6)	f)	37(2)	75 → 33
53.52(15) b)	0.08(2)	–	–	–	–		0.2(1)	86 → 33 g)
61.60(5)	0.51(10)	0.38(6)	–	–	–		1.0(4)	136 → 75 g)
72.52(10) b)	0.15(4)	0.38(8)	–	–	–		0.25(10)	159 → 86 g)
74.78(2)	92(5)	81(2)	98(8)	70	45(9)	E1	123(6)	75 → 0
84.00(2)	100(6)	100	100	100	100	M1	158(9)	159 → 75
86.37(15) b)	0.05(2)	0.07(2)	–	–	–		0.07(3)	86 → 0 g)
119.18(8)	0.13(5)	0.34(5)	–	–	–	f)	0.14(5)	152 → 33 g)
126.15(2)	15(1)	18.0(15)	11(1)	33	10(2)	E1	16(1)	159 → 33
139.92(5) c)	0.35(4)	0.45(7)	<1	1.2	–		0.38(5)	140 → 0 h)
151.88(5) c)	0.61(5)	0.81(4)	<1	2.5	–	E1 e)	0.64(6)	152 → 0
154.00(10) c)	0.061(15)	0.16(3)	–	–	–		0.065(16)	154 → 0 g)
158.87(5) c)	3.2(2)	3.35(14)	2.0(5)	8.2	1.3(3)	f)	3.3(2)	159 → 0

a) For intensities per 100 decays of ^{100}Pd, multiply by 0.53(4) (see text).
b) Energy from present $\gamma\gamma$ coincidence data.
c) $\gamma\gamma$ data indicate coincidence only with Rh K–x rays.
d) Deduced from I_γ (present data) and theoretical internal conversion coefficients. For intensities per 100 decays, multiply by 0.53(4). Transition assumed to be dipole if multipolarity is not known.
e) From ref. [9].
f) Assumed E1 from level scheme.
g) New placement proposed in the decay of ^{100}Pd from present $\gamma\gamma$ coincidence data.
h) Earlier placement [5] from a 214– keV level is not supported by present $\gamma\gamma$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61.60
72.52
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84.00
86.37
119.18
126.15
139.92
151.88
154.00
158.87
B
BS
S
U
÷4
÷20
÷20
÷4
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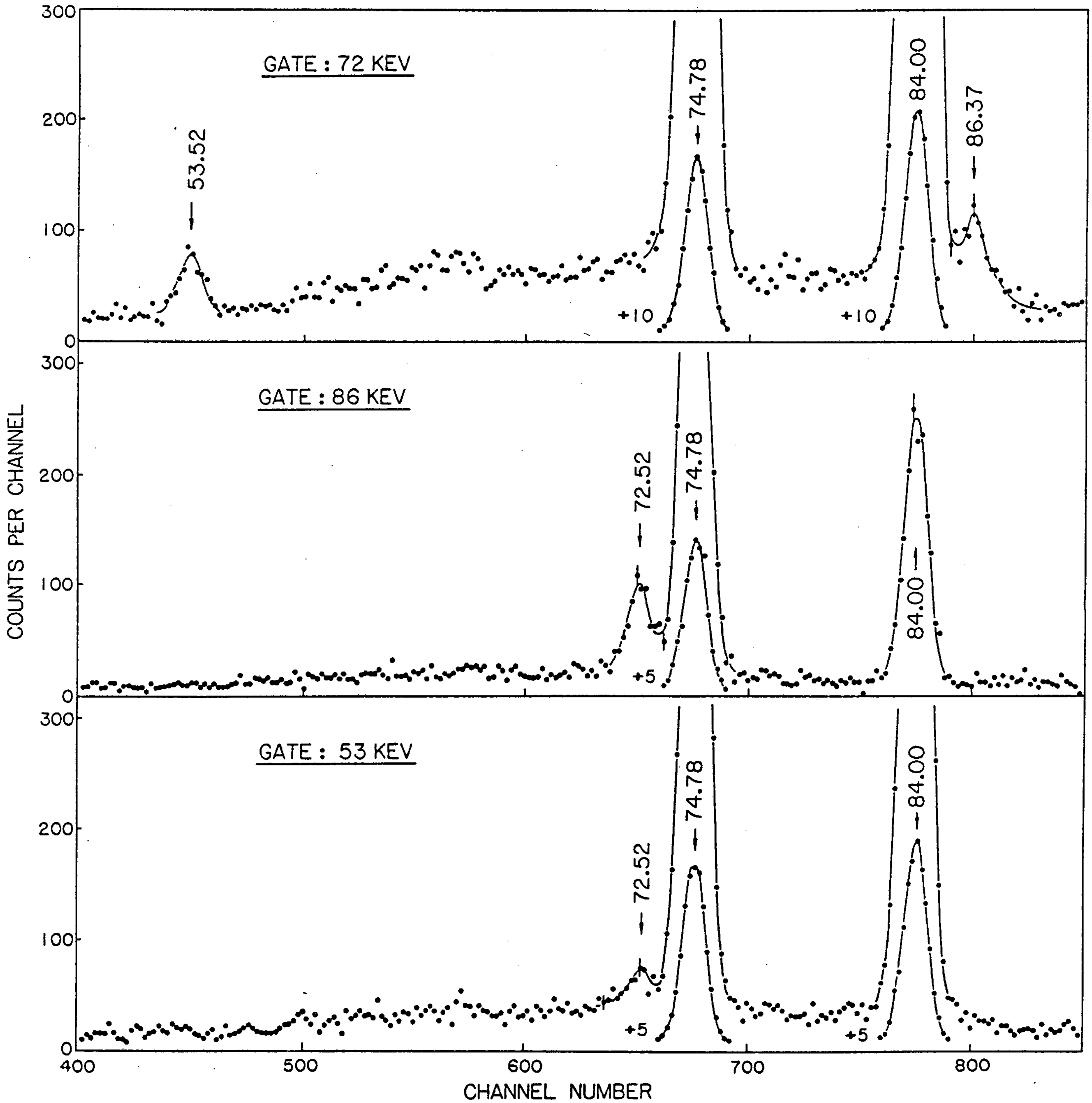
1400
1600
1800

Figure 1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53.52
74.78
84.00
86.37
+10
+10
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72.52
74.78
84.00
+5
GATE : 53 KEV
72.52
74.78
84.00
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+5
COUNTS PER CHANNEL
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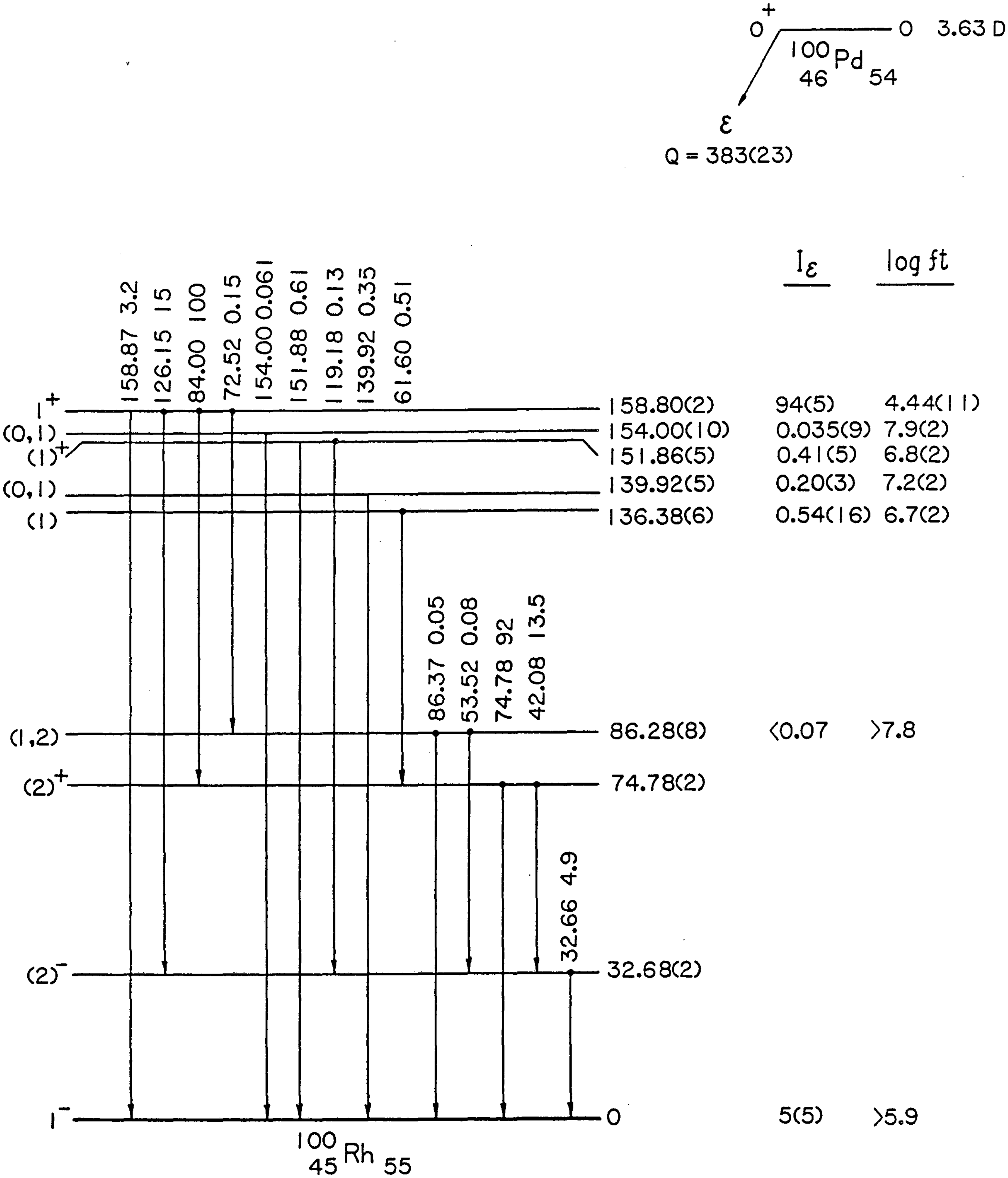
400
500
600
700
800

Figure 2



0+
0 3.63 D
100 46 Pd 54
ε
Q = 383(23)
Iε
log ft
158.87 3.2
126.15 15
84.00 100
72.52 0.15
154.00 0.061
151.88 0.61
119.18 0.13
139.92 0.35
61.60 0.51
1+ 158.80(2) 94(5) 4.44(11)
(0,1) 154.00(10) 0.035(9) 7.9(2)
(1)+ 151.86(5) 0.41(5) 6.8(2)
(0,1) 139.92(5) 0.20(3) 7.2(2)
(1) 136.38(6) 0.54(16) 6.7(2)
86.37 0.05
53.52 0.08
74.78 92
42.08 13.5
(1,2) 86.28(8) <0.07 >7.8
(2)+ 74.78(2)
32.66 4.9
(2)− 32.68(2)
1− 0 5(5) >5.9
100 45 Rh 55

Figure 3

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