ATOMIC MASSES OF RARE-EARTH ISOTOPES

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Abstract

A survey is given of decay energies of rare-earth isotopes measured in electron-capture decay by relative P_K ratios, EC_K/β^+, and EC/β^+ ratios. Atomic masses of A = 147 isotopes and of ^{146}Gd and ^{148}Dy were derevid. The masses of these isotopes and of α-decaying precessors are compared with predictions of current mass formulae. The subshell closure at Z = 64 is shown for N = 82, and 84 isotones.

1. Introduction

The motivation to perform remeasurements of the decay energies of ^{146}Gd[1)], ^{147}Eu and ^{147}Gd[2)] was to resolve discrepancies in earlier Q_{EC} values. Differing results have been reported from studies of the radioactive decay[3-9)] and from nuclear reaction work[10,11)]. A similar situation is found in the decay energies of ^{145}Sm[12,13)] and ^{151}Gd[12,14)] which has also prompted a remeasurement[15)].

The measurement of ^{148}Dy, and ^{148}Tb aimed at determination of the unknown atomic masses[16)]. The isotopes ^{146}Gd and ^{148}Dy represent key masses in the sense that α-decay chains which are ending at these isotopes permit the determination of masses at a distance from the region of ß stability. Six sequential α decays connect ^{172}Pt with ^{148}Dy, and a chain of eight α decays connect ^{178}Hg with ^{146}Gd. Until recently this series was interrupted between ^{162}Hf and ^{158}Yb, but this α decay has now been observed[17)]. By comparison of the experimentally determined masses with calculated ones a test of mass predictions in the region far from ß stability is achieved.

Our recent measurement of the unknown decay energies of 1.9 min and 1.6 h - ^{147}Tb [18)] is performed in order to determine the relative position of the two states. Alpha decay chains are also ending at ^{147}Tb. In case the α decays between nuclear ground states are identified, further atomic masses far from stability become accessible and proton separation energies can be derived from a number of isotonic mass differences.

Our experimental masses can be used to demonstrate a shell closure at Z = 64. This is reflected in the two-proton separation energies by a sudden drop of the regular decrease when the shell is crossed in the direction towards heavier isotopes[16)].

2. Decay energies of ^{146}Gd, ^{147}Eu, ^{147}Gd, and ^{145}Sm

We have listed the contradicting values of earlier measurements of the decay energies of these four nuclides in Table 1 together with our results. In the course of the ^{146}Gd investigation we have established a new γ-decay scheme following the electron capture[1)].

Table 1. Experimental Q_{EC} values

isot.	author	ref.	exp. method	Q_{EC} (keV)
^{146}Gd	Ageev 1970	3	β^+ end-point	1756 ± 30
	Wapstra'77	4	extrapol.	1200
	Alford '79	11	$^{144}Sm(^3He,n)$	1026 ± 30
	Pardo '80	12	$^{144}Sm(^{12}C,^{10}Be)$	1015 ± 26
	present	1	P_K, β^+/EC_K	$950 < Q_{EC} < 1064$
^{147}Gd	Adam 1969	5	β^+end-point	2382 ± 25
	Gromov '65	6	"	2221 ± 15
	Vylov '79	7	"	2184 ± 5
	Pardo '80	11	$^{144}Sm(^{12}C,^{9}Be)$	2070 ± 50^a 2116 ± 45^b
	present	2	$\beta^+/EC=2.18\cdot10^{-2}$	2088 ± 45
^{147}Eu	Adam 1967	8	β^+end-point	1767 ± 10
	Vylov '79	9	"	1723 ± 3
	present	2	$\beta^+/EC=4.13\cdot10^{-3}$	1669 ± 30
^{145}Sm	Sujokowski '68	13	EC bremsspec.	607 ± 7
	present	15	P_{K492}/P_{K60}	630 ± 11^c
^{148}Dy	present	16	$EC_K/\beta^+ = 21.7$	2669^{+65}_{-50} ^{c}
	Spanier'81	30	$EC_K/\beta^+ = 14.7$	2802 ± 60
^{148}Tb (2.2 min)	present	16	$EC/\beta^+ = 2.36$	5805 ± 50
^{148}Tb (60 min)	Wapstra'77	4		5630 ± 80
^{147}Tb (1.9 min)	present	18	$EC_K/\beta^+=.85$	5170 ± 100^c

a) adjusted ^{147}Eu mass from ref. 12
b) with present ^{147}Eu mass
c) preliminary data

The decay energy was measured by comparing the relative K-capture probabilities of the transitions feeding the new level at 690.7 keV and the one at 384.8 keV with calculated values[19,20)]. Since the decay energy turned out to be unfavourably large for this method we were only able to derive a lower limit of the decay energy, $Q_{EC} > 950$ keV[1)]. The upper limit of this decay energy was obtained from the investigation of ^{150}Dy and using the closed cycle of atomic masses of ^{150}Dy, ^{150}Tb, ^{146}Gd and ^{146}Eu. The following relation holds: $Q_{EC}(^{146}Gd) = Q_{EC}(^{150}Dy) + Q_\alpha(^{150}Tb) - Q_\alpha(^{150}Dy) = Q_{EC}(^{150}Dy) - 762.3 \pm$

5.8 keV, where the last value was obtained by inserting α-decay energies from a recent compilation[21]. The isotope ^{150}Dy was produced by the ^{141}Pr(^{14}N,5n) reaction at the heavy-ion accelerator facility at Berlin. In Fig. 1 the simple decay scheme[22] of ^{150}Dy and the detector set-up is given as an inset.

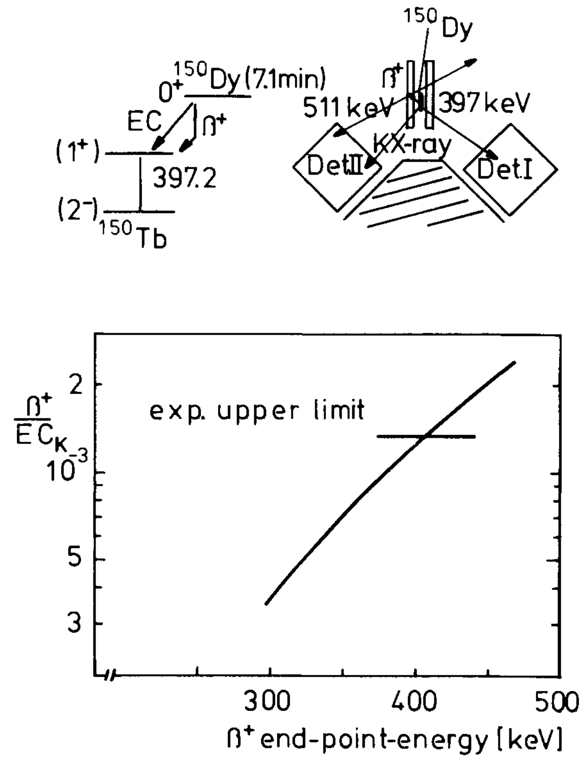

Fig. 1 Comparison of experimental and calculated β^+/EC_K values in ^{150}Dy. Insert: decay scheme of ^{150}Dy, detection geometry

The β^+/EC_K ratio was found by counting of annihilation radiation and Tb-K X-rays in coincidence with 397 keV γ-rays. This method (I) was also applied in the investigation of ^{148}Dy and ^{147}Tb (1.9 min). The experimental ratio is given by

(1) $\beta^+/EC_K = N_{511-397}/N_{K-397} \cdot \varepsilon_K/(2 \cdot \varepsilon_{511}) \cdot \omega_K / \{s \cdot (1-f)\}$.

We denote by N_{i-397} the number of observed coincidences, by ε_i the detection efficiency, and by ω_K the fluorescence yield. The quantity $f \ll 1$ corrects for annihilation in flight[23] while s is a correction faktor introduced to account for the annihilation taking place outside the source. This factor was obtained in two ways, first by mapping of the efficiency as a function of source position and weighing solid angle sections into which the positrons are emitted with these efficiency values, second by placing a calibrated ^{22}Na source, thickness 1 μg/cm² on 10 μm of mylar foil, in the source position. It is important to note that the coincidence efficiency cancels, and only the relative efficiency enters into equation 1. A further small correction of .4% is caused by summing of the back scattered second annihilation quantum.

No β^+ decay was observed in the decay of ^{150}Dy. The experimental upper limit of the ratio, $\beta^+/EC_K < 1.33 \cdot 10^{-3}$, is compared in Fig. 1 with the calculated ratios[24]. The tabulated values[24] were changed to take the exchange correction into account. The comparison gives an upper limit of the β^+ end-point energy of 407 keV, and the value $Q_{EC} < 1826$ keV. Application of the calculated ratios of Gove and Martin[25] result in a value 5 keV lower. In Fig. 1 no additional correction factor[1] has been used for the theoretical ratios.

For the ^{146}Gd decay energy the upper limit of 1064 keV is derived which is listed in Table 1. The energy region of the ^{146}Gd decay energy, found by determination of lower and upper limits, is supporting the reaction data[10,11].

To remeasure the decay of ^{147}Gd, and of ^{147}Eu we have produced ^{147}Gd by ^{144}Sm (α,n) reaction at the Göttingen cyclotron. After aging, the same sources have been used for the ^{147}Eu measurements. In a similar geometry as indicated in the inset of Fig. 1 coincidences were measured. For the measurement of ^{147}Eu, the Ge(Li) gate detector was replaced by an intrinsic Ge X-ray detector, 10 mm diameter and 5 mm thick.

In these decay processes we have determined (β^+/tot)-decay ratios by the measurement of relative γ-ray intensities in the singles, and in the coincidence spectrum. This procedure (II) is discussed for the decay scheme of ^{147}Eu. A partial decay schme of this nucleus is given in Fig. 2. For the branching feeding the 121.3 keV level in ^{147}Sm the ratio of total transitions and of positron decays, $(\beta^+ + EC)_{121}/\beta^+_{121}$, is determined. From the decay scheme it is seen that the balance of the transition intensities feeding and deexciting this level is given by

$$(\beta^+ + EC)_{121} + \sum_i I_i = I_{121}.$$

Herein ΣI_i is the total intensities of transitions from higher excited states feeding the 121 keV level. By a simple arithmetic therefrom the following expression is derived:

(2) $(\beta^+ + EC)_{121}/\beta^+_{121} = I_{677}/\beta^+_{121} \cdot (I_{121}/I_{677} - \Sigma I_i/I_{677})$

The intensity ratios in the brackets of the right side of this equation were obtained by singles counting at 15 cm distance in order to minimize summing effects. Relative detection efficiencies and values of total conversion coefficients were used to arrive at $I_{121}/I_{677} = 4.63 \pm .02$, and $\Sigma I_i/I_{677} = 2.048 \pm .010$. The factor in front of the brackets was obtained from the coincidence measurement. The coincidence spectrum of ^{147}Eu gated by 121.3 keV - rays is given in Fig. 2. Essentially, the wanted intensity ratio is given by the intensity ratio of the 677 keV, and 511 keV line.

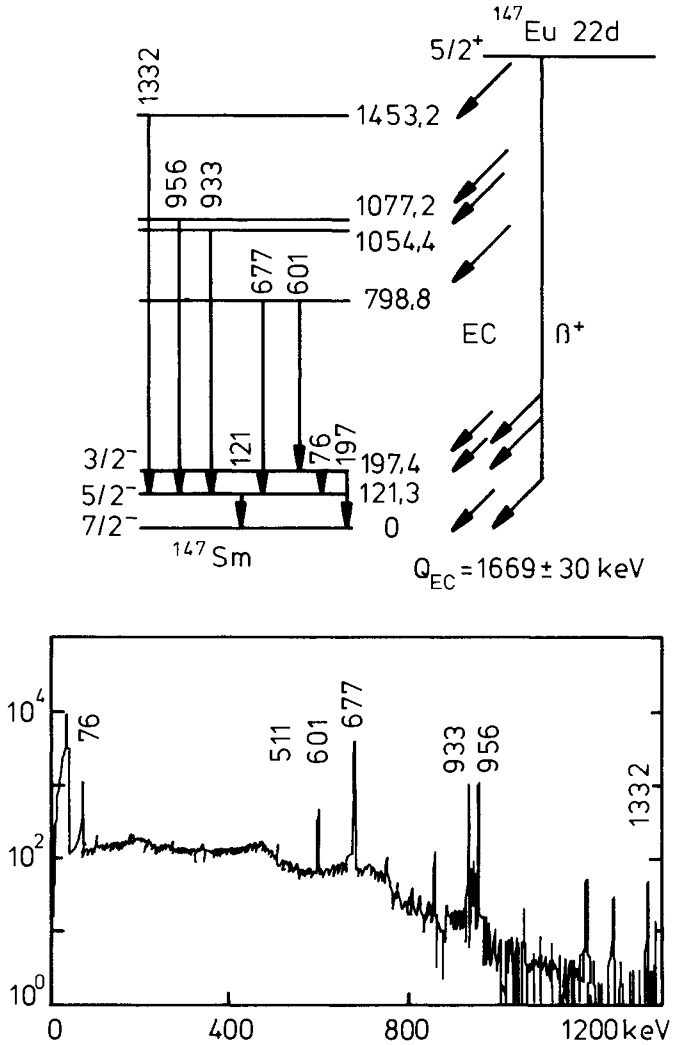

Fig. 2 Partial decay scheme of ^{147}Eu and spectrum gated by 121 keV γ-rays

However, the feeding of the 197 keV level by β^+ decay and the presence of the 76 keV transition in Fig. 2 causes a small correction of the number of 511 keV quanta. It is obtained in an extra coincidence measurement with the gate on 197 keV γ-rays from the intensity ratio of 511 keV, and 601 keV quanta. To obtain the correction the latter ratio was normalized by the intensity of the 601 keV line in Fig. 2 and gave a 5% deduction of the number of 511 keV quanta. If $N_{511-121}$ denotes the corrected number in the coincidenc spectrum of Fig. 2 the factor in equation 2 is given by the expression:

$$I_{677}/\beta^+_{121} = N_{677-121}/N_{511-121} \cdot 2\varepsilon_{511}/\varepsilon_{677} \cdot s \cdot (1-f),$$

where the values ε_i, s, and f have the same meaning as above. We obtained the ratio $I_{677}/\beta^+_{121} = 94.6 \pm 20.1$ and deduced herefrom $(\beta^+/EC)_{121} = (4.13 \pm .86) \cdot 10^{-3}$ (ref. 2).

The same method was used in the investigation of the ^{147}Gd decay[2] to obtain $(\beta^+/EC)_{229} = (2.18 \pm .41) 10^{-2}$. This evaluation was simpler in the sense that no positron branching to a higher level had to be corrected for. These ratios are compared in Fig. 3 with the calculated ratios of Gove and Martin[25], and of Dzhelepov et al.[24]. Using for these first forbidden non-unique transitions the tabulated values of allowed transitions altered to include exchange corrections [24,25].

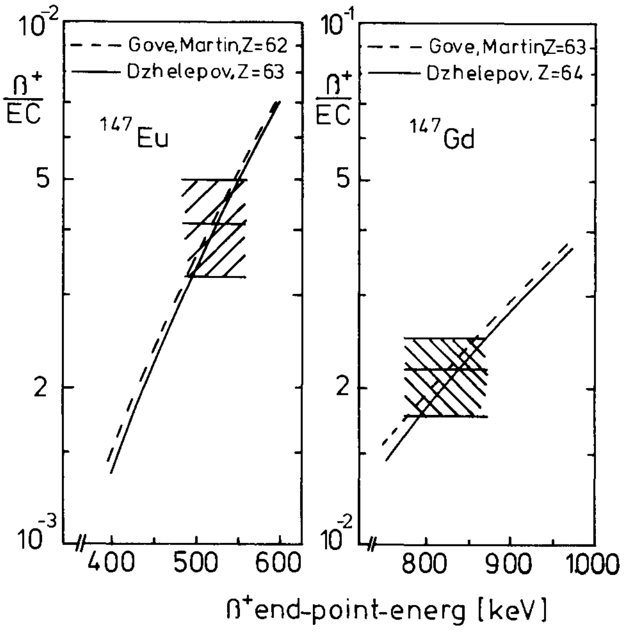

Fig. 3 Comparison of experimental and calculated β^+/EC values in ^{147}Eu, and ^{147}Gd decay as function of β^+ end-point energy

The Z values to be chosen in the tabulations are indicated in Fig. 3. Taking for comparison the ratios of Dzhelepov, decay energies of 1669±30 keV, and of 2088±45 keV were obtained for ^{147}Eu, and ^{147}Gd, respectively[2], and are given in Table 1. Again, our value for ^{147}Gd is supporting the reaction work[11], however, our result for ^{147}Eu is smaller than the earlier extrapolated β^+ end-point energies[8,9].

In the adjustment of experimental data for the table of atomic masses closed cycles of masses are considered[4,12]. The bearing of the ^{145}Sm, and ^{151}Gd data on the ^{147}Eu decay energy and the presently exsisting inconsistencies have recently been refered[12]. We therefore have started the remeasurement of ^{145}Sm, and ^{151}Gd decay energies[15].

In this investigation we are making use of the energy dependent K capture probability[19]. The decay of ^{145}Sm is a favourable case since a capture branching leading to the 492 keV level in ^{145}Pm having a small transition energy is compared with the capture leading to the 61 keV level, having a large energy[26]. This method[26] is also used in ^{151}Gd decay. For ^{145}Sm the relative K-capture probability is expressed by the number of 61, and 492 keV γ-rays in the singles spectrum, N_{61}, and N_{492}, and in coincidence with K X-rays, $\bar{N}_{61-K}$, and $\bar{N}_{492-K}$. It is expressed by

$$P_{K492}/P_{K61} = \bar{N}_{492-K}/\bar{N}_{61-K} \cdot N_{61}/N_{492} \cdot$$

An advantage of this relative measurement is that detector, and coincidence efficiency and also absorption corrections cancle.

Our value[15] for this ratio, 0.46±.07, was compared with ratios deduced from the tabulation[24] taking for the EC decay to the 492 keV level values of a first-forbidden unique, and to the 61 keV level, of an allowed transition.

The comparison gives the decay energy $Q_{EC}(^{145}Sm) = 630 \pm 11$ keV also listed in Table 1.

3. Decay energies of ^{148}Dy, 2.2 min ^{148}Tb and ^{147}Tb

New values of the decay energies of ^{148}Dy, and of 2.2 min ^{148}Tb have recently been measured[16]. These isotopes were produced at the heavy-ion accelerator facility at Berlin by ^{141}Pr(^{14}N,7n), and ^{141}Pr(^{14}N, p6n) reactions using 120 MeV ^{14}N ions, and have been measured in a similar counting array as shown in Fig. 1. The isotope ^{148}Dy has a simple decay scheme[27] as shown in the inset of Fig. 4. The β^+/EC_K ratio was measured in a coincidence experiment (method I) by the counting of annihilation radiation and of Tb-K X-rays gated by 620 keV γ-rays. This coincidence spectrum is given in Fig. 4. Here, also the high energy γ-ray spectrum is displayed in order to show the absence of further γ-rays and to exclude the feeding of a higher state in ^{148}Dy decay[16,28]. From the number of K X-rays and of annihilation quanta the value $(EC_K/\beta^+)_{620} = 21.7 \pm 3.9$ was found using equation 1, and by comparison with calculated ratios[24] (see below) we obtain the β^+ end-point energy of (1027^{+65}_{-50}) keV and the value $Q_{EC} = (2669^{+65}_{-50})$ keV.

The same method was used in a first evaluation of recently measured data in the decay of 1.9 min ^{147}Tb. We have produced ^{147}Tb by the ^{141}Pr(^{12}C,6n) reaction at the Berlin acelerator using 100 MeV ^{12}C ions. The number of 511 keV quanta, and of Gd-K X-rays was measured by gating on 1397.7 keV, and on 1797.8 keV γ-rays. The result for the β^++EC transition leading to the 1397.7 keV level in ^{147}Eu is = $(EC_K/\beta^+)_{1397} = 0.85 \pm .08$ [18].

The comparison with calculated ratios[24] gives an end-point energy of (2750±100) keV and a decay energy of Q_{EC} = (5170±100) keV.

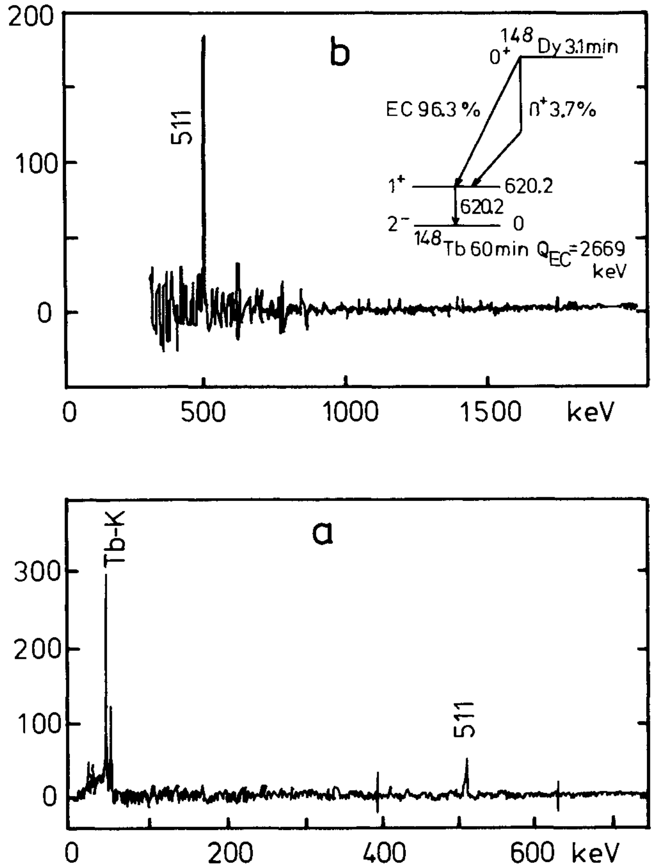

Fig. 4 Quanta spectrum of ^{148}Dy gated by 620 keV transitions, a) low energy, b) high energy part. Insert: decay scheme of ^{148}Dy

An other evaluation method (III) is used in the investigation of 2.2. min ^{148}Tb. In this decay a cascade of 882, 394 keV, and of two more γ-rays with the same transition intensity are directly following the β^++EC transition which is leading to the level at 2693 keV in ^{148}Gd. By gating on 882 keV γ-rays the balance of the number of transitions feeding the 2693 keV, and deexciting the level below at 1810 keV is given by $(\beta^++EC)_{2693} = I_{394}$. From this equation we calculate the EC/β^+ ratio which can be expressed by the number of γ-rays measured in coincidence with the 882 keV line, N_{i-882}, explicitely,

$$(3) \quad (EC/\beta^+)_{2693} = I_{394}/\beta^+{}_{2693}{}^{-1} = N_{394-882}/N_{511-882} \cdot 2\,\varepsilon_{511}/\xi_{394} \cdot (1+\alpha_{394}) \cdot s \cdot (1-f) -1.$$

The factors in eq. 3 have the same meaning as in equation 1 and α_{394} is the total conversion coefficient. A small correction for summing in the 511 keV and in the 394 keV lines was also performed. This coincidence method we have applied successfully before[29]. In ^{148}Tb decay the experimental value is $(EC/\beta^+)_{2693} = 2.36 \pm \cdot 18$[16] and we obtain the total decay energy Q_{EC} = (5805±50) keV. A similar evaluation of our recently measured data of ^{147}Tb (1.6 h) is in progress[18].

In our earlier evaluation[16] of the decay energy of ^{148}Dy, and of 2.2. min ^{148}Tb we have used the tabulated ratios of Gove and Martin[25] which are arranged for the atomic number of the daughter isotopes and in a similar way have used the tabulation of Dzhelepov et al.[24]. We are presently using ratios in the tables of Dzhelepov et al. which are given under the atomic number of the parent isotope. In addition we are applying an exchange correction to these values EC_K/β^+ [24]. This procedure causes a slight shift in the theoretical curve of Dzhelepov et al. and consequently gives different values of the decay energies, e.g. the Q_{EC} value of ^{148}Dy is increased by 17 keV and of 2.2 min ^{148}Tb, by 50 keV. This reevaluation is shown in Fig. 5, and the results are also given in table 1.

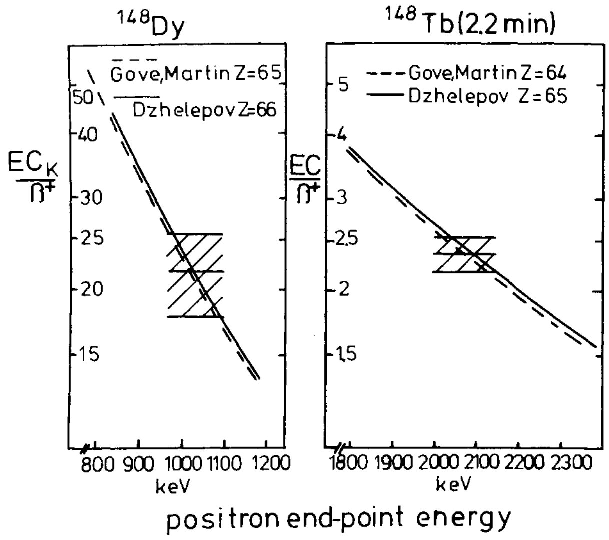

Fig. 5 Reevaluation of β^+ end-point energy in ^{148}Dy, and ^{148}Tb (2.2 min) decay, compare text

New mass values have been obtained from the following experimental information

1) Q_{EC}-values from this survey and from ref. 10, and 11 (Table 1)

2) Q_α-values given in ref. 16, Table 3, and the new experimental value of ^{162}Hf, E_α = 4308±10 keV[17]

3) the mass excess values from ref. 4 of ^{146}Eu (-77111±11 keV), ^{148}Gd (-76268 ± 6 keV), ^{148}Tb (60 min) (-76640±80 keV), and ^{147}Sm (-79265±5 keV).

4. Discussion and conclusion

From experimental EC_K/β^+, EC/β^+ and from relative P_K values, decay energies for a series of isotopes have been obtained and are given in Table 1. The known experimental Q_{EC} value in the decay of ^{148}Tb (60 min) [4] is also given as well as a result obtained for ^{148}Dy recently by a less direct mehtod[30]. Comparing the decay energies of the two isomers we find that the 2.2 min ^{148}Tb state is higher in energy by 175±94 keV and therefore very probably is the isomeric state. We hope that a similar decision can be made from the recorded data taken for the decay of the two ^{147}Tb isomers in a short time.

New mass excess values have been determined and are given in Table 2. We are giving the masses for the A = 147 isobars, for ^{148}Dy, and 2.2 min ^{148}Tb. The ^{146}Gd mass was taken from the reaction data[10,11]. We have also listed the experimental limits of this mass[1].

The masses are compared with the predictions of current mass formulae[31-33]. We found a fair agreement with the liquid droplet masses of Myers[31] and the droplet formula in the version of Hilf et al.[32]. In Table 2 we have listed these mass differences. Included in Table 2 are the masses of the two rows of isotopes within the α-decay chains ending at ^{148}Dy and ^{146}Gd. They represent two sections through the mass surface. These sections as a function of the mass number are given in Fig. 6 by the two bands A, and B. Comparison is done with the calculated masses[31-33]. The width of the bands represent the error of the experimental masses alone which is ±100 keV for the masses in band A, and ±40 keV, in band B. The experimental limits for the ^{146}Gd, and ^{150}Dy masses[1] are indicated by the bars in Fig. 6, representing an independently measured key mass with an approximate error of ±60 keV for the isotopes in band B. The section B through the mass surface is closer to the line of ß stability than the section A. The differences $|M_{calc}-M_{exp}|$ which are smaller for the section B indicate that the calculated masses agree better with the experimental ones in this region of the nuclear chart.

The masses used in Fig. 6a, b are shell and shape corrected liquid droplet masses[31,32]. We find about the same differences, $M_{calc}-M_{exp}$, in the region of shell closures to the left (Z = 64, N = 82) and to the right close to the proton number Z = 82. Inbetween we observe a systematic deviation. The agreement with the masses of Hilf et al.[32] is comparatively good in Fig. 6a, the deviation between the shells is larger for the masses from

Table 2 Present mass-excess data compared to predictions from calculations of Myers (M), and Hilf, Groote, and Takahshi (HGT).

isotope	$M_{exp}-A$ (MeV)	$M_{calc}-M_{exp}$ (MeV) M	$M_{calc}-M_{exp}$ (MeV) HGT
^{147}Eu	-77.60 ± 0.3	0.04	-.09
^{147}Gd	-75.51 ± 0.5	+.14	+.58
^{147}Tb(1.9min)	$-70.34\pm.12$ [a]		
^{148}Tb(2.2min)	$-70.46^{+.07}_{-.06}$	$-.78+E_{is}$	$-.13+E_{is}$ [b]
^{148}Dy	$-67.97^{+.10}_{-.09}$	-.93	+.28
^{152}Er	$-60.62^{+.10}_{-.09}$	-.88	+.27
^{156}Yb	$-53.38\pm.10$	-.58	+.56
^{160}Hf	$-46.06\pm.10$	-.24	+.97
^{164}W	$-38.36\pm.10$	-.18	+1.02
^{168}Os	$-30.13\pm.10$	-.45	+.87
^{172}Pt	$-21.22\pm.10$	-1.22	+.50
^{146}Gd	$-76.09\pm.03$ [c]	-.37	+.37
	-76.16 [d]	-.30	+.44
^{150}Dy	$-69.32\pm.03$ [c]	-.35	+.23
	-69.27 [d]	-.40	+.18
^{154}Er	-62.62 ± 0.3 [c]	-.16	+.36
^{158}Yb	-56.02 ± 0.4 [c]	+.21	+.64
^{162}Hf	-49.17 ± 0.4 [c]	+.13	+.62
^{166}W	$-41.89\pm.04$ [c]	+.01	+.59
^{170}Os	$-33.94\pm.04$ [c]	-.24	+.43
^{174}Pt	$-25.34\pm.04$ [c]	-.71	+.16
^{178}Hg	$-16.33\pm.04$ [c]	-1.72	-.09

a) preliminary mass value
b) E_{is} energy of isomeric level in ^{148}Tb
c) ref. 10, and 11
d) ref. 1

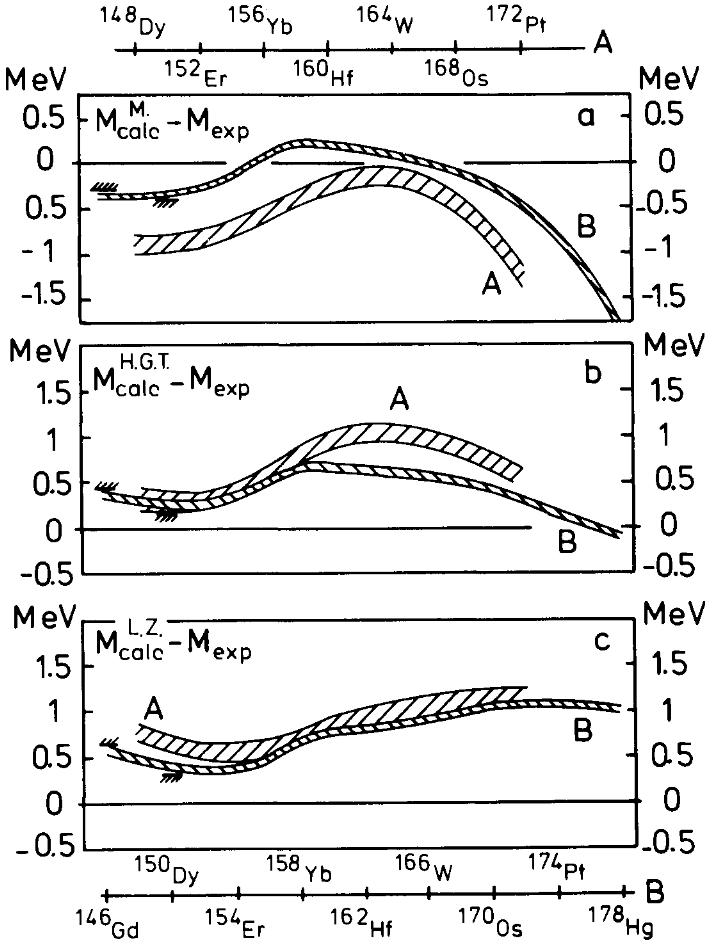

Fig. 6 a-c Comparison of experimental results of Table 2 with predictions of current mass formulae, mass differences in MeV, a) calculated masses of Myers, b) of Hilf et al., c) of Liran and Zeldes

inhomogeneous-partial-difference equations of Jaenecke and Eynon[33] in Fig. 6d which have proven to agree well in the region of neutron deficient Sn isotopes[29]. A fair agreement is observed with the masses of the semiempirical shell model of Liran and Zeldes[33]. No remaining shell effect as in Fig. 6a, d was observed in Fig. 6c. The formula of Comay and Kelson[33] being successful in the region close to stability shows in Fig. 6e large deviations when going to more neutron-deficient nuclei.

As has been discussed in ref. 16 the shell closure at Z = 64 can be observed in a discontinuity in the regular decrease of two-proton separation energies when the shell is crossed in the direction towards heavier nuclei. Using the mass excess values of Table 2 and taking for a try the mass of ^{147}Tb (1.9 min) to be the groundstate mass we have plotted the separation energies in Fig. 7. We observe subshell gaps of ~ .7, and ~ 1 MeV for the N = 84, and N = 82 isotones, respectively.

We intend to investigate mass differences of isotonic atoms after having established the ^{147}Tb masses.

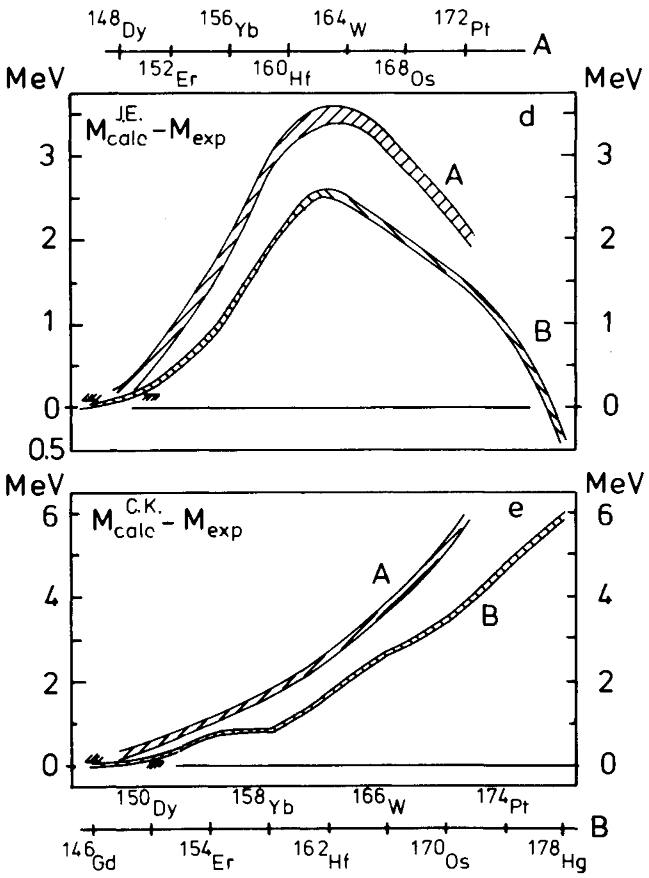

Fig. 6 d,e continued, d) masses of Jaenecke and Eynon, d) of Comay and Kelson

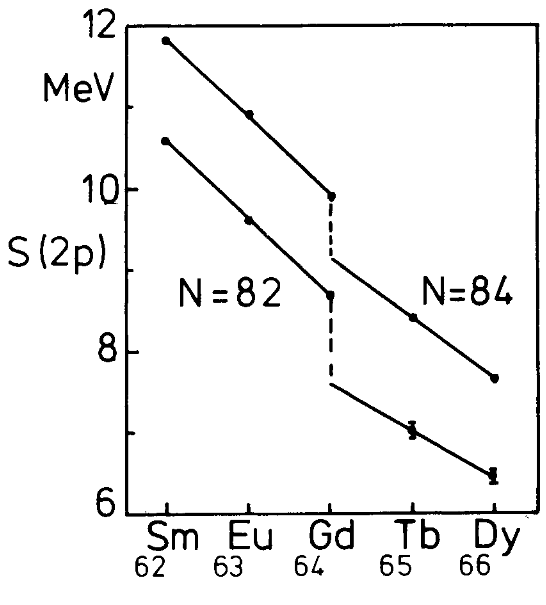

Fig. 7 Two-proton separation energies, S(2p), for N = 82, and 84 isotones. The value S(2p) of ^{147}Tb has to be increased by the isomeric transition if ^{147}Tb (2.2 min) is not ground-state

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DISCUSSION

F. Tondeur: The gap in S_{2p} systematics is not to be compared with the gap in single-particle energy at Z = 64 due to pairing effects. The latter must be higher than the former.

W.-D. Schmidt-Ott: I agree, there are two definitions of the gap.

P. Möller: Many speakers have today discussed new regions of deformation on the chart of the nuclides. I would like to show a colour plot of calculated[1]) ground state deformations for 4023 nuclei plotted versus Z and N. Of particular interest are the new deformed regions a little to the left and right of the N = 50 magic number. Some features in the very lightest region are also of interest. For instance, note that the nucleus $^{31}Na_{20}$ is quite deformed, despite the "magic" neutron number. The colour plot will be submitted to Physica Scripta[2]).

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