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PHYSICAL REVIEW C

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Atomic Masses of ²³²Th, ²³⁵U, and ²³⁸U and a Mass Table for the Heavy Isotopes*

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This paper reports the results of the authors' mass spectroscopic measurements on the masses of the isotopes ²³²Th, ²³⁵U, and ²³⁸U. Notice is taken of recently reported mass-difference measurements and Q-value measurements, and a least-squares process is used to construct a new mass table for the heavy isotopes. This table differs from Wapstra's 1967 table in two major respects: (1) For most isotopes with A > 220 the new mass value is lower than the old mass value by 20-25 μ u, or roughly $1\frac{1}{2}$ times the quoted error in the 1967 table. (2) As a result of the new measurements, the uncertainty in our knowledge of the masses of most of the heavy isotopes has been reduced by very roughly a factor of 3.

I. INTRODUCTION

Until recently the heaviest isotopes for which direct, high precision mass measurements were available were isotopes of bismuth and lead. In this paper the authors will present the results of their measurements on ²³²Th, ²³⁵U, and ²³⁸U, the isotopes which head the natural-radioactive-decay series.

The authors' measurements on these isotopes were made using a 16-in. magnetic radius doublefocusing Nier-Johnson mass spectrometer and the error-signal doublet-peak matching technique. The Minnesota mass-measuring instruments¹⁻³ and the error signal technique⁴ have been described in some detail elsewhere. No further description will be given here. The equipment, when tuned, produced a full-width at half-maximum (FWHM) resolution of 150 000 to 200 000 for these measurements.

Two major problems were encountered in this work. One had to do with sample introduction, and the other was a calibration problem. The calibration problem arose from the fact that the measured values of known mass doublets were consistently too high by about 20 ppm. This consistent discrepancy was discovered by measuring three different types of doublets. Measured values of doublets of the type $C_m H_{n+1} - C_m H_n$ were compared with the accepted value of the hydrogen mass. Measured values of $U^{35}Cl_{m_2}^{37}Cl_{n_2}^{-}-U^{35}Cl_{m_1}^{37}Cl_{n_1}^{-}$ type doublets were compared with the accepted value of the ³⁷Cl-³⁵Cl mass difference. Finally the sum of the measured values for the $C_9H_{10}-\frac{1}{2}^{235}U$ and $\frac{1}{2}^{235}$ U-C_oH_o doublets was compared with the accepted hydrogen mass value. In all three cases the measured values were found to be too high by about 20 ppm. This calibration problem for the Minnesota instrument was ultimately resolved by applying a 20 ± 3 -ppm correction to all doublet measurements. Other mass measurement laboratories have also had to apply corrections of this sort.

This calibration problem is somewhat similar to a problem encountered by Hudson⁵ in 1969 when he used the Minnesota instrument to make measurements on the light rare-earth isotopes. He

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found that measured values of hydrocarbon-hydrocarbon doublets agreed with the accepted hydrogen mass value to within 1 ppm, but closure errors indicated that measured values of metal-metal and metal-hydrocarbon doublets were too high by about 10 ppm. In the present work it was not possible to discern any difference between these three types of doublets. The cause of the earlier difficulty remains uncertain.

The sample problem arose from the fact that uranium and thorium are excellent getters. When heated, these elements and their salts tend to absorb large amounts of any hydrocarbon that is introduced into the instrument as a mass reference. The extent of the gettering depends somewhat on the hydrocarbon used. Eventually two workable doublets were found for each of the three isotopes to be measured.

In the case of ²³⁵U the gettering problem, combined with the low natural abundance of this isotope, made it necessary to use an enriched sample. The Oak Ridge National Laboratories provided the ²³⁵U enriched uranium in the form of U_3O_8 , and it was converted into UCl₄ by means of the process described in U. S. patent number 2 688 529.

II. RESULTS

About half a dozen runs were made on each of the doublets listed in Table I. The mass of each of the isotopes ²³²Th, ²³⁵U, and ²³⁸U was obtained by measuring two different doublets, and in each case the doublets gave answers that agree to within the limits of experimental error. Isotopic masses were calculated from the doublet values by employing the standard atomic masses of ¹H, ³⁵Cl, and ³⁷Cl listed in the 1964 Mass Table by Mattauch, Thiele, and Wapstra.⁶ The errors quoted in Table I are the quadratic sum of the standard deviation of the mean, the uncertainty in calibrating resistors in a precision voltage divider, the uncertainty in the 20 ± 3 -ppm correction, and the uncertainty in the 1964 Mass Table values for ¹H, ³⁵Cl, and ³⁷Cl. The final doublet errors correspond to about 1/1000 of the FWHM of the ion peak.

The doublet measurements listed in Table I include the 20 ± 3 -ppm calibration correction. At this point it might be well to comment on the magnitude of that correction. The doublets involving ²³²Th have widths of about 0.07 u. For such doublets a 20 ± 3 -ppm correction amounts to about 1.5 μ u with an uncertainty of about 0.3 μ u. This uncertainty is rather small compared to the statistical fluctuation of the runs, and it contributes very little to the over-all doublet measurement error. If one takes a weighted average of the two ²³²Th values, the doublet corrections tend to add. The over-all correction is

$$\alpha \frac{(\Delta m_1 / \sigma_1^2 + \Delta m_2 / \sigma_2^2)}{\frac{1}{2}(1 / \sigma_1^2 + 1 / \sigma_2^2)}$$

where α is the 20±3-ppm correction factor, Δm_1 and Δm_2 are the measured doublet values, and σ_1 and σ_2 are the quoted uncertainties in the ²³²Th values corresponding to Δm_1 and Δm_2 . This overall correction for ²³²Th amounts to about 3 μ u with an associated uncertainty of 0.4 μ u. A similar treatment leads to similar results for the doublet measurements involving ²³⁸U.

The case of ²³⁵U is a bit different. From Table I one sees that the doublets involving ²³⁵U have widths of about 0.5 u. For such doublets a 20 ± 3 -ppm calibration correction amounts to about 10 μ u with an associated uncertainty of about 1.5 μ u. This uncertainty accounts for a good deal of the quoted error in the doublet measurements. When one takes a weighted average of the two ²³⁵U measurements, however, one doublet correction tends

TABLE I. Doublet measurements in the present work.

Doublet	Reference hydrocarbon	No. runs	Δm (u)	Atomic mass (u) ^a
$C_9H_8-\frac{1}{2}Th$	Indene	6	0.0435712 ± 1.0	232 Th = 232.0380606 ± 2.4
$C_{12}H_8 - \frac{1}{2}Th \ {}^{37}Cl \ {}^{35}Cl$	Biphenyl	5	0.0761967 ± 0.9	232 Th = 232.0380600 ± 2.8
$\frac{1}{2}^{235}$ U-C ₉ H ₉	lpha methylstyrene	4	0.4515336 ± 1.9	235 U = 235.043 920 6 ± 4.1
$C_9H_{10}-\frac{1}{2}$ ²³⁵ U	lpha methylstyrene	5	0.5562921 ± 2.4	235 U = 235.043 919 6 ± 5.1
$C_9H_{11}-\frac{1}{2}^{238}U$	Durene trimethylbenzene	10	$0.060\ 683\ 0\pm1.1$	238 U = 238.050 788 2 ± 2.8
$C_{12}H_{10} - \frac{1}{2}^{238}U^{35}Cl_2$	Biphenyl	4	$0.084\ 005\ 4\pm 0.7$	238 U = 238.050 790 8 ± 3.1

^a Isotopic masses were calculated from the doublet values by employing the standard atomic masses of ¹H, ³⁵Cl, and ³⁷Cl listed in the 1964 Mass Table by Mattauch, Thiele, and Wapstra (Ref. 6).

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to be subtracted from the other. This pleasant result comes about because ²³⁵U forms the upper member of one doublet and the lower member of the other. The over-all correction is

$$\alpha \frac{(\Delta m_1/\sigma_1^2 - \Delta m_2/\sigma_2^2)}{\frac{1}{2}(1/\sigma_1^2 + 1/\sigma_2^2)}$$

Since the two terms in the numerator nearly cancel, this over-all correction amounts to only 0.1 μ u and is negligible compared to the statistical spread of the runs.

At about the same time the present measurements were being made at the University of Minnesota, a somewhat similar set of measurements was being made at Harvard by Kerr and Bainbridge.⁷ They obtained the following results:

$$\label{eq:238} \begin{split} & {}^{238}U^{-206}Pb^{32}S=0.104\,253\,9~u\pm10~\mu u~(\pm3.7~\mu u)~;\\ & {}^{235}U^{-206}PbC_2H_5=0.030\,341\,0~u\pm10~\mu u~(\pm2.8~\mu u)~;\\ & {}^{238}U^{-235}U=3.006\,858\,6~u\pm10~\mu u~(\pm6~\mu u)~;\\ & {}^{206}PbH^{-207}Pb=0.006\,394\,2~u\pm1.1~\mu u~. \end{split}$$

The $10-\mu u$ quoted errors represent limits of error. The errors listed in parentheses were computed on the "usual statistical basis." They include the uncertainty in a 30 ± 2 -ppm calibration correction. It is these statistical errors which will be used for comparisons in this paper.

By using the ¹H and ³²S mass values from the 1964 Mass Table⁶ one can cast these Harvard measurements in a somewhat more convenient form:

 ${}^{238}U^{-206}Pb = 32.076\ 327\ 6\ u\pm 3.8\ \mu u;$ ${}^{235}U^{-206}Pb = 29.069\ 467\ 0\ u\pm 2.8\ \mu u;$ ${}^{238}U^{-235}U = 3.006\ 858\ 6\ u\pm 6\ \mu u;$ ${}^{207}Pb^{-206}Pb = 1.001\ 431\ 0\ u\pm 1.1\ \mu u.$

Other measurements of interest involve nuclear reaction Q-value measurements. Because the energy gained in a nuclear reaction is related to the mass lost by $E = mc^2$, nuclear reaction Q-value measurements are another method of measuring mass differences. Wapstra's 1967 Mass Table,⁸ a revision of the heavy mass section of the 1964 table, lists a good many nuclear reaction Q-value measurements involving isotopes with A > 229. Table II lists some measurements⁹⁻¹⁵ made since 1967. A least-squares adjustment of all these Qvalue measurements gives the following isotopic mass difference values:

 $^{238}\text{U}\text{-}^{235}\text{U}$ = 3.006 873 1 u \pm 4.4 $\,\mu\text{u}$;

 235 U- 232 Th = 3.005 868 3 u ± 4.3 μ u.

Table III presents a comparison of various mass values. The 12 primary measurement values are

Reference	Reaction	Q exp	σ	Q ₁₉₆₇	σ	Δ^{a}	σ_{Δ}	Δ/σ_{Δ}
9	217 At(α) 213 Bi	7203.0	8	7199.3	3	3.7	9	0.44
	$^{221}\mathrm{Fr}(\alpha)^{217}\mathrm{At}$	6455.0	5	6457.3	3	-2.3	6	-0.39
	$^{225}\mathrm{Ac}(\alpha)^{221}\mathrm{Fr}$	5934.6	2	5931.3	3	3.3	4	0.93
10	228 Th(α) 224 Ra	5516.8	1	5520.9	2	-4.1	2	-1.83
	232 U(α) 228 Th	5413.6	1	5413.6	1	-0.0	1	-0.00
	233 U(α) 229 Th	4907.9	1	4909.4	2	-1.5	2	-0.71
	238 Pu(α) 234 U	5592.9	1	5592.1	1	0.8	1	0.57
	240 Pu(α) 236 U	5255.3	1	5255.1	1	0.2	1	0.12
	$^{242}\mathrm{Pu}(\alpha)^{238}\mathrm{U}$	4982.4	1	4980.8	3	1.6	4	0.43
11	$^{226}{ m Ac}(eta)^{226}{ m Th}$	1105.0	10	1118.0	7	13.0	12	1.07
12	233 U(<i>d</i> , <i>p</i>) ²³⁴ U	4656.0	15	4616.0	4	40.0	15	2,58
	235 U(d,t) 234 U	935.0	15	950.3	4	-15.3	16	-0.98
13	239 Pu(t, p) 241 Pu	3242.0	20	3285.0	4	-43.0	20	-2.11
	240 Pu(t, p) 242 Pu	3043.0	20	3065.7	5	-22.7	21	-1.10
	$^{242}{ m Pu}(t,p)^{244}{ m Pu}$	2576.0	20	2570.2	11	5.8	23	0.26
	$^{235}{ m U}(t,p)^{237}{ m U}$	3178.0	20	3183.1	4	-5.1	20	-0.25
	$^{236}{ m U}(t,p)^{238}{ m U}$	2780.0	20	2791.4	5	-11.4	21	-0.55
	$^{238}{ m U}(t,p)^{240}{ m U}$	2253.0	20	2246.0	12	7.0	23	0.30
	²³² Th(t , p) ²³⁴ Th	2487.0	20	2485.5	7	1.5	21	0.07
14	$^{239}{ m Pu}(n,\gamma)^{240}{ m Pu}$	6533.7	2	6523.9	4	9.8	4	2.33
15	$^{239}\mathrm{Pu}(n$, $\gamma)^{240}\mathrm{Pu}$	6533.1	1	6523.9	4	9.2	4	2.41
$^{a}\Delta = Q_{exp} - Q_{1967}.$								

TABLE II. Some nuclear reaction Q-value measurements made since 1967 [Q (keV)].

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		Mass value	Δ^{a}	
		$(u \pm \mu u)$	(µu)	Δ/σ
²³⁸ U	Present Minn.	$238.050\ 788\ 2\pm2.8\ ^{\mathrm{b}}$	-1.5	-0.54*
		$238.0507908\pm3.1^{ m c}$	1.1	0.39*
	Harvard	238.0507956 ± 8.0	5.9	0.73
	1967 Table	238.0508191 ± 12.4	29.4	2.37
	Minn. LSQ.	$238.050\ 789\ 7\pm1.8$	• • •	• • •
235 U	Present Minn.	$235.0439206\pm4.1~^{\rm d}$	3.8	-0.93*
		$235.043919.6\pm5.1^{\ e}$	-4.8	-0.94*
	Harvard	235.0439350 ± 7.5	10.6	1.41
	1967 Table	235.0439432 ± 11.5	18.8	1.63
	Minn. LSQ.	235.0439244 ± 2.1	•••	•••
232 Th	Present Minn.	$232.0380606 \pm 2.4^{\text{f}}$	1.0	0.41*
		232.0380600 ± 2.8 g	0.4	0.14*
	1967 Table	232.0380793 ± 11.9	19.7	1.66
	Minn. LSQ.	$232.038\ 059\ 6\pm1.7$	•••	•••
²³⁸ U- ²³⁵ U	Nuclear reactions	3.0068731 ± 4.4	7.8	1.77*
	Harvard (direct)	3.0068586 ± 6.0	-6.7	-1.12*
	Harvard (indirect)	3.0068606 ± 4.7	-4.7	-1.00
	Present Minn. (wtd. ave.)	3.0068692 ± 3.8	3.9	1.03
	1967 Table	3.0068759 ± 5.4	10.6	1.96
	Minn. LSQ.	3.0068653 ± 2.2	•••	•••
235 U $^{-232}$ Th	Nuclear reactions	3.0058683 ± 4.3	3.6	0.84*
	Present Minn. (wtd. ave.)	3.0058599 ± 3.7	-5.1	-1.38
	1967 Table	3.0058639 ± 4.7	-0.8	-0.17
	Minn. LSQ.	3.0058647 ± 2.4	•••	• • •
$^{238}U^{-206}Pb$	Harvard	32.0763276 ± 3.8	-2.1	-0.55*
	Present Minn. (wtd. ave.)	32.0763214 ± 7.3	-8.3	-1.14
	1967 Table	32.0763525 ± 11.5	22.8	1.98
	Minn. LSQ.	32.0763297 ± 2.5	• • •	• • •
$^{235}\mathrm{U}$ - $^{206}\mathrm{Pb}$	Harvard	29.0694670 ± 2.6	2.6	0.93*
	Present Minn. (wtd. ave.)	29.0694522 ± 2.8	-12.2	-1.58
	1967 Table	29.0694767 ± 10.4	12.3	1.18
	Minn. LSQ.	29.0694644 ± 2.2	• • •	•••
207 Pb- 206 Pb	Harvard	1.0014310 ± 1.1	-0.4	-0.36*
	1967 Table	1.0014365 ± 4.3	5.1	1.18
	Minn. LSQ.	1.0014314 ± 1.1	• • •	• • •

TABLE III. Comparison of mass values. The asterisk is used to designate primary measurements.

 $^{a}\Delta$ is the difference between the given mass value and the corresponding mass value in the Minnesota least-squares adjustment.

^b From the doublet $C_9H_{11} - \frac{1}{2}^{238}U$. ^c From the doublet $C_{12}H_{10} - \frac{1}{2}^{238}U^{35}Cl_2$. ^d From the doublet $\frac{1}{2}^{235}U - C_9H_9$. ^e From the doublet $C_9H_{10} - \frac{1}{2}^{235}U$.

^f From the doublet $C_9 H_8 - \frac{1}{2}^{10} Th$.

^g From the doublet $C_{12}H_8 - \frac{1}{2}Th^{37}Cl^{35}Cl$.

designated by asterisks. The other values were derived from these. In cases where it was necessary to assume a value for ²⁰⁶Pb to make a comparison, the value used was the value from the 1964 Mass Table.⁶

Probably the most stringent test of data agreement is the test of whether there exists a set of isotopic mass values which is in reasonably good

agreement with all of the measurements. If such a set exists, it can be found through a leastsquares adjustment process. In Table III the least-squares adjustment values are designated "Minn. LSQ.," and other values are compared to them. Note that for 10 of the 12 primary measurements there is agreement to within one σ . In only 1 of the 12 primary measurements is the disagree-

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			1967 Table			1971 Minn. LSQ.	
Series	Reaction ^a	Q _{exp} (keV)	Δ^{b}	Δ/σ	$m{Q}_{exp}$ (keV)	$\Delta^{\rm b}$	Δ/σ
4 n	$^{232}\mathrm{Th}(6lpha+4eta)^{208}\mathrm{Pb}$	$42\ 687.1\pm22$	14.8	0.67	42683.8 ± 22	23.6	1.07
4n + 1	229 Th $(5\alpha + 2\beta)^{209}$ Pb	35071.0 ± 33	-31.8	-0.96	35073.5 ± 33	-19.3	-0.58
4n + 2	$^{230}{ m Th}(5lpha + 3eta)^{210}{ m Bi}$	33525.3 ± 25	-28.4	-1.14	33525 , 3 ± 25	-15.0	-0.60
4n + 3	231 Pa(6 α + 3 β) 207 Pb	41345.5 ± 11	5.3	0.48	41345.5 ± 11	11.6	1.05

TABLE IV. Radioactive decay series.

^a The end points of the series are not the natural end points but rather the points of attachment to the top and bottom structures.

^b Δ = experimental Q-Q computed from mass table (1967 or Minn. LSQ.).

ment more than 1.5 σ , and in no instance is it greater than 2.0 σ . For the group of 12 primary measurements the rms value of Δ/σ is 0.85. In short, the data agreement is just about what one would expect on statistical grounds or perhaps a bit better.

The most serious disagreement seen in Table III is the disagreement with the 1967 table. One can understand how this disagreement comes about if one looks at the way in which the mass table was constructed. Nearly all input data for the 1967 Mass Table fall into three categories:

(1) A rather closely knit network of measurements linking isotopes with $A \ge 229$ (the top structure); (2) a somewhat less closely knit network of measurements linking isotopes with $A \le 210$ (the bottom structure); and

(3) four radioactive decay series linking the top and bottom structures.

From Table IV one can see a disturbing feature of the 1967 Mass Table: It places primary reliance on the 4n+3 decay series. This series, with its 11-keV quoted error, is weighted 4 times as heavily as the 4n series, 5 times as heavily as the 4n+2 series, and 9 times as heavily as the 4n+1series. Indeed the 4n+3 series is weighted nearly twice as heavily as the other three series combined. The 1967 table for the high mass region lists more than 200 pieces of input data and uses these data to compute the masses of 97 isotopes with A > 225; yet in the last analysis the accuracy of these computed values depends largely on a single string of Q values being correct. On the basis of the new measurements it now appears that the Mass Table values for isotopes in the top structure were too high because the 4n+3 decay chain was too long.

The six new measurements from Minnesota and two new measurements from Harvard provide eight precise and compatible links between the top and bottom structures. By continuing to think in terms of these structures one can get a fairly good idea of the effects of these new measurements. First of all note that the new Harvard measurements have effectively short circuited the decay chains and have become the dominant influence in determining the separation of the top structure and the upper part of the bottom structure. In a least-squares adjustment these measurements, with their 3- and 4- μ u quoted errors, are each weighted about 10 times as heavily as is the 4n+3series Q value. Table III shows that both of these links are shorter than predicted by the 1967 table. The ${}^{238}U-{}^{206}Pb$ link is shorter by 25 μ u; the 235 U- 206 Pb link is shorter by 10 μ u. Taken together the Harvard measurements serve to bring the top and bottom structures about 15 μ u closer together. Note also that members of the top structure do not move strictly in unison. The mass value for ²³⁸U tends to fall more than the value for ²³⁵U, and the two isotopic mass values come closer together.

The present Minnesota measurements of atomic masses introduce a new feature into the system, a set of six direct links between the top structure and "ground." From Table III one observes that these links are considerably shorter than the predictions of the 1967 table. For ²³⁸U the differences are 31 and 28 μ u; for ²³⁵U they are 23 and 24 μ u; for 232 Th they are 19 and 19 μ u. Once again note the lack of complete rigidity in the top structure and note that the ²³⁸U and ²³⁵U values are brought closer together. Taken together the present Minnesota measurements serve to lower the top structure by about 23 μ u. In accordance with what was said earlier, one expects that 15 μ u of this change will be absorbed in the links between the top and bottom structures and that 8 μ u of this downward change will be transmitted to the upper part of the bottom structure itself. Later when the authors present a complete tabulation of the Minnesota least-squares adjustment it will be seen that the changes wrought by the new measurements follow mainly this pattern. One noteworthy exception is provided by ²⁰⁷Pb, the lowest member of the 4n+3decay series. Even though this decay series is no

longer a dominant influence in determining the separation of the top and bottom structures, it still retains enough rigidity to exercise a palpable influence on the bottom structure at the point of attachment. Thus as the top structure is lowered by 20 μ u, the ²⁰⁷Pb value is pushed down 12 μ u instead of the usual 8 μ u.

The principal effect of the nuclear reaction Qvalue measurements listed in Table II is to alter relations within the top structure. In most cases the new measurements are in good agreement with the predictions of the 1967 table. In cases where there is disagreement, the quoted errors on the table values are usually substantially less than the quoted errors on the new measurements, so the new measurements have little influence on a new least-squares adjustment. The only exception to this rule is provided by the two new measurements of the ²³⁹Pu (n, γ) reaction Q value. These new measurements by Chrien et al.¹⁶ and Matussek et al.¹⁷ are about 9.5 keV, or 2.4 σ , higher than the prediction of the 1967 table and suggest that the true value of the ²⁴⁰Pu-²³⁹Pu mass difference is about 10 μ u lower than the table value. Precisely measured α -decay Q values link ²³⁹Pu and ²⁴⁰Pu with ^{235}U and $^{236}\text{U}.~$ A lessening of the $^{240}\text{Pu}-^{239}\text{Pu}$ mass difference leads to a corresponding reduction of the ²³⁶U-²³⁵U mass difference, and this effect in turn brings about a reduction of the ²³⁸U-²³⁵U mass difference. Among the Harvard measurements, the present Minnesota measurements, and the nuclear reaction Q-value measurements there seems to be unanimous agreement that the ²³⁸U and ²³⁵U mass values should be brought about 10 μ u closer together.

The new (t, p) reaction Q-value measurements by Britt and Cramer¹⁵ deserve further comment even though they do not bring about any major changes in a new least-squares adjustment. At the time the 1967 Heavy Isotope Mass Table was compiled three relevant (t, p) reaction Q values were available. These had been reported by Middleton

TABLE V. (t, p) reaction Q values.

Reaction	1964 Middleton and Marchant ^a	Q (keV) 1967 Mass Table ^b	1969 Britt and Cramer ^c
235 U(t, p)	$\begin{array}{c} 3138 \pm 20 \\ 2900 \pm 20 \\ 2242 \pm 20 \end{array}$	3183 ± 4	3178 ± 20
236 U(t, p)		2791 ± 5	2780 ± 20
238 U(t, p)		2246 ± 12	2253 ± 20

^a See reference 18.

^b See reference 8.

^c See reference 15.

and Marchant¹⁸ in 1964. In making up the 1967 table Wapstra rejected the ²³⁶U(t, p) Q value because it differed from a least-squares adjustment value of the other data by 110 keV, or $5\frac{1}{2}\sigma$. He kept the ²³⁵U(t, p) value but probably regarded it with some suspicion because it was $2\frac{1}{4}\sigma$ away from the leastsquares adjusted value. The ²³⁸U(t, p) value looked good. Note in Table V that in all three cases the 1969 measurements by Britt and Cramer confirm the prediction of the table. Similar experimental confirmation of the table predictions may be seen in Baranov's remeasurement¹² of the ²³⁸Pu and ²⁴⁰Pu α -decay Q values.

With that bit of advertisement for the leastsquares process the authors now present in Table VI the Minnesota least-squares adjustment for isotopic masses in the high mass region. This adjustment incorporates all the new mass measurements and Q-value measurements listed in this paper together with all experimental results in this region listed by Wapstra⁸ as input data for the 1967 Mass Table. No attempt has been made by the authors to assign their own laboratory weighting factors. The data which appeared as input in the 1967 Mass Table are given the same errors that Wapstra assigned them. New data are given the same errors assigned them in their original publication.

The 1964 Mass Table went astray in the high mass region because there were too few measured links between isotopes of different decay series and because one of the links that did exist was inaccurate. The multitude of links that were measured between 1964 and 1967 cured this problem. The 1967 table got into somewhat less serious trouble because it placed primary reliance on a single decay series. This difficulty has now been eliminated. The recent Minnesota and Harvard measurements provided eight precise and compatible links between the top and bottom structures and place our knowledge of isotopic masses in the high mass region on a considerably more secure foundation. An examination of Table VI shows that for most of the more than 100 isotopes with A > 220 the uncertainty in our knowledge of the isotopic mass has been reduced from roughly 12 μ u to roughly 4 μ u, about a factor of 3 improvement.

Note. Since the preparation of the present table, Wapstra and Gove have published a revision of the 1964 and 1967 Mass Tables. The 1971 Mass Table was published in Nucl. Data <u>A9</u>, 265 (1971). The 1971 Mass Table employed neither the present work nor the work of Kerr and Bainbridge.⁷ Comparison of the 1971 Mass Table results with the earlier results indicates no important changes. The following table lists some selected masses

	Minn ISO	σâ	1967 Table b	a a	V C	
Isotope	(u)	0 (μu)	(u)	(μu)	Δ (μu)	Δ/σ^{d}
	205 977 508 9	22	5157	22	-7	-0.3
TI 206	205.976.097.0	55	1037	7 5	-7	-0.9
Db 206	205.9700970	2.5	1057	5.7	-7	-1.2
PU 200	205,974 400 0	4.0		26	-7	-0.2
DI 200	205.9783825	20	11. 2021	20	-,	
T1 207	206.9774296	6.0	$\cdots 4431$	8.4	-14	-1.6
Pb 207	206.9758914	2.7	$\cdots 9031$	6.1	-12	-1.9
Bi 207	206.9784723	9.0	$\cdots 4839$	10	-12	-1.1
Po 207	206.9815931	14	$\cdots 6048$	15	-12	-0.8
T1 208	207.9820032	5.8	$\cdots 0106$	7.5	-7	-1.0
Pb 208	207.9766422	3.6	$\cdots 6493$	6.0	-7	-1.2
Bi 208	207.9797214	6.3	$\cdots 7302$	7.7	-9	-1.1
Po 208	207.981 238 2	11	$\cdots 2424$	12	-4	-0.4
T1 209	208.9853415	17	3523	18	-11	-0.6
Pb 209	208,9810706	7.7	$\cdots 0799$	9.0	-9	-1.0
Bi 209	208,980 384 3	4.4	3935	6.4	-9	-1.4
Po 209	208,982 420 5	11	$\cdots 4254$	11	-5	-0.4
TI 210	209,990 081 7	13	0935	14	-12	-0.8
Pb 210	209,984 183 0	3.1	··· 1898	6.0	-7	-1.1
Bi 210	209,9841148	2.9	$\cdots 1216$	5.9	-7	-1.2
Po 210	209 982 868 5	2.7	$\cdots 8751$	5.8	-7	-1.1
At 210	209.9870304	26	0370	26	-7	-0.2
Db 911	210 088 752 0	6.0	769.0	11	-17	-1.6
PU 211 D: 911	210,900 102 0	6.1	1030	85	-14	-1.6
DI 211	210,907,270.0	0.1		6.0		-1.8
P0 211	210,986,6451	3.0 0.6		11	-12	-1.1
At 211 Bn 211	210.9874964 210.9906011	9.0 14	•••6127	15	-12	-0.8
Tur 211	210.000 0011		0221			0.0
Pb 212	211.991 884 3	7.1	892 1	8.5	-8	-0.9
Bi 212	211.9912689	5.9	2764	7.5	-7	-1.0
Po 212	211.988 857 9	3.7		6.0	-7	-1.2
At 212	211.9907144	22	$\cdots 7232$	23	-9	-0.4
Rn 212	211.990 702 5	13	$\cdots 7067$	13	-4	-0.3
Bi 213	212.9943656	13	$\cdots 3764$	14	-11	-0.8
Po 213	212.992 839 2	9.2	$\cdots 848.8$	10	-10	-0.9
At 213	212.9930574	210	$\cdots 0666$	210	-9	-0.0
Rn 213	212.993 929 9	24	$\cdots 9348$	24	-5	-0.2
Pb 214	213,9998232	5.8	••• 843 9	12	-21	-1.7
Bi 214	213.998 713 7	13	$\cdots 7255$	14	-12	-0.8
Po 214	213.9951974	3.3	$\cdots 2042$	6.1	-7	-1.1
At 214	213,996 325 2	11	$\cdots 3320$	12	-7	-0.6
Fr 214	213,998 977 8	34	$\cdots 9844$	34	-7	-0.2
Bi 215	215.001 835 9	100	••• 853 9	100	-18	-0.2
Po 215	214.9994322	5.8	$\cdots 4494$	11	-17	-1.6
At 215	214.9986422	12	6559	14	-14	-1.0
Rn 215	214.9986741	110	$\cdots 6857$	110	-12	-0.1
Fr 215	215,000 395 0	34	$\cdots 4066$	34	-12	-0.3
Ra 215	215.002 753 6	26	$\cdots 7652$	26	-12	-0.4
Po 216	216.001 900 4	7.3	··· 908 2	8.7	-8	-0.9
At 216	216,002 409 0	9.5	4165	11	-8	-0.7
Rn 216	216,000 265 3	11	$\cdots 2724$	12	7	-0.6
Ra 216	216.0034830	35	$\cdots 4872$	35	-4	-0.1
At 217	217.0046980	13	$\cdots 7084$	14	-10	-0.7
Rn 217	217,003,910,6	10	••• 920 2	11	-10	-0.9
Fr 217	217.004 742 9	300	$\cdots 7521$	300	-9	-0.0
Do 917	217 006 388 3	40	393 2	40	-5	-0.1

TABLE VI. The Minnesota least-squares adjustment.

	Minn. LSQ	σ ^a	1967 Table ^b	σ ^a	Δ°	
Isotope	(u)	(µu)	(u)	(µu)	(µu)	Δ/σ
Po 218	218,008 987 9	5.7	008 6	12	-21	-1.7
At 218	218.008 697 5	14	$\cdots 7094$	15	-12	-0.8
Rn 218	218.005 598 9	11	605 7	12	-7	-0.6
Fr 218	2180075136	15	$\cdots 5204$	16	7	-0.4
At 219	219 011 299 1	86	••• 3171	87	-18	-0.2
Rn 219	219 009 490 2	5.6	507.6	11	-17	-1.6
Fr 219	219.009 235.8	25	2495	25	-11	-1.0
Ra 219	219.00023500 219.0100374	150	$\cdots 0491$	150	-12	-0.3
Bn 220	220 011 379 3	7.6	387 2	8 9	9	-0.9
Fr 220	220.0123102	10		19	7	_0.6
Ra 220	220.012.010.2	16	0260	16	-7	-0.4
E. 001		14	040.0	14		0.1
FF 221 Do 221		14		14	-11	-0.8
Ra 221		11	••• 912.6	12	-10	-0.8
AC 221	221.015 666 1	370	••• 6753	370	-9	-0.0
Rn 222	222.017 588 9	5.3	· · · 609 7	12	-21	-1.7
Fr 222	222.017 540 6	29	$\cdots 5520$	30	-11	-0.4
Ra 222	222.0153680	15	$\cdots 3749$	16	-7	-0.4
Ac 222	222.0177723	19	$\cdots 7791$	19	-7	-0.3
Fr 223	223.0197424	5.3	$\cdots 7604$	11	-18	-1.6
Ra 223	223.0185093	5.4	$\cdots 5269$	11	-18	-1.6
Ac 223	223.0191188	25	$\cdots 1325$	26	-14	-0.5
Th 223	223.020 907 0	190	$\cdots 9186$	190	-12	-0.1
Ra 224	224.020 195 3	7.8	$\cdots 2033$	9.1	-8	-0.9
Ac 224	224.0216940	14	$\cdots 7014$	15	-7	-0.5
Th 224	224.0214634	19	$\cdots 4704$	20	-7	-0.4
Ra 225	225.0236099	6.8	$\cdots 6295$	13	-20	-1.5
Ac 225	225,0232058	14	$\cdots 2145$	15	-9	-0.6
Th 225	225.023 935 2	13	••• 944 8	13	-10	-0.7
Ra 226	226.0254174	4.9	$\cdots 4382$	12	-21	-1.7
Ac 226	226.0260891	20	$\cdots 100.6$	21	-12	-0.5
Th 226	226.0248935	19	900 3	20	-7	-0.3
Pa 226	226.027 875 3	22	··· 882 1	22	-7	-0.3
Ra 227	227 029 162 1	22	••• 180 1	24	-18	-0.7
Ac 227	227 027 755 7	4.8		11	-18	-0.0
Th 227	227.027.708.9	- 1 .0 5 1		11	-18	-1.0
Pa 227	227,028 787 0	26	800 7	27	-18 -14	-1.0
Ra 228	228 031 076 6	5 5		19	10	1 5
Ac 228	228.0310100	5.5		10	-19	-1.5
Th 228	220.0310100	7 9		10 0	-19	-1.0
Da 228	228.020 022 2	1.0		9.0	-12	-1.2
U 228	228.031 369 9	22	3770	22	-7	-0.8
Th 990	990 091 760 6	4.9	700 4	10		
Do 220	229,031 700 0	4.0		12	-20	-1.6
Ta 229 U 229	229.032 073 2 229.033 486 4	10	•••• U81 9 •••• 496 0	10 14	-9 -10	-0.6
- 440 (77)- 000		10	4000	7-4	-10	-0.7
1n 230	230.0331383	4.4	••• 159 2	12	-21	-1.8
Pa 230 U 230	230.034 530 0 230.033 029 4	20	••• 541 4	21	-11	-0.5
0 400	200.000 928 4	19	••• 935 3	20	-7	-0.4
AC 231	231.038 553 4	110	572 6	110	-19	-0.2
1 II 231	231,0362990	3.1	3181	11	-19	-1.7
ra 231 U 991	231,0358849	4.5	903 1	11	-18	-1.6
U 231 No 991	231,0362703	54	••• 288 1	55	-18	-0.3
146 291	231.038 200 9	00	···2746	60	-14	-0.2
Th 232	232.038 059 6	1.7	0794	12	-20	-1.6

TABLE VI (Continued)

	Minn. LSQ	σ ^a	1967 Table ^b	σα	Δ ^c	
Isotope	(u)	(µu)	(u)	(µu)	(µu)	Δ/σ
Pa 232	232.038 580 8	23	··· 592 3	23	-12	-0.
U 232	232.0371369	7.9	$\cdots 1484$	9.3	-11	-1.
Pu 232	232.041 165 9	58	$\cdots 1730$	58	-7	-0.
Th 233	233.0415814	4.4	$\cdots 6037$	12	-22	-1.
Pa 233	233.040 244 9	3.8	$\cdots 2677$	12	-23	-1.
U 233	233.0396329	4.3	$\cdots 6541$	12	-21	-1.
Pu 233	233.0429775	25	$\cdots 9871$	26	-10	-0.
Th 234	234.0436073	5.2	$\cdots 6356$	13	-28	-2.
Pa 234	234.0433255	5.6	· · · 3536	13	-28	-2.
U 234	234.040 954 3	4.3	$\cdots 9756$	12	-21	-1.
Np 234	234.0428867	17	• • • 908 0	20	-21	-1.
Pu 234	234.0433057	20	$\cdots 3126$	20	-7	-0.
Pa 235	235.0454273	110	$\cdots 4462$	110	-19	-0.
U 235	235.0439244	2.1	$\cdots 9432$	11	-19	-1.0
Np 235	235.0440565	2.3	$\cdots 0754$	11	-19	-1.
Pu 235	235.0452718	63	••• 2896	64	-18	-0.
U 236	236.0455635	2.4	$\cdots 5913$	12	-28	-2.
Np 236	236.046 593 6	14	$\cdots 6051$	15	-12	-0.
Pu 236	236.0460375	9.6	$\cdots 0490$	11	-11	-1.
Pa 237	237.051 194 2	54	$\cdots 2197$	55	-25	-0.
U 237	237.0487250	3.5	· · · 750 5	12	-26	-2.
Np 237	237.0481699	3.3	$\cdots 1946$	12	-25	-2.
Pu 237	237.048 409 6	6.5	$\cdots 4341$	13	-25	-1.
U 238	238.050 789 7	1.8	$\cdots 8191$	12	-29	-2.
Np 238	238.050 949 2	9.0	$\cdots 9700$	14	-21	-1.
Pu 238	238.0495613	4.4	5822	12	-21	-1.
Cm 238	238.053 026 5	38	$\cdots 0334$	38	-7	-0.
U 239	239.0542984	3.1	$\cdots 3273$	13	-29	-2.
Np 239	239.0529315	3.4	9510	12	-19	-1.
Pu 239	239.0521562	2.2	$\cdots 1748$	11	-19	-1.
Am 239	239.053 023 7	22	$\cdots 042.6$	24	-19	-0.
U 240	240.0566015	10	$\cdots 6324$	17	-31	-1.
Np 240	240.0560519	64	0799	65	-28	-0.
Pu 240	240.053 808 1	2.3	$\cdots 8361$	12	-28	-2.
Cm 240	240.055 506 8	9.8	$\cdots 5183$	11	-11	-1.
Np 241	241.0583077	110	· · · 332 6	110	-25	-0.
Pu 241	241.0568476	3.2	··· 872 6	12	-25	-2
Am 241	241.0568255	3.2	850 4	$1\overline{2}$	-25	-2
Cm 241	241.0576544	6.6	6790	13	-25	-1.
Pu 242	242.0587417	2.1	··· 769 5	12	-28	-2.
Am 242	242.0595519	9.0		14	-21	-1.
Cm 242	242.058.838.8	4.5	859 6	12	-21	-1
Cf 242	242.0636599	39	••• 666 8	40	-7	-0.
Pu 243	243.062 004 1	8.2	$\cdots 0307$	15	-27	-1.
Am 243	243.0613740	3.6	••• 393 5	12	-20	-1
Cm 243	243.061.380.8	2.9	$\cdots 4001$	12	-19	-1
Bk 243	243.0630033	22	··· 022 2	25	-19	-0.
Pu 244	244,0642046	9.7	$\cdots 2347$	17	-30	-1
Am 244	244.0642818	3.3	••• 3096	12	-28	-2
Cm 244	244.062.747.7	2.5	••• 7754	12	-28	-2
Cf 244	244.0659770	10	•••9885	11	-11	-1
Am 245	245,0664511	4.9	4765	13	-25	-2.
13111 ATU	210.000 101 1	1.0		19	-25	-2

TABLE VI (Continued)

	Minn. LSQ	σ ^a	1967 Table ^b	σa	Δ ^c	
Isotope	(u)	(µu)	(u)	(µu)	(µu)	Δ/σ^{-d}
Bk 245	245.0663681	6.2	••• 393 0	13	-25	-1.9
Cf 245	245.0680462	6.9	$\cdots 0708$	13	-25	-1.8
Pu 246	246.0700941	55	$\cdots 1215$	56	-27	-0.5
Am 246	246.0696915	54	$\cdots 7189$	55	-27	-0.5
Cm 246	246.0672223	3.5	$\cdots 2497$	13	-27	-2.2
Cf 246	246.0688162	12	8370	16	-21	-1.3
Fm 246	246.0752488	45	$\cdots 2556$	45	-7	-0.2
Cm 247	247.0703508	8.2	$\cdots 3794$	15	-29	-1.9
Bk 247	247.0702682	32	$\cdots 2877$	34	-20	-0.6
Es 247	$247.073\ 604\ 4$	39	$\cdots 6233$	41	-19	-0.5
Cm 248	248.0723490	9.6	$\cdots 3787$	16	-30	-1.8
Cf 248	248.0721894	32	$\cdots 2171$	34	-28	-0.8
Fm 248	248.0771794	34	$\cdots 1909$	34	-11	-0.3
Cm 249	249.0759548	11	$\cdots 9845$	17	-30	-1.7
Bk 249	249.0749794	4.7	$\cdots 0048$	13	-25	-2.0
Cf 249	249.0748443	4.5	•••8697	12	-25	-2.0
Es 249	249.0763574	33	$\cdots 3823$	35	-25	-0.7
Bk 250	250.0783093	11	$\cdots 3367$	16	27	-1.7
Cf 250	250.0764042	6.4	$\cdots 4316$	14	-27	-2.0
Fm 250	250.0795247	34	$\cdots 5455$	36	-21	-0.6
Cf 251	251.0795617	13	$\cdots 5903$	18		-1.6
Es 251	251.0799461	46	$\cdots 9656$	47	-20	-0.4
Cf 252	252.0816265	11	$\cdots 6562$	17	-30	-1.7
Fm 252	252.0824717	39	4994	40	-28	-0.7
No 252	252.0889572	37	• • • 968 7	38	-12	-0.3
Cf 253	253.0851146	54	$\cdots 1400$	55	-25	-0.5
Es 253	253.084 824 8	7.1	$\cdots 8502$	14	-25	-1.9
Es 254	254.0880258	12	$\cdots 0532$	17	-27	-1.6
Fm 254	254.0868551	8.4	$\cdots 8825$	15	-27	-1.9
No 254	254.090 968 6	38	$\cdots 9894$	39	-21	0.5
Fm 255	255.0899406	14	$\cdots 9692$	19	-29	-1.5
Fm 256	256.0917016	34	$\cdots 7313$	36	-30	-0.8
No 256	256.0942581	42	$\cdots 2858$	44	-28	0.6
Fm 257	257.0950867	54	$\cdots 1121$	55	-25	-0.5

TABLE VI (Continued)

^a Represents the rms deviation in μ u for the associated mass.

 $^{\rm b}$ The dots indicate that the leading significant figures are omitted.

 $^{c}\Delta = mass$ value given in the Minnesota least-squares fit minus the mass value given in the 1967 Mass Table (in μu). $^{d}\Delta / \sigma$ represents the difference between the mass values from the Minnesota least-squares fit and the 1967 Mass Table divided by the error assigned to that mass in the 1967 Mass Table.

from the 1967 Mass Table compared with results from the 1971 Mass Table:

²³² Th	1967 1971	232.038 079 ± 12 u 232.038 074 ± 11 u
²³⁵ U	1967 1971	$\begin{array}{c} 235.043\ 943 \pm 11\ u\\ 235.043\ 944 \pm 11\ u\end{array}$
²³⁸ U	1967 1971	$\begin{array}{c} 238.050\ 819 \pm 12 \ u\\ 238.050\ 816 \pm 11 \ u \end{array}$

When measured in terms of the magnitude of the disagreement between the Minnesota Mass Table and the 1967 Wapstra Mass Table, the revisions that have occurred in the new table by Wapstra and Gove are small. The disagreements referred to in the text of this paper, therefore, still exist.

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PHYSICAL REVIEW C

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Nuclear Orientation Studies of the Decays of ¹⁸⁷W and ^{185,191,193}Os[†]

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Angular distributions have been measured for γ rays emitted following the decays of ¹⁸⁷W and ^{185,191,193}Os nuclei polarized at low temperatures in iron. The decay of polarized spin-1/2 nuclear levels was found to show isotropic angular distributions to three parts in 10⁴, in keeping with angular momentum theory, and purity of the accepted nuclear spin values. The magnitudes of the magnetic moments of the ¹⁸⁷W and ¹⁹³Os ground states have been deduced to be $(0.688\pm0.021)\mu_N$ and $(1.30\pm0.19)\mu_N$, respectively, assuming saturation of the hyperfine field at the nucleus; the magnitude of the magnetic moment of the ¹⁹¹Ir 171-keV level has been similarly deduced to be $(3.27\pm0.12)\mu_N$, based in part on the observation that the nuclear spin-lattice relaxation time associated with decays from that level is less than 0.1 sec. E2/M1 multipole mixing ratios have been deduced for a number of ¹⁸⁷Re and ¹⁹³Ir γ rays, and the multipole characters of several of the β radiations emitted by ¹⁸⁷W and ¹⁹³Os have been obtained; these multipolarities are discussed in terms of the nuclear structure. The use of polarized ¹⁹¹Os as an absolute γ -ray anisotropy thermometer is discussed.

I. INTRODUCTION

The observation of the angular distribution of radiation emitted by nuclei polarized at low temperatures is a convenient means of investigating fundamental nuclear symmetries as well as of gathering data on nuclear properties such as spins, moments, and radiation multipolarities.¹ We report here an investigation into the γ rays emitted by ¹⁸⁷W and ^{185, 191, 193}Os polarized at $T \sim 20$ mK in iron. The magnetic moments of the ¹⁸⁷W and ¹⁹³Os ground states and the 171-keV ¹⁹¹Ir 5-

sec excited state have been deduced from the observed angular distributions; the latter measurement was confirmed to be characteristic of the ¹⁹¹Ir level rather than of the ¹⁹¹Os parent by our observation that an upper limit of 0.1 sec can be set on the nuclear-spin-lattice relaxation time associated with decays from that level. Mixing ratios of a number of γ rays following the decays of the parent states have been deduced, and multipolarities of the unobserved β -radiation fields have been obtained.

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