Preferred Ratios of Energies of Excited States of Nuclei

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For the lowest three states in the decay schemes of twenty-eight nuclei, the ratio of the larger to the smaller of the intervals between adjacent states is known within ±0.018. Twenty ratios equal fractions with denominators 1 to 5 approximately within experimental uncertainty.

Integral relations exist in the decay schemes of nuclei with two or three level pairs. Energies of the several known lines of seven nuclei are approximately integral multiples of a single energy, different for each nucleus.

THERE exist many numerical regularities in atomic spectra. The energies of levels of hydrogen and the alkali metals are given approximately by simple formulas. For many complex atoms L–S coupling is a good approximation, and energies of the various terms of a single configuration are linear combinations of a small number of Slater integrals with integral coefficients. The further splitting of a term with given L and S is then in good agreement with the simple Landé interval rule.

Energies of many low excited states of nuclei are now well known. They are accurately obtained from γ-ray transition energies. Most of these, in turn, are determined by energies of conversion electrons; some are measured directly in a crystal spectrometer. With modern β-ray spectrographs the conversion electron energies can be measured to a precision of 0.2 percent or better. Most of the data available now are, however, less accurate. The x-ray absorption edges are accurately known and have recently been tabulated.[1] Since calibration of a spectrograph is difficult, the absolute accuracy of the energy of a line is considerably smaller than the accuracy of a ratio between energies of two lines measured by a given instrument.

The decay schemes of about 140 nuclei were juxtaposed in a recent volume of data about nuclei.[2] Even though level energies of nuclei are much less accurately known than those of atoms, it seems tempting to investigate the possibility of numerical relations among them. Three avenues of approach to this problem are considered here.

1. THE FIRST TWO EXCITED STATES IN DECAY SCHEMES OF NUCLEI

The energies of the first two excited states in the decay schemes of nuclei were examined. The final nuclei are produced by β- or α-decay or by a heavy particle reaction leaving them in an isomeric state. For some nuclei the first two excited states observed in this way may actually be the lowest ones. For others, different sets of low states could probably be excited by other reactions.

An excited state is defined to be well identified if its position in the decay scheme is established, for example, by a γ-ray to the ground state and the end-point energies of β-decays to it and to the ground state. For the set of nuclei whose lowest two excited states are well identified, let a denote the larger and b the smaller of the energy differences between adjacent levels among the first three. The ratio $R=a/b$ was calculated for the nuclei in this set found in the Landolt-Börnstein volume, as well as those in a recent table of isotopes[3] and in a review article on nuclear isomerism.[4] The uncertainty in R, $U(R)$, is defined in terms of $U(a)$ and $U(b)$ as

$$U(R)=R\left[\frac{U(a)}{a}+\frac{U(b)}{b}\right].$$

$U(a)$ and $U(b)$ were taken from original papers or estimated from data presented there whenever possible; otherwise, more arbitrary estimates amounting to at least 0.2 percent of a and b were made, and $U(R)$ is enclosed in parentheses in the tables which follow.

The nuclei in this set for which $U(R)\leq 0.018$ are listed in Table I. There are twenty-eight of them. Twenty of the ratios R are fractions with denominators 1 to 5 within 0.020.

It is not very likely that this is a chance occurrence. The probability that such a result be obtained from random distribution of decimals of twenty-eight numbers may be calculated in the following way. Let $R=I+D$, where I is an integer and D is a decimal. If R were exactly equal to a fraction with denominator 1 to 5, then D would equal some d_i, where $d_1=0$, $d_2=0.2$, $d_3=0.25$, $d_4=0.33\cdots$, $\cdots\cdots$, $d_{10}=0.8$. The absolute value of the difference between D and the nearest d_i is denoted δ and listed in Table I. $\delta\leq 0.020$ for twenty ratios, and they lie in an interval of length 0.40; eight others lie in an interval of length 0.60. The subintervals and a histogram of the decimals of the twenty-eight ratios are drawn in Fig. 1. The probability P that at least twenty out of twenty-eight decimals lie in an

[1] Hill, Church, and Mihelich, Rev. Sci. Instr. **23**, 523 (1952).
[2] Landolt-Börnstein, *Zahlenwerte und Funktionen* (Julius Springer, Berlin, 1952), sixth edition, pp. 205–217 (I. Band, 5. Teil).
[3] Hollander, Perlman, and Seaborg, University of California Radiation Laboratory Report 1928. This table, which contains data published by August 1952, was used in the present investigation. A later edition is to be published in Revs. Modern Phys.
[4] M. Goldhaber and R. D. Hill, Revs. Modern Phys. **24**, 179 (1952).

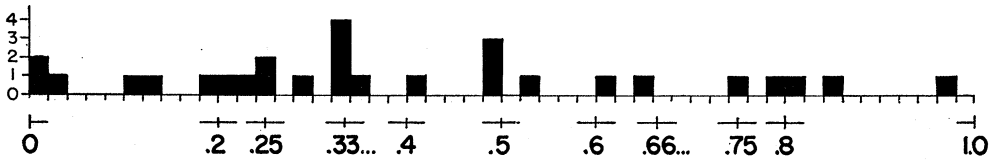

FIG. 1. Histogram of decimals of ratios R. Drawn below it are the intervals from $d_i-0.020$ to $d_i+0.020$.

interval of length 0.40 is given by:

$$P=\sum_{k=20}^{28}\binom{28}{k}0.4^k 0.6^{28-k}=7.6\times10^{-4}.$$

The data indicate only that decimals near some d_i are preferred. It may well be that more accurate data will not yield many decimals closer to the special ones. In fact, for five of the twenty ratios with $\delta\leq0.020$, δ is larger than $U(R)$; for them $U(R)<\delta\leq1.8\cdot U(R)$. It is because accurate integer ratios for all nuclei are not to be expected, and because estimates of experimental uncertainties may be incorrect by a factor of 2 or 3, that the accuracy of an individual ratio was not directly taken into account in the probability calculation.

The distribution of decimals of the ratios in Table I in the intervals determined by the different denominators is given in Table II. Line 1 lists the denominators, z. The statistical weights, s, or total intervals corresponding to denominators 1 to 5 and ≥6 are given in the second line. Denominators larger than 5 cannot be analyzed in a definite way with intervals $d_i-0.02\leq D\leq d_i+0.02$ because of overlapping of such intervals. Line 3 contains $e=28s$. In a random distribution of twenty-eight decimals, let m equal the number of decimals which lie in an interval s. Then e equals the average of m for a large number of such random distributions.

The number of observed ratios in each interval, n, is given in line 4. This number increases as the denominator is increased from 1 to 5; the ratio n/e (line 5), however, decreases monotonically from 2 to 5. The decrease in preference for larger integers is illustrated in a precise way in line 6. $p(n,z)$ is the probability that n or more of the twenty-eight decimals in a random distribution lie in the total interval determined by z. In symbols,

$$p(n,z)=\sum_{k=n}^{28}\binom{28}{k}s^k(1-s)^{28-k}.$$

If rational ratios exist, they may be valid, at least within 0.02, only for some nuclei. Also, larger denominators than 5 may appear; they might account for the four ratios in Table I which do not lie at all near a d_i. There are not sufficient data now available to test such an hypothesis. Some of the ratios which fall near some d_i may do so by accident; for instance, they may actually belong to a denominator larger than 5. Furthermore, some of the decay schemes upon which Table I is based will probably prove to be incomplete or incorrect. That twenty of the twenty-eight decimals of ratios should approximate values of d_i, however, was shown to be unlikely by the probability calculation.

TABLE I. Ratios between energy intervals a and b.

Nucleus	N, Z	Method of excitation[a]	R	$U(R)$ $\times10^3$	δ $\times10^3$	Approximate ratio of integers[b]
Mg^{24}	12, 12	Na β^-	2.001	(9)	1	2/1
Ti^{46}	24, 22	Sc β^-	1.258	13	8	5/4
Ti^{48}	26, 22	Sc β^-, V β^+	1.333[c]	(6)	0	4/3
Fe^{55}	29, 26	Co β^+, EC	1.960	(15)	40	~2/1
Ni^{60}	32, 28	Co β^-	1.137	18		
Ge^{72}	40, 32	Ga β^-	1.323	(16)	10	4/3
Se^{76}	42, 34	As β^-	2.180	17	20	11/5
Br^{80}	45, 35	m	1.325	(13)	8	4/3
Sr^{87}	49, 38	Y β^+, EC	1.244	14	6	5/4
Pd^{106}	60, 46	Rh β^-, Ag β^+, EC	1.216	(17)	16	6/5
Cd^{114}	66, 48	In EC	1.299	4		
Sn^{117}	67, 50	m	1.019	13	19	1/1
Te^{121}	69, 52	m	2.611	11	11	13/5
Te^{123}	71, 52	m	1.795	8	5	9/5
Te^{124}	72, 52	Sb β^-, I β^+	2.814[c]	(16)	14	14/5
Te^{125}	73, 52	m	3.105	14		
Xe^{131}	77, 54	m, I β^-	1.035	13	35	~1/1
Ba^{134}	78, 56	Cs β^-	1.327[c]	14	6	4/3
Pm^{147}	86, 61	Nd β^-	2.488	(15)	12	5/2
Eu^{153}	90, 63	Sm β^-	1.481	(12)	19	3/2
Eu^{155}	92, 63	Sm β^-	2.344	16	11	7/3
Hf^{177}	105, 72	Lu β^-	1.844	4		
Ta^{181}	108, 73	Hf β^-, m	2.534	(13)	34	~5/2
Hg^{197}	117, 80	m	1.232	10	18	5/4
Hg^{198}	118, 80	Au β^-	1.643	10	24	~5/3
Tl^{203}	122, 81	Pb EC	1.758	(18)	8	7/4
Pb^{204}	122, 82	m	2.420	(18)	20	12/5
Pb^{208}	126, 82	Tl β^-	4.492	12	8	9/2

[a] m: The second excited state is metastable; EC: electron capture.
[b] A ~ sign indicates that $\delta\times10^3>20$.
[c] Note added March, 1953: New data indicate that one or more additional states may lie between the ones which were designated first and second excited states here (later edition of reference 3).

2. NUCLEI WITH TWO OR THREE LEVEL PAIRS

A glance at nuclear energy level schemes reveals many pairs of levels. The separation between the two levels of a pair varies with increasing atomic number from about 300 to 20 kev, but it is often only of the order of 1/5th the distance between a pair and the nearest level. For several nuclei two level pairs exist, and there is one nucleus with accurate data and a three-pair scheme.

Let ΔE_l and ΔE_s be the larger and smaller energy

TABLE II. Distribution of denominators of the ratios in Table I.

Denominator	z	1	2	3	4	5	≥6	Total
Statistical weight	s	0.04	0.04	0.08	0.08	0.16	0.60	1.00
Expected number	e	1.12	1.12	2.24	2.24	4.48	16.80	28
Observed number	n	2	3	5	4	6	8	28
n/e		1.79	2.68	2.23	1.79	1.34	0.48	
$p(n,z)$		0.309	0.100	0.069	0.182	0.285	1.000	

TABLE III. Nuclei with two or three level pairs. (The smaller number below each datum is its estimated uncertainty.)

Nucleus	N, Z and method of excitation	ΔE_l and $U(\Delta E_l)$ (kev)	ΔE_s and $U(\Delta E_s)$ (kev)	R and $U(R)$	δ	Approximate ratio of integers	p_k
Sm^{152} [a]	90, 52	244.3	122.0	2.003		2/1	
	Eu EC	0.1	0.2	0.004	0.003		0.08
Ta^{181}	108, 73	135.8	132.9	1.022		1/1	
	Hf β^-	0.4	0.3	0.005	0.022		0.10
Bi^{211}	128, 83	82.9	65.2	1.271		5/4	
	Pb β^-	0.4	0.1	0.008	0.021		0.16
U^{233}	141, 92	28.9	17.4	1.661		5/3	
	Pa β^-	0.1	0.2	0.025	0.006		0.50
U^{233}		40.6	28.9	1.405		7/5	
		0.1	0.1	0.006	0.005		0.12
U^{234} [b]	142, 92	440.9	176.7	2.496		5/2	
	Np EC	1.4	1.1	0.024	0.004		0.48

[a] It is not certain that the decay scheme of Eu^{152} is correct.
[b] The order of lines in the decay scheme is not certain.

differences between the two levels of each of two successive pairs and E the difference in energy between the lower level of the top pair and the higher level of the bottom pair. The ratio $R=\Delta E_l/\Delta E_s$ was calculated for those nuclei in the three summaries[2-4] for which $E>1.6\Delta E_l$. Although there is evidence for about a dozen nuclei with multiple-pair schemes, R is known to within an uncertainty $U(R)$ less than 0.025 for only five of them. They are listed in Table III. The decimals of all six ratios are equal to various d_i (see Part 1) to within 0.022. They lie in an interval of length 0.44; the probability that such a distribution be the result of a random distribution of decimals, calculated as before, is $0.44^6=7.3\times10^{-3}$. For four decimals $\delta<U(R)$; for the other two, however, it is equal to $4.4U(R)$ and $2.6U(R)$. These are much greater discrepancies than any found in the previous section and a calculation which takes into account the uncertainty for each R was made. Column 8 of Table III lists the probability p_k that the decimal of the kth ratio lie within experimental uncertainty of one of the d_i. The probability P that each of some four or more of six decimals assigned a particular p_k lie in an interval of length p_k is given by

$$P=\sum_A p_1p_2p_3p_4(1-p_5)(1-p_6)+\sum_B p_1p_2p_3p_4p_5(1-p_6)+p_1p_2p_3p_4p_5p_6,$$

where the summation A is taken over the 15 four-combinations, and B over the 6 five-combinations of the six subscripts. For the p_k of Table III, $P=0.021$.

The data indicate that the ratios of pair energy intervals of double pair nuclei are nearly equal to fractions with denominators 1 to 5. The three pair intervals of U^{233} are in ratio 3:5:7 within experimental uncertainty.

TABLE IV. γ-ray lines in the spectrum of U^{233}.

Line[a] (kev)	Number of conversion electron lines for the γ-ray	Maximum spread of conversion electron lines (kev)	Line (kev) 5.80 kev	x
28.9[c] (28.7)[b]	4	0.0	4.98±0.00[d]	−1
40.6[c] (40.5)[b]	3	0.1	7.00±0.02	0
58.1[c]	2	0.0	10.02±0.00	1
75.7[c] (75.4)[b]	5	0.1	13.05±0.02	3
87.1[c] (87.0)[b]	5	0.2	15.02±0.03	1
104.5[c]	5	0.2	18.02±0.03	1
272.6	2	0.1	47.00±0.02	0
301.5[c]	3	0.2	51.98±0.03	−1
313.1	4	0.4	53.98±0.07	−1
342.0[c]	4	1.2	58.97±0.21	−2
376.5	2	0.2	64.91±0.03	−5
399.9	3	0.2	68.95±0.03	−3
416.4[c]	3	0.0	71.79±0.00	−12

[a] H. B. Keller and J. M. Cork, reference 5.
[b] C. I. Browne and I. Perlman (unpublished data), Berkeley, 1952. Crystal spectrometer measurements.
[c] Also found by D. G. Karraker (unpublished data), Berkeley, May, 1951. Less accurate β-ray spectroscope measurements.
[d] The numbers which follow the ± signs are based only upon column 3, and thus represent only a part of the total uncertainty.

3. ENERGIES OF ALL OBSERVED γ-RAYS OF SOME NUCLEI ARE APPROXIMATELY INTEGRAL MULTIPLES OF ONE ENERGY

The energies of all of the observed lines of three of the nuclei in Table III are, approximately within experimental uncertainty, integral multiples of a single factor $f(N,Z)$, different for each nucleus. The largest number of lines was observed for U^{233} and its spectrum[5] will be discussed in detail. A scheme with six levels fits the data well. It is drawn to scale in Fig. 2. Thirteen of fifteen possible lines were observed. Their energies are listed in column 1 of Table IV. The number of conversion lines observed and the maximum spread in energy of γ-rays converted in different shells are given in the next two columns. The x-ray absorption edges used in the original paper differ at most by 0.1 kev from the latest values,[1] except for the K absorption edge, which differs by 0.4 kev. Column 4 contains the line energies divided by 5.80 kev. This number leads to the best fit for energy differences between adjacent levels. The uncertainties are based only on the figures in column 3. With two exceptions, the numbers in column 4 are integers ±0.05. There are several groups of equal decimals because the second decimal of the factor happens to be zero. Any

TABLE V. Analysis of the levels of U^{233}.

Level number	Energy assigned to level (kev)	Number of lines +No. of sums of lines used to compute level energy	Maximum spread among lines and sums of lines (kev)	Level energy (kev) / 5.80 kev	Integer assigned to level
0	0			0	0
1	17.4	2	0.2	3.00	3
2	75.7	1	—	13.05	13
3	104.5	3	0.1	18.02	18
4a	376.5	1	—	64.91	
4b	377.1	2	0.1	65.02	65
4[a]	376.9	3	0.7	64.98	
5a	416.4	1	—	71.79	
5b	417.6	4	0.4	72.00	72
5[a]	417.4	5	1.3	71.97	

[a] The energy of this level is the weighted average of the energies of the preceding two levels.

[5] H. B. Keller and J. M. Cork, Phys. Rev. **79**, 1030 (1950).

number given to one decimal, as are the γ-ray energies E_ν, can be expressed as $E_\nu = 5.80[I + (x/58)]$, with I and x integers. x is tabulated in column 5; I was taken as the integer nearest $E_\gamma/5.80$. The maximum possible value of x is 29.

The energy levels are given in Table V. The energy differences between successive levels are in ratio

3.00:10.05:4.97:46.96:7.02.

For the lowest three excited states there is good agreement among the lines and sums of lines which were used to compute the energies of the levels. It is possible that the highest two levels, numbered 4 and 5, may actually be doublets with separations of 0.6 and 1.2 kev. The tentative doublet levels are denoted by a and b. Very narrow doublets, with a ratio of splitting to average level distance of approximately the same magnitude, have been observed in the nuclear spectra of many light elements. Only more accurate and complete data, however, could verify the conjecture of splitting in U^{233}.

TABLE VI. Nuclei for which at least four lines are known and the energies of all lines are approximately integral multiples of one energy.

Nucleus	N, Z and method of excitation	Line a (kev)	$U(a)$ (kev)	$f(N,Z)$ (kev)	$R=\frac{a}{f(N,Z)}$	$U(R)$
Sn^{116}	66, 50	137	1	27.1	5.06	0.04
	In β^-	406	2		14.98	0.07
		1085	5		40.04	0.18
		1274	6		47.01	0.22
		1487	7		54.87	0.26
		2090	10		77.12	0.37
Xe^{131}	77, 54	80.13[a]	0.01	40.3	1.99	0.00
	I β^-	163	2.0		4.04	0.05
		284.1[a]	0.1		7.05	0.00
		364.2[a]	0.1		9.04	0.00
		637	3.0		15.81	0.07
		722	4.0		17.92	0.10
Pr^{144}	85, 59	33.7[b]	0.3	6.71	5.02	0.04
	Ce β^-	53.5[b]	1.0		7.97	0.15
		80.7	0.5		12.03	0.07
		100.3[b]	0.5		14.95	0.07
		134.2	0.5		20.00	0.07
Sm^{152}	90, 62	122.0	0.2	12.20	10.00	0.02
	Eu EC	244.3	0.1		20.02	0.01
		720.4	(1.0)		59.05	(0.08)
		964	1.0		79.02	0.08
		1086	(2.0)		89.02	(0.16)
Ta^{181}	108, 73	132.9	(0.3)	68.2	1.95	(0.00)
	Hf β^-	135.8	(0.4)		1.99	(0.01)
		344.1	(0.7)		5.04	(0.01)
		480.8	(1.0)		7.05	(0.02)
		611.2	(1.2)		8.96	(0.02)
Hg^{199}	119, 80	50.6	(0.3)	52.2	0.97	(0.01)
	Au β^-	157.5	(0.6)		3.02	(0.01)
	m[c]	208.1	(0.7)		3.99	(0.01)
		368	7.0		7.05	0.13

[a] Measured by crystal spectrometer. All others obtained from conversion electron energies.
[b] F. T. Porter and C. S. Cook, Phys. Rev. **87**, 464 (1952). Presumably converted in Pr. Appropriate electron lines from decay of Ce^{144} were not sufficiently distinct to distinguish between Pr and Nd.
[c] Other lines have been reported from Tl EC. H. I. Israel and R. G. Wilkinson, Phys. Rev. **83**, 1051 (1952).

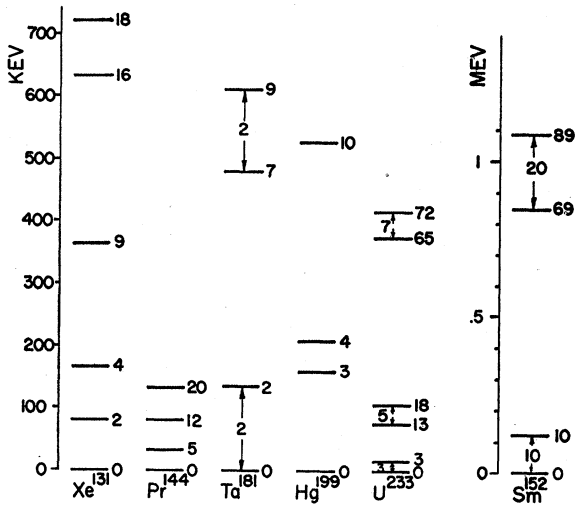

FIG. 2. The energy levels of six nuclei with integer schemes. The energy of each level equals $f(N, Z)$ times an integer, and the level is labeled with this integer.

The decay schemes of about 45 nuclei with four or more lines known to an accuracy of about 2 percent or better were examined. All definitely known lines of U^{233} and six other nuclei listed in Table VI are approximately integral multiples of a single energy. It must be emphasized, however, that for at least some of these nuclei the integral relations may well be accidental. The decay schemes published for six of them are drawn to scale in Fig. 2. Only two of the nuclei meet the criteria for inclusion in Table I.

A few of the most accurately known lines, e.g., three of Xe^{131}, fit an integer scheme, but not by any means to within experimental uncertainty. This is another indication that integer relations are only approximately valid. The deviations from integer schemes, while small for these seven nuclei, may be much larger for others and might be responsible for the fact that such schemes, at least with small integers, are not found for other nuclei.

As early as 1931, Rutherford and Ellis[6] pointed out that energies of excited states of nuclei in the α-decay region are linear combinations of two numbers with integral coefficients. The evidence which they presented is striking, but it indicates such relations among some, not all, of the levels of product nuclei. The level separation energies are small and are not even today known accurately enough to determine whether there exist integer schemes as simple as the ones for some product nuclei of artificial radioactivity.

The possibility that integral relations, especially with small integers, exist only for some of the levels of a nucleus must also be considered. Elliot and Deutsch[7]

[6] E. Rutherford and C. D. Ellis, Proc. Roy. Soc. (London) **A132**, 667 (1931).
[7] L. G. Elliot and M. Deutsch, Phys. Rev. **64**, 321 (1943).

found that energies of five of eight γ-rays in the spectrum of Fe^{56} are proportional to small integers. This appears to be so also for several light nuclei. In B^{10} there is a set of levels with energy ratios 1:3:5 and isotopic spin $T=0$; the state at 1.74 Mev, with $T=1$ does not fit into this scheme.[8] It may be that several series of lines exist in the spectrum of a nucleus as in that of an atom, and that the levels of each series are multiples of a different energy. The data for B^{11} indicate integral relations also, as has recently been pointed out.[9] They do not, however, justify a hypothesis of integral relations among *all* the observed states of this nucleus. The possibility that integral relations appear for other physical quantities has also been put forward.[10]

I am grateful to Professor E. P. Wigner for his advice and encouragement.

[8] Wigner, Ajzenberg, and T. Lauritsen (private communications).

[9] P. J. Grant, Proc. Phys. Soc. (London) **A65**, 150 (1952).
[10] E. E. Witmer, Phys. Rev. **86**, 618 (1952).

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The Half-Life of Co^{60}

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A new determination of the half-life of Co^{60} yields the value 5.21 ± 0.04 years.

RECENTLY Lockett and Thomas[1] published a value for the half-life of Co^{60} of 4.95 ± 0.04 year. This differs markedly from the results of previous measurements,[2,3] most of which lie between 5.2 and 5.3 years. In view of this discrepancy, the value obtained to date from measurements in progress in this Laboratory may be of interest.

The half-life of Co^{60} is being determined by comparing the ionization produced in an ionization chamber of high stability[4] by the gamma-radiation from a 100-mC Co^{60} source with that produced by the gamma-radiation from a radium source of about the same strength. The comparison with radium is made in order to eliminate the effects of any long-range variations in the response of the measuring apparatus. Measurements made at intervals of 6 to 12 months for the past three years show no observable deviation from a simple exponential decay. A least-squares analysis of the data leads to a value for the half-life of

$$T_{\frac{1}{2}}=5.21\pm0.04 \text{ years},$$

where the standard deviation has been calculated from the known reproducibility of measurements made with this apparatus over long periods of time.

The above figure disagrees with the new value found by Lockett and Thomas, but agrees well with what appears to be the most precise of the earlier determinations: the value of 5.27 ± 0.07 years found by Tobailem.[3]

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[1] E. E. Lockett and R. H. Thomas, Nucleonics **11**, No. 3, 14 (1953).
[2] J. J. Livingood and G. T. Seaborg, Phys. Rev. **60**, 913 (1941); E. Segrè and C. E. Wiegand, Phys. Rev. **75**, 39 (1949); G. L. Brownell and C. J. Maletskos, Phys. Rev. **80**, 1102 (1950); W. K. Sinclair and A. F. Holloway, Nature **167**, 365 (1951).
[3] J. Tobailem, Compt. rend. **233**, 1360 (1951).
[4] G. C. Laurence, Can. J. Research **7**, 103 (1932).