The Energy Content of the Heavy Nuclei

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A packing fraction curve is constructed from the atomic masses previously measured and from new measurements for tin, lanthanum and neodymium. The curve is also drawn to agree with the differences in packing fractions for the twenty-five pairs previously reported. The curve has the general shape of Aston's curve, but has a higher minimum value of -7.2×10^{-4} in place of -10×10^{-4} and is from 2×10^{-4} to 3×10^{-4} higher for the heavy elements. For the elements below lead the curve is much less steep than above. This gives an energy content to these elements, such that α-ray emission is impossible. With the values given by the curve for tin and palladium, the differences between the masses of lead and thorium, and lead and uranium are greater than the total mass of the α- and β-rays emitted in the transformations from thorium or uranium to lead, but agree with the differences to be expected if the mass equivalents of the energies of the α- and β-rays are allowed for. The effect of the packing fractions on the computed atomic weights of the elements cassiopeium, neodymium thorium and uranium is discussed. The masses of gold and tin isotopes have been compared, and the masses of neodymium, lanthanum and tin isotopes have been compared with titanium.

THEORETICAL formulae have been given by Weizsäcker,[1] Bethe,[2] and Gamow[3] for the divergence of the atomic masses from integral values. These formulae are based on estimates of the various forces between the particles in the nuclei, thought of as protons and neutrons, and predict a gradual decrease in the packing fractions to a minimum value followed by a gradual increase for the heavy nuclei.

If we take the atomic mass of hydrogen on the physical scale equal to 1.00813, ($0^{16}=16$), and the neutron mass on the same scale[4] as 1.00893, the difference between the atomic mass of an element M, whose nucleus is made up of p protons and n neutrons, and the masses of its constituents is $(M-1.00893n-1.00813p)$. This is the energy content or negative binding energy of 6×10^{23} nuclei in mass units. If $I(=p+n)$ is the nearest integer to M, the packing fraction f is defined[5] as $(M-I)/I$. The energy content of 6×10^{23} nuclei may thus be written $(M-I-0.00893n-0.00813p)$ or $I(f-0.00853-0.0004\times(n-p)/(n+p))$. Per elementary particle in the nucleus, the energy in mass units is given by the bracket. We see that, apart from the uniformly increasing final term, which reaches a maximum value for uranium of -0.9×10^{-4}, the variation in the energy content per particle due to changes in the nuclear structure is given by the packing fraction f.

The packing fraction curve in Fig. 1 is based on the values reported in a recent paper.[6] The determinations referred to oxygen (multiplied by 10^4) were: Ti(48), -7.2 ± 0.2; Fe(56), -7.0 ± 0.4; Cu(63–65), -6.9 ± 0.2; Ag(107–109), -4.9 ± 0.5; Pd(106–110), -5.35 ± 0.6; Pt(195), 2.03 ± 0.3; Au(197), 2.0 ± 0.4 In the case of Cu, Ag, Pd the value given is the mean packing fraction for the two atomic masses. These values are indicated in Fig. 1 by large circles with vertical lines indicating the probable errors.

In addition the curve in Fig. 1 is drawn to agree with the twenty-five differences in packing fractions given in Table II, p. 74 of the former paper,[6] and also with those reported below for Sn, La and Nd. If the value for one mass is taken from the curve, the packing fraction of the second mass is determined by these measurements. These values are indicated in Fig. 1 by a vertical line whose length gives the probable error[7] of the difference measurement. In general this error is less than in the measurements referred to oxygen. Small open circles on the curve indicate the values used in deducing the packing fractions shown by the vertical lines.

No attempt was made to draw the curve for the elements below titanium so as to agree with

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[1] C. F. von Weizsäcker, *Die Atomkerne*, p. 50.
[2] H. A. Bethe, Phys. Rev. **50**, 978 (1936).
[3] G. Gamow, *Structure of Atomic Nuclei*, p. 33.
[4] H. A. Bethe, Phys. Rev. **53**, 314 (1938).
[5] F. W. Aston, *Mass Spectra and Isotopes*, p. 160.
[6] A. J. Dempster, Phys. Rev. **53**, 74 (1938).
[7] Probable errors are given as suggested by R. T. Birge (Phys. Rev. **52**, 241 (1937)) so that the probability of the true value lying in the range indicated is one-half.

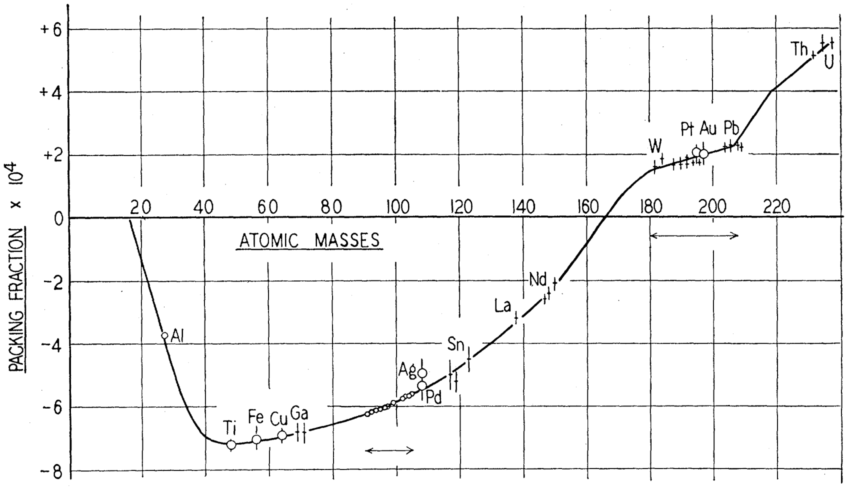

FIG. 1. Packing fraction curve. Comparisons with oxygen are indicated by large circles. Values deduced from differences in packing fractions are indicated by vertical lines. Values assumed in these deductions are indicated by small circles breaking the curve. The lengths of the lines indicate the probable errors. The differences in packing fractions for the ranges indicated by arrows, 91–105 and 182 to 209, are shown in greater detail in Fig. 2.

the values for the lighter elements. Aluminum is shown, as it was used in deducing the values shown for silver and palladium. A plot for the light elements is known to show striking periodic variations[8] with a period of four mass units. For the elements between masses 30 and 40, however, these variations in packing fractions become less prominent and tend to disappear with increasing mass numbers.[9]

The curve drawn in Fig. 1 is not unique; it is, however, the curve with the smallest number of points of inflection that agrees with the data used. It would have been desirable to show the experimental data in relation to a theoretical curve, but none of the theoretical formulae gives the various changes of slope that are required by the experimental data now available.

The curve is seen to have a minimum near mass 50 in agreement with Aston's curve.[10] The minimum value is, however, -7.2×10^{-4} in place of -10×10^{-4}. The three masses near the minimum measured by Aston were those of chromium (52), zinc (64), and nickel (58). For the first two a possible error of 3.0×10^{-4} was estimated, so that his measurements do not diverge from the curve in Fig. 1 by more than his experimental error. For nickel the estimated error was 2.0×10^{-4} in the comparison of Ni (58) and CO_2 (44) by means of the ratio $CH_4 : C$. Aston comments,[11] "it was difficult to get suitable intensities for good comparison." The curve in Fig. 1 also diverges from Aston's curve for the heavy elements in giving packing fractions from 1×10^{-4} to 3×10^{-4} higher than those given by his curve. It crosses the zero axis at atomic mass 166 in place of 200. Aston's observations which gave the main feature of the packing fraction curve for heavy elements were based on measurements of Hg (198), Br (81), and Kr (86), and were made with an instrument with a maximum resolving power of 600 and an accuracy of 1 in 10,000, giving an error of 1.0×10^{-4} in the value of the packing fraction. Recently Aston[12] has redetermined with a new spectrograph the values for Kr(78), Kr(82), Kr(84), Kr(86), and Xe(129) with much greater precision (± 0.10 to ± 0.20) $\times 10^{-4}$. Comparisons were made between doubly and triply ionized atoms of the monatomic gases Kr and Xe, and singly charged hydrocarbon molecules C_3H_3, C_3H_5, C_3H_6 and C_3H_7, formed

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[8] M. L. Oliphant, Nature **137**, 396 (1936).

[9] E. Pollard and C. J. Brasefield, Phys. Rev. **51**, 11 (1937); Nature **137**, 943 (1936).

[10] F. W. Aston, *Mass Spectra and Isotopes*, p. 167.

[11] F. W. Aston, *Mass Spectra and Isotopes*, p. 157.

[12] F. W. Aston, Nature **140**, 149 (1937); Proc. Roy. Soc. **163**, 402 (1937).

from a pentane mixture introduced into the discharge tube. If the atomic mass of carbon is taken[12] as 12.00355, the packing fraction of Xe is -4.46 slightly below the value -4.1 given by the curve in Fig. 1. If we, however, take the value for carbon given by Bainbridge and Jordan[13] 12.00428, the fraction for Xe is -3.94, and if we take their more recent value[14] 12.00398, we find -4.15, in excellent agreement with Fig. 1. Even with this value, however, the fractions deduced for the Kr isotopes are from 0.4 to 0.9×10^{-4} below the curve. Doublets have recently been observed by Mattauch[15] between singly charged strontium atoms at masses 86 and 87, formed by bombardment of strontium bromide salt on the cathode, and charged molecules of SiF_3 formed in the discharge tube. Only one doublet was measured in each case and gave -9×10^{-4} and -8.5×10^{-4}, considerably below the curve in Fig. 1.

THE RANGE TUNGSTEN TO BISMUTH

The fifteen comparisons of atomic masses between 91 and 105, with those between 182 and 209 indicated by arrows in Fig. 1 are shown in greater detail in Fig. 2. Here the observed differences in the packing fractions (f_2-f_1) are given by the lengths of the vertical lines. *It is seen that the differences are practically constant for these atomic masses. Consequently the slope of the packing fraction curve for the smaller masses (91–105) must be twice that for the range covered by the heavy nuclei of twice the mass (182–209).* There must consequently be a point of inflection in the curve between the two. The steep slope shown at the lower end of the radioactive elements is required by the high energy of decay of these elements (see Fig. 3). If an attempt is made to continue this curve to lower masses with a gradually decreasing slope, it can be made to agree only approximately with the values for Au, Sn, Pd, Nd and La. The values for Zr, Mo and Pd must then be made much lower than shown and require the curve to have a point of inflection near mass 105, with a very steep slope between 90 and 105, twice that between 180 and 210, that is, nearly 3×10^{-5}. This sudden rise in the nuclear energy over a short range, making the curve almost as steep as the fall between masses 26 and 36, would be contrary to any present theory of nuclear structure. The alternative shown in Fig. 1 was consequently chosen, which makes a point of inflection occur at mass 208, and which permits a curve with an excellent agreement with the other observations. The comparisons at mass 96 between Os-Mo and Os-Ru suggest that there is very little difference in the packing fractions of these isobars.

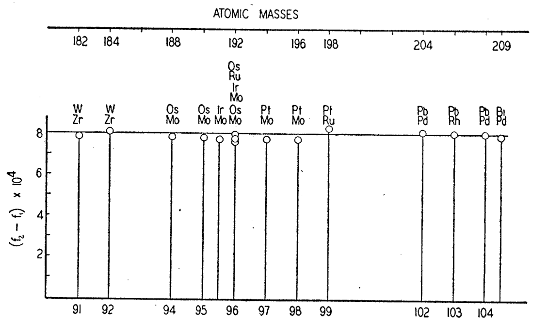

FIG. 2. Differences in packing fractions obtained by comparisons of the atomic masses of the singly charged atoms from zirconium to palladium with the corresponding group of doubly charged atoms from tungsten to bismuth.

ALPHA-RAY STABILITY OF THE HEAVY NUCLEI

If an atom of mass I with a packing fraction f_1 is formed by an α-ray disintegration of an atom of mass $(I+4)(1+f_2)$, then

$$(I+4)(1+f_2)-I(1+f_1)\geqq 4.0039$$

or

$$I(f_2-f_1)+4f_2\geqq 0.0039.$$

For instability the packing fraction curve must thus have a slope greater than that given by

$$I\,df/dI+f=9.7\times 10^{-4}.$$

For $I=206$, $f=2.3\times 10^{-4}$, the critical slope is 3.7×10^{-6}. The slope for the radioactive elements just above lead is 14×10^{-6} showing that they are unstable. For the elements from W to Pb the slope in Fig. 1 is 3×10^{-6} and indicates that the

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[13] K. T. Bainbridge and E. B. Jordan, Phys. Rev. **51**, 384 (1937). In a former paper, Phys. Rev. **53**, 67 (1938), the author pointed out that the lines forming the CH_4-O doublet reproduced by Bainbridge and Jordan, Phys. Rev. **50**, 287 (1936), were not parallel. He wishes to withdraw any implied criticism of these measurements, as the lack of parallelism is not present in the original photographs, but was due entirely to some fault in the reproduction process.

[14] M. S. Livingston and H. A. Bethe, Rev. Mod. Phys. **9**, 373 (1937).

[15] J. Mattauch, Naturwiss. **25**, 170 (1937).

energy content of these nuclei is such that α-rays of finite energy cannot be emitted.

For still lighter elements the formula just given would permit α-ray transformations. For $I=140$, $f=3.2\times 10^{-4}$, the critical slope is consequently 9.2×10^{-6} and the curve in Fig. 1 shows that, since for $I=140$ the slope is 10×10^{-6}, α-ray transformations are possible. Samarium is known to emit α-rays but the other elements are not radioactive. Reasons that may account for the failure to observe α-ray activity in elements between 140 and 180 have been given by Weizsäcker.[16]

Mass Equivalent of Energy in Radioactive Transformations

The curve in Fig. 1 from mass 206 to 238 gives the values of the packing fractions deduced from the maximum energies set free in radioactive transformations. The value for lead is taken as 2.3×10^{-4}. This part of the curve is shown in greater detail in Fig. 3. One striking feature is the occurrence of isobars with different energy content. The slope is less for the heavy radioactive nuclei, since the energy of disintegration is comparatively small at the beginning of a radioactive series and becomes greater for the short lived nuclei, in the latter half of the radioactive series.

The values deduced from the mass comparisons for the packing fractions of thorium, 5.12 ± 0.1, actinouranium, 5.56 ± 0.3, and uranium, 5.56 ± 0.1, agree with Fig. 3. A theoretical curve could also be drawn without allowing for the mass equivalent of the energy radiated. This gives values of 3.06×10^{-4} and 3.3×10^{-6} for thorium and uranium, very far from the fractions deduced from the mass comparisons. The difference would be removed, if the fractions for tin (116 and 119), with which uranium and thorium are compared, were reduced by 2×10^{-4}, or if the fraction for palladium (104), with which lead was compared were increased by 2×10^{-4}. The comparisons of the tin isotopes with titanium exclude values lower by 2×10^{-4} than those deduced in Table I, for if 116 and 119 were lower, the mass at 118 would have to be higher by the same amount, and this is excluded by the uniformity in the spacing of the tin isotopes. Also a fraction for Pd (104) lower by 2×10^{-4} than the value measured for Pd (108) would be very improbable. Unless the packing fractions for the isotopes of palladium fluctuate by 2×10^{-4}, the comparisons show that the mass differences between thorium or uranium and the lead isotopes into which they disintegrate are greater than the total mass of the particles emitted by an amount agreeing with the mass equivalent of the disintegration energy.

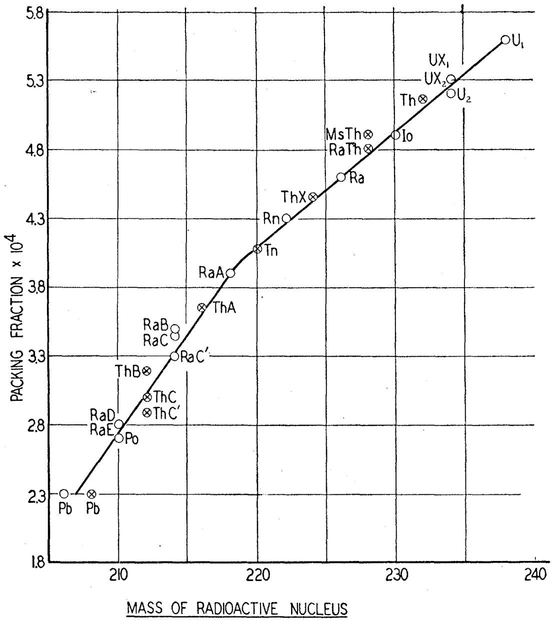

FIG. 3. Theoretical packing fraction curve for the radioactive elements, computed from the energy of disintegration and the mass of helium 4.0039, assuming a value for lead equal to 2.3×10^{-4}.

It is to be expected that the irregularities shown in Fig. 3 would appear in Fig. 1, if the accuracy of the measurements were sufficiently great. It is known, for example, that samarium emits alpha-rays and it is probable that its radioactive isotopes would have an increased mass corresponding to an extra energy content. However even if this extra energy content were 3×10^{6} electron volts, the increase in the atomic mass would be only 0.003 and the increase in the packing fraction only 0.2×10^{-4} which would not be detected in the present experiments. Also in cases of neutron absorption, it is probable that in different cases different fractions of the addi-

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[16] C. F. von Weizsäcker, *Die Atomkerne*, p. 35, 51, 102.

tional mass 1.00897 is accounted for by radiation and by electron or positron emission. We should thus expect an irregularity in the third decimal place of most of the atomic masses. For the heavy elements this requires a precision greater than one part in one hundred thousand in the mass comparisons, if this energy content is to be observed directly.

Atomic Weights of the Heavy Elements

With the packing fraction curve in Fig. 1, the atomic weights of the elements beyond titanium computed from their isotopic structure would all be higher than the values computed from Aston's curve by two or three parts in ten thousand. For thorium the atomic mass given by the packing fraction 5.1×10^{-4} is 232.12. Reduced to the chemical scale the atomic weight is 232.07 in satisfactory agreement with the chemical atomic weight 232.12. The discrepancies in the case of gold[17] and osmium are too large to be accounted for by the change in packing fraction. In neodymium the packing fraction -2.5×10^{-4} in place of -5×10^{-4} assumed by Mattauch and Hauk[18] gives a chemical atomic weight of 144.26 in good agreement with the recent determination[19] 144.27.

A recent determination[20] of the atomic weight of cassiopeium gives a value of 174.98, whereas a packing fraction of -3×10^{-4} and a single isotope gives a value of 174.91. Hönigschmid suggested that this discrepancy could be explained by a new isotope at 177 with an abundance of 2.5 percent. The existence of an isotope at 173 or 177 has been deduced from band spectrum observations[21] but its abundance was estimated at only 1.5 ± 0.5 percent. If, however, we take the packing fraction to be $+1\times10^{-4}$ as given by Fig. 1, the predicted value of the atomic weight with only one isotope at mass number 175 comes out to be 174.97. This is very close to the chemical determination and exact agreement would result with an abundance of only one-half percent for an additional isotope at 177.

For uranium the packing fractions 5.56×10^{-4} and 5.3×10^{-4} give atomic masses of 238.132 for uranium and 235.125 for actino-uranium. If the latter is assumed to constitute 0.4 percent of ordinary uranium, the mean mass is 238.12 which when reduced to the chemical scale gives an atomic weight of 238.06 in excellent agreement with the recent determination of 238.07 by Hönigschmid and Wittner.[22]

New Mass Comparisons for Sn, La and Nd

In addition to the atomic mass comparisons reported in a previous paper,[6] measurements have recently been made of the following masses, which are also plotted in Fig. 1.

Titanium-Neodymium

With electrodes made by packing nickel tubes with a mixture of neodymium and titanium metal (5 : 1), it was found that triply charged ions of the former metal were superimposed on singly charged atoms of the latter. The narrow doublet formed between Nd(150) and Ti(50) was sharply resolved by the mass spectrograph. The mass scale was taken from the separation of the two neodymium isotopes at 148 and 150 assumed to correspond to two mass units. The average of five doublets gave a difference in the packing fractions of Ti and Nd of $(5.27\pm0.1)\times10^{-4}$. If the packing fraction of Ti(50) is taken as $-(7.2\pm0.1)\times10^{-4}$ that of Nd(150) is $-(1.93\pm0.2)\times10^{-4}$.

In addition the triply charged isotopes at $49.33\cdots$ and $48.66\cdots$ formed a bracket about the titanium isotope at 49. The average of nine photographs gave a difference of $(4.6\pm0.1)\times10^{-4}$ between the packing fraction of titanium 49 and the arithmetic mean of the fractions for the two neodymium isotopes at 146 and 148. If we assume $-(7.2\pm0.1)\times10^{-4}$ for the former the mean of the latter two is $-(2.6\pm0.2)\times10^{-4}$.

Furthermore the two titanium isotopes at 49 and 50 formed a bracket about the triply charged isotope at $49.33\cdots$. Nine photographs gave an average of $(4.79\pm0.1)\times10^{-4}$ for the difference between the packing fraction of Nd(148) and

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[17] A. J. Dempster, Nature **136**, 65 (1935).

[18] J. Mattauch and V. Hauk, Naturwiss. **25**, 781 (1937).

[19] O. Hönigschmid and Wittner, Naturwiss. **25**, 701 (1937).

[20] O. Hönigschmid, Naturwiss. **25**, 748 (1937).

[21] H. Gollnow, Zeits. f. Physik **103**, 443 (1936).

[22] O. Hönigschmid and Wittner, Zeits. f. anorg. allgem. Chemie **226**, 289 (1936).

TABLE I. *Differences in packing fractions $f=(M-I)/I$ for different pairs of atoms, m = mass near which measurements were made, n = number of photographs measured. When two masses are given the value of f is a mean for the two masses.*

m	IONS	n	$(f_2-f_1)\,10^4$	ASSUME $f_1\times10^4$	DEDUCE $f_2\times10^4$
50	Nd(150) − Ti(50)	5	5.16 ± 0.1	Ti(50) = −7.2	Nd(150) = -2.04 ± 0.1
49.3	Nd(148) − Ti(49–50)	9	4.79 ± 0.1	Ti(49–50) = −7.2	Nd(148) = -2.41 ± 0.1
49.	Nd(146–148) − Ti(49)	9	4.60 ± 0.1	Ti(49) = −7.2	Nd(146–148) = -2.6 ± 0.1
46.3	La(139) − Ti(46–47)	7	3.97 ± 0.2	Ti(46–47) = −7.2	La(139) = -3.2 ± 0.2
23.5	Sn(116–118) − Ti(47)	5	2.23 ± 0.4	Ti(47) = −7.2	Sn(116–118) = -4.97 ± 0.4
24.5	Sn(122–124) − Ti(49)	2	2.73 ± 0.4	Ti(49) = −7.2	Sn(122–124) = -4.53 ± 0.4
39.4	Au(197) − Sn(118–119)	5	-7.2 ± 0.3	Au(197) = 2.0	Sn(118–119) = -5.2 ± 0.3

the mean of the packing fractions of Ti(48) and Ti(49). Assuming $-(7.2\pm0.1)\times10^{-4}$ for the latter the fraction for Nd(148) is $-(2.4\pm0.2)\times10^{-4}$.

LANTHANUM-TITANIUM

With nickel tube electrodes filled with powdered lanthanum and titanium triply charged ions at 46.33··· were bracketed by the two titanium isotopes at 46 and 47. Seven photographs gave an average difference of $(4.0\pm0.2)\times10^{-4}$ between the packing fraction of lanthanum and the mean of the fraction for the two titanium isotopes. If the latter is taken as $-(7.2\pm0.1)\times10^{-4}$, the packing fraction of lanthanum is $-(3.2\pm0.3)\times10^{-4}$.

TIN-TITANIUM

With a spark between electrodes made by packing gold tubes with a mixture of tin and titanium (20 : 1), tin ions with five charges at masses 23.2, 23.4, 23.6, 23.8 were superimposed on doubly charged titanium ions at 23.5. The lines at 23.2 and 23.6 were measured and five photographs gave an average of $(2.23\pm0.4)\times10^{-4}$ for the difference in the packing fraction of Ti(47) and the mean of the fractions for Sn(116) and 118. If the former is taken as $-(7.2\pm0.1)\times10^{-4}$, the latter is $-(5.0\pm0.5)\times10^{-4}$.

The two tin isotopes at 122 and 124 also gave lines at 24.4 and 24.8 on either side of the doubly charged titanium isotope 49. Two photographs gave $(2.73\pm0.4)\times10^{-4}$ as the difference in the packing fractions. If the fraction for Ti(49) is assumed to be $-(7.2\pm0.1)\times10^{-4}$, the mean for the two tin isotopes at 122 and 124 is $-(4.5\pm0.5)\times10^{-4}$.

Gold ions with five charges at mass 39.4 appeared between two tin isotopes with three charges at 39.33··· and 39.66···. Five plates gave a difference of $(7.2\pm0.2)\times10^{-4}$ between the packing fraction for gold and the mean of the fractions for the two tin isotopes at 118 and 119. If the former is taken to be $(2\pm0.4)\times10^{-4}$ the latter is $-(5.2\pm0.6)\times10^{-4}$.

The agreement of these three different comparisons (two with Ti, and one with Au) suggests that there is no large systematic error.

The summary of the values for the differences in the packing fractions is given in Table I.

The author is indebted to Mrs. G. Monk for carrying out computations in connection with the measurements and to Dr. A. E. Shaw for assistance in making electrodes and in preparing the figures in this paper.