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On Closed Shells in Nuclei*

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Experimental facts are summarized to show that nuclei with 20, 50, 82, or 126 neutrons or protons are particularly stable.

IT has been suggested in the past that special numbers of neutrons or protons in the nucleus form a particularly stable configuration.[1] The complete evidence for this has never been summarized, nor is it generally recognized how convincing this evidence is. That twenty neutrons or protons (Ca^{40}) form a closed shell is predicted by the Hartree model. A number of calculations support this fact.[2] These considerations will not be repeated here. In this paper, the experimental facts indicating a particular stability of shells of 50 and 82 protons and of 50, 82, and 126 neutrons will be listed.

I. ISOTOPIC ABUNDANCES

The discussion in this section will be mostly confined to the heavy elements, which for this purpose may be defined as those with atomic number greater than 33; selenium would be the first "heavy" element. For these elements, the isotopic abundances show a number of striking regularities which are violated in very few cases.

(a) For elements with even Z, the relative abundance of a single isotope is not greater than 60 percent. This becomes more pronounced with increasing Z; for $Z>40$, relative abundances greater than 35 percent are not encountered. The exceptions to this rule are given in Table I.

(b) The isotopic abundances are not symmetrically distributed around the center, but the light, neutron-poor isotopes have low abundances. The concentration of the lightest isotope is, as a rule, less than 2 percent. The exceptions to this rule are listed in Table II.

It is seen that the violations of these two regularities occur practically only at neutron numbers 50 and 82. Only the case of ruthenium in Table II, which is not a very pronounced exception, does not fall into one of these groups.

The case of samarium, where the lightest isotope has an isotopic abundance of 3 percent, is only a bare violation of the rule and may not seem striking. However, what is extraordinary, the next heavier even isotope of samarium, Sm^{146} with 84 neutrons, which one would expect to find in greater concentration, does not exist at all.

II. NUMBER OF ISOTONES

Figures 1 and 2 reproduce the parts of the table by Segrè in the region of nuclei with 50

* This document is based on work performed under Contract Number W-31-109-eng-38 for the Atomic Energy Commission at the Argonne National Laboratory. Submitted for declassification on February 13th, 1948.

[1] W. Elsasser, J. de phys. et rad. 5, 625 (1934).

[2] E. Wigner, Phys. Rev. 51, 947 (1937); W. H. Barkas, Phys. Rev. 55, 691 (1939).

TABLE I. Even nuclei with $Z>32$ with isotopic abundance greater than 60 percent.

Element	Abundance in percent	Number of neutrons
Sr^{88}	82	50
Ba^{138}	71.66	82
Ce^{140}	90	82

TABLE II. Lightest isotopes of elements with even $Z>32$ with isotopic abundance greater than 2 percent.

Element	Abundance in percent	Number of neutrons
Zr^{96}	48	50
Mo^{92}	15.5	50
Ru^{96}	5	52
Nd^{142}	25.95	82
Sm^{144}	3	82

and 82 neutrons, respectively. For 82 neutrons, there exist seven stable nuclei, which, for convenience, shall be called isotones. For neutron number 50 there exist six naturally occurring isotones, of which one, Rb^{87}, is β-active, however, with a lifetime of 10^{11} years and a maximum β-energy of 0.25 Mev. The average number of isotones for odd neutrons number is somewhat less than one; the same number for even N varies as a rule between three and four. The greatest number of isotones, attained only once in the periodic table, is seven for neutron number 82; six isotones are encountered once only, and for neutron number 50. Five isotones are found five times, namely, for $N=20$, 28, 58, 74, and 78. The frequency of $N=28$ is probably due to the stability of Ca^{48}, with 20 protons, that of $N=74$ to the stability of Sn^{124}, with 50 protons. As few as two isotones for even N are found only three times for heavy nuclei, namely, for neutron numbers 84, 86, and 120.

III. THE SLOPE OF THE CENTER AND THE EDGES OF THE STABILITY CURVE

In the case of neutron number $N=82$ two isotones of odd Z are found, La and Pr. The same is the case for $N=50$, where the unstable but long-lived Rb^{87} and Y^{89} differ only in proton number. Only one other case where nuclei of different odd Z have the same number of neutrons is encountered in the periodic table, namely, that of Cl^{37} (abundance 24.6 percent) and K^{39} (abundance 93.3 percent); this is the case of 20 neutrons. The case of 82 neutrons is most pronounced, since the La and Pr isotopes in question have isotopic abundances of 100 percent.

As Fig. 2 shows, the isotones Nd^{142} and Sm^{144} are both the lightest isotopes of their respective elements. Here, the limit of the stability for neutron-poor isotopes stays at constant neutron number. Exactly the same is true for $N=50$ (Fig. 1). This situation does not occur anywhere else in the periodic table.

The limit of stability for neutron-rich isotopes also stays at constant neutron number for $N=50$ and $N=82$, namely, the pairs of isotones, Kr^{86}, Sr^{88} and Xe^{136}, Ba^{138} are the heaviest isotopes of their elements. Such a case is encountered once more in the periodic table: Ca^{48} and Ti^{50} are the heaviest isotopes of their respective elements and have the same neutron number $N=28$.

IV. THE CASE OF 20 AND 50 PROTONS

Ca, with 20 protons, has five isotopes, which is not too unusual for this region of the periodic table. The difference in mass number between its heaviest and lightest isotope is eight mass numbers, which is quite outstanding, since this difference does not exceed four for elements in this neighborhood.

Sn, $Z=50$, has without exception the greatest number of isotopes of any element, namely, 10. Its heaviest and lightest nuclei differ by 12 neutrons. Such a spread of isotopes is encountered in only one other case, namely, at Xe, where it may be attributed to the stability of Xe^{136} with 82 neutrons.

Incidentally, the next largest difference, 10, in mass number between heaviest and lightest isotope of an element, is encountered once only, in samarium, and may be attributed to the unusual stability of Sm^{144} with 82 neutrons.

V. THE CASE OF 82 PROTONS AND 126 NEUTRONS

Lead, $Z=82$, is the end of all radioactive chains. It has only four stable isotopes, of which the heaviest one, Pb^{208}, has 126 neutrons.

Evidence for the stability of 82 protons and 126 neutrons can be obtained from the energies of radioactive decay. If, for constant value of the charge of the resultant nucleus the energies of α-decay are plotted against the neutron num-

										Gd^{152} 0.2		Gd^{154} 1.5
										Eu^{151} 49		Eu^{153} 50.9
				Sm^{144} 3			Sm^{147} 17	Sm^{148} 14	Sm^{149} 15	Sm^{150} 5		Sm^{152} 26
				Nd^{142} 25.95	Nd^{143} 13	Nd^{144} 22.6	Nd^{145} 9.2	Nd^{146} 16.5		Nd^{148} 6.8		Nd^{150} 5.95
				Pr^{141} 100								
Ce^{136} <1		Ce^{138} <1		Ce^{140} 90		Ce^{142} 10						
				La^{139} 100								
Ba^{134} 2.43	Ba^{135} 6.59	Ba^{136} 7.81	Ba^{137} 11.32	Ba^{138} 71.66								
Cs^{133} 100												
Xe^{132} 26.96		Xe^{134} 10.54		Xe^{136} 8.95								
Te^{130} 33.1												
$N=78$	79	80	81	82	83	84	85	86	87	88	89	90

FIG. 1.

ber of the resultant nucleus, a sharp dip in energy is encountered when N drops below 126, indicating a larger binding energy for the 126th neutron. From these considerations, Elsasser[1] estimates the discontinuity in neutron binding energy at 126 neutrons to be 2.2 Mev or larger, the discontinuity in proton binding energy at $Z=82$ to be 1.6 Mev. These relations have been studied in detail by A. Berthelot.[2a]

VI. ABSOLUTE ABUNDANCE

Absolute abundances are notoriously uncertain. The best estimates are probably contained in the book by Goldschmidt.[3] For the light elements, the abundances vary erratically; for heavy elements, from about Se on, they remain roughly constant. In the region of heavy elements, the following abundance peaks are apparent. At Zr (50 neutrons), at Sn (50 protons); at Ba (82 neutrons), at W and at lead (82 protons or 126 neutrons). In Goldschmidt's plot of abundance against neutron number, page 127, the Zr and Ba peaks are seen to be at neutron number 50 and 82 and become much more pronounced and narrow, whereas the peak at Sn, $Z=50$, as well as the peak at W, become much broader than in the plot against Z.

Most trustworthy among absolute abundances is probably the relative abundance of the rare earths, since these are not likely to have been appreciably fractionated in the earth's crust. The case of 82 neutrons falls just on the edge of this region. According to Goldschmidt's data on the abundance of rare earths in eruptive rocks (which are probably more reliable chemical analyses than the abundance in meteorites), the

[2a] A. Berthelot, J. de phys. et rad. (8) **3**, 17, 52 (1942).
[3] V. M. Goldschmidt, *Geochemische Verteilungsgesetze der Elemente* (Norske Videnskaps Akademi, Oslo, 1938), Fig. 1, page 117, or Fig. 2, page 118.

						Ru^{96} 5		Ru^{98} 2.2	Ru^{99} 12	Ru^{100} 14	Ru^{101} 22	Ru^{102} 30
				Mo^{92} 15.5		Mo^{94} 8.7	Mo^{95} 16.3	Mo^{96} 16.8	Mo^{97} 8.7	Mo^{98} 25.4		Mo^{100} 8.6
						Cb^{93} 100						
				Zr^{90} 48	Zr^{91} 11.5	Zr^{92} 22		Zr^{94} 17		Zr^{96} 1.5		
				Y^{89} 100								
Sr^{84} 0.56		Sr^{86} 9.86	Sr^{87} 7.02	Sr^{88} 82.56								
		Rb^{85} 72.3		Rb^{87} 27.7								
Kr^{82} 11.53	Kr^{83} 11.53	Kr^{84} 57.1		Kr^{86} 17.47								
Br^{81} 49.4												
Se^{80} 48.0		Se^{82} 9.3										
$N=46$	47	48	49	50	51	52	53	54	55	56	57	58

FIG. 2.

abundances of rare earths heavier than samarium are reasonably constant, except that the elements with even Z are about 5.7 times as abundant as those with odd Z. Of the lighter rare earths, however, praesodymium ($N=82$) is about 8 times and lanthanum ($N=82$) about 27 times as abundant as the average of the odd rare earth with greater Z. Nd, with a 26 percent isotopic composition of isotopes with $N=82$, is about five times as abundant; cerium, with 90 percent composition of isotopes with $N=82$, is about twelve times as abundant as the average of the heavier even rare earths. In the composition of meteors the differences are not quite as striking, but still very pronounced.

VII. DELAYED NEUTRON EMITTERS

If 50 or 82 neutrons form a closed shell, and the 51st and 83rd neutrons have less than average binding energy, one would expect especially low binding energies for the last neutron in Kr^{87} and Xe^{137}, which have 51 and 83 neutrons, respectively, and the smallest charge compatible with a stable nucleus with 50 or 82 neutrons, respectively. It so happens that the only two delayed neutron emitters identified are these two nuclei.[4]

The fission products Br^{87} ($N=52$), as well as I^{137} ($N=84$), have not enough energy to evaporate a neutron, and undergo β-decay; in the resultant nuclei, Kr^{87} and Xe^{137}, the binding energy of the last neutron is small enough to allow neutron evaporation.

VIII. ABSORPTION CROSS SECTIONS[5]

The neutron absorption cross sections for nuclei containing 50, 82, or 126 neutrons seem all to be unusually low. This is seen very clearly in the measurements of Griffiths[6] with Ra γ-Be neutrons, and those of Mescheryakov[7] with neutrons from $a(d,d)$ reaction. These measurements extend from mass number 51 to 209. In general,

[4] A. H. Snell, Y. S. Levinger, E. D. Meiners, Jr., M. B. Sampson, and R. G. Wilkinson, Phys. Rev. **72**, 545 (1947).

[5] The author is indebted to Dr. Katherine Way, who pointed out the connection of the closed shells with neutron absorption cross sections.

[6] J. H. E. Griffiths, Proc. Roy. Soc. **170**, 513 (1939).

[7] C. R. Mescheryakov, C. R. U.S.S.R. **48**, 555 (1945).

the cross sections increase with increasing mass number. Griffiths investigates, of the nuclei in question, yttrium (89) with 50 neutrons and lanthanum and praesodymium with 82 neutrons. The activation cross section for yttrium is the smallest he observes for any element; it is about 20 to 30 times smaller than the cross sections in that region of mass number. There is a very pronounced dip of cross sections for lanthanum and praesodymium; the cross section of Pr^{141} is about one-seventh of the average of this region, and that of La^{139} is still smaller by a factor 3. Mescheryakov investigates, among others, La, Pr, barium (138), and bismuth (209). He finds a similar dip at La and Pr, and finds that the cross section of Ba^{138} with 82 neutrons is even lower, namely, less than 0.03 of that of lanthanum. The cross section of bismuth with 126 neutrons is even smaller. The only other unusually small cross section which Griffiths finds is that of thallium (122 or 124 neutrons), which is about the same as that for praesodymium.

Recent experiments by Hughes[8] with fission neutrons have shown exceptionally low neutron absorption cross sections for Pb^{208}, Bi^{209} (126 neutrons) and for Ba^{136} (82 neutrons).

IX. ASYMMETRIC FISSION

It is somewhat tempting to associate the existence of the closed shells of 50 and 82 neutrons with the dissymmetry of masses encountered in the fission process. U^{235} contains $143=82+50+11$ neutrons. It appears that the probable fissions are such that one fragment has at least 82, one other at least 50, neutrons.

X. THEORETICAL ESTIMATE OF THE DISCONTINUITY IN BINDING ENERGIES

It is possible to make an estimate of the change in neutron binding energy at, for instance, 82 neutrons. There exists the semi-empirical formula for the mass of an atom[9] with mass number A and charge Z.

$$M_{A,Z}=A-0.00081Z-0.00611A+0.014A^{\frac{2}{3}}+0.083(A/2-Z)^2A^{-1}-0.000627Z^2A^{-\frac{1}{3}}+\delta; \quad (1)$$

with $\delta=0$ for A odd, $\delta=-0.036A^{-\frac{3}{4}}$ for A even, Z even, $\delta=+0.036A^{-\frac{3}{4}}$ for A odd, Z odd. For odd A, this formula permits the calculation of the value of Z for which the energy is a minimum. For Z less than 50 and for neutron numbers greater than 82, the calculated curve is in good agreement with the position of, for instance, the nuclei of odd Z. Between $Z=50$ and $N=82$, however, the experimental values of Z seem to be below the theoretical curve. The disagreement can be explained by a definite shift of the stability line at 82 neutrons. This shift of the stability line can be explained by a change in binding energy of about 2 Mev. Also, according to the formula (1), xenon (136), with 82 neutrons, should be unstable by about 2 Mev, whereas it is undoubtedly stable; Sm^{144} should be unstable against K-capture by 0.6 Mev, whereas Ba^{140}, with 84 neutrons, which is unstable, would be just stable according to formula (1).

Whereas these calculations are undoubtedly very uncertain, they may serve as an estimate of the order of magnitude of the discontinuity in the binding energies. Since the average neutron binding energy in this region of the periodic table is about 6 Mev, the discontinuities represent only a variation of the order of 30 percent. This situation is very different from that encountered at the closed shells of electrons in atoms where the ionization energy varies by several hundred percent. Nevertheless, the effect of closed shells in the nuclei seems very pronounced.

[8] D. J. Hughes, private communication.

[9] N. Bohr and J. A. Wheeler, Phys. Rev. **56**, 426 (1939); G. B. von Albada, Astrophys. J. **105**, 393 (1947).