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## 78. ON NUCLEAR ENERGETICS AND BETA-ACTIVITY\*

M. N. Saha and A. K. Saha

(Nature, 158, 6, 1946)

Ever since the discovery by Joliot and Curie of the phenomenon of induced radioactivity in 1934, a very large number of radioactive nuclei, exceeding 450, has been prepared in the laboratory. In recent years, new types of radioactive nuclei have been obtained from fission of heavy nuclei like 92U, 90Th and 91Pa, and possibly also of 93Np and 94Pu; which have, however, not yet been released for publication. Extensive studies of the  $\beta$ - and B+ activities of these nuclei have been made in the various laboratories of the world. The data so far collected, though they have reached vast magnitudes, are by no means sufficient, but already they form a rather bewildering mass (for example, see tables by Seaborg<sup>1</sup>), reminding one of the vast collection of spectroscopical data, before the rise of the modern theories of the electron-structure of the atom reduced them to a few simple laws like Pauli's exclusion principle.

In addition to the nuclei like C<sup>14</sup> which have been prepared in the laboratory, we have nuclei, mostly stable, occurring in Nature the number of which now reach nearly 250. In the case of nuclei derived from fission, the designation of a new isotope by mass is occasionally not quite unambiguous; indeed, this is a very acute problem for fission products.

Several attempts at a regularization of data have been made by previous workers, but here we shall refer to a chart prepared by Saha, Sarkar and Mukherjee<sup>2</sup>, a section of which is shown in Fig. 1; the full chart is too large to be reproduced here. This is a synthesis of several charts

already published by different authors. In this, the abscissa represents mass-number M, the ordinate represents the isotope number  $I=\mathcal{N}-Z$ , which represents the excess of neutrons over protons in any nucleus. M ranges from 1 to 239, and I from -1 to about 54. The section of the chart reproduced here extends from I=-1 to I=8. We have attempted to make the chart as up to date as possible. The parallel lines at 45°, henceforth to be called the Z-lines, represent the atomic number Z. Thus all isotopes of the element Z are to be found on the same Z-line. Each isotope is represented by a circle. Solid circles, , represent 'stable nuclei'. Hollow circles, ô, with an arrow pointing upwards represent  $\beta$ +- (positron)-emitting nuclei. When the arrow points down, o, it indicates that the nucleus is  $\beta$ --(electron)-emitting. Circles with arrow pointing both up and down, \$\dip\$, indicate that the nucleus emits both nega trons and positrons, for example, Cu<sup>64</sup>. <sup>†</sup> denotes that the nucleus decays by K-capture only. A denotes that the nucleus decays by K-capture as well as by positronemission. 9 denotes that the nucleus has been obtained in 'fission'; such nuclei are all  $\beta$ --emitting. If any particular isotope has two different half-lives (isomers), both half-lives are given (compare Co<sup>62</sup>). Saha, Sirkar and Mukherjee<sup>2</sup> gave the following rules of stability, some of which were previously known:

Rule 1: I is even.

When I is even, say 4, we get alternation of stable and  $\beta$ -active nuclei, as shown below:

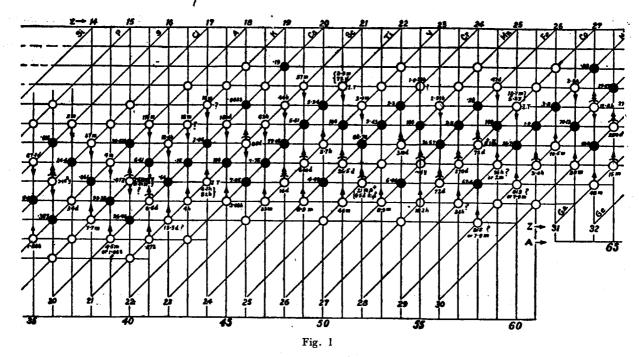
Z: 15\* 16 17 18 19 20 21 22 23 24 25 26 27

p<sup>34</sup> S<sup>36</sup> K<sup>38</sup> A<sup>40</sup> K Ca<sup>44</sup> Sc<sup>46</sup> Ti<sup>48</sup> V<sup>50</sup> Cr<sup>52</sup> Mn<sup>54</sup> Fe<sup>56</sup> Co<sup>58</sup>

— S — S — S — S — S — S — S —

<sup>\*</sup>Summary of a paper by M. N. Saha and A. K. Saha, "On Nuclear Energetics and  $\beta$ -Activity," Trans. Nat. Inst. Sci. India, 2, 193 (1946.)

0.3



S denotes stable, — denotes negatron-emitting, + denotes positron-emitting.

Exceptions. There are certain exceptions:

(a) He<sup>6</sup>, B<sup>10</sup>, C<sup>14</sup> are  $\beta$ -active, though both N and Z are even. (b) H<sup>4</sup>, Li<sup>6</sup>, B<sup>10</sup>, N<sup>14</sup> are stable, though both N and Z are odd. These rules have been given by Bethe and others.

Rule 2. I is odd, say 5: We have

(-) denotes negatron-emitting, (+) denotes positronemitting.

The above is illustrative of all groups having I odd. If in any of the odd groups we arrange the nuclei in order of their mass numbers Z, we first get  $\beta$ --emitting nuclei, then a succession of stable nuclei, which are followed by  $\beta$ +-emitting nuclei.

These rules apply only to known nuclei. If in future it be possible to extend the series on both flanks, will the new nuclei follow these rules? We need not consider the nuclei with odd I because evidently rule 2 will continue to apply to them. Let us consider nuclei with I even: they may be illustrated by a typical group, say I=4; we may think of the possibility of forming some day in the laboratory  $_{12}\text{Mg}^{28}$  and  $_{14}\text{Si}^{32}$ , on the left flank, and  $_{32}\text{Ge}^{68}$ ,  $_{34}\text{Se}^{72}$ , on the right flank, though these are at

present unknown. Will they be stable, or unstable like  ${}_{6}C^{14}$  with a long life?

An explanation to these rules, and a clue to the energetics of  $\beta$ -emission, is afforded in a general way if we write out the formula for mass-defect of nuclei in the form  $\triangle M(Z,A) = \varphi(Z,A) + \chi(Z,A)$ , where  $\varphi(Z,A)$  is the spin-independent part, which is given by one of the various mass-defect formulae due to Weizsäcker, Bethe or Wigner. We have taken, for the sake of simplicity, the Weizsäcker-

Bethe form 
$$\varphi(Z,A) = \alpha A - \beta \frac{I^2}{A} - \gamma A^{\frac{1}{2}} - \frac{\delta Z^2}{A^{\frac{1}{2}}}$$
 (1)

with Bethe's values of the constants  $\alpha$ ,  $\beta$ ,  $\gamma$ ,  $\delta$ . These values were based on older data on masses of nuclei, but probably the newer data will give better values. But these have not been available to us. We have sometimes adjusted the value of  $\beta$ , but this is an unsatisfactory procedure from the theoretical point of view. The Weizsäcker-Bethe formula holds only for Z even, N even, for which the nuclear spin i=0, and the spin-dependent term  $\chi(Z, A)=0$ .

## THE SPIN-DEPENDENT TERM

When I is even and Z odd,  $\mathcal{N}$  odd, for example, in  ${}_{7}\mathrm{N}^{16}$ , we have found uniformly that  $\chi(Z, A)$  is negative and of the order of a few million electron-volts. For odd I, no general principle has yet been found.

From the general mass-defect formula, expressions for the energy-release in  $\beta$ -transitions have been worked out. Let  $E^-$  be total energy-release in a  $\beta$ --transition in

which a nucleus  $zM^A$  changes to  $z_{+1}M^A$ ; then it can be easily shown that

$$E^- = A^- + \chi(Z + 1, A) - \chi(Z, A),$$

where

$$A^{-} = 0.766 + \frac{4\beta(I-1)}{A} - \frac{\delta(A-I+1)}{A^{\frac{1}{2}}}$$
in Mev. (2)

The  $\chi$ -functions may be multi-valued, corresponding to the different nuclear levels of both parent and daughter nuclei. For  $\beta$ +-emission, let us denote by E+ the total energy-release in a  $\beta$ +-transition when the nucleus  $zM^A$  changes to  $z_{-1}M^A$ . It can be easily shown that

$$E^+ = A^+ + \chi(Z - 1, A) - \chi(Z, A),$$

where  $A^+$  is the spin-independent part given by

$$A^{+} = -1.788 - \frac{4\beta(I+1)}{A} + \frac{\delta(A-I-1)}{A^{\frac{1}{4}}} .$$
 (3)

For K-capture, the energy  $E^K$  released in the process, which is taken to be carried by the neutrino, is given by

$$E^{K} = E^{+} + mc^{2} \{1 + \sqrt{1 - c^{2}Z^{2}}\} \simeq E^{+} + 2mc^{2}$$
 (4)

## ENERGY-RELEASE

In cases where the  $\beta$ -transitions are of a simple allowed type, the energy-release is simply equivalent to the endenergy of the  $\beta$ -spectrum, for in general in such cases no  $\gamma$ -rays are emitted. Where such is not the case, but  $\beta$ -rays are of the forbidden type, for example, in Na<sup>24</sup>, and  $\gamma$ -rays are also emitted, the task of finding the energy-release from the  $\beta$ -and  $\gamma$ -radiations emitted is a rather

difficult one, and every case has to be dealt with separately. A typical case is afforded by  $Na^{24}$ , which has been discussed by Krüger and Ogle<sup>3</sup> and by Maier-Leibnitz<sup>4</sup>. In such cases we have to examine with great care the decay scheme of the  $\beta$ -transitions as given by different authors; but this is not easy, for the data are very often contradictory.

According to these formulae, the values of  $E^-$ ,  $E^+$  and  $E^K$  depend on I and A, and on the spin-dependent terms  $\chi(Z, A)$ . The classification of nuclei according to I-values is therefore justified.

It is obvious that a nucleus is stable with respect to  $\beta$ --emission when  $E^-$  is negative, and with respect to  $\beta$ +-emission when  $E^+$  is negative. But for the right hand nuclei of the nuclear chart, even when  $E^+$  is negative, the nuclei may decay by K-capture, provided  $E^K$  is greater than 0, or  $E^+$  is greater than  $-2 mc^2$ , which is approximately 1 Mev. The line of stability is therefore depressed by -1 Mev. below the zero-line, for the right-hand nuclei.

The operation of these rules is best illustrated by taking one or two typical examples for I odd and I even separately. We first take I odd, and choose the group I=3. This is illustrated in Fig. 2. Here the abscissa represents the mass number M, the ordinate represents  $A^-$  and  $A^+$ , calculated from formulae (2), (3). The known nuclei extend from  $O^{19}$  to  $Ga^{65}$ . If the  $\chi$ -terms contributed nothing to  $E^-$  or  $E^+$ , all nuclei for which  $A^-$ -values were below the zeroline, and  $A^+$ -values below the line (-1) would have been stable, namely, from  $S^{35}$  to  $Ti^{47}$ , and the energy-release would be given by the values of  $A^-$  and  $A^+$  against each mass number.

Actually we have all nuclei from Cl<sup>37</sup> to Ti<sup>47</sup> stable, with the exception of A<sup>39</sup>, about which we have extremely meagre data and probably wrong information. S<sup>35</sup> is very

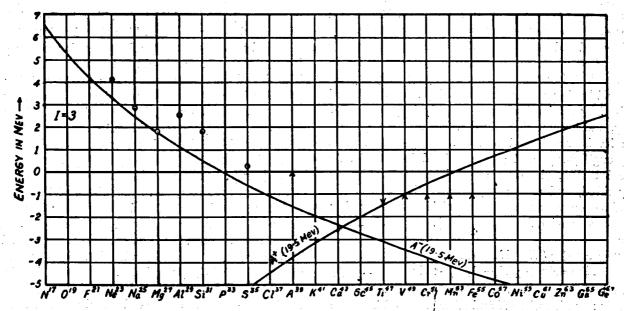


Fig. 2

nearly stable—it has a long life of 87,5 days. We have for S35,

$$E^- = A^- + \chi(18, 35) - \chi(17, 35).$$

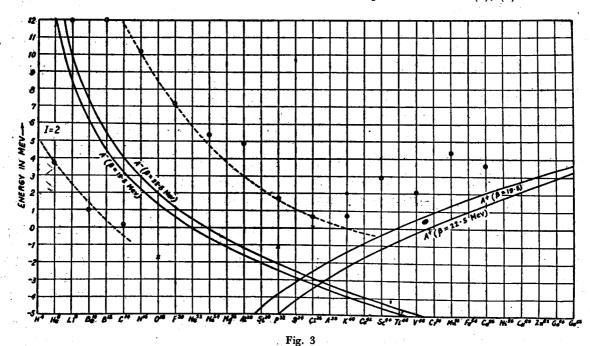
The  $A^-$  is below the zero line,  $\chi(18, 35) - \chi(17, 35)$  is evidently positive, and these terms take  $E^-$  slightly above the zero line.  $E^-$  is only 0.103 Mev. according to the measurements of one of us (A. K. Saha), but if there is a  $\gamma$ -ray emitted by S<sup>35</sup> in cascade, as claimed by Giebert et al.<sup>5</sup>,  $E^-$  is 0.233 Mev.

2. Odd-odd nuclei (N odd, Z odd). For such nuclei we have, since Z is odd,

$$E^{-} = A^{-} - \chi(Z, A) > A^{-},$$
  
 $E^{+} = A^{+} - \chi(Z, A) > A^{+}$  (6)

The operation of these rules is illustrated by the case I=2.

The known nuclei for I=2 range from He<sup>6</sup> to Ga<sup>64</sup> and the operation of rules (2), (3) is illustrated in Fig. 3.



The energy-release in the case of  $\beta$ --active and  $\beta$ +-active nuclei generally follows the  $A^-$  and  $A^+$ -curves. The ringed points give the actual values, wherever available. On the right-hand side, however, data are generally very uncertain; but whatever is available appears to be in agreement. V<sup>49</sup> is nearly stable, decaying only by K-capture and having a long life of 600 days. As we proceed outwards the nuclei release progressively larger amounts of energy, and become short-lived. When E is large, intermediate levels are formed and the energy-release takes place through  $\beta$ -rays and  $\gamma$ -rays emitted in cascade. As mentioned earlier, the data on such cases have to be carefully sifted, and most are yet uncertain.

When I is even, we can place the nuclei in two categories: 1. Even-even nuclei ( $\mathcal{N}$  even, Z even). For such nuclei we have

$$E^{-} = A^{-} + \chi(Z + 1, A) < A^{-},$$
  
 $E^{+} = A^{+} + \chi(Z - 1, A) < A^{+}$  .. (5)

for  $\chi(Z, A) = 0$ , when  $Z, \mathcal{N}$  are both even, and  $\chi(Z, A)$  is uniformly negative when Z and  $\mathcal{N}$  are both odd. Hence Z being even here  $\chi(Z+1, A) < 0$ .

The A-- and A+-curves have been drawn for two values of  $\beta$ , namely, 19.5 and 22.5 Mev. The first is Bethe's value, the second is a trial value. The ringed points give us the observed values of energy-release. It shows that  $\chi(Z, A)$ is negative whenever Z is odd and is of the order of 3-5 Mev. The dotted curve has been drawn for  $\beta$ --emitting nuclei with Z odd. For even-even nuclei, such a curve would be parallel to the A-curve, but according to (5), as much below it, roughly, as the odd-odd nuclei values are above. Only a section of it passing through the points He<sup>6</sup>, Be<sup>10</sup> and C<sup>14</sup> is shown. Here the  $|\chi(Z,A)|$  terms are smaller than | A- |, which is large on account of small values of M, the mass number, and hence the nuclei are negatronemitting, though both Z and  $\mathcal N$  are even; but the value of E- is far smaller than those of contiguous odd-odd nuclei like B12 or N16, where the two terms are additive. The curves give a rough indication as to the value of energy-release in a  $\beta$ --emission. From O<sup>18</sup> the A--values become smaller as the atomic mass is large, though y's continue to be of the same order and all even-even nuclei have  $E^-$  negative and therefore the nuclei are stable.

On the right-hand side, the ringed dots show the energy-release wherever known. The stability of even-even nuclei up to Ni<sup>58</sup> is easily explained. The next even-even nuclei like Zn<sup>62</sup> or Ge<sup>66</sup> will prove to be either stable or decaying by K-capture with a long life.

. Cl<sup>36</sup> and K<sup>40</sup>, which are in the middle of the group, are especially interesting. Cl<sup>36</sup>, like Cu<sup>64</sup>, has been found (Grahame and Walke<sup>6</sup>) to decay both by  $\beta$ --emission to A<sup>36</sup> as well as by K-capture to S<sup>36</sup>. K<sup>40</sup>, which occurs naturally, is similar, as it decays by negatron emission to Ca<sup>40</sup> and by K-capture, but probably also by positron emission to A<sup>40</sup>, as several investigators have found, but its spectrum requires re-investigation. Positron emission by K<sup>40</sup> is indicated by the annihilation radiation of 0.5 Mev., found by Klemperer.

It has been found for other groups that all nuclei which decay by simultaneous emission of negatrons on one hand, and positrons or K-capture on the other hand, like  $Cu^{64}$ , occur round about the points where the  $A^-$ -curves cut the  $A^+$ -curves.

So far, we have examined the data available for nuclei belonging to the groups I=-1 to I=6, and in more recent work by A. K. Saha and S. Ghosal, the method has been extended from I=7 to I=20. We have generally confirmed that the stability of nuclei, and the energetics of  $\beta$ -emissions, can be explained in the same general way; but it is not so easy to deal with level-formations in the nucleus. Probably the agreement will be far better when a more accurate mass-defect formula for even-even stable nuclei is available. A few important deductions arising out of the present investigation may be noted.

- (a) Li<sup>5</sup> is expected to be a nucleus having a long life of several years, and decaying by K-capture to He<sup>5</sup>, which should be stable.
- (b)  $A^{39}$ , reported as a  $\beta$ --nucleus with a life of 4 min., is a glaring anomaly:  $A^{39}$  is expected to be a long-lived nucleus decaying by K-capture to  $K^{39}$ .
- (c) Even-even nuclei may not always be stable, if the nucleus is a bit off the main line of stability. Thus Ti<sup>44</sup>, Cr<sup>48</sup>, etc., if they could be prepared, they would be found,

like  $C^{14}$ , to be long-lived products, decaying in these cases by K-capture.

- (d) It is probable that  $Ca^{48}$ , stated by Nier to be a rare stable isotope of calcium with a frequency of 0·2 per cent, is illusory, like  $Co^{57}$ , which was once believed to be stable, but was predicted by us to be unstable. This prediction has been confirmed; recently I. Curie<sup>7</sup> has independently come to the view that a stable  $Ca^{48}$  does not exist. This nucleus, and  $Ti^{52}$ ,  $Cr^{56}$ ,  $Fe^{60}$ , if they could be made, would be found unstable, decaying like  $C^{14}$  with the emission of slow  $\beta^-$ -rays. The first stable nucleus in the group I=4 should be  $Ni^{64}$ .
- (e) The radioactive nuclei  $V^{50}$ ,  $Mn^{54}$ ,  $Co^{58}$  should decay with both  $\beta^+$  and  $\beta^-$ -emissions and K-capture, but only  $\beta^+$ -emission and K-capture activity have been reported so far. If they are found not to emit  $\beta^-$ -rays a fundamental difficulty arises in the theory of  $\beta$ -emission.
- (f) The spin-dependent part of the nuclear binding energy, namely,  $\chi(Z,A)$  is found generally to be of the order of a few million electron volts, while according to classical theory it should be  $\simeq \frac{Zev}{cr^2} \mu_k$ , where Z is number of charges in the nucleus, v is the velocity of a nucleon inside in the nucleus, and  $\mu_k$  is the nuclear magnetic moment. Making plausible assumptions about these quantities, we find that  $\chi(Z, A)$  is of the order of about 10-20 kilovolts. This shows that we have to apply some type of meson theory to find out the right order of value of the spin-dependent part of the nuclear binding energy. Actually, D. Bose has deduced the right order of value for  $\chi(Z, A)$  using the vector meson theory of Kemmer, but as theories of meson field as Louis de Broglie has said, are not yet definitive, this point has not been pursued further.

<sup>&</sup>lt;sup>1</sup> Seaborg, Rev. Mod. Phys., 16, 1 (1944).

<sup>&</sup>lt;sup>2</sup> Saha, Sarkar and Mukherjee, Proc. Nat. Inst. Sci. India, 6, 45 (1940).

<sup>&</sup>lt;sup>3</sup> Kruger and Ogle, Phys. Rev., 67, 273 (1945).

<sup>&</sup>lt;sup>4</sup> Maier-Leibnitz, Z. Phys., 122, 233 (1944).

<sup>&</sup>lt;sup>5</sup> Giebert, Roggen, Rossel, Helv. Phys. Acta, 17, 97 (1944).

<sup>&</sup>lt;sup>6</sup> Grahame and Walke, Phys. Rev., **60**, 909 (1941).

<sup>&</sup>lt;sup>7</sup> Curie, J. Phys., (8), 6, 209 (1945).