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#### NUCLEAR EVAPORATION FROM URANIUM

Warren Heckrotte

April 7, 1953

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A nuclear reaction can be divided into two essentially independent phases. The first phase is the interaction of the incident particle (proton, neutron, deuteron, etc.) with the nucleus. This can lead to the immediate absorption of the incident particle; or for sufficiently high energies the incident nucleon can produce a number of high energy nucleons through a series of individual nucleon-mucleon collisions. In either case the nucleus will be left with a certain excitation energy. The second phase is the degradation of this excitation energy through the emission of nucleons. This phase can be considered as independent of the first phase. That is, the mode of decay of the excited nucleus bears no relationship to the mode of excitation except in so far as general conservation theorems hold. This concept is based on the assumption of strong interactions of short range between the particles of the nucleus. The energy which is transferred to the nucleus is quickly transferred through the nucleus as a whole; and the nucleus must go through many periods of motion before enough energy is again localized on a given particle for it to escape. Because this period of time before which a particle will be emitted occupies many nuclear periods, it is assumed that any phase relationships which might have existed initially are averaged out. Thus the nucleus "forgets" how it was excited.

The purpose of this paper is to investigate this second phase as it applies to uranium. For a given excitation energy we will calculate the number and kind of nucleons emitted and their energy distribution. To do this we will make use of the existing theories on the emission or evaporation of nucleons from an excited nucleus.

#### II. Evaporation Theory

The purpose of this section is to sketch briefly the theory of nuclear evaporation.

For very low excitation energies of the nucleus, the nucleus possesses rather well defined energy levels which decay by Y-ray emission. For high excitation energies, however, the levels become very closely spaced with large widths and decay principally by particle emission. For this region of closely spaced levels the basic approach is to use statistical considerations. One assumes the existence of the average values of certain quantities over small energy intervals. It is assumed that these quantities will be slowly varying functions of the energy. This can be expected to be so if the energy interval for averaging can be taken much smaller than the excitation energy. This will be so for a sufficiently dense distribution of levels. For the heavy nuclei this would correspond to an excitation energy of about 10 Mev above the ground state (or to an incident neutron of 3 or 4 Mev).

\* For a more thorough presentation of the material of this section the following references can be consulted:

- V. F. Weisskopf, Phys. Rev. <u>52</u>, 295 (1937)
- V. F. Weisskopf and D. H. Ewing, Phys. Rev. <u>57</u>, 472, 935 (1940)

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K. J. LeCouteur, Proc. Phys. Soc. A, 63, 498 (1950)



For charged particles the factor  $\in$  is replaced by  $\in + V$  where V is the potential barrier. In that case  $\in > V$ .

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Eq. (3) becomes then

$$T(E_A, \epsilon)d\epsilon = \gamma e^{-S_A(E_A) - S_B(E_A - B)} \epsilon e^{-\overline{T(E_A - B)}} d\epsilon$$
 (6)

The energy spectrum thus exhibits the Maxwellian form

where, however, it is to be noted, the temperature is that of the residual nucleus for an excitation energy  $(B_A - B)$ . The above is of course only valid for large aexcitation energies and small  $\in$ . However, the contribution for large  $\in$  is so small that the above expression can be considered valid for all  $\in$  for sufficiently large excitation energies.

It remains now to adopt a specific nuclear model which will relate S, T, and E. The most usual model to adopt for the present purposes is that of the Fermi degenerate gas for the nucleus. The nucleus is regarded as being made up of a collection of independent particles moving in a potential well, which represents the average effect of all nucleons upon one another. The collection of particles is taken to be com-pletely degenerate---the nucleus is at temperature T = 0 in the ground state. For an excitation energy E, the relation between E and  $\mathcal T$  is given by

$$=\frac{\pi^{2}A}{4\xi_{0}}\tau^{2}=\frac{\Lambda^{2}}{4}\tau^{2}$$

(7)

$$\Lambda^2 = \frac{\pi^2 A}{\xi_0}$$

where A is the number of muclear particles and  $\xi_0$  is the Fermi energy of the distribution. For the ground state,  $\xi_0 \sim 20$  or 30 MeV which yields

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From Eq. (2b),

 $s = \int \frac{dE}{T}$ 

which yields, by using Eq. (7),

$$S = \Lambda \sqrt{E}$$
.

Substituting Eq. (8) into Eq. (1),

$$W(\epsilon)d\epsilon = \gamma e^{-S_A(E_A)} e^{\Lambda_B \sqrt{E_A - B - \epsilon}} \epsilon d\epsilon .$$
(9)

Now because of the radical on the exponent, Eq. (9) is rather troublesome to handle; on the other hand Eq. (6) is comparatively simple. Eq. (6) is valid for  $\epsilon << E_A - B$ . We will now evaluate the average energy of emission  $\bar{\epsilon}$  according to Eq. (9) in order to obtain a criterion for the validity of Eq. (6).  $\bar{\epsilon}$  is given by

$$E_{A} - B_{EW(E)AE}$$

$$\int_{0}^{E_{A}} E_{A} - B_{W(E)AE}$$

$$\int_{0}^{E_{A}} W(E)AE$$

and one obtains

$$= 2 \mathcal{T}_{E_{A} - B} - \frac{6}{\Lambda_{B}^{2}} + \frac{6}{\Lambda_{B}^{3} \sqrt{E_{A} - B}} - \dots$$
(10)

where  $\mathcal{T}_{E_A=B}$  is the temperature of the residual nucleus with excitation energy  $(E_A = B)$ . For heavy nuclei  $\wedge \sim 10$  and thus for  $\mathcal{T} > 1/2$  MeV,

$$\overline{\epsilon} = 2 \mathcal{T}_{E_A - B} \tag{11}^*$$

For  $\mathcal{T}_{E_A=B} \sim 1/2$ ,  $E_A \sim 15$  MeV. Thus, since  $\overline{\epsilon} << E_A = B$  for  $E_A$  greater than 15 or 20 MeV, Eq. (6) will serve as an adequate representation of the energy spectrum if we are principally concerned with large excitation energies."

For the case of charged particles  $\overline{\epsilon} = 2 \overline{\tau} + \overline{v}$ . \*\* Actually Eq. (6) seems to be in better agreement with experiment for low excitation energies that Sq. (9), the more grade expression.

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It is apparent in the above discussion that the constant  $\wedge$  is a fundamental parameter in the theory. Although the value of  $\wedge$  has been calculated theoretically, it is better for purposes of application to regard it as an empirical parameter to be fixed by experiment. It is found, for instance, for low excitation energies that  $\wedge^2$  does not show the simple variation with A that the theory indicates, although the shape of the energy spectrum of emitted particles does agree with the Maxwellian form given in Eq. (6). The value that has been indicated for  $\wedge^2$  for uranium does agree approximately with the low energy data and such high energy data as is interpretable. Accordingly we will take the value of  $\wedge^2$  to be given by

$$\frac{\Lambda^2}{\Lambda} = \frac{\Lambda}{10}$$

for the very heavy nuclei.

It has been indicated in the previous discussion how the theory was to be modified for the case of charged particles. The height of the potential barrier is given by

$$v = \frac{Z}{A^{1/3}}$$

However, there is some tunneling through the barrier when the energy of the charged particles is less than this (E < V). This effect can be taken into account approximately by defining an effective potential barrier which will be lower than that given by Eq. (13). Letting the effective barrier by V', then it can be shown that

V' = KV

where.

K = 0.7 for protons = 0.77 for deuterons = 1.66 for alpha particles = 0.8 for H<sup>3</sup> = 1.6 for He<sup>3</sup>.

Redefining the potential barrier in this fashion will yield a better value for the probability of charged particle emission; it will on the other hand leave the shape of the low energy end of the energy spectrum of charged particles somewhat in error.

Another effect of importance is the variation in height of the potential barrier with excitation energy. For large excitation energies, the potential barrier shows a marked decrease in height, which increases the probability of charged particle emission and shifts the energy spectra of charged particles to lower energies. The effect seems to be marked for excitation energies as low as 200 Mey. However, since we will be concerned mainly with lower excitation energies, this effect will be neglected in this report.

A more detailed calculation shows that  $\wedge^2$  depends also on the neutron excess; However, this does not change the variation with A to any great extent. It would also be expected to show a variation from even-even to even-odd for low excitation energies.

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$$\mathbf{R} = \frac{\int_{\mathbf{X}}}{\int_{\mathbf{y}}} = \frac{Y_{\mathbf{x}} \mathbf{R}_{\mathbf{x}}}{Y_{\mathbf{y}} \mathbf{R}_{\mathbf{y}}} \stackrel{\wedge}{\bullet} \frac{\wedge (\sqrt{\mathbf{R}_{\mathbf{x}}} - \sqrt{\mathbf{R}_{\mathbf{y}}})}{\mathbf{R}_{\mathbf{y}} \mathbf{R}_{\mathbf{y}}}$$

where any difference between  $\Lambda_{\rm Bx}$  and  $\Lambda_{\rm By}$  has been neglected.

If particle x is a neutron and particle y a charged particle then, because of the coulomb potential, it will usually be true that R > 1. It is true that if a sufficient difference in binding energies existed then R could be less than 1.

Eq. (17) has been plotted in Figs. 1 and 2 for the ratio of neutron emission to that of protons, deuterons, alphas and He<sup>3</sup> and for the two cases A = 238, Z = 92 and A = 228, Z = 92. The variation of the potential barrier with excitation energy has been neglected. The inclusion of this variation would have a marked influence on these ratios above 200 MeV excitation energy.

For these values of A and Z the ratio of neutron emission to charged particle emission is generally >>1 for not too large excitation energies (<200 Mev). However, it is also true that as A becomes smaller for a given Z and a given excitation energy the charged particle emission becomes more and more probable. If A is reduced sufficiently for constant Z, charged particle emission will become more favorable, since the binding energy of the neutron will be constantly increasing while that of the charged particles will be decreasing.

For  $U^{238}$  it can be concluded that for not too high excitation energies, only neutron emission will effectively take place. A more quantitative criterion for this can be



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obtained easily. An examination of the binding energies of uranium as a function of A (constant Z = 92) shows that the effective binding energy (binding energy and coulomb barrier height) of protons and alpha particles does not become equal to that of the neutrons until A is about 224. This is 14 neutrons less than natural uranium. Now the average binding energy of neutrons in this region is about 6.5 Mev and the average kinetic energy carried out by a neutron is about 3 or 4 Mev for excitation energies of the order of 100 Mev. Thus if the excitation energy is  $14 \times (6.5 + 3.5) =$ 140 MeV, we can expect that the principal contribution to particle emission will be neutrons and that we can safely neglect charged particle emission. Since we can expect a spread about the average number of neutrons emitted, a more reasonable criterion on the excitation energy for considering only neutron emission. is 100 Mev.

To sum up the above considerations, the picture of particle emission that is indicated is the following. For not too high excitation energies—say of the order of a few hundred Mev—uranium will first emit a sequence of neutrons. After this initial sequence of neutron emission, charged particle emission becomes favorable and the remaining excitation energy will be degraded through charged particle and neutron emission together. This may be a sequence of alpha particles or a proton-neutron sequence. The general course of this emission process is sketched in Fig. 3.

IV. <u>Distribution in Number and Energy of Neutrons Evaporated from U<sup>235</sup> for</u> <u>Excitation Energies of 100 and 50 Mey</u>

(A) Before considering the detailed solution of the problem, the average number of neutrons emitted from uranium, neglecting charged particle emission, will be calculated.

The average change in excitation energy of the nucleus when a neutron is evaporated is given by

 $\frac{dE}{dA} = 2T + B,$ 

and since

$$E = \frac{\Lambda^2}{4} T = KA T^2,$$

$$= \frac{d\tau}{dA} + \kappa \tau^2.$$

Combining Eqs. (18) and (19),

dE dA

$$\frac{dA}{A} = \frac{2K\mathcal{T}}{B+2\mathcal{T}-K\mathcal{T}^2} a\mathcal{T}.$$



(19)

(20)

(18)



$$A_{f} = \frac{A_{0}}{2B} \left\{ K T_{0}^{2} - 2 T_{0} - B \right\} \left\{ \left( \frac{T_{0} - x}{T_{0} + y} \right) \frac{x}{T_{0} + y} \right\}^{\frac{1}{2}}$$
(22)

For  $K \sim 1/10$  and B = 6 MeV, the average number of neutrons evaporated is given in the following table. It will be noted that the average number of neutrons boiled off is not quite proportional to the excitation energy but increases somewhat less rapidly. The average number given for E = 200 is not to be taken seriously since charged particle emission would take place after the initial sequence of neutron evaporation.

E	n
50	6
100	11
200	20

(B) The distribution in number of evaporated neutrons and their energy distribution for a given initial excitation energy will now be calculated. Instead of an analytical treatment of the problem, the calculation will be done using the Monte Carlo method of solution. We will confine our attention to excitation energies of 50 and 100 Mev so that charged particle emission can be neglected.

For the Monte Carlo method for this problem one follows the sequence of neutron evaporation in detail. For a given excitation energy, the energy that the first neutron carries off is determined directly from the probability distribution of emission energies (Eq. (6)) by a choice of a random number. This energy is noted. Then for the residual nucleus, the excitation energy is E - B - T where T is the kinetic energy carried off (we are using T instead of  $\in$  in this section). Then the kinetic energy of the second neutron is determined as before. One continues in this fashion step by step until the excitation energy is insufficient to emit another



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neutron (E < B). This whole process is then repeated as many times as necessary to obtain good statistics on the distribution in number and energy of the emitted neutrons.

This mode of calculation will now be discussed in detail. The energy distribution for neutrons evaporated from a nucleus with excitation energy E is given by

T . T dT

where T is the neutron energy and  $\mathcal{T}$  is the temperature corresponding to the excitation energy E - B (p. 3). For large enough excitation energies, the normalized form of this equation is

$$P(T)aT = \frac{T}{T} e^{-\frac{T}{T}} \frac{dT}{dT}, \qquad (23)$$

which can be written as

$$x^{o} = x = x b(x)^{o}$$

The probability that x lies between  $x_1$  and  $x_{i+1}$  is

$$P_{i} = \int_{x_{i}}^{x_{i+1}} x e^{-x} dx$$

Dividing the total range of x into increments  $x_1 - x_0$ ,  $x_2 - x_1$ ,  $x_3 - x_2$ , ...,  $x_{1+1} - x_1$ , ..., one can associate a  $P_1$  with each increment of x. We say then that the probability of having the value of x equal to

$$x_1 = \frac{x_1 + x_{1+1}}{2}$$
 is  $P_1$ 

At this stage a correspondence between the integers 1, 2, 3, ...,  $n_k$ ,  $n_k$ 

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$$\frac{x_i + x_{i+1}}{2}$$
 is  $P_i$ .

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At this stage a correspondence between the integers 1, 2, 3, ...,  $n_x$ ,  $p_{00}$ , N and the  $x_1$ 's is made in the following manner. Starting with  $x_0$ , one associates the first  $n_0$  integers with  $x_0$ , where  $n_0$  is chosen to be proportional to  $P_0$ . The next  $n_1$  integers are associated with  $P_1$ , where  $n_1$  is proportional to  $P_1$ , and so on. For the particular case at hand, we chose the smallest value of  $x_1$  to be 0.1, and the smallest unit of probability to be 0.1. This is consistent with limiting ourselves to the first 200 integers (N = 100). The correspondence between these integers and the  $x_1$ 's is given in Table I.

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The next stop in the procedure is to construct a nonograph relating to x and T to the excitation energy E. Since

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and

this is easily accomplished and the nonograph is given in Fig. 4. Thus, if one knows x and E, T is found by laying a straight-edge across the figure for a given x and E which then selects the proper T.

In constructing the nonograph we have taken the average value of B which is given by 6.3 May in this region.  $\land$  is defined by

$$\frac{\Delta^2}{4} = \frac{A}{10} = \frac{233}{10}$$

 $\mathcal{T} = \frac{2}{\Lambda} \sqrt{E - B}$ ,

where we choose A = 233 as representing the average value of A.

With the correspondence between the  $x_1$ 's and the integers established and the nonograph prepared, the neutron spectrum can be solved for. We will start with the initial excitation energy 100 Mev. We now select from the group of integers one of them in a random fashion. This will correspond to a cortain x. For this x and E = 100, we determine  $T_1$  from the nonograph. This is the energy with which the first neutron is boiled off. This is noted. The excitation energy for the residual nucleus is now  $E = B = T_1$ . We select another number in a random fashion which gives us another x. For this x and the excitation energy  $E = B = T_1$ , we can determine  $T_2$ . This noted, and the excitation energy is too small to boil off another neutron. We will have obtained then the number of neutrons boiled off by this nucleus and the energy of each neutron. This whole process is then repeated for another nucleus with 100 Mev excitation and so on. It was found necessary to repeat the process about 200 times to obtain adequate statistics.

Figs. 5 and 6 give the distribution in number of neutrons evaporated for excitation energies of 50 and 100 Mev. Fig. 7 gives the distribution in kinetic energy of the neutrons for excitation energies of 50 and 100 Mev. The curves given in Fig. 7 represent a least square fit to the data.

It was noted that if the neutron energy spectrum for excitation energies 50 and 100 Mov were plotted on the same graph as functions of  $x = T/T_0$  where  $T_0$  is the initial temperature, the two spectra were similar. This is given in Fig. 8. It is not known at the present time whether this is accidental or not. However, this suggests that

" Kondall, M. G., Tablos of Random Sampling Numbors, London University, Dept. of Statistics, Tracts for Computers, No. 24 (1939)

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in the region of excitation energies of 50 and 100 MeV, the neutron energy sportra can be considered as a function of  $x = T/T_{c}$  only.

The distribution in final excitation energies (E < B) is given in Fig. 9. This distribution may be of assistance in determining the probability that the final mucleus fissions. It should be recognized however that because of the approximations made and inadequacies of the theory for low excitation energies, this distribution in final excitation energy may be considerably in error.

It should be noted that the great simplicity of this treatment was made possible by two factors. The first is that the energy distribution of evaporated neutrons could be written as a function of (T/T) and secondly, that T did not vary from nucleus to nucleus in the evaporation chain (an avorage A and B was taken). These two factors made it possible to set up the probability distribution and the nonograph for all nuclei. If this had not been the case the problem could still have been solved by the Monte Carlo method, of course, but not with the same dispatch.

The method could now be extended to take into account charged particle emission. It is only necessary to establish at each point in the sequence what type of particle is emitted. This is done by calculating the emission probabilities (Eq. 16) and then deciding by a choice of random numbers what type of particle is emitted at each stop. Encopt for this modification the procedure is the same as before. The principal difficulty facing this procedure at the moment is lack of knowledge of the binding energies far off the region of stability (Fig. 3).

Another point to consider is that fission may also play a role in these considerations. This could be taken into account easily enough if one know the fission probability as a function of excitation energy. This of course is not known. Most probably, however, fission will only play a role for the lower excitation energies. This would have the effect of modifying the low energy end of the neutron spectrum on Figs. 6 and 8, by reducing the contribution from the evaporation process and adding to it the contribution from the fission process. The high energy end (to the right of the peak) would remain essentially the same.

Finally, it should be noted that those quantitative results cannot be compared directly with experiment (except in a qualitative way). It would first be necessary to analyze the first phase of the nuclear reaction-i.e., the interaction of the incident particle with the nucleus. This would lead to an expression for the distribution in excitation energy of the nucleus. That result combined with the above results would lead then to a complete description of the nuclear reaction.









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Most probable decay path for 2238 for excitation energies of a few hundred Mev.

Fig. 3

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