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Warren Heckrotte

December 18, 1953

Berkeley, California

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ABSTRACT

The standard nuclear evaporation theory is applied to the excited uranium nucleus to determine the number and kind of nucleons emitted and their energy distribution. For excitation energies of about 100 Mev and less charged particle emission can be neglected. The neutron spectrum is obtained by using the Monte Carlo method. Simple analytical expressions for the neutron energy distribution and the mean number of evaporated neutrons which fit these results are given. The results of the calculation are compared with the measured neutron spectrum from the 190 Mev deuteron bombardment of uranium.

The role of fission at these excitation energies is considered qualitatively in the light of several high energy experiments. The experiments indicate that neutrons are emitted before the fissioning nucleus actually separates into fragments. This suggests two possible hypotheses on the relation of the fission probability to that of the neutron emission probability, which are discussed briefly.

EVAPORATION OF NEUTRONS FROM THE EXCITED URANIUM NUCLEUS

Warren Heckrotte

I. Introduction

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-nucleon 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 that the energy which is transferred to the nucleus is quickly spread through the nucleus as a whole; and that the nucleus will 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 theory of the emission or evaporation of nucleons from an excited nucleus. In addition, we will consider in a qualitative sense the possible relation between fission and the evaporation process.

II. Evaporation Theory

The purpose of this section is to sketch briefly the theory of nuclear evaporation[1].

For very low excitation energies of the nucleus, the nucleus possesses rather well defined energy levels which decay by γ -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 smoothly varying functions of the energy. This can be expected if the energy interval for averaging can be taken much smaller than the excitation energy; which will be so for a sufficiently dense distribution of levels. For the heavy nuclei this would correspond to an excitation energy of about 8 Mev above the ground state (or to an incident neutron of about 2 or 3 Mev).

The pertinent quantity which describes the evaporation process is the probability per unit time that the nucleus A, with excitation energy E_A, emits a nuclear particle with kinetic energy between T and $T + dT$, leaving the residual nucleus with energy $E_B = E_A - T - B$, where B is the binding energy of the emitted particle to nucleus A.

This probability can be expressed as a function of the reverse process by means of the principle of detailed balance. Application of this leads to the following equation for the probability of emission[1]

$$W(E_A, T)\,dT = \gamma\,\sigma(E_A, T)\,\frac{\rho_B(E_B)}{\rho_A(E_A)}\,T\,dT \qquad (1)$$

$\rho(E)dE$ represents the number of excited levels of a nucleus in the energy interval dE for an excitation energy E. The factor

$$\gamma = \frac{(2s+1)m}{\pi^2\,\hbar^3}$$

where m is the mass and s is the spin of the emitted particle. $\sigma(E_A, T)$ is the cross section for the inverse process; that is, the cross section for the absorption of the same nucleon with energy T by the nucleus B with excitation energy E_B to yield nucleus A with excitation energy $E_A = E_B + B + T$. For E_B of the order of the binding energy or more, and for T in the usual range of a few Mev, $\sigma(E_A, T)$ may be taken to be the nuclear cross section $= \pi R^2$. It is this simplifying fact which makes the evaporation theory so useful. For the case of charged particles, the factor T is replaced by $(T - V)$, where V is the potential barrier and $T > V$.

Having Eq. (1), the problem is to relate $\rho(E)$ to the specific nuclear properties. To do this one must construct a specific nuclear

model and calculate $\rho(E)$. However, one can proceed several steps further before specializing.

The functions S and τ are introduced which are defined by the equations

$$S(E) = \ln \rho(E) \tag{2a}$$

$$\frac{1}{\tau} = \frac{\partial S}{\partial E} \tag{2b}$$

In analogy with thermodynamics, S is identified with the entropy and τ with temperature of the nucleus. In terms of S, Eq. (1) becomes

$$W(E_A, T)\,dT = \gamma\sigma \exp\left[-S_A(E_A) + S_B(E_B)\right] T\,dT. \tag{3}$$

S_B can be expanded about $(E_A - B)$ in powers of T. Neglecting higher powers of T than the first, and utilizing Eq. (2b), Eq. (3) becomes

$$W(E_A, T)\,dT = \gamma\sigma \exp\left[-S_A(E_A) + S_B(E_A - B)\right] \exp\left[-T/\tau_{E_A - B}\right] T\,dT. \tag{4a}$$

The energy spectrum given by Eq. (4a) thus exhibits the simple form

$$T \exp\left[-T/\tau_{E_A - B}\right] T\,dT, \tag{4b}$$

where, it is to be noted, the temperature is that of the residual nucleus for an excitation energy $(E_A - B)$[1].

It remains now to adopt a specific nuclear model which will relate S, τ, and E. The most usual model to adopt for the present purposes is that of the degenerate Fermi 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 completely degenerate --the nucleus is at temperature $\tau = 0$ in the ground state. For an excitation energy E, the relation between E and τ is given by[2]

$$E = \frac{\pi^2 A}{4\mu_0}\tau^2 = \frac{\lambda^2}{4}\tau^2, \qquad \lambda^2 = \frac{\pi^2 A}{\mu_0}, \tag{5}$$

where A is the number of nuclear particles and μ_0 is the Fermi energy of the distribution. For the ground state, μ_0 = 20 to 30 Mev, which yields

$$E \simeq \frac{A}{10}\tau^2.$$

From Eq. (2b), and using Eq. (5),

$$S = \int \frac{dE}{\tau} = \lambda E^{1/2} \tag{6}$$

Substituting Eq. (6) into Eq. (1) yields,

$$W(T)\,dT = \gamma\sigma \exp\left[-S_A(E_A) + \lambda_B (E_A - B - T)^{1/2}\right] T\,dT \tag{7}$$

Because of the radical in the exponent, Eq. (7) is rather troublesome to handle; on the other hand Eq. (4) is comparatively simple. For $(E_A - B)$ greater than about 8 or 10 Mev, Eq. (4) is an acceptable approximation to the energy distribution. Accordingly Eq. (4) will be used for the energy distribution where S and τ are given by Eqs. (5) and (6). Experimentally it is found that the shape of the energy spectrum of emitted neutrons is of the form indicated by Eq. (4) for excitation energies of about 20 Mev[3].

The average kinetic energy of emission, T, is easily evaluated from Eq. (4) and is given by

$$T = 2\tau_{E_{A-B}} \qquad (8)$$

For the case of charged particles, this is replaced by $T = 2\tau + V$.

It is apparent from Eqs. (5) and (6) that the constant Λ is a fundamental parameter in the theory. Although the value of Λ has been calculated theoretically*, it is better for purposes of application to regard it as an empirical parameter to be fixed by experiment, if possible. This, cannot, however, be done in an unambiguous way. It is found, for instance, from the study of the energy spectrum of neutrons from various nuclei[3] that, although the shape is given by Eq. (4), the value of Λ^2 obtained does not show the simple variation in A

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* A more detailed calculation[4] shows that Λ^2 depends on the neutron excess and several other parameters, though this does not change the variation with A to any extent. Also, a variation in the level density from even-even to even-odd nuclei would be expected for low excitation energies.

that the theory indicates. On the other hand, a study of the density of nuclear levels just above the neutron threshold[5] does seem to show that λ^2 is roughly proportional to A. However, the numerical value of this constant for the heavy nuclei from both studies is roughly in agreement with the value indicated earlier; and the detailed analysis of high energy nuclear stars[4] for nuclei of A about 100 yields a value of λ^2 of about the value calculated. Since there is no unambiguous choice and the calculated value does not disagree too violently with any of the data, the value of λ^2 will be taken to be given by

$$\frac{\lambda^2}{4} = \frac{A}{10} \qquad (9)$$

for the 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 for protons is given by

$$V \simeq \frac{Z}{A^{1/3}} \qquad (10)$$

However, there is some tunneling through the barrier when the energy of the charged particles is less than this ($T < V$). This effect can be taken into account approximately by defining an effective potential barrier which will be lower than that given by Eq. (10). Letting the effective barrier be V', then it can be shown that[6]

$$V' = KV \qquad (11)$$

where

$K = 0.7$ for protons
$= 0.77$ for deuterons
$= 1.66$ for alpha particles
$= 0.8$ for H^3
$= 1.6$ for He^3.

Another effect of importance is the variation in height of the potential barrier with excitation energy which is caused by the distortion in the nuclear shape with increasing excitation energy[7]. For large excitation energies, this 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 Mev[4]. However, we will be concerned mainly with lower excitation energies and will neglect this effect.

III. Relative Probabilities of Neutron Emission, Charged Particle Emission, and Fission.

In this section the relative probability of the various processes which a highly excited heavy nucleus, such as U^{238}, can undergo will be estimated. These processes are neutron emission, charged particle emission, and fission. Evaporation theory provides good estimates of the relative particle emission rates; there is, however, no adequate theory of the high energy fission process which provides reliable estimates of the rate of fission. Accordingly, only a qualitative description of the high energy fission suggested by the available (and rather limited) data will be made. This will at least establish a working hypothesis.

(A) Particle Emission

The relative rates of neutron emission to charged particle emission will be considered first. This will be done by a straight-forward application of the evaporation theory and without regard to any possible modifications which the fission process may imply.

The probability of emission of a given kind of particle from an excited nucleus is given by

$$\frac{\Gamma}{\hbar} = \int_{V'}^{E_A - B} W(E_A, T)\, dT \qquad (12)$$

Γ = transition probability (in energy units)

V' = effective potential barrier; for neutrons $V' = 0$.

Using Eq. (9), this becomes

$$\frac{\Gamma}{\hbar} = \frac{4\gamma\sigma G}{\lambda_B^2} \exp\left[-S_A(E_A) + S_B(G)\right], \qquad (13)$$

$$G = E_A - B - V'.$$

The ratio of emission probabilities for two particles x and y is

$$R = \frac{\Gamma_x}{\Gamma_y} = \frac{\gamma_x G_x}{\gamma_y G_y} \exp\left[-\lambda\left(G_x^{1/2} - G_y^{1/2}\right)\right] \qquad (14)$$

where any difference between λ_{Bx} and λ_{By} has been neglected.

If particle x is a neutron and particle y a charged particle, then, because of the coulomb barrier, it will generally be true that $R > 1$. R may be less than unity if the binding energy of the charged particle is sufficiently less than the binding energy of the neutron, or if there exists large variations in the level density in neighboring nuclei. This latter effect has been neglected here, but can play a role in nuclear reactions for nuclei of intermediate mass[8].

For values of A and Z in the region of the heavy nuclei, the ratio of neutron emission to that of charged particles is generally much greater than unity for not too high excitation energies. As A becomes smaller for a given Z and a given excitation energy, the charged particle emission rate becomes more favorable since the binding energy of the neutron will be increasing, while that of the charged particles will be decreasing. This is shown in Fig. 1, when the ratio of neutron width to proton width for the nuclei U^{238} and U^{228} is plotted as a function of excitation energy. The ratios for other charged particles are comparable.

For U^{238} it can be concluded that for not too high excitation energies, charged particle emission can reasonably be neglected*. An examination of the binding energies of particles in uranium, shows that the effective binding energy (binding energy plus coulomb barrier height)

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* It should be noted though that reactions involving the emission of charged particles may occur with a much larger cross section than would be indicated from the above considerations by virtue of some other mechanism than the formation of a compound nucleus; e.g., direct knock-ons, pickup reactions, stripping reactions, etc.

of protons and alpha particles is approaching that of neutrons for A $\simeq$ 222-224. This is about 15 neutrons less than natural uranium. The average binding energy of neutrons in this region is about 6.5 Mev and the average kinetic energy carried out by a neutron (Eq. (8)) is about 3 or 4 Mev for excitation energies of the order of 100 Mev. Thus, if the excitation energy is 15 x (6.5 + 3.5) = 150 Mev or more, charged particle emission would start to play an important role. A reasonable criterion on the excitation energy for neglecting charged particle emission is 100 Mev. This corresponds to the emission of about ten neutrons, and an estimate on the basis of Eq. (14) shows that about 96% of the time, ten neutrons will be emitted--the other 4% of the time, one or more charged particles will be emitted in the sequence.

These considerations suggest that at considerably higher energies (and still ignoring fission) that the excitation energy is degraded first by the emission of a sequence of neutrons followed by a sequence of protons and neutrons in about equal number and alpha particles. However, the uncertainties in the fission process and the nature of the coulomb barrier at high excitation energies prevent any definite conclusion.

(B) Fission

The above considerations have ignored the fission process and it will be attempted here to state its possible relationship to particle emission for high excitation energies (10 to 100 Mev).

The fission width according to Bohr and Wheeler[9], is given by

$$\Gamma_f = \frac{1}{2\pi} \frac{\int_{E_f}^{E} \rho^*(E'-E_f)\,dE'}{\rho(E)} \qquad (15)$$

where E is the excitation energy, E_f the fission threshold, and ρ^* is the density of levels at the critical configuration (saddle point in the potential energy surface) leading to fission. Assuming that the form of ρ^* is the same as ρ (Eqs. (2a) and (6)),

$$\Gamma_f \simeq \frac{\tau(E-E_f)}{2\pi} \exp\left[\lambda\left(E^{1/2}-(E-E_f)^{1/2}\right)\right], \qquad (16a)$$

and the ratio $\left(\Gamma_f / \Gamma_n\right)$ is given by

$$\frac{\Gamma_f}{\Gamma_n} = \frac{1}{2\pi\hbar\gamma\sigma} \frac{\tau(E-E_f)}{\tau^2(E-B)} \exp\left[\lambda\left((E-E_f)^{1/2}-(E-B)^{1/2}\right)\right], \qquad (16b)$$

$$\frac{1}{2\pi\hbar\gamma\sigma} \simeq 0.12$$

This function, for most values of the parameters B and E_f, shows a fairly rapid change with energy. This can be compared with the available data which is confined, on the whole, to excitation energies less than 12 Mev. The photo induced fission in U^{238} shows a fission

width which is essentially constant for excitation energies from 8 to 11 Mev[10]. The value of (Γ_f / Γ_n) is about 0.30. Eq. (16b), on the other hand, shows a change in value from 2.5 to 0.9 for this energy range where $E_f = 5.3$ [10] and $B_n = 5.9$. This constancy of the ratio (Γ_f / Γ_n) seems to hold for other nuclei also. An examination of the neutron induced fission cross sections[11] for many of the nuclei in this region shows that they are fairly constant in this energy region. Since the absorption or interaction cross section is about $\pi (R + \lambda)^2$ in this region and thus almost constant, this implies that (Γ_f / Γ_n) is also constant. The values of (Γ_f / Γ_n) vary considerably among the nuclei. This variation is not reproduced by Eq. (16). The principal point to be made here is that Eq. (16) does not serve as an adequate description of the fission rate. This may follow from an assortment of reasons in proceeding from Eq. (15) to Eq. (16)--such as an inadequate representation of ρ^*, or even of ρ at these energies or possibly from a variation of E_f with excitation energy. This serves to illustrate the lack of a reliable estimate in such simple terms of the fission rate at these or higher energies.

Since above this medium energy range, the dependence of the ratio (Γ_f / Γ_n) can only be surmised, this will be done on the light of several high energy experiments, which, though by no means conclusive, do suggest possible models.

The excitation functions of the reactions Th^{232} (p, xn) [xn = x neutrons out] in the energy range 30 to 300 Mev have been investigated by Meinke, Wick and Seaborg[12]. They have found that the

shape of the excitation functions can be understood in terms of the evaporation theory, if it is assumed that the ratio of fission width to neutron width is either a constant or a slowly varying function of the energy in the energy region from about 30 to 100 Mev. About the magnitude of the ratio nothing definite can be concluded from their results.

Douthett[13] has measured the range of fission fragments from the 340 Mev proton bombardment of uranium. He finds that the fission fragment ranges are about 5% shorter than the ranges from the thermal neutron fission. He shows that this shortening of the range is consistent with the assumption that (1) about twenty neutrons or (2) about four protons or (3) about ten neutrons and two protons were emitted before the fission took place. The mean excitation energy of the uranium nucleus under 340 Mev proton bombardment is about 100 Mev[14]. Thus, on the basis of the evaporation theory and the known binding energies, statement (3) is to be preferred.

From these experimental results it could be conjectured that the ratio (Γ_f/Γ_n) decreases quite rapidly from the medium energy value and stays small at higher energies. The usual evaporation theory could be applied then without consideration of fission at excitation energies above 25 or 30 Mev.

This is not the only possibility that one can suggest which is consistent with the above (limited) data. Suppose that $\Gamma_f \simeq \Gamma_n$ at the higher excitation energies. The quantity $(\hbar/\Gamma_f)$ is a measure of the time it takes for the nucleus to reach the critical configuration which then leads to the actual separation. Now the time between reaching this critical separation and the time of the actual separation may be

much longer than the time given by $(\hbar/\Gamma_f)$. This, for instance, is the main assumption in the statistical treatment of fission at low excitation energies[15]. If this holds more or less true, then, since $\Gamma_n \simeq \Gamma_f$, neutrons would be evaporated during this period (i.e., before the actual separation). This mechanism is consistent with the above experimental data.

The third possibility which exists is that $\Gamma_f \simeq \Gamma_n$, and that on the whole, most of the neutrons are emitted after the actual separation by the fission fragments. However, this possibility is in contradiction to Douthett's conclusions and we shall assume that this presents no effective competition. It should be noted however that the usual fission neutrons associated with low energy fission are not being referred to, since they are indeed emitted by the fragments. In that case there is not sufficient excitation energy in the original nucleus to emit the neutrons.

In the subsequent calculations to be made, the usual evaporation theory will be applied to the emission of neutrons from the excited nucleus. If the assumption that fission does not compete effectively with neutron emission above 25 or 30 Mev, the evaporation theory will serve as an adequate description above this excitation energy. Below this energy, one encounters the same uncertainties as with the second possibility sketched above. In this case one expects the fission process to have some effect on the evaporation process. If, for instance, the time during which the separation is taking place is long compared to $(\hbar/\Gamma_n)$, the principal effect would be to change the effective excitation energy by the amount necessary to achieve the distortion of the nucleus--this would lead to a somewhat smaller nuclear temperature, and might reduce the

mean number of evaporated nucleons before the actual separation. It would also undoubtedly increase the possibility of charge particle emission. However, the actual circumstances are undoubtedly more complicated, and it can only be hoped they do not affect the simpler theory too markedly.

IV. Distribution in Number and Energy of Neutrons Evaporated from U^{238} for Excitation Energies of 100 and 50 Mev.

(A) Monte Carlo Solution.

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 reasonably be neglected.

In 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. (4)) 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. 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 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

$$T \exp [-T/\tau] \, dT$$

where T is the neutron energy and is the temperature corresponding to the excitation energy (E - B). For large enough excitation energies, the normalized form of this equation is

$$P(T)\,dT = \frac{T}{\tau} \exp [-T/\tau] \, \frac{dT}{\tau} \qquad (17)$$

which can be written as

$$P(x)\,dx = x \exp [-x] \, dx$$
$$x = T/\tau \qquad (18)$$

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

$$P_i = \int_{x_i}^{x_{i+1}} x \exp [-x] \, dx$$

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

$$\bar{x}_i = \frac{x_i + x_{i+1}}{2} \quad \text{is } P_i \, .$$

At this stage a correspondence between the integers 1, 2, 3, ..., n_k, ..., N and the $\bar{x}_i$'s is made in the following manner. Starting with $\bar{x}_o$, one associates the first n_o integers with $\bar{x}_o$, where n_o is chosen to be proportional to P_o. 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_i to be 0.1, and the smallest unit of probability to be 0.1. This is consistent with limiting ourselves to the first 100 integers (N = 100). The correspondence between these integers and the $\bar{x}_i$'s is given in Table I.

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

$$x = T/\tau$$

and

$$\tau = \frac{2}{\Lambda}(E-B)^{1/2}$$

this is easily accomplished and the nomograph is given in Fig. 2. 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 nomograph we have taken the average value of B which is given by 6.3 Mev in this region[16]. Λ is defined by

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

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

With the correspondence between the x_i's and the integers established and the nomograph 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[17]. This will correspond to a certain x. For this x and E = 100, we determine T_1 from the nomograph. This is the energy with which the first neutron is foiled 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 of the residual nucleus is $E - 2B - T_1 - T_2$. We then proceed until 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.

Fig. 3 gives the distribution in number of neutrons evaporated for excitation energies of 50 and 100 Mev. The mean numbers evaporated are 5.7 and 10.6, respectively. The variance, $(\overline{n^2} - \overline{n}^2)^{\frac{1}{2}}$, of these numbers is comparatively small; this results, of course, because most of the excitation energy is carried off as binding energy, rather than kinetic energy. An analytical expression for the mean value will be obtained later. Fig. 4 gives the distribution in kinetic energy of the neutrons for excitation energies of 50 and 100 Mev. These curves represent a least square fit to the data. In Fig. 5 the neutron energy spectra of Fig. 4 have been plotted as a function of $x = T/\tau_0$, where τ_0 is

the initial temperature. The two curves are essentially the same, which suggests that the neutron energy spectrum can be represented as a function of $x = T/\tau_o$ in this energy region. This point will be considered later.

In Fig. 6, the neutron energy spectrum resulting from the 190 Mev deuteron bombardment of uranium[18] is plotted along with the calculated neutron spectrum for 100 Mev excitation. The normalization is somewhat arbitrary and can only be regarded as approximate. The two match fairly well; however, this cannot be regarded as particularly significant. The experimental situation is quite complicated since there is a considerable spread in excitation energies and there is undoubtedly more than one type of initial excited nucleus due to knock-ons. In addition the experimental curve naturally includes some fission neutrons; that is, neutrons from fragments. The high peak of the experimental curve around one to two Mev may be due to these fission neutrons, or the sharp peak may be caused by the possible double-peaked nature of the distribution of initial excitation energy transferred to the nucleus by the deuteron[19].

It should be noted that the 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/τ_o) and secondly, that the form of τ_o did not vary from nucleus to nucleus in the evaporation chain (an average A and B was taken). These two factors made it possible to set up the probability distribution and the nomograph 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 step. Except for this modification the procedure is the same as before.

(B) Approximate Analytical Solution*

The Monte Carlo solution yields a full solution to the evaporation problem at any given energy. From the data that is obtained every pertinent type of distribution can be extracted. However it is possible to obtain in a simple way analytical expressions for the mean number of neutrons evaporated at a given energy, and the mean value and variance of the neutron energy spectrum, which are the most pertinent quantities. This will be done and the results compared to the Monte Carlo solution.

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

$$\frac{dE}{dA} = 2\tau + B \qquad (19)$$

and since

$$E = \frac{A}{10}\tau^2 = \lambda\tau^2,$$

$$\frac{dE}{dA} \simeq 2\lambda\tau\frac{d\tau}{dA}, \qquad (20)$$

where the variation in A of the coefficient of τ^2 has been neglected.

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* A much more extensive treatment of this nature has been given by LeCouteur[20]. The more limited results obtained in this section are derived in a more direct fashion however.

This simplifies the results without effecting the results appreciably. Combining Eq. (19) and (20),

$$dA = \frac{2\lambda\tau\, d\tau}{2\tau + B} \qquad (21)$$

The solution to Eq. (21) is

$$A_o - A_f = \bar{n} = \frac{E_o - B}{2}\left[1 - \frac{2}{3}\left(\frac{2\tau_o}{B}\right) + \frac{2}{4}\left(\frac{2\tau_o}{B}\right)^2 - \cdots\right],$$

where $\bar{n}$ is the mean number of evaporated neutrons and $\tau_o = (E_o - B/\lambda)^{1/2}$ is the initial temperature.

A somewhat better result is obtained if the coefficient $(E_o - B)$ is replaced by $(E_o - B/2)$ which corresponds to normalizing n to 1/2 at $E_o = B$ rather than zero as above. We have then

$$\bar{n} = \frac{E_o - B/2}{B}\left\{1 - \frac{2}{3}\left(\frac{2\tau_o}{B}\right) + \frac{2}{4}\left(\frac{2\tau_o}{B}\right)^2 - \cdots\right\}. \qquad (22)$$

For $\lambda = 233/10$ and $B = 6.3$ Mev, the average number of neutrons evaporated is given in the following table. The average number for $E = 200$ Mev is not to be taken too seriously since charged particle

emission would compete favorably for this energy. The values obtained

E	$\bar{n}$
50	5.7
100	10.6
150	15.1
200	19.2

agree with the Monte Carlo calculation.

From Eq. (22) the mean energy of the emitted neutrons can be obtained. The mean kinetic energy carried off per nucleon is

$$\overline{T} = B\left\{\frac{1}{1-\frac{2}{3}\left(\frac{2T_0}{B}\right)+\cdots} - 1\right\}.$$

This may be reduced to

$$\overline{T} = \left(\frac{4}{3}T_0 - \frac{2}{9}\frac{T_0^2}{B}\right), \qquad (23)$$

where only the two leading terms have been retained.

To determine the mean square kinetic energy, we introduce the function J_n which is given by

$$J_n = \overline{T_1^2} + \overline{T_2^2} + \overline{T_3^2} + \cdots + \overline{T_n^2} \qquad (24)$$

where $\overline{T_i^2}$ is the mean square kinetic energy of the i'th evaporated nucleon. Then

$$\Delta J_n = J_n - J_{n-1} = \overline{T_n^2}$$

or

$$\frac{\Delta J}{\Delta n} = \overline{T_n^2} = 6\tau^2(E_0 - B - E) \quad *$$

Since $\Delta n = \dfrac{\Delta E}{2\tau(E_0 - B - E) + B}$ and $\Delta E = 2\lambda\tau\, d\tau$,

$$J = \int \Delta J = 12\lambda \int_0^{\tau_0} \frac{\tau^3\, d\tau}{2\tau + B} \qquad (26)$$

The solution of Eq. (26) is

$$J = 6\,\frac{E_0}{B}\,\frac{\tau_0^2}{2}\left[1 - \frac{4}{5}\left(\frac{2\tau_0}{B}\right) + \frac{2}{3}\left(\frac{2\tau_0}{B}\right)^2 - \cdots\right]. \qquad (27)$$

The average mean square kinetic energy per particle is given by

$$\overline{T^2} = \frac{J}{n} \simeq 6\,\frac{\tau_0^2}{2}\left[1 - \frac{2}{15}\left(\frac{2\tau_0}{B}\right)\right]. \qquad (28)$$

---

* This may be easily verified from Eq. (4).

The variance , σ, of T is given by

$$\sigma^2(T) = \overline{T^2} - \overline{T}^2 \simeq \tau_0^2 \left\{ \frac{11}{9} - \frac{14}{135} \left(\frac{2\tau_0}{B} \right) \right\}.$$

Assuming the following Poisson like distribution for T,

$$P(T)dT = \frac{\left(\frac{\overline{T}}{\sigma^2}\right)^{\frac{\overline{T}^2}{\sigma^2}}}{\Gamma\left(\frac{\overline{T}^2}{\sigma^2}\right)} T^{\left(\frac{\overline{T}^2}{\sigma^2} - 1\right)} \exp\left[-\overline{T}T/\sigma^2\right] dT$$

and letting $\overline{T} = \frac{4}{3}\tau_0$ and $\sigma^2 = \frac{11}{9}\tau_0^2$,

the distribution for T becomes

$$P(T)dT = \frac{\left(\frac{12}{11}\right)^{\frac{16}{11}}}{\Gamma\left(\frac{16}{11}\right)} X^{\frac{5}{11}} \exp\left[-\frac{12}{11}X\right] dX \quad , \quad (29)$$

where $X = T/\tau_0$

This bears out the result of the Monte Carlo calculation that the neutron energy spectrum could be represented as a function of T/τ_0 in that energy region.

Eq. (29) is plotted in Fig. 5. Although the agreement with the Monte Carlo calculation is not too satisfactory for $x < 1$ *, Eq. (26)

* The statistics were not quite sufficient to render the shape of the Monte Carlo curve too accurately in this region where it changes rather rapidly with energy. Some of the discrepancy may be due to this.

can be regarded as an adequate representation of the energy spectrum of the evaporated neutrons. It is to be noted that the spectrum does not refer to any specific nucleus, but depends solely on the temperature .

V. Conclusion

A highly excited uranium nucleus can undergo neutron emission, charged particle emission, and fission. Good estimates of the particle emission rates are provided by the usual nuclear evaporation theory. For excitation energies of about 100 Mev and less, charged particle emission is greatly inhibited by the coulomb barrier, so that principally neutrons are emitted. On the other hand, the fission process at high excitation energies presents an essentially unsolved problem. The present experimental data suggests two possible alternatives for the role of fission. The first of these assumes that the fission becomes negligible for energies above 25 or 30 Mev compared with neutron emission. The second assumes that although the fission competes effectively with neutron emission at high energies, the excitation energy of the nucleus is degraded by a sequence of evaporation neutrons during the time between the critical configuration leading to fission and the actual separation of the nucleus into fragments. This latter circumstance would undoubtedly have an effect on the evaporation process--this has not been considered though. The principal point in either case is that the neutrons are apparently emitted before the separation.

It should be noted that in either case the probability that the uranium nucleus of 100 Mev excitation has fissioned in very close to unity. Either because of the increase of Z^2/A from the evaporation of the neutrons or because of the chance to fission at each step in the evaporation sequence.

The evaporation process for sufficiently high energies involves a sequence of emitted particles, which can be treated most naturally by the Monte Carlo method of solution. For the heavy nuclei where charged particle emission is small or negligible, the calculation becomes particularly simple. The most pertinent results obtained were the distribution in number of neutrons emitted and their energy distribution for excitation energies of 50 and 100 Mev. In addition, simple formulae for the mean number of neutrons emitted, and the energy distribution were obtained, which agreed with the Monte Carlo results.

The only quantitative result which would offer some chance of evaluating the calculation by comparison with experiment is that of the neutron energy spectrum. The only data available however is the spectrum resulting from the 190 Mev deuteron bombardment of uranium. As was pointed out there are too many uncertainties in this spectrum to make a significant comparison. The spectrum resulting from the proton bombardment in the 50 - 100 Mev range would probably enable a more valid comparison.

With respect to the role of fission, it is possible that the angular correlation of the neutrons with the fission fragments would yield further information on when the neutrons are evaporated. Also a study of the cross section of the spallation products and the fission cross section resulting from the bombardment with protons of 100 Mev or somewhat less energy might throw some light on the competition between fission and neutron evaporation. The advantage of limiting the proton energy to this value is that the spread in initial excitation energy will be less than for higher energy protons.

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FIGURE CAPTION

Figure 1: The ratio (Γ_n/Γ_p) for U^{238} and U^{228} plotted as a function of excitation energy.

Figure 2: Nomograph relating x, T, and E where $x = T/\tau_o$ and $\tau_o = 2(E - B)^{\frac{1}{2}}/a$.

Figure 3: The probability distributions in the number of evaporated neutrons for excitation energies 50 and 100 Mev.

Figure 4: The distribution in kinetic energy of the evaporated neutrons from uranium for 50 and 100 Mev excitation energies. The curves are least square fits to the results of the Monte Carlo calculation. The histogram for the result of the 50 Mev calculation is also shown. The areas under the curves are proportional to the total number of particles emitted.

Figure 5: The distribution of the evaporated neutrons as a function of $x = T/\tau_o$ where T is the kinetic energy of the neutrons and τ_o is the initial nuclear temperature. All three curves are normalized to unity.

Figure 6: A comparison of the measured neutron energy spectrum from the 190 Mev deuteron bombardment of U^{238} and the calculated neutron energy distribution for 100 Mev excitation energy.

TABLE I

Correspondence between set of integers N, and X established by the probability distribution, Eq. (18).

N	X	N	X	N	X	N	X
1	0.1	26	0.9	51	1.7	76	2.7
2		27	1.1	52		77	
3	0.3	28		53		78	2.9
4		29		54		79	
5		30		55	1.9	80	
6		31		56		81	3.1
7	0.5	32		57		82	
8		33		58		83	
9		34		59		84	3.3
10		35	1.3	60	2.1	85	
11		36		61		86	3.5
12		37		62		87	
13	0.7	38		63		88	3.7
14		39		64		89	
15		40		65	2.3	90	3.9
16		41		66		91	
17		42	1.5	67		92	4.2
18		43		68		93	4.4
19		44		69		94	4.6
20	0.9	45		70	2.5	95	4.8
21		46		71		96	5.0
22		47		72		97	5.4
23		48		73		98	5.8
24		49	1.7	74	2.7	99	6.9
25		50		75		100	8.0

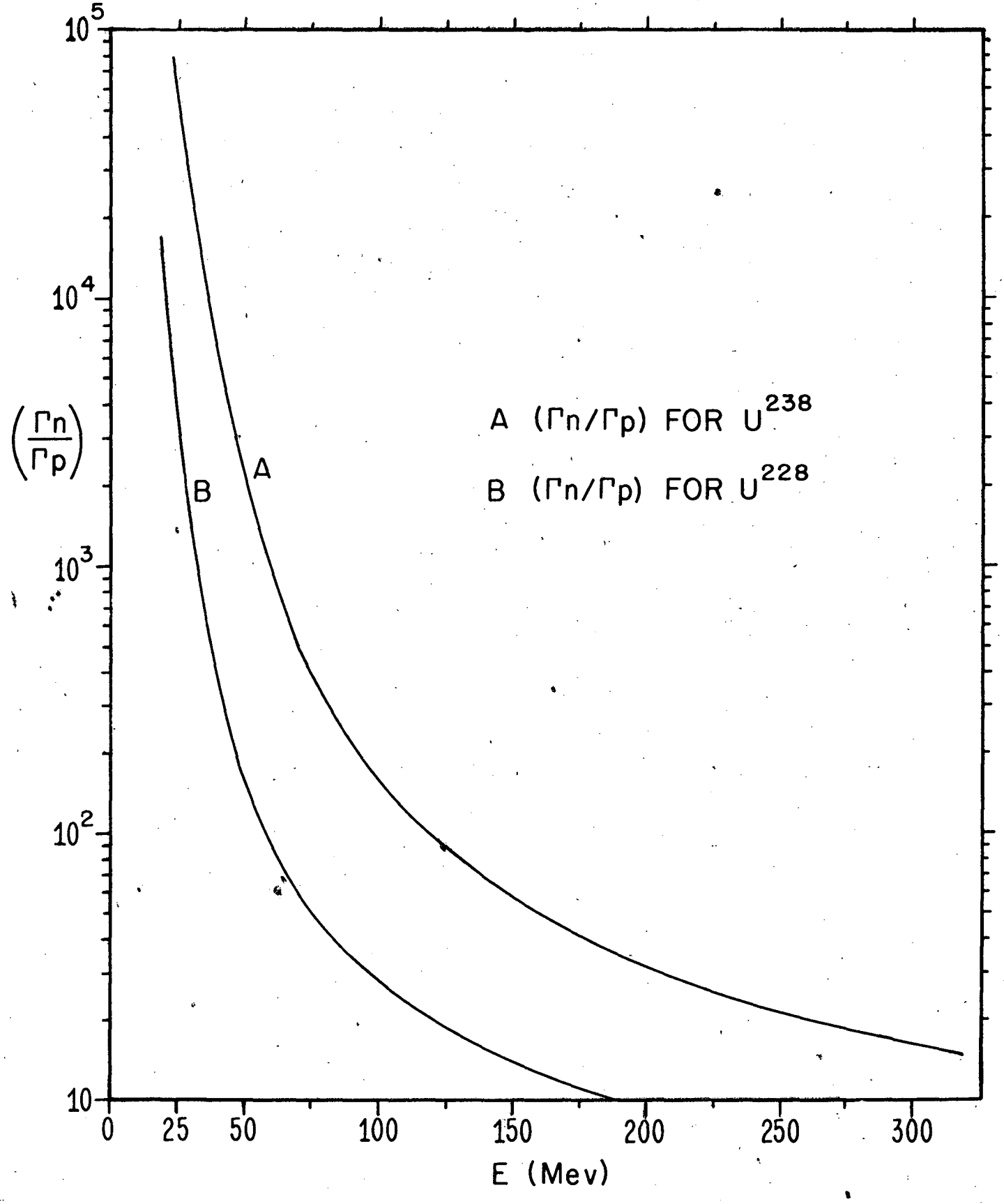
10^5
10^4
10^3
10^2
10
$\left(\frac{\Gamma n}{\Gamma p}\right)$
A
B
A $(\Gamma n/\Gamma p)$ FOR U^{238}
B $(\Gamma n/\Gamma p)$ FOR U^{228}
0
25
50
100
150
200
250
300
E (Mev)

Fig. 1

22019-1

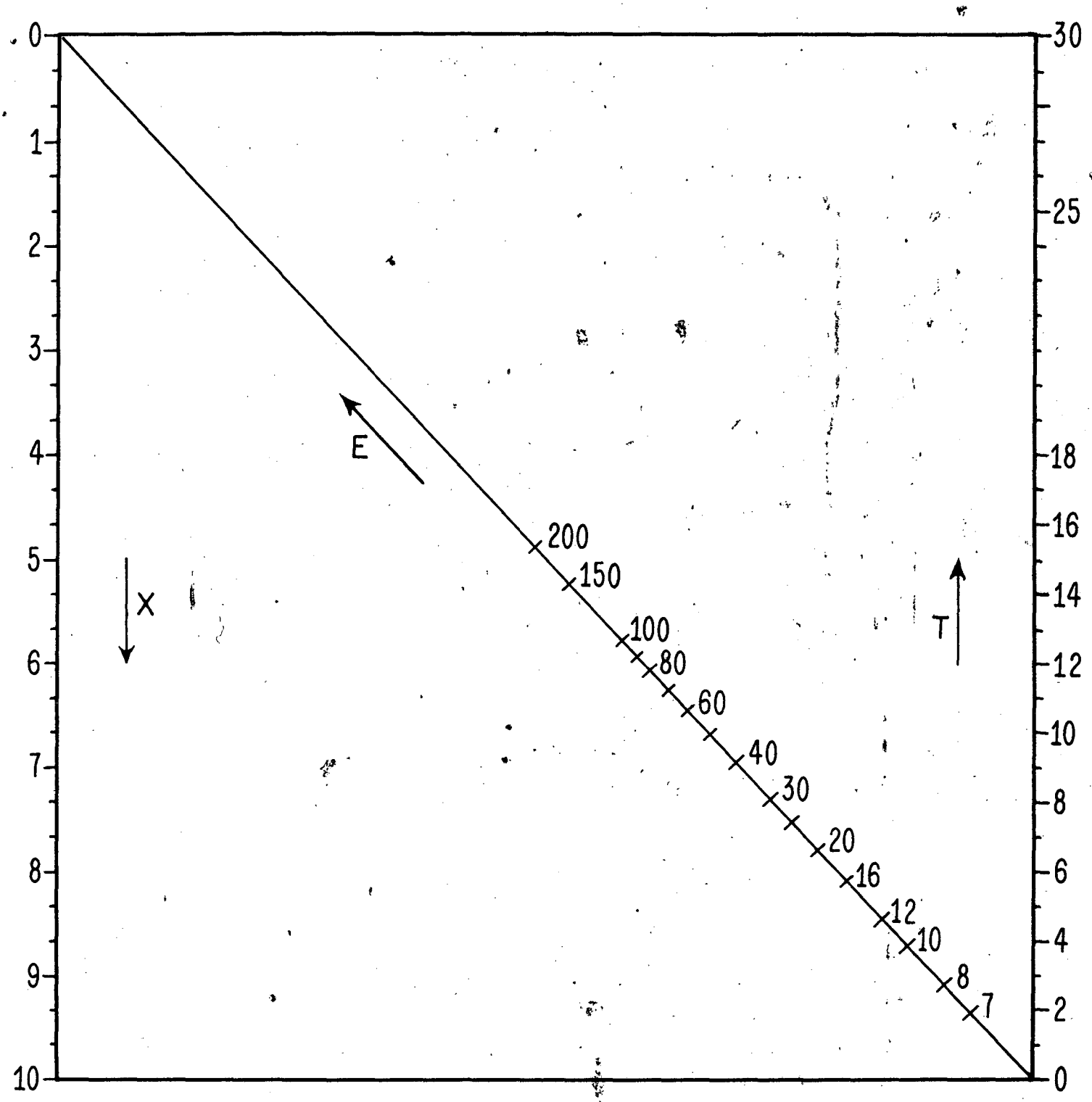
0
1
2
3
4
5
6
7
8
9
10
30
25
18
16
14
12
10
8
6
4
2
0
E
X
T
200
150
100
80
60
40
30
20
16
12
10
8
7

Fig 2

22016-1

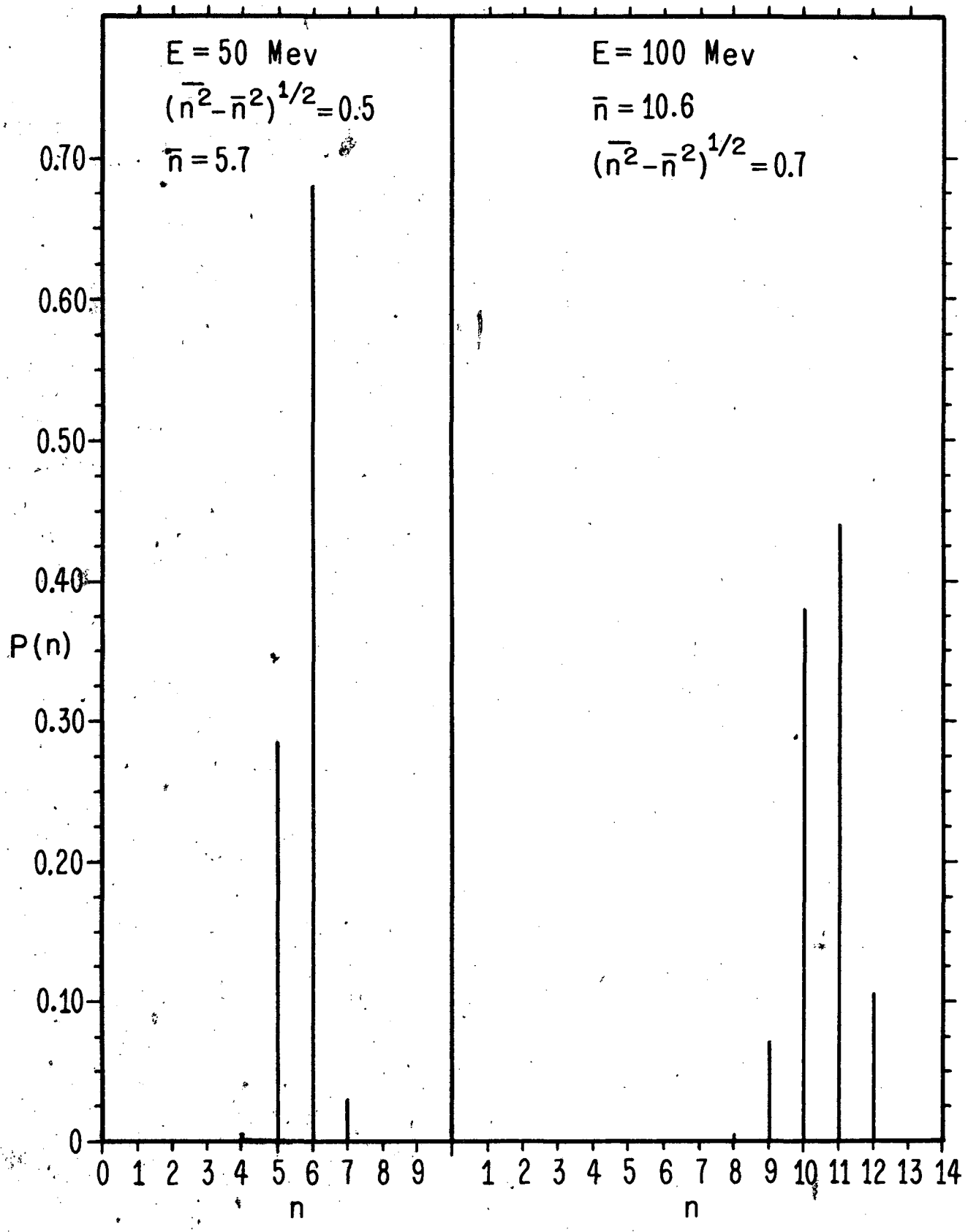
E = 50 Mev
$(\overline{n^2}-\bar{n}^2)^{1/2}=0.5$
$\bar{n}=5.7$
E = 100 Mev
$\bar{n}=10.6$
$(\overline{n^2}-\bar{n}^2)^{1/2}=0.7$
P(n)
0.70
0.60
0.50
0.40
0.30
0.20
0.10
0
0 1 2 3 4 5 6 7 8 9
1 2 3 4 5 6 7 8 9 10 11 12 13 14
n
n

Fig. 3

22017 – 1

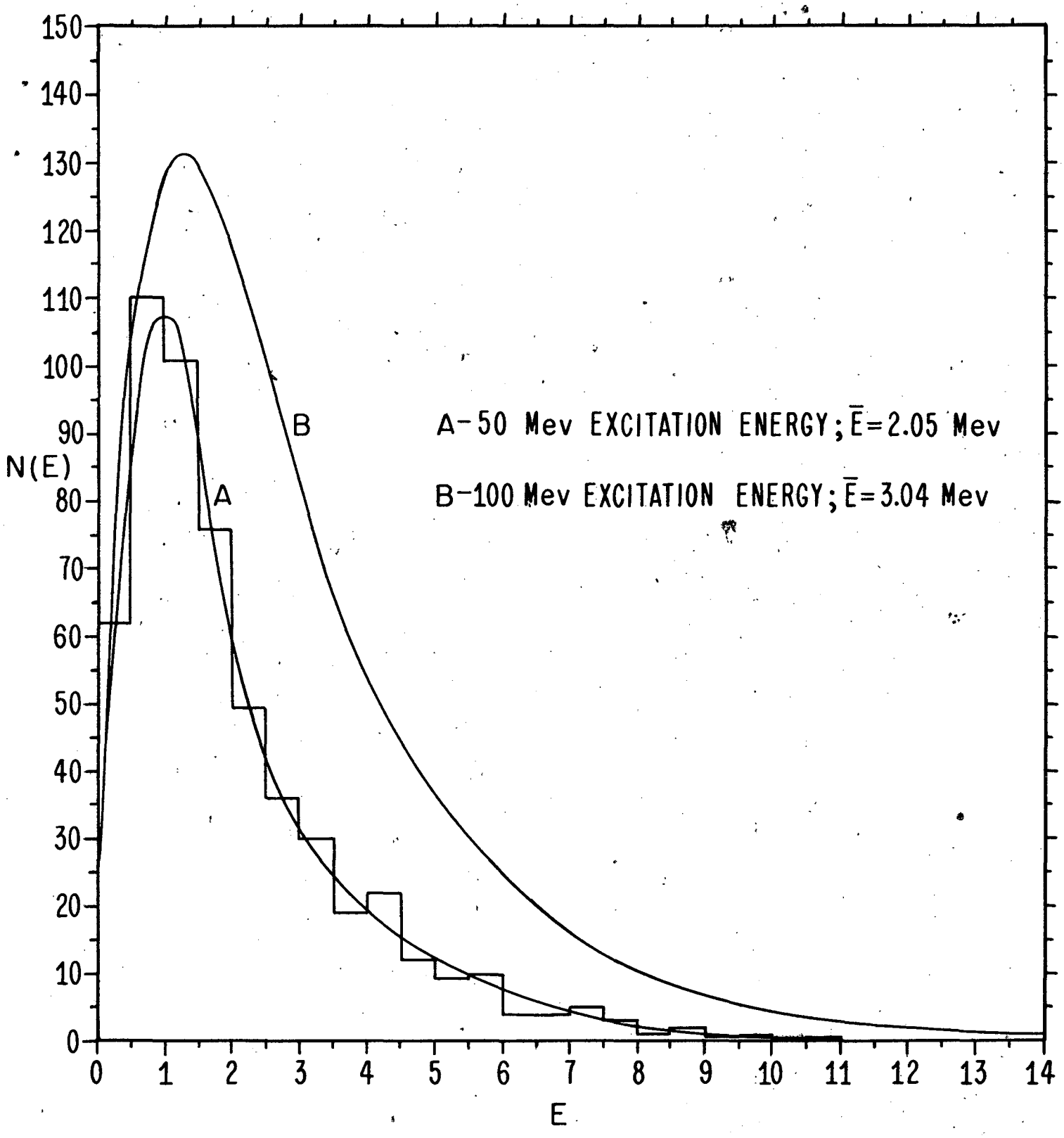
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
N(E)
0
1
2
3
4
5
6
7
8
9
10
11
12
13
14
E
A
B
A-50 Mev EXCITATION ENERGY; $\bar{E}$=2.05 Mev
B-100 Mev EXCITATION ENERGY; $\bar{E}$=3.04 Mev

Fig. 4

22018-1

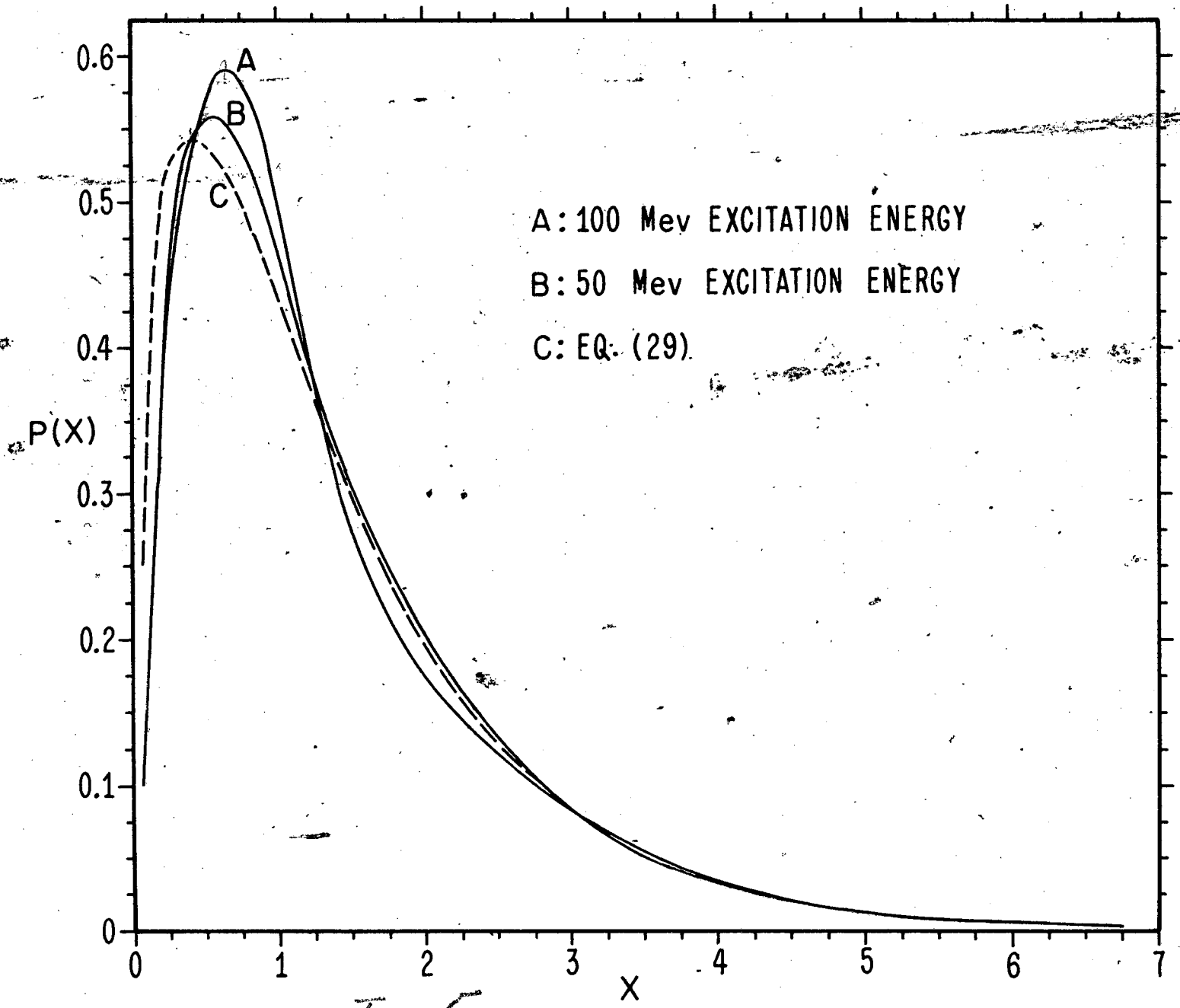
A
B
C
A: 100 Mev EXCITATION ENERGY
B: 50 Mev EXCITATION ENERGY
C: EQ. (29)
P(X)
X
0.6
0.5
0.4
0.3
0.2
0.1
0
0
1
2
3
4
5
6
7

Fig. 5

22015-1

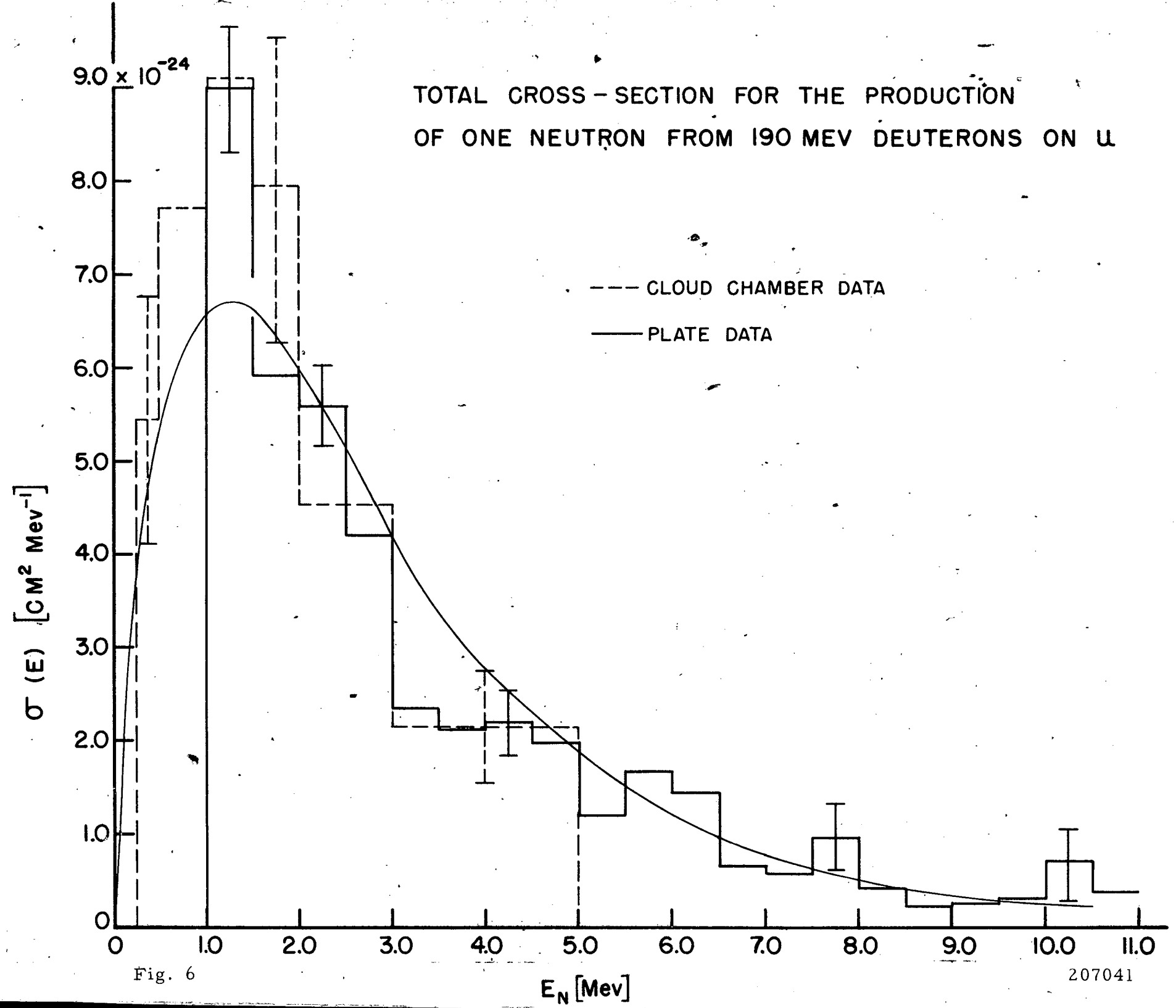
TOTAL CROSS-SECTION FOR THE PRODUCTION
OF ONE NEUTRON FROM 190 MEV DEUTERONS ON U
CLOUD CHAMBER DATA
PLATE DATA
9.0 x 10^-24
8.0
7.0
6.0
5.0
4.0
3.0
2.0
1.0
0
σ (E) [CM² Mev⁻¹]
0
1.0
2.0
3.0
4.0
5.0
6.0
7.0
8.0
9.0
10.0
11.0
E_N [Mev]

Fig. 6

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