# Ratios of disintegration rates for distinct decay modes of an excited nucleus

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(Received 18 May 2008; published 12 November 2008)

This paper examines a prevalent departure from the standard transition-state treatment of $\Gamma_n/\Gamma_f$, the relative rate of disintegration of an excited nucleus by neutron emission or fission. This departure is caused by what we believe is an erroneous treatment of shell structure corrections. According to the transition-state theory the shell correction in the excited compound nucleus cancels out identically in the ratio $\Gamma_n/\Gamma_f$, whereas in the deviant treatment it leads to an energy-dependent fission barrier that modifies the expression for the partial width $\Gamma_f$. Moreover, according to the transition-state theory, the partial width $\Gamma_n$ depends on the shell effect in the residual nucleus that emitted the neutron, whereas in the deviant treatment this dependence is ignored. We illustrate explicitly the magnitude of the errors that the deviant treatment of $\Gamma_n/\Gamma_f$ generates in typical nuclear reactions, errors that can reach orders of magnitude at low excitation energies. We take the opportunity to describe an accurate algebraic method of evaluating integrals over shell-affected level densities that appear in the transition-state theory. We also present a new derivation of Weisskopf's nucleon evaporation formula, based on the transition-state method rather than on the statistical principle of detailed balance used by Weisskopf. This unifies the theoretical treatments of fission and nucleon evaporation.

DOI: 10.1103/PhysRevC.78.054604 PACS number(s): 24.10.Pa, 24.60.Dr, 24.75.+i, 25.70.Gh

## I. INTRODUCTION

An important branch of nuclear physics is concerned with estimating the disintegration rates of an excited nucleus by one or more distinct decay modes. A typical example with wide-ranging applications deals with estimating the ratio of the decay rates by neutron (or other particle) emission and fission. A statistical estimate of such ratios, based on the "transition-state" theory of chemical reaction rates was introduced in 1939 by Bohr and Wheeler [1]. This theory has served as a starting point for a number of refinements, but the underlying physical framework has not changed. For some time, however, a number of publications have appeared [2–22] in which the transition-state formalism is applied in what seems to be a modified, not to say erroneous, way. These are publications concerned with shell structure corrections to integrals over nuclear level densities that enter in the transition-state theory. A conceptual error appears to be involved, which leads to formulas for the ratio of neutron to fission decay rates that no longer agree with those of the transition-state theory.

The purpose of the present paper is to rederive in a few lines the relevant transition-state decay rate formulas, thus identifying the aforementioned error, and to describe how to introduce shell structure corrections in a way consistent with that theory. We will illustrate typical deviations from transition-state predictions that are present in a number of recent publications. We will also describe a novel accurate algebraic method of evaluating certain complicated integrals over nuclear level densities, in particular those introduced by Ignatyuk *et al*. to accommodate shell structure corrections [23]. In an appendix we will provide a new derivation of Weisskopf's nucleon evaporation formula [24] by using the transition-state theory rather than the statistical principle of detailed balance, thus unifying the theoretical treatments of fission and particle emission.

We hope that our paper will help to eliminate from future publications unjustified departures from the transition-state theory's description of the competition between particle emission and fission.

## II. TRANSITION-STATE THEORY

We are concerned with a physical system (be it an excited chemical compound or an atomic nucleus) whose potential energy considered as a function of the system's degrees of freedom can be approximated as a multidimensional potential energy hollow surrounded by a mountain range that ensures the system's (meta)stability against disintegration. There may be one or more saddle point passes in the mountain range, corresponding to different modes of disintegration of the system. The transition-state theory of disintegration rates is based on introducing a microcanonical ensemble of $\rho(E - V_g)\Delta E$ systems, where $\rho(E - V_g)$, to be abbreviated simply as $\rho$ in what follows, is the level density of the system at the excitation energy $E - V_g$ above the system's ground state $V_g$. [We shall denote by the symbol $V$ the potential energy of a system's configutation, with $V_g$ for ground state and $V_s$ for saddle. The symbol $V$ may be thought of as the configuration's mass (or mass excess) in energy units.] A calculation is then

 

performed of the number of systems that disintegrate per unit time. The ratio

$$\frac{1}{t} = \frac{\text{number of disintegrations per unit time}}{\text{total number of systems } [=\rho\Delta E]} \tag{1}$$

is equal to the reciprocal of the average disintegration lifetime $t$, and $\hbar/t$, where $\hbar$ is Planck's constant divided by $2\pi$, is the partial disintegration width $\Gamma$.

The number of disintegrations per unit time is calculated by focusing attention on the microcanonical ensemble near the saddle-point configuration. The total number of degrees of freedom of the system (say $n$) is imagined to be split into two sets: a single degree of freedom $q$ in the disintegration direction and the remaining $n-1$ degrees of freedom, which, in the terminology of chemical reaction rates, constitute the "activated complex." If $p$ is the momentum conjugate to $q$ then the density of states in the two-dimensional phase space $(p, q)$ is $g/h$, where $g$ is the degeneracy of the state in question (two in the case of neutrons with spin 1/2). The number of states of the total system with $q$ in the interval $\mathrm{d}q$, $p$ in the interval $\mathrm{d}p$, and energy in the interval $\Delta E$ is then

$$gh^{-1}\rho_s(x)\Delta E\,\mathrm{d}p\,\mathrm{d}q, \tag{2}$$

where $\rho_s(x)$ is the level density of the activated complex at excitation $x$. This excitation is given by

$$x = E - V_s - K, \tag{3}$$

where $V_s$ is the potential energy of the relevant saddle point and $K$ is the kinetic energy of the system in the disintegration degree of freedom $q$. We note the relations

$$K = p^2/2m, \tag{4}$$

$$\mathrm{d}K = (p\,\mathrm{d}p)/m = v\,\mathrm{d}p = -\mathrm{d}x, \tag{5}$$

where $v$ is the velocity and $m$ is the effective mass associated with $q$. (The effective mass will not appear in the final rate formula.)

The subspace $(p, q)$ is displayed in Fig. 1. The line $q = 0$ corresponds to the location of the saddle point. (In the case of neutron emission, when a well-defined saddle point does not exist for finite $q$, $q = 0$ locates the neutron's "point of no return" where its interaction with the emitting nucleus has become ineffective.) The possible values of $p$ range from $-P$ to $+P$, where

$$P^2/2m = E - V_s. \tag{6}$$

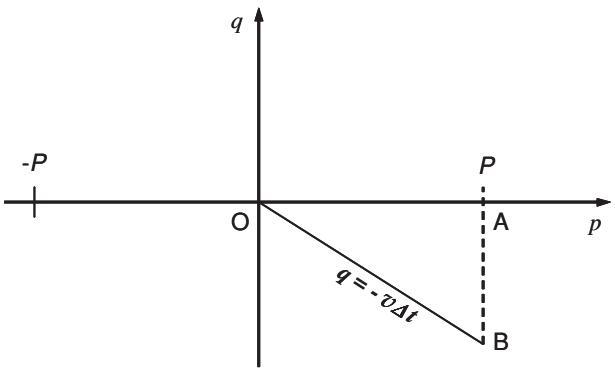


FIG. 1. Phase space of the disintegration degree of freedom $q$ and its conjugate momentum $p$.

Consider now a time interval $\Delta t$. The number of representative points in the full phase space that cross the saddle point or point of no return at $q = 0$ in the positive direction (and, in the simplest version of the transition-state theory, are assumed to lead to disintegration in the time $\Delta t$) is given by $\rho_s(x)\Delta E$ multiplied by the number of systems inside the triangle OAB in Fig. 1, defined by $0 < p < P$ and $0 < q < -v\Delta t$; thus,

$$\begin{aligned}\int_0^P \mathrm{d}p \int_{-v\Delta t}^0 \mathrm{d}q\; gh^{-1}\rho_s(x)\,\Delta E &= gh^{-1}\Delta t\;\Delta E\int_0^P \mathrm{d}p\; v\;\rho_s(x)\\ &= gh^{-1}\Delta t\;\Delta E\int_0^{E-V_s} \mathrm{d}x\;\rho_s(x),\end{aligned} \tag{7}$$

where we have made use of Eqs. (4)–(6).

The number of disintegrations per unit time follows by dividing by $\Delta t$, and the reciprocal of the lifetime by further division by $\rho\Delta E$. Hence the final canonical transition-state lifetime and decay width formulas are

$$1/t = N/h\rho, \tag{8}$$

$$\Gamma = N/2\pi\rho, \tag{9}$$

where

$$N = g\int_0^X \mathrm{d}x\;\rho_s(x) \tag{10}$$

is the number of levels ("decay channels") of the saddle-point configuration in the slot of excitation energies $E - V_s$, which we denote by $X$.

## III. RATIO OF TWO DECAY WIDTHS

Consider in particular the ratio of the neutron decay width $\Gamma_n$ to the fission decay width $\Gamma_f$. Writing $\Gamma_n = N_n/2\pi\rho$ and $\Gamma_f = N_f/2\pi\rho$ we obtain the central transition-state formula for the ratio of two partial decay widths,

$$\Gamma_n/\Gamma_f = N_n/N_f, \tag{11}$$

where $N_n$ and $N_f$ are the numbers of levels (decay channels) of the two saddle configurations in the excitation energy slots $E - V_n$ and $E - V_f$, where $V_n$ and $V_f$ are the potential energies of the two saddle points (or points of no return) in question. Note that although the individual partial decay widths depend on the compound nucleus level density $\rho$, in their ratio this factor cancels out identically. Note also that if, in evaluating $N_n$ in Eq. (11), one were to use Eq. (10) as it stands, this would correspond to adopting the "symmetrical" expression for $\Gamma_n/\Gamma_f$, discussed in Ref. [25]. For the sake of clarity we shall denote by $\mathcal{N}_n$ this symmetrical version and by $N_n$ the commonly used formula for the number of neutron decay channels, namely,

$$N_n = \frac{2gm\sigma}{\pi\hbar^2}\int_0^{X_n} \mathrm{d}x\;(X_n - x)\,\rho_n(x). \tag{12}$$

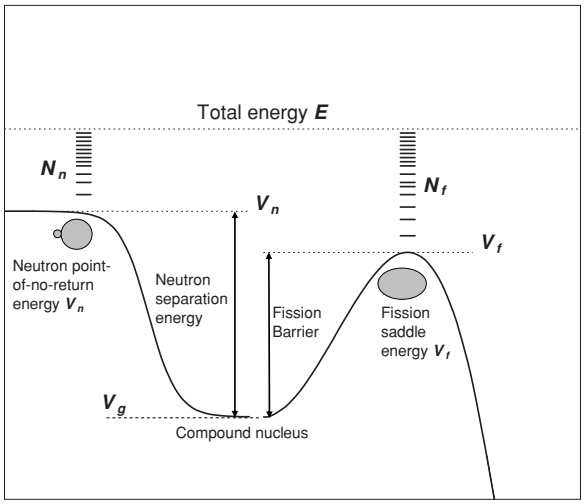


FIG. 2. The decay of a nucleus by two distinct disintegration modes. The compound nucleus, with ground-state potential energy (or ground-state mass in energy units) $V_g$, can decay by way of a fission saddle point (with potential energy $V_f$) on the right and, on the left, by a neutron point of no return with potential energy $V_n$. The total energy of the excited compound nucleus will be denoted by $E$. (In a nuclear reaction it is equal to the sum of the potential energies or masses in energy units of the target and projectile, augmented by the center-of-mass collision energy.) The ratio $\Gamma_n/\Gamma_f$ of the two decay widths is equal to the ratio of the number of levels (decay channels) $N_n$ and $N_f$ in the two energy slots $E - V_n$ and $E - V_f$, respectively. The barrier for fission, $B_f$, is equal to $V_f - V_g$ and the barrier for neutron emission, $B_n$ (the neutron separation energy), is equal to $V_n - V_g$. The excitation energy of the compound nucleus is $E - V_g$, often denoted by $E^*_{\rm CN}$.

Here $m$ is the neutron mass and $\sigma$ stands for the cross section for neutron capture, the inverse of neutron evaporation. This form of the equation for $N_n$ follows from Weisskopf's nucleon evaporation theory. As shown in Appendix B the appearance of the extra factor $(2\,{\rm gm}\,\sigma/\pi\hbar^2)\,(X_n - x)$ in Eq. (12) results from singling out the neutron's two transverse degrees of freedom (those orthogonal to the disintegration direction) for special treatment, rather than combining them with the degrees of freedom of the remainder of the activated complex, treated in terms of a statistical level density.

Figure 2 illustrates the quantities that enter in the ratio of the partial decay widths for neutron emission and fission. For a given total energy $E$ of the compound nucleus this ratio requires knowledge of the potential energies of the fission saddle point and of the neutron's point of no return, as well as of the level densities of these configurations in the slots $E - V_f$ and $E - V_n$. Note in particular that, as already mentioned, this ratio does not involve the compound nucleus level density $\rho$. More generally, the ratio $\Gamma_n/\Gamma_f$ knows nothing about *any* property of the compound nucleus, such as the height of its fission barrier $V_f - V_g$ or the value of its neutron separation energy $V_n - V_g$. The middle portion of Fig. 2 could, in fact, be erased without loss of any information relevant for the calculation of $\Gamma_n/\Gamma_f$. We stress this elementary feature of the transition-state theory because, as we shall see in the following, the error in the aforementioned publications [2–22] is immediately revealed by the dependence of the erroneous formulas on the fission barrier height. The next section will trace the origin of this error, which is related to the incorrect introduction of shell structure corrections to the relevant nuclear level densities.

We note in passing that since the ratio of two decay widths does not depend on the ground-state energy $V_g$, nor on the compound nucleus level density $\rho$ or the fission barrier $B_f$, it is often a practical advantage to estimate the saddle-point energy $V_f$ without reference to the irrelevant quantities $V_g$ and $B_f$. For heavy and superheavy nuclei, measured values or good estimates of $V_g$, $B_f$, and the ground-state shell correction may not be available, whereas a fair estimate of $V_f$ may nevertheless be made. This is due to the "topographic theorem" [26], confirmed by extensive data, which shows that saddle-point masses are much less sensitive to shell effects than ground-state masses and fission barriers. As a result, a relatively simple macroscopic calculation of $V_f$ without shell corrections can give a fair first estimate of this quantity.

## IV. THE ERROR

The number $N_f$ of fission decay channels, which determines the fission decay width $\Gamma_f$, will be taken as

$$N_f = \int_0^{X_f} {\rm d}x\, \rho_f(x), \tag{13}$$

where $X_f = E - V_f$. We note again that this expression makes no reference to the *height* of the fission barrier $B_f$. However, the energy $E$ is often assumed, implicitly or explicitly, to be specified with respect to the ground state of the compound nucleus, in which case the upper limit of integration can be rewritten in terms of $B_f$ as $(E - V_g) - (V_f - V_g) = E^* - B_f$, where $E^*$ stands for $E - V_g$ (also denoted by $E^*_{\rm CN}$ in the literature), the excitation energy of the compound nucleus. Thus

$$N_f = \int_0^{E^* - B_f} {\rm d}x\, \rho_f(x). \tag{14}$$

Now when a ground-state shell correction $D$ is present, $V_g$ can be written as

$$V_g = V_{\rm LD} + D, \tag{15}$$

where $V_{\rm LD}$ stands for the macroscopic (liquid-drop-like) part of the ground-state energy. This does not affect the validity of Eq. (14), but a subtle error may creep in when the shell correction is considered to "depend on the excitation energy," and such a dependence (a damping out of the correction with increasing excitation) is inserted in Eq. (15):

$$V_g(E^*) = V_{\rm LD} + D(E^*). \tag{16}$$

The barrier $B_f$ now seems to acquire an energy dependence according to

$$B_f(E^*) = V_f - V_g = B_{\rm LD} - D(E^*), \tag{17}$$

where $B_{\rm LD} = V_f - V_{\rm LD}$ is the macroscopic fission barrier. When Eq. (17) is inserted in Eq. (14) the erroneous conclusion might be drawn that the number of fission decay channels and

the partial fission decay width $\Gamma_f$ increase as the negative shell correction $D(E^*)$ becomes less negative with excitation, the barrier $B_f$ decreases, and the range of integration in Eq. (14) increases. This would become especially noticeable in the case of heavy nuclei, where $B_{\rm LD}$ is close to zero and the vanishing of $D(E^*)$ would imply the vanishing of the fission barrier. Such a conclusion is wrong. The origin of the error is Eq. (16), which implies that the *ground-state potential energy* (or *mass*) of the compound nucleus is affected by excitation. One needs to remember that the expression "excitation-dependent shell correction" is shorthand for describing the circumstance that, with increasing excitation, the effect of a shell correction on the density of levels becomes less and less important relative to the main macroscopic contribution to the level density. To describe this phenomenon as a modification of the ground-state mass of a nucleus has no basis whatever. A ground state is a *ground* state and it knows nothing about the decreasing effect of shell structure on the level density with increasing excitation. Thus Eq. (16) should be restored to its original form $V_g = V_{\rm LD} + D$ and Eq. (17) should read $B_f = B_{\rm LD} - D$, where $D$ is the *energy-independent* ground-state shell correction.

The simplest way to avoid this kind of error is to use Eq. (13), which does not mention $V_g$ or $B_f$ in the first place.

## V. CORRECT AND INCORRECT FORMULAS FOR $\Gamma_n/\Gamma_f$

By using Eq. (12) for $N_n$ and Eq. (13) for $N_f$ the ratio $\Gamma_n/\Gamma_f$ becomes

$$\frac{\Gamma_n}{\Gamma_f} = \frac{2gm\sigma}{\pi\hbar^2}\frac{\int_0^{X_n} {\rm d}x\,(X_n - x)\,\rho_n(x)}{\int_0^{X_f} {\rm d}x\,\rho_f(x)} = \frac{2gm\sigma}{\pi\hbar^2}\frac{\int_0^{X_n} {\rm d}x\,(X_n - x)\,\exp[s_n(x)]}{\int_0^{X_f} {\rm d}x\,\exp[s_f(x)]}, \qquad (18)$$

where we have written the level densities $\rho(x)$ in terms of the corresponding entropies $s(x)$.

To make contact with the frequently quoted equation (VII-7) in Ref. [27] for $\Gamma_n/\Gamma_f$ appropriate for a Fermi gas without shell corrections we evaluate the integrals in Eq. (18) to leading order in the entropies, using $s_n(x) = 2\sqrt{a_n x}$ and $s_f(x) = 2\sqrt{a_f x}$. The level density parameters $a_{n,f}$ are macroscopic coefficients independent of excitation. We find

$$\frac{\Gamma_n}{\Gamma_f} = \frac{2gmr_0^2A^{2/3}}{\hbar^2}\frac{X_n}{a_n}\sqrt{\frac{a_f}{X_f}}\exp\left[2(\sqrt{a_n X_n} - \sqrt{a_f X_f})\right], \qquad (19)$$

where we have written $\sigma$ as $\pi r_0^2 A^{2/3}$ in terms of a radius constant $r_0$ and the nuclear mass number $A$. This equation, which is the starting point of many current discussions of the ratio $\Gamma_n/\Gamma_f$, agrees with Eq. (VII-7) in Ref. [27] taken consistently to leading order in the entropies (i.e., by disregarding the $-1$ in the denominator in Eq. (VII-7) in Ref. [27]). It also agrees with Eq. (6) in Ref. [2], to which reference we shall return presently. For what is to follow note that the fission barrier appears in Eq. (19) in the excitation energy $X_f$ given by $E - V_g - B_f$, or $E^*_{\rm CN} - B_f$ in the notation of Ref. [2], but this dependence of $\Gamma_n/\Gamma_f$ on $B_f$ is spurious, since $E^*_{\rm CN} - B_f = E - V_f$, as noted in Sec. IV.

Shell corrections to the level densities may be taken into account approximately by using Fermi gas entropies modified by the formula due to Ignatyuk *et al.* [23]:

$$s_{n,f}(x) = 2\sqrt{a_{n,f}}\sqrt{x + D_{n,f}[1 - \exp(-x/d)]}, \qquad (20)$$

where $D_{n,f}$ are the shell corrections to the saddle energies $V_{n,f}$, and $d$ is a shell damping parameter. The integrals in Eq. (18) can no longer be evaluated in closed form and either numerical integrations or the accurate approximations outlined in Sec. VI have to be used to calculate the relevant values of $N_n$, $N_f$, and their ratio.

This would be the standard transition-state procedure. What is done instead in Ref. [2] and in a number of subsequent references is to argue as follows: "Since the fission barrier $B_f$ in the heaviest nuclei is defined by the shell correction, its value depends on the excitation energy $E^*_{\rm CN}$ of the compound nucleus as

$$B_f = B_f(E^*_{\rm CN}{=}0)\cdot\exp(-E^*_{\rm CN}/E_d), \qquad (21)$$

where $E_d$" ... "is the shell-damping energy." The resulting energy-dependent fission barrier is then inserted in the excitation energy in Eq. (19) according to $X_f = E^*_{\rm CN} - B_f$.

It is to be expected that the values of $\Gamma_n/\Gamma_f$ deduced in this way will disagree with the predictions of the transition-state theory. The disagreement actually concerns *two* features. First, as explained in Sec. IV, an energy-dependent fission barrier arises from an unjustified assumption of an energy dependence of the ground-state energy of the compound nucleus, the bottom of the barrier. (And certainly the *top* of the barrier—the energy of the fission saddle—cannot depend on a shell effect in the level density of the compound nucleus.) Second, the existence of a shell correction in the neutron emission rate, an integral part of the transition-state formula for $\Gamma_n/\Gamma_f$, is ignored altogether. We are thus presented with a very different scenario from that of the transition-state theory: The fission decay rate is made to depend on the shell effect in the ground state rather than in the saddle configuration, and the shell effect on the level density at the neutron saddle (point of no return) is disregarded. It is true that these two errors act in opposite directions, and the authors of Ref. [4] actually attempt on p. 4 to justify the use of Eq. (19) with an energy-dependent fission barrier as a rough estimate of $\Gamma_n/\Gamma_f$. To us it seems a pity to encourage in this way the use of an intrinsically flawed treatment of shell effects, rather than to recall the standard unambiguous transition-state Eq. (18) to be used, when shell effects are present, with the entropies given by Eq. (20).

In the following we give results of numerical calculations that demonstrate the serious deviations from the transition-state values of $\Gamma_n/\Gamma_f$ that result from using a combination of Eq. (19) with an energy-dependent fission barrier. First we focus on Ref. [7], representative of the series of papers [2–11]. In Fig. 3 we show a comparison of the $\Gamma_n/\Gamma_f$ ratios for the decay of a typical heavy nucleus $^{266}$Hs produced in the reaction $^{58}$Fe + $^{208}$Pb, calculated according to the prescription of Ref. [7] with results of the correct (numerical) integration of Eq. (18) [with the Ignatyuk entropies of Eq. (20) included]. For the latter calculation we used the fission saddle energy

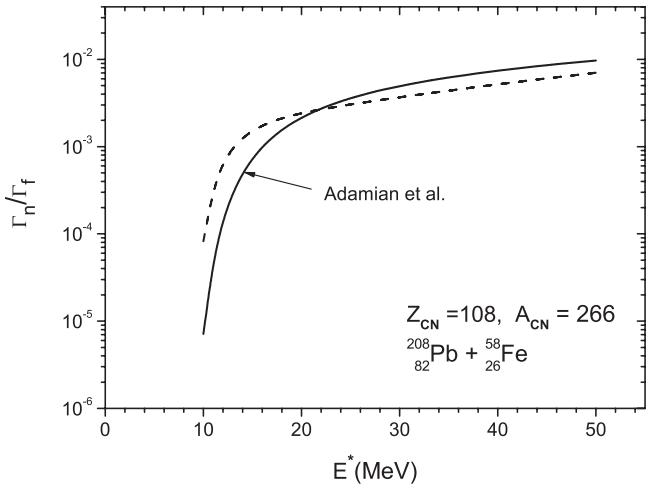


FIG. 3. The ratio $\Gamma_n/\Gamma_f$ for the $^{266}$Hs nucleus obtained by using the scheme and parameters of Refs. [2–11] with an excitation-energy-dependent fission barrier (solid curve) compared with the ratio $\Gamma_n/\Gamma_f$ calculated with the transition state method [Eqs. (18) and (20)] with precise numerical integrations over level densities, for parameters as in Ref. [7], but for a standard value of the shell damping parameter $d = 18.5$ MeV (dashed curve).

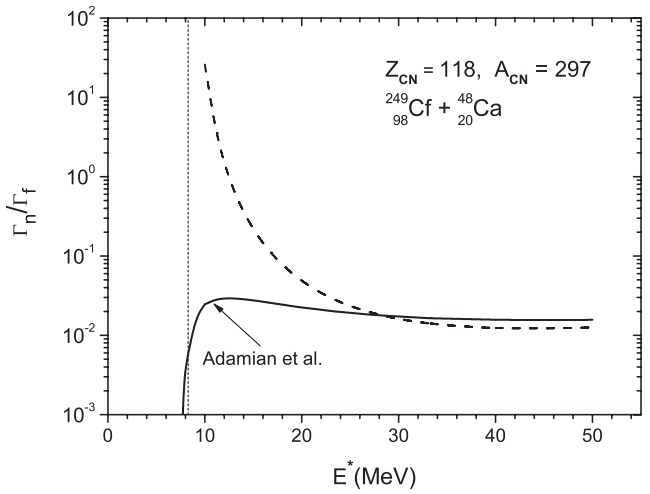


FIG. 4. Same as Fig. 3 but for a compound nucleus $^{297}$118 formed in the $^{48}$Ca + $^{249}$Cf reaction. The excitation energy threshold $E^* = V_f - V_g = 8.27$ MeV is indicated by the vertical dashed line.

corresponding to the value $B_f(E^*_{CN}=0) = -D$ assumed in Ref. [7], and we took the other parameters $a_n$, $a_f$, and pairing corrections exactly as given in Ref. [7]. For compound nucleus excitations $E^*$ from 21 to 50 MeV the authors of Ref. [7] overestimate $\Gamma_n/\Gamma_f$ by an amount that levels out at about 50%. For energies below 21 MeV this turns into an underestimate, which reaches an order of magnitude at $E^* = 10$ MeV. This low-energy underestimate is especially serious because it affects not only low-energy "cold fusion" reactions but also reactions that start at high excitations, since a de-excitation cascade must eventually pass through the low-energy region. The reason why both curves in Fig. 3 precipitously tend to zero for low energies is that $\Gamma_n$ tends to zero at the threshold for neutron emission given by $E^* = V_n - V_g = B_n = 8.22$ MeV. (The nuclear masses underlying this and the following estimates are taken from Ref. [28].)

In Fig. 4 we show a similar comparison for the reaction $^{48}$Ca + $^{249}$Cf leading to the compound nucleus $^{297}$118. In this case the estimated fission saddle energy $V_f$ is higher than $V_n$ and the excitation energy threshold for the reaction is determined by $E^* = V_f - V_g = B_f = 8.27$ MeV. But now the approach of $\Gamma_n/\Gamma_f$ (calculated correctly) to the threshold is dramatically different: The curve explodes toward infinity as the probability for fission tends to zero whereas that for neutron emission remains finite. And here emerges a disaster for the scenario in which the fission barrier is made erroneously energy dependent. With the fission barrier reduced by excitation, the incorrectly calculated curve for $\Gamma_n/\Gamma_f$ is allowed to continue to energies below the threshold set by the fission saddle energy $V_f$ (i.e., to excitation energies below $V_f - V_g$!). According to this scenario the threshold for the reaction is determined by the point where the excitation-dependent barrier $B_f(E^*)$ has become equal to the excitation, namely at $E^* = 6.40$ MeV according to Eq. (21). This is well below the true threshold at 8.27 MeV. This fact shows at once, even without reference to the transition-state method, that the assumption of an excitation-dependent fission barrier is not acceptable. As seen from Fig. 4, this assumption leads to predictions that at low excitation energies may deviate from the correct $\Gamma_n/\Gamma_f$ values by a factor of 1000 or more. As already mentioned, these low-energy errors affect all de-excitation cascades, including those starting at higher excitation energies.

In three recent publications [20–22] the scheme with an energy-dependent fission barrier was used with parameters somewhat different from those of Ref. [7]. Results of a comparison of the recipe used in Refs. [20–22] with correctly calculated $\Gamma_n/\Gamma_f$ ratios are shown in Figs. 5 and 6. The calculations are presented for the same pair of compound nuclei as in the comparisons with Ref. [7], namely for $^{266}$Hs and $^{297}$118. The correct curves are calculated with Eqs. (18) and (20) in two versions: assuming a standard value of Ignatyuk's shell damping parameter $d = 18.5$ MeV and also assuming $d = 5.48A^{1/3}/(1 + 1.3A^{-1/3})$ MeV, as used in Refs. [20–22].

These examples illustrate the range of errors resulting from the method of calculating the $\Gamma_n/\Gamma_f$ ratios used in Refs. [2–11, 20–22]. Similar errors may also be present in works of other groups, for example those of Refs. [12–19], because in all these papers an expression for an excitation-energy-dependent fission barrier, similar to Eq. (21), was used. Unfortunately, quantitative illustrations of the errors in Refs. [12–19] are not possible owing to insufficient information on details of the calculations.

## VI. EVALUATION OF INTEGRALS OVER LEVEL DENSITIES

In the previous section we arrived at the final integral expression for the $\Gamma_n/\Gamma_f$ ratio, Eq. (18), which includes Ignatyuk's energy-dependent shell corrections to the level

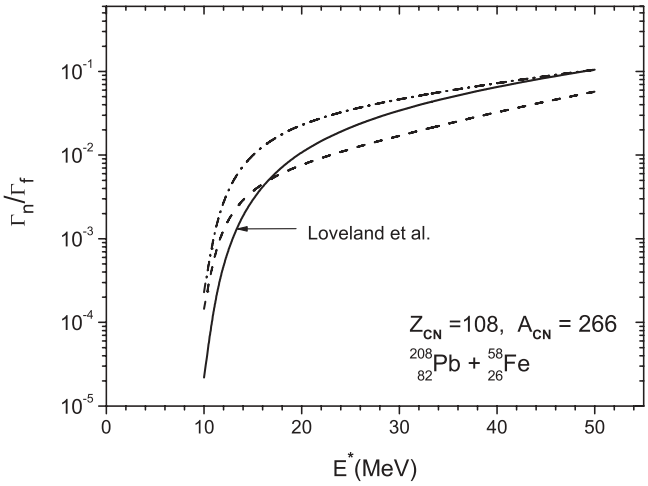


FIG. 5. The ratio $\Gamma_n/\Gamma_f$ for the $^{266}$Hs nucleus obtained by using the scheme and parameters of Refs. [20–22] with an excitation-energy-dependent fission barrier (solid curve) compared with the ratio $\Gamma_n/\Gamma_f$ calculated with the transition-state method [Eqs. (18) and (20)] with precise numerical integrations over level densities, for parameters as in Refs. [20–22], but for a standard value of the shell damping parameter $d = 18.5$ MeV (dashed line), and also for a value $d = 29.32$ MeV (dotted-dashed curve) assumed in Refs. [20–22].

densities given by Eq. (20). This expression cannot be evaluated in closed form. In the following we give simple and very accurate approximations to integrals for the relevant numbers of channels $N_f$, $N_n$, and $\mathcal{N}_n$. These approximations may be of considerable importance for speeding up the very time consuming Monte Carlo statistical calculations, especially in simulations of extremely low probability decay cascades that lead to the formation of superheavy elements.

As previously outlined we shall be concerned with two types of integrals: For fission, we have

$$N_f = C\int_0^X \mathrm{d}x e^{s(x)} \tag{22}$$

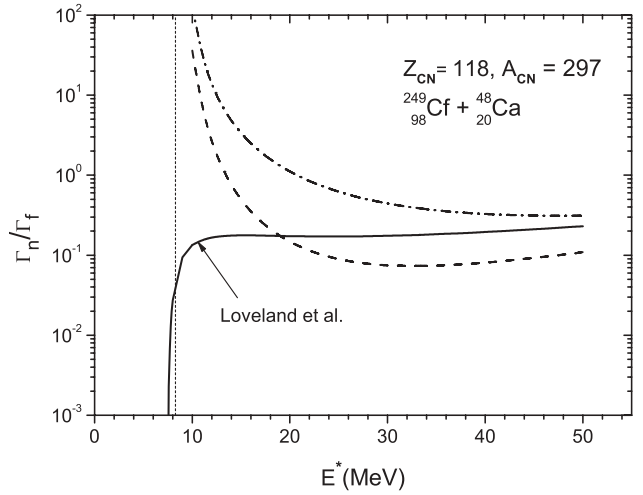


FIG. 6. Same as Fig. 5 but for a compound nucleus $^{297}$118 formed in the $^{48}$Ca + $^{249}$Cf reaction. The excitation energy threshold $E^* = V_f - V_g = 8.27$ MeV is indicated by the vertical dashed line.

and for neutrons either

$$\mathcal{N}_n = C\int_0^X \mathrm{d}x e^{s(x)} \tag{23}$$

or

$$N_n = C\int_0^X \mathrm{d}x (X-x) e^{s(x)}. \tag{24}$$

Here $C$ is an appropriate constant, $s(x)$ is the entropy (related to the logarithm of the level density), and $X$ is the maximum excitation energy, equal to $E - V_s$. (All these quantities may be different in the three cases considered.) As already mentioned, the appearance of the factor $(X - x)$ in Eq. (24) results from singling out for special treatment the neutron's two transverse degrees of freedom (those orthogonal to the disintegration direction) rather than combining them with the degrees of freedom of the remainder of the activated complex, treated in terms of a statistical entropy expression (see Appendix B). If this is *not* done and Eq. (23) is used, there results a "symmetrical" expression for $\Gamma_n/\Gamma_f$, discussed in Ref. [25]. Reference [29] provides experimental evidence for the superiority of the formula with the factor $X - x$ included.

In the simplest approximation to the level density of a degenerate Fermi gas we have

$$\rho(x) = C\, e^{2\sqrt{ax}}, \tag{25}$$

where $a$ is a level density parameter independent of excitation. In this case the integrals in Eqs. (22)–(24) can be evaluated in closed form. This integrability is usually spoiled by an energy-dependent "pre-exponential" factor in the level density as well as by shell effect corrections that modify the function $\sqrt{x}$ into a more complicated expression. Fortunately, both modifications, even though not necessarily small, do not change significantly the qualitative appearance of the dependence of $s$ on $x$ (or of $x$ on $s$). The result is that even in the general case the dependence of $x$ on $s$, which for a Fermi gas is a parabola centered on the origin, can be accurately approximated by a quadratic polynomial in $s$ passing through the origin (this time with a finite slope) and adjusted to have value and derivative equal to the value and derivative of the postulated entropy $s(x)$ at the upper limit of integration (from where an exponentially dominant contribution to the integral comes). In this approximation these integrals are readily evaluated by a change of variable from $x$ to $s$, with the result (see Appendix A)

$$\begin{aligned} N_f\ (\text{or}\,\mathcal{N}_n) &= CX\{e^S[(2+\epsilon)/S - 2/S^2] \\ &\quad -\epsilon/S + 2/S^2\}/(1+\epsilon) \\ &= CXe^S[(2+\epsilon)/S - 2/S^2]/(1+\epsilon) \end{aligned} \tag{26}$$

if the exponentially smaller terms are neglected. Here, $S \equiv s(X)$ and $\epsilon$ is a measure of the shell correction $D$ in Eq. (20). (Except for very light nuclei, $S$ is typically in the range 7 to 20, with $e^S$ of order $10^3$ to $10^9$.)

Similarly, when exponentially small terms are neglected, we find

$$N_n = Ce^S X^2[(2+\epsilon)^2/S^2 - 6(2+\epsilon)/S^3 + 12/S^4]/(1+\epsilon)^2. \tag{27}$$

In these equations the parameter $\epsilon$ is shown in Appendix A to be given by

$$\epsilon = (2 - \Lambda)/(\Lambda - 1), \tag{28}$$

where $\Lambda = S/XS'$ and $S' \equiv s'(X)$ is the derivative of $s(x)$ evaluated at the upper limit of integration.

What we have achieved is that, for a class of level density functions of interest, we have derived algebraic approximations (accurate to a couple of percent—see Appendix A) for the number of levels (decay channels) in a slot of excitation energies between zero and $X$. These approximations require only knowledge of the postulated level density and its derivative at $X$.

When the entropy $s(x)$ is that of a degenerate Fermi gas modified by a shell correction in the form suggested by Ignatyuk *et al.* [23] [see Eq. (20)], the quantities $S, S'$ are given by

$$S = 2\sqrt{a}\,\sqrt{X + D(1 - e^{-\chi})}, \tag{29}$$

$$S' = \frac{\sqrt{a}\,(1 + \delta e^{-\chi})}{\sqrt{X + D(1 - \mathrm{e}^{-\chi})}}, \tag{30}$$

and Eq. (28) leads to

$$\epsilon = \frac{2(-\delta)[1 - (1 + \chi)\mathrm{e}^{-\chi}]}{\chi + \delta[2 - (2 + \chi)e^{-\chi}]}, \tag{31}$$

where $\chi = X/d$ and $\delta = D/d$ are the values of $X$ and $D$ in units of $d$.

The parameter $\epsilon$, approximately proportional to $-\delta$, is seen to be a measure of minus the shell effect. For vanishing $\epsilon$ Eqs. (26) and (27) give the values of $N_f$ and $N_n$ for a simple degenerate Fermi gas.

## VII. SUMMARY AND CONCLUSIONS

We have shown that a commonly used scheme of calculating the ratio of neutron emission to fission, one that introduces an excitation-energy-dependent fission barrier, is inconsistent with the transition-state theory of nuclear disintegration rates. In some cases it even leads to unphysical predictions for the existence of fission at energetically forbidden subthreshold excitation energies. When shell effects are present the ratio of the erroneous predictions to those obtained by the transition-state method can reach orders of magnitude at low excitation energies.

We have included an account of an accurate algebraic treatment of shell effects, as well as a new derivation of Weisskopf's nucleon evaporation formula in which we use the transition-state method rather than the statistical principle of detailed balance.

## ACKNOWLEDGMENTS

This work was supported by the U.S. Department of Energy under Contract No. DE-AC0376SF00098 (LBNL) and by the Polish Ministry of Science and Higher Education under Grant MNiSW No. 1 P03B 090 29.

## APPENDIX A

To approximate an integral of the type

$$N = C \int_0^X \mathrm{d}x e^{s(x)}, \tag{A1}$$

where the entropy $s(x)$ is in the form of a modified Fermi gas expression that makes Eq. (A1) no longer integrable, write the dependence of $x(s)$ on $s$ as a quadratic polynomial in $s$ that passes though the origin and is adjusted so that it agrees in value and slope at $x = X$ with the postulated $s(x)$. Write the approximating polynomial in the form

$$\frac{x}{X} = \frac{\epsilon(s/S) + (s/S)^2}{1 + \epsilon}, \tag{A2}$$

where $S \equiv s(X)$ and $\epsilon$ is a parameter. It is clear by inspection that $s = S$ when $x = X$. To make the derivatives also equal at $x = X$ equate $\mathrm{d}x/\mathrm{d}s$ at $x = X$ to $1/S'$, where $S' \equiv s'(X)$:

$$\left(\frac{\mathrm{d}x}{\mathrm{d}s}\right)_{x=X} = \frac{X}{S}\,\frac{(\epsilon + 2)}{(1 + \epsilon)} = \frac{1}{S'}, \tag{A3}$$

which gives

$$\epsilon = (2XS' - S)/(S - XS'), \tag{A4}$$

equivalent to Eq. (28). Changing the variable of integration in Eq. (A1) from $x$ to $s$ according to Eq. (A2) one arrives after some algebra at Eqs. (26) and (27).

Solving Eq. (A2) for the approximating function $s(x)$ we find

$$s(x) = \frac{S}{2}\left(-\epsilon + \sqrt{\epsilon^2 + 4(1 + \epsilon)(x/X)}\right). \tag{A5}$$

This may be compared with the postulated Ignatyuk entropy given by Eq. (20) in the text, to which Eq. (A5) is supposed to be an approximation. Figure 7 displays the difference between the corresponding level densities $\rho(x) \sim \exp[s(x)]$ for a nucleus with mass number $A = 250$ in the range of excitation energies between 0 and $X = 10$ MeV, and with four assumed shell corrections $D = 0, -3, -6$, and $-9$ MeV. It will be seen that, even for the largest shell correction, the inaccuracy of the polynomial approximation for $N_f$ or $\mathcal{N}_n$ would be only of the order of 1%. Similarly, Fig. 8 displays the difference between the postulated and approximating integrands in Eq. (24). In this case the inaccuracy in $N_n$ would be no more than about 2.5%. Figure 9 shows, as a function of the excitation energy $X$, the ratio of $N_n$ calculated with the polynomial approximation, to $N_n$ calculated by numerical integration. Figure 10 shows a similar comparison of $\Gamma_n/\Gamma_f$ calculated in two ways for parameters appropriate to the reaction $^{58}\mathrm{Fe} + {}^{208}\mathrm{Pb}$. As expected, the discrepancy tends to zero for large excitation energies, where the shell effect is attenuated. A more extensive study showed that, in a large range of nuclear masses, excitations, and shell effects of practical interest, the use of our approximation in calculating $\Gamma_n/\Gamma_f$ would only rarely exceed a couple of percent.

## APPENDIX B

Using the transition-state theory we shall derive the formula for the neutron decay width $\Gamma_n$ in the case when all the

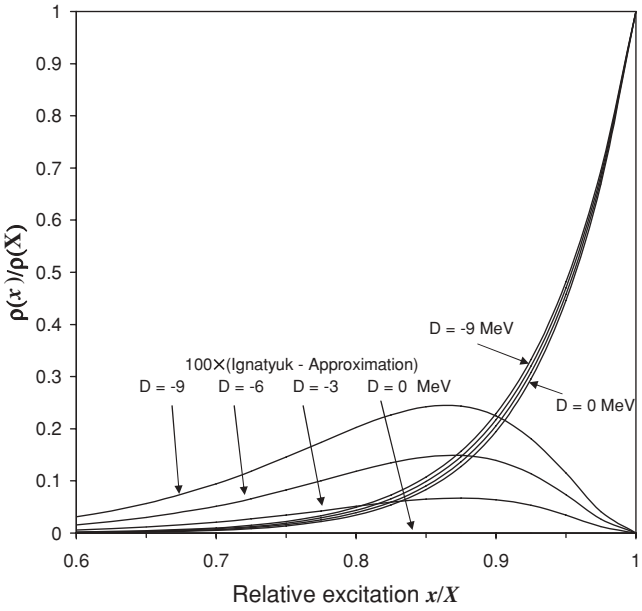


FIG. 7. The normalized level density $\rho(x)/\rho(X) = e^{s(x)}/e^{s(X)}$, where $s(x)$ is the Ignatyuk entropy, Eq. (20), plotted against the relative excitation energy $x/X$ for a nucleus with $A = 250$, an excitation energy $X = 10$ MeV, and shell corrections $D = 0, -3, -6$, and $-9$ MeV. This quantity is proportional to the integrand in the expression for the number of fission decay channels, Eq. (22), or to the number of neutron decay channels in the "symmetrical" treatment, Eq. (23). Also shown are the differences, multiplied by 100, between the postulated Ignatyuk relative level densities and the polynomial approximations. For the largest shell correction, $-9$ MeV, the error in the number of decay channels would be about 1%.

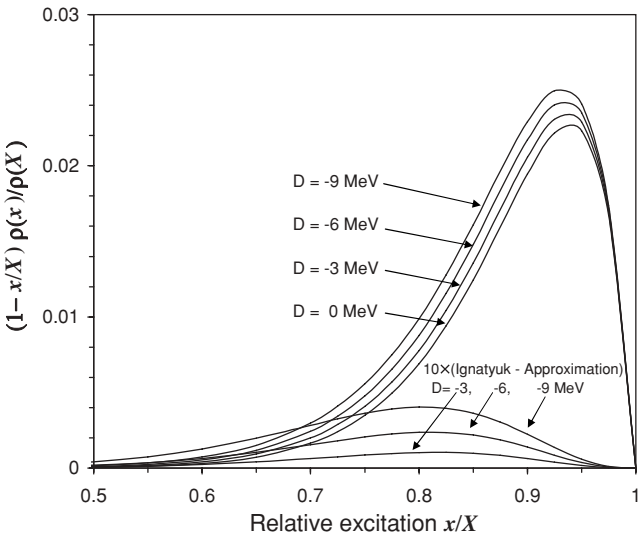


FIG. 8. Plot for the same hypothetical nucleus as in Fig. 7, but the quantity plotted is $(1 - x/X)\rho(x)/\rho(X)$, which is proportional to the integrand in the conventional expression for the number of neutron decay channels, Eq. (24). Also shown are the differences, multiplied by 10, between the Ignatyuk integrands and the polynomial approximations. For the largest shell correction the error in the number of decay channels calculated using that approximation would be about 2.5%.

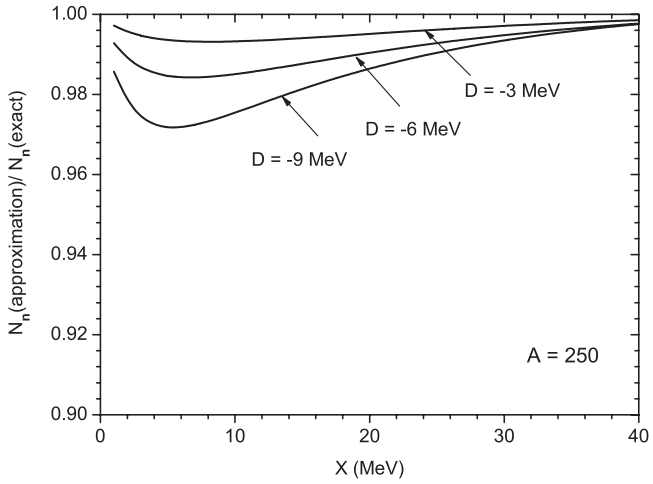


FIG. 9. Plot of the ratio of $N_n$ calculated using the polynomial approximation to $N_n$ calculated by numerical integration, as a function of excitation energy $X$, for a nucleus with $A = 250$ and Ignatyuk level densities with shell effects $D = -3, -6$, and $-9$ MeV.

three neutron degrees of freedom are singled out for special treatment. The result turns out to be identical with the formula due to Weisskopf [24] based on the principle of detailed balance between neutrons evaporated from and captured by a nucleus in equilibrium with its vapor.

Consider as in Sec. II a microcanonical ensemble of $\rho\Delta E$ systems at the neutron's "point of no return." Again consider the total number of degrees of freedom split into the neutron's disintegration degree of freedom $q_1$ normal to the surface of the emitting nucleus and the remainder, the activated complex. But now subdivide the activated complex into the neutron's remaining transverse degrees of freedom $q_2$ and $q_3$ orthogonal to $q_1$, to be accorded special treatment, and the rest, to be treated by a statistical formula. Let $p_1$, $p_2$, $p_3$ be the momenta

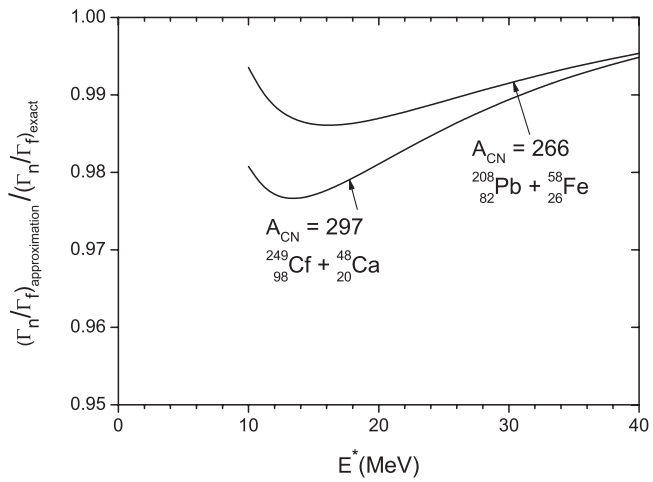


FIG. 10. The ratio of $\Gamma_n/\Gamma_f$ calculated using the polynomial approximation to $\Gamma_n/\Gamma_f$ calculated by numerical integration for the $^{266}$Hs and $^{297}$118 compound nuclei, as a function of excitation energy. The parameters used for this comparison are the same as for Figs. 3 and 4.

conjugate to $q_1, q_2, q_3$, so that

$$K = p^2/2m = \left(p_1^2 + p_2^2 + p_3^2\right)/2m \tag{B1}$$

is the neutron's kinetic energy, where $m$ is the neutron's mass. We shall denote by $x$ the left-over energy that serves as the excitation energy of the "statistical" subsystem with $n-3$ degrees of freedom:

$$x = E - V_n - p^2/2m = E - V_n - K. \tag{B2}$$

We shall write the level density of the statistical system at excitation $x$ as $\rho_n(x)$.

Consider now an infinitesimal element of the total phase space, defined by the product of a configuration space volume $4\pi R^2 \mathrm{d}q_1$ in the form of a thin spherical shell surrounding the emitting nucleus (assumed for simplicity to be spherical) at the "no return distance" $R$ times a momentum space volume specified by the intersection of a thin spherical shell between $p$ and $\mathrm{p} + \mathrm{d}p$ and a thin slab between $p_1$ and $p_1 + \mathrm{d}p_1$ in the momentum space $p_1, p_2, p_3$. Since the choice of $p$ specifies $x$ by Eq. (B2) the level density $\rho_n(x)$ is also fixed by this choice. The number of representative points in element of phase space thus specified and in the energy slot $\Delta E$ is then

$$\Delta E\,(4\pi R^2\,\mathrm{d}q_1)(2\pi p\,\mathrm{d}p\,\mathrm{d}p_1)(2/h^3)\rho_n(x). \tag{B3}$$

Here $2\pi p\,\mathrm{d}p\,\mathrm{d}p_1$ is readily verified to be the volume of the intersection of the thin spherical shell and the thin slab in momentum space, and $2/h^3$ is the density of doubly degenerate states in the neutron's phase space. As in Sec. II focus attention on the two-dimensional phase space depicted in Fig. 1 (but with $p, q$ replaced by $p_1, q_1$ and $P$ replaced by $p$), and obtain the number of disintegrations in time $\Delta t$ by an integration of Eq. (B3) first over $q_1$ from $-v\Delta t$ to 0, followed by an integration over $p_1$ from 0 to $p$, followed by an integration over $p$ from 0 to its maximum value $P$ given by $P^2/2m = E - V_n$:

$$\begin{aligned}
&\int_0^P \mathrm{d}p \int_0^p \mathrm{d}p_1 \int_{-v\Delta t}^0 \mathrm{d}q_1\, \Delta E(4\pi R^2)(2\pi p)(2/h^3)\rho_n(x) \\
&= \int_0^P \mathrm{d}p \int_0^p \mathrm{d}p_1\, v_1 \Delta t\, \Delta E(4\pi R^2)(2\pi p)(2/h^3)\rho_n(x) \\
&= \Delta E\,\Delta t(4\pi R^2)(2\pi)(2/h^3)m^{-1} \int_0^P \mathrm{d}p\; p \int_0^p \mathrm{d}p_1\; p_1 \rho_n(x) \\
&= \Delta E\,\Delta t(4\pi R^2)\pi(2/h^3)m^{-1} \int_0^P \mathrm{d}p\; p \cdot p^2 \rho_n(x).
\end{aligned} \tag{B4}$$

Now change variable from $p$ to $x$ using $p^2 = 2m(E - V_n - x)$ and $\mathrm{d}pp = -m\mathrm{d}x$. Changing the limits of integration appropriately we obtain

$$\Delta E\,\Delta t(4\pi R^2)2\pi(2/h^3)m \int_0^{X_n} \mathrm{d}x(X_n - x)\rho_n(x), \tag{B5}$$

where the maximum excitation energy is denoted by $X_n = E - V_n$.

The neutron disintegration width is obtained by division by $\rho\Delta t\,\Delta E$ and multiplication by $\hbar$, with the final result

$$\Gamma_n = \frac{2m\sigma}{\pi^2\hbar^2\rho} \int_0^{X_n} \mathrm{d}x(X_n - x)\,\rho_n(x), \tag{B6}$$

where $\sigma$ stands for $\pi R^2$, the cross sectional area of the "sphere of no return." Equation (B6) becomes identical with Weisskopf's neutron evaporation formula if this cross section is identified with the cross section for neutron capture, the reaction inverse to neutron evaporation. Multiplication of Eq. (B6) by $2\pi\rho$ gives the corresponding expression for the number of neutron decay channels:

$$N_n = \frac{4m\sigma}{\pi\hbar^2} \int_0^{X_n} \mathrm{d}x(X_n - x)\,\rho_n(x). \tag{B7}$$


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