### Effect of Competition between Gamma-Ray and Particle Emission on Excitation Functions\*

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A procedure has been devised for calculating cross sections for nuclear reactions within about 2 Mev of threshold, where the effect of competition between gamma-ray and particle emission is often important. The requisite formulas depend upon assumptions embodied in the spin-dependent statistical theory of nuclear reactions, so the treatment is most valid for medium to heavy nuclei at moderate bombarding energies. Input data required by the formulas are (1) the level density parameter a, (2) an effective nuclear moment of inertia  $\mathfrak{s}$ , (3) the ratio of radiation width to level spacing  $(\Gamma_{\gamma}/D)$  evaluated at some convenient energy, spin, and parity in the excited nucleus immediately preceding the product, (4) transmission coefficients  $T_{l}(\epsilon)$  for the range of ener-

#### INTRODUCTION

NE of the approximations most often used in the analyses of excitation functions is that de-excitation of an excited nucleus by gamma-ray emission may be neglected if the same excited nucleus can also decay by particle emission. This approximation is often used when it is not justified. The results of the analyses of two sets of experimental data are used to demonstrate that the correction for competitive gamma-ray emission can be very important within about 2 Mev of reaction thresholds. An argument is also advanced to show that in certain (not unusual) cases, the correction is still important even many Mev above the threshold.

Important competition between gamma-ray emission and particle emission arises because the excited nucleus immediately preceding the product nucleus is, in general, characterized not only by a distribution in excitation energies, but also by a distribution of spins and parities. In that zone of excitation energies where energetic restrictions are such that only a few states of the product nucleus can be populated by particle emission, it often happens that the emission of any other type of particle is similarly restricted or energetically forbidden. Then, for those particle-unstable states characterized by spins greatly different from those of any available state in the product(s), the rate of particle emission is strongly diminished by the "centrifugal barrier" because the particles must be emitted with large values of the orbital angular-momentum quantum number l. The gamma-ray de-excitation rate is under no such strong restriction, however, and may then compete very favorably with particle emission.

Considering the ranges of bombarding energies (10–50 Mev) and projectiles  $(1 \leq \text{projectile mass number} \leq 4)$ within which most such excitation functions are measured, the range of spins significantly populated in typical compound nuclei (i.e.,  $J \sim$  from 0 up to 5–20) is large

gies and type of particle in the final evaporation step, and (5) the energies, spins, and parities of the first few excited states in the product nucleus. Using reasonable estimates for  $\mathfrak{I}$ ,  $\Gamma_{\gamma}/D$ , and  $T_l(\epsilon)$ , experimental excitation functions near threshold for the reactions  $\operatorname{Bi}^{209}(p,2n)\operatorname{Po}^{208}$  and  $\operatorname{Sm}^{144}(\alpha,3n)\operatorname{Gd}^{145}$  were analyzed to find the corresponding values of a. The results are consistent with  $a \sim 0.1 A$  Mev<sup>-1</sup> (A is the mass number), but inconsistent with  $a \sim 2$  to 3 Mev<sup>-1</sup> (independent of A), in contrast to the result often obtained when competition from gamma-ray emission is ignored. Also, a semiqualitative argument is given to suggest that competitive gamma-ray emission often seriously influences excitation functions even several Mev above threshold.

enough that the above argument should be taken seriously.

Considerations similar to those above have been advanced qualitatively by Flerov,<sup>1</sup> Thomas,<sup>2</sup> Kammuri,<sup>3</sup> and Mollenauer,<sup>4</sup> and more quantitatively by Pik-Pichak.<sup>5</sup> These workers were concerned with the dissipation of energy from systems arising from the interactions between complex nuclei at energies up to  $\sim 200$ Mev, where very large values of nuclear spin (i.e., as high as 50–100) are attained with high probability.

The apparent anomaly in measurements of the energy dependence of nuclear-state density pointed out by Igo and Wegner<sup>6</sup> is probably at least partly a consequence of the neglect of gamma-ray emission. In particular, values of the parameter a in Weisskopf's Fermi gasstate density formula<sup>7</sup>

# $\rho(E) \approx (\text{const}) \exp[2(aE)^{\frac{1}{2}}]$

(most workers neglect the energy dependence of the preexponential factor), which are derived from excitationfunction measurements are usually anomalously small, of the order of  $a \sim 2$  to 3 Mev<sup>-1</sup> (independent of mass number A), while values of a extracted from measurements on the energy spectra of emitted particles<sup>8</sup> usually (but not always) roughly obey the expected approxi-

J. F. Mollenauer (private communication).

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<sup>&</sup>lt;sup>1</sup>G. N. Flerov, Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, (United Nations, Geneva, 1958), Vol. 14, p. 151, Paper No. P/2299. <sup>2</sup> T. D. Thomas, in *Proceedings of the Second Conference on Re-*

actions Between Complex Nuclei, Gatlinburg, Tennessee, edited by A. Zucker, F. T. Howard, and E. C. Halbert (John Wiley & Sons, Inc., New York, 1960), p. 223. <sup>3</sup> T. Kammuri, Physics Department, Osaka University, pri-

vately circulated report.

<sup>&</sup>lt;sup>4</sup> J. F. Mollenauer (private communication).
<sup>5</sup> G. A. Pik-Pichak, Soviet Phys.—JETP 11, 557 (1960).
<sup>6</sup> G. Igo and H. Wegner, Phys. Rev. 102, 1364 (1956).
<sup>7</sup> J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley & Sons, Inc., New York, 1952), Chap. VIII.
<sup>8</sup> As recent examples, see, R. L. Bramblett and T. W. Bonner, Nuclear Phys. 20, 395 (1960); R. D. Albert, J. D. Anderson, and C. Wieng Phys. Phys. 12140 (1960). C. Wong, Phys. Rev. 120, 2149 (1960).

mate law<sup>9</sup>  $a \sim 0.1 A$  Mev<sup>-1</sup>. The introduction of competitive gamma-ray emission into the analyses helps to remove the disagreement, because it increases the resulting values of *a* obtained from excitation functions. Two experimental excitation functions (near threshold) were so analyzed, by a method described in this paper, and values of a were derived which are inconsistent with  $a \sim 3$  Mev<sup>-1</sup> but consistent with  $a \sim 0.1 A$  Mev<sup>-1</sup>.

#### CALCULATIONAL PROCEDURE

### Definition of Terms and Outline of Problem

The calculation is done within the framework of the spin-dependent statistical theory of nuclear reactions, so its validity is restricted to medium to heavy nuclei at moderate bombarding energies. The calculation is conveniently separated into two parts which will be discussed separately in Parts I and II.

In Part I, a procedure is outlined for estimating the distribution of spins, parities, and excitation energies in the excited nucleus immediately preceding the product. For calculational brevity, only neutron emission is specifically considered here, although the formulas are also appropriate for the emission of charged particles. The procedure used is that of Thomas,<sup>10</sup> suitably adapted for the present purpose; however, see also reference 7 and the papers by Ericson,<sup>11</sup> and Vandenbosch and Huizenga.12

In Part II is considered the role of the competition between gamma-ray and particle emission in determining the proportion of the preceding excited nucleus which will form the product of interest. This part of the calculation is very similar to the theory and procedures developed by Margolis,<sup>13</sup> by Lane and Lynn,<sup>14</sup> and by Rae, Margolis, and Troubetzkoy<sup>15</sup> for calculating  $(n, \gamma)$ cross sections, and by Axel and Fox<sup>16</sup> for analyzing photonuclear activation functions near threshold.

Diagram (1) below describes the system of nuclear reactions to which the following formulas refer, and introduces symbols that are used throughout.

$$T \xrightarrow{t} C \xrightarrow{n} U_1 \xrightarrow{n} U_2 \xrightarrow{n} \cdots \xrightarrow{n} F \to P \quad \text{or} \quad F \to N.$$
 (1)

The target nucleus T amalgamates with the incident particle t to form the compound nucleus C. C emits a neutron to form nucleus  $U_1$ .  $U_1$ , in turn, emits a neutron to form  $U_2$ , and so on. F is the last nucleus in the chain which can possess enough excitation energy to emit a particle. P is the specific product nucleus whose excitation function is being discussed. N represents all products (including P) which can be formed by particle emission from F. The competition between particle emission and gamma-ray de-excitation is assumed to be important only in the transition from F to P (or N).

It is convenient to express the desired excitation function  $H(\mathcal{E})$  as the product of an excitation function calculated by conventional methods,  $G(\mathcal{E})$ , and a correction factor  $K(\mathcal{E})$ .

$$H(\mathcal{E}) = K(\mathcal{E})G(\mathcal{E}). \tag{2}$$

Here,  $\mathcal{E}$  is that amount of energy by which the center-ofmass bombarding energy exceeds the reaction threshold. The excitation function  $G(\mathcal{E})$  is calculated as if P were the only possible product; the function  $K(\mathcal{E})$  must then account not only for competitive gamma-ray emission, but also for the competitive formation of any other particle-emission products. The value of  $K(\mathcal{E})$  depends upon the relative distribution of population in spin parity  $J_F^i$  and excitation energy  $E_F$  in nucleus F [superscripts on the J's indicate parity, and subscripts specify the nucleus according to diagram (1)]. It is, therefore, convenient to define an average channel fraction  $k(J_F^i, \epsilon_x, J_{Px}^{jx})$ , which gives the fraction of F, with spin parity  $J_F^i$ , that de-excites by the emission of the appropriate particle to populate the *x*th excited state of P, with spin parity  $J_{Px}^{jx}$  and excitation energy  $E_{Px}$ . Here,  $\epsilon_x = E_F - B_{FP} - E_{Px}$ , where  $B_{FP}$  is the binding energy of the particle in nucleus F. Also, let  $S(J_F^i, E_F)$ represent the average relative distribution of populations of  $J_F^i$  and  $E_F$  in F, being normalized so that

$$\int_{B_{FP}}^{B_{FP}+\epsilon} \sum_{i=+}^{-} \sum_{J_{F}=0}^{\infty} S(J_{F}^{i}, E_{F}) dE_{F} = 1.$$
(3)

The functions  $k(J_F^i, \epsilon_x, J_{Px}^{jx})$  and  $S(J_F^i, E_F)$  are averages over many individual levels in the excited nucleus F in a limited region of excitation energy around  $E_F$ . Then.

$$K(\mathcal{E}) \approx \int_{B_{FP}}^{B_{FP}+\varepsilon} \sum_{i=+}^{-} \sum_{J_F=0}^{\infty} \sum_{x} S(J_F^i, E_F) \times k(J_F^i, \epsilon_x, J_{P_x}^{jx}) dE_F, \quad (4)$$

where the summation over x includes all states in P for which  $\epsilon_x > 0$ . This equation neglects the small correction which arises from those cases in which gamma-ray deexcitation takes place to levels with energies  $E_F'$  in F for which  $E_F' > B_{FP}$ , allowing yet another opportunity for particle emission to form the product P.

The summand-integrand in Eq. (4) is written as a product of averages when it should be the average product. The error introduced by this approximation is discussed briefly in a following section (Part II). Ignoring the error, the calculations of  $S(J_F^i, E_F)$  and

<sup>&</sup>lt;sup>9</sup> J. M. B. Lang and K. J. LeCouteur, Proc. Phys. Soc. (London) A67, 586 (1954), and many others (see references 8, 19).

<sup>&</sup>lt;sup>10</sup> T. D. Thomas (unpublished work). <sup>11</sup> T. Ericson, Nuclear Phys. **17**, 250 (1960)

<sup>&</sup>lt;sup>12</sup> R. Vandenbosch and J. R. Huizenga, Phys. Rev. 120, 1313 (1960)<sup>13</sup> B. Margolis, Phys. Rev. 88, 327 (1952).

<sup>&</sup>lt;sup>14</sup> A. M. Lane and J. E. Lynn, Proc. Phys. Soc. (London) A70, 557 (1957).

<sup>&</sup>lt;sup>15</sup> E. R. Rae, B. Margolis, and E. S. Troubetzkoy, Phys. Rev. 112, 492 (1958).

<sup>&</sup>lt;sup>16</sup> P. Axel and J. D. Fox, Phys. Rev. 102, 400 (1956).

 $k(J_F^i, \epsilon_x, J_{Px}^{jx})$  are done separately, as described in 1 Parts I and II.

#### Part I. Calculation of $S(J_F^i, E_F)$

The function  $S(J_F^i, E_F)$  can be numerically calculated in a step-by-step procedure. The first step is to calculate the relative population of spin parity  $J_e^i$  in C. The cross section for forming the compound nucleus with a given spin parity  $J_e^i$  is given by

$$\sigma_c(J_T^{j},\epsilon,J_c^{i}) = \pi \lambda^2 \frac{2J_c+1}{(2s+1)(2J_T+1)} T(J_T^{j},\epsilon,J_c^{i}), \quad (5)$$

where the spin parity of the target nucleus is  $J_T{}^j$  and the spin of the incident particle is *s* (positive parity is assumed). The function  $T(J_T{}^j,\epsilon,J_e{}^i)$  is defined, in general terms, by

$$T(J_1{}^j,\epsilon,J_2{}^i) = \sum_{S=|J_1-s|}^{J_1+s} \sum_{l=|J_2-S|}^{J_2+s} T_l(\epsilon),$$
(6)

where the  $T_l(\epsilon)$  are the appropriate particle-transmission coefficients defined in reference 7. The dependence of the  $T_l(\epsilon)$  upon  $J_2$  and S is not expected to be important,<sup>17</sup> and is therefore neglected. In computing the  $T(J_1{}^{i}, \epsilon, J_2{}^{i})$ , only even values of l are included if i = j, and only odd values if  $i \neq j$ . Whenever an argument contains  $\epsilon$  between two subscripted symbols, as above, it refers to the energy of that species of particle appropriate to the transition between the specific nuclei designated by the subscripts. Equation (5) is essentially just Eq. (10.23) in Chap. VIII of reference 7.

The second step is to calculate the relative population of spin parity and energy in  $U_1$  resulting from the neutron evaporation from C. This may be done using the following formula,<sup>10-12</sup> which expresses the emission rate of particles with respect to both initial and final values of spin parity and excitation energy.

$$P(J_{c}^{i}, E_{c}, \epsilon, J_{U}^{j}, E_{U})d\epsilon = \frac{1}{h} \frac{\omega(J_{U}^{j}, E_{U})}{\omega(J_{c}^{i}, E_{c})} T(J_{U}^{j}, \epsilon, J_{c}^{i})d\epsilon.$$
(7)

The level density as a function of spin parity and excitation energy is  $\omega(J^i, E)$ , and the  $T_l(\epsilon)$  appropriate for neutron emission are used. The state density is related to the level density by

$$\rho(E) = \sum_{J=0}^{\infty} (2J+1)\omega(J,E),$$
 (8)

where it is assumed<sup>18</sup> that  $\omega(J^+,E) = \omega(J^-,E) = \frac{1}{2}\omega(J,E)$ . The dependence of level density on spin is taken to be<sup>9,19</sup>

$$\omega(J,E) \approx \omega(0,E)(2J+1) \exp[-J(J+1)/2\sigma^2], \quad (9)$$

where the parameter  $\sigma^2$  is

$$\sigma^2 = (\mathfrak{G}/\hbar^2) (E/a)^{\frac{1}{2}},\tag{10}$$

 $\mathcal{G}$  being an effective nuclear moment of inertia. Equation (7) is obtainable from a "detailed balance" calculation, employing Eq. (5) for the inverse cross section.

Equation (7) may be repeatedly applied to follow the neutron evaporation from  $U_1$  to  $U_2$ , from  $U_2$  to  $U_3$ , etc., until nucleus F is reached. The last step provides the requisite knowledge of the relative distribution of population in spin parity and excitation energy in F, for the determination of  $S(J_F^i, E_F)$ .

After making certain approximations, the tedious procedure which has just been described may be sufficiently simplified that it is practical to perform the computations with a desk calculator, if only neutron emission is permitted. This simplification is described in the Appendix.

### Part II. Calculation of $k(J_F^i, \varepsilon_x, J_{Px}^{jx})$

The channel fraction  $k(J_F^i, \epsilon_x, J_{Px}^{jx})$  may be expressed in terms of emission probabilities as follows:

$$\approx \frac{P(J_F^i, \epsilon_x, J_{P_x}^{jx})}{P_{\gamma}(J_F^i, E_F) + \sum_{N,y} P(J_F^i, \epsilon_{N,y}, J_{N,y}^{jN,y})}.$$
 (11)

The symbol  $P(J_F^i, \epsilon_x, J_{Px}^{jx})$  represents the average particle-emission rate from F to the xth excited level in P, and  $P_{\gamma}(J_F^i, E_F)$  expresses the average rate of deexcitation of F by gamma-ray emission. The summation over N, y includes all particle-emitting channels open from excited nucleus F to all the possible product nuclei N, including P. Equation (11) is only an approximation because the right side is written as a ratio of averages when it should be an average ratio. The error involved in Eq. (11) should be combined with the similar error (already mentioned) in Eq. (4). Lane and Lynn<sup>14</sup> and Dresner<sup>20</sup> have estimated the magnitude of the error introduced by such approximations, in situations rather similar to this one; guided by their results, one sees that the multiplicative correction function that one might include in the integrand-summand of Eq. (4) can take values as far from unity as 0.65 for certain combinations of values of the various arguments. However, because of the averaging effect of the integrations and summations in Eq. (4), the resulting error in the calculated value of

<sup>&</sup>lt;sup>17</sup> A. M. Lane and R. G. Thomas, Revs. Modern Phys. **30**, 257, 311 (1958). However, see also, K. K. Seth, Nuclear Phys. **24**, 169 (1961).

<sup>&</sup>lt;sup>18</sup> T. Ericson, Advances in Physics, edited by N. F. Mott (Taylor and Francis, Ltd., London, 1960), Vol. 9, p. 425.

 <sup>&</sup>lt;sup>19</sup> H. A. Bethe, Revs. Modern Phys. 9, 84 (1937); C. Bloch, Phys. Rev. 93, 1094 (1954); T. Ericson and V. Strutinski, Nuclear Phys. 8, 284 (1958).
 <sup>20</sup> L. Dresner, Proceedings of the International Conference on Co

<sup>&</sup>lt;sup>20</sup> L. Dresner, Proceedings of the International Conference on the Neutron Interactions with the Nucleus, September 9–13, 1957 [Atomic Energy Commission Report TID-7547 (unpublished)], p. 71.

 $K(\mathcal{E})$  should be somewhat less serious, and, being difficult to estimate, has been ignored.

From Eq. (7)

$$P(J_F^i, \epsilon_x, J_{Px}^{jx}) \approx h^{-1} D(J_F^i, E_F) T(J_{Px}^{jx}, \epsilon_x, J_F^i), \quad (12)$$

where  $D(J_F^i, E_F) = \omega^{-1}(J_F^i, E_F)$  is the level spacing, and where  $P(J_F^i, \epsilon_x, J_{Px}^{jx})$  is expressed in units of sec<sup>-1</sup>.

In order to introduce the gamma-ray emission rate into Eq. (11), it is convenient to make the following definitions13:

$$b(J_{F}^{i}) = \frac{\Gamma_{\gamma}(J_{F}^{i}, B_{FP})}{D(J_{F}^{i}, B_{FP})},$$
(13)

where  $\Gamma_{\gamma}(J^{i}, E)$  is the average radiation width, and

$$f(E_F) = \frac{D(J_F^i, B_{FP})}{D(J_F^i, E_F)} \frac{\Gamma_{\gamma}(J_F^i, E_F)}{\Gamma_{\gamma}(J_F^i, B_{FP})}.$$
 (14)

For  $E_F - B_{FP}$  not too large, the evaluation of  $f(E_F)$  can be performed using the following rough formula,<sup>21</sup> where it is assumed that the first step in the gamma-ray deexcitation cascade is dipole radiation.

$$\Gamma_{\gamma}(J,E) \propto D(J,E) \int_{0}^{E} \epsilon^{3} \omega(J, E-\epsilon) d\epsilon.$$
 (15)

In this narrow region of  $E_F$ ,  $f(E_F)$  is nearly independent of spin, as may be seen from the following easily derived approximation:

$$f(E_F) \approx e^{(E_F - B_F P)/\theta},\tag{16}$$

which is valid for  $B_{FP} \gg \theta$ , for  $(E_F - B_{FP})/\theta < 3$ , and where  $\theta \equiv (E_F/a)^{\frac{1}{2}} - \frac{3}{2}(1/a)$ .

Employing the above definitions, the gamma-ray emission rate is introduced into Eq. (12) through

$$P_{\gamma}(J_F^{i}, E_F) = \hbar^{-1} \Gamma_{\gamma}(J_F^{i}, E_F)$$
  

$$\approx \hbar^{-1} D(J_F^{i}, E_F) 2\pi b(J_F^{i}) f(E_F). \quad (17)$$

Combining Eqs. (11), (12), and (17) gives, finally,

$$k(J_F^{i},\epsilon_x,J_{P_x}^{jx}) = \frac{T(J_{P_x}^{jx},\epsilon_x,J_F^{i})}{2\pi b(J_F^{i})f(E_F) + \sum_{N,y}T(J_{N,y}^{jN,y},\epsilon_{N,y},J_F^{i})}.$$
 (18)

The values of the  $b(J_F^i)$  remain as parameters which must either be estimated somehow, or else be determined in a separate experiment. As will be seen in the sample calculations in the next section, an excitation function seems not to provide data of a kind from which both  $b(J_F^i)$  and a can be determined simultaneously. Current workers regard the value of  $\Gamma_{\gamma}(J^{i},E)$  to be roughly independent of spin.22 The spin dependence of  $b(J_F^i)$  must then be principally through the level spacing, and one therefore needs a knowledge of  $b(J_F^i)$ at only one spin. Measurements or estimates of  $b(J_F^i)$ can be obtained from several sources, such as from compilations of slow-neutron resonances and radiation widths,<sup>23</sup> from analyses of  $(n,\gamma)$  excitation functions,<sup>24</sup> or from the various published empirical and semiempirical procedures for estimating level spacings<sup>25</sup> and radiation widths.<sup>26</sup>

### COMPARISON WITH EXPERIMENT

The calculation of an excitation function by the above-described procedure requires that the spins and parities of all the energetically permissible productstates N, y be known, in order to calculate all of the necessary  $k(J_F^i, \epsilon_{N,y}, J_{N,y}^{iN,y})$ . The limited knowledge of the energy states in most medium to heavy nuclei therefore restricts such calculated excitation functions to within about 1 or 2 Mev of the reaction threshold. The two sample calculations were accordingly performed for comparison with some experimental excitation functions measured in the threshold region.

The most serious uncertainty in the analyses of the experimental data is a lack of accurate knowledge of the important input functions  $b(J_F^i)$  and  $T_l(\epsilon)$ . The  $b(J_F^i)$ are usually not known from experiment, and at this time can only be estimated to within about a factor three. The degree of uncertainty in the knowledge of the transmission coefficients may be roughly appraised (for neutrons) by an intercomparison of the strong-interaction values of Feld et al.,27 the values of Beyster et al.,28 and the values of Campbell et al.29 The two last-named tables are for a diffuse-surface, complex potential well. Corresponding values very often disagree by a factor of two to three or even more, in the energy region of most interest. The magnitudes of the effects of such uncertainties may be surmised from Eq. (18).

Although accurate values of a cannot presently be determined from the data, for the reasons mentioned above, the results of the calculations can be used to demonstrate the importance of the effect of competitive gamma-ray de-excitation on excitation functions.

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<sup>&</sup>lt;sup>21</sup> Reference 7, Chap. XII. <sup>22</sup> D. J. Hughes and R. L. Zimmerman, *Nuclear Reactions*, edited by P. M. Endt and M. Demeur (Interscience Publishers, Inc., New York, 1959), Vol. I, Chap. VIII, p. 369.

<sup>23</sup> Neutron Cross Sections, compiled by D. J. Hughes and R. B. Schwartz, Brookhaven National Laboratory Report, BNL-325 (U. S. Government Printing Office, Washington, D. C., 1958), 2nd ed.

<sup>&</sup>lt;sup>24</sup> B. Margolis, Phys. Rev. 88, 327 (1952); C. Mossin-Kotin, B. Margolis, and E. S. Troubetzkoy, *ibid.* 116, 937 (1959).
<sup>25</sup> A. G. W. Cameron, Can. J. Phys. 36, 1040 (1958).
<sup>26</sup> A. Stolovy and J. A. Harvey, Phys. Rev. 108, 353 (1957).
<sup>27</sup> B. T. Feld, H. Feshbach, M. L. Goldberger, H. Goldstein, and V. F. Weisskopf, Atomic Energy Commission Report NYO-636, January 31, 1951 (unpublished).
<sup>28</sup> J. R. Beyster, R. G. Schrandt, M. Walt, and E. W. Salmi, Los Alamos Scientific Laboratory Report LA-2099, April 29, 1957 (unpublished).

<sup>(</sup>unpublished).

<sup>&</sup>lt;sup>29</sup> E. J. Campbell, H. Feshbach, C. E. Porter, and V. F. Weisskopf, Massachusetts Institute of Technology Laboratory for Nuclear Science Technical Report No. 73, February 8, 1960 (unpublished).

## $Bi^{209}(p,2n)Po^{208}$

Andre et al.30 measured excitation functions for the reactions  $Bi^{209}(p,\gamma)Po^{210}$ ,  $Bi^{209}(p,n)Po^{209}$ , and  $Bi^{209}$ -(p,2n)Po<sup>208</sup>, for bombarding energies up to about 1 Mev above the (p,2n) threshold. An estimate of a from these data was made the object of the first sample calculation.

It was assumed that only the ground state  $(J_P^i=0+)$ of Po<sup>208</sup> is populated. The first, second, and third excited states are thought to be at 0.66, 0.82, and 0.91 (or 1.1) Mev, respectively,<sup>31</sup> so the excitation function was calculated only up to  $\sim 1$  Mev above threshold. Both proton and alpha-particle emission are energetically permitted, but the magnitudes of the relevant transmission coefficients<sup>32</sup> are so small that this competition was neglected. Since  $b(J_F^i)$  has not been measured for Po<sup>209</sup>, it was estimated<sup>25,26</sup> to be roughly  $b(\frac{1}{2}) \sim 1.5 \times 10^{-4}$ , where  $B_{FP} = 6.85 \text{ Mev}^{33} \left[ \text{assuming } b(\frac{1}{2}) = b(\frac{1}{2}+) = b(\frac{1}{2}-) \right].$  In performing the calculations, it was found that the results are sensitive to the exact value of  $b(J_F)$  only in the immediate neighborhood of  $J_F = \frac{3}{2}, \frac{5}{2}$ . The simplifying approximation was made that  $b(J_F) \approx b(2) \approx 4 \times 10^{-3}$ , independent of  $J_F$ . The neutron transmission coeffi-



FIG. 1. Comparison of some calculated excitation functions with the experimental data of Andre  $et \ al.^{30}$  (open circles) for the reaction  $Bi^{209}(p,2n)Po^{208}$ .

<sup>30</sup> C. G. Andre, J. R. Huizenga, J. F. Mech, W. J. Ramler, E. G. Rauh, and S. R. Rocklin, Phys. Rev. **101**, 645 (1956).

 <sup>31</sup> K. Way, Nuclear Data Sheets, National Academy of Sciences (National Research Council, Washington, D. C., 1960).
 <sup>32</sup> H. Feshbach, M. M. Shapiro, and V. F. Weisskopf, Atomic Energy Commission Report NYO-3077, June 15, 1953 (unpublication) lished).

<sup>33</sup> B. M. Foreman, Jr., and G. T. Seaborg, J. Inorg. & Nuclear Chem. 7, 305 (1958).



FIG. 2. Locus of pairs of values of  $b(\frac{1}{2})$  and a corresponding to calculated excitation functions which fit the experimental data for the reaction  $\operatorname{Bi}^{209}(p,2n)\operatorname{Po}^{208}$ .

cients used in Part II of the calculation were those given by Beyster et al.28 for Bi. Those used in Part I were taken from Feshbach et al.<sup>32</sup> for charged particles, and from Feld et al.27 for neutrons because it was felt that the "strong-interaction" coefficients might be more appropriate in the intermediate evaporation steps. The relationship between the spin of the product state in  $Po^{208}$  (J=0) and the distribution of spins in excited  $Po^{209}$  (most of the population is within  $\pm 2$  units of  $J_F = 9/2$ ) is such that it was suspected that the calculated excitation function might be rather sensitive to the value of the effective nuclear moment of inertia in this case. Accordingly, the calculation was performed for  $\mathcal{I} = 0.1 \mathcal{I}_{rigid}^{34}$  and for  $\mathcal{I} = \infty$  for which  $\omega(J, E)$  $\approx (2J+1)\omega(0,E)$  (the latter convention gives a result nearly the same as for  $\mathcal{I} = \mathcal{I}_{rigid}$ , see the Appendix).

Figure 1 shows a comparison with experiment of some typical calculated excitation functions. It was found that no unique combination of a and  $b(\frac{1}{2})$  could be found which would give a significantly better fit to the data than any other appropriate combination, over wide ranges of their respective values. This means that it is not possible to determine both  $b(\frac{1}{2})$  and a from these data.

Figure 2 shows the locus of values of a and  $b(\frac{1}{2})$  which will give calculated excitation functions that "fit" the experimental data at a point 0.75 Mev above the (p,2n)threshold. For reasonable estimates of  $b(\frac{1}{2})$ , represented by the shaded zone, the corresponding range in values

<sup>&</sup>lt;sup>34</sup> Various workers report values of  $\mathcal{I}$  between  $\sim 0.1 \mathcal{I}_{rigid}$  and ~1.6 $g_{rigid}$ ; a review and discussion of this problem is found in A. C. Douglas and N. MacDonald, Nuclear Phys. 13, 382 (1959). In addition, see also, reference 12 and J. R. Huizenga and R. Vandenbosch, Phys. Rev. 120, 1305 (1960); J. H. Carver and G. A. Vandenbosch, Phys. Rev. 104 (1960) Jones, Nuclear Phys. 19, 184 (1960).

of a is inconsistent with  $a \sim 3$  Mev<sup>-1</sup>, but is clearly consistent with  $a \sim 20$  Mev<sup>-1</sup>, i.e. with  $a \sim 0.1 A$  Mev<sup>-1</sup>.

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#### $Sm^{144}(\alpha, 3n)Gd^{145}$

At Brookhaven, the author is measuring excitation functions for the reactions of 15- to 40-Mev helium ions with Sm<sup>144</sup>, with special attention to the Sm<sup>144</sup>( $\alpha$ , 3n)Gd<sup>145</sup> reaction within 2 Mev of threshold. Early results of the latter experiment have been used for another sample calculation.

Unfortunately, nothing is directly known about the first few excited states of the product Gd<sup>145</sup>. However, the first three levels in  $Gd^{145}$  are expected to have spin parities of  $\frac{3}{2}$ +,  $\frac{1}{2}$ +, and 11/2- because corresponding levels are systematically found in the other known even-odd 81-neutron isotones.<sup>35</sup> For the same reason, it is assumed that there are no other levels lying below  $\sim 1$ Mev; this very convenient circumstance is plausible because of the close proximity of the closed 82-neutron shell. It is apparent from the large positron-decay energy of  $Gd^{145}$  (~5.3 Mev)<sup>36</sup> that proton emission from excited Gd<sup>146</sup> must also be considered in the calculation. The lowest states in the proton emission product Eu<sup>145</sup> were assigned with the help of the "systematics" of lowlying levels in the other known odd-even 82-neutron isotones, and made to agree with what little is known<sup>36</sup> of the decay characteristics of Gd<sup>145</sup>. Figure 3 gives the assumptions concerning the relevant nuclides and their



FIG. 3. Energies, spins, and parities of the first few excited states of Gd<sup>145</sup> and Eu<sup>145</sup> which were assumed for the calculation of excitation functions near threshold for the reaction  $Sm^{144}(\alpha, 3n)Gd^{145}$ .

excited states, which were adopted for the sake of the calculation.

It is not known whether the 24-min activity observed for Gd<sup>145</sup> is due to the expected isomeric state or to the ground state. The calculation was therefore performed for the three extreme possibilities, i.e., that the observed activity represents (i) ground state only, (ii) upper state only, or (iii) both states together.

The neutron transmission coefficients of Beyster et al.28 for Sm, and the charged-particle transmission coefficients calculated by Feshbach et al.32 were used in Part II of the calculation. The estimate<sup>25,26</sup>  $b(0) \sim 6$ 



FIG. 4. Comparison of some calculated excitation functions with experimental data (open circles) for the reaction  $Sm^{144}(\alpha,3n)Gd^{145}$  near threshold. The dashed and dotted curves to either side of the unbroken curves represent the uncertainty in the determination of the reaction threshold in this experiment.

 $\times 10^{-3}$  was obtained using Cameron's calculated binding energy<sup>37</sup> of the last neutron in Gd<sup>146</sup>,  $B_{FP} = 10.88$  Mev. As in the previous example, the simplifying approximation  $b(J_F) \approx b(2) \approx 3 \times 10^{-2}$  (independent of  $J_F$ ) was employed in the calculations.

Part I of the calculation was performed taking full advantage of the simplifying assumptions described in the Appendix. The spin-dependent part of  $S(J_F^i, E_F)$ was calculated assuming only s-wave neutron emission, and with  $\mathfrak{I} = \infty$ . The function  $G(\mathfrak{E})$  and the energydependent part of  $S(J_F^i, E_F)$  were calculated using Jackson's formula,<sup>38</sup> since the effect of proton emission

<sup>&</sup>lt;sup>35</sup> R. A. James and C. D. Bingham, Phys. Rev. 117, 810 (1960); K. Kotajima and H. Morinaga, Nuclear Phys. 16, 231 (1960). <sup>36</sup> J. R. Grover, Phys. Rev. 116, 406 (1959).

<sup>&</sup>lt;sup>37</sup> A. G. W. Cameron, Chalk River Laboratory Report CRP-690, March, 1957 (unpublished).
<sup>38</sup> J. P. Jackson, Can. J. Phys. 34, 767 (1956).

on the evaporation chain is expected to be relatively small. In Jackson's formula, the nuclear temperature  $\tau$  was taken to be

$$\tau = \frac{1}{2} \left[ \left( \frac{B_{FP} + 0.5}{a} \right)^{\frac{1}{2}} + \left( \frac{B_{FP} + B_{UF} + 0.5}{a} \right)^{\frac{1}{2}} \right],$$

where  $B_{UF}$  is the binding energy of the last neutron in Gd<sup>147</sup>. Cameron's calculated value<sup>37</sup> ( $B_{UF}$ =8.25 Mev) was used. The distribution of spins in the compound nucleus was estimated by using in Eq. (5) the extreme assumption that  $T_l(\epsilon)=1$  for  $l \leq l_{\max}$  and  $T_l(\epsilon)=0$  for  $l > l_{\max}$ . This leads to

$$\sigma_c(E) = \sum_{J_c} \sigma_c(J_T^i, \epsilon, J_c^i) = \pi \lambda^2 (l_{\max} + 1)^2$$



FIG. 5. Locus of pairs of values of a and b(0) corresponding to calculated excitation functions which fit the experimental data for the reaction Sm<sup>144</sup>( $\alpha$ ,3n)Gd<sup>145</sup>.

The total cross section for compound-nucleus production  $\sigma_c(E)$  was estimated from other measurements made in this experiment to be  $1.23\pm0.12$  b at a centerof-mass bombarding energy of 32.0 Mev, from which  $l_{\max} \approx 14$ .

In Fig. 4, the experimental data are compared with calculated excitation functions. As in the first example, it was found not possible to determine both a and b(0) with the same data. The locus of values of a and b(0) which fit the data at a point 1.5 Mev above threshold is illustrated in Fig. 5.

Despite the crudeness of the calculations in this example, it is apparent that the value of a is very unlikely to fall as low as  $a\sim 3$  for any set of reasonable assumptions and estimates. Setting b(0)=0 gives  $a\approx 2.8$ , a value very similar in magnitude to the a's which have been obtained from the analyses of many excitation functions when competitive gamma-ray de-excitation



FIG. 6. Schematic drawing of the distribution of population with respect to excitation energy in nucleus F, about 5 Mev above threshold for a (p,2n) reaction. The figure is drawn roughly to scale for  $a \sim 10$  Mev<sup>-1</sup>. (See text.)

has been neglected. On the other hand, the value  $a \sim 0.1 A$  is consistent with the calculated curves plotted in Fig. 5.

Rather unfortunately, both of the experimental examples involve reaction products having nearly magic nucleon numbers. This situation reflects the paucity of threshold nuclear-excitation functions suitable for testing the calculation described in this paper. The wide level spacing near the ground states of the magic and near-magic nuclei permits an effective widening of the threshold region in nuclear reactions where these nuclei are products, making the experimental measurements much easier. Because of the near-magic character of the products, the "law"  $a \sim 0.1 A$  Mev<sup>-1</sup> must not be taken too seriously for the cited examples, because the nearmagic and magic nuclei are just the ones which probably have an atypical dependence of level density on excitation energy.<sup>39</sup> Also, because of the widened threshold region, the effect of the competitive emission of gamma rays is somewhat larger for these examples than is expected for more "normal" reaction products. However, the above qualifications do not modify the main result suggested by the calculations, namely, that in the first Mey or so above threshold in many nuclear reactions in which at least two nucleons are evaporated, the effect of competitive gamma-ray emission is very important.

### DISCUSSION

That the competition between gamma-ray de-excitation and particle emission can strongly influence the course of an excitation function several Mev above the threshold may be seen schematically from Figs. 6 and 7. The unbroken curve in Fig. 6 illustrates, for a typical example, the population distribution in excitation energy (after summing over all spins and parities) in nucleus F,

<sup>&</sup>lt;sup>39</sup> T. D. Newton, Can. J. Phys. 34, 804 (1956).



FIG. 7. Schematic drawing of some (p, xn) excitation functions (roughly to scale for  $a \sim 10$  Mev<sup>-1</sup>) illustrating the apparent 'shift" of the functions to higher energies due to competitive gamma-ray emission (see text).

for a bombarding energy several Mev above the threshold for the formation of P. If the competitive gammaray de-excitation of F is neglected, all of the population to the right of  $B_{FP}$  is assumed to represent that portion of the total population which goes by neutron emission (ignoring charged-particle emission for brevity) to form the (p,2n) reaction product (nucleus P), while all the population to the left of  $B_{FP}$  goes by gamma-ray deexcitation to form the (p,n) reaction product. However, the sample calculations have shown that there can easily be a zone around one or two Mev wide at energies just above  $B_{FP}$  in which most of the population is lost to the formation of P, because it is here that gamma-ray de-excitation competes effectively with neutron emission. The dashed line and shaded region suggest the diminished magnitude of the corrected contribution to the formation of P. At the same time, the calculated cross section for the formation of F (as a product) will be correspondingly increased. In proceeding from  $B_{FP}$ to higher values of  $E_F$ , the dashed line turns upward and finally approaches the unbroken curve as more and more levels, including a greater and greater variety of spins, become energetically available to neutron emission. The influence of the centrifugal barrier in inhibiting neutron emission consequently decreases until the competitive emission of gamma rays loses its importance.

The effect on calculated excitation functions is shown schematically in Fig. 7 for the example of (p,xn) reactions. The unbroken curves represent excitation functions as calculated for a given value of a, but neglecting competitive gamma-ray de-excitation. The dashed and dotted curves represent the same excitation functions, calculated using the same value of a, but with the effect of competitive gamma-ray emission included. The lowenergy side (dashed curve) of each excitation function is depressed by the competition, and the high-energy side (dotted curve) is correspondingly raised. The apparent over-all effect is that the entire excitation function is "shifted" to higher energies, and one sees that the effect is not just localized to the vicinity of the threshold.

If data representing such a shifted excitation function are used to calculate a value of a, ignoring the effect of the gamma-ray competition, the resulting value of awill be erroneously low, even though (as has already been suggested by the two examples) passable-looking curve-fits to the data can be achieved.

In general, the apparent shift will be different for corresponding excitation functions arising from the "same" compound nucleus made by different targetprojectile combinations, because the distributions of population in the spin  $J_c$  of the compound nucleus will be different. Such an effect may account for part of the energy mismatch reported in the experiments of Ghoshal,<sup>40</sup> John,<sup>41</sup> and perhaps, more strikingly, in recent work with energetic heavy ions.42

That the experimental cross sections for reactions in which only one or two particles are emitted  $\lceil e.g.$ , for  $(\alpha, n)$  reactions, etc.] are often much too high, at energies well above their maxima, to be explained by the compound-nucleus theory,43,44 may be partially explained by the effect of the gamma-ray competition. However, such competition appears to be inadequate to fully explain such large discrepancies.

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#### APPENDIX

The lengthy calculational procedure described in Part I may be greatly simplified for many cases when estimates of only moderate accuracy are desired. This is because the J dependence of the function  $S(J_F^i, E_F)$ does not usually depend strongly either on emitted particle energy or on  $\mathcal{I}$  in the narrow regions of  $J_F$  and  $E_F$  of greatest interest in threshold calculations. These simplified calculations and their shortcomings are described in this Appendix.

Making the plausible assumption<sup>18</sup> that  $\omega(J^-,E)$  $=\omega(J^+,E)$ , and summing the right side of Eq. (7) over  $J_{U}$ , one obtains for emitted particle energies restricted to  $\epsilon \leq 3$  MeV, and for  $\mathfrak{I} \geq 0.1 \mathfrak{I}_{rigid}$ ,

$$P(J_{c}, E_{c}, \epsilon, E_{U})d\epsilon$$

$$\sim \frac{1}{\hbar}(2s+1)\frac{\omega(0, E_{U})}{\omega(0, E_{c})}\exp\left\{-\frac{J_{c}(J_{c}+1)}{2\sigma_{U}^{2}}\left[1-\left(\frac{E_{U}}{E_{c}}\right)^{\frac{1}{2}}\right]\right\}$$

$$\times \sum_{l=0}^{\infty}(2l+1)T_{l}(\epsilon)d\epsilon, \quad (19)$$

<sup>40</sup> S. N. Ghoshal, Phys. Rev. 80, 939 (1950).
 <sup>41</sup> W. John, Phys. Rev. 103, 704 (1956).
 <sup>42</sup> A. S. Karamyan, Y. B. Gerlit, and B. F. Myasoedov, Soviet Phys.—JETP 9, 431 (1959).
 <sup>43</sup> J. J. Pinajian and M. L. Halbert, Phys. Rev. 113, 589 (1959).

44 I. Dostrovsky, Z. Fraenkel, and G. Friedlander, Phys. Rev. 116, 683 (1959).

where the summation over  $T_l(\epsilon)$  now includes *all* values of *l*, both even and odd. The above restrictions on  $\epsilon$  and  $\mathscr{G}$  are introduced to make it possible to evaluate (approximately) the sums over  $J_U$  and S which appear during the derivation of Eq. (19).  $\mathscr{G}_{\text{rigid}}$  is the nuclear moment of inertia calculated as if the nucleus were a rigid sphere. Because of the restriction on  $\epsilon$ , this equation is useful only for neutron emission if  $\sigma_U$  is small, although, if  $\sigma_U$  is large enough (i.e. if  $\mathscr{G} \sim \mathscr{G}_{\text{rigid}}$ ), the restriction can be relaxed somewhat, to include proton emission also.

An idea of the inaccuracy of Eq. (19) may be obtained from the following report of the results of a sample calculation. For  $J_c = \frac{7}{2}$ , a = 20 Mev<sup>-1</sup>,  $E_U = 7$  Mev, and  $\mathfrak{s} = 0.1 \mathfrak{s}_{\text{rigid}}$ , the calculated value of  $P(J_c, E_c, \epsilon, E_U)$  for neutron emission from Po<sup>209</sup> is ~6% high at  $\epsilon = 0.6$  Mev (near the maximum in the spectrum of emitted neutrons) and ~35% high at  $\epsilon = 3$  Mev.

Equation (19) can be further simplified for heavy nuclei (A > 100) if it is permissible to set  $\mathscr{I} \sim \mathscr{I}_{rigid}$  in calculating  $\sigma_U$ . For most ordinary situations then, the exponential factor in Eq. (19) seldom drops below 0.7, because we are seldom interested in excitation energies much below about  $E_U \sim 6$  Mev, or in the details of spin population distribution much above J=7. With this rough justification, the exponential factor is simply replaced with unity (i.e., implying that  $\mathscr{I} = \infty$ ).

Finally, performing the indicated summation in Eq. (8) and solving for  $\omega(0,E)$  leads to

$$\omega(0,E) \approx \{2(2\pi)^{\frac{1}{2}} \sigma_U^3 \exp[1/(8\sigma_U^2)]\}^{-1} \rho(E_U) \sim (\operatorname{const}) \rho(E_U), \quad (20)$$

neglecting the relatively weak energy dependence of  $\sigma_U^3$  compared to  $\rho(E_U)$  (the latter approximation requires that  $E_U \gg 0$ ). Substitution of Eq. (20) into Eq. (19), and replacing the exponential factor with unity, as suggested above, gives

 $P(E_c,\epsilon,E_U)d\epsilon$ 

$$\sim (\text{const})\rho(E_U)\epsilon[\pi\lambda^2\sum_{l=0}^{\infty}(2l+1)T_l(\epsilon)]d\epsilon,$$
 (21)

in which the argument  $J_c$  is dropped, as it no longer appears on the right side. Equation 21 is just the Weisskopf spin-independent particle evaporation spectrum.<sup>7</sup> This same result has previously been mentioned by Hauser and Feshbach<sup>45</sup> and by Lane and Thomas.<sup>17</sup>

The spin independence of Eq. (21) leads to the very useful simplification that the energy-dependent part of  $S(J_F^i, E_F)$  may usually be calculated separately from the spin-dependent part, if  $g \gtrsim g_{\text{rigid}}$ , and that, to a reasonable approximation, one may calculate the energy-dependent part of  $S(J_F^i, E_F)$ , and also calculate  $G(\mathcal{E})$ , using the ordinary formulas and methods customarily employed for calculating excitation functions,<sup>7,38,44</sup> e.g., Jackson's formula.<sup>38</sup>

The spin-dependent part of  $S(J_F^i, E_F)$  is calculated using Eq. (7), starting with the calculated relative population distribution in the  $J_c^i$ . It may often be sufficiently accurate to perform the calculation as if the particles were being emitted at one single representative energy,<sup>12</sup> e.g., for an  $(\alpha, xn)$  reaction near threshold choose  $\epsilon \sim (1/x) \mathcal{E}$  if  $\mathcal{E} < x\theta$ . It is only slightly less accurate in some cases to choose  $\epsilon = 0$  and assume only *s*-wave particle emission, as was done in the illustrative example for the reaction Sm<sup>144</sup> $(\alpha, 3n)$ Gd<sup>145</sup>.

The great simplifications described in this Appendix are not applicable to all cases, of course, and are meant only as suggestions. For example, in the calculation for the reaction  $Bi^{209}(p,2n)Po^{208}$ , it was necessary to perform the calculation in considerable detail in order to achieve even moderate accuracy. No definite rules can be formulated; each case must be individually examined to ascertain the detail with which the calculation must be done to achieve a desired accuracy.

<sup>45</sup> W. Hauser and H. Feshbach, Phys. Rev. 87, 366 (1952).