Interpretation of Experimental $(n,2n)$ Excitation Functions*

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Radiochemically determined $(n,2n)$ excitation functions for Sc^{45}, Ti^{46}, Ni^{58}, Cu^{65}, Ge^{70}, As^{75}, Sr^{84}, Rb^{85}, Rb^{87}, Y^{89}, Zr^{90}, Sn^{112}, Cd^{116}, Sb^{123}, and U^{238} have been interpreted in terms of the statistical model of nuclear reactions. Values of the level density parameter a are obtained and correlated with mass number. A procedure is outlined for predicting the magnitude of any $(n,2n)$ cross section from the nuclear content of the target material. Two level density formulations are studied, and approximations customarily made in calculations of this sort are examined quantitatively.

I. INTRODUCTION

SUFFICIENT experimental evidence has been accumulated[1] to confirm the Bohr compound-nucleus idea[2] as a useful concept for describing nuclear reactions for the range of incoming particle energies up to 20 Mev. The estimation of nuclear reaction cross sections based on this idea is due to Weisskopf[3] and collaborators and has been summarized recently by Moore[4] and LeCouteur.[5] In this formulation, it is necessary to know the density of levels, ω, in an excited nucleus. The level density has been derived in general form by Bethe,[6] and for the Fermi-gas nuclear model is given by

$$\omega(E)=C\exp[2(aE)^{\frac{1}{2}}], \qquad (1)$$

where C and a are adjustable constants and E is the excitation energy of the nucleus. The groups of experimental observations summarized by Hurwitz and Bethe[7] and Feld *et al.*[8] have been incorporated in an alternate level density formula proposed by Weinberg and Blatt.[9] This expression, applied to experimental data by Kaufman,[10] is

$$\omega(E)=C\exp[2(aE')^{\frac{1}{2}}], \qquad (2)$$

where

$$E'=(E-\delta)/\{1-\exp[-a(E-\delta)]\},$$

with δ equal to the pairing energy of the nucleus, a term commonly used in atomic mass formulas to express the increased stability of nuclei with paired neutrons and protons.[11] The purpose of this paper is to determine values of the parameter a in expressions (1) and (2) from the shape of experimental excitation functions of several $(n,2n)$ reactions on target nuclei ranging in mass number from 45 to 238. The experimental data of Prestwood and Bayhurst,[12] excluding those measurements which involved only the partial $(n,2n)$ yield of an isomeric or ground state, were used for the analysis along with the data of Knight *et al.*[13] on the reaction $U^{238}(n,2n)U^{237}$. These data were chosen on the basis that they are recent measurements over an extensive energy and mass number range performed within the same laboratory by people utilizing consistent experimental methods.[14] All computations referred to in this paper were performed on an IBM electronic data processing machine, type 704.

II. METHODOLOGY—LEAST SQUARES CALCULATION

Within the framework of the statistical model, the expression for an $(n,2n)$ cross section[4] with threshold at E_t is

$$\sigma_{n,2n}=\sigma_{n,M}\int_0^{E_n-E_t}x\sigma_c\omega dx\Big/\int_0^{E_n}x\sigma_c\omega dx, \qquad (3)$$

where E_n is the incident neutron energy, $\sigma_c=\sigma_c(x)$ is the compound nucleus formation cross section for a neutron with an excited target nucleus whose level density is $\omega=\omega(E_n-x)$, and $\sigma_{n,M}$ is the cross section for neutron emission from the compound nucleus. Inserting Eq. (1) for the level density and neglecting

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the variation of σ_c with energy yields the formula[15]

$$\sigma_{n,2n}/\sigma_{n,M}=1-\frac{\{[1-2(aE_t)^{\frac{1}{2}}][E_t-E_n+3/2a]+2E_t\}}{2E_n\{1-[3/2(aE_n)^{\frac{1}{2}}]+(3/4aE_n)\}}\times\exp\{-2[(aE_n)^{\frac{1}{2}}-(aE_t)^{\frac{1}{2}}]\}. \quad (4)$$

The Appendix contains a comparison of this result with the formula given by Blatt and Weisskopf[3] as well as an estimation of the error involved in neglecting the variation of σ_c with energy.

The experimental data of reference 12 were fitted by the method of least squares to Eq. (4) with a and $\sigma_{n,M}$ as the adjustable constants. This procedure amounts to determining a from the shape of the excitation function rather than from any consideration based on the absolute magnitude of $\sigma_{n,M}$. The significance of these parameters and the reasons for their choice as the ones to be fitted in the calculation will now be discussed and will be followed by pertinent details concerning the least-squares computation.

A. Discussion of Parameters

The magnitude of the parameter a in Eq. (4) is a direct measure of the steepness of ascent of the excitation function. Stated in another way, it is inversely related to the energy above threshold required for the $(n,2n)$ cross section to reach some fraction, say 0.90, of $\sigma_{n,M}$. This energy excess is about 7 Mev for $a=3.5$ and approximately 3 Mev for $a=10$. The a value is quite dependent on the $(n,2n)$ threshold, E_t, and several of the thresholds applicable in this study are uncertain to as much as 0.2 Mev according to the most recent tabulations.[16,17] However, in general, the literature values are better than those obtainable from an analysis of the $(n,2n)$ data including a variable threshold. This fact was ascertained during preliminary calculations, the results of which gave thresholds agreeing with literature values within their standard deviations, but these deviations were frequently 0.3 Mev or more. It was on the basis of this experience that the final calculations were done with the $(n,2n)$ thresholds fixed.

It will clarify discussion of the parameter $\sigma_{n,M}$ to first examine the limitations on the radiochemical method of determining an absolute cross section. For a thin target, the experimentally measured quantity is

$$\sigma_{n,2n}=A^0/(\epsilon B)\lambda n\phi', \quad (5)$$

where A^0=nuclide activity at end of bombardment corrected for decay during irradiation, (ϵB)=product of the counting efficiency and the ratio of observed radiation(s) to total disintegrations in the nuclide decay, λ=nuclide decay constant, n=atoms of target material per square centimeter of target, and ϕ'=total neutrons impinged on the target during irradiation.

In the type of experiments described in reference 12, A^0, λ, n, and ϕ' can each be determined in the range of a few percent accuracy. If β-particle counting is done, ϵ can be determined to good accuracy.[18] B is unity for negatron counting, and an accurate cross section measurement is possible. For positron counting, however, B is not always well known as is the case for $Sr^{84}(n,2n)Sr^{83}$. In counting gross gamma rays, the factor (ϵB) is difficult to estimate, thus the data on the Y^{89}, Sn^{112}, and Sb^{123} $(n,2n)$ reactions are accurate relatively, but absolute cross-section scales are not available at this writing.

The limiting assumption involved in this interpretive treatment of $(n,2n)$ excitation functions is that the parameter $\sigma_{n,M}$ is a constant. Deviations of its value from the nonelastic cross section for the element under consideration, a quantity observed to be essentially flat over the energy region of interest here,[19] must be due to one of the following reasons:

(1) The experimental data are not on an absolute cross-section scale for one of the reasons described in the preceding paragraph.

(2) One or more reactions may be competing with the $(n,2n)$ process in the energy range studied.

The latter is certainly true in the cases of the lighter mass elements studied, but the assumption of constancy of $\sigma_{n,M}$ could still be true. Without complete excitation function data on such reactions as (n,p) and (n,α), the severity of this assumption and its directional effect on the values of a obtained cannot be estimated. Competition within the neutron exit channel category, specifically from the (n,np) reaction, could be important at the higher energies in the lighter mass elements, leading to high values of a obtained.

Thus, inclusion of $\sigma_{n,M}$ as a variable in the least-squares calculation was necessary in order to include the Sr^{84}, Y^{89}, Sn^{112}, and Sb^{123} data, since their cross-section scales are only relative. However, this method of analysis was used throughout for the sake of consistency and to avoid errors in the conversion of the experimental data to absolute cross sections. For the remainder of the nuclides studied, it must suffice to define the significance of the value of $\sigma_{n,M}$ as the nonelastic cross section minus the sum of the cross sections for all processes

[15] The denominator in the integrated expression is strictly

$$\frac{E_n}{a}\left(1-\frac{3}{2(aE_n)^{\frac{1}{2}}}+\frac{3}{4aE_n}\right)\exp(2[aE_n]^{\frac{1}{2}})+\frac{(E_n-3/2a)}{2a}.$$

A comparison of magnitudes for values of E_n and a applicable in this study shows that the second term may be neglected without introducing significant error; hence the final result, Eq. (4).

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TABLE I. Results of analysis of experimental $(n,2n)$ excitation functions.

Target nucleus	$(n,2n)$ threshold[a] (Mev)	a from Eq. (4) (Mev^{-1})	Δa / $\Delta E_t=+0.1$ (Mev^{-2})	Δa / $\Delta E_t=-0.1$ (Mev^{-2})	$\sigma_{n,M}$ from Eq. (4) (mb)	$\Delta\sigma_{n,M}$ / $\Delta E_t=\pm0.1$ (mb Mev^{-1})	Pairing energy[b] (Mev)	a Including pairing energy (Mev^{-1})
Sc^{45}	11.57	3.51±0.37	0.62	−0.56	599±27	20	1.41	2.90
Ti^{46}	13.48	3.50±0.46	1.35	−1.16	280±13	26	3.14	2.49
Ni^{58}	12.14	3.13±0.32	0.58	−0.49	85.7±3.7	2.4	3.69	2.00
Cu^{65}	10.06	4.94±0.34	0.74	−0.65	1143±22	24	1.46	4.01
Ge^{70}	11.77	7.74±0.36	1.00	−0.88	950±15	8	2.88	5.55
As^{75}	10.29	5.73±0.90	0.80	−0.70	1346±57	22	1.47	4.66
Sr^{84}	12.04	6.63±0.63	1.19	−0.99	291±13[c]	7	2.48	5.04
Rb^{85}	10.49	6.29±1.37	0.82	−0.70	1814±108	25	1.23	5.35
Rb^{87}	10.03	6.57±1.00	0.88	−0.77	1372±48	18	2.27	4.90
Y^{89}	11.56	7.60±1.21	1.79	−1.51	19.09±1.22[c]	0.92	2.27	5.89
Zr^{90}	11.95	6.96±0.34	1.12	−0.98	1272±26	24	3.27	4.83
Sn^{112}	11.19	7.84±0.96	1.26	−1.10	6.673±0.260[c]	0.14	3.68	5.05
Cd^{116}	8.71	8.12±1.83	0.73	−1.24	1728±46	6	2.62	5.63
Sb^{123}	9.02	8.74±3.26	0.73	−0.95	1311±51[c]	4	1.24	7.47
U^{238}	6.13	9.70±1.05	2.70	−2.17	1585±40[d]	43	1.36	7.54

[a] Best value estimates from data of references 11, 16, 17, 21–24.
[b] Taken from reference 25 with 1.0 Mev added for magic-number nuclides.
[c] Cross-section scale is not absolute.
[d] Fission cross section in 7–10 Mev region is flat at 1.0 barn.

other than inelastic scattering and $(n,2n)$, on the assumption that these other reactions are flat in the energy region of $(n,2n)$ data accumulation.

B. Calculational Details

Uncertainty weighting[20] on both the cross sections and energies was included in the least-squares calculations. The cross-section uncertainties quoted in reference 12 are the statistical or random-error estimates, which include the errors in the neutron flux and in the chemical and counting procedures. The errors on the average energies were assessed on the basis of target geometry. They are sufficiently large to validate the assumption that the experimentally determined quantity, Eq. (5), with ϕ' given by

$$\phi'=\int_{E_L}^{E_H}\varphi(E)dE,$$

where E_H and E_L= the maximum and minimum energies, respectively, of the neutron distribution $\varphi(E)$, may be expressed as a function of the average incoming neutron energy, which is strictly true only if Eq. (4) is linear in the range of E_L to E_H.

In all instances, the only selective elimination of data from the computations were energy points above the $(n,3n)$ thresholds for the reactions studied, since Eq. (4) does not include the eventual decrease in the $(n,2n)$ excitation function due to competition from the tertiary reaction. Q-value data were taken from references 11, 16, 17, and 21–24. Best value estimates of the laboratory $(n,2n)$ thresholds used in the calculations are given in Table I. Standard deviations on the fitted parameters are based on the external consistency criterion described by Deming.[20]

III. METHODOLOGY—CALCULATIONS BY NUMERICAL INTEGRATION

The calculations for the level density parameter a with Eq. (2) inserted into (3) were done by numerical integration. The pairing energies used are listed in Table I. They were taken from Cameron[25] with 1 Mev added for magic-number nuclides after Kaufman.[10] The neutron compound-nucleus formation cross sections were computed from the formulas of Blatt and Weisskopf[3] at low energies, using a sufficient number of orbital angular momentum values[26] and interpolated from plots[3,8] at higher energies. Nuclear radii were computed from the formula[27]

$$R=(1.2A^{\frac{1}{3}}+2.1)\times10^{-13}\text{ cm}.$$

The Appendix contains a comparison of the $(n,2n)$ excitation function shape based on this formulation with Eq. (4). Final values of a were obtained assuming the least-squares results for $\sigma_{n,M}$.

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IV. RESULTS

Table I is a presentation of the results of the calculations which have been described. The values of a and $\sigma_{n,M}$ obtained from the least-squares analysis are listed in columns 3 and 6. The standard errors given on the a values do not include any estimate of the uncertainty in the $(n,2n)$ thresholds used (column 2), but columns 4 and 5 give the least-squares solution change produced in a for a 0.1-Mev increase and a 0.1-Mev decrease, respectively, in the listed thresholds. The corresponding changes in $\sigma_{n,M}$ are small and may be taken as varying linearly over the same energy region so that

$$[\sigma_{n,M}]_{E_t\pm 0.10}=[\sigma_{n,M}]_{E_t}(1\mp\Delta\sigma_{n,M}),$$

where the $\Delta\sigma_{n,M}$'s are the entries in column 7. The results for the level density parameter a, including pairing energies (column 8), are given in column 9 of the Table.

V. PARAMETER CORRELATIONS

A. The Parameter $\sigma_{n,M}$

On the basis of the definition put forth in Sec. II.A for this parameter, one would predict a correlation between $\sigma_{n,M}$, the nonelastic cross section σ_{nx}, and some parameter which is a measure of the probability for competition from other reactions. Such a quantity is $(N-Z)/A$, where N is the neutron number, Z the proton number, and A the mass number of a nuclide. This factor predicts the general trends observed in the barrier and threshold effects which govern charged particle emission. Figure 1 shows the ratio $\sigma_{n,M}/\sigma_{nx}$ plotted against $(N-Z)/A$ for the nuclides studied which have

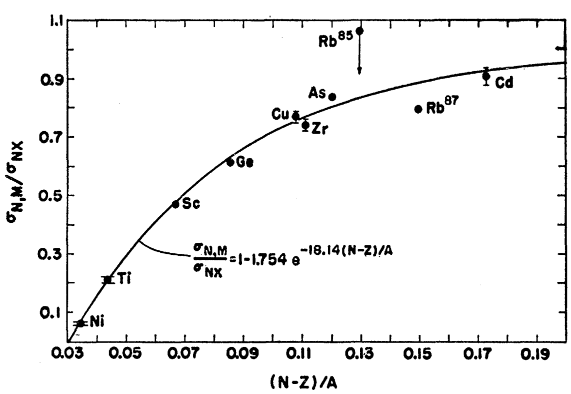

FIG. 1. Empirical correlation of the least-squares-fitted parameter, $\sigma_{n,M}$. The ratio of this parameter to the nonelastic cross section of the target element is plotted against the ratio of the target neutron excess to its mass number, taken as a measure of the probability for competing reactions [see Sec. V.A of the main text]. It should be noted that the ordinate scale supersedes the limiting value of unity in order to include the Rb^{85} result obtained from the direct analysis of the data of reference 12. The direction and magnitude of the change shown in this point is based on a more recent decay scheme[16] than that used by the authors of reference 12. The solid line, given by the equation shown on the plot, is an empirical formula which adequately represents the results.

an absolute $(n,2n)$ cross-section scale. The points with error limits are based on measured nonelastic cross sections,[28] but these limits do not include any uncertainty in the (ϵB) factors used to fix the cross-section scales. For the points without error limits, the formula[27]

$$\sigma_{nx}=\pi(0.12A^{\frac{1}{3}}+0.21)^2 \text{ barns} \qquad (6)$$

was used to compute the nonelastic cross secton. The correlation is good, considering the limited amount of data available and the fact that (ϵB) errors would cause deviations from a smooth relationship. The empirical formula indicated on the graph gives a reasonable fit to the data and yields a β^+/E.C. branching ratio of 0.3 for Sr^{83} as well as credible gross γ-counting (ϵB) factors for Y^{88}, Sn^{111}, and Sb^{122}. In addition, the nonelastic cross section for U^{238} is calculated to be 2.64 barns in the 6–10-Mev region, a value compatible with existing measurements.[26]

B. The Level Density Parameter a

Simple nuclear models predict a direct proportionality of a with mass number A. Figure 2 is a plot of a from the least-squares calculations against mass number of the target nucleus. The dashed line given by $a=A/13$ fits the results up to $A=120$, in accord with the findings of others.[29–31] The full line which fits the lower mass numbers equally well, but also includes the U^{238} result, is an empirical formula not far different from one given by Weisskopf,[32]

$$a=0.85(A-40)^{\frac{1}{2}} \quad \text{for} \quad A>60.$$

C. Predictions

Figures 1 and 2, along with Eq. (4) and a knowledge of the $(n,2n)$ threshold, are presumably all that is necessary to compute any $(n,2n)$ cross section up to the maximum of its excitation function. The task of comparing the predictions of the systematics with every $(n,2n)$ cross section published in the literature was not undertaken, especially in the light of occasional large discrepancies in purported duplicate measurements. However, the pair of examples given in Table II will illustrate the potential utility of this systematic procedure for predicting an $(n,2n)$ cross section from only the neutron and proton content of the target nucleus.

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TABLE II. Comparison of predicted and observed (n, 2n) cross sections.[d]

Nuclide	$(N-Z)/A$	$\sigma_{n,M}/\sigma_{nx}$ from Fig. 1	σ_{nx} from Eq. (6)[a] (mb)	$\sigma_{n,M}$ (mb)	a from Fig. 2 (Mev^{-1})	(n,2n) threshold[b] (Mev)	(n,2n) cross section at 14.5 Mev (in mb) Calculated from Eq. (4)	Observed[c]
$_{29}Cu_{34}^{63}$	0.079	0.585	1490	872	4.8	11.00	600	650
$_{42}Mo_{50}^{92}$	0.087	0.635	1780	1130	7.0	13.27	275	280

[a] See reference 27.
[b] From reference 17.
[c] These values are taken from *Neutron Cross Sections*, compiled by D. J. Hughes and R. Schwartz, Brookhaven National Laboratory Report BNL-325 (Superintendent of Documents, U. S. Government Printing Office, Washington, D. C., 1958), 2nd ed.
[d] The predictions of the systematics described herein have been compared with the recent data of L. A. Rayburn, Phys. Rev. 122, 168 (1961) at 14.4±0.3 Mev. Out of thirteen total (n,2n) cross-section measurements over the mass region 50 to 144, where the systematics are expected to apply, nine of the predicted cross sections were within ±20% of the experimental numbers. This is considered satisfactory agreement based on the comparison of Rayburn's data with that of Prestwood and Bayhurst for seven reactions common to the two studies. Of the remaining four cases, only $Fe^{54}(n,2n)Fe^{53}$ had a deviation greater than ±45%.

VI. A CONCLUSION CONCERNING THE OPTIMUM EXPERIMENTAL MEASUREMENT

A study of function (8) in the Appendix and its derivative along with definition (9) leads to certain conclusions regarding the inherent sensitivity for determining a. Assuming that the (n,2n) threshold, E_t, is known and that $\sigma_{n,M}$ has been fixed through measurements made near the maximum of the excitation function, then if all (n,2n) cross sections could be measured to the same magnitude of uncertainty, the measurement at

$$(E_n-E_t)=2T\simeq 2(E_t/a)^{\frac{1}{2}}$$

would minimize the error in a. If, however, all cross-section measurements could be made to the same percentage accuracy, then the closest one possible to E_t would give the smallest error in a. In practice, the relative experimental uncertainties are usually such that the minimum error in the value of a is attained when the measured (n,2n) cross section is in the energy region

$$T<(E_n-E_t)<2T;\quad T\simeq(E_t/a)^{\frac{1}{2}}.$$

Based on this criterion, the optimum cross-section measurements for the Au and Tl data of reference 12 would be in the 9–10-Mev region. The available data begin at 12 Mev, however, at which energy it can be shown that a cross section with 5% standard error is only capable of fixing the value of a to 37%. For this reason, and since the 12-Mev points are the only ones indicating any significant rise in the excitation functions for Au and Tl, these nuclides were omitted from the analysis presented in this paper.

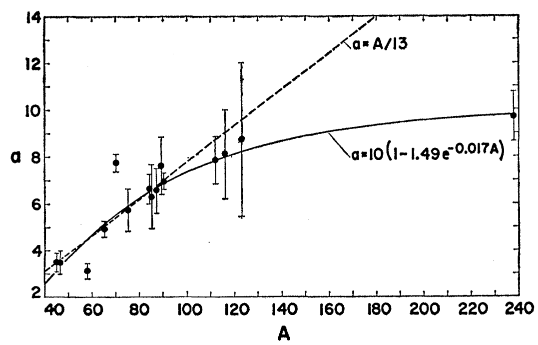

FIG. 2. Correlation of the level density parameter, a, from the least squares calculations with target mass number. The dotted line is the fit of these results assuming the Fermi-gas model prediction[33] that $a=\text{constant}\times A$. The full line, given by the equation shown, is an empirical formula which fits the data better in that it includes the U^{238} result.

VII. SUMMARY

It must be pointed out that due to the uncertain weighting on the cross sections and energies, the least-squares goodness of fit criterion[20] cannot be interpreted as completely confirming the validity of Eq. (4). However, the reduced sum of the squares[20] on all solutions lay between a few tenths and unity, indicating that the shape function, Eq. (4), is a good representation of the observed data, the errors on which have probably been slightly overassessed by the authors of reference 12. Thus, the statistical model formalism is adequate to account for observed shapes of (n,2n) excitation functions in the energy range explored herein, and accurate measurements in the proper E_n-E_t region have the capacity to determine the level density parameter a with reasonable accuracy. Extremely precise data would be necessary to differentiate between similar excitation function shapes arising from near-equivalent level density formulations. The results for a up to mass 120 may be interpreted as giving at least a partial confirmation of the Fermi-gas model prediction[33] that $a=\text{constant}\times A$, a result in accord with recent work on neutron spectra from (p,n) reactions.[30,31]

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APPENDIX

A generalized expression for an $(n,2n)$ cross section given by Blatt and Weisskopf[3] is

$$\sigma_{n,2n}=\sigma_{n,M}\left\{1-\left(1+\frac{(E_n-E_t)}{\theta}\right)\exp\left[-\frac{(E_n-E_t)}{\theta}\right]\right\},$$

where $\theta=\theta(E_n)$ is the nuclear "temperature" and the other terms have the same significance described in the main text of this paper. The derivation of this formula utilizes the following thermodynamic definitions of entropy S and temperature θ:

$$S(E)=\ln\omega(E);$$
$$1/\theta(E)=dS/dE. \qquad (7)$$

In addition, the approximations are made that (a) the entropy may be expanded in a Taylor's series with neglect of higher derivatives in accord with the single definition (7); (b) a subtractive term in the denominator of the integrated expression for the $(n,2n)$ cross section is negligible; (c) the variation of σ_c with energy in Eq. (3) may be neglected. If the temperature is regarded as a constant, T, (a) above is exact and the formula,

$$\sigma_{n,2n}/\sigma_{n,M}=1-\left(1+\frac{(E_n-E_t)}{T}\right)\exp\left[-\frac{(E_n-E_t)}{T}\right], \qquad (8)$$

involves only assumptions (b) and (c). The corresponding level density is

$$\omega(E)=C\exp(E/T).$$

If the temperature is given by[3]

$$\theta(E_n)=(E_n/a)^{\frac{1}{2}}, \qquad (9)$$

which is equivalent to defining the level density as in Eq. (1), the formula

$$\sigma_{n,2n}/\sigma_{n,M}=1-\left[1+\frac{(E_n-E_t)}{(E_n/a)^{\frac{1}{2}}}\right]\exp\left[-\frac{(E_n-E_t)}{(E_n/a)^{\frac{1}{2}}}\right] \qquad (10)$$

contains approximations (a), (b), and (c) above. Equation (4) used in this paper involves only assumption (c). Curves A, B, and C of Fig. 3 illustrate the magnitude of these approximations for a hypothetical nuclide with $(n,2n)$ threshold at 10 Mev and an a value of 5.5. The conclusion is that assumption (c) is good and that Eq. (4) is a significant improvement over the approximate Eq. (10) insofar as interpreting the value of a as the parameter in level density Eq. (1). Inclusion of the pairing energy, that is, level density expression (2) inserted into (3), gives curve D for the same hypothetical case and $\delta=1.5$. In this latter formulation, the combination of $a=4.5$ and $\delta=1.5$ yields a curve identical to C. Thus for a given set of experimental data, the value of a obtained from an analysis of that data decreases with a span of 2–3 units as one proceeds from curves A to D or, strictly, in the formulations leading to those curves.

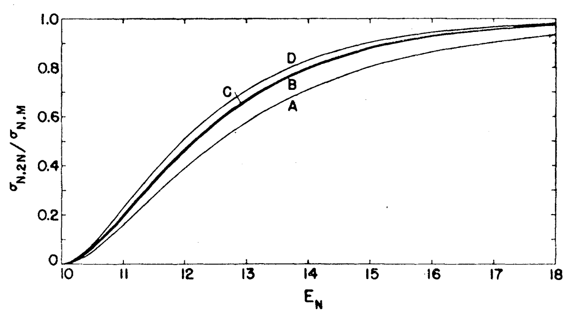

FIG. 3. Comparison of $(n,2n)$ excitation function shapes based on calculational approximations and two level density formulations. Curve A: Eq. (10) with $E_t=10$ Mev and $a=5.5$ Mev^{-1}. Curve B: Eq. (4) with $E_t=10$ Mev and $a=5.5$ Mev^{-1}. Curve C: Eq. (3) including $\sigma_c(x)$ with ω given by Eq. (1) for $E_t=10$ Mev and $a=5.5$ Mev^{-1}. Curve D: Eq. (3) including $\sigma_c(x)$ with ω given by Eq. (2) for $E_t=10$ Mev, $a=5.5$ Mev^{-1}, and $\delta=1.5$ Mev.