The Statistical Theory of Nuclear Reactions

Bernard L. Cohen Oak Ridge National Laboratory, Oak Ridge, Tennessee (Received July 9, 1953)

It is pointed out that the discrepancies between experimental results and the predictions of the statistical theory of nuclear reactions can be traced to peculiarities in the energy distribution of particles emitted in the breakup of the compound nucleus. Experimental energy distributions are analyzed, and it is shown that very strange assumptions-susceptible to experimental check-are necessary to explain them in any way without the introduction of a new selection rule in nuclear transitions. Experiments are suggested to clarify these problems.

T has by now become quite clear that large discrepancies exist between the predictions of the statistical theory of nuclear reactions and the results of experiments designed to test them. In particular, it appears that nuclear temperatures calculated from experimental-level densities¹ are much higher than those obtained from measurements of energy spectra of emitted particles,²⁻⁵ much lower than those required to explain the cross sections of (n,p), (n,α) , (p,pn), and $(\alpha, pn)^{6-10}$ reactions, but nevertheless, approximately equal to those found from $(n,2n)^{6-8}$ and $(\alpha,2n)^{11,12}$ cross-section measurements. In an effort to clarify this situation, the theory is here developed to a form more suitable for comparison with the experimental results, the experiments are analyzed in this light, and some conclusions about the difficulties with nuclear reaction theory are pointed out.

We begin with a derivation of the equation for the energy spectrum of particles emitted in the breakup of a compound nucleus. Consider the reversible reaction,

$$C(E_0+B) \leftrightarrows A(E_0-E) + a + E, \tag{1}$$

where C, A, and a are the compound nucleus, residual nucleus, and emitted particle, respectively, the quantities in parentheses are their excitation energies (E_0 is maximum possible kinetic energy available, B is binding energy of a to A), and E is the kinetic energy released, practically all of it appearing as the kinetic energy of a. The probability per unit time, $\lambda(\rightarrow)$, of (1) proceeding to the right into energy interval ΔE , is

$$\lambda(\rightarrow) = (2\pi/\hbar) |M|^2 \omega_A (E_0 - E) \omega_a(E) \Delta E, \qquad (2)$$

where |M| is the average matrix element for the transition (averaged over the levels within energy

¹J. Blatt and V. F. Weisskopf, Theoretical Nuclear Physics (John Wiley and Sons, Inc., New York, 1952).
²P. C. Gugelot, Phys. Rev. 81, 51 (1951).
³P. C. Gugelot and H. A. Lauter, Phys. Rev. 91, 486 (1953). (The author is indebted to Dr. Gugelot for permission to use this data in advance of publication.)
⁴P. H. Stelson and C. Goodman, Phys. Rev. 52, 69 (1951).
⁵E. R. Graves and L. Rosen, Phys. Rev. 89, 343 (1953).
⁶H. Wäffler, Helv. Phys. Acta 23, 239 (1950).
⁷B. L. Cohen, Phys. Rev. 81, 184 (1951).
⁸E. B. Paul and R. L. Clarke, Can. J. Phys. 31, 267 (1953).
⁹S. N. Ghoshal, Phys. Rev. 80, 939 (1950).
¹⁰ Newman, Cohen, and Handley, Phys. Rev. 91, 486 (1953).

- ¹⁰ Newman, Cohen, and Handley, Phys. Rev. **91**, 486 (1953). ¹¹ J. Bradt and D. J. Tendam, Phys. Rev. **72**, 1118 (1947).
- ¹² Bleuler, Stebbens, and Tendam, Phys. Rev. 90, 460 (1953).

interval ΔE), $\omega_A(E_0-E)$ is the level density of A at excitation $E_0 - E$, and $\omega_a(E)$ is the degeneracy of a (in spin, momentum, and physical space). By the same token, the probability per unit time of (1) proceeding to the left is

$$\lambda(\leftarrow) = 2\pi/\hbar |M|^2 \omega_C(E_0 + B) \Delta E.$$
(3)

However, $\lambda(\leftarrow)$ may be expressed in terms of the cross section for (1) proceeding to the left, σ , as

$$\lambda(\leftarrow) = \sigma v / V, \tag{4}$$

where v is the velocity of a and V is the volume in which the system is contained. Using (4) and (3),

$$|M|^{2} = \frac{\hbar}{2\pi} \frac{\sigma v}{V} \frac{1}{\omega_{C}(E_{0}+B)\Delta E},$$
(5)

whence (2) becomes

$$\lambda(\to) = \frac{\sigma v}{V} \frac{\omega_A(E_0 - E)}{\omega_C(E_0 + B)} \omega_a(E).$$
(6)

The physical space degeneracy of a is proportional to V. the momentum space degeneracy is proportional to $E^{\frac{1}{2}}\Delta E$, and $v \propto E^{\frac{1}{2}}$, so that $I(E)\Delta E$, the dependence of $\lambda(\rightarrow)$ on *E*, may be expressed as

$$I(E) = \sigma E \omega_A(E_0 - E). \tag{7}$$

Formula (7), which was originally due to Weisskopf,¹³ was derived here without taking into account angular momentum and parity selection rules. This problem, however, was treated in detail by Wolfenstein,14 who found that the energy distribution, while similar to (7), also depends on the density of energy levels with respect to spin. With any reasonable assumption about this quantity, (7) is altered only slightly; however, it is important to bear in mind that (7) is not an exact formula.

For purposes of the discussion below, we introduce the "sticking probability," η , by

$$\eta = \sigma / \sigma_0, \tag{8}$$

where σ_0 is the reaction cross section given in reference 1 with all $\eta_l = 0$. For neutrons, σ_0 is approximately con-

¹³ V. F. Weisskopf, Phys. Rev. 52, 295 (1937).
 ¹⁴ L. Wolfenstein, Phys. Rev. 82, 690 (1951).

stant $(=\pi r^2)$ for energies above about 1 Mev, while for charged particles it is less by the Coulomb barrierpenetration factor. From (8) and (5) we see that η must contain any selection rules that are effective in nuclear transitions.

In the measurement of energy spectra of emitted particles, the quantity determined is the rate of change of I(E) with E. Thus, a quantity very directly obtainable from the measurements is

$$\Sigma = \left[-\frac{\partial \log(I(E)/\sigma_0 E)}{\partial E} \right]^{-1}.$$
 (9)

From (7) and (8), the measured quantity Σ may be interpreted as

$$\Sigma^{-1} = -\frac{\partial \log \eta}{\partial E} - \frac{\partial \log \omega (E_0 - E)}{\partial E}.$$
 (10)

Using the conventional thermodynamic analogies,

$$S = \log \omega, \quad \partial S / \partial \epsilon = 1/T(\epsilon),$$
 (11)

Equation (10) becomes

$$\frac{1}{\Sigma} = -\frac{\partial \log \eta}{\partial E} + \frac{1}{T(E_0 - E)}.$$
(12)

Equation (12) may be compared with three types of experimental results. Measurements of energy distributions of particles emitted from nuclear reactions give I(E), and therefore Σ , directly. The total cross section for nuclear reactions in which charged particles are emitted is given by¹

$$\sigma(a, b) \simeq \sigma_a f_b / \sum_i f_i, \tag{13}$$

where σ_a is the cross section for capture of a to form a compound nucleus, f_i is





FIG. 1. Σ vs $(E_0 - E)$ from inelastic proton scattering. According to the usual theory, the slopes of these curves should be positive rather than negative. Σ was calculated from the lines drawn through the 150° data by the authors of reference 3. We are indebted to them for permission to use their data in advance of publication.

 $(m_i$ is the mass of particle i), and the summation in (13) is over all particles that can be emitted. Thus, cross sections for nuclear reactions are determined by I(E) and, therefore, give information about Σ . Somewhat better information is obtained from (x, 2n) reactions; the cross sections for these are¹

$$\sigma(x, 2n) = \sigma_x \int_0^{\epsilon - B} I_n(E) dE \bigg/ \int_0^{\infty} I_n(E) dE, \quad (15)$$

where ϵ is the incident energy of x, and B is the threshold for the (x, 2n) reaction. Experimental excitation functions of (x, 2n) reactions can therefore be differentiated to give relatively direct information on I(E).

In comparing (12) with the experimental results, we first take the point of view of the usual theory, as presented in reference 1. The basic assumption of this theory is that η is identically unity (except for reflections due to sudden change of wavelength which are negligible above about 1 Mev). With this assumption, (12) becomes

$$\Sigma = T(E_0 - E). \tag{16}$$

To investigate (16), an analysis was made of published experimental results to determine values of Σ . Figures 1 and 2 show the values of Σ obtained from Gugelot's data^{2,3} on energy spectra of inelasticallyscattered protons and of neutrons from (p,n) reactions. The most striking feature of these curves is that in every one of the twelve cases, Σ decreases as $(E_0 - E)$ increases. This feature was verified by performing similar analyses on every other applicable work; on the inelastic proton scattering energy spectrum of Levinthal et al.¹⁵ (see Fig. 3); on the spectra of neutrons emitted from (d,n) reactions at 90°16,17 (in the latter reference there is good evidence that at angles as large as 90° , compound nucleus interaction is predominant); and on



FIG. 2. Σ vs $(E_0 - E)$ from (p,n) reactions (see caption for Fig. 1). Σ was calculated from the data of reference 2.

¹⁵ Levinthal, Martinelli, and Silverman, Phys. Rev. 78, 199 (1950).
 ¹⁶ C. Grosskreutz, Phys. Rev. **76**, 482 (1949).
 ¹⁷ B. L. Cohen and C. E. Falk, Phys. Rev. **84**, 173 (1951).

1246

inelastically scattered neutron spectra^{4,5} (although the statistics are somewhat poor in this case). In each case, without a single exception, Σ decreases with increasing (E_0-E) . In the light of (16), this result is very surprising; it indicates that temperatures decrease as excitation energy increases. This runs contrary to the most basic tenets of the thermodynamic analogy. But even if we are unwilling to assign any thermodynamic properties whatsoever to the nuclear temperature, this result demands that temperatures measured, for example, by Gugelot's method,² would increase as the energy of the bombarding particle decreases. Extrapolated to higher energies, it would mean that energy distributions of particles emitted from stars produced by cosmic rays and synchrocyclotrons would have very low temperatures; this is contrary to the findings of many experimenters.

Another requirement for the validity of (16) is that, for a given final nucleus, Σ must be a function of (E_0-E) only. The most directly applicable data on this is the case of Al²⁷ which is the final nucleus in inelastic scattering of either protons or neutrons from aluminum. Figure 3 shows the results of three inelasticscattering experiments in which the incident particles were 30-Mev protons,¹⁵ 18-Mev protons,² and 14-Mev neutrons.⁵ In each case the values of Σ for a given value of $(E_0 - E)$ are very different.

An additional, though less reliable, check of this type is available from the neutron-energy spectra from (d,n)reactions on copper induced by 10-Mev¹⁶ and by 16-Mev¹⁷ deuterons. The values of Σ differ by at least 40 percent for the same values of $(E_0 - E)$.

If one is willing to assume that temperatures of nuclei of approximately the same mass are equal, there are several other tests available. Data on (n,p), (n,α) , (n,2n), and $(\alpha,2n)$ cross sections, neutron energy spectra, and proton energy spectra (see Figs. 1 and 2) provide several cases where, for the same value of (E_0-E) and approximately the same nuclear mass, experimental values of Σ vary widely. Thus, certainly for nuclei up to mass 30, probably for nuclei up to mass 60, and possibly for nuclei up to mass 100, there is good experimental evidence that Σ is not a function of $(E_0 - E)$ alone, as is indicated by (16).

In summary, the validity of (16) is supportable only if one is willing to accept *each* of the following seemingly improbable consequences.

(a) Nuclear temperatures, as defined in (11), do not increase with excitation energy in the energy region investigated here.¹⁸ This demands, for example, that if Gugelot's neutron spectra² were measured with 12-Mev incident protons rather than with 16 Mev, the temperatures obtained would be higher.



FIG. 3. Σ vs $(E_0 - E)$ for the final nucleus Al²⁷. These curves were calculated from the data of references 3, 5, and 15.

(b) For some reason, the derivation of Eq. (7) breaks down badly for aluminum and probably for elements at least up to mass 60, causing Σ not to be a function of $(E_0 - E)$ only.

To test whether (16) is tenable at all, measurements of neutron energy spectra similar to Gugelot's² with \sim 12-Mev incident protons [for higher proton energies, (p,2n) reactions interfere], and of inelastic proton energy spectra for incident protons of about 14 Mev, 24 Mev, and 30 Mev should be performed. Actually, any one of these experiments would probably settle the matter fairly conclusively, although the neutron energy spectra might not be sufficiently accurate for heavy elements.

If these experiments should indicate that (16) is untenable, two alternatives remain. One is that other interactions besides compound nucleus formation are important in the reactions under consideration. Mc-Manus and Sharpe¹⁹ have proposed that they may be due to a direct interaction ("knock-out" process). It would seem that this should influence only the highest energy portion of the spectrum of emitted particles, whereas the difficulties described above are in evidence at all energies. In addition, it would influence the spectrum only at small angles, whereas the data for Fig. 1 were taken at 150°. (Actually, there is some evidence in reference 2 for more high-energy inelastically scattered protons at 60° than at 150°. However, the effect is not large and might be explained, for example, as inelastic scattering by electric interactions). The matter can be settled experimentally, by looking for a forward peak in the angular distributions. Allen et al.20,21 have measured angular distributions of neutrons from (α, n) reactions for several elements, and their data shows no evidence for a forward peak. Similar measurements for (n,p), (n,α) , (p,n), and (p,α) reactions would give a conclusive answer to this problem. However, as mentioned above, even if it were proved that direct interactions are important, the difficulties in explaining energy spectra at large angles

¹⁸ V. F. Weisskopf (private communication) has pointed out, however, that the nuclear spectrum might change character when going from one energy region to another (independent particle character to statistical character). Then the level density might increase so strongly in that energy region that the nuclear temper-ature defined by (11) is decreasing with increasing excitation.

¹⁹ H. McManus and W. T. Sharpe, Phys. Rev. 87, 188 (1952).

 ²⁴ Allen, Nechaj, Sun, and Jennings, Phys. Rev. 81, 526 (1951).
 ²⁴ B. L. Cohen, Phys. Rev. 81, 632 (1951).

would still remain. Also, the experiments of Ghoshal⁹ lend strong support to the general validity of the compound nucleus model at these energies.

The only remaining alternative that seems obvious is to retain the compound nucleus model, but abandon (16) in favor of the more general formula (12). If the $\partial \log \eta / \partial E$ term in (12) is to contribute appreciably, it must be of the order of an inverse nuclear temperature; that is, η must decrease by a factor of e(2.7) for a decrease in the emitted particle energies of a few Mev. In view of (5) and (8), this would mean that there are selection rules operating in nuclear transitions such as to make transitions between distant levels more probable than transitions between levels of nearly equal energy.

One objection to this alternative is that the many published measurements of total reaction cross sections for neutrons, protons, alphas, and pi mesons are actually direct measurements of the sticking probability, η ; and in every case, they indicate that η is its maximum theoretical value, unity. In these cases, of course, only

ground-state nuclei were bombarded whereas the η in (12), which entered through the reciprocity theorem, is the sticking probability when excited nuclei are bombarded. Obviously, there can be no direct experimental measurements of η for such cases.

Another objection to this alternative, pointed out by Weisskopf,¹⁸ is that it requires that sticking probabilities be less for excited than for ground state nuclei, whereas according to current theories of nuclear structure, the opposite should be the case.

On the other hand, Wigner²² has pointed out independent evidence for the selection rule mentioned above in heavy elements from comparison of known absolute level densities with Gugelot's data.³

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²² E. P. Wigner (private communication).

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The Radionuclides of Arsenic Produced by Deuteron Bombardment of Germanium*

H. J. WATTERS[†][‡] AND J. F. FAGAN, JR.[‡][§] Department of Physics and Radioactivity Center, Laboratory for Nuclear Science, Massachusetts Institute of Technology, Cambridge, Massachusetts

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The arsenic produced by a deuteron bombardment of germanium has been studied to determine the nuclides present in the mixture. Identification of the isotopes was made by comparing measured values of half-life and maximum β energy with published values. Counting rates were measured with 4π and coincidence counters, obtaining half-lives which indicated that the nuclidic mixture was made up of As⁷¹, As⁷², As⁷³, As⁷⁴, and As⁷⁷. These findings were confirmed by maximum β energy values obtained by absorption measurements and by γ -energy values found using a γ -ray scintillation spectrometer. Measurements indicated that the 40-hr half-life reported for As⁷⁷ is in error by a significant amount, and that no As⁷⁶ was obtained from this bombardment. Thick target yield data were determined for each nuclide from the 4π counter measurements.

N investigation was made of the radionuclides of arsenic produced by the cyclotron bombardment of a thick germanium target with 15-Mev deuterons. After chemical separation of the arsenic,¹ identification of the isotopes present was made by correlating measured values of γ , β^+ , and total β half-lives with maximum β energy and γ -ray energy.

Lieutenant, U. S. Navy

Arsenic activity was measured continuously for a period of 53 days with a 4π proportional counter, a γ - γ coincidence counter, and with a thin end-window Geiger Muller tube using calibrated aluminum absorbers. Gamma-ray energy measurements were made using a thallium-activated sodium iodide scintillation spectrometer. The energy spectrum up to 3 Mev was scanned continuously for the first 72 hours (Fig. 1) and an additional spectrum was obtained 52 days after bombardment.

By application of the method of least squares to 4π and coincidence counter data, the decay curves were analyzed in a total of four periods: 25.8 hours, 48.2 hours, 17.8 days, and 88.9 days. Comparison of total β -decay curves with those due only to positron disintegration yielded an additional period slightly greater

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¹ Lieutenant Commander, U. S. Navy. 1 Now on sea duty, U. S. Navy. Investigation performed while a U. S. naval postgraduate student at Massachusetts Institute of Technology, Cambridge, Massachusetts.

¹ Brownell, Backofen, White, and Irvine, Massachusetts Institute of Technology Progress Report, May 1953 (unpublished).