Collision Matrix for (n,d) and (p,d) Reactions^{*}

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The contributions to (n,d), (p,d) reactions and their inverses from the pickup and stripping mechanisms are considered as corrections to the compound-nucleus or R-matrix theory of nuclear reactions. In an (n,d) reaction, for example, the *R*-matrix theory neglects the interaction of the incident neutron with the targetnucleus proton "tails" which extend beyond the nuclear radius. The pickup correction to the collision-matrix component, or reaction amplitude, appears as the matrix element of the neglected interaction involving an exact wave function and the approximate wave function of the compound-nucleus system not having the interaction; a distorted-wave Born approximation is used in which the former exact wave function is replaced by one of the latter type with the appropriate radiation condition. An explicit expression

I. INTRODUCTION

HE theories of deuteron stripping and formation by pickup, which were first formulated by Butler¹ and by Bhatia, Huang, Huby, and Newms² for applications at intermediate energies where it is possible to observe the angular distributions of the individual particle groups,³ have been the subject of numerous theoretical studies.⁴ However, it does not appear that any of these investigations have revealed how the compound-nucleus contribution to these deuteron reactions can be fitted into a general theory in a manner which is also useful for interpretations. The purpose of this note is to show formally how this can be done and to give an explicit form of the stripping or pickup contribution to the collision matrix which, together

butions at high energies where the resolution of the individual particle groups is not of concern. See J. R. Oppenheimer and M. Phillips, Phys. Rev. **48**, 500 (1935); H. A. Bethe, Phys. Rev. **53**, 39 (1938); R. Serber, Phys. Rev. **72**, 1008 (1947); D. C. Peaslee, Phys. Rev. **74**, 1001 (1948); G. F. Chew and M. L. Goldberger, Phys. Rev. **77**, 470 (1950). ⁴ P. B. Daitch and J. B. French, Phys. Rev. **87**, 900 (1952); R. Huby, Proc. Roy. Soc. (London) **A215**, 385 (1952); N. Austern, Phys. Rev. **89**, 318 (1953); E. Gerjuoy, Phys. Rev. **91**, 645 (1953); J. Horowitz and A. M. L. Messiah, Phys. Rev. **92**, 1326 (1953) and J. phys. et radium **14**, 695 (1953); N. C. Francis and K. M. Watson, Phys. Rev. **93**, 313 (1954); S. Yoshida, Progr. Theoret. and J. phys. et radium 14, 695 (1953); N. C. Francis and K. M. Watson, Phys. Rev. 93, 313 (1954); S. Yoshida, Progr. Theoret. Phys. (Japan) 10, 1, 370 (1953); Fujimoto, Hayakawa, and Nishijima, Progr. Theoret. Phys. (Japan) 10, 113 (1953); R. D. Dalitz, Proc. Phys. Soc. (London) A66, 28 (1953); M. Gell-Mann and M. L. Goldberger, Phys. Rev. 91, 398 (1953); E. Clementel, Nuovo cimento 11, 412 (1954); S. T. Butler and N. Austern, Phys. Rev. 93, 355 (1954); W. Tobocman, Phys. Rev. 94, 1655 (1954); J. Yocoz, Proc. Phys. Soc. (London) A67, 813 (1954); I. P. Grant, Proc. Phys. Soc. (London) A67, 981 (1954) and A68, 244 (1955). A review of the experimental material and of the 244 (1955). A review of the experimental material and of the theoretical work has been presented by R. Huby, Progr. Nuclear Phys. 3, 177 (1953).

is given for the collision-matrix component which, together with the compound-nucleus contribution, can be substituted directly into the formulas of Blatt and Biedenharn for total reaction cross sections and angular distributions. In general, the angular distributions contain interference terms in addition to the straight pickup and compound-nucleus contributions. If the distorted neutron and deuteron spherical partial waves are assumed to depend only on the angular momenta, and not explicitly on the total spin and the channel spins, the formula of Tobocman is obtained for the pickup contribution, while Butler's formula is obtained if plane waves are used instead of distorted waves. There are discussions of the various approximations, the exchange terms, and the question of the nuclear radius.

with the compound-nucleus contribution, can be substituted directly into the formulas of Blatt and Biedenharn⁵ for the total reaction cross sections and angular distributions. The present derivation of the stripping or pickup contribution is also simpler than some of the previous ones in that the desired collision-matrix components are obtained directly by an application of Green's theorem to the wave equations involved, rather than by introducing Green's functions, such functions not being necessary for the usual approximate procedures (plane-wave or distorted-wave Born approximations). With the present results it is possible, for example, to interpret a stripping reaction in the vicinity of an isolated resonance of the compound nucleus,⁶ which resonance may also contribute an appreciable number of reaction products, and it is possible to consider the distortion by the nucleus of the wave functions involved in the evaluation of the stripping contribution.⁷ The present formulation should also be applicable to a more detailed interpretation of other "surface" or "direct" interactions, such as the (n,p) and (n,n')reaction mechanisms proposed by Austern, Butler, and McManus.8

Wigner, Eisenbud, and Teichmann⁹ have developed a general theory of nuclear reactions, the R-matrix theory, which is particularly useful for considering reactions which proceed through one or two isolated

⁶ Berthelot, Cohen, Cotton, Faraggi, Grjebine, Levêque, Naggiar, Roclawski-Conjeaud, and Szteinsznaider, Compt. rend. 238, 1312 (1954); Holmgren, Blair, Simmons, Stratton, and Stuart, Phys. Rev. 95, 1544 (1954); Jones, McEllistrem, Douglas, Herring, and Silverstein, Phys. Rev. 98, 241(A) (1954).

⁷ Numerical evaluations of the effects of such distortions have been made by W. Tobocman and M. H. Kalos, Phys. Rev. 97, 132 (1955).

⁸ Austern, Butler, and McManus, Phys. Rev. **92**, 350 (1953). ⁹ E. P. Wigner and L. Eisenbud, Phys. Rev. **72**, 29 (1947); T. Teichmann and E. P. Wigner, Phys. Rev. **87**, 123 (1952). Additional references on the development of this theory are cited in these papers.

^{*} Work performed under the auspices of the U.S. Atomic Energy Commission. ¹S. T. Butler, Phys. Rev. 80, 1095 (1950); Proc. Roy. Soc.

⁽London) A208, 559 (1951); Phys. Rev. 88, 685 (1952)

Bhatia, Huang, Huby, and Newms, Phil. Mag. 43, 485 (1952). ³ The earlier work on deuteron reactions was primarily concerned with the excitation functions or with the angular distributions at high energies where the resolution of the individual

⁵ John M. Blatt and L. C. Biedenharn, Revs. Modern Phys. 24, 258 (1952).

levels of the compound nucleus. This theory is also readily applicable to the region of nuclear excitations where many levels overlap, provided that the signs of the reduced-width amplitudes $\gamma_{\lambda c}$, for the levels λ and channels c, are uncorrelated,¹⁰ such a lack of correlations presumably being the case if the interactions in the internal, or compound-nucleus region, are relatively strong. In this application the compound nucleus is found to manifest many of the reaction properties which have been well known from the work of Weisskopf and his collaborators.¹¹ The salient features of such compound-nucleus reactions are the symmetry of the angular distributions and the Maxwellian nature of the energy distributions. In contrast, the stripping and pickup angular distributions are generally peaked forward and the energy distributions are concentrated at the higher energies. These departures could of course be attributed to correlations of the signs of the $\gamma_{\lambda c}$. However, in an examination of the derivation of the *R*-matrix theory one notices that the derivation assumes that the nuclear interactions are confined to an internal or compound-nucleus region of the configuration space and thereby neglects, for example, the interaction of an incident neutron, while it is in the external neutron channel, with the "tail" of the wave function of a proton from the target nucleus, which tail extends to infinity though in an exponentially decaying manner. The present theories of pickup (or stripping) do indeed suggest that it is just such interactions which give rise to the deuterons in an (n,d) reaction. Since the total neutron cross sections are satisfactorily described by the compound-nucleus theory and since they are observed to be an order of magnitude larger than the total (n,d) reaction contribution, it seems reasonable to use the *R*-matrix theory for the interpretation of the interaction of the incident neutron with the bulk of the nucleus and then to correct for the external interaction with the proton by a first-order perturbation calculation using the compound-nucleus wave functions as first approximations. The collision-matrix component $U_{n;d}$ referring to an (n,d) reaction appears then as a sum $U_{n;d} = \mathfrak{U}_{n;d} + \Delta U_{n;d}$, where $\mathfrak{U}_{n;d}$ represents the compound-nucleus contribution and $\Delta U_{n;d}$ the external pickup correction term. The cross section therefore contains terms proportional to $|\mathfrak{U}_{n;d}|^2$ for the straight compound-nucleus contribution, to $|\Delta U_{n:d}|^2$ for the pickup contribution, and to interference terms of the form $\mathfrak{U}_{n;d}\Delta U_{n;d}^*+$ c.c. At nuclear excitations where the levels overlap, the interference terms will vanish if averaged over a sufficiently wide energy interval, provided that the signs of the $\gamma_{\lambda c}$ are uncorrelated.¹⁰ On

the other hand, these terms may be important in the vicinity of an isolated resonance level.

The pick-up term $\Delta U_{n;d}$ appears as the matrix element of the perturbing np interaction of the external region. This matrix element involves the wave function of the complete Hamiltonian which includes the perturbing interaction, and the wave function of the Hamiltonian with just the internal or compound-nucleus interactions. An approximation for its evaluation is the replacement of the former type wave function by one of the latter type with the same radiation condition, as in a Born approximation. Since the "distorted" waves of the compound nucleus are used rather than plane waves, this evaluation is expected to be more accurate than the usual "plane-wave" Born-approximation calculation. The distorted-wave method has been presented for general applications by Mott and Massey¹²; it has also been discussed and applied in various forms to stripping and pickup reactions by Horowitz and Messiah,⁴ by Francis and Watson,⁴ by Tobocman,⁴ and by Kalos and Tobocman.⁷ It was first noted by Francis and Watson,⁴ that at nuclear excitations where the levels overlap, the complex potential representations of the nucleonnucleus interaction^{10,13} may be useful for constructing the external wave functions which are involved in the correction matrix element.

II. DERIVATION BY MEANS OF GREEN'S THEOREM

The derivation proceeds by considering the two wave equations

$$H\Psi = E\Psi,$$
 (1a)

$$\mathcal{K}\Phi = E\Phi.$$
 (1b)

Here H is the complete Hamiltonian and $\mathcal{R}=H-\Delta V$ is the Hamiltonian for the system not having the interaction ΔV between the neutron and proton in the external region, that is, in the region where both are situated at distances from the center of the residual nucleus which are larger than the nuclear radius a; Ψ and Φ are the respective wave functions with the hereinafter specified radiation conditions. By application of Green's theorem to the integral over the configuration space τ of the difference of (1a) multiplied on the left by Φ^* and the complex conjugate of (1b) multiplied on the right by Ψ , one obtains¹⁴

$$\int_{\tau} \Phi^* \Delta V \Psi d\tau = \int_{S} \left(\hbar^2 / 2M_o \right) \times \left(\Psi \operatorname{grad}_n \Phi^* - \Phi^* \operatorname{grad}_n \Psi \right) dS, \quad (2)$$

¹⁰ R. G. Thomas, Phys. Rev. **97**, 224 (1955). ¹¹ V. F. Weisskopf, Phys. Rev. **52**, 295 (1937); V. F. Weisskopf and D. H. Ewing, Phys. Rev. **57**, 472, 935 (1940); H. Feshbach and V. F. Weisskopf, Phys. Rev. **76**, 1550 (1949); and, in particu-lar, J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, Inc., New York, 1952), Chaps. VIII and IX.

¹² N. F. Mott and H. S. W. Massey, The Theory of Atomic Collisions (Oxford University Press, London, 1949), Chap. VIII,

 ¹⁰ Store 5.
 ¹³ N. C. Francis and K. M. Watson, Phys. Rev. 92, 291 (1953);
 ¹³ N. C. Francis and Weisskopf, Phys. Rev. 96, 448 (1954);
 ¹⁴ G. Breit, Phys. Rev. 58, 1068 (1940); E. P. Wigner, Phys. Rev. 70, 15 (1946).

provided that the surface S is far enough out so that the nuclear systems can be regarded as decomposed into groups of two-particle subsystems of reduced masses M_c ; groups involving three or more subsystems are ignored.

For the consideration of an (n,d) reaction, the example to be used throughout, spherical waves of the following asymptotic form are substituted in (2):

$$\Psi_d = \sum_c (\delta_{cd} \mathcal{G}_c - U_{c;d} \mathcal{O}_c), \qquad (3a)$$

$$\Phi_n = \sum_c (\delta_{cn} \mathfrak{O}_c - \mathfrak{U}_{c;n}^* \mathfrak{G}_c), \qquad (3b)$$

where \mathcal{I}_c and \mathcal{O}_c are wave functions for the various channels c representing unit-flux incoming and outgoing waves, respectively; U and \mathfrak{u} are the collision matrices associated with the Hamiltonians H and \mathfrak{K} , respectively. The radiation condition imposed on the solution Ψ_d is that only the deuteron channel d has an *incoming* wave, while the condition imposed on Φ_n is that only the neutron channel *n* has an *outgoing* wave. The Φ_n of (3b) is thus a solution which is the time-reversed of the usual type, such as (3a), having an *incoming* wave in only one channel.^{12,15} The expansion of Φ_n is valid throughout the entire external part of the configuration space, whereas the expansion of Ψ_d is the asymptotic form which is valid only at distances far from the origin where even the interaction ΔV is negligible.

As in the work of Wigner and Eisenbud,⁹ \mathcal{I}_c and \mathcal{O}_c wave functions are constructed for each channel chaving a definite value of the channel spin j, which is a vector sum of the spins of the two particles of the group, and of the total spin J, which is a vector sum of j and the relative angular momentum $\hbar l$ of the two particles, and of the total spin component M along the beam direction; the channel subscript c thus refers to the set of quantities $\alpha j l J M$, where α denotes the nature of the pair and their respective states of excitation. These wave functions are therefore of the form

$$\mathcal{G}_{\alpha j l J M} = \sum_{\nu m} (j l \nu m | J M) \psi(\alpha j \nu) \times (i^{l} Y_{l m}(\Omega_{\alpha})) v_{\alpha}^{-\frac{1}{2}} r_{\alpha}^{-1} I_{\alpha l}(r_{\alpha}), \quad (4a)$$

$$\mathcal{O}_{\alpha j l J M} = \sum_{\nu m} (j l \nu m | J M) \psi(\alpha j \nu) \\ \times (i^{l} Y_{lm}(\Omega_{\alpha})) v_{\alpha}^{-\frac{1}{2}} r_{\alpha}^{-1} O_{\alpha l}(r_{\alpha}).$$
(4b)

Here $\psi(\alpha j\nu)$ is the sum of the products of the wave functions of the two members of the pair α , which sum has a definite channel spin j, component ν ; Y_{lm} is the spherical harmonic with the usual property Y_{lm}^* $=(-1)^m Y_{l-m}$; and v_{α} is the relative velocity.¹⁶ The ¹⁵ See K. M. Watson, Phys. Rev. **88**, 1163 (1952); G. Breit and H. A. Bethe, Phys. Rev. **93**, 888 (1954). ¹⁶ The behaviors under the time-reversal operation K of the

wave functions in (4) are

$$K\psi_{(\alpha j\nu)} = (-1)^{j-\nu}\psi_{(\alpha j-\nu)}, \quad K(i^{l}Y_{lm}) = (-1)^{l-m}(i^{l}Y_{l-m})$$

radial functions in (4) are taken to be

$$I_{\alpha l} = (G_{\alpha l} - iF_{\alpha l}) \exp(i\psi_{\alpha l}) \sim \exp[-i(\rho_{\alpha} - \eta_{\alpha}\log 2\rho_{\alpha} - \frac{1}{2}l\pi + \sigma_{\alpha 0})], \quad (5a) O_{\alpha l} = I_{\alpha l}^{*}, \quad (5b)$$

where $\sigma_{\alpha 0} = \arg \Gamma(1+i\eta_{\alpha})$ and $\psi_{\alpha l} = \sum_{s=1}^{l} \tan^{-1}(\eta_{\alpha}/s)$ are Coulomb phases, $\eta_{\alpha} = Z_{1\alpha} Z_{2\alpha} e^2 / \hbar v_{\alpha}$ is the Coulomb field parameter, and $\rho_{\alpha} = M_{\alpha} v_{\alpha} r_{\alpha} / \hbar = k_{\alpha} r_{\alpha}$; F and G are the usual regular and irregular external radial wave functions.

By substituting the expansions (3) into the right side of (2) with the surface S at infinity and by noting that the Wronskian $I(dO/d\rho) - O(dI/d\rho) = 2i$, one immediately finds that

$$\Delta U_{n;d} = U_{n;d} - \mathfrak{U}_{n;d} = (i/\hbar) \int_{\tau} \Phi_n^* \Delta V \Psi_d d\tau.$$
 (6)

The matrix element on the right gives the correction $\Delta U_{n;d}$ which is to be added to the compound-nucleus contribution $\mathfrak{U}_{n;d}$ in order to obtain the total collision matrix component $U_{n;d}$. As noted by Watson and by Breit and Bethe,¹⁵ the use of a wave function having an incoming wave in only one channel, rather than the Φ_n of (3b), would not give $\Delta U_{n;d}$, but a sum over channels c of the products of the components $U_{c;d}$ and U_{n; c}.

III. APPROXIMATIONS AND EVALUATIONS OF THE COMPONENT SUMS

As mentioned in the Introduction, the basic approximation of the present approach is the replacement of the exact wave function Ψ_d in (6) by a solution with the same radiation condition to the partial Hamiltonian 3°C. It is also assumed that the contributions to the matrix element (6) from the reaction channels of the Ψ_d and Φ_n solutions can be neglected; that is, these wave functions are approximated in the external region by

$$\Psi_d = \mathcal{J}_d - \mathfrak{U}_{d;d} \mathfrak{O}_d, \tag{7a}$$

$$\Phi_n = \mathfrak{O}_n - \mathfrak{U}_{n;n}^* \mathfrak{I}_n. \tag{7b}$$

The second approximation neglects, for example, the possibility that a neutron, which is inelastically scattered by a compound-nucleus process, could on emergence pick up a proton to form a deuteron.¹⁷ The inclusion of such contributions does not present any formal difficulties, but does make the final formulas more cumbersome. A third approximation is the neglect of the tensor np force which gives rise to the ³D component of the deuteron wave function; this neglect may be justified by a plane-wave, Born-approximation cal-

This representation was suggested by L. C. Biedenharn and M. E. Rose, Revs. Modern Phys. **25**, 736 (1953), and differs from the one used by Wigner and Eisenbud. As noted by R. Huby, Proc. Phys. Soc. (London) **A67**, 1103 (1954), the use of the present representation requires that the Z coefficients in Eq. (4.6) of Blatt and

Biedenham for the differential cross sections be replaced by the coefficients $\overline{Z}(l_1J_1l_2J_2; jL) = i^{l_1-l_2-L}Z(l_1J_1l_2J_2; jL)$. ¹⁷ According to calculations by B. H. Bransden, Proc. Phys. Soc. (London) A65, 738 (1952), such processes are likely to be important at energies above 200 Mev. See also W. N. Hess and B. J. Marry Dh. De 66 (2004) (1074) B. J. Moyer, Phys. Rev. 96, 859(A) (1954).

culation which shows that the contribution from this component is incoherent and therefore negligible compared with the ${}^{3}S$ contribution.¹⁸ As in the work of Horowitz and Messiah,⁴ it is assumed as a fourth approximation that the interaction ΔV between neutron and proton can be replaced by a zero-range (centralforce) interaction¹⁹; if $\chi(\mathbf{r})$ is the wave function for the relative motion of the neutron and proton in the ground state of the deuteron, then

$$\Delta V(\mathbf{r})\chi(\mathbf{r}) = -\left(\hbar^2/M\right)(8\pi\alpha)^{\frac{1}{2}}\delta(\mathbf{r}),\tag{8}$$

where $\alpha^{-1} = (\hbar^2/MB)^{\frac{1}{2}} = 4.4 \times 10^{-13}$ cm is the "radius" of the deuteron associated with its binding energy B, $\delta(\mathbf{r})$ is the three-dimensional delta function, and M is the nucleon mass.

The final bit of information needed for the evaluation of (6) is an expansion of the wave function of the target nucleus t in the external region of the configuration space where the contributions occur; that is, it is necessary to know the form of the proton "tails" which can be picked up. This wave function appears as one of the factors in the $\psi(\alpha i\nu)$ of (4) when the latter is used in (7b). The expansion is taken to be

$$\psi(tI_{t}i_{t}) = \sum (I_{f} \frac{1}{2} i_{f}i_{p} | j_{p} \nu_{p}) (j_{p} l_{p} \nu_{p} m_{p} | I_{t}i_{t})$$

$$\times \psi(\alpha_{f} I_{f}i_{f}) \psi(p \frac{1}{2} i_{p}) u(\alpha_{f} j_{p} l_{p}; r_{p}) r_{p}^{-1}$$

$$\times \lceil i^{l_{p}} Y_{l_{p} m_{p}}(\Omega_{p}) \rceil, \quad (9)$$

the sum being with respect to $\alpha_f j_p \nu_p l_p m_p i_f i_p$. Here I_t , i_t and I_f , i_f are the spins and components of the states of the target and final nucleus, respectively; $\psi(\alpha_f I_f i_f)$ is the wave function of the final nucleus in the excitation state α_t and $\psi(p \cdot \frac{1}{2} i_p)$ is the wave function representing the pickedup proton in the spin state i_p ; l_p and m_p designate the orbital angular momenta and components of the proton tails; the channel spins j_p are the vector sums of the proton spin $\frac{1}{2}$ and I_f , and their components are ν_p ; the $(i^{l_p} Y_{l_p m_p})$ are the angular parts of the proton wave functions and the $u(r_p)$ are r_p -times the radial parts, the latter being considered as real because they may be defined as scalar products.²⁰ It is convenient to express the radial parts as

$$\begin{aligned} u(\alpha_{f}j_{p}l_{p};r_{p}) &= \gamma(\alpha_{f}j_{p}l_{p})(2M\epsilon a/\hbar^{2})^{\frac{1}{2}} \\ \times \left[W(-\eta_{p},l_{p}+\frac{1}{2};2k_{p}r_{p}) / \\ W(-\eta_{p},l_{p}+\frac{1}{2};2k_{p}a) \right]. \end{aligned}$$
(10)

Here $\gamma(\alpha_f j_p l_p)$ is the reduced-width amplitude for the separation in the external region of the ground state of the target nucleus into a proton and a state α_f of the final nucleus; M is the nucleon mass; $W(-\eta, l+\frac{1}{2}; 2kr)$ is the exponentially decaying Whittaker function; $k_p = (2M\epsilon B_{pf}/\hbar^2)^{\frac{1}{2}}$ is the wave number associated with the binding energy B_{pf} of the proton in the ground state of the target to the state α_f of the final nucleus; $\eta_p = Z_f e^2 M \epsilon / \hbar^2 k_p$ is the corresponding Coulomb field parameter; $\epsilon = A_t - 1/A_t$ is the ratio of the masses of the final and target nuclei.²¹

With the above information and approximations the matrix element (6) can readily be evaluated, the methods of Racah²² being used to perform the sums over the various spin components. The result is that

$$\Delta U(nj_{n}l_{n}; dj_{d}l_{d}; J)$$

$$= 2if(-1)^{I_{f}-I_{t}+J}(2I_{t}+1)^{\frac{1}{2}}\sum_{j_{p}l_{p}}(-1)^{l_{p}-j_{p}}(2j_{p}+1)^{\frac{1}{2}}$$

$$\times \gamma(j_{p}l_{p})W(\frac{1}{2}1j_{p}j_{d};\frac{1}{2}I_{f})W(I_{l}l_{p}\frac{1}{2}j_{d};j_{p}j_{n})$$

$$\times \bar{Z}(l_{n}j_{n}l_{d}j_{d};Jl_{p})R(j_{n}l_{n},j_{d}l_{d};l_{p}J), \quad (11)$$
where
$$f = (3\alpha a/M\epsilon v_{n}v_{d})^{\frac{1}{2}},$$

and the (dimensionless) radial integral is

$$R(j_{n}l_{n}, j_{d}l_{d}; l_{p}J)$$

$$=i^{l_{p}-l_{n}+l_{d}} \int_{a}^{\infty} \frac{dr}{r} \cdot \frac{W(-\eta_{p}, l_{p}+\frac{1}{2}; 2k_{p}r)}{W(-\eta_{p}, l_{p}+\frac{1}{2}; 2k_{p}a)}$$

$$\times [Il_{n}(\epsilon r) - \mathfrak{U}(nj_{n}l_{n}; nj_{n}l_{n}; J)Ol_{n}(\epsilon r)]$$

$$\times [Il_{d}(r) - \mathfrak{U}(dj_{d}l_{d}; dj_{d}l_{d}; J)Ol_{d}(r)]; \quad (11a)$$

W is the Racah coefficient and the relation between the $ar{Z}$ coefficient and the Z coefficient of Blatt and Biedenharn is given in reference 16. The angular distributions may be obtained by substitution of these collisionmatrix components, together with those for the compound-nucleus contribution, into the formulas given by Blatt and Biedenharn,⁵ and polarizations may be deduced from the formulas of Simon and Welton.23 Since ΔU is symmetrical, this result applies also to (d,n) reactions; the modifications required for (p,d)and (d, p) reactions should be obvious.

The presence of the Racah and \bar{Z} coefficients in (11) implies that there are selection rules. In addition to the

¹⁸ R. D. Dalitz, reference 4; R. G. Thomas (unpublished). ¹⁹ See G. Breit, Phys. Rev. 71, 215 (1947) where the validity of such an approximation is investigated in detail in connection with the problem of the interaction of slow neutrons with bound ²⁰ See Eq. (4) of Wigner and Eisenbud, reference 9.

²¹ The dimension of the reduced-width amplitude used here is the square root of energy (see Sec. II of reference 10); it is normalized with respect to all configuration space, rather than just the internal region as in the resonance applications, the difference which is involved in a (p,d) reaction, $\eta_p = 0$ and $W(0, l + \frac{1}{2}; 2s) = (2z/\pi)^{\frac{1}{2}} K_{l+\frac{1}{2}}(z)$, where $K_{l+\frac{1}{2}}(z)$ is the modified Bessel function [see Sec. II-B of R. G. Thomas, Phys. Rev. 88, 1109 (1952)].

²² Biedenharn, Blatt, and Rose, Revs. Modern Phys. 24, 249 (1952).

²³ A. Simon and T. A. Welton, Phys. Rev. **90**, 1036 (1953); Albert Simon, Phys. Rev. **92**, 1050 (1953). The remarks in footnote 16 concerning the modification in the convention for time reversal To concerning the modification in the convention for time reversal also apply to these papers; the *i* factors involving angular mo-mentum exponents should be deleted and the Z coefficients replaced by \overline{Z} coefficients. Polarizations in stripping and pickup processes have been investigated by W. B. Cheston, Phys. Rev. 96, 1590 (1954), by J. Horowitz and A. M. L. Messiah, J. phys. radium 14, 731 (1953), and by H. C. Newms, Proc. Phys. Soc. (London) A66, 477 (1953).

expected ones: $\mathbf{j}_n + \mathbf{l}_n = \mathbf{J}$; $\mathbf{j}_d + \mathbf{l}_d = \mathbf{J}$; $\mathbf{j}_2 + \mathbf{l}_2 = \mathbf{1}$; $\mathbf{l}_2 + \mathbf{I}_f = \mathbf{j}_p$; $\mathbf{l} + \mathbf{I}_f = \mathbf{j}_d$; $\mathbf{l}_2 + \mathbf{I}_t = \mathbf{j}_n$; $\mathbf{l}_p + \mathbf{j}_p = \mathbf{I}_t$; there are also the relations $\mathbf{j}_n + \mathbf{j}_d = \mathbf{l}_p$; $\mathbf{l}_2 + \mathbf{j}_d = \mathbf{j}_p$; $\mathbf{l}_n + \mathbf{l}_d = \mathbf{l}_p$. There is also the parity rule $l_n + l_d + l_p = \text{even}$.

At intermediate and high energies where many partial waves l_n and l_d contribute to the pickup process, the cross section evaluations will in general be very tedious. Fortunately, some simplification is possible if it is assumed that the diagonal collision-matrix components in the radial integrals of (11) are independent of the total spin and of the channel spins, in which case the radial integrals depend only on l_n , l_d , and l_p . This simplification is quite reasonable for applications at excitation energies where the levels overlap. Indeed, at such excitations it is the practice to represent the neutron-nucleus or deuteron-nucleus interactions by models which depend on only the angular momentum, such as the strong-coupling or complex potential models (without a spin-orbit coupling dependence). With this simplification the sums over the total spin J and the channel spins j_n , j_d in the formulas of Blatt and Biedenharn can be evaluated using the unitary properties of the transformation coefficients and the Racah sum rules.²² One finds that the pickup contribution to the (n,d) differential cross sections is given by

$$\begin{bmatrix} d\sigma_{n;d}(\theta_d)/d\Omega_d \end{bmatrix}_{\text{pickup}}$$
$$= \frac{1}{4}k_n^{-2}f^2 \sum_l (2l_p+1)^{-2}\gamma(l_p)^2 Q(l_p), \quad (12)$$

where

$$\gamma(l_p)^2 = \sum_{jp} \gamma(j_p l_p)^2$$

and $Q(l_p) = \sum_{m=-l_d}^{l_d} \left| \sum_{l_n l_d} (2l_n + 1)(2l_d + 1) \left[\frac{(l_d - |m|)!}{(l_d + |m|)!} \right]^{\frac{1}{2}}$

 $\times (l_n l_d 00 | l_p 0) (l_n l_d 0m | l_p m)$

 $\times R(l_n, l_d; l_p) P_{l_d}|^{m|}(\cos\theta_d)|^2,$

$$= (-1)^{l_p} (2l_p + 1) \sum_{L} (-1)^{L} P_L(\cos\theta_d)$$

$$\times \sum_{l_n l_n' l_d l_d'} (2l_n + 1) (2l_n' + 1) (2l_d + 1) (2l_d' + 1)$$

$$\times (l_n l_d 00 | l_p 0) (l_n' l_d' 00 | l_p 0) (l_n l_n' 00 | L 0)$$

$$\times (l_d l_d' 00 | L 0) W (l_n l_n' l_d l_d'; L l_p)$$

$$\times R(l_n, l_d; l_p) R(l_n', l_d'; l_p)^*.$$

Apart from a slight difference entering the factor f, this cross-section formula with the first alternative expression for the quantity $Q(l_p)$ is the same as the one derived by Tobocman.^{4,7} The second alternative form of $Q(l_p)$ is obtained by forming the absolute square in the first and carrying out the sum over the deuteron angular momentum component m; for numerical work

it is probably not as useful as the first. Note that the incoherence of the various contributing j_p and l_p terms is a consequence of the simplification.

The compound-nucleus contribution to the differential cross section can be obtained directly from the formulas of Blatt and Biedenharn. The interference terms can also be simplified if the pickup radial integrals are again assumed to be independent of J. By carrying out the J sum in the pickup factor, one finds that for a single compound-nucleus collision matrix component $\mathfrak{U}(nj_n'l_n'; dj_d'l_d'; J')$ these terms are given by

$$\left[d\sigma_{n;d}(\theta_d)/d\Omega_d \right]_{\text{interference}}$$

$$= -\frac{1}{4}k_{n}^{-2i}f(2I_{t}+1)^{-\frac{1}{2}}(-1)^{I_{f}+I_{t}+J'}(2J'+1)$$

$$\times \sum_{L}(-1)^{L}P_{L}(\cos\theta_{d}) \sum_{i_{p}l_{p}} (-1)^{i_{p}}(2j_{p}+1)^{\frac{1}{2}}$$

$$\times \gamma(j_{p}l_{p}) \sum_{l_{n}l_{d}} [(2l_{n}+1)(2l_{d}+1)/(2l_{n}'+1)(2l_{d}'+1)]^{\frac{1}{2}}$$

$$\times (l_{d}l_{d}'00|L0)(l_{n}l_{d}00|l_{p}0)(l_{n}'l_{d}'00|l_{p}0)^{-1}$$

$$\times \overline{Z}(l_{n}'j_{n}'l_{d}'j_{d}';J'l_{p})\overline{Z}(l_{n}l_{d}l_{n}'l_{d}';l_{p}L)$$

$$\times W(\frac{1}{2}1j_{p}j_{d}';\frac{1}{2}I_{f})W(I_{d}l_{p}\frac{1}{2}j_{d}';j_{p}j_{n}')$$

$$\times R(l_{n},l_{d};l_{p})^{*}\mathfrak{U}(nj_{n}'l_{n}'j_{d}';d'_{d}';J')+\text{c.c.} (13)$$

One-level formulas for the collision-matrix componenst have been given by Wigner and Eisenbud⁹ and by Blatt and Biedenharn.⁵

Much simpler expressions are obtained for the straight pickup and the interference contributions if the plane-wave Born approximation is used rather than the distorted-wave Born approximation. These expressions are given in the Appendix.

IV. DISCUSSIONS

The reader may have noticed that the above development contains a flaw in the assertion that the *R*-matrix theory can be used to obtain the compound-nucleus contribution to the collision matrix component $U_{n;d}$ for a nuclear system not having the interaction ΔV between the neutron and proton when they are in the external region. For if ΔV were completely neglected in the *R*-matrix formalism the deuterons which appear at the nuclear surface would cease to interact there and consequently would appear at infinity as free neutrons and protons. This contribution to $U_{n;d}$ would thus vanish, and the "compound-nucleus" deuterons which appear at the nuclear surface would contribute instead to the component $U_{n;np}$. This difficulty may be resolved by noting that the effect of the interaction ΔV is not only to give the $\Delta U_{n;d}$ contribution of (6) but also to convert the above compound-nucleus component $U_{n;np}$ to a compound-nucleus component $U_{n;d}$, as implied in the development. It is evident then that the derivation of the *R*-matrix theory does not entirely neglect ΔV but only that contribution from it which was used to calculate $\Delta U_{n;d}$.

There is also an exchange contribution to $U_{n;d}$ from the interactions of the external region. This contribution arises from additional terms in the expansion (9) corresponding to the dissociation of the target nucleus in the external region into a deuteron and a core of $A_t - 2$ particles. The perturbing interactions are those between the incident neutron n_1 and both the neutron n_2 and the proton ϕ of the deuteron when the deuteron is in the external region. As a result of these interactions the deuteron can be ejected and the recoiling neutron n_1 captured by the core. (The incident neutron could also pick up the core, forming the residual nucleus and leaving the deuteron behind.) A similar mechanism was considered by Austern, Butler, and McManus⁸ for the contribution to (n,p) reactions from the external region, in which the incident neutron ejects a proton from the target and the recoiling neutron is subsequently captured by the core. Because of the fact that in the (n,d)exchange calculation there are essentially four interacting particles involved (the core and the nucleons n_1, n_2, p), rather than just the three particles of the direct term, the exchange term is considerably more difficult to compute. Fortunately, it may be negligible for a number of reasons: First, and perhaps the most significant, is that the reduced width (or probability) for the dissociation of the target into a deuteron and a core may be expected to be small compared with the proton reduced width since two particles are involved. Secondly, the radial wave function for the dissociation, which is involved in a "radial" integral similar to (11a), will decay rather more rapidly than a proton function, which is involved in the direct term, because deuteron binding energies are generally larger than proton binding energies and because a deuteron has twice the mass. Finally, the "radial" integral will also be relatively smaller because it involves two, rather than just one, exponentially decreasing function and because the momenta transfers involved are relatively larger.

In the calculation of the pickup contribution we have neglected the contribution from the interaction between the incident neutron and the core which takes place when the proton of the target and the core are separated by distances greater than the channel radius. One can associate with this interaction a mechanism in which the incident neutron ejects the core with the result that the freed proton can combine with the neutron to form a deuteron. This mechanism is essentially the same as the above-mentioned one considered by Austern et al.8 However, there is an important difference in detail in that in the (n,p) case the ejected particle is a light one whereas in the (n,d) case it is a heavy one, that is, the roles of the proton and core are reversed; and it is this difference which leads one to believe that although the former mechanism is apt to be important, the latter is

not. Unfortunately, we cannot give a really satisfactory justification for this remark, although the impulse type of approximation, such as the one used by Austern et al., suggests that in an (n,d) reaction, the core ejection mechanism is apt to be less important than the pickup mechanism. First, the radial integral of the former involves two, rather than just one, exponentially decaying radial function and, secondly, the momentum transfer involved in producing forward deuterons is much larger in the ejection process than in the pickup process. In fact, according to such a calculation, the deuterons from the ejection process are concentrated in the backwards directions (as one would expect for a process which ejects the core in the forward directions). rather than in the forward directions, as in the pickup process.

It is well to emphasize that although the present approach utilizes distorted partial waves in the matrix elements, it is still a Born approximation evaluation and subject to the limitations of that approximation. In particular, the absolute magnitude of the calculated collision-matrix component $U_{n;d}$ of (11) can exceed unity in certain circumstances, thus violating the unitarity or flux-conservation requirement. If the waves involved in the radial integrals are not distorted by a sharp resonance, it is not likely that the absolute magnitude of those integrals will exceed unity, and it would then be sufficient to require that the factor $f\gamma(j_p l_p)$ of (11) be less than unity. Since the factor αa of f is of the order unity, this requirement is essentially that the geometric mean of the neutron and deuteron energies be greater than the reduced widths $\gamma(j_p l_p)^2$, these having an upper limit of a few Mev according to the sum rule. On the other hand, near a sharp resonance, the absolute value of the radial integral can become very large compared with unity because the irregular functions G, which are generated by the resonance, would be large. However, in such a case it is probably not necessary to include an external contribution to $U_{n;d}$ because the internal contribution dominates and, anyway, the channel radii for the resonant partial waves could be taken large enough to include in the "internal" region most of the "external" interaction so that there is effectively no "external" contribution to $U_{n;d}$ for those partial waves. The radial integrals could also be large if the binding and barrier of the picked-up proton are very small; the Whittaker function would then decay very slowly and contributions to the integral could arise from a large radial interval. The Born approximation will also be invalid if there is appreciable polarization of the deuteron by the Coulomb field, as expected at low deuteron energies.

Finally, it is appropriate to remark on the important matter of what value of the nuclear radius should be used in the external calculation. In order that the above Born approximation have some validity, it is clear that the radius should not be too small. On the

other hand, for the applicability, in the case of overlapping levels, of the compound-nucleus theory to the internal region, it is necessary that the radius be small enough for the signs of the reduced-width amplitudes to be uncorrelated, thus indicating that the interactions of that region are strong right up to the surface. In other words, the interactions of the internal region must be strong enough so that the multiple processes dominate, whereas those of the external region must be sufficiently weak so that multiple processes can be ignored (Born approximation). Clearly these requirements are not compatible with a unique boundary, and there is probably an intermediate region, or shell, which is not properly accounted for by either the compoundnucleus calculation or by the external perturbation calculation. Although there may be no simple way of dealing with this region, the observed near equality of the nuclear radii inferred from total neutron cross sections and stripping processes²⁴ is an indication that it may be small or not very important.

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APPENDIX

The results of the text are, of course, much simpler if undistorted rather than distorted waves are used, that is, if the $\mathfrak{U}_{n;n}$ and $\mathfrak{U}_{d;d}$ of (7) are assumed to be unity, and if, in addition, the Coulomb fields are neglected. With these simplifications the various neutron and deuteron partial waves combine to form a simple exponential function, and the radial integrals corresponding to (11a) become simply the Fourier transforms of the radial functions $u(\alpha_f j_p l_p; r_p)$ of (9). By application of the wave equation and by an integration by parts, these transforms may be expressed as wronskians evaluated at r=a. Furthermore, if the compoundnucleus contribution is neglected, the cross section formula reduces to the relatively simple one which was originally given by Butler. Although these approximate methods do appear to give fairly accurate predictions of the angular distributions in circumstances where the Coulomb barriers are not strong, they are found to predict absolute yields that are much too large.^{7,25}

In the plane-wave Born approximation, the pickup contributions ΔA to the (n,d) reaction amplitudes $A = \alpha + \Delta A$ for initial and final channel spin states j_{n}, ν_n and j_d, ν_d , respectively, are

$$\begin{split} \Delta A (nj_{n}\nu_{n}; dj_{d}\nu_{d}) \\ &= -4\pi^{\frac{1}{2}}fk_{d}(2j_{n}+1)^{\frac{1}{2}}(2j_{d}+1)^{\frac{1}{2}}(2I_{t}+1)^{\frac{1}{2}} \\ &\times (-1)^{I_{f}-I_{t}-j_{d}+j_{n}+\nu_{n}}(\alpha^{2}+k_{pn}^{2})^{-1} \\ &\times \sum_{j_{p}l_{p}m_{p}} (-1)^{m_{p}-j_{p}}(2j_{p}+1)^{\frac{1}{2}}(2l_{p}+1)^{-\frac{1}{2}} \\ &\times W(I_{t}l_{p}\frac{1}{2}j_{d}; j_{p}j_{n})W(\frac{1}{2}1j_{p}j_{d}; \frac{1}{2}I_{f}) \\ &\times (j_{n}j_{d}\nu_{n}-\nu_{d}|l_{p}m_{p})\gamma(j_{p}l_{p})^{c} \psi_{l_{p}}V_{l_{p}m_{p}}(\Theta,\Phi). \end{split}$$
(14)

In addition to the quantities already defined in connection with (11), there are the following: The characteristic stripping momentum transfer wave numbers are $\mathbf{k}_{pn} = \mathbf{k}_n - \frac{1}{2}\mathbf{k}_d$ and $\mathbf{k}_{pf} = \mathbf{k}_d - \epsilon \mathbf{k}_n$, so that

$$k_{pn}^2 = k_n^2 + \frac{1}{4}k_d^2 - k_nk_d\cos\theta_d,$$
$$k_{pf}^2 = \epsilon^2 k_n^2 + k_d^2 - 2\epsilon k_nk_d\cos\theta_d;$$

 Θ is the angle between the direction of \mathbf{k}_{pf} and the incident beam and may be obtained from the relation $\sin \Theta = (k_d/k_{pf}) \sin \theta_d$. The Wronskian is

$$W_l = F_{l-1}(x) - (l+\Lambda)x^{-1}F_l(x),$$

where $x = k_{pl}a$, and Λ is the radius *a* times the logarithmic derivative at *a* of *r* times the radial part of the wave function of the proton bound to the target nucleus; when the Coulomb field is neglected, the expressions for $(l+\Lambda)$ in terms of $z=k_pa$ for the proton angular momenta *l* are



Butler's formula for straight pickup is obtained by summing the absolute square of the amplitudes (14) over all final channel spin states and averaging over all initial ones; it is

$$\left[d\sigma_{n;d}(\theta_d) / d\Omega_d \right]_{\text{pickup}} = k_d^2 f^2 (k_{pn}^2 + \alpha^2)^{-2}$$

$$\times \sum_{lp} \gamma(l_p)^{2\pi} W_p^2, \quad (15)$$

²⁴ J. R. Holt and T. N. Marsham, Proc. Phys. Soc. (London) **A66**, 1032 (1953). These authors show that the stripping radii are actually larger, as one would expect.

A00, 1052 (1953). These authors show that the schepping radiu are actually larger, as one would expect. ²⁵ R. G. Thomas, Phys. Rev. **91**, 453(A) (1953); Horowitz and Messiah, reference 4; Fujimoto, Kikuchi, and Yoshida, Progr. Theoret. Phys. (Japan) **11**, 264 (1954); G. Abraham, Proc. Phys. Soc. (London) **A67**, 273 (1954). The proportionality of stripping and pickup cross sections to the reduced widths $\gamma(l_p)^2$, which is also evident in (12), is brought out in these papers; see also A. M. Lane and D. H. Wilkinson, Phys. Rev. **97**, 1199 (1955).

Note that the angular dependent factors $Y_{l_pm_p}(\Theta, \Phi)$ in the amplitude expressions do not appear in (15).

The contributions to the reaction amplitudes from a particular collision-matrix component

$$\mathfrak{U}(nj_n'l_n';dj_d'l_d';J')$$

are given by

$$\begin{aligned} &\alpha(nj_{n}'\nu_{n}';dj_{d}'\nu_{d}') \\ &= -i\pi^{\frac{1}{2}}k_{n}^{-1}(2l_{n}'+1)^{\frac{1}{2}}(j_{n}'l_{n}'\nu_{n}'0|J'\nu_{n}') \\ &\times (j_{d}'l_{d}'\nu_{d}'\nu_{n}'-\nu_{d}'|J'\nu_{n}') \\ &\times \mathfrak{U}(nj_{n}'l_{n}';dj_{d}'l_{d}';J')Y_{l_{d}'\nu_{n}'-\nu_{d}'}(\Omega_{d}). \end{aligned}$$
(16)

By summing the absolute squares of the sum of terms from (16) and (14) over all possible ν_n' , ν_d' and by dividing by the number of initial spin states, one obtains in addition to the straight pickup and resonance contributions, the interference contribution to the differential cross section which is

$$\lfloor d\sigma_{n;d}(\theta_d)/d\Omega_d \rfloor$$
interference

$$= \frac{1}{2} i f k_d k_n^{-1} (-1)^{I_f + I_t + J'} (\alpha^2 + k_{pn}^2)^{-1} (2J' + 1) \\ \times (2I_t + 1)^{-\frac{1}{2}} \mathfrak{U}(nj_n'l_n'; dj_d'l_d'; J') \sum_{j_p l_p} (-1)^{j_p} \\ \times (2j_p + 1)^{\frac{1}{2}} \gamma(j_p l_p)^{\mathfrak{W}} l_p W(I_t l_p \frac{1}{2} j_d'; j_p j_n') \\ \times W(\frac{1}{2} 1 j_p j_d'; \frac{1}{2} I_f) \bar{Z}(l_n'j_n'l_d'j_d'; J'l_p) \\ \times \sum_m [(l_n'l_d' 0m | l_pm)/(l_n'l_d' 00 | l_p 0)] \\ \times [(l_d' - |m|)! (l_p - |m|)!/(l_d' + |m|)! (l_p + |m|)!]^{\frac{1}{2}} \\ \times P_{l_d'} [\mathfrak{m}|(\theta_d) P_{l_p}]^{\mathfrak{m}|}(\Theta) + \text{c.c.} \quad (17)$$

The *m* sum extends from $-l_p$ to l_p , or from $-l_d'$ to l_d' , whichever range is smaller.

The pickup reaction amplitude (14) was calculated for the deuteron ¹S wave function of the zero-range potential. For a Chew-type wave function of the form $\exp(-\alpha r) - \exp(-\beta r)$, where $\beta \approx 7\alpha$, (14), (16), and (17) should be multiplied by $\beta^{\frac{1}{2}}(\alpha + \beta)^{\frac{3}{2}}/(\beta^2 + k_{pn}^2)$, and (15) should be multiplied by its square.

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Reactions $C^{12}(d,n)N^{13}_{\text{ground state}}$ and $C^{12}(d,t)C^{11}$ up to $E_d = 20 \text{ Mev}^*$

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The course of the partial cross section for the direct formation of N¹³ in its ground state only in the reaction $C^{12}(d,n)N^{13}$ has been followed up to $E_d=20$ Mev by observing the N¹³ activity induced in a stack of polyethylene foils (only the ground state is stable against proton emission). The cross section falls appreciably less rapidly than would be expected for compound nucleus formation and also less rapidly than predicted by simple stripping theory. The cross section at $E_d=8$ Mev is 100 mb which is not so large relative to that for the mirror (d, p) reaction as is predicted by simple stripping theory. The reaction $C^{12}(d,t)C^{11}$ has been detected and its total cross section measured from its threshold $(E_d=14.5 \text{ Mev})$ to $E_d=20$ Mev where it is 10 mb. The magnitude of this cross section indicates that this is a pickup reaction.

INTRODUCTION

THE many measurements of "stripping" angular distributions in recent years have given ample grounds for believing that (d,p) and (d,n) reactions induced by deuterons of energy greater than three or four Mev do not as a rule involve the strong formation of a compound nucleus but that the reactions proceed most frequently by a direct interaction in which a nucleon of the impinging particle simply severs its "deuteron bond" at the nuclear surface and attaches itself to the existing (and undisturbed) structure of the target nucleus thereby forming one or other of those states of the residual nucleus of which the target nucleus is a parent.¹ Although fair to good qualitative agreement between experimental and theoretical stripping patterns can usually be obtained (albeit by a somewhat

cavalier approach to the problem of the nuclear radius), it is abundantly clear that neither the details of the patterns predicted by simple stripping theory nor the theoretical absolute cross sections are reproduced by experiment and that considerable refinements to the theory are needed. Examples of such refinements are the taking account of Coulomb effects, scattering of the incident deuteron wave, exchange effects, and boundary conditions for the outgoing particle.² As soon as such refinements are introduced the fit with experiment may, of course, be much improved since it is not usually clear which of many alternative procedures should be followed at each stage, and advantage may be taken of this uncertainty. Thus, although some empirical working recipe may emerge for the fitting of experimental angular distributions and reduced widths, we cannot feel confident that the particular constellation of param-

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¹ On leave from Cavendish Laboratory, Cambridge, England. ¹ S. T. Butler, Proc. Roy. Soc. (London) **A208**, 559 (1951).

² See, for example, W. Tobocman and M. H. Kalos, Phys. Rev. **97**, 132 (1955).