Theoretical Considerations Concerning the $D+D$ Reactions

E. J. KONOPINSKI, *Indiana University, Bloomington, Indiana*

AND

E. TELLER, *University of Chicago, Chicago, Illinois*

(Received January 12, 1948)

Some possibilities are explored for the theoretical explanation of the angular distribution of $D+D$ reaction products. The variation with energy is ascribed entirely to differences in the centrifugal barriers encountered by the bombarding particles responsible for the isotropic and non-isotropic components. The detailed data on the proton distributions at low energies appear to be explained in terms of an asymmetric part produced by P-waves superposed on isotropic emissions caused by S-waves. However, ordinary extrapolation of this to higher energies gives much less symmetric distributions than experimentally found at such energies for the emission of neutrons. If the experiments are correct as interpreted, then they appear to show that spin-orbit coupling plays a large part in the reactions. In that case, an isotropic component produced by incoming P-waves grows in importance with energy and accounts for the increasingly parallel growth of the isotropic and asymmetric emissions.

1. INTRODUCTION

THEORETICAL treatments of the reactions of deuterons with deuterons have been offered by Dolch,[1] Schiff,[2] Flügge,[3] and Myers.[4]

The first three authors made attempts to to derive an absolute value for the reaction cross section from the magnitude of the nuclear forces. Flügge improved on the others in that he included the effect of the nuclear forces on the relative motion of the two deuterons (a Hartree-Fock approximation), instead of assuming that motion to be influenced only by the electrostatic repulsion (a modified Born approximation). He succeeded in obtaining the right order of magnitude (cross section $\sim 10^{-26}$ cm^2 at ~ 150 kev). Difficulties of calculation caused Flügge to consider only the production of particles without relative orbital angular momentum. With the type of nuclear forces assumed, such emissions can arise only from singlet collisions. This resulted in the introduction of a weight factor 1/9, representing the fraction of the collisions in which the deuterons come together in a singlet state.

Schiff, using the modified Born approximation, was able to consider particles with non-vanishing angular momenta. This made possible a discussion of the angular distribution of the product particles. No spin changes during the reaction were permitted by the types of nuclear forces assumed, these types being the same as those considered by Dolch and Flügge.

Myers made a tentative application of the Breit-Wigner resonance theory. He argues that, in general, symmetry of the angular distribution with respect to a plane normal to the direction of incidence indicates the formation of a compound nucleus. This argument is not binding here, where such a symmetry follows from the identity of the collision partners.

None of the work mentioned discusses the *energy-dependence* of the asymmetry in the angular distribution. This is the primary object of the present paper.

The experiments give for the angular distribution of the product protons the form:

$$1+A(E)\cos^2\theta, \tag{1}$$

where θ is the angle of emission relative to the incident deuteron beam, in the center of mass system (CMS), and $A(E)$ is the "asymmetry coefficient," which depends on the deuteron bombarding energy, E. Extensive measurements of $A(E)$ for protons were carried out at New York University,[5] for E between 90 kev and 250 kev. Bretscher, French, and Seidl[6] supplemented

[1] H. Dolch, Zeits. f. Physik **100**, 401 (1936).
[2] L. T. Schiff, Phys. Rev. **51**, 783 (1937).
[3] S. Flügge, Zeits. f. Physik **108**, 545 (1938).
[4] R. D. Myers, Phys. Rev. **54**, 361 (1938).
[5] H. P. Manning, R. D. Huntoon, F. Myers, and V. Young, Phys. Rev. **61**, 371 (1942); also R. D. Huntoon, A. Ellett, D. S. Bayley, and J. Van Allen, Phys. Rev. **58**, 97 (1940).
[6] E. Bretscher, A. French, and F. Seidl, Phys. Rev. **73**, (1948).

these with points between $E=20$ kev and 80 kev. Both sets of data are represented by the experimental points in Fig. 1. Bennett, Mandeville, and Richards[7] made measurements of the relative dependence of A on energy for neutrons. This was for E in the range from 0.5 Mev to 2 Mev. Their results are shown in Fig. 2, in which the data already in Fig. 1 are included for comparison.

In exploring the possibilities for explaining the results for A, it is helpful to consider also the integral cross section, including all angles. Experimental data[6,8] on this are represented by the points in Fig. 3. Actually plotted is the quantity $\sigma E/(1+\frac{1}{3}A)$ where σ is the total proton production cross section in barns (1 barn $=10^{-24}$ cm^2) and E is the energy in kev. The abscissa of Fig. 3 is $E^{-\frac{1}{2}}$, with E expressed in Mev.

The present theoretical treatment proceeds on the assumption that the relative penetrability of the electrostatic and centrifugal force barriers can entirely account for the energy dependence of the asymmetry coefficient, A. Thus, no effort will be made to find absolute magnitudes dependent on the interactions between particles after their penetration within the barriers. Usually, the only strong variation with energy which results from the specifically nuclear interactions within the barriers is due to resonance effects. The observed[6,8] dependence of yield on energy shows no resonance in the D$-$D process, at least in the energy range of interest here (in which the reliable measurements of A have been made). Moreover, Flügge seems to have shown that a "first-order," non-resonance reaction should have the observed order of magnitude.

In treating the penetration of the barriers, one may consider using the precise solutions of the Schrödinger equation with a Coulomb potential. To justify such precision, care would have to be taken with the fitting of the "regular" and "irregular" solutions at the nuclear surface. The conditions for such fitting are at least as obscure as the current status of the nuclear forces. We shall be content with the usual (WKB) approximations for the penetration probabilities.

2. THE PENETRABILITY

The distance R to which two deuterons may approach before nuclear forces take hold will be given the value:

$$R=7\times 10^{-13}\text{ cm.} \tag{2}$$

This is compounded from a deuteron diameter ($\sim 4.2\times 10^{-13}$ cm) plus the range of nuclear forces ($\sim 2.8\times 10^{-13}$ cm). The value here leads to a Coulomb barrier of height $e^2/R=205$ kev in the CMS equivalent to a bombarding energy

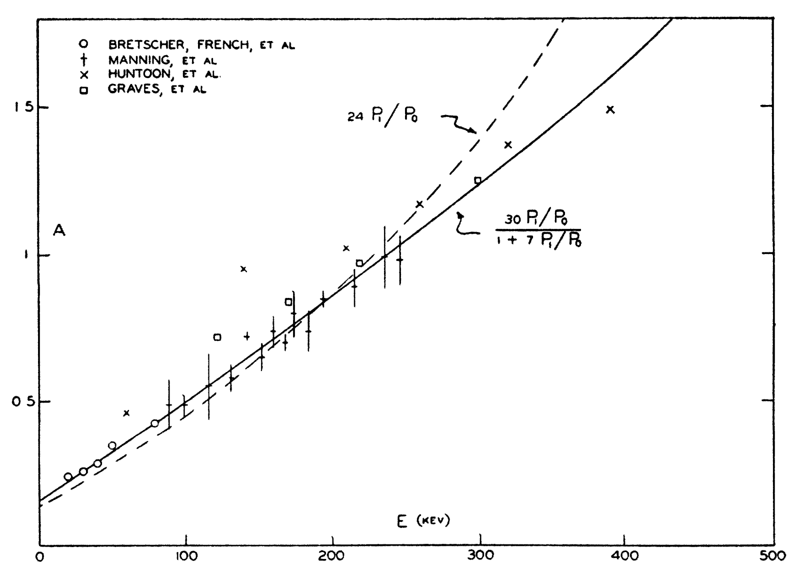

FIG. 1. The asymmetry coefficient A for proton emission as a function of bombarding energy, E. The experimental points are due to the groups indicated.[5,6,8] The numerical coefficient "24" for the steeper theoretical curve was adjusted to the data at $\sim$200 kev. This represents the case 4(a) in which all the isotropic emission is presumed to be due to incident S-waves. The second theoretical curve has had the coefficients "30" and "7" chosen so that the data at 40 kev and 200 kev are fitted. It represents case 4(c), in which some of the isotropic emission is ascribed to incident P-waves, indicating spin-orbit coupling in the transition.

[7] W. E. Bennett, C. Mandeville, and H. T. Richards, Phys. Rev. 69, 418 (1946).
[8] A. Graves, E. Graves, J. Coon, and J. Manley, Phys. Rev. 70, 101 (1946).

$E_B=410$ kev. The centrifugal barrier has the height

$$l(l+1)\hbar^2/2\mu R^2=2.05l(l+1)e^2/R\approx 420l(l+1) \text{ kev.}$$

Here l is the orbital angular momentum quantum number ($l=0, 1, 2, \cdots$), and μ is the reduced mass of the two deuterons.

Usually, centrifugal barriers are of great importance only for neutrons. Charged particles with moderate angular momentum, such as are responsible for most of the observed nuclear reactions, are most affected by electrostatic repulsion outside the nucleus. The $D+D$ reactions present a peculiar case in that the centrifugal barrier is higher than the Coulomb barrier even for P-waves ($l=1$). Actually, a comparison of the barrier heights is not quite fair because the centrifugal barrier is thinner ($\sim 1/r^2$) than the electrostatic barrier ($\sim 1/r$). The centrifugal barrier will be dominant if it manages to equal the Coulomb barrier even at the distance to which the electrostatic repulsion by itself would allow particles to approach:

$$r_0=e^2/W, \tag{3}$$

$W=\frac{1}{2}E$ being the relative kinetic energy of the two deuterons. This leads to

$$E=2W>(4\mu e^4/\hbar^2)/l(l+1)=200/l(l+1) \text{ kev}$$

for the energies at which the Coulomb barrier is negligible compared to the angular momentum barrier.

The penetration probability P_l for a pair of deuterons possessing the relative orbital angular momentum $[l(l+1)]^{\frac{1}{2}}\hbar$ is given by:

$$P_l=e^{-2C_l},$$

$$C_l=\frac{(2\mu)^{\frac{1}{2}}}{\hbar}\int_R^{r_l}\left[\frac{e^2}{r}+\frac{l(l+1)\hbar^2}{2\mu r^2}-W\right]^{\frac{1}{2}}dr. \tag{4}$$

The radius r_l is the "classical distance of closest

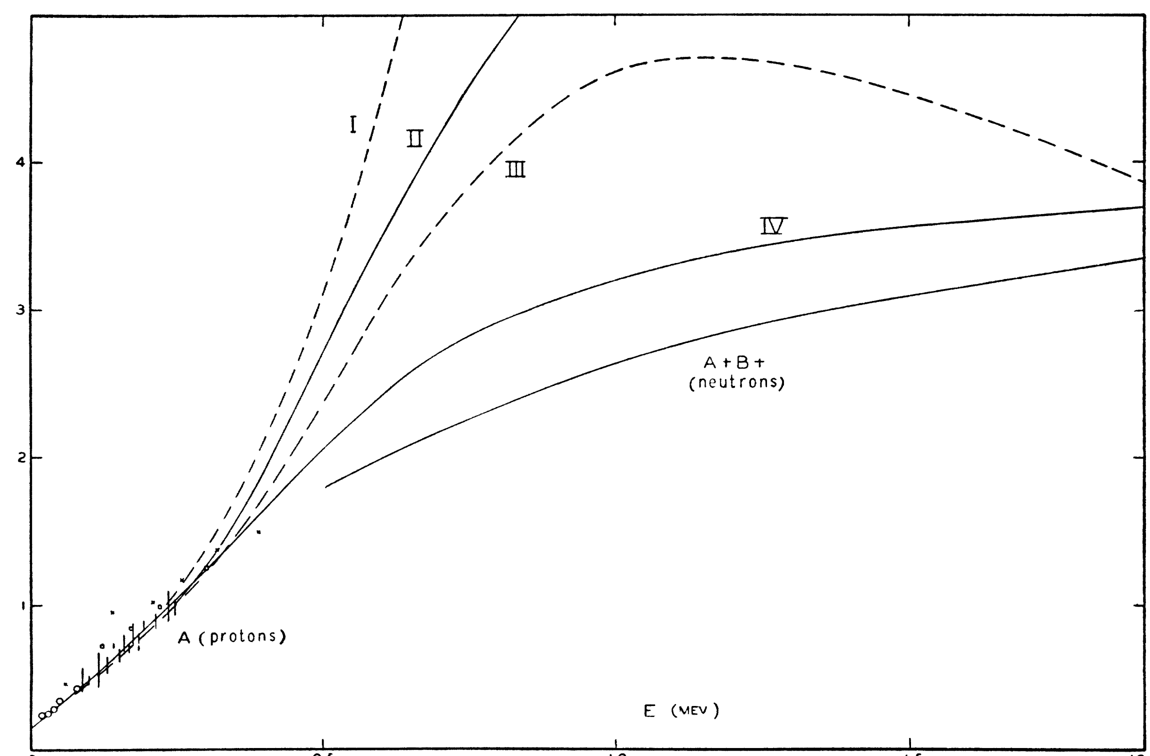

FIG. 2. The experimental points are repeated from Fig. 1. The solid curve starting at 0.5 Mev represents $A+B+\cdots$ (see 4) as measured[7] for neutron emission. The theoretical curves I and IV are extensions from Fig. 1. Curve III is A, curve II is $A+B$, according to the assumption 4(b). These two curves involved the adjusting of three parameters to fit the data at 40 kev, 250 kev and to give as low $A+B$ values at high energies as is consistent with the experimental upper limit on B at 246 kev.

approach" and is the root of the integrand. If one introduces the de Broglie wave-length (divided by 2π) :$\lambda\!\!\!^{-}=\hbar/(2\mu W)^{\frac{1}{2}}$, and the collision parameter $b=[l(l+1)]^{\frac{1}{2}}\lambda\!\!\!^{-}$, one has

$$C_l=\int_R^{r_l}[(r_0/r)+(b/r)^2-1]^{\frac{1}{2}}(dr/\lambda\!\!\!^{-}),$$
$$r_l=\tfrac{1}{2}[r_0+(r_0^2+4b^2)^{\frac{1}{2}}]. \tag{5}$$

The integrated form of C_l may be found in Bethe's article.[9] It has the well-known value $\pi e^2/\hbar v$, with v the relative velocity, for vanishingly small energy and $l=0$.

It is illuminating to notice that when the Coulomb barrier is negligible and if

$$E\ll l(l+1)\hbar^2/\mu R^2=840l(l+1)\text{ kev},$$
$$P_l\approx(\epsilon/2l+1)^{2l+1}(R/\lambda\!\!\!^{-})^{2l+1}\sim E^{l+\frac{1}{2}}. \tag{6}$$

Here ϵ is the base of the natural logarithms. (6) exhibits the well-known dependence on energy of the l-wave intensity near the nucleus, which is thus seen to be automatically included by the present procedure.

The penetration formulae presented here can be applied also to the outgoing particles, with an appropriate change of the reduced mass μ. The energy release is high enough (4 Mev) so that the Coulomb barrier for the protons can be neglected. The outgoing S-, P-, and D-waves surmount their respective barriers. The F-waves have a penetrability: $P_3\approx 0.55$, the G-waves: $P_4\approx 0.07$, values which will change little with deuteron energy.

3. CHARACTER OF THE TRANSITIONS

Two deuterons may collide in singlet, triplet, or quintet states. The triplet is antisymmetric in the deuteron spins, the others are symmetric. Thus the Bose statistics will have the consequence that the initial triplet states are odd (as to orbital quantum number) while the singlet and quintet states are even, thus: ^1S, ^5S, ^3P, ^1D, ^5D, ^3F, $\cdots$. The states ^5S and ^5D will be omitted since, in order to get reaction in these states, close approach of neutrons (and protons) with parallel spins is required.

The final states contain pairs of unlike particles of spin $\frac{1}{2}$, so that singlet and triplet states of arbitrary orbital angular momentum may be formed (the state ^3S, which requires the close approach of parallel protons or neutrons will be omitted).

If one assumes that spins must be conserved (see 1) in addition to parity and total angular momentum, the following transitions ($l\leq 2$) may occur:

$$^1S_0\rightarrow{}^1S_0;\quad {}^3P\rightarrow{}^3P;\quad {}^1D_2\rightarrow{}^1D_2. \tag{7a}$$

Quintet collisions would be totally ineffective; odd-singlet and even-triplet final states would not be formed.

Spin conservation is only approximate at best, since there is evidence for spin-orbit coupling in nuclear interactions (e.g., the quadrupole moment of the deuteron). If spin changes are admitted, one has in addition to (7a), the following possibilities:

$$^3P_1\rightarrow{}^1P_1;\quad {}^3P_2\rightarrow{}^3F_2;\quad {}^1D_2\rightarrow{}^3D_2. \tag{7b}$$

These might be expected to be weak in comparison to the transitions (7a).

No initial orbital quantum numbers higher than $l=2$ are included in the lists (7). Particles with higher l have greater centrifugal barriers to penetrate, and it can be shown that this fact makes it impossible already for $l=2$ to account for more than a small fraction of the observed reactions, as follows. The cross section for penetration to the nuclear surface is given by

$$\sigma_l=\pi\lambda\!\!\!^{-2}(2l+1)P_l \tag{8}$$

for particles with orbital quantum number l. The highest energy for which fairly reliable measurements on both the neutron producing and proton producing cross section have been made is $E\approx 300$ kev. There the total is found to be about 0.08 barn, with the proton production accounting for 0.034 barn of this. For 300 kev, $\pi\lambda\!\!\!^{-2}=\pi\hbar^2/\mu E=4.32$ barns, $P_0=0.845$, $P_1=0.0488$, and $P_2=0.00108$. This makes $\sigma_0=3.66$ b and $\sigma_1=0.634$ b. Thus, even though only $\frac{1}{9}$ of the S collisions are in the required singlet state, only $\frac{1}{3}$ of the P collisions triplet, still only a fraction of either need be effective in producing reaction to yield the observed cross section. On the other hand, $\sigma_2=0.0234$ b, so that ^1D collisions could not account for more than $0.0026/0.08\approx 3$ per-

[9] H. A. Bethe, Rev. Mod. Phys. 9, 178 (1937).

cent of the observed cross section, while ${}^5D$ collisions could only give 15 percent of the observed magnitude if every one led to a reaction. These figures are based on what is regarded as a generous nuclear radius ($R=7\times10^{-13}$ cm) which would tend to exaggerate the importance of high l-values.

Of course, the effective fraction of D collisions becomes larger relative to S and P with increasing energies. Raising the energy gives a greater increase in the penetration of the higher barrier. For this reason, the D collisions will not be left entirely out of consideration, and are included in the lists (7). An even more important reason is that S, D interference makes the D-waves more effective.

It is of interest that A is an increasing function

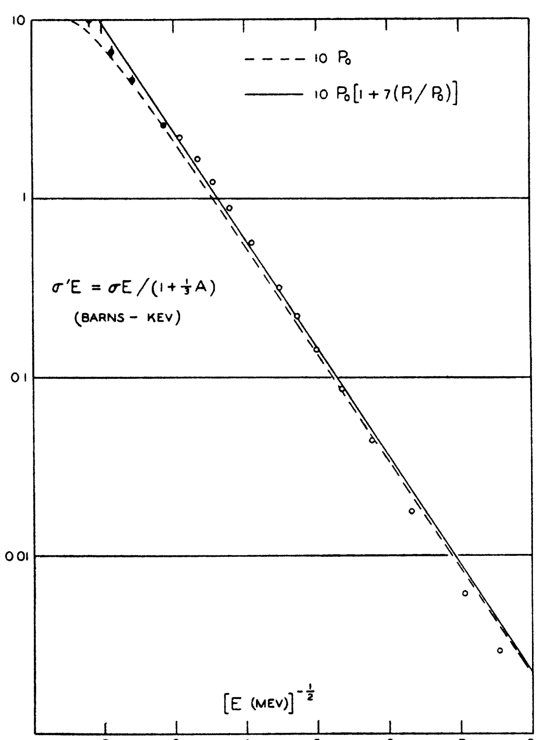

FIG. 3. The product of bombarding energy E in kev and isotropic cross section σ' in barns (10^{-24} cm^2) plotted logarithmically *vs.* $E^{-\frac{1}{2}}$ with E in Mev. The open circles (○) represent experimental points due to Bretscher, French, and Seidl.[6] The dots (●) are due to Graves, *et al.*[8] The dashed curve (---) represents the theoretical penetrability P_0 for S-waves adjusted to the data at 80 kev. The second theoretical curve represents the superposed penetration of S- and P-waves also fitted at 80 kev. The coefficient "7" indicated was obtained from an adjustment in Fig. 1.

of E (Figs. 1 and 2). This suggests that the collisions responsible for the asymmetry probably have to penetrate greater barriers than the ones giving rise to the isotropic emissions.

4. THE ANGULAR DISTRIBUTION

The S, P, $D\cdots$ states formed in the reaction are represented by de Broglie wave components having amplitudes proportional, respectively, to spherical harmonics of order $l'=0, 1, 2, \cdots$. The resultant wave intensity determines the angular distributions of the outgoing particles.

A first general statement which can be made concerning the outgoing intensity is that it cannot contain interference terms between harmonics of even (l') and odd (l') order. Such an interference would result in asymmetry with respect to the plane which is equidistant from the two deuterons. No such distinction between the forward and backward directions can arise from the reaction of two identical deuterons, which approach each other symmetrically from opposite directions in the center of mass system. Reflection through the center of mass changes the phase difference between the initial odd and even waves by π. Accordingly, with each phase difference between the odd and even states another phase difference greater by π must occur. These states give rise to product waves with similar pairs of phase differences between their odd and even components. There will result a cancellation of interference effects just as in the initial configuration.

A second general statement concerning the product angular distribution can be made when one considers the effect of the incoming wave component described by the lth spherical harmonic and when the incoming spins can be regarded as oriented at random. In that case, one can show that the outgoing intensity is described by the spherical harmonics of order $2l$, $2l-2$, $2l-4$, etc., or any superposition of these.[10]

First conservation of spin will be assumed. This assumption leads to disagreement with experiments.

Next we shall investigate how much spin-orbit coupling must be admitted to obtain agreement with the observed facts.

[10] R. G. Sachs and E. Eisner, Phys. Rev. 72, 680 (1948).

If the spin is conserved, the orbital angular momentum vector also remains unchanged. It will have the same magnitude ($l'=l$) and the same orientation ($m=0$) in the initial and final states (7a). Accordingly, the differential cross section for the emission of reaction products into the solid angle $d\omega$ will have the form:

$$d\sigma=|\sum_l \alpha_l\sigma_l^{\frac{1}{2}}g_l^{\frac{1}{2}}Y_{l0}|^2 d\omega. \tag{9}$$

The Y's are normalized spherical harmonics, appropriate to the various components of the outgoing wave amplitude. A component with quantum number l is expected to have an amplitude proportional to $\sigma_l^{\frac{1}{2}}$, since it arises from an initial l-wave having a cross section for reaching the nucleus as given by (8). g_l is a weight factor determined by the initial spin configuration; $g_l=1/9$ for the singlet collisions, 3/9 for the triplet states. The α's are coefficients, possibly complex, which determine the relative magnitudes of the outgoing wave components. Integration of $d\sigma$ over all directions of emission gives

$$\sigma=\sum_l \sigma_l g_l|\alpha_l|^2. \tag{10}$$

From the significance of σ_l and g_l it is clear that if σ is to be interpreted as the total reaction cross section then $|\alpha_l|^2\leq 1$ for each l. An appropriate name for $|\alpha_l|^2$ may be "the intrinsic reaction probability" for l-waves.

(a) $^1S_0\rightarrow {}^1S_0$ plus $^3P\rightarrow {}^3P$

If only the first two of the transitions (7a) are taken into account, the expression

$$d\sigma=(d\omega/4\pi)(\pi\lambda\!\!\!^{-2}/9)|\alpha_0|^2P_0 \times[1+27|\alpha_1/\alpha_0|^2(P_1/P_0)\cos^2\theta] \tag{11}$$

is obtained for the differential cross section. The consequent expression for the asymmetry coefficient is $A=27|\alpha_1/\alpha_0|^2(P_1/P_0)$. In order to evaluate $|\alpha_1/\alpha_0|^2$, this expression is compared with the experimental values in Fig. 1. One sees that $A=24(P_1/P_0)$ may be regarded as fitting the experimental points within the possibly large uncertainties. This corresponds to

$$|\alpha_1|^2=0.89|\alpha_0|^2.$$

To evaluate $|\alpha_0|^2$, one compares the isotropic part of the cross section resulting from (11), $\sigma'=(\pi\lambda\!\!\!^{-2}/9)|\alpha_0|^2P_0$, with the isotropic part of the proton producing cross section shown in Fig. 3. A fair fit with the experimental points occurs if $\sigma'E=10P_0$ barns-kev. This leads to $|\alpha_0|^2=0.0694$ and $|\alpha_1|^2=0.063$. Each of these quantities is expected to be less than $\frac{1}{2}$, since about half the reactions may be expected to produce neutrons instead of protons.

A striking feature of the fair agreement between the low energy experimental points and the present theoretical standpoint (a) is the comparatively large finite value of the asymmetry coefficient A for vanishing energy. One might have expected A to vanish for vanishing energy, since a vanishing velocity might be considered incapable of establishing a direction relative to which asymmetry could appear. The theory, however, gives for $E\rightarrow 0(r_0/\lambda\!\!\!^{-2}\rightarrow 2\mu e^2/\hbar^2 \equiv K)$:

$$P_l/P_0\rightarrow \exp -2\int_R^\infty \{[(K/r)+(l+\tfrac{1}{2}/r)^2]^{\frac{1}{2}} -[K/r]^{\frac{1}{2}}\}dr\approx[\epsilon^2KR/(2l+1)^2]^{2l+1}$$

if

$$KR\ll(2l+1)^2. \tag{12}$$

This is non-vanishing only in the presence of a Coulomb field ($K\neq 0$). Without it, the S-waves would predominate completely because an l-wave of vanishing energy requires an infinite "lever arm" to maintain its angular momentum of $l\hbar$, and would miss collision with the nucleus. When the Coulomb repulsion acts, r_0 becomes greater than the lever arm for low energies. The S-, P-, D-, $\cdots$ waves are all treated alike for $r<r_0$, since the centrifugal potential is there negligible in comparison to the electrostatic barrier.

Two points concerning the standpoint represented by (a) may be regarded as unsatisfactory. In the first place, the experimental values of $\sigma'E$ in Fig. 3 seem to change more rapidly with energy than can be expected from the penetrability, P_0, for S-waves. This lack of agreement persists to the lowest energies investigated by Bretscher, French, and Seidl, where the variation of the penetrability is quite insensitive to the choice of nuclear radius.

The second point at which the present theoretical explanations (a) may be unsatisfactory is concerned with the behavior of the asymmetry coefficient $A(E)$ beyond the range of energies included in Fig. 1. According to (11), the coefficient $A\sim P_1/P_0$ continues to increase rapidly (up to the top of the P-barrier, $\sim$1.7 Mev) as shown by curve I in Fig. 2. There are no measurements on the angular distribution of protons at these energies. However, one might expect them to differ little from the neutron data shown in Fig. 2, especially since the latter appear to be a smooth continuation of the proton data. We shall therefore seek other possibilities than (a), in order to explain the leveling off exhibited by the neutron data.

One precaution is necessary in dealing with the neutron data presented in Fig. 2. These data are based on measurements at two angles only, with $\cos\theta\approx 0$ and 1. The distribution (1) was assumed so that the ratio of observed intensities was equated to $1:(1+A)$. If the true distribution had been $1+A\cos^2\theta+B\cos^4\theta+\cdots$ instead, this ratio would be $1:(1+A+B+\cdots)$. Thus the measurement can be considered to have given $A+B+\cdots$ rather than A, as indicated in Fig. 2.

(b) ${}^1S\rightarrow S^1$ plus ${}^3P\rightarrow{}^3P$ plus ${}^1D\rightarrow{}^1D$

The contribution of the ${}^1D\rightarrow{}^1D$ transition to the angular distribution may be important because of the interference between the S- and D-waves. If this is taken into account, a term proportional to $(\sigma_2\sigma_0)^{\frac{1}{2}}$ appears in the expression for the differential cross section. The quantity $(\sigma_2\sigma_0)^{\frac{1}{2}}$ differs little in its energy dependence from σ_1 over most of the range; at low energies the σ_1 vanishes less rapidly.

In this case the differential cross section (9) becomes:

$$\begin{aligned}d\sigma &= (d\omega/4\pi)(|\alpha_0|^2/9)\{\sigma_0+9\sigma_1\rho_1{}^2\mu^2\\ &\quad+(5\sigma_0\sigma_2)^{\frac{1}{2}}\rho_2\cos\chi(3\mu^2-1)\\ &\qquad+\sigma_2\rho_2{}^2(5/4)(3\mu^2-1)^2\}\\ &=(d\omega/4\pi)\sigma'\{1+A\mu^2+B\mu^4\}. \qquad (13)\end{aligned}$$

Here $\rho_1{}^2=|\alpha_1/\alpha_0|^2$, $\rho_2e^{i\chi}=(\alpha_2/\alpha_0)$, $\mu\equiv\cos\theta$, and the isotropic cross section $\sigma'=(|\alpha_0|^2/9)C\sigma_0$. Thus:

$$\begin{aligned}A&=[27(P_1/P_0)\rho_1{}^2+15(P_2/P_0)^{\frac{1}{2}}\rho_2\cos\chi\\ &\qquad-(75/2)(P_2/P_0)\rho_2{}^2]/C, \qquad (14a)\end{aligned}$$

$$B=(225/4)(P_2/P_0)\rho_2{}^2/C, \qquad (14b)$$

with

$$\begin{aligned}C&=1-5(P_2/P_0)^{\frac{1}{2}}\rho_2\cos\chi\\ &\qquad+(25/4)(P_2/P_0)\rho_2{}^2. \qquad (14c)\end{aligned}$$

When a matching of the formula (14a) for A to the experimental data is attempted by giving as arbitrary values as possible to the intrinsic probabilities $|\alpha|^2$, the following facts become plain. The magnitude of $|\alpha_2|^2$ is most severely limited through the second asymmetry coefficient, B, as given by (14b). The detailed angular distribution measurement[5] of highest energy ($E\approx 246$ kev) showed no definite trace of B, but it appears that $B\leq 0.2$ at that energy could not be excluded. Now, any value of $|\alpha_2|^2$ consistent with this turns out to be too small (even though it approached the largest possible value $\sim\frac{1}{2}$) to affect much the low energy values predicted for A. This means that $|\alpha_0|^2$ and $|\alpha_1|^2$ are not significantly changed from the values found in Part (a). The small value which $|\alpha_2|^2$ must be given also limits its influence at the high energies regardless of the value of the phase χ. In Fig. 2 are shown the theoretical curves (II and III) for A and $A+B$ according to formulae (14), when $|\alpha_0|^2=1/15$, $|\alpha_1|^2=1/10$, $|\alpha_2|^2=1/10$ and $\cos\chi=-1$. These are the values which make $A+B$ a minimum at $E=600$ kev when it is required that A be fitted at the energies below 246 kev and that the value of B at 246 kev be not more than 0.2.

The conclusion is that transitions in which spin is conserved cannot account for the small value of the asymmetry coefficient, $A+B$, apparently observed for high energies, as against the comparatively large values of A at low energy. There is still the possibility that spin-orbit coupling plays a large part in the transitions.

(c) ${}^1S$ and ${}^3P$, with Spin-Orbit Coupling

It seems plausible that the spin-orbit corrections to the D-transitions, having the status of "corrections upon corrections," cannot play an important role. Accordingly, we attempt to find how large the coupling of spin and orbit must be for S and P collisions alone in order to achieve an explanation of the apparent experimental facts.

In the present case, the differential cross section, replacing (9), will be

$$d\sigma = |\alpha_0(\sigma_0/9)^{\frac{1}{2}} Y_{00} + (\sigma_1/3)^{\frac{1}{2}} \sum m\alpha_{1m} Y_{1m}|^2 d\omega$$
$$= (d\omega/4\pi)(|\alpha_0|^2/9)\{\sigma_0 + 9\sigma_1[\rho_{10}{}^2\mu^2 + \rho_{11}{}^2(1-\mu^2)]\} \quad (15)$$

in which

$$\rho_{10}{}^2 = |\alpha_{10}/\alpha_0|^2,$$
$$\rho_{11}{}^2 = \tfrac{1}{2}(|\alpha_{11}/\alpha_0|^2 + |\alpha_{1-1}/\alpha_0|^2).$$

Thus,

$$A = 27(P_1/P_0)(\rho_{10}{}^2 - \rho_{11}{}^2)/C, \quad (16a)$$

$$C = 1 + 27(P_1/P_0)\rho_{11}{}^2. \quad (16b)$$

Ostensibly, only ${}^1S \rightarrow {}^1S$ and ${}^3P \rightarrow {}^{1,3}P$ transitions are so far included. The P-waves can also give rise (7b) to ${}^3P_2 \rightarrow {}^3F_2$ transitions which are *a priori* expected to be somewhat weaker for two reasons: only the particular initial configuration with $j=2$ can produce them (thus a factor 5/9) and, further, the F-waves must penetrate an outgoing barrier (a factor ~ 0.5, see **2**). According to the argument presented above, the $P \rightarrow F$ transitions cannot alter the type of resultant angular distribution (15); such transitions will only contribute both to the isotropic and μ^2 terms. The expressions (16) for A and C would then be retained with somewhat altered meanings for ρ_{10} and ρ_{11}.

The curve IV in Fig. 2 has the form (16a) with $\rho_{10}{}^2 = 1.37$ and $\rho_{11}{}^2 = 0.26$. These values were adjusted to give the very good agreement at low energies shown both in Fig. 2 and in Fig. 1. Thus the rapid leveling off of the values of A at higher energies is a characteristic of the assumption (c). Better agreement with the experimental curve shown at high energy could have been obtained by making use of the wide margin of error permissible at the low energies. As the concluding discussion will show, it is doubtful that a closer agreement would have more significance than the present comparison.

The consequences for the isotropic cross section are shown by Fig. 3. With the present assumptions, it is found that $\sigma' E \approx 10P_0 \cdot (1+7P_1/P_0)$ barns-kev fits the data somewhat better than was the case when the S-wave alone was made responsible for the isotropic part of the product distribution. This is a result of the steeper energy dependence of the P contribution, which deals with a higher barrier. The curve calculated on the basis of (c), shown in Fig. 3, is based on the value $|\alpha_0|^2 \approx 1/15$, for the intrinsic reaction probability of the S-wave. The intrinsic probability for the P reactions is the sum of $|\alpha_{10}|^2 = 0.09$, and $|\alpha_{11}|^2 = 0.017$ and $|\alpha_{1,-1}|^2 = 0.017$.

5. CONCLUSION

The large asymmetry of the $D+D$ reaction product angular distribution *at low energies* is almost certainly due largely to the effects of P-waves superposed on isotropic emissions resulting from the incident S-waves. The *a priori* probabilities are such that the intrinsic reaction probability of P-waves need not be greater in order of magnitude than that of S-waves in order to account for the striking results. These facts hold whether assumptions 4(a) or 4(c) are made, that is, without or with the contribution of the P-wave to the isotropic component which may come from spin-orbit coupling. The presence of the spin-orbit coupling, as in 4(c), seems to provide somewhat the better agreement (Fig. 1) at the low energies inasmuch as the margins of error need not be strained.

The incident D-wave can have little effect in the low energy region because of its small penetration to the nuclear surface. It has greater effect in the region above 0.5 Mev but still much too small to account for the leveling off in the asymmetry observed for neutrons, as shown in 4(b). To produce this observed result it seems essential to assume rather large spin-orbit coupling, as done in 4(c).

The assumption 4(c), which ascribes a substantial part of the isotropic emission to incident P-waves, seems to be able to account for all the known experimental facts within their margins of error. The result shown in Fig. 2 as curve IV was produced with no attempt to fit the high energy data through the evaluation of the two parameters available with assumption 4(c). If such an attempt were regarded as significant in connection with the available experimental data, then either the wide margins of error at the low energies or the small effects of D-waves at high energies could be made use of in providing complete agreement. Such a procedure is, however, probably without significance because of the uncertainty in the relation between proton and neutron angular distributions and because of the lack of separation of the angular distribution into $\cos^2\theta$, $\cos^4\theta$, etc., terms in the high energy experiments.

A measurement of the value of B, the coefficient for the $\cos^4\theta$ asymmetry, would have the greatest significance for determining the magnitude of the spin-orbit coupling in the nuclear interactions. It would determine just how large the spin-orbit coupling must be made to account for results such as those of Fig. 2.

The authors are grateful to Mr. Geoffrey Chew for helpful calculations in connection with this work.

This document is partially based on work begun at the Los Alamos Scientific Laboratory under Government Contract W-7405-eng-36.

PHYSICAL REVIEW VOLUME 73, NUMBER 8 APRIL 15, 1948

The Diffraction of Neutrons by Crystalline Powders

E. O. WOLLAN AND C. G. SHULL
Oak Ridge National Laboratory, Oak Ridge, Tennessee

(Received January 5, 1948)

The powdered crystal technique, which has been shown for the case of x-rays to be best suited for accurate, integrated intensity measurements, has been applied to neutron diffraction. Neutrons from the Clinton pile are monochromatized by reflection from a single crystal and the diffraction patterns produced when these neutrons fall on specimens of crystalline powders have been studied. These studies have given (a) a check on some aspects of the diffraction theory, (b) the magnitude and sign of the scattering amplitudes from various nuclear species from which information is obtained on the spin dependence of the scattering, which has a bearing on the magnitude and range of nuclear forces, and (c) an improvement in the techniques and a better understanding of the problems involved so that results can be more readily obtained on the diffraction by other crystals, such as those containing hydrogen or deuterium.

Diffraction measurements have been obtained on diamond, graphite, Al, Na, NaBr, NaCl, and NaF in which all intensity measurements were standardized against diamond to which a definite cross section was assigned on the basis of total cross-section measurements. The scattering of carbon, Al, and F was found to have no measurable spin dependence. The scattering by Na, however, shows a considerable spin dependence as evidenced by a Bragg scattering cross section of 1.51 barns as against a total scattering cross section of 3.7 barns. Measurements have been made on a number of other crystals with the purpose of determining the phase of scattering. A table showing the scattering phase for a number of elements is given.

1. INTRODUCTION

EXPERIMENTAL work on the diffraction of neutrons by crystals, directed towards obtaining information about the diffraction process and its dependence on the crystal and nuclear properties of various substances, was started at Clinton Laboratories in 1945 by E. O. Wollan and R. B. Sawyer. The first measurements along this line were made with single crystals; the results gave information regarding the phase of nuclear scattering. It was found, however, that with single crystals it would be difficult to make measurements of the diffracted intensity with sufficient accuracy to permit reliable conclusions to be drawn regarding the effect on the intensity of various factors such as nuclear spin, presence of more than one isotope, characteristic temperature of the crystals, crystal structure, etc.

By using the powdered crystal method one can almost completely eliminate the effects of extinction and of crystal distortion and, as has been shown in the case of x-rays, accurate intensity measurements can be made if a sufficiently intense source of monochromatic radiation is available. The flux from the Clinton pile was found to be just sufficient to permit one to use the powder diffraction method. However, since no monochromatic lines are available with neutron sources, it is necessary to first reflect