

(*d,p*) Reaction on Heavy Elements at Low Deuteron Energies*

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Deuterons of 9.1, 8.3, and 7.4 Mev were used to produce (*d,p*) reactions in Pb^{206} and Bi^{209} targets. The proton differential cross section was measured for different Q values, each of which corresponds to a final state of known assignment. With one exception, all of the observed angular distributions were broad peaks with maxima near 180° . The theoretical approximations which apply for low deuteron energy predict a Gaussian distribution peaked in the backward direction. Although the measured distributions are not of Gaussian form, a comparison of the measured and predicted width variation with Q shows fair agreement with one theoretical result and poor agreement with the other. For the reaction with the highest Q (≈ 4.5 Mev) a peak near 120° was observed. This more forward peak would be expected both from a reduced Coulomb effect and from the influence of the nuclear potential on the proton. As expected when the Coulomb field is dominant, there was only a small observed correlation between the measured angular distribution and the angular momentum of the captured neutron. In a few cases, triton angular distributions from (*d,t*) reactions were measured, and these also showed peaks at large scattering angles.

INTRODUCTION

IN the past decade there have been many studies in which the predictions of the theory of stripping reactions have been compared with experimental results. For the lightest nuclei the agreement with the simple theory is often very good, and the Butler method¹ has allowed the assignment of parity and spin to many nuclear levels. In some cases disagreement with the theory occurs, and in other cases agreements occur which seem fortuitous.

There are two strong assumptions which were originally made in the theory. First, the outgoing proton is assumed not to interact with the nuclear potential. Wilkinson² has explained how this condition is best satisfied for deuteron stripping reactions which have low Q values. For a low- Q (*d,p*) reaction the most probable deuteron configuration is that in which the two constituent nucleons are well separated at the moment of neutron capture. Thus, when the reaction occurs, the proton need not be within range of the nuclear potential. Generally, the attractive nuclear potential, if not negligible, would have the effect of producing a more forward-peaked angular distribution. Quantitatively, this has been shown by Tobocman and Kalos.³

The second assumption made in the original theory is that the Coulomb force on both the incoming and outgoing particles can be neglected. If not negligible, the repulsive Coulomb field has been shown³ to produce an effect on the angular distribution opposite to that caused by the nuclear attractive force. In some cases the balance of these two neglected effects may produce agreement with the uncorrected theoretical results. Tobocman and Kalos point out that the predicted angular distributions from their more exact theory are

not sensitive to Q . This could be interpreted as a manifestation of a balance between these opposite effects. The extensive computer calculations required for a comparison of theory and experiment have not yet been made. Thus, except for the lightest elements with deuteron energies well above the Coulomb barrier, interpretation of the measured angular distributions may be difficult. Wilkinson shows, however, that better stripping patterns may be expected at low Q and low deuteron energies. Apparently, as the deuteron energy is decreased, the reduction of nuclear effects on the proton is more important than the increasing Coulomb effects. When investigating medium and heavy elements, the use of higher deuteron energy may not always be a simplification.

The present measurements were made in an attempt to gain further understanding of the (*d,p*) reaction, as well as to compare them with two theoretical results which specifically apply to the experiment. The experimental conditions are unusual in that the deuteron energy is well below the Coulomb barrier of the target. Under this condition Oppenheimer and Phillips⁴ first explained the high radiochemical yield from the (*d,p*) reaction. In the present work the emphasis has been on measuring the proton differential cross section. With deuterons of low enough energy, the reaction mechanism must be of a direct nature. Both formation of a compound nucleus by deuteron capture and nuclear effects on the outgoing proton will be small. In the heavy targets used, even if a compound nucleus were formed by deuteron capture, it is much more likely that a neutron rather than a proton be emitted in the de-excitation process.

Papers by Ter-Martirosian⁵ (T-M) and by Biedenharn, Boyer, and Goldstein⁶ (B) predict the angular

* Work performed under the auspices of the U. S. Atomic Energy Commission.

¹ S. T. Butler and O. H. Hittmair, *Nuclear Stripping Reactions* (John Wiley & Sons, Inc., New York, 1957).

² D. H. Wilkinson, *Phil. Mag.* **3**, 1185 (1958).

³ W. Tobocman and M. H. Kalos, *Phys. Rev.* **97**, 132 (1955).

⁴ J. R. Oppenheimer and M. Phillips, *Phys. Rev.* **48**, 500 (1935).

⁵ K. A. Ter-Martirosian, *J. Exptl. Theoret. Phys. U.S.S.R.* **29**, 713 (1955) [translation: *Soviet Phys.—JETP* **2**, 620 (1956)].

⁶ L. C. Biedenharn, K. Boyer, and M. Goldstein, *Phys. Rev.* **104**, 383 (1956).

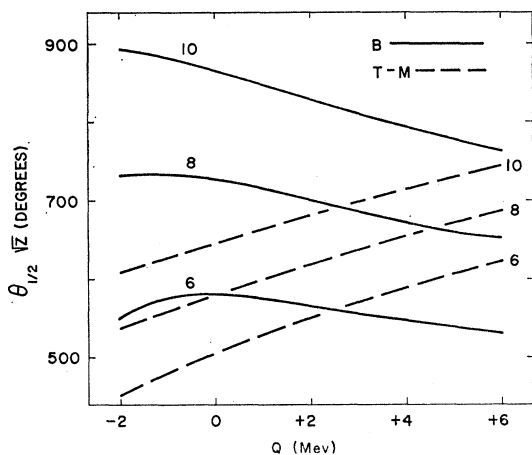


FIG. 1. Half the full-width at half maximum intensity multiplied by the square root of the target atomic number plotted against the reaction Q . These curves are the result of numerical calculations using the theoretical results of Ter-Martirosian (T-M) and of Biedenharn, Boyer, and Goldstein (B) for (d,p) reactions. The areas where these results are expected to be valid are discussed in the text.

distribution of protons under the conditions of the present experiment. Both papers predict a Gaussian proton angular distribution centered about a scattering angle of 180° . In T-M the required condition is satisfied if the deuteron energy lies below the barrier, where the barrier is considered to be Ze^2/R and R is the radius of the target nucleus of charge Z . In addition, it was necessary for Z to be greater than 50. In order for the explicit expression for the width given by B to be valid, the Coulomb parameter $\eta \equiv Ze^2/\hbar v$ was required to be much greater than unity for both the incoming deuteron and the outgoing proton, and a value of $\eta = 3$ was not considered large enough. For the (d,p) reactions described later, η_d had the range 6.2 to 6.8, and η_p had values between 3.7 and 5.0.

Although the theories seem to be of identical content, the explicit formulas given for the width of the backward-directed Gaussian proton distribution are different. Both expressions predict a narrower width as the deuteron energy decreases, but as Q decreases, for a fixed deuteron energy, B predicts an increasing width, whereas T-M predicts a narrower distribution. This is illustrated by Fig. 1, where the theoretical widths multiplied by the square root of the target Z are plotted against the reaction Q . For high values of η , both B and T-M predict little dependence of the angular distribution on the orbital angular momentum of the captured neutron.

When η is much larger than unity, the collision of charged particles can be regarded nearly classically. A large Coulomb parameter means that the reduced wavelength is small with respect to the radius of the classical turning point, and a description using the impact parameter and classical orbits is valid. As emphasized by T-M, when η is large, completely justifi-

able perturbation calculations can be made for (d,p) reactions, in contrast to the somewhat questionable use of the Born approximation or its equivalent. The Gamow penetration factor $e^{-2\pi\eta}$ shows that capture of the deuteron will be strongly inhibited for large η ; because of the nature of the deuteron wave function, the direct (d,p) reaction is reduced to a lesser extent. Ordinarily, the direct mode of a nuclear reaction becomes more probable relative to the compound mode as the energy of the incoming particle increases. The reactions of the present work are examples of the opposite case where the direct mode becomes more predominant as the deuteron energy is lowered. The enhancement of direct reactions relative to compound nucleus formation as well as effects of deuteron distortion have been discussed in a recent paper by Biedenharn and Satchler.⁷

There have been several experimental investigations made in a region lying between the present work and what might be called the Butler region. Measurements at Indiana University^{8,9} on isotopes of lead and bismuth have been made at 11 Mev. At this deuteron energy, great sensitivity of the proton angular distribution to the captured neutron angular momentum is not expected. However, a correlation of the angular position of maximum intensity with l_n gave consistent results which agreed well with predictions of the shell model. All the observed angular distributions had low forward

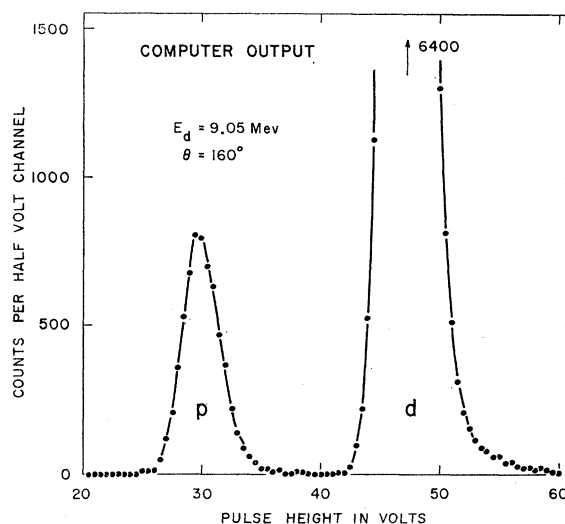


FIG. 2. An example of the pulse height spectrum from the electronic computer used to identify the mass of the reaction products from a Pb^{206} target. The left peak contains protons of all energies from the (d,p) reaction, and the other peak contains all the deuterons, most of which are the result of elastic scattering.

⁷ L. C. Biedenharn and G. R. Satchler, paper given at the International Conference on Polarization Phenomena of Nucleons, Basel, 1960 (unpublished).

⁸ M. T. McEllistrem, H. J. Martin, D. W. Miller, and M. B. Sampson, Phys. Rev. **111**, 1636 (1958).

⁹ G. B. Holm, J. R. Burwell, and D. W. Miller, Phys. Rev. **118**, 1247 (1960).

intensity and a peak in the range 60° to 120° . Schiffer and Lee¹⁰ have measured the proton angular distribution from (d, p) reactions in elements between titanium and nickel. At deuteron energies of 3.8 and 4.5 Mev, the distributions were rather broad with a peak near 60° . In their experiment the maximum value of η_d was 3.2. Pratt¹¹ has measured the angular distribution of protons from the $\text{Ti}^{48}(\text{d}, \text{p})$ reaction at a deuteron energy of 2.6 Mev. These measurements have been fitted by various refinements^{3,7} of stripping theory.

There have been instances¹² where a backward peak was observed in the center-of-mass angular distribution. These usually occur in addition to a more forward-peaked component. In these cases it is not clear whether the backward-peaked protons were from the deuteron or were the result of heavy-particle stripping.¹³ In the present experiment where heavy targets were used, the center-of-mass system and the laboratory system have only a small relative velocity, and, for the Q values concerned, heavy-particle stripping cannot contribute. All of the measured proton groups had Q values greater than -2.23 Mev; therefore, protons from the electric breakup of the deuteron in the Coulomb field of the target were excluded because of energy.

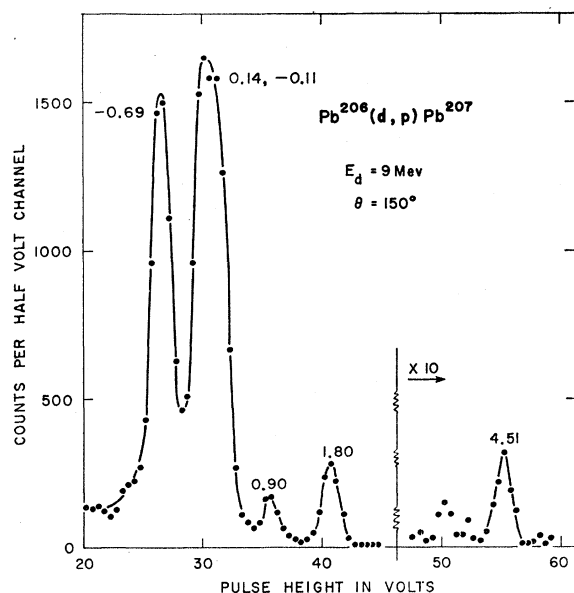


FIG. 3. Proton energy spectrum as measured by a NaI scintillation spectrometer. The number near each strong group is the associated Q value in Mev.

¹⁰ J. P. Schiffer and L. L. Lee, Jr., Phys. Rev. **115**, 1705 (1959).

¹¹ W. W. Pratt, Phys. Rev. **97**, 131 (1955).

¹² G. C. Phillips, Phys. Rev. **80**, 164 (1950); D. de Jong, P. M. Endt, and L. J. G. Simons, Physica **18**, 676 (1952); E. Baumgartner and H. W. Fulbright, Phys. Rev. **107**, 219 (1957); I. B. Teplov and B. A. Iur'ev, J. Exptl. Theoret. Phys. U.S.S.R. **34**, 334 (1958) [translation: Soviet Phys.—JETP **7**, 233 (1958)].

¹³ L. Madansky and G. E. Owen, Phys. Rev. **99**, 1608 (1955).

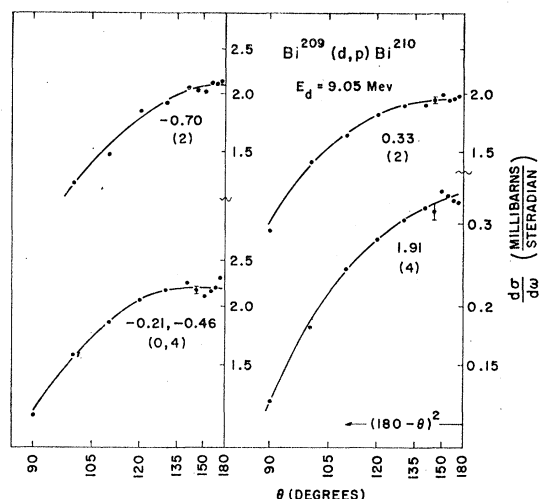


FIG. 4. Measured proton differential cross sections from a Bi^{209} target with a deuteron energy of 9.05 Mev.

EXPERIMENTAL PROCEDURE

The Los Alamos variable-energy cyclotron was used to produce deuteron beams of 7.35, 8.30, 9.05, and 11.9 Mev. A beam $\frac{5}{32}$ in. high and $\frac{1}{16}$ in. wide traversed a target at the center of a reaction chamber.¹⁴ The targets of lead and bismuth were self-supporting evaporated metal foils a few milligrams per square centimeter thick. The Pb^{206} target was made from radiogenic lead and contained¹⁵ 88% Pb^{206} . In the (d, t) measurement on U^{235} , the target consisted of ≈ 1 mg/cm² of oxide which had been evaporated onto 200- $\mu\text{g}/\text{cm}^2$ gold leaf.

Two counters were used to identify and measure the energy of the charged reaction products. The particles first passed through the ΔE counter which measured their relative specific ionization and then they were stopped in the E counter which measured the residual energy. The ΔE counter was an ion chamber and the E counter a NaI scintillator. Pulses from these counters were amplified and then went to an electronic computer.^{16,17} This computer generated a pulse whose height was characteristic of the mass of the particular charged particle. A pulse-height discriminator then allowed either protons or tritons to be distinguished from the strong flux of elastically scattered deuterons. As an example, the computer output spectrum for the $\text{Pb}^{206}(\text{d}, \text{p})$ reaction is shown in Fig. 2. In spite of the strong elastic component, it is easy to identify protons of all energies which arise from the reaction. Figure 3, which gives a representative energy spectrum, shows that the modest resolution of the E counter is adequate except for the one pair of closely spaced levels.

¹⁴ H. E. Wegner and W. S. Hall, Phys. Rev. **119**, 1654 (1960).

¹⁵ R. E. Peterson, R. K. Adair, and H. H. Barschall, Phys. Rev. **79**, 935 (1950).

¹⁶ R. H. Stokes, Rev. Sci. Instr. **31**, 768 (1960).

¹⁷ R. H. Stokes, J. A. Northrop, and K. Boyer, Rev. Sci. Instr. **29**, 61 (1958).

The beam energy was measured¹⁸ and continuously monitored by slowing an elastically scattered fraction in the proper thickness absorber to reduce the energy to a standard value. A meter, with reading proportional to the energy of the slowed beam, was used by the cyclotron operator as a guide in holding constant energy during the period data were collected. The relative amount of beam, for data taken at different angles, was measured by a scintillator which counted elastically scattered deuterons from the target. The absolute cross section was obtained, using the elastically scattered deuterons as a reference. At these low deuteron energies and with high- Z targets, it was easy to move the counter system forward to an angular position where the elastic scattering was Rutherford. In this way, provided the deuteron energy is known, an absolute cross section can be obtained without measuring either the target thickness or the solid angle of the counters. In one case, where this method was compared to a determination of cross section from direct measurements of beam current, counter solid angle, and target thickness, results were obtained which differed by about 20%. Most of this disagreement could have arisen from the target thickness measurement which was an average taken over an area much larger than the beam spot.

At low deuteron energies and with high- Z targets, the Coulomb scattering of deuterons is very strong and the reaction cross section small. Also, reactions from the small amounts of target contaminants such as carbon and oxygen are relatively emphasized since the deuteron energy is far over their Coulomb barrier. The first effect tends to make protons more difficult to identify, and the second contributes an unwanted background to the proton spectrum of interest. The protons from heavy targets

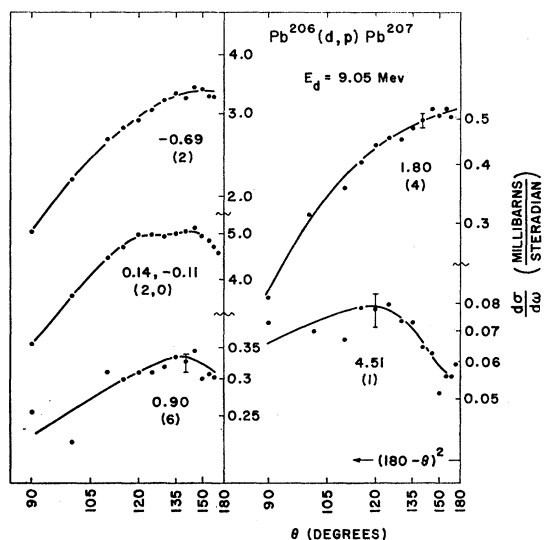


FIG. 5. Measured proton differential cross sections from a Pb^{206} target with a deuteron energy of 9.05 Mev.

¹⁸ J. A. Northrop and R. H. Stokes, Rev. Sci. Instr. 29, 287 (1958).

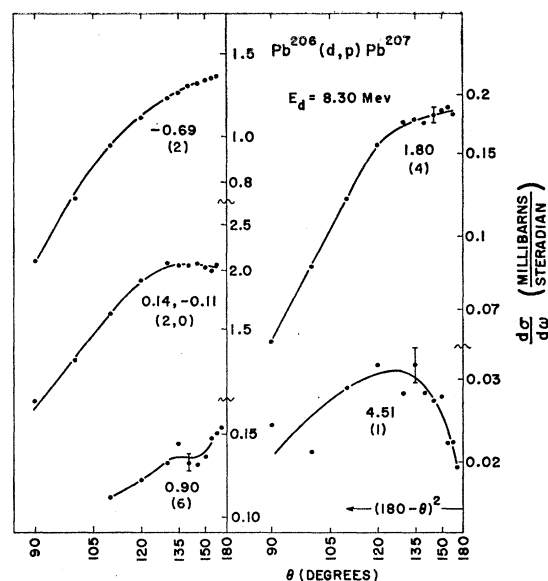


FIG. 6. Measured proton differential cross section from a Pb^{206} target with a deuteron energy of 8.30 Mev.

are emitted mainly in the rear hemisphere while Coulomb scattered deuterons, reaction protons from light elements, and recoil protons from hydrogen contamination are emitted predominantly forward. In addition, the background of neutrons and gamma rays from the gold collimators and from the cyclotron decrease rapidly as the cyclotron energy is lowered. For these reasons, it was easier than was expected to measure such low cross sections with counters.

A few triton angular distributions were measured for the $\text{Bi}^{209}(d,t)$ and the $\text{U}^{235}(d,t)$ reactions. In this case the deuteron energy was raised to 11.9 Mev to increase the yield and to allow better triton identification. In the U^{235} measurement, the target contained a large amount of oxygen as well as some carbon. The (d,t) ground state Q values for both carbon and oxygen are highly negative, and the triton energy is further reduced by the center-of-mass effect. This means that in heavy elements, where the (d,t) ground state Q values are often nearly zero or only slightly negative, it is easy to observe many levels with no background from these contaminants. Avoiding background by this method is important when investigating targets such as uranium or plutonium, which are very difficult to obtain as thin metallic foils. Choosing the (d,t) reaction instead of the (d,p) , which has positive Q values for carbon and oxygen, affords great simplification.

RESULTS

The differential cross section data are shown in Figs. 4 to 7. Since both the theory of T-M and of B predict Gaussian distributions, the data were plotted so that, if the results were Gaussian, the points would fall on a straight line. The ordinate has a logarithmic scale and

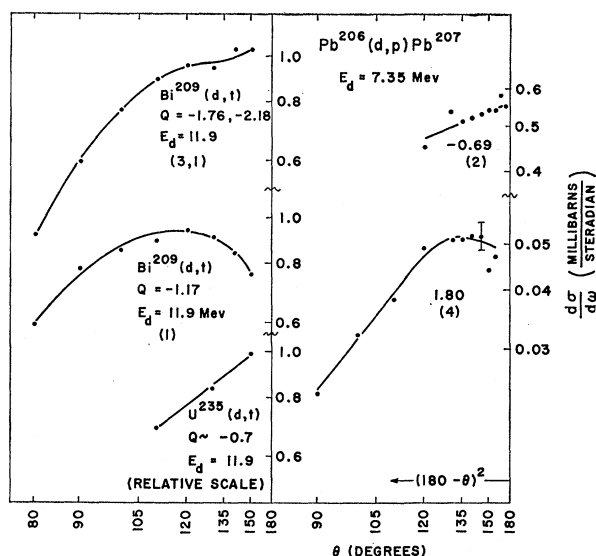


FIG. 7. Measured proton and triton differential cross sections. The ordinate scale is relative for the $U^{235}(d,t)$ reaction only.

the square of the angular deviation from 180° is plotted horizontally. θ is the laboratory scattering angle. In all the distributions where a representative error point is not shown, the statistical errors lie between 1 and 2%. For each distribution, the reaction Q value in Mev is given, and the value of l_n is given in parentheses. As can be seen from the four figures, most of the distributions deviate considerably from a Gaussian shape. These figures show an angular range of 90° to 165° laboratory scattering angle. In a few cases, points were also taken at smaller angles including 60° . Although these data were not accurate, they indicated that the intensity continues to decrease as the angle decreased. Thus, the distributions were peaked in the backward hemisphere and were not symmetric about 90° .

Since the shapes of the observed distributions are not as predicted by the theories, it is difficult to make a valid comparison of width. Arbitrarily, half the full-width at half-maximum was chosen to characterize the width of the measured distributions. Figures 8 and 9 show these experimental widths as well as the predictions of the two theories. Little significance should be attached to the absolute magnitude since the predicted shape is different from that measured. However, the variation of width as a function of Q does seem to favor the approximate result of theory B. The $Q=4.51$ -Mev distribution was not used in this comparison since a consistent measure of its width was lacking.

If the (d,p) reaction on targets near doubly magic Pb^{208} produces single-particle neutron states, the value of the orbital angular momentum of the captured neutron l_n is unique if the assignment of the final state is known. In the figures giving the proton distributions from Pb^{206} and Bi^{209} , the Q value and the l_n value (in parentheses) are taken from the work of Holm, Burwell, and Miller.⁹ Although the observed distributions arise

from a rather large range of l_n values, the shape of the distributions is not strikingly different. The unresolved pair at $Q=0.14$ and -0.11 Mev from Pb^{206} , as well as the pair at $Q=-0.21$ and -0.46 Mev from Bi^{209} , show either a tendency to peak away from 180° or a small forward bump as well as a more backward peak. Since one member of each pair arises from $l_n=0$, this behavior is consistent with a small residual tendency for more forward peaking at low l_n . The $Q=4.51$ -Mev level shows a definite peak in the vicinity of 120° . This level requires $l_n=1$, which may explain the shape, or, as discussed later, dynamical factors which are characteristic of high Q may produce such an effect. The $Bi^{209}(d,t)$ data taken at $E_d=11.9$ Mev (Fig. 7) also show angular distributions peaked in the backward hemisphere. Assuming that the (d,t) reaction produces hole states in the neutron core of the target, the value of the orbital angular momentum of the picked-up neutron is determined if the level assignments of Bi^{208} are known. In Fig. 7, the Q values and the l_n values (in parentheses) are taken from the work of Harvey.¹⁹

DISCUSSION

The most striking feature of the data is the consistent appearance of backward peaking in the angular distributions. Although the conditions of T-M are fulfilled and the conditions of B are rather well met, the angular distributions are not distributed about 180° in a Gaussian manner. Whether the values of η are not high enough or whether there is a more serious difficulty in the theory is not clear. It has been pointed out²⁰ that this disagreement is not surprising. The various momentum-transfer factors which are assumed constant in obtaining the Gaussian approximation are in fact angle-dependent. For example, the form factor which multiplies the Coulomb integral varies by a factor of

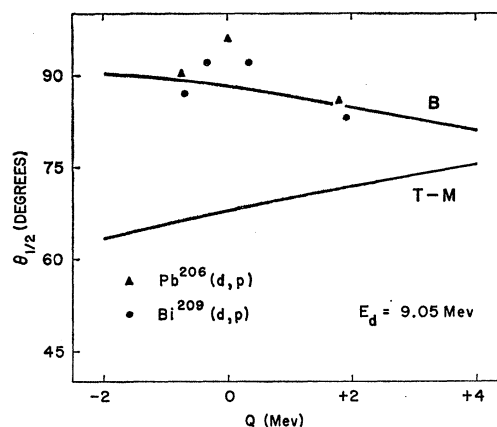


FIG. 8. Comparison of the measured widths (points) at $E_d=9.05$ Mev with curves showing the theoretical predictions. As explained in the text, significance should be attached only to the variation of the measurements with Q and not to their absolute magnitude.

¹⁹ J. A. Harvey, Can. J. Phys. **31**, 278 (1953).

²⁰ L. S. Rodberg (private communication).

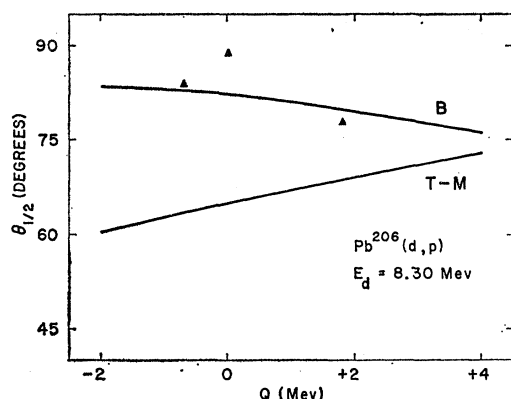


Fig. 9. Comparison of the measured widths (points) at $E_d = 8.30$ Mev with curves showing the theoretical predictions.

nearly four between 90° and 180° . Another possible explanation arises from the neglected effect of deuteron distortion. If the internal wave function of the incoming deuteron is appreciably distorted by the Coulomb field of the target, it is reasonable to expect that the proton angular distribution would be of different shape than if the deuteron were a more rigid body.

The $Q = 4.51$ -Mev distribution from Pb^{206} shows a peak at $\approx 120^\circ$ at both deuteron energies where it was measured. In this case $l_n = 1$, which may partly explain this result; however, it is interesting to consider the possible effects of a large Q on the angular distribution. In a (d, p) reaction on a heavy element just following the moment of neutron capture, it is most probable that the proton have a kinetic energy $\approx \frac{1}{2}(E_d - \epsilon_d - B)$, where ϵ_d is the deuteron binding energy and B is the Coulomb energy of the proton. Depending on its position at the time of neutron capture, the proton then gains additional kinetic energy as it is repelled from the product nucleus. To have high energy, the proton must be at small radius at the time of the reaction, not only because of the subsequent energy added by the Coulomb field, but also because of the added kinetic energy from the deuteron internal wave function when the neutron-proton separation is small. Thus, high Q reactions are expected to occur with the proton closer to the nucleus. In this case the proton can be attracted by the nuclear force and will be ejected at a more forward scattering angle. This mechanism for a high Q reaction is supported by the observed rapid decrease in the proton energy spectrum as Q increases. At $E_d = 8.3$ Mev the cross section decreases by a factor of 40 as Q changes by 5.2 Mev. Part of this decrease would be expected from the internal momentum distribution of the deuteron, but a large part must arise from Coulomb effects. There is another possible contribution to the shape of the

$Q = 4.51$ -Mev distribution. Deuteron capture which is most likely for a head-on collision, would tend to remove those protons which would have been emitted in the backward direction. This effect could partly explain the minima at 180° .

It is of interest to ask if low-energy reactions on medium and heavy elements could be used to determine the spin of nuclear levels. One possibility would be for the excitation function to have different slopes according to the orbital angular momentum of the transferred nucleon. If this could be observed, the parity and the spin of the final nucleus could be determined experimentally. The present data were examined for such an effect; however, none was found. The highest Q groups from (d, p) reactions on heavy elements decrease most rapidly in cross section as the deuteron energy is lowered, which makes such a correlation difficult to find.

The low-energy reactions discussed here have similarities to processes in the field of heavy-ion physics. It would seem that some of the objects of heavy-ion investigations could be accomplished by studying reactions of low-energy protons or alpha particles. The repulsion of the incoming particle by the Coulomb field makes interactions with surface nucleons most probable. The distortion of protons and alpha particles by the target is small and more efficient momentum transfer to the surface nucleons is possible. Inelastic scattering of low-energy protons or alpha particles, or reactions with these particles in or out, could give information on the position-momentum distribution of the target nucleons. As previously discussed, measurement of the inherently low cross sections is not as difficult as might be expected, and the acceleration, detection, and identification of these lighter particles is easier than for heavy ions.

A few measurements were made in an attempt to observe the effects of the electric breakup of the deuteron in the Coulomb field of the target. The results were inconclusive because of background effects in the low-energy range where breakup protons are expected. The angular distribution of all protons from electric breakup has been calculated by Landau and Lifshitz.²¹ The width of their Gaussian distribution differs from the width of the proton distribution given by T-M. For instance, at $E_d = 9$ Mev, Landau and Lifshitz predict a 36% greater width than that calculated from T-M at $Q = -3$ Mev. However, when the width for electric breakup is compared with the width given by B, the results are in much closer agreement, and it would be difficult to determine the difference experimentally.

²¹ L. Landau and E. Lifshitz, *Zhur. Eksp. i Theoret. Fiz. U.S.S.R.* 18, 750 (1948).