Measurement of branching ratios of low energy deuteron-induced nuclear reactions on ^{2}H, ^{6}Li, and ^{10}B

F. E. Cecil, H. Liu, and J. S. Yan
Department of Physics, Colorado School of Mines, Golden, Colorado 80401

G. M. Hale
Los Alamos National Laboratory, Los Alamos, New Mexico 87545
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We have measured the branching ratios ^{2}H$(d,p)^{3}$H/^{2}H$(d,n)^{3}$He, ^{6}Li$(d,p)^{7}$Li/^{6}Li$(d,\alpha)^{4}$He, and ^{10}B$(d,p)^{11}$B/^{10}B$(d,\alpha)^{8}$Be between c.m. energies of 3 and 15 keV, 20 and 135 keV, and 58 and 142 keV, respectively. Our measurements of the ^{2}H-d reaction are in good agreement with R-matrix calculations of the branching ratio. We find no enhancement of the (d,p) branches of these reactions at the lowest observed energies. Implications of our findings to recent claims of anomalous production of heat from deuterium-metal systems are presented.

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I. INTRODUCTION

The Oppenheimer-Phillips (OP) [1] effect suggests that the interactions between a projectile deuteron and target nucleus at energies below the Coulomb barrier may be affected by the electric polarization of the deuteron in the electric field of the target. The static polarizability of the deuteron has been the subject of considerable theoretical effort [2], and the consensus appears to acknowledge a polarizability of about 0.62 fm^3. This theoretical estimate is in good agreement with the polarizability which was inferred experimentally by careful measurements of low-energy elastic deuteron scattering by heavy nuclei [3].

Another manifestation of the OP effect would be an asymmetry between the single nucleon transfer reactions (d,p) and (d,n) at energies well below the deuteron-target Coulomb barrier. The asymmetry may be naively imagined to result from the fact that, when electrically polarized by the target nucleus, the proton in the projectile deuteron will be farther from the target than will the neutron.

Confirmation of this naive picture is muddled by conflicting experimental evidence and serious theoretical calculations. Experimentally, Lawrence, McMillan, and Thornton [4] investigated the (d,p) reactions on Al, Na, Si, and Cu as functions of deuteron bombarding energy by measuring the yield of the respective short-lived activation products. They found that the energy dependence of these reaction yields was inconsistent with the Coulomb barrier penetration probability by a point deuteron, but *was* consistent with the adiabatic approximation to a polarizable deuteron as calculated by Oppenheimer and Phillips [1]. This discrepancy was most pronounced in the lighter targets with the measured yields exceeding the yields expected on the basis of mere Coulomb barrier penetration probabilities by 50% or more.

In subsequent distorted wave Born approximation (DWBA) calculations, Dar, de-Shalit, and Reiner [5] concluded that the polarizability of the deuteron would not effect low-energy deuteron stripping reactions by more than a few percent. The most sensitive experimental test of the difference between (d,p) and (d,n) reactions would be a measurement of the ratio of the cross sections for these processes since such a measurement would eliminate the dependence on such factors as charge collection, target thickness, stopping powers, etc, all factors contributing to the difficulty of measuring absolute cross sections. Paul [6], for example, measured the relative yields of the gamma rays from the mirror reactions ^{6}Li$(d,p1)^{7}$Li(478 keV) and ^{6}Li$(d,n1)^{7}$Be(429 keV) between bombarding energies of 120 keV and 2.5 MeV. In an earlier report from our laboratory [7], the same relative yields were measured between deuteron bombarding energies of 60 and 170 keV. Our measurements agreed with those of Paul in the region between 120 and 170 keV, and both sets of measurements clearly indicated that the ratio of the (d,n) to (d,p) decreased with decreasing energies by as much as 15–20% at the lowest energies. Similar results were reported in a later study of the (d,n) and (d,p) reaction ratios on ^{9}Be between deuteron bombarding energies of 80 and 170 keV [8]; again, the (d,n) to (d,p) ratio decreased by about 10% at the lowest energies.

Recently, Koonin and Mukerjee [9] have carried out a series of second-order Born approximation calculations of the (d,n) to (d,p) reaction cross section ratios on ^{2}H, ^{6}Li, and ^{27}Al from deuteron energies of a few MeV down to essentially zero energy. They find no significant energy-dependent asymmetry between the two reaction branches on these three targets. We are thus left in something of a quandary; we recognize three fairly recent measurements, indicating a significant energy dependence of the (d,p) to (d,n) ratio on light nuclei, and two fairly recently calculations, concluding definitively that there should be no such dependence.

In an effort to extricate ourselves from this quandary, we addressed the asymmetry in the (d,p) to (d,n) ratio from two perspectives. Experimentally, we have measured the deuteron-induced reactions on ^{2}H, ^{6}Li, and ^{10}B down to deuteron bombarding energies of 6, 27.5, and 70 keV, respectively. In the case of the reaction on ^{2}H, the charged particles corresponding to the (d,p) and (d,n) reactions were observed directly. In the case of the reactions on ^{6}Li and ^{10}B, we investigated the possible enhancement of the (d,p) reactions by comparing the yields of the (d,p) to (d,α) reactions. Theoretically, we have carried out exact, nonperturbative, calculations of the (d,p) and (d,n) reactions on ^{2}H using the R-matrix formalism.

II. EXPERIMENTAL PROCEDURE AND RESULTS

The measurements of the branching ratios for deuteron-induced reactions on ^{2}H, ^{6}Li, and ^{10}B were carried out with magnetically analyzed deuteron beams from the low-energy charged particle accelerator at the Colorado School of Mines [10]. As discussed in Ref. [10], the energy of the beam was calibrated at the higher energies using the resonance in the reaction ^{11}B$(p,\gamma)^{12}$C at a proton bombarding energy of 163.1 keV. At energies below about 25 keV, the energy of the beam was determined by measuring the potential difference between the ion source and the grounded scattering chamber with a commercial precision voltage divider and a voltmeter. The potential difference was determined to an accuracy of about 10 eV. The pressure along the 2-m beam line during the runs was about 5×10^{-7} Torr, and consequently the measured potential difference was taken to be an accurate measure of the beam energy. The targets for the measurements on ^{2}H, ^{6}Li, and ^{10}B consisted of pressed wafers of polyethelene, enriched to 98% in CD_2, rolled foils of 98% enriched metallic ^{6}Li, and pressed pellets of 90% enriched ^{10}B powder. The chief contaminants in the targets were ordinary polyethelene, ^{7}Li, and ^{11}B, respectively. No reactions from these contaminants yielded charged particles which interfered with the particles under consideration. All other listed contaminants were at the parts per 10^{6} level. The targets for all measurements were typically several hundred micrometers in thickness, and hence the ion beams of even the highest energies stopped in the targets. At the lowest energies, beam currents of up to 100 μA with beam spot sizes of about 1 cm^{2} were used.

For the ^{2}H measurement, the detector consisted of a circular 300-mm^{2} Ortec ruggedized surface barrier silicon detector at a distance of 5 cm from the target and at an angle of 135° from the beam. The detector was used with no additional protective foil on the front. Consequently, we were able to measure all three of the charged reaction products of the d-d reaction: the 3.09-MeV proton and the 1.01-MeV triton from the ^{2}H$(d,p)^{3}$H reaction and the 0.86-MeV ^{3}He ion from the ^{2}H$(d,n)^{3}$He reaction. Above a deuteron bombarding energy of about 30 keV, however, the elastically scattered deuterons created an intense low-level background which severely deteriorated the detector resolution and precluded any measurement of the reaction at higher energies. The spectrum measured

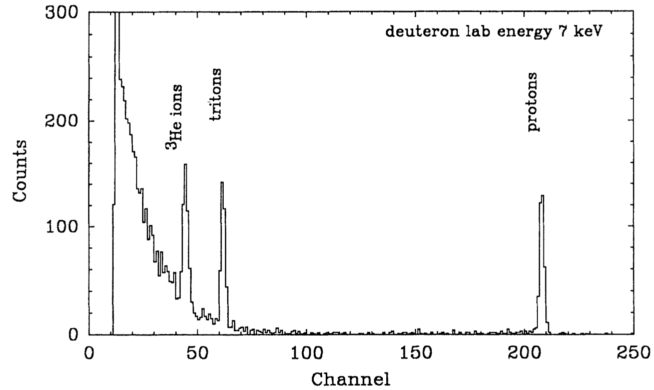

FIG. 1. Energy spectrum of charged particles during bombardment of a CD_2 target with a 7-keV deuteron beam.

at a deuteron bombarding energy of 7 keV is shown in Fig. 1. The reaction ratio was taken directly from the ^{3}He and triton peak yields. The ratios measured between c.m. energies of 17 and 3 keV are plotted in Fig. 2. Although no angular distributions were measured in the present work, the angular distributions for both branches of the (d,d) reaction have been established at a c.m. energy of 7.5 keV [11] and are isotropic at the 10% level. We assume that this near isotropy is maintained down to the lowest energy of the present work, and consequently the ratios of the yields measured at a single angle are used to infer the total cross section ratio. The present measurements are compared to total reaction cross section ratios determined from published cross section measurements of the ^{2}H$(d,p)^{3}$H and ^{2}H$(d,n)^{3}$He reactions [11,12]. From our measurements as well as the previous measurements, we find no evidence for an enhancement of the (d,p) branch of the d-d reaction. The measured yields are likewise in good absolute agreement with our R-matrix calculation and in good relative agreement with the second-order DWBA calculation of Koonin and Mukerjee [9].

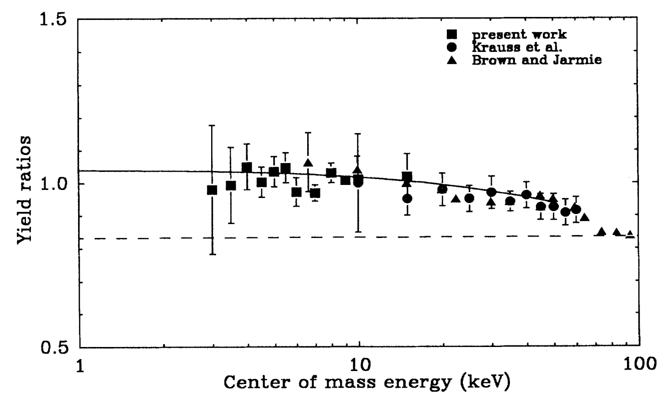

FIG. 2. Ratio of yields of ^{2}H$(d,p)^{3}$H and ^{2}H$(d,n)^{3}$He reactions. Solid symbols are measured ratios; lines are calculated ratios. The circles are from Ref. [11]; the triangles are from Ref. [12]. The solid line is the present calculation and the dashed line is from Ref. [9].

Our R-matrix calculations of the d-d reaction comes from a charge-independent analysis of the $A=4$ system that has been in progress for some time at Los Alamos National Laboratory [13]. The calculation is charge independent in the sense that the $T=1$ parameters are first determined to fit the data for the $p+{}^3$He elastic scattering, checked by predicting $n+{}^3$H scattering results [14], and then used in a large analysis of reaction in the ^{4}He system in which only the $T=0$ parameters are varied. In addition, isospin constraints are used to relate the p-^{3}H and n-^{3}He widths for both the $T=0$ and 1 levels. A small amount of internal isospin mixing is introduced by allowing slightly nonzero d-d widths (less than 0.1% of the single particle value) to occur in the $T=1$ levels.

These nonzero widths, which seem to be consistent with internal Coulomb mixing, become greatly amplified in the external Coulomb field by the proximity of broad P-wave levels having opposite isospin, causing large differences in the transition matrix elements for the two branches to occur in the $L=1$ initial d-d states. In fact, the P-wave cross section branching ratio from this calculation [13],

$$[\sigma_{d,n}^{L=1}/\sigma_{d,p}^{L=1}]_{\text{calc}}=1.43\pm0.03 ,$$

is in excellent agreement with the results of recent muon-catalyzed fusion and polarization experiments [15,16].

Although the effect of the P waves on the integrated cross sections is masked at low energies by large S-wave contributions, it is this enhancement of the neutron in the P-wave states that causes the branching ratio shown in Fig. 2 to drop below unity at c.m. energies above 15 keV. At low energies the dominant S-wave transitions are not strongly isospin mixed and, in fact, favor the proton branch, as would be expected from considering only the outgoing-channel penetrability factors, resulting in a low-energy branching ratio value (1.04) slightly greater than unity.

For the ^{6}Li measurements, the bombarding energies were too high to operate the detector without a front protective foil, and consequently we were unable to measure the ratio of the (d,p) and (d,n) cross sections by measuring the recoil ^{7}Li and ^{7}Be. We therefore sought evidence of an enhancement of the (d,p) cross section by comparing the yields of the 5-MeV protons from the reaction ^{6}Li$(d,p)^7$Li to the yields of the 11-MeV alpha particle from the reaction ^{6}Li$(d,\alpha)^4$He. This comparison is justified by the assumption, as first pointed out by Lawrence, McMillan and Thornton [4], that the (d,α) reaction would involve the penetration of the entire deuteron through the Coulomb barrier set up by the target and as such would not be affected by the electric polarization of the projectile deuteron. At high energies the yields of the 5-MeV protons and 11-meV alpha particles were measured with a single 300-mm^2 and 300-μm-thick detector in a setup identical to the measurement of the d-d reaction. The spectrum measured at a deuteron bombarding energy of 120 keV is shown in Fig. 3. At lower energies it was not possible to measure the yield of the 5-MeV proton from the ^{6}Li$(d,p)^7$Li reaction because of the pileup continuum from the 3-MeV protons from the d-d reaction. The reaction occurs because of the buildup of deuterium in the target from the deuteron beam, and the pileup extends to 6 MeV. In an effort to eliminate this deleterious pile-up, the reaction was studied with a detector telescope consisting of a 150-μm-thick front detector followed by a 500-μm-thick back detector. By gating the summed signal of the two detectors on a pulse from the back detector, the pile-up signals were excluded from the

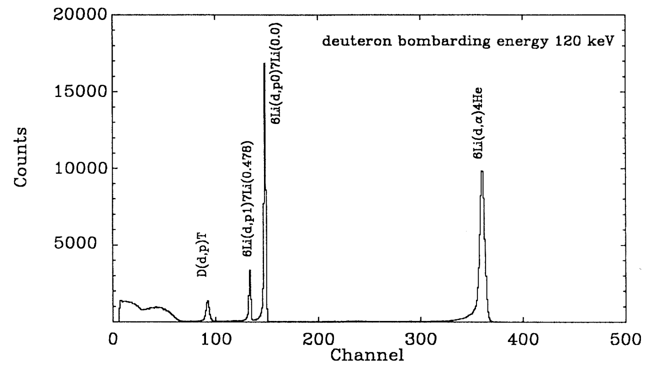

FIG. 3. Energy spectrum measured during bombardment of ^{6}Li with a 120-keV deuteron beam with a single detector at an angle of 135°.

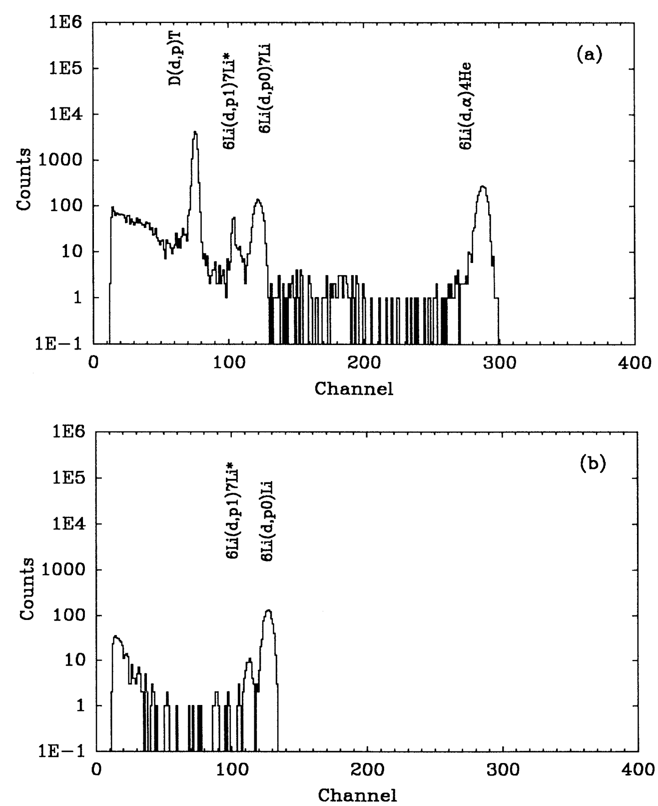

FIG. 4. Energy spectra during bombardment of a ^{6}Li reaction with 50-keV deuterons using a two-detector telescope. (a) Spectrum not gated on a signal from the back detector; (b) spectrum gated on a signal from the back detector.

spectra. The gated spectra were used to determine the yields of the 5-MeV protons, and the ungated spectra were used to determine the yields of the 11-MeV alphas. Because of straggling in the front detector, we were unable to measure reliably the yield to the ^{7}Li excited state at 478 keV. The gated and ungated spectra measured at a bombarding energy of 50 keV, for which the pileup was not a serious problem, are shown in Figs. 4(a) and 4(b). These figures indicate that the yield of the (d,p) reaction to the ^{7}Li ground state is not affected by the gating, whereas the yield to the ^{7}Li excited state is, as discussed above, affected. The gated and ungated spectra measured at 27.5 keV are shown in Figs. 5(a) and 5(b), respectively. The magnitude of the pile-up problem is evident in Fig. 5(a). The yield ratios measured between deuteron bombarding energies of 27.5 and 170 keV are plotted in Fig. 6. [Note that the reaction ratio is twice the yield ratio since there are two alpha particles of equal energy in the c.m. given off by the reaction ^{6}Li$(d,\alpha)^{4}$He.] Our yield ratios are compared to the ratios calculated from the published cross sections by Elwyn *et al.* [17]. In the energy range between 120 and 170 keV where our measurements overlap with those of Ref. [17], the yield ratios are in

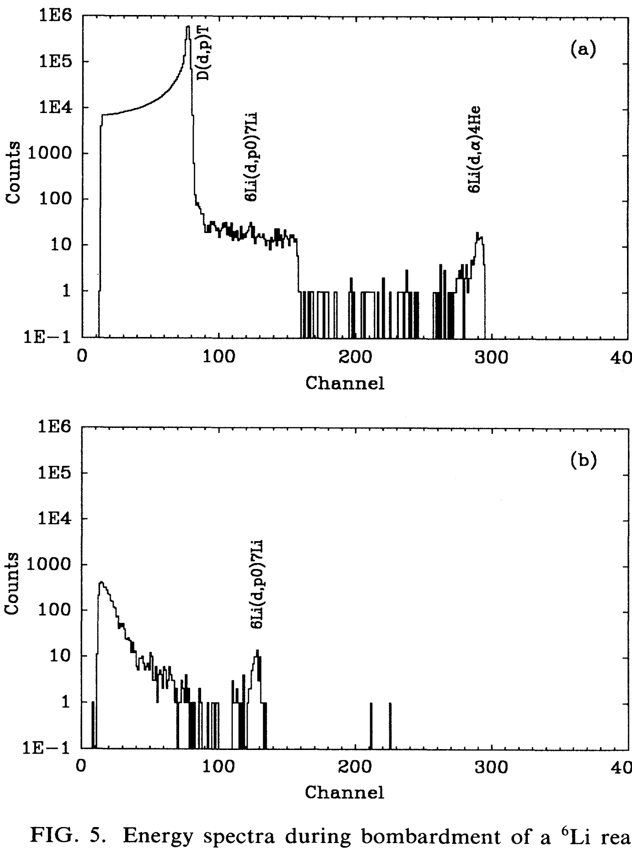

FIG. 5. Energy spectra during bombardment of a ^{6}Li reaction with 27.5-keV deuterons using a two-detector telescope. (a) Spectrum not gated on a signal from the back detector; (b) spectrum gated on a signal from the back detector. The magnitude of the pileup problem from the (d,d) reaction is evident in the continuum extending up to channel 160 (an energy of about 6 MeV) in (a).

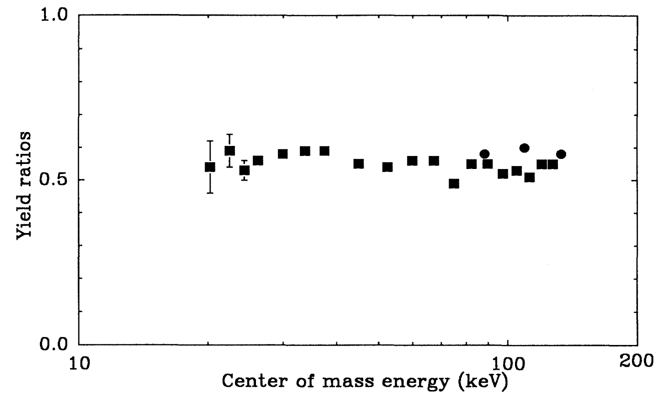

FIG. 6. Ratio of yields of ^{6}Li$(d,p)^{7}$Li and ^{6}Li$(d,\alpha)^{4}$He reactions. Solid squares are the present work; solid circles are from Ref. [17].

agreement. The angular distributions of both the (d,p) and (d,α) reactions reported in Ref. [17] are each isotropic at the 8% level at a deuteron bombarding energy of 118 keV, which is the lowest energy at which the angular distributions were reported in this reference. As in the case of the d-d reaction, we therefore interpret the yield ratios as measures of the total cross section ratios. Likewise, as with the d-d reaction, there is no evidence for an enhancement of the (d,p) reaction at low energies. The R-matrix formalism used in calculating the d-d reaction ratio has not been extended to the d-^{6}Li system.

In the case of the ^{10}B measurement, as with the ^{6}Li measurement, we compared the yield of the protons from the reaction ^{10}B$(d,p)^{11}$B to the alpha particles from the reaction ^{10}B$(d,\alpha)^{8}$Be. A detector telescope was likewise used, in this case to separate the protons to the ground

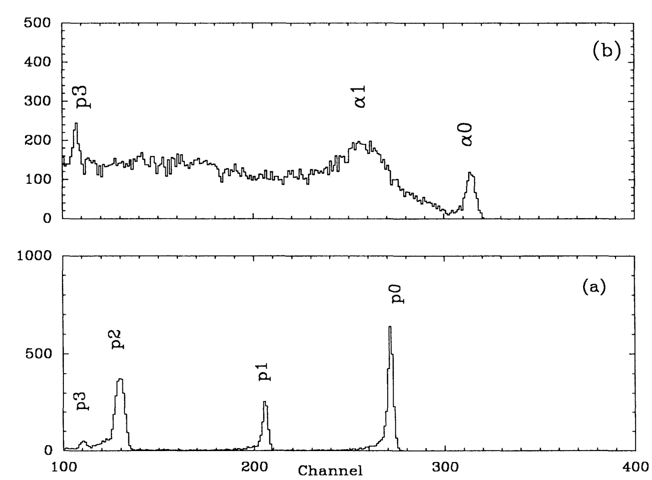

FIG. 7. Energy spectra measured during bombardment of ^{10}B with deuteron beams using a two-detector telescope. (a) Spectrum gated on signals from back detector of protons with energy greater than about 4.5 MeV; (b) spectrum vetoed by signals from the back detector of all alpha particles and of protons with energy less than about 4.5 MeV.

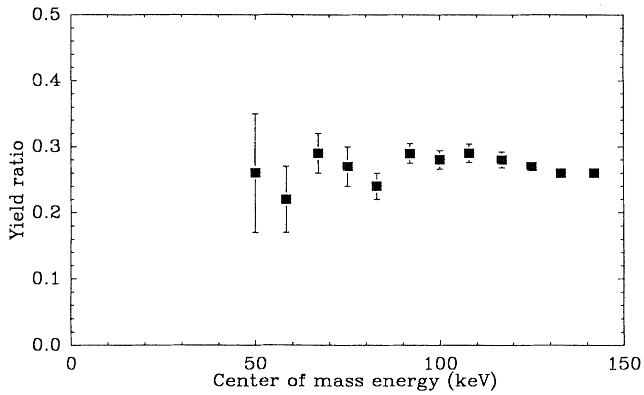

FIG. 8. Ratio of yield of protons to the ^{11}B ground state to the yield of alpha particles to the ^{8}Be first excited state.

state of ^{11}B from the alpha particles to the broad first excited state of ^{8}Be. The spectrum gated on a signal from the back detector recorded protons from the ground and first two excited states of ^{11}B. Because of straggling in the front detector, we were unable to measure reliably the yield to the third excited state of ^{11}B. The spectra of the alpha particles were measured by vetoing those summed signals for which there was a signal from the back detector. The proton and alpha spectra thus measured at a bombarding energy of 150 keV are shown in Figs. 7(a) and 7(b). The ratio of the yields to the ^{11}B ground state and the ^{8}Be first excited state are shown in Fig. 8. While there have been previous measurements of the ^{10}B$(d,p)^{11}$B reaction at energies overlapping with the present measurements [18], there are no reported measurements of the ^{10}B$(d,\alpha)^{8}$Be reaction at low energies. Unlike the ^{2}H and ^{6}Li results presented above, we are therefore unable to compare our measured yield ratios to independent cross section determinations at overlapping energies. The angular distributions for the ^{10}B$(d,p)^{11}$B reaction measured in Ref. [18] are isotropic at the 10% level at the lowest reported deuteron bombarding energy of 170 keV. We assume a similar level of isotropy for the ^{10}B$(d,\alpha)^{8}$Be reaction and interpret the yield ratios as measures of the total cross section ratios. As with the measurements on ^{2}H and ^{6}Li, Fig. 8 indicates no enhancement of the (d,p) reaction at low energies. The other ratios calculated by comparing the yields to the ^{11}B excited states and ^{8}Be ground state likewise were independent of energy.

III. CONCLUSIONS

In conclusion, there is no evidence for the enhanced yield of the (d,p) reaction relative to the (d,n) reaction for the d-d measurement or relative to the (d,α) reactions for the d-^{6}Li and d-^{10}B measurements. This lack of enhancement is, moreover, consistent, in the case of the d-d and d-^{6}Li reactions, with the second-order DWBA calculations of Koonin and Mukerjee [9] and in the case of the d-d reaction with the present R-matrix calculations. If the present results are to be viewed as consistent with the previous observations of an enhanced (d,p) to (d,n) ratio on ^{6}Li [6,7], then we must assume that the (d,n) reactions are being suppressed relative to both the (d,p) and (d,α) branches of the d-^{6}Li reaction. A number of future projects are suggested by the present results: measurement of the (d,p) and (d,α) yields on ^{9}Be to complement the above-mentioned $(d,p\gamma)/(d,n\gamma)$ ratio measurement [8], a measurement of the $(d,p\gamma)/(d,n\gamma)$ ratio on ^{10}B to complement the present measurement of the (d,p) and (d,α) yields, and R-matrix calculations of the d-^{6}Li and d-^{10}B reactions.

There is one area of investigation upon which the present results have a profound implication. This area includes the recent claims of significant heat production from deuterium-metal systems, cold nuclear fusion [19,20]. Remarkable to these claims is the absence or near absence of the production of energetic neutrons concurrent with the production of heat. Specifically, we would expect, based on the near equality of the (d,p) and (d,n) branches of the d-d reaction at low energies as indicated in Fig. 2, that if the d-d nuclear reactions were responsible for the reported heat production, then there would be about 10^{12} fast neutrons per watt of heat generated. We must conclude either that the (d,p) to (d,n) ratio for the d-d reaction changes by many orders of magnitude as the energy drops from a few keV to room temperature or that some other nuclear or non-nuclear reaction is responsible for the heat production. Similar conclusions obtain for the suggested possibility that the source of heat in electrochemical experiments involving LiOD electrolyte is the d-^{6}Li reactions [20]. If our observation of the lack of enhancement of the (d,p) to (d,α) branches of the d-^{6}Li reaction may be used to infer that there is a corresponding lack of suppression of the (d,n) branch of the reaction, then we could similarly conclude that the production of heat from the d-^{6}Li reactions must be accompanied by enormous fluxes of fast neutrons. On the other hand, if the earlier reports of the energy dependence of the $(d,n)/(d,p)$ ratios in the d-^{6}Li and d-^{9}Be reactions may be interpreted as suppression of the (d,n) reactions at very low energies, then the possibility of a near aneutronic d-^{6}Li reaction at very low energies should be considered.

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