Neutrons from Proton Bombardment of Lithium

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Relative cross sections for the production of neutrons from the proton bombardment of Li^7 have been measured from 3 to 13 MeV bombarding energy. Time-of-flight spectrometry was used to resolve the groups of neutrons going to various states in Be^7. The cross section for neutrons leaving Be^7 in its ground state shows a peak at 5 MeV and then decreases rapidly as the energy is increased, while the number of neutrons leaving Be^7 in its first excited state decreases fairly uniformly over the whole energy range. The angular distributions for both groups show considerable structure below 6 MeV but become isotropic at higher bombarding energies. From 9 to 13 MeV the zero-degree yield of neutrons leaving Be^7 in its excited state at 4.53 MeV was measured. At the higher proton energies several breakup reactions are energetically possible, but no continuum neutrons from them were observed at 10 MeV bombarding energy.

INTRODUCTION

THE $Li^7(p,n)Be^7$ reaction has long been one of the most widely used and studied sources of neutrons. For proton energies between 1.9 and 2.4 MeV, the neutrons produced are monoergic and have an energy ranging between 120 and 650 keV at zero degree At bombarding energies greater than 2.4 MeV, the residual nucleus Be^7 can be left in its first excited state at 0.43 MeV, as well as in the ground state. Consequently, two groups of neutrons are produced. As the bombarding energy is increased more neutron-producing reactions are energetically possible.[1]

If the $Li^7(p,n)Be^7$ reaction is to be used as a neutron source at bombarding energies above 2.4 MeV, an energy-sensitive detector must be used, or otherwise a correction for the presence of the low-energy neutrons must be made. The latter requires a knowledge of the relative cross sections of the various possible neutron-producing reactions. The purpose of this investigation was the determination of the relative cross sections for the different neutron groups, and the investigation of the utility of the $Li^7(p,n)Be^7$ reaction as a neutron source in the range of proton energies from 3 to 10 MeV. Information can also be obtained about the level structure of Be^8, the compound nucleus, and Be^7, the final nucleus.

Below 5 MeV bombarding energy several measurements of the $Li^7(p,n)$ cross section have been made. Taschek and Hemmendinger[2] have made an absolute measurement of the total reaction cross section and relative measurements of the differential cross section for bombarding energies below 2.55 MeV. Macklin and Gibbons[3] and Newson *et al.*[4] have measured the absolute total reaction cross section from threshold to 5.5 MeV bombarding energy and their values are in disagreement with those of Taschek and Hemmendinger. The total reaction cross section determined from an angular distribution of the absolute differential cross section at 2.27 MeV proton energy taken by Gabbard, Davis, and Bonner[5] is in agreement with the value obtained by Macklin and Gibbons. Gabbard, Davis, and Bonner also measured the absolute zero-degree yield up to 3.25 MeV and their data are consistent with the shape of the yield as reported by Bevington *et al.*[6] and Austin.[7]

Studies of the relative yields of the neutrons to the first two states of Be^7 have been made by Batchelor and Morrison[8] from 2.4 to 2.8 MeV proton energy and by Bevington *et al.* from 2.6 to 4.1 MeV. Cranberg[9] has determined the ratio of the number of neutrons to the first two states of Be^7 at several energies near 5 MeV and Bogdanov[10] has measured an angular distribution at 9.6 MeV in which the ground-state and first excited-state groups are resolved and neutrons to the second excited state of Be^7 are also seen. At 10.45 MeV Ajzenberg-Selove *et al.*[11] have made some measurements using emulsions in which neutrons to Be^7 (4.53 MeV) were seen. The only systematic measurements at high energies are some data on the total reaction cross section using activation techniques with stacked foils.[12]

EXPERIMENTAL METHOD

Targets of natural lithium metal were evaporated in place on a 0.7-mm-thick Ta backing which was insulated from the rest of the target assembly. No attempt

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[1] F. Ajzenberg-Selove and T. Lauritsen, Nucl. Phys. **11**, 37 (1959).
[2] R. Taschek and A. Hemmendinger, Phys. Rev. **74**, 373 (1948).
[3] R. L. Macklin and J. H. Gibbons, Phys. Rev. **109**, 105 (1958); J. H. Gibbons and R. L. Macklin, *ibid.* **114**, 571 (1959).
[4] H. W. Newson, R. M. Williamson, K. W. Jones, J. H. Gibbons, and H. Marshak, Phys. Rev. **108**, 1294 (1957).
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[6] P. R. Bevington, W. W. Roland, and H. W. Lewis, Phys. Rev. **121**, 871 (1961).
[7] S. M. Austin, Bull. Am. Phys. Soc. **7**, 269 (1962).
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[9] L. Cranberg, Los Alamos Scientific Laboratory Report LA-1654, 1954 (unpublished).
[10] G. F. Bogdanov, N. A. Vlasov, S. P. Kalinin, B. V. Rybakov, and V. A. Sidorov, Soviet J. At. Energy **3**, 987 (1959).
[11] F. Ajzenberg-Selove, C. F. Osgood, and C. P. Baker, Phys. Rev. **116**, 1521 (1959).
[12] S. P. Kalinin, A. A. Oglobin, and Yu. M. Petrov, Soviet J. At. Energy **2**, 193 (1957).

was made to measure absolute target thickness, but the relative thickness of several targets was determined by comparing the neutron yield from different targets at several bombarding energies and neutron emission angles.

Protons were accelerated by a tandem electrostatic accelerator and neutron spectra were obtained by the pulsed-beam time-of-flight method. The spectrometer is described in a paper by Lefevre *et al.*[13]

Neutrons were detected by observing proton recoils in an organic scintillator. In order to maintain a stable and reproducible detection efficiency, the proton recoil bias was set at a fixed fraction of the pulse height corresponding to the maximum energy Compton electrons from the Cs^{137} γ ray. The relative detector efficiency as a function of neutron energy was determined by observing the zero-degree yield of the monoergic neutrons from the $T(p,n)He^3$ reaction, for which the cross section has been measured by Perry *et al.*[14]

Background neutrons from the target assembly and beam stop were observed by bombarding only the Ta backing. Background not coming directly from the target was measured by placing a brass shadow bar between the Li target and the detector. The indirect background was found to be uncorrelated in time with the beam bursts, and was the only background present in the region of the spectrum near the peaks from the groups to the ground state and first excited states. The region of the spectrum near these peaks was therefore used as a measure of the flat background. The neutron group to Be^7(4.53 MeV), however, was superimposed on a continuum of neutrons from the Ta beam stop. This background was subtracted graphically.

Figure 1 shows the portions of three spectra containing the peaks from the neutrons to the ground state and first excited state of Be^7 with the flat background subtracted. At the lower energies the two peaks are sufficiently well separated to allow simple integration of the areas, but at higher energies the separation of the peaks was done graphically. The uncertainty introduced in the unfolding process depends on the degree of overlap but was always less than 15% of the area of the smaller peak.

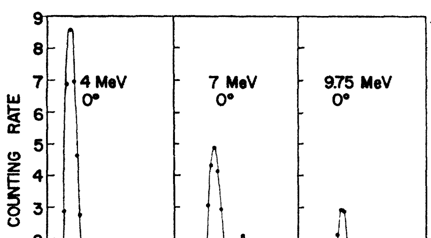

FIG. 1. Portions of time spectra taken at zero degree and three bombarding energies. The two peaks are caused by neutrons to the ground state and first excited state of Be^7. Time increases from left to right about 1 nsec/channel in the 4-MeV spectrum and 0.4 nsec/channel in the other two.

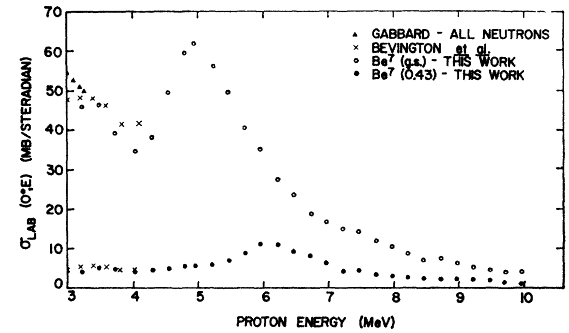

FIG. 2. Absolute zero degree differential cross sections. The upper curve is for neutrons to the ground state, the lower curve for neutrons to Be^7(0.43 MeV).

The measured detector efficiency and relative target thicknesses were used to convert all of the data to relative cross sections. The angular distributions at 4.0, 4.5, and 5.0 MeV proton energy were then integrated to obtain relative total reaction cross sections for the groups to the ground state and first excited state of Be^7. Since only neutrons to the ground and first excited states were present below 5 MeV, the sum of the two cross sections could be compared to the absolute measurement of Macklin and Gibbons.[3] The ratio obtained was used to normalize all of the present data to an absolute scale.

RESULTS

Figure 2 shows the zero-degree yield for both the group to the ground state and first excited state of Be^7. The results obtained by both Gabbard *et al.*[5] and Bevington *et al.*[6] are also plotted. Gabbard *et al.* performed an absolute measurement of the cross section for all neutrons. This cross section should be compared to the sum of the present measurements of the ground state and first excited state cross sections.

In the present measurements the uncertainties of the points below 6 MeV are about $\pm 7\%$ for the ground state group and $\pm 10\%$ for the first excited state group. Above 6 MeV the graphical unfolding of the peaks could introduce uncertainties as high as $\pm 10\%$ for the ground state and $\pm 15\%$ for the first excited state group. These uncertainties do not include the absolute error in the measurements of Macklin and Gibbons.[3]

Figure 3 shows the ratio of the measurements of the cross sections obtained in the present work and the

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[14] J. E. Perry, Jr., E. Haddad, R. L. Henkel, G. A. Jarvis, and R. J. Smith (unpublished), reported by J. D. Seagrave, at *Conference on Nuclear Forces and the Few Nucleon Problem, London, 1959*, edited by T. C. Griffith and E. A. Power (Pergamon Press, New York, 1960).

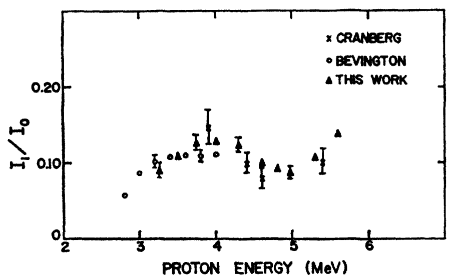

FIG. 3. The ratio of the number of neutrons going to the first excited state (I_1) to the number going to the ground state (I_0), at 0°.

ratio obtained by Bevington *et al.*[6] and Cranberg[9] for bombarding energies between 2.4 and 5.8 MeV. The agreement is good.

The angular distribution measurements were made at 15° intervals and the cross sections obtained were converted to the center of mass system. The center-of-mass angular distributions were then fitted with a Legendre polynomial series using an IBM 704 computer.[15]

Figures 4 and 5 show the center-of-mass angular distributions of neutrons to the ground state and first excited state. The curves shown are the Legendre polynomial fits to the center-of-mass cross sections and are extrapolations for $\cos\theta \leqslant -0.9$. Polynomials up to $n=4$ (i.e., $\cos^4\theta$) were used.

The total reaction cross sections for the two groups were obtained by integrating the Legendre polynomial fits. These cross sections are plotted along with the data of Bevington *et al.*[6] and Macklin and Gibbons[3] in Fig. 6. The measurements of Macklin and Gibbons[3] for all neutrons produced are shown as the upper curve. The solid triangles at 4.0, 4.5, and 5.0 MeV near the upper curve indicate the sum of the cross sections for the first and second groups from the present work. The lower two curves are the individual total reaction cross

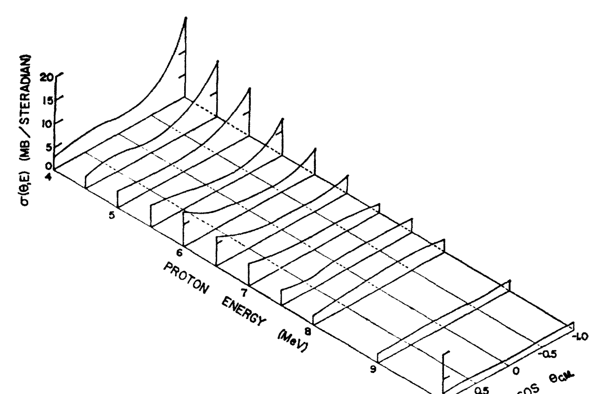

FIG. 4. Center-of-mass angular distributions of neutrons to the ground state of Be^7. The curves are the Legendre polynomial fits and are extrapolations for $\cos\theta \leqslant -0.9$.

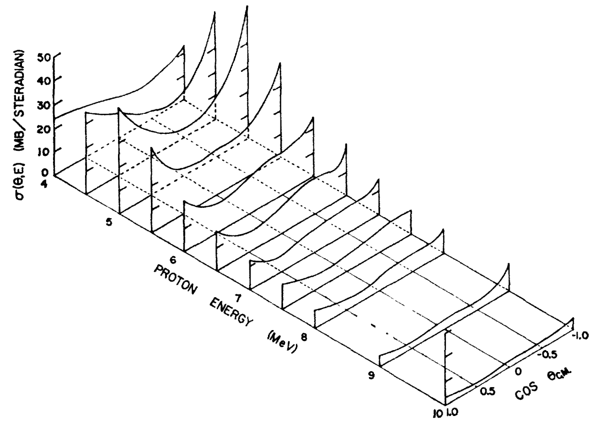

FIG. 5. Center-of-mass angular distributions of neutrons to the first excited state of Be^7. The curves are the Legendre polynomial fits to the data and are extrapolations for $\cos\theta \leqslant -0.9$.

sections for the ground-state and first excited-state groups. The lines are simply drawn through the data points. Uncertainties in the relative measurements are probably less than $\pm 10\%$ for the ground state group and $\pm 15\%$ for the first excited state group.

Above 7 MeV proton energy it is energetically possible to produce neutrons to the second excited state of Be^7.[1] The zero-degree differential cross section for these neutrons and for the unresolved groups to the ground state and first excited state of Be^7 were measured at 9, 10, 11, 12, and 13 MeV proton energy and are shown in Fig. 7.

Cross sections were obtained by normalizing the yield of the unresolved groups at 10 MeV to the sum of the cross sections for neutrons to the ground state and first excited state. The squares are the sum of the cross sections shown in Fig. 2 and the triangles represent the normalized yield of the unresolved lines. In computing the cross sections for the unresolved lines

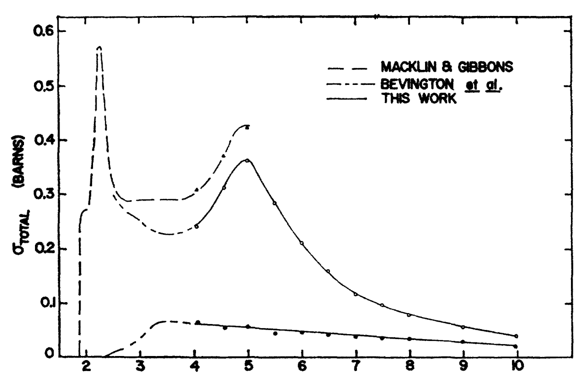

FIG. 6. Absolute total reaction cross sections. The dashed curve is the work of Macklin and Gibbons and the triangles are the sum of the cross sections obtained in the present work and normalized to the data of reference 3. The upper curve is the cross section for neutrons to the ground state and the lower curve for neutrons to the first excited state.

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[15] SHARE Program No. 424, Argonne National Laboratory E201.

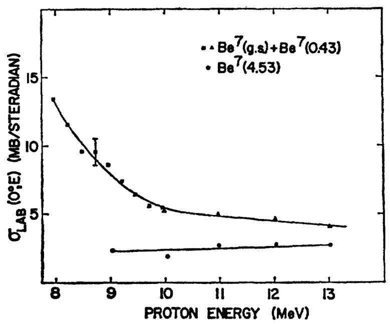

FIG. 7. High-energy zero degree yield of the unresolved groups to the ground state and first excited state (upper curve) and the group to the excited state of Be^7 at 4.53 MeV excitation.

the average detector efficiency for the two groups was used. The combined uncertainty from statistics, efficiency determination, background subtraction and normalization is about 20%.

At the higher energies three-body breakup reactions are also possible[1] but few neutrons from these reactions were seen, as illustrated in Fig. 8, which shows foreground and background spectra taken at about 10 MeV proton energy. A neutron energy scale determined from the known energy of the ground state group is shown along the top of the figure. Four points along the axis are marked by arrows. Arrow 1 shows the position of the peak from the unresolved ground state and the first excited state groups, 3 indicates the peak caused by neutrons going to Be^7(4.53 MeV), and arrows 2 and 4 indicate the positions of the maximum energy neutrons from the reactions $Li^7(p,n\alpha)He^3$ and $Li^7(p,np)Li^6$, respectively. The neutron yield per unit energy from these reactions is small in comparison to the monoergic groups. This conclusion is in disagreement with the results obtained by both Bogdanov *et al.*[10] and Ajzenberg-Selove *et al.*[11]

No neutrons attributable to the $Li^6(p,n)Be^6$ reaction were seen during this investigation. This is consistent with the fact that the isotopic abundance of Li^6 is only 7.5% and the cross section for the $Li^6(p,n)Be^6$ reaction is known to be small.[10,11]

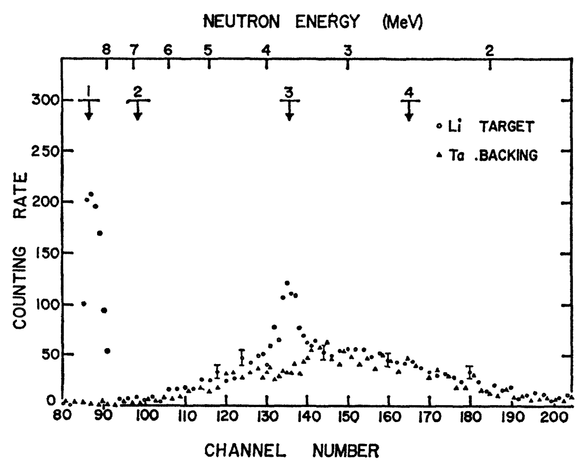

FIG. 8. Time spectra at 10 MeV. The arrows 1 to 4 are discussed in the text. The open circles are two-channel averages of the foreground and the triangles are the background taken by bombarding only the Ta backing.

DISCUSSION

Even for proton energies above the threshold for the second group, the $Li^7(p,n)Be^7$ reaction is a useful source of neutrons. The present data indicate that the cross section for the ground-state neutrons remains comparable to that at lower energies up to about 6 MeV, but decreases rapidly as the bombarding energy is increased above this point. Above 5.5 MeV the relative contribution of the neutrons to the first excited state, which is always less than 15% at lower energies, increases rapidly. It seems, therefore, that the $Li^7(p,n)Be^7$ reaction is of limited value as a neutron source for proton energies above 5.5 MeV.

Numerous attempts to fit the experimental data on $Li^7(p,n)Be^7$ assuming levels in Be^8 have been reported.[3,4,6,16,17] In the present case no attempt was made to fit all the data. Two peaks were seen in the measurements which could arise from levels in Be^8. The first appears only in the cross section for neutrons to the ground state at a bombarding energy of about 5 MeV. The peak occurs in both the zero-degree yield (Fig. 2) and the total reaction cross section (Fig. 6). A peak near this energy has been previously reported in the zero-degree yield[18] and in the total reaction cross section.[3] Both measurements included the contribution of all neutrons produced. Macklin and Gibbons[3] point out that the variation of the cross section near this peak requires the total angular momentum of the compound state to be greater than three units if only one level is present. An attempt was made to fit the peak in the total reaction cross section using the Breit-Wigner single-level formula assuming $J=3$. If the parity of the level is assumed even, the state can decay to the Be^7 and Li^7 ground state ($J^\pi=\frac{3}{2}^-$), with one unit of angular momentum. However, two more units of angular momentum are required for decay to the first excited states. Thus the decay to the first excited states should be inhibited by the decreased penetrability. This is in agreement with observation. Assuming $\gamma_n^2=\gamma_p^2$, the best fit was obtained with a resonant energy in the laboratory system of 4.9 MeV and a total width of 1.1 MeV.

A peak occurs in the zero-degree cross section for neutrons to Be^7(0.43 MeV) at a bombarding energy of about 6 MeV (Fig. 2), but is not seen in the total reaction cross section (Fig. 6).

The angular distributions vary considerably but

[16] S. M. Austin, S. E. Darden, A. Okazaki, and Z. Wilhelmi, Nucl. Phys. **22**, 451 (1961).
[17] R. K. Adair, Phys. Rev. **96**, 709 (1954).
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slowly with energy and the angular distribution of neutrons to both the ground state and first excited states becomes quite isotropic at higher energies. The possibility of a direct-interaction mechanism has been suggested at lower energies[19] but the angular distributions, especially at high energies, show none of the strong forward or backward peaking usually associated with direct processes.

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Overlap and Exchange Effects in Beta Decay*

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The change in nuclear charge by one unit in beta decay causes initial and final atomic states to overlap imperfectly. The effect of this imperfect overlap on the shape of allowed electron and positron emission spectra is calculated. The calculated change in the spectrum shape can be simulated by including the average excitation energy of the final atom in the energy balance. The inhibition, due to imperfect atomic overlap, of electron-capture rates, as well as total electron and positron-emission rates, is also determined. In all known cases, imperfect atomic overlap increases beta-decay lifetimes by at most a few percent and usually by an amount less than a few tenths of one percent. Antisymmetrization between decay and bound atomic electrons, in conjunction with the change in nuclear charge, gives rise to exchange effects in electron emission and electron capture. Due to exchange terms, the usual allowed electron spectrum is multiplied by a quantity that is of the order of $1-2Z^{-1}$ for energies less than the binding energy of a K electron in the initial atom. This exchange correction is negligible for higher energies of the emitted continuum electron. A simple approximate formula is derived that predicts the effect of exchange on L to K capture ratios; this formula predicts a 22% increase over the usual theoretical value for the L to K ratio of Ar^{37}. The Ar^{37} prediction is in excellent agreement with recent experiments and with a more complicated calculation by Odiot and Daudel. Exchange effects change total electron emission and electron capture rates by at most a few percent.

I. INTRODUCTION

HOW does the change in nuclear charge by one unit from initial to final atomic states affect beta decay? How much does the imperfect overlap of initial and final atomic states inhibit beta-decay rates? Does the possibility of exchange between bound and decay electrons significantly affect electron emission and electron capture probabilities? This paper is an attempt to answer the above questions.

Benoist-Gueutal[1] first emphasized that a correct specification of the initial and final states of a radioactive system must include a description of the atomic electrons. The overlap between initial and final atomic states is not equal to one since the initial and final states are eigenstates of zero-order Hamiltonians with different nuclear charges. Thus, one expects the theoretical decay rate to be decreased if atomic states are included in the description of the radioactive system. If this decrease were large, one would have to know the magnitude of the decrease in order to calculate nuclear matrix elements from experimentally determined parameters.

Benoist-Gueutal[1] estimated the effect of imperfect atomic overlap on the total electron capture rate of Be^7 by calculating the electron capture probability for various final atomic states. She concluded that the decrease in the total decay rate was between 0 and 30%; her calculation was limited by the lack of accurately known wave functions for an excited lithium atom. For heavier atoms, good atomic wave functions are even more difficult to obtain than for lithium. Moreover, the problem of evaluating the decay probability to all final states of a heavy atom is prohibitively complicated.

We calculate the effect of the change in nuclear charge by expanding the energy conserving delta function as a power series in the excitation energy of the final atom and then use closure to sum the beta-decay transition probability over all possible final atomic states. Explicit results are presented for allowed electron and positron emission and for allowed electron capture.

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