New Chain Barium-126—Cesium-126

M. I. KALKSTEIN AND J. M. HOLLANDER
Radiation Laboratory and Department of Chemistry, University of California, Berkeley, California
(Received July 9, 1954)

The neutron-deficient chain $Ba^{126}-Cs^{126}$ has been produced from nitrogen-ion bombardments of indium in the 60-inch cyclotron, by the reaction $In^{115}(N, 3n)$. Studies have been made of this new chain with a 50-channel scintillation spectrometer, a scintillation coincidence spectrometer, and a time-of-flight mass spectrograph. Element assignments and genetic relations have been verified chemically, and the mass number assigned with the isotope separator. Ba^{126} decays principally by orbital electron capture with a half-life of 96.5±2.0 minutes, and its daughter Cs^{126} is a positron emitter of 1.6±0.2 minute half-life and electron capture branching of 18±4 percent. The decay of Cs^{126} proceeds by allowed transitions, ~62 percent to the ground state of Xe^{126} and ~38 percent to the first excited state at 385 kev. The positron spectrum has a maximum energy of 3.8±0.4 Mev. On the basis of its decay properties, Cs^{126} appears to have a ground-state configuration of (1+).

INTRODUCTION

A PROGRAM has been underway in this laboratory to study the nuclear reactions of cyclotron-accelerated heavy ions and to utilize these ions in the synthesis of new isotopes of the heaviest elements.[1–4] In principle at least, heavy-ion bombardment should also find application to the study of neutron deficient isotopes of medium and low Z elements, because it provides an almost unique way to prepare these isotopes free of heavier ones. To date, the major limitation on this application has been the low beam intensities available, which has severely restricted the feasibility of spectroscopic investigations. Nonetheless, it was felt that the possible advantages of using heavy ions for the production of neutron deficient nuclei should be examined.

In the work reported here, a new neutron-deficient chain, $_{56}Ba^{126}-_{55}Cs^{126}$, has been produced by the irradiation of indium ($Z=49$) targets in the nitrogen ion beam of the Crocker 60-inch cyclotron. The genetic relationship between these nuclides has been verified by means of chemical "milking" experiments, and their radiations have been studied with NaI and anthracene scintillation detectors coupled to a 50-channel differential analyzer. Some coincidence studies have also been made by using the scintillation coincidence spectrometer. Assignment to the mass-126 chain has been accomplished by means of a time-of-flight mass spectrometer. Barium-126 is found to have a half-life of 96.5 minutes, and to decay by orbital electron capture. Its daughter, $_{55}Cs^{126}$, decays with a 1.6-minute half-life, chiefly by the emission of positrons of maximum energy about 3.8 Mev.

A 12-minute barium activity, which may be Ba^{127} or possibly Ba^{124}, has also been observed in some short nitrogen ion bombardments of indium. This will be reported only briefly, since no detailed study of this nuclide has been undertaken.

[1] Miller, Hamilton, Putnam, Haymond, and Rossi, Phys. Rev. **80**, 486 (1950).
[2] Ghiorso, Thompson, Street, and Seaborg, Phys. Rev. **81**, 154 (1951).
[3] Rossi, Jones, Hollander, and Hamilton, Phys. Rev. **93**, 256 (1954).
[4] Ghiorso, Rossi, Harvey, and Thompson, Phys. Rev. **93**, 257 (1954).

BOMBARDMENT CONDITIONS

In most of the bombardments, targets of indium oxide (In_2O_3)[5] weighing ~100 mg were mounted in 0.25- or 0.5-mil platinum envelopes for use with the internal heavy ion probe. A few runs were also made using targets of indium metal melted to the copper backing plate of a water-cooled probe. Irradiations were made at a dee radius of 23.5 inches. The energy of the nitrogen ion beam has a continuous distribution with a maximum of ~140 Mev.[3] Because of the thick targets used, it was not possible to monitor the beam during bombardments, but nitrogen ion currents measured separately have averaged around 0.1 microampere. Bombardment times were either short (two to five minutes) or of the order of an hour, depending upon the half-lives of the barium activities being studied.

CHEMICAL PURIFICATIONS

A pure barium fraction was prepared initially from the indium metal or oxide targets by a procedure involving the precipitation of $Ba(NO_3)_2$ with fuming HNO_3 followed by precipitation of $BaCl_2$ with "ether-HCl" reagent.

The rapid "milkings" of cesium from solutions containing the $Ba^{126}-Cs^{126}$ equilibrium mixture made use of sodium cobaltinitrite as a precipitant for cesium. The $Cs_3Co(NO_2)_6$ precipitate was quickly dissolved in fuming HNO_3, and several $Ba(NO_3)_2$ scavenges performed. The entire procedure consumed less than three minutes.

EXPERIMENTAL RESULTS

Half-Lives

The barium chemical fractions from the nitrogen ion bombardments of indium exhibited, after the decay of an

[5] The In_2O_3 powder was prepared from Indium Corporation of America 99.97 percent metal by HNO_3 solution, hydroxide precipitation, and finally ignition to the oxide.

initial ~12-minute component, a single activity which followed a period of 96.5±2.0 minutes through as many as ten half-lives without deviation. When the chemical purification was performed more than an hour after the end of bombardment, only the 97-minute activity appeared in the barium fraction, proving its chemical identity as an isotope of barium.

By making a series of rapid chemical separations of cesium away from a solution containing the 97-minute barium activity, a cesium daughter has been isolated whose half-life was measured from five milkings to be 1.5±0.3 minutes.

Mass Assignment

With the aid of M. C. Michel, the mass assignment of the new Ba–Cs chain has been made on a medium-resolution, high-transmission (~20 percent) isotope separator employing the time-of-flight principle.[6]

The sample, containing ~10^5 dis/min of the 97-minute barium in equilibrium with its cesium daughter, was deposited onto a tungsten filament in the form of ~50 μg $BaSO_4$. Because of the weakness of this source, it did not seem feasible to attempt collection of barium ions, consequently advantage was taken of the much greater ionization efficiency of the short-lived cesium daughter. By "flashing" the ion source at a temperature sufficient to ionize cesium but not vaporize barium it was possible to collect cesium ions alone, accomplishing simultaneously both chemical and mass separation. By so retaining barium on the filament, the experiment could be repeated several times with essentially no loss in yield of cesium at the collector end.

After flashing the source for ~10 seconds, the vacuum system was quickly let down to air, and the collector plate removed for counting. This procedure consumed about two minutes. Activity was found to collect only at the mass-126 position, and this decayed with a half-life of 1.6±0.2 minutes. The new chain is thus identified as Ba^{126}–Cs^{126}, produced by the reaction $In^{115}(N, 3n)Ba^{126}$.

Gamma Spectra

Nitrogen ion bombardments of around an hour's duration yielded less than 10^6 disintegrations per minute of the Ba^{126}–Cs^{126} chain, so it was not possible to utilize high resolution magnetic spectrometers in an examination of the beta and gamma spectra. Consequently, we have made use principally of a NaI(Tl) scintillation detector coupled to a 50-channel differential analyzer designed by Ghiorso and Larsh.[7]

TABLE I. Positron and x-ray abundances.

	Ba^{126}+Cs^{126} equilibrium	− Cs^{126}	= Ba^{126}
X-rays	~1.2	0.18	~1
β^+	~0.8	0.82	~0
Total	2.0	1.0	1.0

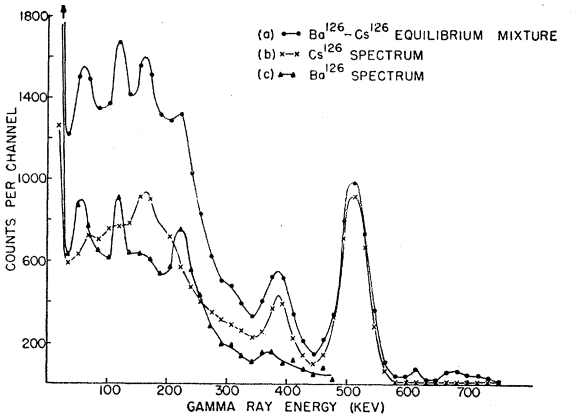

FIG. 1. (a) Gamma spectrum of Ba^{126}–Cs^{126} equilibrium mixture. Ordinate at 30-kev channel equals 6100 counts. (b) Gamma spectrum of Cs^{126}. (c) Gamma spectrum of Ba^{126}, obtained by subtracting *b* from *a*. [The 510-kev peak of Fig. 1(b) has been normalized to the 510-kev peak of Fig. 1(a).]

In these experiments, the gamma-ray spectrum of the barium-cesium equilibrium mixture was first studied, and it was verified that all peaks decayed with a 97-minute half-life. Then a pure sample of the 1.5-minute cesium daughter was prepared by the "milking" procedure described above, and its gamma spectrum was examined. The spectrum due to the 97-minute barium parent was then obtained by making the appropriate subtraction of the cesium spectrum from that of the equilibrium mixture. Figure 1 shows the spectrum between 0 and 750 kev of the equilibrium mixture, the pure Cs^{126}, and the subtracted spectrum obtained for the Ba^{126} parent. It is noted, from the observed relative abundances and from the equilibrium conditions prevailing, that the x-rays (at ~30 kev) belong mainly to the Ba^{126} while the annihilation radiation accompanies the decay of Cs^{126}.

The gamma spectrum of Cs^{126} is quite simple; other than annihilation radiation and ~30-kev x-rays only a gamma ray of 385±5 kev is in evidence. The peak (in Fig. 1(b)) at ~170 kev is due to 180° backscattered annihilation radiation, and that at ~75 kev is the lead fluorescent x-ray generated in the counter shield. From the observed[8] photopeak abundances, it is calculated that Cs^{126} decays principally by positron emission, with an electron capture branching of 18±4 percent. The 385-kev gamma ray appears in 38±7 percent of the disintegrations. No gamma ray of energy greater than 600 kev is observed with an abundance higher than 5 percent of the disintegrations.

The gamma spectrum of Ba^{126}, obtained by sub-

[6] W. E. Glenn, University of California Radiation Laboratory Report UCRL-1628, January, 1952 (unpublished); M. C. Michel and D. H. Templeton, Phys. Rev. 93, 1422 (1954).

[7] A. Ghiorso and A. E. Larsh (to be published).

[8] The calibration of the scintillation spectrometer is described in the Appendix.

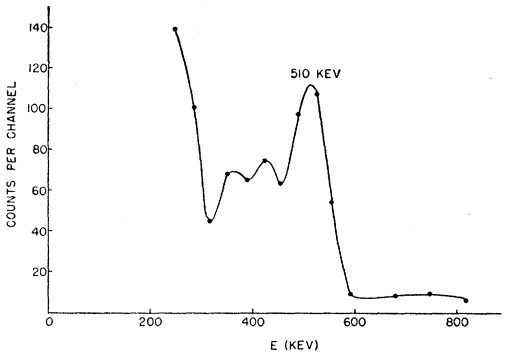

FIG. 2. Gamma-gamma coincidence counting rate of 385-kev γ rays as a function of gate setting.

traction, is more complex. Gamma rays of 225±10 kev and 700±30 kev are definitely observed, with relative intensities 3:1. A weak gamma ray at ~900 kev is probable. The peak at 115 kev appears to be largely backscattered 225-kev radiation, since its relative intensity varies with position of the source in the counter shield. X-rays are the predominant feature of the decay of Ba^{126}, but because of the uncertainties accompanying the subtraction of the gamma spectra and because of the unknown contribution of x-rays from internal conversion of the gamma-rays, a ratio of electron capture to positron emission in Ba^{126} cannot be given with confidence.

The direct decay of the gamma-ray lines from the 1.6-minute Cs^{126} was observed by performing the fast cesium "milking" from the parent, placing the separated cesium fraction into the scintillation spectrometer, and photographing the dials of the spectrometer with a Leica camera at half-minute intervals following each "milking." By subtraction of the dial readings of one negative from those of the next negative, the spectrum during each half-minute period could be examined. In this way, decay curves have been constructed for the integrated photopeaks of the annihilation gamma ray, the 385-kev gamma ray, and for the K x-rays; all exhibit a half-life of 1.6±0.2 minutes, through five half-lives.

Beta Spectrum

The end point of the positron spectrum was measured in a sample of the Ba—Cs equilibrium mixture by means of an anthracene scintillation crystal coupled to the 50-channel analyzer, and also by absorption in beryllium. Both methods gave a value of 3.8±0.4 Mev. As noted above, these positrons are associated with the decay of Cs^{126}.

Coincidence Measurements

Several experiments were done in which the Ba^{126}—Cs^{126} chain was examined with a coincidence scintillation spectrometer designed by J. O. Rasmussen. The apparatus consists of a NaI or anthracene gate detector coupled to a single-channel analyzer, and a NaI coincidence detector coupled to the 50-channel analyzer. In these experiments, the resolving time was $\approx 8\times10^{-8}$ second. The anthracene detector in the gate circuit was employed when positron-gamma coincidences were being examined; for gamma-gamma coincidences NaI crystals were used in both circuits. The two crystals were shielded from each other by a 2-inch lead brick and beryllium absorbers were placed in front of the gamma-ray detector to absorb electrons.

Although the short half-life of the activity under study prevented the accumulation of sufficient events per experiment to give good statistics, several qualitative results were obtained by sweeping the gate channel over the gamma spectrum in increments of ~35 kev and recording the coincident gamma spectrum at 90° on the 50-channel analyzer. Figure 2 is a plot of the coincidence rate at ~385 kev on the 50-channel analyzer as a function of gate setting, showing a maximum rate at a gate setting of ~510 kev. Figure 3 similarly indicates the coincidence rate at ~510 kev, showing a maximum at a gate setting of ~385 kev. These experiments verified that the 385-kev gamma ray follows positron decay in Cs^{126}. They also serve to point out that there is no gamma ray of ~500 kev following positron decay of Cs^{126}, since few 500-500 gamma-gamma coincidences are observed. An upper limit of 4 percent per disintegration can be set for a possible gamma ray at ~500 kev. This fact could not be deduced from the gross gamma spectrum alone, because of interference from the prominent 510-kev annihilation peak. Even those few 510γ-510γ coincidences that are observed at 90° might be explained as being due to annihilation-annihilation coincidences, since an appreciable fraction of positrons annihilate in flight, giving annihilation quanta at angles other than 180° with respect to each other.

In scanning high energies on the gate, a maximum of gamma-gamma coincidences in the region of 225 kev was obtained when the gate was set at ~700 kev, offering some evidence that the 225-kev gamma ray in

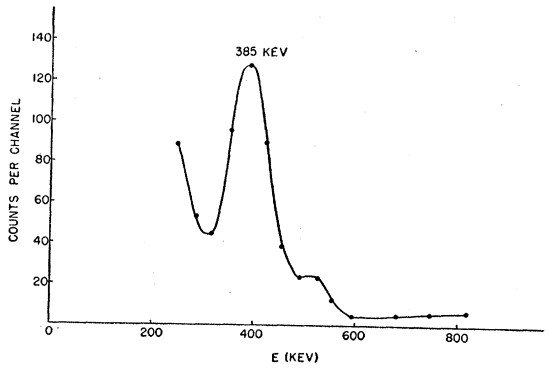

FIG. 3. Gamma-gamma coincidence counting rate of 510-kev γ rays as a function of gate setting.

Ba^{126} may be in coincidence with the 700-kev gamma ray.

Using the anthracene crystal in the gate circuit, positron-gamma coincidences were run at 180°. With a gate setting $\ll 3$ Mev, coincidences were observed at ~ 390 kev and ~ 510 kev. At a gate setting of ~ 3.5 Mev, the ~ 390-kev peak was not observed, but the positron-annihilation coincidences remained. Since no gamma ray other than that at 385 kev is observed in the decay of Cs^{126}, it is inferred from the above that a ground-state positron transition takes place between Cs^{126} and Xe^{126}.

CONCLUSIONS

From the observed positron and gamma-ray abundances in Cs^{126}, some conclusions can be drawn about its decay scheme. Logft values of 4.7 and 4.8 are calculated[9] for the positron decay to the ground and first excited states of Xe^{126}, respectively. These are both within the range of values expected for "allowed" transitions, and suggest that the configuration of the ground state of Cs^{126} is (1+), since (0+) and (2+) doubtless represent the configurations of the first two states in Xe^{126}. The energy of the first excited state of Xe^{126} found in this work, 385±5 kev, is in agreement with that reported from studies of the beta decay of I^{126} by Perlman and Friedlander[10] (386±2 kev) and by Mitchell *et al.*[11] (395±5 kev).

In a further study of the radiations of I^{126}, Perlman and Welker[12] have reported a second excited state in Xe^{126} at ~ 870 kev, with a configuration also (2+). It is of interest to note that in the present study this state was not observed from the decay of Cs^{126}. A limit of 4 percent for the population of this state was set by the absence of coincidences between the 510-kev gamma ray and a gamma of 485 kev which would be expected to de-excite an 870-kev state if the spin sequence were (2+)→(2+)→(0+) as reported by Perlman and Welker. If the same selection rules are operative for decay of Cs^{126} to all these states, one would expect an 870 kev (2+) state in Xe^{126} to be populated from Cs^{126} to the extent of perhaps 20 percent, which is substantially above our experimental upper limit. A cross-over gamma ray of 870 kev was also not observed in the gamma spectrum of Cs^{126}, but its expected abundance might be as low as the upper limit of 5 percent set from the gross gamma spectrum.

Because only a rough measurement of the positron energy of Cs^{126} could be obtained in these experiments, the disintegration energy cannot be specified precisely. However, the value of 4.8±0.4 Mev follows the general trend of the energy surface shown by the "beta-systematics" curves of Way and Wood.[13]

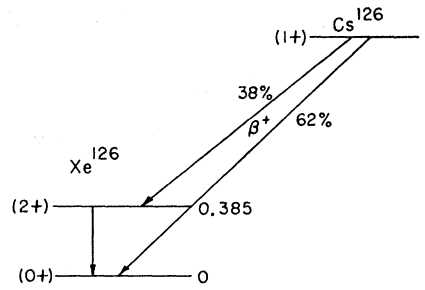

FIG. 4. Decay scheme of Cs^{126}. (β^+ should read β^++EC.)

The electron capture branching of Cs^{126}, 18±4 percent, compares with a figure of ~ 16 percent predicted from the theoretical curves of Feenberg and Trigg[14] for allowed transitions with $Z=54$ and positron abundances and energies as reported here.

A decay scheme consistent with these data is presented in Fig. 4.

THE 12-MINUTE BARIUM ACTIVITY

In several short nitrogen-ion bombardments of indium, an approximately 12-minute activity appeared in the barium fractions in lower yield than the 97-minute Ba^{126}. This activity may be Ba^{127}, produced by the In^{115}(N, 2n) reaction. Barium-127 has been observed by Lindner and Osborne,[15] who have shown that it decays with a 12-minute half-life into 6.3-hour Cs^{127}. Although no 6-hour "tail" has been seen in the Geiger counter decay curves of our barium fractions, the low β+/EC branching ratio (<10 percent) of Cs^{127} reported by Mathur and Hyde[16] would indicate that this isotope could have a counting efficiency low enough to have escaped detection in our gross decay curves.

On the other hand, it is possible that the half-life of the unknown isotope Ba^{124} could be of the order of 12 minutes, if it decays by an allowed transition. Such a half-life would require a disintegration energy of ~ 3 Mev, which is not an unreasonable value.[13] Barium-124 would be produced in these experiments by In^{113}(N, 3n) and In^{115}(N, 5n) reactions.

ACKNOWLEDGMENTS

We would like to thank A. Ghiorso, Professor J. G. Hamilton, W. B. Jones, and G. B. Rossi, and the crew of the 60-inch cyclotron for their constant efforts to develop and improve the heavy ion beams, and for their assistance to us in making these bombardments. It is also a pleasure to acknowledge helpful discussions with Professor J. O. Rasmussen and Dr. E. K. Hyde,

[9] S. A. Moszkowski, Phys. Rev. **82**, 35 (1951).
[10] M. L. Perlman and G. Friedlander, Phys. Rev. **82**, 449 (1951).
[11] Mitchell, Mei, Maienschein, and Peacock, Phys. Rev. **76**, 1450 (1949).
[12] M. L. Perlman and J. Welker, Phys. Rev. **95**, 133 (1954).
[13] K. Way and M. Wood, Phys. Rev. **94**, 119 (1954).
[14] E. Feenberg and G. Trigg, Revs. Modern Phys. **22**, 406 (1950).
[15] M. Lindner and R. Osborne, July, 1952 (private communication).
[16] H. Mathur and E. K. Hyde, Phys. Rev. **95**, 708 (1954).

and to thank Dr. M. C. Michel and Dr. F. A. Asaro for their help with several of the experiments.

APPENDIX

Calibration of the NaI(Tl) Spectrometer

A $1\frac{1}{2}$-inch diameter by 1-inch thick thallium activated sodium iodide crystal was used in conjunction with the 50-channel analyzer. A diffuse reflector of MgO surrounded the crystal, and a thin beryllium window separated it from the sample. The crystal was bonded to a thin quartz disk which in turn was optically coupled to a Dumont 6292 photomultiplier tube by a layer of mineral oil. With this arrangement, a resolution of 8 percent was obtained at 661 kev. An aluminum-lined lead shield housed the detector assembly, and samples were mounted in a standard G-M tube five position shelf holder. In these experiments, most measurements were made at a distance of ~1.5 inches from the crystal, which represented a geometry of 5.1 percent (as determined with the 60-kev gamma ray of a standardized Am^{241} sample).

Energy calibration of the spectrometer was made during each experiment, with the following gamma-ray standards: Am^{241} (60 kev), U^{235} (143, 184 kev), Au^{198} (412 kev), Cs^{137} (32, 661 kev), and Na^{22} (510 kev, 1.28 Mev). In addition, the annihilation radiation of Cs^{128} served as an internal standard, so that any shift in the energy scale could be observed and corrected.

The following counting efficiencies were used to calculate the relative intensities of the various gamma rays:

X-rays (Cs, Xe)	100 percent
225 kev	63
385 kev	28
510 kev	17
700 kev	11

We have determined the counting efficiencies of gamma rays at 412 kev (Au^{198}), 510 kev (Na^{22}), 661 kev (Cs^{137}), 1.28 Mev (Na^{22}), and 1.33 Mev (Co^{60}), using a 4-π counter to determine the absolute disintegration rates of the standard samples. The results of these measurements were all within 5 percent of the efficiencies obtained from the data of Bell *et al.*[17,18] for the experimental conditions used here. Because of this agreement, the Oak Ridge curves were used to determine the efficiencies at other gamma-ray energies.

The observed x-ray intensities were corrected for fluorescence yield and absorption by comparison with the x-rays from a Cs^{137} standard examined under identical experimental conditions.

[17] Bell, Heath, and Davis, Oak Ridge National Laboratory Report ORNL-1415, 1952 (unpublished).
[18] Bell, Hughes, Davis, Jordan, and Randall, Oak Ridge National Laboratory Report ORNL-1415, 1952 (unpublished).

PHYSICAL REVIEW VOLUME 96, NUMBER 3 NOVEMBER 1, 1954

Neutral Pion-Deuteron Production in 400-Mev n-p Collisions*

R. A. SCHLUTER
Institute for Nuclear Studies, The University of Chicago, Chicago, Illinois
(Received June 17, 1954)

The reaction $n+p\rightarrow d+\pi^0$ has been studied in a high-pressure hydrogen-filled diffusion cloud chamber. A total of 102 deuterons were identified by a technique of ionization measurement in conjunction with momentum measured in the 10 500-gauss magnetic field. From the laboratory deuteron angle and momentum, the pion angle and the incident neutron energy can be deduced with good resolution, provided the pion is emitted backward in the center-of-mass system. Proton recoils were observed simultaneously, from which the incident neutron flux and energy spectrum were determined. The energy spectrum is centered at 400 Mev with a spread of ±25 Mev. From 52 events occurring in the backward direction the angular distribution of pion emission was found to be consistent with $(0.28_{-0.14}{}^{+0.26}+\cos^2\bar{\psi}_\pi)$, where the limits include 70 percent of the probability and $\bar{\psi}_\pi$ is the pion angle in the center-of-mass system. The observed excitation function and total cross section determined from 60 events is consistent with $(0.47\pm0.08)\eta^3$ millibarn, where η is the center-of-mass pion momentum in rest mass units. These results agree, within the accuracy of the measurements, with the prediction of charge independence that the ratio of this reaction to $p+p\rightarrow d+\pi^+$ should be $\frac{1}{2}$.

Eight cases of internal conversion of the neutral pion were seen, four accompanied by deuterons and four by protons, the later giving evidence on neutral pion creation with unbound final nucleons.

I. INTRODUCTION

THE experiment reported here is a study of the reaction $n+p\rightarrow d+\pi^0$ in a high-pressure diffusion cloud chamber, with 400±25-Mev neutrons produced by the University of Chicago 170-inch synchrocyclotron. The total cross section as well as the angular distribution were measured. Since deuterons rather than neutral pions were detected, there is in this experiment no ambiguity due to the presence of pions produced with unbound final nucleons. The general plan of the experiment is this: deuterons produced in collisions of 400-Mev neutrons with protons in a high-pressure hydrogen

* Research supported by a joint program of the U. S. Office of Naval Research and the U. S. Atomic Energy Commission.