$(n,2n)$ Excitation Functions of Several Nuclei from 12.0 to 19.8 Mev*

R. J. PRESTWOOD AND B. P. BAYHURST
Los Alamos Scientific Laboratory, University of California, Los Alamos, New Mexico
(Received October 3, 1960)

$(n,2n)$ excitation functions have been obtained for Sc^{45}, Ti^{46}, Ni^{58}, Cu^{65}, Ge^{70}, As^{75}, Rb^{85}, Rb^{87}, Sr^{84}, Y^{89}, Zr^{90}, Nb^{93}, Ag^{107}, In^{115}, Sn^{112}, Sb^{121}, Sb^{123}, Ta^{181}, Au^{197}, Tl^{203}, and Th^{232} at incident neutron energies of 12.00 to 19.76 Mev. The target elements were exposed to neutrons from the $T(d,n)He^4$ reaction in Zr—T and T_2 gas targets, and the products were measured by radiochemical methods.

I. INTRODUCTION

OVER the span of time since tritium became available as a target material for particle accelerators, a number of measurements have been made of $(n,2n)$ cross sections with D-T neutrons.[1–19] With the exception of a few recent investigations, these measurements have been made with the D-T neutrons as a single group, without energy differentiation, i.e., they have yielded for each of the target nuclei a single value in the vicinity of 14.5 Mev. However, even with incident deuteron energies in the range of 0.3 Mev, characteristic of the Cockcroft-Walton accelerators commonly used for the D-T reaction, the neutron energy span from 0° to 180° is almost 2 Mev.

The work reported here was undertaken to measure accurately the energy dependence of the $(n,2n)$ cross sections of a variety of target nuclei by exploitation of the energy vs angle variation at a Cockcroft-Walton accelerator and at a 6-Mev Van de Graaff accelerator.

II. EXPERIMENTAL

Two neutron energy spectra were employed. The source of 13.34- to 14.95-Mev neutrons was the $T(d,n)He^4$ reaction produced by 350-kev deuterons on a Zr-T target[20] at the Los Alamos Cockcroft-Walton accelerator. The energies were obtained by exposure of samples from 0° to 165° with respect to the deuteron beam. The samples to be irradiated ($\frac{3}{8}$-in. diameter foils of metal or of a compound of the element rolled with polythene) were positioned on an accurately molded thin plastic hemispherical shell of 5-cm radius which had been scribed with lines representing various angles with respect to the deuteron beam (Fig. 1). Aluminum monitor foils, also of $\frac{3}{8}$-in. diameter, were placed under each sample. In addition, an aluminum foil was placed at 90° and 10.0±0.1 cm distance to serve as a primary monitor. The Na^{24} produced in these aluminum foils from the $Al^{27}(n,\alpha)Na^{24}$ reaction was counted on a β-proportional counter. The activity on the primary monitor foil was related to the flux number obtained from an α counter monitor. The α counter measures the α particles from the $T(d,n)He^4$ reaction and has been calibrated in terms of an accurately known fraction of the number of such events in the target. The uncertainty (4%) in number of D-T events is mainly in the measurement of the area of the diaphragm which determines the fraction of total α particles accepted by the counter. The flux at any sample position on the hemisphere can then be calculated from a ratio of activity of an aluminum foil at the sample position to the activity in the primary monitor foil. This technique reduced the error caused by inaccurate measurement of distance from the target or beam wandering to less than 3% and eliminated error due to Zr-T target shadowing. A correction was made for the $Al^{27}(n,\alpha)Na^{24}$ excitation function which was carefully measured several times in this energy range. The integrated neutron flux was usually 10^{12} cm^{-2} at our sample position in an approximately 4-hour run.

The neutrons of energies of 12.0, 16.5, 18.0, and 19.8 Mev were produced by deuteron bombardment of T_2 gas targets at the large electrostatic generator at Los Alamos (Fig. 2). Neutrons produced in the forward direction were used in the runs at 16.5, 18.0, and 19.8 Mev; the 12.0-Mev neutrons were obtained in the 19.8-Mev run by using the neutrons in the backward direction. The $U^{238}(n,\text{fission})$ process was used to monitor neutron flux. A carefully calibrated fission counter in conjunction with a U^{238} foil was placed

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

[1] J. E. Brolley, J. L. Fowler, and L. K. Schlacks, Phys. Rev. **88**, 618 (1952).
[2] J. D. Knight, R. K. Smith, and B. Warren, Phys. Rev. **112**, 259 (1958).
[3] A. Poularikas and R. W. Fink, Phys. Rev. **115**, 989 (1959).
[4] E. B. Paul and R. L. Clarke, Can. J. Phys. **31**, 267 (1953).
[5] C. S. Khuranu and H. S. Hans, Nuclear Phys. **13**, 88 (1959).
[6] L. A. Rayburn, Bull. Am. Phys. Soc. **3**, 337, 365 (1959).
[7] S. G. Forbes, Phys. Rev. **88**, 1309 (1952).
[8] H. A. Tewes, A. A. Caretto, and A. E. Miller, Bull. Am. Phys. Soc. **4**, 445 (1959).
[9] B. L. Cohen, Phys. Rev. **81**, 184 (1951).
[10] D. R. Koehler and W. L. Alford, Phys. Rev. **119**, 311 (1960).
[11] V. J. Ashby, H. C. Catron, L. L. Newkirk, and C. J. Taylor, Phys. Rev. **111**, 616 (1958).
[12] J. J. Dudley and C. M. Class, Phys. Rev. **94**, 807 (A) (1954).
[13] H. C. Martin and B. C. Diven, Phys. Rev. **86**, 565 (1952).
[14] J. L. Fowler and J. M. Slye, Jr., Phys. Rev. **77**, 787 (1950).
[15] H. C. Martin and R. J. Taschek, Phys. Rev. **89**, 1302 (1953).
[16] A. V. Cohen and P. H. White, Nuclear Phys. **1**, 73 (1956).
[17] J. M. Ferguson and W. E. Thompson, Phys. Rev. **118**, 228 (1960).
[18] R. G. Wille and R. W. Fink, Phys. Rev. **118**, 242 (1960).
[19] L. P. O'Conner and J. L. Perkin, J. Inorg. Nuclear Chem. **13**, 5 (1960).
[20] E. R. Graves, A. A. Rodriquez, M. Goldblatt, and D. I. Meyer, Rev. Sci. Instr. **20**, 579 (1949).

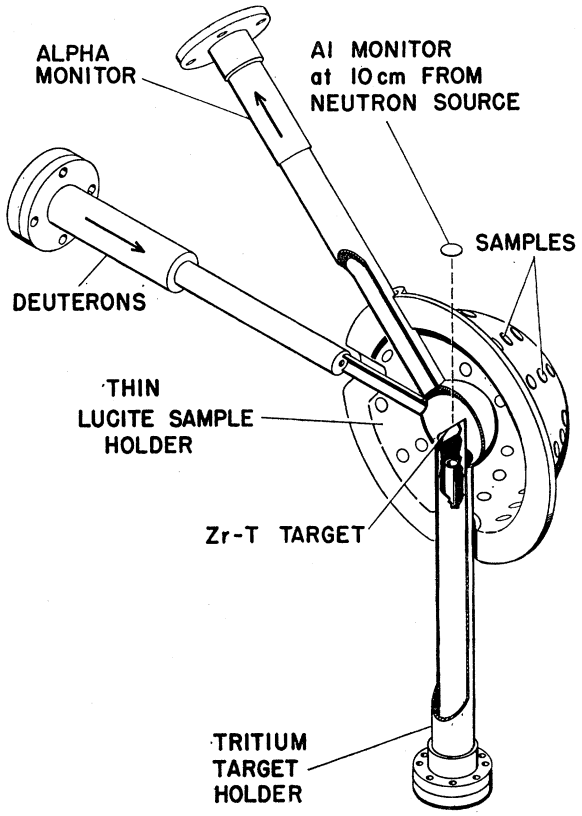

FIG. 1. Cockcroft-Walton target and sample assembly.

between the target and sample. This monitoring system was checked by radiochemical analyses of the fission product Mo^{99} in U^{238} foils irradiated at 7 and 8 Mev. At these energies both the ratio of counts per minute of Mo^{99} to total fissions[2] and the fission cross section[21] are known.

U^{238} foils were also used for determining flux attenuation throughout the stack of target foils. The Mo^{99} activity in U^{238} foils, which were alternated with target foils (Fig. 2), was related to the Mo^{99} activity in a U^{238} foil placed immediately behind the fission counter. In all cases corrections were made for fission fragment loss. The integrated neutron flux varied from 5×10^{10} to 10^{12} cm^{-2} at our sample positions for 2 to 8 hour runs.

The irradiated samples were prepared for measurement by radiochemical techniques, involving the addition of appropriate standardized carrier for (n,p) and (n,α) products, chemical separations, decontamination, and purification of each product.[22] The mounting and counting techniques and measurement of absolute disintegration rates were done by the method of Bayhurst and Prestwood.[23] Only relative numbers were obtainable in cases of insufficient knowledge of the decay scheme and in some gamma counted samples where the counting efficiency was not known.

III. DISCUSSION OF RESULTS

The experimental cross sections and energy data are summarized in Table I. Errors were grouped in two classes: (1) absolute errors, including error in the calibration of the α counter or the fission counter monitoring systems, half-life, decay scheme (which would effect counting efficiency), and any error in the $U^{238}(n,\text{fission})$ cross section; (2) relative errors, including any α or fission counting variations during a run, chemical or weighing errors, calculations of counting efficiencies for thick or uneven samples, and statistical counting errors (low count rates). The latter also includes any errors in resolving counting data representing more than one activity and calculations of decay during irradiations. Both of these latter calculations were made on an IBM Model 704 data processing

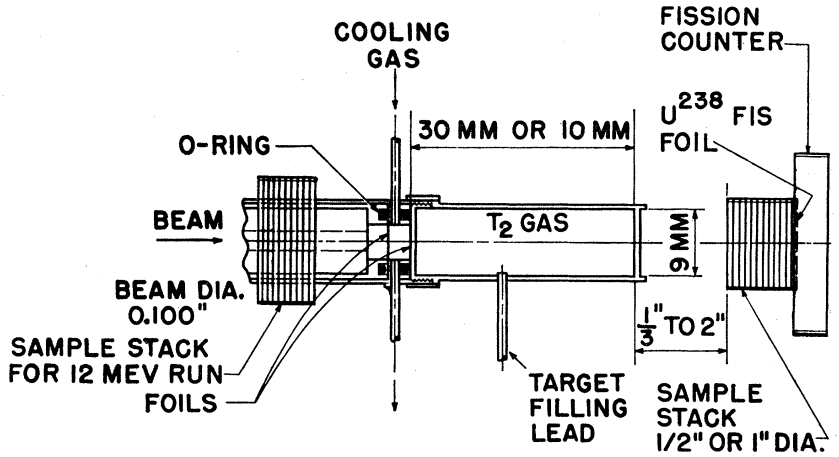

FIG. 2. Van de Graaff target and sample assembly.

[21] R. K. Smith, R. L. Henkel, and R. A. Nobles, Bull. Am. Phys. Soc. 2, 196 (1957).
[22] In most cases the (n,p) and (n,α) products were also studied and the data will be presented in a later paper.
[23] B. P. Bayhurst and R. J. Prestwood, Nucleonics 17, No. 3, 82 (1959).

TABLE I. Summary of $(n,2n)$ cross-section values.[a]

Reaction	Product half-life[e]	Particle counted[d] / $\bar{E}_n$ (Mev)	12[b]	13.33±0.23	13.40±0.20	13.52±0.15	13.69±0.10	13.88±0.10	14.01±0.10	14.09±0.10
$Sc^{45}(n,2n)Sc^{44m}$	2.44 days	β	2.5±0.1		74.0±2.4		91.9±3.0		107.4±3.5	116.2±3.7
$Sc^{45}(n,2n)Sc^{44g}$	3.92 hr	β	6.5±0.3		87.0±3.3		111.9±4.2		138.5±5.2	150.0±5.7
$Sc^{45}(n,2n)Sc^{44\,total}$			9.0±0.3		161.0±4.1		203.8±5.2		245.9±6.3	266.2±6.8
$Ti^{46}(n,2n)Ti^{45}$	3.09 hr	β		<1.0	1.0±0.5		2.2±1	7.0±1		13.0±3
$Ni^{58}(n,2n)Ni^{57}$	36 hr	β				13.9±0.7		21.4±1.1		23.5±1.2
$Cu^{65}(n,2n)Cu^{64}$	12.68 hr	β	504±25	778±31	771±31	814±33	830±33	879±35	879±35	906±36
$Ge^{70}(n,2n)Ge^{69}$	40.4 hr	β			379±11		461±14	509±15	508±15	
$As^{75}(n,2n)As^{74}$	17.8 days	β	515±21		907±36		991±40		1070±43	
$Rb^{85}(n,2n)Rb^{84e}$	33.0 days	β	726±73		1099±55		1356±68			1447±72
$Rb^{87}(n,2n)Rb^{86f}$	18.66 days	β	700±70		1056±53		1107±55			1170±59
$Sr^{84}(n,2n)Sr^{83g}$	33 hr	β		71.7±3.6	84.3±4.2		111.1±5.6	115.9±5.8		142.4±7.1
$Y^{89}(n,2n)Y^{88}$	104 days	γ^h	2.88±0.14	8.20±0.41	8.74±0.44	8.63±0.43	10.54±0.69	10.70±0.54	12.01±0.60	11.96±0.60
$Zr^{90}(n,2n)Zr^{89i}$	79.3 hr	β	28±1.4		398±12	457±14	527±16	585±18	604±18	623±19
$Nb^{93}(n,2n)Nb^{92j}$	10.1 days	γ^h	15.62±0.78		17.99±0.90		18.01±0.90		18.43±0.92	
$Ag^{107}(n,2n)Ag^{106k}$	8.2 days	γ^h	6.34±0.32	8.65±0.43	9.30±0.47	9.23±0.46	9.78±0.49	9.71±0.49		10.49±0.52
$Cd^{116}(n,2n)Cd^{115m}$	43 days	β	721±72		780±78		795±80		840±84	769±77
$Cd^{116}(n,2n)Cd^{115g}$	53 hr	β	721±72		821±82		830±83		850±85	835±84
$Cd^{116}(n,2n)Cd^{115\,total}$			1442±102		1601±113		1625±115		1690±119	1604±114
$In^{115}(n,2n)In^{114m}$	50.0 days	β	1132±57	1471±74	1428±71	1506±75	1468±73	1523±76	1557±78	
$Sn^{112}(n,2n)Sn^{111}(In^{111})$	2.81 days	γ^h	1.53±0.15		3.67±0.18	3.91±0.20	4.03±0.20			4.72±0.24
$Sb^{121}(n,2n)Sb^{120k}$	5.8 days	γ^h	798±40	1116±56	1077±54	1098±55	1114±56		1106±55	1272±64
$Sb^{123}(n,2n)Sb^{122f}$	2.80 days	γ^h	1089±109	1191±60	1145±57	1161±58	1269±63		1279±64	1335±67
$Ta^{181}(n,2n)Ta^{180m}$	8.15 hr	β	1116±56			1126±56		1118±56		1132±57
$Au^{197}(n,2n)Au^{196m}$	9.83 hr	γ	68.3±3.4		118.8±5.9		128.1±6.4		134.3±6.7	
$Au^{197}(n,2n)Au^{196\,total}$	6.06 days	γ	2081±104		2330±117		2369±118		2403±120	
$Tl^{203}(n,2n)Tl^{202}$	12.5 days	γ	867±43			1268±63		1302±65		1302±65
$Tl^{203}(n,3n)Tl^{201}$	72 hr	γ								
$Th^{232}(n,2n)Th^{231}$	25.64 hr	γ^l	1760±176	1610±161	1680±168	1635±164	1630±163	1580±158		1560±156

Reaction	Product half-life[e]	Particle counted[d] / $\bar{E}_n$ (Mev)	14.31±0.13	14.50±0.20	14.68±0.26	14.81±0.31	14.93±0.36	16.50±0.30	17.95±0.32	19.76±0.43
$Sc^{45}(n,2n)Sc^{44m}$	2.44 days	β	127.3±4.1	134.3±4.3	144.7±4.7			170.3±9.2	189.6±10.2	185.7±10.0
$Sc^{45}(n,2n)Sc^{44g}$	3.92 hr	β	169.0±6.4	181.4±6.9	204.3±7.7			283.7±13.6	318.3±15.2	322.2±15.4
$Sc^{45}(n,2n)Sc^{44\,total}$			296.3±7.6	315.7±8.1	349.0±9.0			454.0±16.4	507.9±18.3	507.9±18.3
$Ti^{46}(n,2n)Ti^{45}$	3.09 hr	β		28.0±3			44.5±3	145.0±7	185.0±9	233.9±12
$Ni^{58}(n,2n)Ni^{57}$	36 hr	β	31.1±1.6	34.3±1.7		39.3±2.0		53.3±4.3	67.6±3.4	77.4±3.9
$Cu^{65}(n,2n)Cu^{64}$	12.68 hr	β	892±36	937±37	953±38	968±39	975±39	997±50	930±47	986±49
$Ge^{70}(n,2n)Ge^{69}$	40.4 hr	β	607±18	621±19	664±20	716±21	681±20	843±42	895±45	941±47
$As^{75}(n,2n)As^{74}$	17.8 days	β	1113±45		1149±46		1123±45	1210±61	1143±57	1176±59
$Rb^{85}(n,2n)Rb^{84e}$	33.0 days	β		1498±75	1520±76	1530±77			1659±166	1767±177
$Rb^{87}(n,2n)Rb^{86f}$	18.66 days	β		1211±61	1194±60	1191±60			1141±114	1131±113
$Sr^{84}(n,2n)Sr^{83g}$	33 hr	β	149.9±7.5	171.7±8.6	176.8±8.8		180.6±9.0	230±23	351±35	295±30
$Y^{89}(n,2n)Y^{88}$	104 days	γ^h	13.17±0.66	13.46±1.35	13.74±0.69	14.16±0.71	13.89±0.70	16.35±1.6	16.23±1.6	17.36±1.7
$Zr^{90}(n,2n)Zr^{89i}$	79.3 hr	β	716±21	768±23	822±25	838±25	856±26	1115±56	1173±59	1169±58
$Nb^{93}(n,2n)Nb^{92j}$	10.1 days	γ^h	18.32±0.92	18.46±0.92	18.20±0.91			15.82±1.6	13.91±1.4	12.06±1.2
$Ag^{107}(n,2n)Ag^{106k}$	8.2 days	γ^h	10.73±0.54	10.87±0.54	10.72±0.54	10.90±0.55	11.07±0.55	11.76±1.2	12.50±1.3	12.02±1.2
$Cd^{116}(n,2n)Cd^{115m}$	43 days	β	885±89	808±81	825±83	807±81	836±84	822±82	666±67	566±57
$Cd^{116}(n,2n)Cd^{115g}$	53 hr	β	863±86	826±83	817±82	781±78	798±80	658±66	488±49	355±36
$Cd^{116}(n,2n)Cd^{115\,total}$			1748±124	1634±116	1642±117	1588±112	1634±116	1480±105	1154±83	921±67
$In^{115}(n,2n)In^{114m}$	50.0 days	β	1500±75	1539±77		1585±79	1503±75	1534±153	1310±131	1360±136
$Sn^{112}(n,2n)Sn^{111}(In^{111})$	2.81 days	γ^h	5.07±0.25	5.16±0.26	5.36±0.27	5.34±0.27	5.65±0.28	5.86±0.6	6.65±0.7	6.44±0.6
$Sb^{121}(n,2n)Sb^{120k}$	5.8 days	γ^h	1128±56	1309±65	1260±63	1297±65	1214±61	1339±134	1259±126	1426±143
$Sb^{123}(n,2n)Sb^{122f}$	2.80 days	γ^h	1263±63	1343±67	1256±63	1281±64	1194±60	1447±145	1266±127	1080±108
$Ta^{181}(n,2n)Ta^{180m}$	8.15 hr	β	1115±56	1116±56	1087±54			701±70	394±39	341±34
$Au^{197}(n,2n)Au^{196m}$	9.83 hr	γ	137.1±6.9	142.1±7.1		145.1±7.3		166.8±17	164.1±16	136.2±14
$Au^{197}(n,2n)Au^{196\,total}$	6.06 days	γ	2420±121	2403±120		2356±118		1860±186	1398±140	1111±111
$Tl^{203}(n,2n)Tl^{202}$	12.5 days	γ	1329±66	1321±66	1305±65			1134±113	943±94	818±82
$Tl^{203}(n,3n)Tl^{201}$	72 hr	γ						<25	124±12	280±28
$Th^{232}(n,2n)Th^{231}$	25.64 hr	γ^l	1520±152	1440±144	1400±140	1280±128	1255±126	<480		

[a] Cross sections in millibarns except where noted.
[b] The average neutron energies and their standard deviations at various foil positions for the 12-Mev run are: Sc, Sb=11.97±0.08; Cu, Cd, Sn=12.06±0.11; Ta, Au, Th, Zr, As=12.13±0.15; In, Ag, Nb, Rb=12.20±0.18; Sr, Y, Tl=12.29±0.21.
[c] Values from D. Strominger, J. M. Hollander, and G. T. Seaborg, Revs. Modern Phys. **30**, 585 (1958) except for Cu^{64}, As^{74}, Au^{196m}, Au^{196g}, and Tl^{202} which are from unpublished work by the authors.
[d] Decay schemes of all except Au^{196m} and Ti^{45} from footnote c; Au^{196m} decay scheme from R. Van Lieshout, R. K. Girgis, R. A. Ricci, A. H. Wapstra, and C. Ythier, Physica **25**, 703 (1959); Ti^{45} decay scheme from K. Way, R. W. King, C. L. McGinnis, and R. Van Lieshout, *Nuclear Level Schemes, A=40–A=92*, Atomic Energy Commission Report TID-5300 (U. S. Government Printing Office, Washington, D. C., 1955).
[e] Includes the fraction of Rb^{84m} that decays by isomeric transition.
[f] Includes isomeric transition from upper state.
[g] Cross section calculated assumes 100% β^+ emission.
[h] Relative numbers taken from gross gamma counts whose efficiencies were not known.
[i] Includes 93% of Zr^{89m}.
[j] Only 10.1-day Nb^{92} observed.
[k] Cross section for listed state only.
[l] Gamma counts normalized to one absolute β count.

machines.[24] The relative errors of the electrostatic generator runs would also include any error arising from degradation of neutron energy through the stack of foils.

The standard deviations of the neutron energies of the Cockcroft-Walton accelerator were obtained by taking $\frac{1}{3}$ of the energy spread at various laboratory angles for thick-target yields[25] except in the cases where the neutron energy spread due to deuteron scattering or sample geometry exceeded the above. Those of the Van de Graaff accelerator were obtained by geometrical relationships of the target and samples.[26] The standard deviations of cross sections used in Table I were those which gave the best fit of the data to a theoretical excitation function based on the statistical model concept of the compound nucleus and reflect the relative consistency of the data for a given function. The absolute errors are difficult to evaluate, but probably the greatest sources of error are in the decay schemes and in estimating γ-counting efficiencies.

Note added in proof. Linear plots of individual curves of the data in Table I will be available in Los Alamos Scientific Laboratory Rept. LA 2493 and will be available at a later date from the Office of Technical Services, U. S. Department of Commerce, Washington 25, D. C.

ACKNOWLEDGMENTS

The authors wish to express their appreciation to Dr. G. A. Cowan and Dr. J. D. Knight for their interest and advice, to R. K. Smith for the Van de Graaff irradiations, to the personnel at the Cockcroft-Walton accelerator, and to T. T. Shull for some of the titanium chemistry.

[24] Code for resolving counting data from more than one activity is a least squares solution, and the Los Alamos Scientific Laboratory designation for this code is J-11-GPF-002. The code for decay corrections during irradiation is designated as J-11-GFW-010 and evaluates the expression

$$(A/A_0)=\sum_{i=1}^{N} F_i[1-\exp(-\lambda\tau_i)\exp(-\lambda T_i)],$$

where A = number of atoms remaining at $T_{(0)}$, A_0 = total number of atoms produced during the N irradiation intervals, T_i = time from any interval to end of irradiation (T_0), and F_i = fraction of the isotope produced during the interval τ_i whose integrated irradiation level is P_i and is given by $F_i=P_i/\Sigma_i P_i$.

[25] Tabulated in Los Alamos Scientific Laboratory Report LAMS-2162 (unpublished).

[26] L. Blumberg and S. I. Schlesinger, Atomic Energy Commission Report AECU-3118, May, 1956 (unpublished).

PHYSICAL REVIEW VOLUME 121, NUMBER 5 MARCH 1, 1961

Nuclear Structure Studies in the Tin Isotopes with (d,p) and (d,t) Reactions*

BERNARD L. COHEN AND ROBERT E. PRICE
Radiation Laboratory, University of Pittsburgh, Pittsburgh, Pennsylvania

(Received July 22, 1960)

The neutron single-particle states in the odd isotopes of tin are identified by (d,p) angular distribution studies. The cross sections for exciting these states by (d,p) and (d,t) reactions are measured, and the results are analyzed to give values of V_j^2 (in Kisslinger-Sorenson notation), the fraction by which each of the single-particle states is full, for each subshell in each isotope. These are used to calculate ϵ_j, the unperturbed single-particle energies; the results are reasonably consistent. If the observed energies of single-particle states are used to predict the V_j, the agreement is generally good, but some discrepancies are noted and an explanation is offered.

Other weakly excited states are found in the region of the single-particle states. At higher excitation energies, several rather sharp levels are strongly excited in (d,p) reactions. Their energy, cross section, and regularities among the isotopes suggests that these are single-particle levels from the next major shell ($82<N\leq126$); however, their angular distributions cannot be used for identification as they are the same for all levels in this region and show little structure. This last fact is not easily explained.

Some of the two quasi-particle excitation states in the even isotopes of Sn are identified and the apparent pairing energy is thereby measured; it is surprisingly found to vary rapidly with mass number. Spectra from (d,p) and (d,t) reactions in isotonic pairs Cd^{114}-Sn^{116} and Cd^{116}-Sn^{118} are compared to show that the single-particle neutron states are much more radically affected by the addition of two protons than by the addition of two neutrons, contrary to the usual assumption in shell model theory.

Q values for (d,p) and (d,t) reactions on the major isotopes of tin are measured.

I. INTRODUCTION AND THEORY

FROM the simple shell-model viewpoint, the structure of nuclei with more than three particles (or holes) outside of closed shells is extremely complicated, and good theoretical calculations are essentially impossible. However, in the pairing theory approximation,[1,2] the structure becomes simple again provided, at least, that either the neutrons or protons have a closed shell. Such a situation arises among the isotopes

* Work done in the Sarah Mellon Scaife Radiation Laboratory and assisted by the National Science Foundation and the joint program of the Office of Naval Research and the U. S. Atomic Energy Commission.

[1] L. S. Kisslinger and R. A. Sorenson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. (to be published).
[2] M. Baranger (to be published).