

The configuration  $4f^{11}6s^2$  should show isotope splittings roughly twice that of configurations with only a single  $6s$  valence electron. The splittings are somewhat less than twice the normal value because of the mutual screening of the  $s$ -electrons. The screening effects cannot be estimated very well in this case because of the lack of an analysis, but in the case Tl II, Crawford and Schawlow<sup>6</sup> estimated that the shift in the  $6s^2$  configuration should be about 7 percent less than twice the shift of a single  $6s$  electron. It is possible that the line  $\lambda 4888$  arises from the double electron transition  $4f^{11}5d6p$  to  $4f^{11}6s^2$ , although the mean value of the shift ( $72.9 \text{ cm}^{-1}$ ) is somewhat less than 93 percent of twice the value of the normal positive shifts ( $45.5 \text{ cm}^{-1}$ ). The transition  $4f^{11}6s6p$  to  $4f^{11}6s^2$  should show shifts which are about 14 percent less than the other positive shifts ( $4f^{11}5d6p$  to  $4f^{11}5d6s$  and  $4f^{12}6p$  to  $4f^{12}6s$ ). The shifts around either  $0.033 \text{ cm}^{-1}$  or  $0.042 \text{ cm}^{-1}$  (see Fig. 1) may arise from this transition.

In summary, the negative shifts almost certainly arise from the double electron transition  $4f^{11}6s6p$  to  $4f^{11}5d^2$ . Most of the positive shifts probably arise from the transitions  $4f^{11}5d6p$  to  $4f^{11}5d6s$  and  $4f^{12}6p$  to  $4f^{12}6s$ . The shifts arising from the transition  $4f^{11}6s6p$  to  $4f^{11}6s^2$  are probably smaller than the normal positive shifts. The double electron transition  $4f^{11}5d6p$  to  $4f^{11}6s^2$  should give a shift nearly twice the normal positive shift, and one line of this transition may have been observed.

It is interesting to note in which regions most of these various shifts occur. The normal positive shifts are distributed quite uniformly throughout the region from 6600 to 4500A, but the negative shifts appear more abundantly between 5125 and 4250A. Between 6600 and 4300A, the number of strong lines which show structure exceeds those which do not, but below 4000A there are very few lines which show structure at all, although some of the most intense lines in the erbium spectrum are to be found in this region.

## Neutrons from the Disintegration of Phosphorus by Deuterons\*

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The neutron spectrum at  $0^\circ$  and  $90^\circ$  from the  $P^{31}(d,n)S^{32}$  reaction has been investigated using nuclear emulsions. The energies of the excited states in  $S^{32}$  are 0.5, 1.5, 2.2, 2.7, 3.3, 3.8, 4.1, 4.5, 4.9, 5.2, and 5.5-Mev, each with an uncertainty of about  $\pm 0.1$ -Mev.

### INTRODUCTION

TWO previous measurements of the energies of the excited states in  $S^{32}$  have been reported. Dicke and Marshall<sup>1</sup> measured the inelastic scattering of protons by sulfur giving excited states in  $S^{32}$  at 2.25 Mev and 4.34 Mev. Grace, Beghian, Preston, and Halban<sup>2</sup> using absorption techniques found a 2.35-Mev gamma-ray when fast neutrons were inelastically scattered from  $S^{32}$ . In this work the energy levels in  $S^{32}$  have been found using the  $P^{31}(d,n)S^{32}$  reaction and the photographic plate technique.

### EXPERIMENTAL PROCEDURE

A thick target of 99+ percent pure red phosphorus<sup>3</sup> was bombarded by 1.60-Mev deuterons from the Bartol electrostatic generator. The resultant neutron spectrum was found by observing the proton recoils in  $100\mu$

Eastman NTA nuclear emulsions mounted 3 cm from the target at angles of  $0^\circ$  and  $90^\circ$  to the direction of the beam. The proton recoils were observed only in a narrow band in the front half of the plate such that an acceptance angle of  $\pm 12^\circ$  could be used. A total of 1700 tracks were measured on the  $0^\circ$  plates and 1100 tracks on the  $90^\circ$  plates. In order to improve the statistical accuracy, 3.19 times as many fields of view were observed above 3.0 Mev on the  $90^\circ$  plates as were observed in the same range when tracks of all energies were counted; on the  $0^\circ$  plates, the factor was 1.79 for the same energy range. The data, corrected for the variation of neutron-proton scattering cross section,<sup>4</sup> geometry,<sup>5</sup> and the above mentioned statistical weights, are shown in Fig. 1 and Fig. 2. The vertical line through each of the observed points has a total length equal to twice the estimated standard deviation of the measurement.

The only likely impurity in the phosphorus was boron since the red phosphorus was prepared from distilled

\* Assisted by the joint program of the ONR and AEC.

<sup>1</sup> R. H. Dicke and J. Marshall, Jr., Phys. Rev. **63**, 86 (1943).

<sup>2</sup> Grace, Beghian, Preston, and Halban, Phys. Rev. **82**, 969 (1951).

<sup>3</sup> Courtesy of J. H. Walthall, Tennessee Valley Authority, Wilson Dam, Alabama. No chemical analysis supplied; however, a statement as to the method of purification was supplied.

<sup>4</sup> R. K. Adair, Revs. Modern Phys. **22**, 249 (1950).

<sup>5</sup> H. T. Richards, Phys. Rev. **59**, 796 (1941).

white phosphorus in a boro-silicate glass apparatus.<sup>3</sup> A normal boron ( $d,n$ ) neutron spectrum previously had been run in this laboratory using  $E_D = 1.30$ -Mev and had shown a rather high intensity of tracks at 10-Mev.<sup>6</sup> The relative intensity of the remaining groups was no more than five times as intense as this group. Therefore, from the fact that no tracks corresponding to the 10-Mev group appeared on the present plates, it may be concluded that the boron reaction contributes a negligible distortion to the observed spectrum. Since the lowest energy peak in the spectrum occurs at about the position of  $C^{12}(d,n)N^{13}$  neutrons the phosphorus spectrum will be distorted by the presence of the carbon surface contaminant. A distortion due to D-D neutrons is not likely with our bombarding currents (0.5–1.0  $\mu a$ ).

In order to reduce the carbon contaminant three fresh targets were used during the 0.05-coulomb bombardment. No background run was made to check other possible contaminants. However, a similar experiment<sup>7</sup> in which  $Si^{28}$  was bombarded by deuterons for a similar length of time showed that no appreciable number of neutrons in the energy range 2.0 Mev to 9.0 Mev was present. The analysis of the  $P^{31}(d,n)S^{32}$  spectrum was made assuming that carbon was the only possible contaminant.

#### ANALYSIS OF DATA

As may be seen from Figs. 1 and 2, the statistical accuracy of each of the  $0^\circ$  and  $90^\circ$  determinations of the proton recoil spectrum is rather poor. In order to extract the maximum amount of information from the data as a whole, two things need to be done. First, a method of plotting all of the data as a unit must be found. Secondly, one must know the theoretical line shape of the recoil spectrum of a single neutron group. Strictly speaking, one cannot combine the  $0^\circ$  and  $90^\circ$  data on to a single energy scale using any transformation, since the shape of the proton recoil range distribution, corresponding to a monoenergetic neutron, changes as the neutron energy is varied. However, since  $P^{31}$  is relatively heavy, an artifice may be employed to effect the transformation approximately. It can be shown that, for the  $P^{31}(d,n)S^{32}$  reaction, the errors involved in treating any particular range as an actually existing energy of a neutron through the range energy relation for photographic emulsions leads to discrepancies in the center-of-mass distribution that are well within the uncertainties allowed by the statistical accuracy involved. The result then is that, if the range scales of the  $0^\circ$  and  $90^\circ$  data are transformed to energy scales by the range-energy relation,<sup>8</sup> the corresponding proton and hence neutron energies may each be transformed to the center-of-mass energy scale of the

<sup>6</sup> C. P. Swann (private communication).

<sup>7</sup> Mandeville, Swann, Chatterjee, and Van Patter, Phys. Rev. 85, 193 (1952).

<sup>8</sup> Richards, Johnson, Ajzenberg, and Laubenstein, Phys. Rev. 83, 994 (1951).

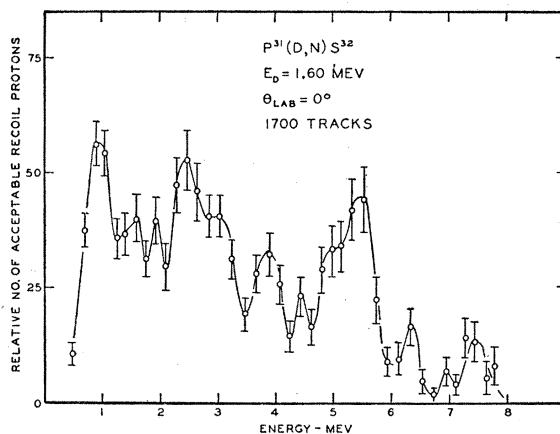


Fig. 1. Relative number of acceptable recoil protons per 200-kev interval versus the recoil proton or neutron energy at  $0^\circ$ . Standard deviation of the measurements are indicated at each point.

$P^{31}(d,n)S^{32}$  reaction. When this is done the two sets of data may be added together to improve the statistical accuracy. No meaning can be attached to the resulting intensities other than that of the sum of the  $0^\circ$  and  $90^\circ$  data of the laboratory coordinate system.

The line shape of the recoil proton spectrum that corresponds to a single neutron group may now be found by employing several assumptions. It is assumed first that the Gamow penetrability factor<sup>9</sup> gives the shape of the yield of each neutron group. This assumption should be justified for our low bombarding energy (1.6 Mev). Secondly, it is assumed that the range straggling parameter for monoenergetic protons is given for our detecting geometry by the root mean square of the deviation of all track lengths in the group as given by Richards *et al.*<sup>8</sup> Since the range-energy relation in

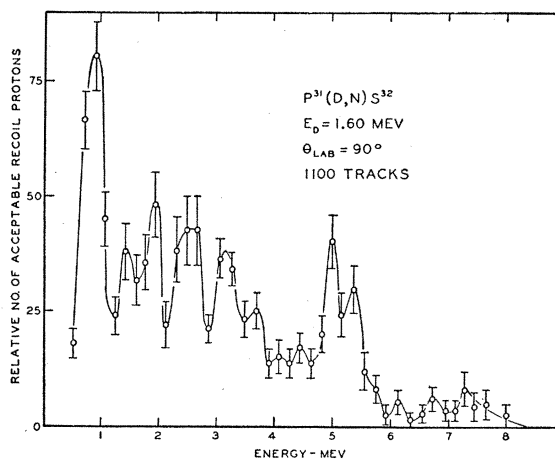


Fig. 2. Relative number of acceptable recoil protons per 200-kev interval versus the recoil proton or neutron energy at  $90^\circ$ . Standard deviation of the measurements are indicated at each point.

<sup>9</sup> J. Mattauch and S. Fluegge, *Nuclear Physics Tables* (Interscience Publishers, Inc., New York, 1946), p. 65.

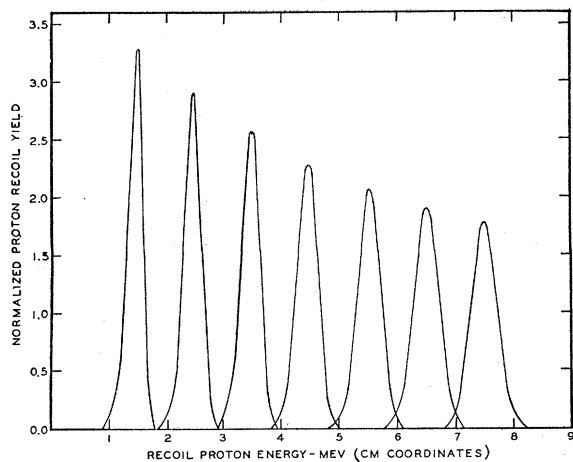


FIG. 3. Plot of typical resolution functions,  $R(x, a_k)$ , showing the combined effect of the thick target and the range straggling of the recoil protons in the nuclear emulsion on the resolution of the various energy neutron groups.

Richard's paper was found using  $(p, n)$  and  $(d, n)$  reactions, the corresponding width of the proton recoil groups should be a good measure of that part of the width of our groups that is caused by the detection of a monoenergetic neutron in a photographic emulsion.

Since our targets may be considered as infinitely thick, the observed line shape of a single neutron group may be found by folding a Gaussian distribution having the above-mentioned straggling parameter into a Gamow yield function each suitably displaced in energy to give the result in the center-of-mass energy scale. Typical line shapes for different neutron groups each normalized to unit area are shown in Fig. 3. This function is designated by  $R(x, a_k)$ , where  $x$  is the energy in the center-of-mass coordinates. The parameter  $a_k$  is related to the  $k$ th  $Q$  value by the equation

$$a_k = M_R Q_k / (M_T + M_B) + M_T M_R E_B / (M_T + M_B)^2, \quad (1)$$

where  $M_B$ ,  $M_T$ , and  $M_R$  are the masses of the bombarding, target, and residual nuclei, respectively.  $E_B$  is the bombarding voltage and  $Q_k$  is the  $Q$  value for the  $k$ th neutron group. The tabular form of the function used is given in Table I. The observed proton recoil spectrum then should be a superposition of the preceding line shapes each adjusted in intensity to account for the relative amounts of each group of neutrons present. Thus, the observed proton recoil spectrum should be written as

$$y = \sum_{k=1}^{\nu} A_k R(x, a_k). \quad (2)$$

The theory of least squares<sup>10</sup> may be used to fit Eq. (2) to the observed points and thereby unfold the data to give the relative intensity  $A_k$  and the energy  $a_k$  of the  $k$ th neutron group. The method also gives the uncer-

tainties in the adjusted parameters  $A_k$  and  $a_k$ . However, it is not possible on a *priori* grounds to choose the correct number of groups  $\nu$ . A number may be selected for  $\nu$  and the statistical  $\chi^2$  test may be employed to judge the over-all suitability of the resulting fit.

### EXPERIMENTAL RESULTS

The  $0^\circ$  and  $90^\circ$  laboratory coordinate data of Figs. 1 and 2 were plotted after applying a correction for the angular straggling (2.28 percent). As explained in the previous section, the relative yields in 0.20-Mev intervals were found in the center-of-mass energy scale. Figure 4 represents the sum of these two sets of transformed data except that the doubtful carbon contamination peak has been subtracted out. Reasonable choices for  $A_k$ ,  $a_k$ , and  $\nu$  were next assumed, and the resulting value of the weighted sum of the squares of the deviations,  $S_0$ , was found. By inspection the values of  $A_k$  and  $a_k$  were adjusted and a new value of  $S_0$  was computed. This process was continued until  $S_0$  could not be reduced further. This smallest value of  $S_0$  was taken as  $\chi^2$ , since the weight of each observation was set equal to the reciprocal of the square of its standard deviation. The trial and error method by which the parameters were found leads one to a fair estimate of the uncertainties involved. In general,  $A_k$  is uncertain by about 15 percent and the uncertainty in  $a_k$  is about  $\pm 0.1$ -Mev.

TABLE I. Resolution function  $R(x, a_k)$ . Note that  $a_k$  is connected to  $Q_k$  by Eq. (1); however, since the chief effect of  $Q_k$  is included in  $x - a_k$ , the changing shape of the function with  $a_k$  of  $Q_k$  is indicated only in gross intervals of  $\Delta Q_k = 1$ -Mev. Units of  $x$ ,  $a_k$ , and  $Q$  are Mev.

$Q$	$x - a_k$						
	0.0	1.0	2.0	3.0	4.0	5.0	6.0
-0.80	0.002	0.005	0.010	0.017	0.026	0.036	0.048
-0.75	0.007	0.012	0.020	0.031	0.045	0.061	0.077
-0.70	0.022	0.030	0.043	0.059	0.076	0.097	0.121
-0.65	0.050	0.061	0.077	0.098	0.123	0.154	0.184
-0.60	0.094	0.105	0.128	0.158	0.193	0.231	0.269
-0.55	0.154	0.172	0.205	0.248	0.291	0.337	0.383
-0.50	0.242	0.269	0.316	0.374	0.427	0.479	0.528
-0.45	0.373	0.415	0.474	0.542	0.607	0.668	0.721
-0.40	0.566	0.630	0.696	0.763	0.834	0.884	0.912
-0.35	0.843	0.918	0.987	1.050	1.103	1.128	1.134
-0.30	1.222	1.300	1.353	1.384	1.397	1.386	1.358
-0.25	1.705	1.765	1.770	1.745	1.685	1.608	1.558
-0.20	2.268	2.243	2.180	2.071	1.924	1.789	1.708
-0.15	2.862	2.687	2.488	2.277	2.067	1.912	1.789
-0.10	3.302	2.912	2.579	2.279	2.042	1.877	1.782
-0.05	3.170	2.650	2.378	2.142	1.938	1.778	1.684
0.00	2.211	2.055	1.917	1.788	1.677	1.582	1.510
0.05	1.015	1.249	1.330	1.345	1.338	1.318	1.283
0.10	0.292	0.612	0.786	0.894	0.981	1.019	1.028
0.15	0.051	0.234	0.392	0.533	0.660	0.747	0.779
0.20	0.004	0.090	0.165	0.271	0.406	0.507	0.556
0.25		0.014	0.057	0.131	0.229	0.330	0.374
0.30		0.003	0.017	0.052	0.117	0.190	0.237
0.35		0.001	0.005	0.022	0.054	0.095	0.141
0.40			0.001	0.008	0.023	0.047	0.079
0.45				0.001	0.008	0.021	0.041
0.50					0.003		

<sup>10</sup> W. E. Deming, *Statistical Adjustment of Data* (John Wiley and Sons, Inc., New York, 1943).

Figure 4 and Table II give the important results of this paper. The resulting curve through the points was drawn by using Eq. (2) with the constants  $A_k$  and  $a_k$  given in Table II. Let the groups be labeled 1, 2, 3, etc., beginning with the ground-state group. If the first 12 groups are tested for goodness of fit by the  $\chi^2$  test,<sup>11</sup> the probability  $P$  that the observed value of  $\chi^2=15.1$  will be exceeded for  $9(=33-2\times 12)$  degrees of freedom is 0.06. If the 13th group of neutrons is included,  $\chi^2=59.8$  and, for  $12(=38-2\times 13)$  degrees of freedom, the value of  $P$  is less than 0.01. In interpreting these values of  $P$  it is customary to say that if  $P$  lies between 0.9 and 0.1 the assumed distribution very probably corresponds to the observed one, while if  $P$  is less than 0.02 or more than 0.98 the assumed distribution is unlikely and is to be questioned seriously. Hence, we may say that there is reasonable assurance that the first 12 groups are present, but that in order to include the 13th group, an additional group would be needed between the 12th and 13th group to account for the observed high point. We have adopted the procedure of including only those groups that seem to persist in Fig. 1, Fig. 2, and Fig. 4. In order for a group to persist, however, it is not necessary that its detailed structure be in evidence. If a group seemed too wide to be admitted as a single group in all the figures, it was analyzed into two or more groups in the center-of-mass system. The sensitivity of the  $\chi^2$  test is also made evident when the one point that does not fall reasonably close to the assumed distribution causes the goodness of fit test to question seriously the assumed distribution.

Table II gives the results of the inelastic scattering determination of the levels in  $S^{32}$  by Dicke and Marshall<sup>1</sup>

TABLE II. Neutron groups from the  $P^{31}(d,n)S^{32}$  reaction and the energy levels in  $S^{32}$ . To identify the groups,  $k$  runs from 1 to 12 beginning with  $k=1$  for the ground-state group.  $A_k$  is the relative intensity of the  $k$ th group,  $a_k$  is the energy of the  $k$ th group in the center-of-mass system of coordinates, and  $Q_k$  is the corresponding  $Q$  value.  $E_k$  is the excitation energy of  $S^{32}$ . Uncertainty in  $E_k$  is  $\pm 0.1$  percent.

$k$	Present work				Ref. 1 $E_k$ (Mev)	Ref. 2 $E_k$ (Mev)
	$A_k$	$a_k$ (Mev)	$Q_k$ (Mev)	$E_k$ (Mev)		
1	5.3	7.5	6.2	0.0		
2	8.3	7.0	5.7	0.5		
3	7.9	6.0	4.7	1.5		
4	29.0	5.3	4.0	2.2	2.25 ( $pp'$ )	2.35 ( $nm', \gamma$ )
5	26.1	4.8	3.5	2.7		
6	17.4	4.3	2.9	3.3		
7	15.6	3.8	2.4	3.8		
8	15.6	3.5	2.1	4.1		
9	23.7	3.1	1.7	4.5	4.34 ( $pp'$ )	
10	24.1	2.7	1.3	4.9		
11	21.6	2.4	1.0	5.2		
12	24.1	2.1	0.7	5.5		

<sup>11</sup> R. A. Fisher, *Statistical Methods for Research Workers* (Oliver and Boyd, Edinburgh, 1932).

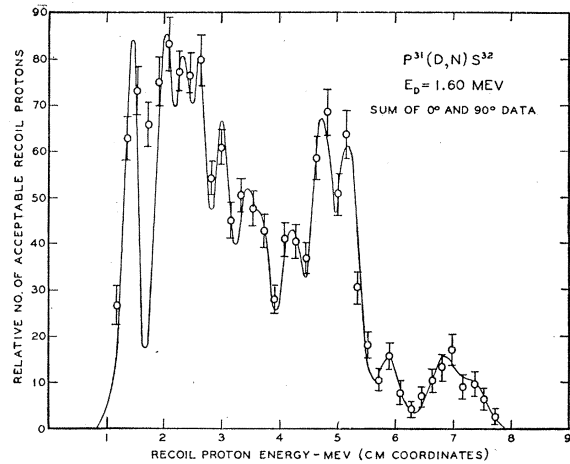


FIG. 4. Plot of the sum of the  $0^\circ$  and  $90^\circ$  laboratory coordinate data transformed to the center-of-mass energy scale. The probable error ( $0.675\times$  standard deviation) of the observed points are shown with each observed point.

and by Grace, Beghian, Preston, and Halban.<sup>2</sup> The agreement of the excited states determined by the various methods is within the uncertainties of the experiments. Finally the ground-state  $Q$  value was found to be 6.2 Mev. Using the latest determination of the masses of  $P^{31}$  and  $S^{32}$  reported by Motz<sup>12</sup> this  $Q$  value should be  $6.63\pm 0.07$  Mev. It should be noted that if the ground-state group was missed, then the  $\gamma$ -ray observed by Grace *et al.*<sup>2</sup> cannot be to the ground state if it is to be associated with the strong group ( $k=4$ ). The preferred interpretation is that the  $\gamma$ -ray is associated with the ( $k=4$ ) group and that the relative accuracy of the determination of the groups is much better than the absolute accuracy. Because of the difficulty in maintaining strictly the  $\pm 12^\circ$  acceptance angle criterion, there is a rather large uncertainty in correcting for the group shifts. In particular it was felt that the uncertainty in correcting for the group shifts due to the angles of emission of the neutrons around  $0^\circ$  and the angular straggling of the recoil protons in the nuclear emulsions could easily cause the energy scale to be four or five percent low. Hence it is felt that the excited states in  $S^{32}$  have the uncertainties given in the analysis but that the ground-state  $Q$  values of 6.2 Mev are subject to a larger absolute error.

In conclusion the author wishes to acknowledge his indebtedness to Mr. T. A. Haddad, who helped with the plate reading, to Mrs. E. A. Seaman for the computational assistance, to Dr. L. Eisenbud for several discussions concerning the chi-test, and finally to Dr. W. F. G. Swann, Director of the Bartol Research Foundation for his continued interest in this problem, especially in its curve fitting aspect.

<sup>12</sup> H. T. Motz, *Phys. Rev.* **81**, 1061 (1951).