

Isotope Shifts in Erbium*

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(Received May 28, 1952)

An investigation of the erbium isotope shifts has been conducted in the region between 4000 and 7000 Å using the dispersion of a Fabry-Perot interferometer crossed with a Steinheil three-prism glass spectrograph. Structures were measured in 68 lines, and components due to the even isotopes Er^{166} , Er^{168} , and Er^{170} have been identified. The ratio of the isotope shifts $(\nu_{168} - \nu_{170})/(\nu_{166} - \nu_{168})$ is close to unity in nearly all lines measured, and it is concluded that the undisturbed value of the ratio probably does not differ from unity by more than 4 percent. Electron configurations have been proposed to describe the observed shifts, and in particular those lines, in which the component due to Er^{170} is shifted to larger wave numbers than the components due to the other even isotopes, probably arise from the two-electron transitions $4f^{11}6s6p$ to $4f^{11}5d^2$.

I. INTRODUCTION

THERE are two general effects which give rise to isotope shifts: the (finite) nuclear mass and the extension of the nuclear charge over a finite volume. The nuclear mass effect, which has been divided into the normal (reduced) mass and specific mass effects for convenience, dominates the isotope shifts among the light elements but decreases in magnitude with increasing atomic weight and is apparently very small among the heavy elements. The nuclear effects dominate among the heavy elements; these arise primarily from the nuclear volume,¹⁻³ nuclear polarization,⁴ and the nonelectrical neutron-electron interaction.⁵ The neutron-electron interaction is very small; the nuclear polarization has not been calculated quantitatively but could account for the even-odd staggering of the isotope shift patterns; the nuclear volume effect probably is the most important. The observed isotope shifts among the heavy elements are in qualitative agreement with the predictions of the theory of the volume effect, but the theory predicts shifts which are twice as large as the observed ones⁶ if one assumes a charge distribution either uniform throughout the nucleus or uniform over the surface and a change in nuclear radius which follows the law $r_0 = 1.5 \times 10^{-13} A^{\frac{1}{3}}$. The discrepancy may be due to the incomplete redistribution of protons when neutrons are added to a nucleus.⁶

Studies of isotope shifts have a twofold interest. First, because the observed shifts are proportional to the change in an effective nuclear (or more properly, nuclear charge) radius, they yield valuable information about the changes in nuclear radius with the addition

of neutrons for a fixed Z . The second interest in the isotope shifts lies in the possibility of identification of electron configuration. This is particularly true among the rare earth elements, which have defied spectral analysis with the exception of a few of the elements which lie at the beginning, at the end, or at the very middle of the group. Even when terms have been identified, it has often been difficult or impossible to identify the electron configuration. The present work was undertaken with both of these points of interest in mind.

II. EXPERIMENTAL APPARATUS

The source of spectral lines was a modification of the liquid-air cooled hollow cathode discharge of the type described by Arroee and Mack.⁷ An initial charge of 30 mg of erbium oxide (Er_2O_3) gave very intense erbium lines, which persisted strongly after more than 35 hours running time at currents of about 100 ma.

A quartz Fabry-Perot interferometer supplied the resolving power of the optical system. The etalon plates were 42 mm in diameter and were silvered by evaporation in vacuum. Auxiliary dispersion was supplied by a Steinheil three-prism glass spectrograph.

III. EXPERIMENTAL RESULTS

A. Observed Structures

With the four spacers available, interferograms of erbium lines were taken in the region of the spectrum from 4000 to 7000 Å. More than 100 lines showed structure, and measurements were made on 68. The structure in each case consists of three components, decreasing in intensity either toward the large or toward the small wave numbers. The components are designated A , B , and C , where A is the most intense and C is the least intense of the three components. The separations, which are given in Table I, are designated as positive if the order of the components with respect to increasing frequency is $C-B-A$, and negative if the order is $A-B-C$. The reason for this choice will be discussed later.

⁷ O. H. Arroee and J. E. Mack, *J. Opt. Soc. Am.* **40**, 386 (1950).

* This article is based upon a thesis by Mr. Willets presented to the Faculty of Princeton University in candidacy for the degree of Doctor of Philosophy. A preliminary report appeared in *Phys. Rev.* **84**, 1055 (1951).

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¹ G. Racah, *Nature* **129**, 723 (1932).

² J. E. Rosenthal and G. Breit, *Phys. Rev.* **41**, 459 (1932).

³ G. Breit, *Phys. Rev.* **42**, 348 (1932).

⁴ Breit, Arfken, and Clendenin, *Phys. Rev.* **78**, 390 (1950); Breit, Arfken, and Clendenin, *Phys. Rev.* **77**, 569 (1950).

⁵ L. Willets and L. C. Bradley, III, *Phys. Rev.* **78**, 390 (1950).

⁶ M. F. Crawford and A. L. Schawlow, *Phys. Rev.* **76**, 1310 (1949).

The limits of accuracy for each measurement given in the table are based upon the internal consistency of the measurements and not upon possible systematic disturbances, which are discussed below.

Figure 1 shows a histogram of the average values of the separations CB and BA for all lines. Although there is a considerable spread for both positive and negative separations, the positive values are grouped around 0.045 and 0.046 cm^{-1} and the negative values are grouped around 0.047 and 0.048 cm^{-1} .

An exploration with a quartz 2-prism spectrograph below 4000Å uncovered very few lines with structure, although the spectrum was quite dense and included some of the strongest lines of the spectrum.

B. Interpretation of the Isotope Shifts

Natural erbium ($Z=68$) contains six isotopes with the following relative abundances:

Atomic weight	Relative abundance, %
162	0.136
164	1.56
166	33.4
167	22.9
168	27.1
170	14.9

The identification of the components A , B , and C with the erbium isotopes 166, 168, and 170 on the basis of intensity and relative abundance is unambiguous. In most lines, the intensities agree quite well with the values to be expected from the relative abundances of the isotopes. In some cases, however, the intensities do vary considerably from the expected values, and this is probably due to the disturbing influence of Er^{167} .

In several lines a fourth component was observed beyond A . Its position could be measured fairly well in $\lambda 5826$. On the basis of position and relative intensity, the component could be attributed to Er^{164} , but it is also possible that it is due to a component of the hfs of the odd isotope.

No components in the structure could be definitely attributed to Er^{167} in any case observed. It is evident from the appearance of the lines that the structure of the odd isotope is not localized between Er^{166} and Er^{168} . Er^{167} is reported⁸ to have a spin of $7/2$.

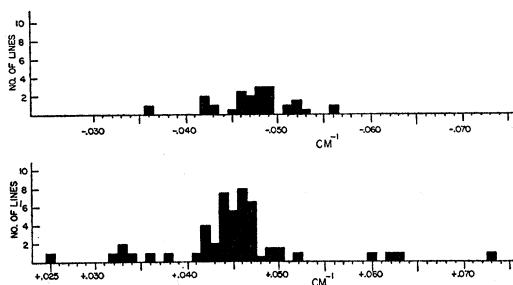


FIG. 1. Distribution of the mean values of the isotope shifts, $\frac{1}{2}(\nu_{166} - \nu_{170})$.

TABLE I. Isotope shifts in the spectrum of Er. The values quoted are averages of the measurements made with various sizes of interferometer spacers. i indicates that the intensities differ from the expected values.

$\lambda(\text{Å})$	Mean shift $\nu_{168} - \nu_{170}$ (10^{-3} cm^{-1})	Mean shift $\nu_{166} - \nu_{168}$ (10^{-3} cm^{-1})	Ratio
6583.46	-48.6±1.0	-46.7±1.0	1.04
6541.53	42.7±1.2	44.2±1.2	0.97
6326.11	42.0±1.2	42.4±1.2	0.99
6308.79	-47.7±1.2	-48.5±1.2	0.98
6299.41	46.6±2.0	41.3±1.6	1.13
6286.86	65.8±3.0	58.9±3.0	
6262.56	43.8±1.2	44.6±1.2	0.98
6076.44	42.3±2.0	42.6±1.6	0.98
6022.56	41.9±2.5	41.9±2.5	1.00
6014.83	50.0±2.0	49.0±2.0	1.02
6006.80	45 ±5		
5855.32	34.6±1.4	33.9±1.2	1.02
5853.6 (?)	31.7±2.5	32.3±2.5	0.98
5826.79	32.6±1.0	33.4±1.0	0.98
5800.77	36.4±1.2	35.8±1.0	1.02
5782.81	42 ±7		
5769.92	24.2±2.5	26.4±2.5	0.92
5762.79	i 33.2±1.2	i 33.1±1.0	1.00
5739.18	48 ±5	45 ±5	
5726.99	-47.0±1.5	-46.4±1.5	1.01
5719.53	44 ±5	44 ±5	
5675.82	48 ±5	41 ±5	
5664.95	42.1±1.5	43.6±1.5	0.97
5640.34	42.7±2.5	44.7±1.5	0.96
5622.02	48.1±3.5	43.4±2.5	
5503.78	43 ±4	40.9±2.5	
5485.93	44 ±5	48 ±5	
5468.32	47.4±2.0	46.1±1.2	1.03
5456.60	45.7±1.0	46.3±1.0	0.99
5350.44	46.9±2.5	40.2±1.5	1.17
5348.04	44.5±1.5	43.9±1.5	1.01
5285.65	-45 ±5	-45.5±4	
5277.70	50.0±1.3	48.5±1.2	1.02
5246.11	i 46.6±1.2	i 42.3±1.2	1.10
5211.16	50 ±5		
5172.75	39.3±1.0	37.2±1.0	1.06
5164.77	45.3±1.4	43.9±1.4	1.03
5160.35	46.1±2.5	46.9±1.4	0.98
5131.52	44.4±1.2	46.7±1.2	0.95
5124.56	-48.2±1.2	-49.0±1.2	0.98
5080.51	-49 ±5	-46.8±2.5	
5058.62	i -56.5±2.0	i -55.6±2.0	1.02
5035.94	48.1±1.2	45.4±1.2	1.06
5028.32	44.6±1.2	47.4±1.5	0.94
5007.25	-43.1±2.5	-43.4±1.5	0.99
5004.1 (?)	46 ±5	49 ±5	
4966.99	-46.1±3.0	-38.3±3.0	1.20
4944.36	46.8±1.4	47.2±1.4	0.99
4928.86	62.7±1.2	57.2±1.2	1.09
4888.86	76.6±1.3	69.6±1.3	1.10
4861.10	53 ±5		
4857.43	47.4±1.2	45.0±1.2	1.05
4848.83	46.6±1.2	43.0±1.3	1.08
4820.75	-52.3±1.5	-51.0±1.5	1.02
4816.63	62.1±2.5	63.1±1.5	0.98
4739.2 (?)	-49 ±5		
4729.04	46.6±1.0	46.6±1.0	1.00
4722.77	-50.6±1.0	-51.3±1.0	0.99
4673.16	-49.5±1.0	-46.9±1.0	1.06
4606.62	-45.6±1.0	-45.6±1.0	1.00
4552.13	45.8±1.0	47.6±1.0	0.96
4531.11	47.5±1.0	44.6±1.0	1.07
4496.38	44.7±1.0	45.9±1.0	0.97
4426.77	-47.2±1.0	-50.0±1.0	0.94
4424.57	-43.8±1.0	-40.4±1.0	1.08
4409.35	-45.1±1.0	-46.2±1.0	0.98
4331.35	-53.0±1.0	-52.0±1.0	1.02
4286.52	-37.3±2.0	-34.3±2.0	1.09

⁸ B. Bleany and H. E. Scovil, Proc. Phys. Soc. (London) A64, 204 (1951).

The case⁹ of ytterbium supplies an example of what may be expected in erbium. The hyperfine structure of the two odd isotopes Yb¹⁷¹ ($I=1/2$) and Yb¹⁷³ ($I=5/2$) is much larger than the isotope shifts of the three even isotopes, even though the magnetic moments of the odd ytterbium isotopes are relatively small (0.45 and -0.65 nuclear magnetons).

Large hyperfine structure splittings may be expected from the odd erbium isotope also, but because of the large spin and because of the large J -values which may be expected in the strong lines of the erbium spectrum, a great number of hfs components are probable. Although the abundance of the odd isotope is comparable with that of the most abundant even isotopes, distribution of the intensity among the numerous hfs components can logically account for the absence of observable structure due to it. If a line should be found which connects levels of low J -values, the hfs of Er¹⁶⁷ should be observable.

Even though components of the odd isotope are not visible, they may be expected to influence measurements on the even isotopes, and this will be discussed later.

Recently there has appeared a report by Murakawa and Suwa¹⁰ on measurements of isotope shifts in a number of Er lines. The results are qualitatively in agreement with our own, but disagreements exist which lie outside the assigned limits of error. We have measured four of the five lines on which Murakawa and Suwa have reported; the fifth line, $\lambda 4086.7$, was not observed although we did observe (but did not measure) the strong line $\lambda 4087.6$ which has a structure similar to that reported for $\lambda 4086.7$. They quote an average ratio of the isotope shifts which corresponds in our convention to a ratio of 1.09 ± 0.05 (their ratio is the inverse of ours); such a large value seems unlikely on the basis of our measurements. Only in the case of $\lambda 5172.8$ do we obtain a ratio in close agreement with their value for the same line. Since in every case reported their measurement of the shift (168-166) is distinctively smaller than ours, the possibility is suggested that the centers of gravity of these two components have shifted towards one another in their plates because of insufficient resolution, possibly influenced by the components of Er¹⁶⁷. That this may indeed be the case is further suggested by the fact that their source was

water-cooled, whereas ours was cooled with liquid nitrogen. Our measurements on Er¹⁶⁴ are not considered sufficiently accurate for any meaningful comparison.

According to the volume picture of isotope shifts, the shifts should be proportional to the change in effective nuclear radius. It is not possible to calculate the change in effective nuclear radius in these investigations since no analysis of the spectrum is available. However, it is possible to investigate whether or not the change in radius is uniform for the three even isotopes, since the ratio of the isotope shifts $(\nu_{168} - \nu_{170}) / (\nu_{166} - \nu_{168})$, should be proportional to the changes in a nuclear radius for the corresponding isotopes.

The ratios of the isotope shifts are given in the table wherever both splittings are known to $\pm 0.0025 \text{ cm}^{-1}$ or better. The average of these ratios is 1.015. If the average is restricted to those lines whose splittings are known to $\pm 0.0015 \text{ cm}^{-1}$ or better, the ratio is 1.010.

A histogram of the ratios is shown in Fig. 2. Although the ratios in the individual lines are in most cases equal to unity within experimental error, the spread of values must be attributed more to the disturbing effects of the odd isotope than to the inaccuracy of the measurements. In many cases the deviation from unity, above or below, is outside of experimental error.

It is quite possible that the disturbing effects of the odd isotope do not cancel out when the results from many lines are averaged; rather, there may be a systematic shifting of the ratios. It is perhaps significant that one of the largest ratios observed, 1.10, comes from the line ($\lambda 4888$) which shows the largest isotope shift. When the shifts are large, as they are in this particular line, the disturbing effects of the odd isotope would be expected to have less influence on the ratio than in the case of smaller splittings, simply because for the same disturbing effect upon an individual components, the relative disturbance of the shift is less.

Except for the one case mentioned above, there appears to be no evidence to indicate that the ratio of the even isotope shifts differs from unity by more than 0.04 in either direction.

C. Assignment of Electron Configuration

Very little in the way of analysis has been accomplished in the rare earth spectra. Although some classifications have been made at both ends and in the very middle of the group, the accomplishments elsewhere have been fragmentary or nonexistent.

Zeeman and hyperfine structure patterns are very useful in spectral analysis, particularly in the identification of terms and J -values. Isotope shift data can be equally useful in the assignment of electron configurations, which frequently defy identification even after the terms are known.

Roughly speaking, the rare earths are formed by the filling in of $4f$ electrons inside the valence electrons. Lanthanum ($Z=57$) begins the group with a structure of $5d6s^2$ outside the closed xenon shell of 54 electrons.

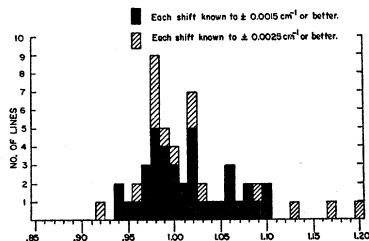


FIG. 2. Distribution of the ratios of the isotope shifts, $(\nu_{168} - \nu_{170}) / (\nu_{166} - \nu_{168})$.

⁹ Schüler, Raig, and Korsching, Z. Physik **111**, 165 (1938); H. Schüler and H. Korsching, Z. Physik **111**, 386 (1938).

¹⁰ K. Murakawa and S. Suwa, Phys. Rev. **85**, 683 (1952).

At lutecium the f -shell is filled and the final structure is $4f^{14}5d6s^2$. The single ion is obtained by the removal of one of the valence electrons.

The unfilled f -shell gives rise to a tremendous number of terms¹¹ for the valence electrons to build on. The spectra are further complicated by the competition of the $4f$, $5d$, and $6s$ electrons which are nearly equal in energy and which give rise to many electron configurations of comparable energy. The number of strong lines which can arise is tremendous: thirteen of the rare earths (Pm omitted) account for more than one-fourth of the atomic lines in the *MIT Wavelength Tables*,¹² while cerium alone accounts for more than 5 percent with 5755 lines out of the more than 100,000 listed.

Configuration interactions are quite common in rare earth spectra, and arise because of the competition of the f , d , and s electrons. The angular momenta of the individual electrons are not constants of the motion and the quantum numbers of a particular electron are not "good" quantum numbers. Generally the admixture of different configurations is not necessary in order to obtain a good approximation to the state of the atom, but where the electron energies corresponding to different l -values are nearly equal, the mixing of configurations of the same parity may become quite strong. In Gd II, for example, Russell¹³ could not distinguish between the P terms arising from the $4f^76s6p$ configuration and those arising from $4f^75d6p$.

A relatively small admixture of a second configuration may impart much of its own characteristics to the primary configuration. A common result is the appearance of double electron transitions. For example, a pd configuration admixed with some ps could make a transition to s^2 . Such double transitions are relatively common in rare earth spectra and will be seen to play an important role in the interpretation of the origin of the erbium isotope shifts.

In those rare earths where analyses have been made it is often not certain what the lowest levels are, for there often exist a set of low even and a set of low odd levels for which no intercombinations have been discovered, probably because they lie too far into the infrared. The low configurations of one group are $4f^n5d^2$, $4f^n5d6s$, $4f^n6s^2$, and for the other group, $4f^{n+1}5d$ and $4f^{n+1}6s$. The intermediate configurations which can combine with these are found by replacing either a $5d$ or a $6s$ electron with a $6p$.

It is most probable that the shifts observed arise from the second spectrum of erbium (Er II), although it is possible that some of the smaller shifts in the red may be due to the first spectrum. There are several reasons for believing this to be true: First, hollow cathodes usually excite the ion of rare earth spectra,

¹¹ See, for example, L. Pauling and S. A. Goudsmit, *Structure of Line Spectra* (McGraw-Hill Book Company, Inc., New York, 1930), p. 156.

¹² G. R. Harrison, *MIT Wavelength Tables* (J. Wiley and Son, New York, 1939).

¹³ H. N. Russell, *J. Opt. Soc. Am.* **40**, 550 (1950).

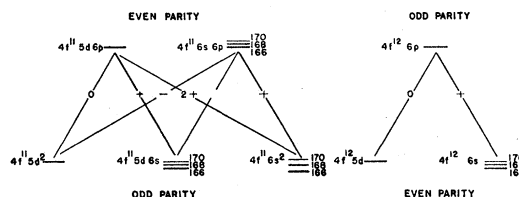


FIG. 3. Proposed electron configurations and transitions in Er II.

particularly when He is the carrier gas. Secondly, the intensities of the lines which show structure were found to agree more closely with spark spectra data¹⁴ than with arc, in the regions where both were available. This was confirmed by an arc spectrogram, which showed that the lines in question were enhanced near the poles of the arc. Finally, the isotope shifts are of the same magnitude as shifts due to the second spectrum of other elements in the region.

The diagram in Fig. 3 shows the more probable configurations of the erbium spectrum, and what combinations may be expected among them. Each configuration contains many terms and even more levels. In this schematic representation, the levels of all isotopes are adjusted to coincide for these configurations where there is no s valence electron. The isotope shifts are roughly the same for all levels with one $6s$ electron and roughly twice this amount for two $6s$ electrons. Account has not been taken of the $6p_{3/2}$ electrons, which should contribute shifts about 5 percent as great as the $6s$ electrons. When a p electron is designated in a term, it may be a $p_{3/2}$, a $p_{1/2}$ or a linear combination of the two. In general, the p electrons may be said to contribute a shift 0 to 5 percent of the normal $6s$ shift. The lightest isotope (Er¹⁶⁶) is always lowest, and, of course, combinations can only occur between levels of the same isotope.

The p to s transitions give rise to the normal isotope shifts, with the component due to the heaviest isotope (Er¹⁷⁰) appearing displaced to the small wave numbers; shifts of this type were designated as positive in the table. According to the scheme proposed in Fig. 3 there are three different transitions which could give rise to such positive shifts, and also one double transition which could give a positive shift about twice the magnitude.

The negative shifts observed undoubtedly arise from the double electron transition $4f^{11}6s6p$ to $4f^{11}5d^2$, since no other probable transitions among low terms of Er II can account for them. The large number of rather strong lines with negative shifts indicates that configuration mixing is taking place quite strongly. Perhaps the mixing of configurations which have isotope shifts with those which do not accounts for part of the spread in values of the isotope shifts (Fig. 1). On the other hand, the spread of values is small enough to justify the assignment of a single configuration to most levels.

¹⁴ A. Gatterer and J. Junkes, *Spektren der Seltenen Erden* (Specola Vaticana, Vatican City, 1945).

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 Schawlow⁶ 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 cm^{-1}) is somewhat less than 93 percent of twice the value of the normal positive shifts (45.5 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 cm^{-1} or 0.042 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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(Received July 23, 1951)

The neutron spectrum at 0° and 90° 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 ± 0.1 -Mev.

INTRODUCTION

TWO previous measurements of the energies of the excited states in S^{32} have been reported. Dicke and Marshall¹ 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 Halban² 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 phosphorus³ was bombarded by 1.60-Mev deuterons from the Bartol electrostatic generator. The resultant neutron spectrum was found by observing the proton recoils in 100μ

Eastman NTA nuclear emulsions mounted 3 cm from the target at angles of 0° and 90° 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° plates and 1100 tracks on the 90° 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° plates as were observed in the same range when tracks of all energies were counted; on the 0° plates, the factor was 1.79 for the same energy range. The data, corrected for the variation of neutron-proton scattering cross section,⁴ geometry,⁵ 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.

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

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

³ 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.

⁴ R. K. Adair, Revs. Modern Phys. **22**, 249 (1950).

⁵ H. T. Richards, Phys. Rev. **59**, 796 (1941).