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Levels in ${}_{75}Re^{188}$ from Studies of the Decay of ${}_{74}W^{188}$ (69.4 day), ${}_{75}Re^{188}$ (18 h), and ${}_{75}Re^{188m}$ (18.5 min)*

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Sources of the 69.4-day β^- emitter 74W¹⁸⁸ were produced by successive capture of two neutrons in W¹⁸⁶. A study with a double-lens spectrometer revealed the presence of a 349±3-keV beta ray decaying to the ground state of 75Re¹⁸⁸. A 285-keV beta-ray component to the 63.7-keV level in Re¹⁸⁸ was observed in coincidence experiments. The 59-keV beta-ray branch populating the 290.3-keV level in Re¹⁸⁸ was not directly observed. The source used for the study of the beta-ray spectrum of W188 contained an equilibrium concentration of the 18-h daughter Re¹⁸³; hence, some of the properties of this latter beta decay were also examined. Scintillation and coincidence studies on sources of W188 from which the daughter activity had been removed chemically showed the presence of three gamma transitions: 63.7, 226.7, and 290.3 keV. The former two are found to be in coincidence. Internal-conversion-electron studies with photographic spectrographs showed lines from all of the above transitions. Scintillation measurements on the gamma-ray spectrum of the 18.5-min isomeric activity in Re¹⁸⁸ showed the presence of three gamma transitions with energies of 63.7, 92.8, and 105.9 keV. The latter two gamma rays are both in coincidence with the 63.7-keV transition but not in coincidence with each other. In addition to internal-conversion-electron lines from these three transitions, spectrographic studies revealed one line at 11 keV which is interpreted as the M line of a transition of 14 ± 1.0 keV. It is concluded that the 92.8- and 105.9-keV transitions depopulate energy levels at 156.5 and 169.6 keV. All of the transitions observed in both activites, i.e., 63.7, 92.8, 105.9, 226.7, and 290.3 keV, are assigned M1 character. From this fact and a consideration of transition rates and possible spin assignments, we conclude that both the 156.5- and the 169.6-keV states have spin $I^{*}=3^{-}$ and that the isomeric state must lie slightly above the 169-keV level (170 keV < E < 183 keV). To the extent that they overlap, both our experimental results and our theoretical conclusions are in agreement with the other two pertinent studies that have been conducted concurrently on W188 and Re188m by Roy and by Takahashi, McKeown, and Scharff-Goldhaber. For the purpose of this report, the energy of 171.5 keV for the isomeric state, directly observed by the latter authors, is assumed to be the best value. The ground state and five excited states are assigned the following spins: ground state, $I = 1^-$; 63.7 keV, $I = 2^-$; 156.5 keV, $I = 3^-$; 169.6 keV, $I = 3^-$; 171.5 keV, $I = 6^-$; and 290.3 keV, $I = 1^-$. An interpretation of the level scheme is presented in terms of the spin-spin coupling model for odd-odd nuclei proposed by Gallagher and Moszkowski. All of the observed levels can be explained by various couplings of a proton in the $\frac{5}{2}$ [402] Nilsson level to a neutron in one of three different available neutron levels. The first two excited states are interpreted as members of a K=1 rotational band based on the ground state.

I. INTRODUCTION

THE radioactive nuclide W¹⁸⁸ was identified by Lindner and Coleman¹ by means of the radiochemical separation of rhenium from samples of neutronirradiated natural tungsten. The radioactive daughter Re¹⁸⁸ was identified by its half-life; and by observing

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¹ Present address: Ripon College, Ripon, Wisconsin. ¹ M. Lindner and J. S. Coleman, J. Am. Chem. Soc. 73, 1610 (1951). the decay of successively separated rhenium fractions, they were able to assign a half-life of 65 ± 5 days to the precursor W¹⁸⁸. They also determined the half-life of Re¹⁸⁸ to be 16.9 h.

For the investigation reported here, samples of tungsten oxide (enriched in W^{186}) were irradiated in the MTR reactor, Arco, Idaho, for periods of 3-6 weeks. After chemical purification of the active tungsten, study of the radiations was commenced. In a preliminary report of this work,² the authors noted the presence of two betaray components (342 and 431 keV) which could be

²S. B. Burson, D. Zei, and T. Gedayloo, Bull. Am. Phys. Soc. 7, 35 (1962).

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^{*} Work performed under the auspices of the U. S. Atomic Energy Commission.

assigned to the tungsten activity. Five gamma-ray peaks corresponding to transitions of about 57, 142, 165, 222, and 290 keV were also observed in the scintillation spectrum. The beta-ray component listed as 431 keV was found to come from W¹⁸⁵ present as a contaminant and the component listed as 342 keV was identified as the ground-state transition from W188 to Re¹⁸⁸. (Two of the gamma rays originally reported at 142 and 265 keV were very weak and were subsequently found to be spurious.)

Roy,³ in a study of the decay of W¹⁸⁸, measured the half-life of the activity to be 69.4 ± 0.5 days. We have not attempted to measure this quantity with higher precision and have adopted Roy's value for the halflife. His value of 360 ± 40 keV for the end point of the ground-state beta transition (measured by the absorption technique) agrees with our spectrometer value of 349 ± 3 keV. Roy confirmed the presence of the three strong gamma transitions and assigned energies of 63, 227, and 290 keV to them. Further details of this investigation will be compared with our own results at appropriate points in the ensuing discussion.

The isomer Re^{188m} was studied by Flammersfeld⁴ and Mihelich⁵ independently. The former reported a gamma transition of about 60 keV which he concluded to be the isomeric transition to the Re188 ground state. He measured the ratio of charged to uncharged radiation to be about 2 and assumed this number to represent the total conversion coefficient of the 63-keV transition. (Examination of the reported data in the light of our present knowledge of the decay scheme of the Re^{188m} activity shows that the number reported by Flammersfeld cannot be interpreted as the internal-conversion coefficient of the 63-keV transition.) Mihelich measured the internal-conversion-electron spectrum of Re188m and reported transitions of 63.5 and 105 keV. Warner and Sheline⁶ found no evidence of direct beta decay from the Re¹⁸⁸ isomeric state to any of the levels in Os¹⁸⁸. In a tentative interpretation⁷ of these data, it was pointed out that the lifetime of the metastable state was about 10⁹ times as long as the single-particle estimate based on a spin assignment of 4⁺. An assignment of 6⁻ brings the lifetime to within an order of magnitude of the expected value. Recently, the results of a study of Re^{188m} have been reported by Takahashi et al.⁸ These measurements agree with our own results and will be presented for comparison in conjunction with the appropriate sections of this report.

The present investigation was directed primarily at determining the properties of the states in the odd-odd nucleus Re¹⁸⁸. At the time the work was commenced, no studies of the radiations associated with the W188 had been reported and little work had been done on the 18.5-min isomeric activity. The ground state was believed to have $I^{\pi} = 1^{-}$ because of its beta-decay properties, and two excited states at 63 and 169 keV were known to exist. To the extent that they overlap, our findings seem to agree with those of the other two independent studies^{3,8} that have been conducted concurrently.

The experimental details are presented in two sections, those relating to the studies made on the 69.4-day parent sources of W¹⁸⁸ (Sec. II) and those relating to the isomeric activity of Re¹⁸⁸ (Sec. III) The results are combined in the section justifying the decay scheme (Sec. IV). In Sec. V, an attempt is made to present a plausible interpretation of the results within the framework of the spin-spin coupling model which Gallagher and Moszkowski⁹ postulate for odd-odd nuclei.

II. STUDIES OF THE 69.4-DAY ACTIVITY IN W188

A. Production and Purification of Sources

The samples, 5–10 mg of tungsten oxide (enriched 97% in W186), were irradiated in a flux of approximately 1.7×10^{14} neutrons/cm²-sec in the MTR reactor, Aroco, Idaho, for periods of 3-6 weeks. The successive capture of two neutrons produced substantial amounts of W188.

Preliminary examination of the gamma-ray spectrum revealed peaks suggesting contamination from 60-day Sb¹²⁴. To eliminate this, the source was dissolved in dilute NaOH, a few milligrams of antimony carrier was added, and finally the antimony was precipated from the solution with H₂S according to the procedure described by Hildbrand and Lundell.¹⁰ Examination of the gamma-ray spectrum of the precipitate confirmed identification of the contaminant. The antimony was completely purged from the tungsten after two such precipitations. After this purification, the source exhibited a spectrum with few features not attributable to the decay of 18-h Re¹⁸⁸. Study of β -ray and γ -ray spectra associated with the source was then commenced.

B. Beta-Ray Spectrum

The beta-ray spectrum was measured with the doublelens spectrometer of the Argonne Chemistry Division.¹¹ The detector window was sufficiently thin to obviate the necessity for window corrections above 35 keV. The spectrometer was operated at a resolution of 2.6%. The data were appropriately corrected for decay of the source.

³ J. C. Roy, Can. J. Phys. 40, 677 (1962).
⁴ A. Flammersfeld, Z. Naturforsch. 89, 217 (1953).
⁵ J. A. Mihelich, Phys. Rev. 89, 407A (1953).
⁶ L. B. Warner and R. K. Sheline, Nucl. Phys. 36, 207 (1962).
⁷ Nuclear Data Sheets, compiled by K. Way et al. (Printing and Publishing Office, National Academy of Sciences—National Research Council, Washington 25, D. C.), NRC 59-3-125.
⁸ K. Takahashi, M. McKeown, and G. Scharff-Goldhaber, Bull. Am. Phys. Soc. 8, 595 (1963).

⁹ C. J. Gallagher, Jr., and S. A. Moszkowski, Phys. Rev. 111, 1282 (1958).

¹⁰ W. F. Hildebrand and G. E. F. Lundell, Applied Inorganic Analysis (John Wiley & Sons, Inc., New York, 1953).

¹¹ We are indebted to Paul Day for his cooperation in making these measurements.

FIG. 1. Fermi plot of the beta-ray spectrum of W¹⁸⁸ in equilibrium with Re¹⁸⁸. The Fermi functions appropriate to the decay of Re have been used. The region indicated as Fit I was fitted with two components for the purpose of determining the endpoint energies of the two principal rhenium components. The values appear in Table I. The region desig-The values nated as Fit II was fitted with a single component for the purpose of extrapolation under the tungsten spectrum. The solid line is the theoretical spectrum resulting from Fit II.



1. Source Preparation

The sources were prepared by vacuum evaporation of small amounts of the purified tungsten oxide onto a 0.18-mg/cm² aluminum backing. In order that the parent source be in equilibrium with its daughter Re¹⁸⁸, counting was not commenced for several days after the evaporation. It was then only necessary to correct the spectrum for the 69.4-day decay of the parent.

2. Method of Analysis

The data were processed by use of a computer program for the analysis of continuous complex beta-ray spectra. (The program is described in detail elsewhere.¹²) This program makes a least-squares fit of up to 13 components to a single spectrum without use of the conventional extrapolation prodecure. Provision is made for only two types of spectral distributions, namely the allowed shape and the unique first-forbidden shape. For flexibility, each component is presumed to comprise a mixture of two contributions, one representing each type. A factor α defines the amount of each shape present in the component. Each of the components being fitted is then completely specified by three variable parameters, the end-point energy ϵ_0 , the intensity m^2 , and the shape-partition factor α . A spectrum comprising J components is presumed to be represented by the equation:

$$\begin{split} \vec{N}_i &= \sum_{j=1}^J m_j^2 f_i (\epsilon_{0j} - \epsilon_i)^2 \\ &\times \left\{ (1 - \alpha_j) L_{0i} + \frac{\alpha_j}{12} \left[(\epsilon_{0j} - \epsilon_i)^2 L_{0i} + 9 L_{1i} \right] \right\}, \end{split}$$

where the f_i are Fermi functions, L_{0_i} is the shape correction factor for an allowed transition, the expression $\frac{1}{12}[(\epsilon_{0j}-\epsilon_i)^2L_{0i}+9L_{1i}]$ represents the shape-correction factor for a unique first-forbidden transition, and Jis the number of components.

In the present case, the analysis is complicated by the fact that the spectrum contains particle distributions associated with the decay of two different species-the parent 74W188 and its daughter 75Re188. Therefore, the spectrum was analyzed in two steps. It was concluded from preliminary scintillation studies that the tungsten spectrum contained no components above about 450 keV. Also, it is known from previous investigations of the spectrum of Re¹⁸⁸ that this distribution contains only negligible contributions from components below about 500 keV. The spectrum above 500 keV is thus deemed to represent only the decay of Re¹⁸⁸; it was therefore fitted with the Fermi functions appropriate to that decay. The remainder of this spectrum was then generated by the computer and subtracted from the data. The resulting distribution was presumed to represent only the spectrum associated with the parent tungsten. This distribution was then reentered into the computer with the Fermi functions for Z=75, and the analysis was continued.

3. Beta-Ray Spectrum of 18-h Re¹⁸⁸

Figure 1 shows a Fermi plot of the data. Above the tungsten end point, the curve is concave toward the energy axis as indicated by previous investigation of the Re¹⁸⁸ spectrum.^{13,14} Because of this property, the distribution defied straightforward analysis by the computer program which is limited to either allowed or unique first-forbidden shapes. The Re¹⁸⁸ beta spectrum was analyzed by making three independent fits to this portion of the data, as described below.

A determination of the total decay energy of Re¹⁸⁸ was made by fitting two components to the spectrum above 1.75 MeV (Fig. 1, Fit I). Thus, the values com-

¹² S. B. Burson, R. G. Helmer, and T. Gedayloo, in Applications of Computers to Nuclear and Radio Chemistry, NAS-Sn 3107 Chemistry, 1963 (unpublished).

¹³ M. W. Johns, C. C. McMullen, I. R. Williams, and S. V. Nablo, Can. J. Phys. **34**, 69 (1956). ¹⁴ K. O. Nielsen and O. B. Nielsen, Nucl. Phys. **5**, 319 (1958).



FIG. 2. Momentum plot of the beta-ray spectrum of W¹⁸⁸ in equilibrium with Re¹⁸⁸. Fit III: A single component fitted to the spectrum thus computed to approximate the rhenium spectrum. Fit IV: A two-component fit to the spectrum obtained by subtracting curve A from the data. Curve B is the theoretical spectrum computed in this fit. Curves C and D represent the decomposition of the tungsten spectrum B into its two components W188 and , respectively. Curve E, obtained by the addition of A and B, compares the calculated total spectrum with the data.

puted for the end points were not adversely affected by the inability of the function to fit the data exactly at lower energy. The energy values so derived for the two principal components of the Re¹⁸⁸ beta spectrum are in agreement with other measurements¹⁴ and are entered in Table I. (This fit was not used to measure the branching ratio.)

Fit II (Fig. 1) was made for the purpose of generating the rhenium distribution under the tungsten portion of the spectrum. A single allowed component was fitted to the sequence of data points immediately above the tungsten end point. The solid line in Fig. 1 shows where this approximation to the rhenium distribution lies with respect to the data points. The difference spectrum was then entered into the computer and analyzed with the Fermi functions for Z=75.

It was desired to make use of the genetic relationship between tungsten and rhenium in the analysis, so a value for the intensity of the rhenium contribution to the spectrum was necessary. This intensity was obtained by fitting a single allowed component to the

TABLE I. Summary of measurements on the β -ray spectra of W¹⁸⁸ in equilibrium with Re¹⁸⁸. The indicated branching of the Re¹⁸⁸ spectrum is based on the internal-conversion data. Branching between the 285- and 59-keV components is calculated from the γ -ray intensities (Table V) on the assumption that all transitions have pure M1 character.

Com- ponent	Energy (keV)	Branching ratio (%)	Intensity	Logft	ΔI
Re ¹⁸⁸ $\begin{cases} \beta_1 \\ \beta_2 \\ \beta_3 \end{cases}$	2153 ± 5 1998 ± 5 β 's>800	72 ± 1.5 24.5 ± 1.5 ~3.5	0.99 ± 0.015ª	8.12 8.59	$\begin{array}{c} (1^- \rightarrow 0^+) \\ (1^- \rightarrow 2^+) \end{array}$
$ W^{185} \begin{cases} \beta_4 \\ \beta_5 \\ \beta_6 \end{cases} $	433 ± 3 349 ± 3 285 ± 3 59 ± 3	98.6 0.51±0.1 0.85±0.1	0.725 ± 0.03 1.00 ± 0.02	7.53 7.19 9.2 6.9	$\begin{array}{c} (\frac{3}{2} \longrightarrow \frac{5}{2}^{+}) \\ (0^{+} \longrightarrow 1^{-}) \\ (0^{+} \longrightarrow 2^{-}) \\ (0^{+} \longrightarrow 1^{-}) \end{array}$

* From Fit III, Fig. 2.

data points on the rhenium spectrum, i.e., to the spectral region between about 0.5 and 2.0 MeV. The region used for this purpose is indicated as Fit III in Fig. 2 and the calculated distribution is designated as curve A. The area under this component is considered to represent the intensity of the Re¹⁸⁸ distribution within 2%.

4. Beta-Ray Spectrum of W¹⁸⁸

A Fermi plot of the tungsten spectrum (not illustrated) showed a break at about 350 keV. Also, the first fits made to the tungsten spectrum indicated that its total area was considerably in excess of that calculated for the Re spectrum by Fit III. These facts suggested the presence of an impurity in the tungsten spectrum. The most likely contaminant was W185 with an end-point energy of about 430 keV. It was presumed that the tungsten spectrum comprised two components, the ground-state transition from W188 to Re188 and a contaminant of W185. The region of the spectrum used in this calculation is indicated as Fit IV in Fig. 2. The result of this fit is shown as curve B. Curve C is the calculated spectrum of the W188 ground-state transition. Curve D is the computed distribution of the W¹⁸⁵ contaminant. Curve E is the sum of curves A and B, i.e., it is the composite calculated spectrum.

5. β-Ray Spectrum of W¹⁸⁸ in Equilibrium with Re¹⁸⁸

The conclusions derived from the various fits to the rhenium and tungsten portions of the spectrum are summarized in Table I. The branching ratio for the two main components of Re^{188} is based upon the measured areas of the internal-conversion lines of the 155.03-keV transition in Os¹⁸⁸ and the theoretical internal-conversion coefficients for that transition. (The E2 character of the transition was assumed.) The branching between the 155.03-keV level and the higher excited states



FIG. 3. Internal-conversion lines of the 155.03-keV transition in Os¹⁸⁸. The beta-ray continuum has been subtracted by computer from the data.

was taken from the report by Arns *et al.*¹⁵ The value 0.245 ± 0.015 so deduced for the branching ratio to the 155.03-keV level is in excellent agreement with that which Nielsen *et al.*¹⁴ obtained by "peeling" the Fermi plot. The log*ft* values for these transitions are in excellent agreement with the previous measurements.

The end-point energies and intensities for the W¹⁸⁵ and W¹⁸⁸ components are based on the results of Fit IV. All the intensity values have been normalized to unity for the W¹⁸³ component in order to demonstrate the genetic relation between it and the Re¹⁸⁶ daughter. The log*ft* value of 7.19 for the transition from the W¹⁸⁸ to the Re¹⁸⁸ ground state indicates first-forbidden character (i.e., $\Delta I = 0, 1; \Delta \pi = \text{yes}$). This result for the ground state of Re¹⁸⁸ confirms the assignment of 1⁻ based on earlier studies. (In this calculation, beta branching to the excited states of Re¹⁸⁸ was considered to be negligible.)

C. Internal-Conversion-Electron Spectra

Two different experimental measurements of the internal-conversion spectrum associated with the W¹⁸⁸ decay were carried out; in both experiments the sources used were in equilibrium with the rhenium daughter. The first data described are those taken in conjunction with the measurements on the beta-ray spectrum; the second data are the results of studies made with photographic internal-conversion electron spectrographs.

1. Beta-Ray Spectrometer Measurements

a. Search for internal-conversion lines from transitions in Re^{188} . In the course of the measurements of the continuous beta-ray spectrum, we thoroughly searched the regions where one might expect to find lines resulting from internal conversion of the three principal transitions seen in the scintillation spectrum, i.e., 63, 227, and 290 keV. The beta spectrum showed no features that could be interpreted as due to internal conversion of any of these transitions.

b. Internal-conversion lines of the 155.03-keV transition in Os^{188} . During this search (between about 35 and 300 keV), the strong lines due to the 155.03-keV transition in Os^{188} were carefully measured.

Figure 3 shows a plot of the data spanning the conversion-electron lines from the 155.03-keV transition. The computer was used to ascertain the spectral background beneath the conversion lines. The continuous spectrum was assumed to consist of the two components found in Fit IV, i.e., the spectra of W¹⁸⁵ and W¹⁸⁸. The tungsten spectrum was again entered into the computer and fitted with these two components. However, only the data points in the immediate proximity of the conversion lines were fitted as shown in Fig. 3. The purpose of this approach was to compensate for possible scatter-

TABLE II. Internal-conversion-electron data from measurements with a β -ray spectrometer.

Shell	Intensity $(e^{-}/\text{total }\beta^{-})$	Measured	Theoretical α(155 keV)
K	0.049 ± 0.001		0.327
L_1			0.038
L_2	0.075 (0.002		0.195
M	0.073 ± 0.002		0.143
K/(L+M+N)		0.653 ± 0.02	
$\frac{(M+N)}{L}{K/L}$		0.32^{a} 0.86 ± 0.05	0.865

* Reference 13.

¹⁵ R. G. Arns, R. O. Riggs, and M. L. Wiedenbeck, Nucl. Phys. 15, 125 (1960).



FIG. 4. Gamma-ray spectrum of W¹⁸⁸ (69.4 day) in equilibrium with Re¹⁸⁸ (18 h). A spectrum of pure Re¹⁸⁸ normalized to these data passes through the points except in the region where the curve is dashed in.

ing in the low-energy region of the spectrum. Figure 3 is a plot of the residual counting rates obtained when the beta-ray continuum found in this fit was subtracted from the data. Notice that the points below, between and above the peaks (i.e., the points fitted) are distributed statistically about zero. The areas of the lines were evaluated from the smooth curve drawn through the points.

Table II summarizes the results of these measurements. The only intensities measured directly are for the K line and for the composite L+M+N line. The resolution of the spectrometer was insufficient to justify an attempt to partition this group. These values are normalized to unity for the area of the total rehenium spectrum as measured in Fit III. As mentioned earlier, the measured value for the intensity of the K line, taken together with the theoretical value¹⁶ of α_K for the 155.03keV transition in Os¹⁸⁸, leads to the beta-branching ratio shown in Table I for the 2.653- and 1.998-MeV beta transitions. Our value for the intensity of the K line is only slightly higher than that reported by Nielsen and Nielsen.¹⁴ It is nearly 50% higher than that reported by Johns *et al.*¹³

However, in the report by Johns et al., the higher resolution of the spectrometer permitted separation of

	Line energy (keV)		Interpretation			
		Shell	Energy (keV)	Transition energy (keV)		
1	51.27	Re L1	63.79)			
2	53.48	$\operatorname{Re} L_3$	64.01 }	63.9 ± 0.2	63.7ª	
3	60.75	Re M_1	63.58			
4	81.13	Os K	155.0			
5	142.51	Os L_2	154.89			
6	144.06	Os L_3	154.92 }	154.9 ± 0.2	155.03 ^b	
7	152.42	Os M_3	154.87			
8	154.51	Os N_8	154.97			
9	155.01	$\int Os O_3$	155.03			
		Re K	226.68	226.7		
10	219.04	`Re <i>K</i>	290.7	290.7 ± 1.0		

TABLE III. Internal-conversion-electron data from spectrographic measurements on W¹⁸⁸ (69.4 h).

^a Reference 17. ^b Reference 18.

the L and M lines. In order to carry the analysis forward, we have assumed the value reported by Johns for the ratio L/(M+N). (Although we note and inordinate depression of the K line in that work, we consider the L, M, and N lines to be sufficiently close together in energy so that their ratio would be independent of variations in spectrometer efficiency.) The value for this ratio taken from Johns is included in the table. With the use of this number, our measured value of K/(L+M+N) has been decomposed to obtain K/L= 0.86 which is in excellent agreement with the theoretical value of 0.865.

2. Spectrographic Measurements

Photographic magnetic spectrographs were employed to search for internal-conversion-electron groups from the gamma-ray transitions in Re¹⁸⁸. Table III summarizes the results of these measurements. Most of the observed lines are attributed to the 155.03-keV transition in Os¹⁸⁸. The K line of the 290-keV transition was identified as well as L and M lines of the 63-keV transition (suggesting M1 character for the latter transition). One other line at 155.0 keV may be the K line of the 227-keV transition but it may also be interpreted as an O line from the 155.03-keV transition. The crystal spectrometer values for the 63.7-keV¹⁷ and the 155.03-keV¹⁸ transitions are considered to be the best values.

D. Gamma-Ray Measurements

1. Apparatus and Method of Analysis

The gamma-ray spectrum was examined with a 4×4 -in. NaI(Tl) crystal. The radiations from the source were confined to the axial region of the crystal by a lead collimator (see insert in Fig. 4). This arrangement approximately halves the intensity of the Compton distribution relative to the full-energy peak at 662 keV.

¹⁶ M. E. Rose, *Internal-Conversion Coefficients* (North-Holland Publishing Company, Amsterdam, 1958); L. A. Sliv and I. M. Band, Report Nos. 57ICCKI and 58ICCLI, Physics Department, University of Illinois, 1957–1958 (unpublished).

¹⁷ R. Hardell and S. Nilsson, Nucl. Phys. 39, 286 (1962).

¹⁸ I. Marklund and B. Lindstrom, Nucl. Phys. 49, 329 (1963).

The central cone of the collimator as well as the end of the cylinder facing the crystal is surfaced with a graded filter to eliminate the characteristic x rays of lead. The pulse distributions were analyzed with a multichannel analyzer and the data were processed with the aid of several computer programs that will be mentioned in the ensuing discussion.

2. Calibration

The energy calibration is specified by the correspondence between the centroid of the full-energy peak of the gamma ray and the associated channel number in the analyzer. The centroids of the photopeaks, both in the sources and for the calibration standards, are determined by fitting a Gaussian function to the data points included within each peak.

The spectrometer was calibrated by use of a number of accurately known gamma-ray standards. The calibration was based on the assumption that the relationship between channel number and energy could be represented by the quadratic function

$E = a + bx + cx^2$,

where E is the energy of a gamma ray whose fullenergy peak has its centroid at the point x on the channel axis. The coefficients were determined by fitting the function to the centroid values associated with the standard gamma rays.

3. Spectrum Analysis

In order to "peel" the gamma-ray spectra, two computer programs are used. A number of representative spectral shapes were accumulated by counting a series of monochromatic gamma-ray sources. The first computer program interpolates these shapes to generate a spectral shape for which the full-energy-peak centroid falls at the required point on the energy axis. The desired spectral shapes are then introduced as a library into the computer along with the data. The intensity of each shape is then adjusted by the second program so that the sum of the components is least-squares fitted to the data. The residual spectrum is then examined and appropriate adjustments made in the positions of the centroids. This procedure is repeated until the data are satisfactorily fitted. The intensities are then calculated from the computed relative amounts of each known shape fitted to the data.

4. Chemical Separation of Tungsten and Rhenium

From the absence of any pronounced peaks (except those of Os¹⁸⁸) in the equilibrium gamma-ray spectrum, it was evident that beta branching to any excited states in Re¹⁸⁸ must be very weak. A source of pure Re¹⁸⁸ was prepared by neutron irradiation of Re¹⁸⁷ in the Argonne CP-5 reactor. The spectrum of this source was compared with that of the 69.4-day W¹⁸⁸ sample. The only visible difference between the two spectra was the presence of two small peaks at about 227 and 290 keV. It was evident that the tungsten spectrum could be examined satisfactorily only if the tungsten was chemically freed from its daughter.

The literature describes a number of methods for separating rhenium from tungsten. Several of these were tried but none were found very suitable because of the rapidity with which the separations had to be carried out. In explanation of this statement, it should be mentioned that within 10 min (less then 1% of the growth period of the daughter) after a good separation, the 155-keV peak that characterizes the spectrum of the 18-h Re¹⁸⁸ daughter has grown to a height comparable to that of the 227-keV peak of the W¹⁸⁸ parent. To obtain a satisfactory gamma spectrum of W¹⁸⁸, it was found necessary to make a series of rapid chemical separations and to accumulate data for only a few minutes after each separation.

In the method finally adopted, the tungsten oxide (about 10 mg) was dissolved in a few drops of 3N NaOH. Next 2-3 ml of concentrated nitric acid were added, and the tube was placed in a hot bath for 1-2 min (until precipitation had commenced) and then centrifuged for about 1 min. The supernatant liquid was poured off and counting of the precipitate was begun 4-5 min after the acid was added. When this method was used, the 155-keV peak corresponding to the ground-state transition in Os¹⁸⁸ was almost completely unobservable for the first 1-2 min of counting. This indicated that the separation from rhenium was quite complete. Unfortunately, a considerable amount of the source material did not come down with the precipitate because of insufficient digestion time. After several such separations, it was necessary to reclaim most of the source from the accumulated supernatant acid.

5. Gamma-Ray Spectrum

The intensities of the γ rays (relative to the total β decay) were determined by making use of the genetic relationship between W¹⁸⁸ and its daughter Re¹⁸⁸. Three independent spectra were obtained with the detection system described above: one of W188 in equilibrium with Re188, one of pure Re188, and one of W188 chemically freed from its daughter. The spectra were accumulated in immediate succession, and calibrations were made before and after the counting of each spectrum to provide a check on the stability of the apparatus. Figure 4 shows the gamma-ray spectrum of a source that had aged for several weeks so that the Re¹⁸⁸ daughter activity had reached a condition of equilibrium with the W¹⁸⁸ parent. The second spectrum of the series was that of a sample of pure Re¹⁸⁸ freshly separated from the W188 source material. This spectrum was then superposed on the data from the equilibrium source, and the two spectra were compared. The only visible difference is in the region between approximately 190 and 320



FIG. 5. Gamma-ray spectrum of W¹⁸⁸ chemically separated from Re¹⁸⁸.

keV. In Fig. 4 the shape of the pure rhenium spectrum has been dashed in for comparison. Peaks associated with the W^{188} decay are visible at 226.7 and 290.3 keV. The spectrum includes the 478- and 633-keV peaks of Os¹⁸⁸ in order to facilitate the analysis.

Also the x-ray peak of the equilibrium source was enhanced about 2% relative to its intensity in the pure rhenium source, but this departure is not visible on the graph.

The third spectrum (shown in Fig. 5) is that of the gamma rays emitted by a W^{188} source that has been chemically separated from its rhenium daughter. The data shown are the accumulated result of seven successive chemical separations; each precipitate was counted for 4 min. For each sample, the counting was begun 4–5 min after the precipitation commenced. (In a preliminary separation before the actual accumulation of data was begun, the digestion was permitted to continue for about 30 min. The supernatant acid from this preliminary separation was not used during the run. This procedure served to eliminate most of the 18-h activity that had accumulated in the source before the experiment.)

In the spectrum shown, the contribution from Re¹⁸⁸ that grew in during the counting period has been subtracted. After the data were accumulated, a pure Re¹⁸⁸ source was placed in the source holder, and its spectrum was subtracted from the data until the 155-keV peak had disappeared. (In the original data the 155-keV peak was about half the height of the 227-keV peak.) The energy values assigned to the gamma rays were derived by fitting Gaussian curves to these data as described above (Sec. II. D.2).

6. Gamma-Ray Intensities

The spectrum shown in Fig. 5 was used to represent the shape of a pure W¹⁸⁸ spectrum. A similar spectrum of pure Re¹⁸⁸ (obtained under identical geometric conditions and spectrometer calibration) was used to represent the pure Re^{188} shape. The computer was then employed to determine the relative amounts of these two spectra that were necessary to synthesize the spectrum of the equilibrium source (Fig. 4). For each of the reference spectra, the individual gamma-ray intensities were computed from the integrated areas of the fullenergy peaks. Corrections for crystal efficiency were made by use of a Monte Carlo calculation developed by W. F. Miller and W. J. Snow.¹⁹

The composite peak at about 62 keV in Fig. 5 is due to the 63.7-keV gamma ray, the Re x ray due to internal conversion of the 227- and 290-keV transitions, and a small contribution from the W^{135} contaminant. The tungsten spectrum (Fig. 5) was "peeled" independently in order to separate the W^{185} contribution. (The spectrum from a pure W^{185} source was used to provide a spectral shape in this fitting procedure.)

Table IV summarizes the results of these measurements. In column 1, the energies of the various transitions are listed. These values were derived by applying the procedure described earlier. The more precise values for the 63.7- and 155.03-keV transitions are taken from the literature.^{17,18} In column 2, the intensities are listed on an arbitrary scale, normalized to unity for the 290-keV gamma ray. Appropriate corrections for detection efficiency, escape peak, and fluorescence yield have been made. In column 3, information derived from measurements on the beta-ray spectrum has been invoked (i.e., the number of 155.03-keV quanta per total beta decay); and the intensities listed represent gamma-ray quanta per 100 decays of the source.

7. Coincidence Measurements

a. Gamma-gamma coincidence. Gamma-gamma coincidence experiments were carried out in order to determine the relationships between the various transitions. Two 2×2 -in. NaI(Tl) crystals were used; the angle between their axes was about 60°, and an "antiscattering" shield was placed between them. The resolving time of the coincidence circuit was $2\tau \approx 50$ nsec. Chemically

TABLE IV. Gamma rays in W^{188} activity. The 62-keV peak is decomposed on the assumption that the 227- and 290-keV transitions have M1 character.

	Energy (keV)	Intensity normalized to γ (290.3)	Gamma rays per 100 decays
W ¹⁸⁸	${}^{\{290.3\pm0.7}_{\{226.7\pm0.7}$	$1.0 \pm 0.02 \\ 0.552 \pm 0.01$	0.403 ± 0.017 0.222 ± 0.010
	(63.7ª	$0.883 {\pm} 0.04$	0.183 ± 0.02
Re ¹⁸⁸	+Re K x ray) 155.03 ^b	37.23 ±1.0	0.173 ± 0.02 15.0 ± 0.5

^a Reference 17. ^b Reference 18.

¹⁹ W. F. Miller and W. J. Snow, Argonne National Laboratory Report ANL-6318, 1961 (unpublished). FIG. 6. Spectrum of internal-conversion electrons in coincidence with the 63.7-keV gamma ray. Curve A represents the spectrum corrected for the Re¹⁸⁸ continuum and accidental coincidences. Curve B shows the results following subtraction of the 285keV beta component of W¹⁸⁸.



separated sources were used, and counting intervals were limited to less than 30 min, in order to minimize the effects of the daughter activity. All portions of the spectrum from about 30 to 350 keV were searched. Two γ - γ coincidences were observed. The first was between the 227-keV peak and the composite peak at 62 keV; the second was self-coincidences within the latter peak. The results of these experiments conclusively demonstrate that the 227-keV gamma ray is in cascade with the 63.7-keV transition. This conclusion is supported by the 290-keV crossover transition.

The self-coincidence peak at 62 keV is readily attributed to coincidences between the 63.7-keV gamma ray and the rhenium x rays following internal conversion of the 227-keV transition. None of the gamma-gamma coincidence data are displayed.

b. Beta-gamma coincidence measurements. A twoparameter 4096-channel analyzer was used to examine the spectrum of electrons in coincidence with the 62-keV gamma-ray peak. This experiment was performed in an effort to obtain information concerning the internalconversion coefficient of the 227-keV transition. A 2×2 -in NaI(Tl) crystal was used to detect the gamma rays, while a lithium-drifted silicon detector was employed to detect the charged radiation. The gain of the gamma-ray detector system was adjusted so that the 64 channels along one axis of the 64×64 storage array spanned the range from zero to about 350 keV. The gain of the beta-detector system was set so that full scale on the other axis corresponded to about 500 keV.

Chemically separated sources were used; counting was limited to about 20 min each. After each precipitation, the precipitate was immediately redissolved in NaOH, and the source was prepared by evaporating a small drop of this solution of a cellulose tape backing. Despite this effort, it is felt that the source thickness contributed seriously to degradation of the electron spectrum. The distribution shown in Fig. 6 is the result of adding together three spectra corresponding to the gamma-ray channels that contained the 62-keV peak. The data have been corrected for random coincidences and the contribution from Re¹⁸⁸ that grew into the source during counting.

In an effort to analyze this distribution, a thin source of Hg²⁰³ was used to generate the spectral shapes that were believed to represent the components of the experimental distribution. The spectrum of beta rays (end point at 212 keV) taken in coincidence with the 279-keV gamma ray of Hg²⁰³ was linearly expanded and presumed to represent the 285-keV beta-ray branch to the 63.7-keV excited state in Re¹⁸⁸. The spectrum of the 194-keV internal-conversion-electron group (arising from K conversion of the 279-keV transition in Hg^{203}) was translated along the energy axis and used to represent the internal-conversion-electron groups corresponding to K conversion of the 290- and 227-keV transitions in Re¹⁸⁸. The intensities of these three representative spectra were then adjusted in order to find the best fit to the data. This synthesized spectrum is shown as the solid line in Fig. 6.

The results of this experiment are considered to be qualitative in nature because of the large uncertainties in the preliminary corrections that were made to the data. However, the ratio of the intensity of the internalconversion lines at about 150 keV to those at 220 keV has been estimated to be about 1. This result is discussed in IV. B.5.

E. Directional-Correlation Measurements

1. Experimental Method

The gamma-gamma directional correlation of the 227–63.7-keV cascade was measured with an automated angular-correlation apparatus. This apparatus will be described elsewhere. The source used for the directional-correlation measurements consisted of a dilute NaOH solution of the equilibrium $W^{188} - Re^{188}$ activity. The source, $\frac{3}{16}$ in. in diameter by $\frac{1}{2}$ in. long, was contained in a sealed Teflon tube, mounted 9.3 cm from two 2×2 -in. NaI(Tl) detectors. The crystals were shielded by $\frac{3}{8}$ -in. Lucite beta absorbers. One of the detectors is attached to a motor-driven arm which can be rotated about an axis through the source.

Coincidences were detected by a conventional fastslow coincidence circuit with a resolving time of $2\tau \approx 40$ nsec. Accidental coincidences were negligible for the source strength used in the experiment. The signal from the stationary counter was fed to a single-channel analyzer whose window was set to span the 62-keV region (Region I in Fig. 7). The discriminator of the single-channel analyzer connected to the movable counter was set to a low level so as to exclude only detector noise. The signal from the counter was also fed to a 256-channel pulse-height analyzer. The analyzer was divided into two 128-channel subgroups ar-



FIG. 7. Spectrum of β rays in coincidence with the 62keV peak.

ranged so that all pulses were normally recorded in the lower subgroup. A routing signal, derived from the slow coincidence circuit, was used to momentarily transfer storage to the upper subgroup so that pulses coincident with the 63.7-keV gamma ray were stored separately. This technique, which records the entire singles spectrum as well as the complete spectrum in coincidence with a selected photopeak, is particularly convenient when the measured correlation must be corrected for interference of other γ rays.

In accumulating the data, the movable counter was rotated sequentially to the angles 90°, 135°, 180°, 225°, 270°, and 180° with respect to the fixed counter. The counter was then returned to 90° and the cycle was repeated. Data from the latter three angles were analyzed separately and compared with the data from the first three angles to reveal any left-right aysmmetry in the apparatus. Since no statistically significant difference was found between the two sets of data, the quoted results were obtained by combining both sets.

In order to minimize the effect of drift in the electronic equipment, the angular position of the counter was changed at 1-h intervals. After each 6-h sequence the position of the window defining the 62-keV region in the stationary detector was recorded automatically to reveal any electronic drift during the preceding counting period. (If such drift was noticeable, the entire run was discarded.) A total of 42 runs, representing 252 h of counting time, were recorded in this fashion.

2. Analysis of Data

The multichannel analyzer spectra corresponding to each angular position of the movable counter were examined, and the channel numbers corresponding to the 227-keV region and to a region above the 227-keV peak (Fig. 7, Regions II and III) were located. A computer was used to sum the counts between the indicated limits for each region, both in the singles spectrum $N_s(\theta_i)$ and in the coincidence spectrum $N_c(\theta_i)$. The normalized counting rate $n(\theta_i) = N_c(\theta_i)/N_s(\theta_i)$ at each angular position was thus obtained. For each run, the rates $n(\theta_i)$ $(i=1, \dots, 6)$ were least-squares fitted by computer to the usual Legendre polynomial expansion

$$n(\theta)/A_0 = 1 + A_2 P_2(\cos\theta) + A_4 P_4(\cos\theta).$$

The coefficients A_i and their associated errors were calculated according to the method described by Rose.²⁰ The values thus obtained for each run were then combined, with each value weighted according to its calculated error, to form the measured correlation coefficients for the two regions.

The results obtained for Region III were taken to represent the correlation of the spectral background included in the 227-keV region. The backgroundcorrected correlation coefficients A_{ν} were obtained from the measured coefficients A_{ν}^{m} for the region of interest and the measured coefficients A_{ν}^{b} for the spectral background by use of the relation

$$A_{\nu} = (1+K)A_{\nu}^{m} - KA_{\nu}^{b},$$

where K represents the ratio of photopeak area to background area included in the region of interest. (Because of the directional correlation, K is a function of the angle between the two counters, and the value used must represent an average over all angles.) The value for K was obtained by adding together the coincidence spectra taken at several angles and then decomposing this composite spectrum (Fig. 7) into a Gaussian photopeak and a smooth spectral background.

The background-corrected values of A_2 and A_4 for the 227-63.7-keV cascade were corrected for the detector solid angle by use of the calculations of Stanford and Rivers.²¹ The final corrected coefficients for the 227-63.7-keV directional correlation are

$$A_2 = 0.047 \pm 0.05$$
,
 $A_4 = -0.041 \pm 0.06$.

3. Interpretation

Evidence for the spins of the various states in Re¹⁸⁸ will be discussed in Sec. IV. For the present purpose, we shall assume that the ground state and first excited state have $I^{\pi}=1^-$ and 2^- , respectively. From beta decay evidence, the spin of the 290-keV level appears to be either 0 or 1. The calculated directional-correlation coefficients for the spin sequences 1(1,2)2(1,2)1 and 0(2)2(1,2)1, corresponding to spin 1 and spin 0, respectively, are shown in Fig. 8. This figure presents a

²⁰ M. E. Rose, Phys. Rev. 91, 610 (1953).

²¹ A. L. Stanford, Jr., and W. K. Rivers, Jr., Rev. Sci. Instr. 30, 719 (1959).





parametric representation of A_2 versus A_4 for various values of δ_1 and δ_2 , where δ for each transition is defined as the ratio of the reduced matrix element for emission of (L+1) radiation to that for emission of L radiation.

From the figure it is seen that the two spin sequences can be distinguished by the sign of A_4 if $A_4 \neq 0$. The corrected experimental result obtained for the 227-63.7keV directional correlation is shown as the hatched area in Fig. 8. Because the measured value of the coefficient A_4 in this experiment is not statistically different from zero, this experimental value does not uniquely define the spin sequence. However, for each sequence the measurement specifies a possible range for the mixing parameters. For the 0(2)2(1,2)1 sequence, δ_2 is confined to the range $0.17 < \delta_2 < 0.28$. For a 1(1,2)2(1,2)1 sequence, both transitions can be mixed, and the mixing parameters determined by these measurements are not independent. An inspection of Fig. 8 reveals that, in general, for any value of $\delta_1(\text{or } \delta_2)$, then $\delta_2(\text{or } \delta_1)$ must lie in the approximate range $0.1 < \delta < 0.3$.

In measuring the directional correlation of a cascade involving a low-energy transition from the intermediate state, there is always the possibility that the measured correlation will be attenuated. Such attentuation results from the interaction of the intermediate state with extranuclear fields and is influenced by the lifetime of the intermediate state as well as the chemical environment of the source. In the present experiment, a liquid source of low viscosity was used to minimize the attenuation; but the lifetime of the 63.7-keV level is not known to be sufficiently short to completely preclude the possibility of this effect.

III. STUDY OF Re^{188m} (18.5 MIN)

A. Introduction

At the time this work was undertaken, little had been reported on the 18-min isomeric activity in Re¹⁸⁸. Early studies by Mihelich⁵ and by Flammersfeld⁴ had indicated the presence of two gamma transitions at 63 and 105 keV. Flammersfeld, whose absorption methods did not reveal the higher energy radiation, concluded that there was a level about 60 keV above the ground state and assumed it to be the isomeric state. He also measured a value of about 2 for the ratio of charged-touncharged radiations and accepted this as representing the total internal-conversion coefficient of the 63-keV transition. A tentative assignment⁷ of spins, based in part on this coefficient, was made. In point of fact, the photons at 62 keV are largely $K \ge 10^{-10}$ x rays resulting from internal conversion of two transitions (with energies of 93 and 106 keV) which populate the 63.7-keV state. In the light of what is now known of the decay scheme. it is clear that the ratio of charged-to-uncharged radiations published by Flammersfeld cannot be used as the internal-conversion coefficient of the 63.7-keV transition. Instead, it more nearly represents an average of the total conversion of all three principal gamma transitions. The theoretical L+M internal-conversion coefficient¹⁶ for the 63.7-keV transition is about 3.6 (on the assumption of pure M1 character). The equations that represent the decay relationships show that a measurement of the total conversion is relatively insensitive to the conversion of the 63.7-keV transition. The fact that Flammersfeld measured a value close



to the correct one must be looked upon as fortuitous. We have therefore avoided the use of that datum in our analysis.

Mihelich,⁵ in his study of the Re¹⁸⁸ isomer, reported observing internal-conversion-electron groups corresponding to two transitions of 63.5 and 105 keV. From scintillation measurements he also noted two gammaray peaks, one corresponding to the 105-keV transition and the other corresponding to the combination of a 63.5-keV gamma ray and K x rays accompanying the conversion of the 105-keV transition. He comments that his data are insufficient to specify the sequence of the cascade from the state at 169 keV. Since Flammersfeld did not observe the 105-keV transition, his conclusion that a state exists at about 60 keV must be reexamined in the light of Mihelich's observation. Likewise, the tentative interpretation of these data included in the Nuclear Data Sheets must be re-examined.

Studying sources of Re^{188m} with a bent-crystal spectrometer, Hardell¹⁷ assigned an energy of 63.7 keV to the principal gamma ray present in that activity. This is considered to be the most precise measurement of the energy of this radiation and is accepted for the purpose of this report. Hardell was unable to observe the 105-keV gamma ray (or any other radiation) with his instrument.

Sources of Re^{188m} were prepared in the Argonne CP-5 reactor by neutron activation of samples of rhenium enriched in Re¹⁸⁷. The gamma-ray singles and coincidence spectra were examined with the apparatus previously described (Sec. II.D.1). Internal-conversionelectron spectra were studied with photographic spectrographs located at the MTR Reactor, Arco, Idaho. From these experiments it is concluded that the isomeric activity involves four excited states in Re¹⁸⁸.

B. Scintillation Studies of Re^{188m}

1. The Gamma-Ray Spectrum

The gamma-ray spectrum obtained with the collimated 4×4 -in. NaI(Tl) crystal is shown in Fig. 9. The recorded spectrum included channels covering the 478-

and 633-keV peaks associated with the 18-h groundstate activity. After the 18-min spectrum was accumulated, the source was allowed to remain undisturbed in the source holder for several hours until only the 18-h activity remained. The residual radiations were then subtracted electronically from the spectrum stored in the memory. Subtraction was continued until no evidence of the 478- and 633-keV Os188 peaks remained. The spectrum was then examined in the region of 150-180 keV. As a result of this experiment, an upper limit of less than 1.0% (relative to the 106-keV gamma ray) is placed upon any possible transition to the ground state from levels at 156, 169, or 171 keV. After this subtraction of the 18-h background, the spectrum shown in Fig. 9 remained. These experiments included an examination of the low-energy region as well. However, no radiations could be found between about 15 keV and the 62-keV peak.

This spectrum (Fig. 9) was analyzed by computer in the manner previously described. The results of this analysis of the scintillation spectrum yielded the values of 92.8 ± 0.6 and 105.9 ± 0.3 keV for the radiations constituting the upper peak. In the figure, the smooth curve represents the synthesized spectrum that resulted from fitting the composite 93-106-keV gamma peak.

The measured gamma-ray energies and intensities are listed in Table V. The intensities are corrected for detector response and are normalized to unity for the composite peak representing the 63.7-keV gamma ray plus the K x ray.

2. Gamma-Gamma Coincidence Measurements

The 4096-channel two-parameter analyzer was used to explore the coincidence relationships between the three gamma transitions with energies of 63.7, 93, and 106 keV. Examination of the spectrum showed conclusively that the 93- and 106-keV radiations were both in coincidence with the 62-keV peak. It is also evident that the 93- and 106-keV radiations were not in coincidence with each other. Self-coincidences were observed in the 62-keV peak. These are attributed to coincidences between the 63.7-keV gamma ray and Kx rays following internal conversion of the two higher energy coincident gamma rays. (The experimental data from which the foregoing conclusions are derived are not displayed.)

TABLE V. Gamma rays in Re188m.

Energy (keV)	Intensity	
$ \begin{array}{c} 105.9 \pm 0.3 \\ 92.8 \pm 0.6 \\ 63.7^{a} \\ +K x ray \end{array} $	$\begin{array}{c} 0.116 {\pm} 0.006 \\ 0.055 {\pm} 0.003 \\ 1.0 \ \pm 0.02 \end{array}$	

* Reference 17.



FIG. 10. Decay scheme for $_{74}W^{188}$ and the energy levels in $_{75}Re^{188}$.

3. Internal-Conversion-Electron Measurements

The internal-conversion-electron spectrum of the 18.5-min activity was investigated with photographic electron spectrographs. Table VI summarizes the results. In addition to lines from the 155.03-keV transition in Os¹⁸⁸, groups corresponding to four transitions in Re¹⁸⁸ were observed. The K line of the 155.03-keV and the L_1 line of the 63.7-keV transitions (both measured by a crystal spectrometer)^{17,18} serve as fiducial marks for the region of the plate in which most of the lines are located. However, there is some uncertainty concerning the very low-energy end as evidenced by the disagreement between the K and L lines of the 93-keV transition.

The line at 11 keV is very weak and subject to considerable uncertainty because of the low energy; it is

TABLE VI. Internal-conversion-electron data on Re^{188m} from spectrographic measurements.

	Tino		Interpretation			
Line energy (keV) 1 11.0	Shell	Energy (keV)	Transition energy (keV)			
	$M_1 + M_3$	14	14.0 ± 1.0			
2 3 4	51.0 60.6 63.0	L_1 M_1 M_2	$\begin{array}{c} 63.5 \\ 63.4 \\ 63.6 \end{array}$	63.5±0.2	63.7	
5	22.9 79.8	\widetilde{K}	94.6	92.3±0.8		
7 8	34.4 93.0	\tilde{K}	106.07	105.8 ± 0.6	105.9	
9 10	81.1 142.3	\overline{K} L ₂	155.0	154.8 ± 0.2	155.03	
11 12	143.8 152.1	$L_3 \\ M_3$	154.8 154.6			

interpreted only as evidence of a transition of about 14 ± 1 keV.

C. Lifetime of the 63.7-keV Level

The lifetime of the 63.7-keV level in Re^{188m} has been investigated by Bolotin in cooperation with the present authors. An upper limit $T_{1/2} \leq 2 \times 10^{-9}$ sec was obtained by observing the 106-63.7-keV gamma-ray cascade. NaI detectors were used in conjunction with a time-to-pulse-height converter of conventional design.

IV. EXPERIMENTAL JUSTIFICATION OF THE DECAY SCHEME

In evolving a self-consistent decay scheme (Fig. 10), the results of the various experimental studies are invoked when they are applicable.

A. Specification of Energy Levels in Re¹⁸⁸

The experiments confirm placement of a level at 63.7 keV. Both the internal-conversion and the scintillation studies clearly show the presence of a 63.7-keV transition in both the 18.5-min and in the 69.4-day activities. Since it is the only transition common to both activities, it must represent decay to the ground state from a level at 63.7 keV in Re¹⁸⁸.

In addition to the persuasive evidence outlined in the previous paragraph, several other experimental results support the existence of a 63.7-keV level. An absorption study of the charged radiation in coincidence with the 62-keV peak (data not shown) indicated the presence of a beta-ray component of approximately 290 keV. Similarly, the beta-gamma coincidence spectrum (Fig. 6) obtained with the two-parameter analyzer also shows the presence of this beta component. The implication that follows these observations is evident considering the energy of 349 keV for the ground-state beta transition.

The level at 290 keV is specified by the scintillation studies carried out on the 69.4-day activity. Coincidences between the 227- and 63.7-keV gamma rays suggest that the 290-keV gamma ray represents a transition to the ground state. Placement of this level is also supported by failure to observe coincidences between the 290-keV radiation and any other gamma rays. The existence of a 290-keV level is also inferred by Roy³ from similar reasoning.

The 106-keV gamma ray is found to be in coincidence with the 63.7-keV transition. There is no evidence of any other transition that might occur between the 106-keV and the 63.7-keV transitions. Therefore a state is specified at 169.6 keV.

The 93-keV gamma ray is also in coincidence with the 63.7-keV transition. As with the 106-keV transition, the absence of any other visible radiations suggests that a level should be placed at 156.5 keV. However, the weak internal-conversion line at 11 keV would tend to suggest one of two alternatives: There is either a level at 156 keV, as postulated here, or possibly a level at 77 keV. The arguments upon which this latter alternative is rejected are based upon a knowledge of the character of the 63.7-, 93-, and 106-keV radiations.

Specification of the isomeric state at 171 keV is based upon inferences drawn from detailed analysis[®] of the data and from consideration of its lifetime and spin. We have no direct experimental observation of the transition to the 169-keV state and only the weak 11-keV conversion line from the 14-keV transition to the 156-keV state. Therefore, since our observations permit us only to specify the energy of the isomeric state to lie between 170 and 183 keV, we have assumed the value of 171.5 keV based upon the measurements of Takahashi⁸ *et al.* The arguments supporting the placement of the isomeric state will be presented in the following sections.

B. Character of the Transitions

1. Character of the 63.7-keV Transition

The upper limit of about 2 nsec for the lifetime of the 63.7-keV state precludes the possibility of any type of transition except E1, M1, or E2, or a mixture of the latter two. Interpretation of the internal-conversionelectron data confirms the assignment⁷ of M1 character to the 63.7-keV transition. The L_1 line is the strongest group seen, both in the 18.5-min and in the 69.4-day activities. A line that may be the L_3 line is weakly visible on the 69.4-day plate $(L_1/L_3>6)$; this is attributed to a small (<2%) admixture of E2 in the transition.

2. Character of the 106-keV Transition

The M1 character of the 106-keV transition can be deduced without any assumptions concerning the 63.7keV transition (i.e., regardless of the internal-conversion coefficient of the 63.7-keV transition). An upper limit of the multipole order of the 106-keV transition can be specified by assuming that the entire 62-keV photopeak is due to $K \ge 100$ arising from conversion of the 106-keV transition. (That is, assume that the 63.7-keV transition is completely converted and that the 93-keV transition is completely unconverted.) After appropriate detector corrections, the intensity ratio of the two radiations, $I_{63}/I_{106} = 8$, represents an upper limit for the K internalconversion coefficient of the 106-keV transition. The theoretical value for an M2 transition of this energy is 24, so that all magnetic transitions except M1 are ruled out.

Only K and L_1 internal-conversion lines are observed for the 106-keV transition. This fact eliminates quadrupole and all higher order electric transitions. For E1 transitions of this energy, the L_2 and L_3 lines are somewhat weaker than the L_1 line; hence, the E1 possibility cannot be ruled out on the basis of the conversion data alone. If the 106-keV transition is postulated to have E1 character, only 5% of the 62-keV photopeak can be attributed to K conversion of the 106-keV transition. The 93-keV transition would then have to have at least M2 character in order to account for the residual intensity in the 62-keV peak (even if the 63.7-keV transition were totally unconverted). Under these conditions, one would find the L_1 line of the 93-keV transition at least ten times as intense as the L_1 line of the 106-keV transition. In point of fact, the conversion lines of the 93-keV transition are somewhat weaker than those of the 106-keV transition. Thus the possibility that the 106-keV transition has E1 character is also eliminated. It is therefore concluded that this transition is of M1character.

3. Character of the 93-keV Transition

The M1 character of the 93-keV transition is deduced through a line of reasoning similar to that presented in the previous two paragraphs for the 106-keV transition.

4. Relative Intensity of the 106- and 93-keV Transitions and the Total Internal-Conversion Coefficient of the 63.7-keV Gamma Ray

The relative branching of the 106- and 93-keV transitions to the 63.7-keV level is calculated to be 60 and 40%, respectively. These values are based on the conclusion that both transitions have pure M1 character.

The total internal-conversion coefficient for the 63.7keV transition can be calculated from the relative intensities listed in Table V, and from the conclusion that both transitions populating this level have M1 character. The value of 3.0 so calculated is in satisfactory agreement with a theoretical value of 3.6 for a pure M1 transition of 64 keV.

5. Character of the 227- and 290-keV Transitions

The spin and parity of the 290-keV state is restricted to 0^- , 1^- , 2^- , 0^+ , or 1^+ ; any other choice is definitely excluded by a cursory consideration of the log*ft* value of the beta branch populating this state.

Examination of the relative intensities of the gamma rays in the singles spectrum shows that the 0^+ possibility can be eliminated. In this case the 227-keV transition would have pure M2 character and the composite 62-keV peak would have nearly twice the observed intensity, even in the absence of beta branching directly to the 63.7-keV level. Further, the 0⁺ possibility can be ruled out on the grounds that the 227-keV M2 transition would not compete with the E1 290-keV groundstate transition. The 1⁺ possibility for the spin of the 290-keV level cannot be eliminated from consideration of the gamma-ray intensities, since any residual intensity in the 62-keV peak can readily be ascribed to beta branching directly to the 63.7-keV level, and there is considerable uncertainty as to the true intensity of this branch. The gamma-gamma coincidence data were used to determine the K-shell internal-conversion coefficient for the 227-keV transition; the value so measured for α_{κ} is 0.6±0.3. Although there is sufficient uncertainty in this measurement to discourage making a choice between M1 and E2 character, it is adequate evidence to exclude the possibility of E1 and thus eliminates the spin assignment of 1+.

Among the remaining choices $(0^-, 1^-, \text{ or } 2^-)$ for the spin of the 290-keV state, the third choice can be rejected with reasonable confidence on the basis of the $\log ft$ for the 59-keV beta branch. The remaining choice then hinges entirely upon the character of the 227-keV transition.

If the spin and parity of the 290-keV state are restricted to either 0^- or 1^- and the M1 character of the 290-keV transition is invoked, the nature of the 227keV transition can be deduced from a qualitative examination of the beta-gamma coincidence data (Fig. 6). Whether or not the 227-keV transition is M1 or E2, its total L conversion coefficient is only about 0.07. For either case, only about 15% of the conversion line at 220 keV can be attributed to the 227-keV transition (from the consideration of relative gamma-ray intensities). The two peaks at about 150 and 220 keV (Fig.6) are nearly equal in area. This is the condition expected if the 227-keV transition is of M1 character; if it were E2, the lower peak would be only about one-fourth as large. Therefore, the 227-keV transition is probably also of M1 character.

C. Assignment of Spins

1. Ground State of Re^{188} ($I^{\pi} = I^{-}$)

The ground state of Re^{188} is assigned spin and parity $I^{\pi}=1^{-}$. As described in the section on the beta decay (Sec. II. B.5), our experimental results confirm the earlier assignment of 1⁻. This conclusion, based on all of these measurements, is now substantiated by a direct atomic-beam measurement by Doyle and Marrus.²² They find the ground state of Re^{188} to have spin I=1.

2. 63.7-keV Level $(I^{\pi}=2^{-})$

The spin $I^{\pi} = 1^{-}$ of the ground state together with the M1 character of the 63.7-keV transition, restricts the spin of the 63.7-keV state to 0, 1, or 2, with negative parity. The intensity of the 285-keV beta branch could not be measured directly. The calculation of the branching into the 63.7-keV level employed the measured intensities of the 290- and 227-keV gamma rays and the intensity of the composite 62-keV radiation. To provide for either possible spin assignment $(0^{-} \text{ or } 1^{-})$ for the 290-keV level, the logft was computed on the assumption that the 227-keV transition is M1 and also under the alternate assumption that it is E2. If the 290-keV state has spin $I^{\pi}=0^{-}$, then $\log ft=9.0$ for the 285-keV beta branch. If the spin of the upper state is 1⁻, then $\log ft = 9.2$. Under either assumption, the transition falls into the class of unique first-forbidden transitions, $\Delta I^{\Delta \pi} = 2$, yes. Thus the spin and parity of the first excited state is well defined. This interpretation agrees with Roy's value for the $\log ft$ of the 285-keV beta branch.

3. Level at 156 keV $(I^{\pi}=3^{-})$

The M1 character of the 93-keV gamma transition restricts the possible spin assignment for the 156-keV level to 1⁻, 2⁻, or 3⁻. A preference for the choice 3⁻ follows from failure to observe the 156-keV crossover transition to the ground state. The choice 1⁻ is also clearly excluded from considerations of the beta decay; if this level had spin 1, it would most certainly be populated directly from the ground state of W¹⁸⁸. Although with considerably less force, the same argument also applies to the choice of 2⁻.

4. Level at 169 keV $(I^{\pi}=3^{-})$

The assignment of 3^- for the 169-keV level follows from a series of arguments identical to those in the previous paragraph for the 156-keV level.

5. Level at 171 keV $(I^{\pi}=6^{-})$

Assignment of the spin $I^{\pi}=3^{-}$ to the 169-keV level clearly indicates that it cannot be the isomeric state. From lifetime considerations alone, it is necessary to postulate another more energetic level with higher spin.

²² W. M. Doyle and R. Marrus, Nucl. Phys. 49, 449 (1963).

The fact that we observed no low-energy gamma rays and only the single 11-keV conversion line implies that the isomeric state must lie but slightly above the 169keV level. The only type of transition compatible with these conditions requires that the isomeric state have spin and parity $I^{\pi} = 6^{-}$.

Since the 156-keV level must be populated by a transition with a minimum energy of at least 13 keV, the 14 ± 1.0 -keV transition was interpreted as a transition to this level. Because of the energy uncertainty in the 14-keV transition, it could not be stated whether this decay occurred from the 169-keV level or directly from the isomeric state. If the first of these two possibilities is postulated, it is clear that the isomeric level could lie anywhere between 170 and 183 keV. (This conclusion is based on the assumption that conversion electrons from the isomeric transition to the 169-keV level were either unresolved from those of the 11-keV line or so low in energy as to undetectable.) If the second postulate is invoked (i.e., that the 14 ± 1.0 -keV transition represents decay directly from the isomeric level to the 156-keV state), then it is clear that the isomeric level must lie between 170 and 172 keV. Both of these possibilities present to anomaly of nearly equal competition between two similar gamma transitions of widely differing energy. One other conclusion is readily evident from the data. The energies and spins of the first two excited states at 63.7 and 156.5 keV indicate that these are probably members of a rotational band based on the ground state. This observation indicates some preference for interpreting the 14-keV transition as decay directly from the isomeric level to the 156-keV state but not with sufficient persuasion to justify this choice with certainty.

It was at this point in the analysis of our data that the results of the experiments by Takahashi et al.8 appeared in the literature. Their direct observation of internal-conversion electron groups representing transitions directly from a level (isomeric) at 171.5 keV to both the 169.6- and the 156.5-keV levels resolved the dilemma present in our data.

6. Level at 290 keV $(I^{\pi}=1^{-})$

Unfortunately, the directional-correlation measurement on the 227-63.7-keV cascade (Sec. II.E) proved not be definitive in determining the spin of the 290-keV level. The choice of $I^{\pi} = 1^{-}$ for the spin of the 290-keV level is indicated by the fact that both the 290- and the 227-keV transitions have M1 character. Also, if the 290-keV state has spin $I^{\pi}=0^{-}$, one would expect that the 227-keV γ branch to the 63.7-keV level would probably be considerably depressed.

V. INTERPRETATION OF THE DECAY SCHEME

A. Introduction

There are few signposts leading to the interpretation of the observable properties of odd-odd nuclei. Except

for some treatments of the ground states, there is no comprehensive review of the available data and there is no general agreement regarding the propriety of possible nuclear models. Coupling rules based on the asymptotic quantum-number description²³ of nuclei in the deformed region have achieved a measure of success with regard to ground states. It seems not unreasonable to attempt to apply the model to the Re¹⁸⁸ nucleus, although its applicability might be challenged on the ground that Re¹⁸⁸ is too near the upper end of the deformed region. However, rotational properties have been found in a number of nuclides of equal and greater mass, such as Os¹⁸⁸, Os¹⁹⁰, Ir¹⁸⁹, and perhaps even Ir¹⁹¹. The following interpretation is thus based upon the available Nilsson levels in the neighborhood of the 75th proton and the 113th neutron.

Our data substantiate the earlier assignment of 1as the spin and parity of the ground state of Re¹⁸⁸. In following the usual practice of investigating the probable character of the last odd proton and neutron, an examination of the available data shows that the ground states of W187, Os189, Pt191, and Hg193 all have groundstate spins of $\frac{3}{2}$. For the first two, the assignments are based on direct measurements, while those for the latter two are inferred from decay-scheme studies. The 113th neutron is thus assigned to the Nilsson level $\frac{3}{2}$ [512].

Spin and parity of $\frac{5}{2}$ are assigned to both Re¹⁸⁵ and Re¹⁸⁷; the 75th proton is thus associated with the Nilsson level $\frac{5}{2}$ [402]. Unlike the earlier Nordheim rules²⁴ based on a *jj*-coupling model, the coupling rules for odd-odd nuclei postulated by Gallagher and Moszkowski⁹ assume that the coupling of the last odd neutron and proton is governed by the asymptotic quantum numbers applicable to deformed nuclei. For convenience these rules are restated here.

Rule I:
$$I = \Omega_p + \Omega_n$$
 if $\Omega_p = \Lambda_p \pm \Sigma_p$ and $\Omega_n = \Lambda_n \pm \Sigma_n$.
Rule II: $I = |\Omega_p - \Omega_n|$ if $\Omega_p = \Lambda_p \pm \Sigma_p$ and $\Omega_n = \Lambda_n \mp \Sigma_n$.

Here Λ_p and Λ_n are the projections of the orbital angular momentum of the odd proton and neutron on the symmetry axis, Σ_p and Σ_n are the projections $(\pm \frac{1}{2})$ of their intrinisic spins along the nuclear symmetry axis, and Ω_n and Ω_n are the sums of the axial components for each particle.

The effective intrinsic state of the odd-odd nucleus is formed by coupling the Ω_p and Ω_n either parallel or antiparallel. It is assumed that those states in which the spin projections (Σ_p, Σ_n) lie parallel are more tightly bound. The interpretations which follow are examined in the light of these rules. (We use the notation of Gallagher and Moszkowski; the arrow up or down, following the aymptotic quantum numbers, indicates whether Σ and Λ are parallel or antiparallel.)

²³ B. R. Mottelson and S. G. Nilsson, Kgl. Danske Videnskab. Selskab, Mat. Fys. Skrifter I, No. 8 (1959). ²⁴ L. W. Nordheim, Phys. Rev. 78, 294 (1950).

B. Interpretation of the Levels in Re¹⁸⁸

1. Ground State

The ground-state spin $I^{\pi}=1^{-}$ is explained by antiparallel coupling of the $\frac{5}{2}+[402]\uparrow$ proton to the $\frac{3}{2}-[512]\downarrow$ neutron in conformity with Rule 2.

2. Levels at 63.7 and 156 keV

The two excited states at 63.7 and 156 keV are concluded to represent the first and second levels of a rotational band based on the K=1 ground state. Their energy separation conforms to the I(I+1) rule within about 2%.

The ratio of reduced transition probabilities for gamma transitions of identical multipolarity L between an initial state J_i , K_i and two final states J_f , K_f and J_f' , K_f of the same rotational band is given by the ratio of two Clebsch-Gordan coefficients, i.e.,

$$\frac{B(L,J_i \to J_f)}{B(L,J_i \to J_f')} = \left| \frac{(J_i L K_i K_f - K_i | J_i L J_f K_f)}{(J_i L K_i K_f - K_i | J_i L J_f' K_f)} \right|^2$$

The theoretical value for the ratio of the reduced transition probability for the 227-keV to that for the 290-keV transitions is $B(1,1 \rightarrow 2)/B(1,1 \rightarrow 1)=1$, the experimental ratio is 1.17.

3. Level at 169 keV

There are three other nearby Nilsson levels in the region of deformation where the 113th neutron occupies the [512] orbital. The remaining excited states in Re¹⁸⁸ can be explained by assuming promotion of a neutron to one or another of these nearby levels.

The 3⁻ state at 169 keV can be formed by coupling a neutron in the $\frac{1}{2}$ -[510] \uparrow orbital to a proton in the $\frac{5}{2}$ +[402] \uparrow orbital. The transition rate from the 6⁻ state at 171-keV to the 169-keV level is sensitive to any uncertainty in the measurement of the 2-keV energy difference between these states. Considerable uncertainty also exists in the correction for internal conversion of a transition of this low energy. However, the

observed rate for the 2-keV transition is within an order of magnitude of the single-particle estimate. Conversely, the 15-keV transition to the 156-keV state with I=3, K=1 is hindered by a factor of about 10⁶. Such a retardation for this transition is not unexpected in consideration of the K selection rule.

4. Level at 171.5 keV

The 6⁻ isomeric state at 171.5 keV is believed to arise from coupling the $\frac{5}{2}$ +[402]↑ proton to a neutron promoted to the $\frac{7}{2}$ -[503]↑ level. When the coupling is parallel, these particles conform to Rule I.

5. State at 290 keV

When the two particles that form the 6^- state are coupled antiparallel, they can form the 290-keV level with spin 1⁻. In conformity with the predictions of the rules, the state formed by coupling the spins antiparallel lies higher than that formed by the parallel coupling.

From energy and spin considerations alone, one would not expect to observe transitions from the 290-keV state to either of the 3^- levels.

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