

New 20-Hour Electron-Capturing Rhenium Isotope, Re^{181} †

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A new 20 ± 2 hour electron-capturing rhenium isotope has been investigated. A mass assignment to Re^{181} is made from the energy threshold for its production by alpha-particle bombardment of Ta^{181} and by the chemical separation and identification of its radioactive daughter, W^{181} . A very prominent 365.5-keV transition is observed in the gamma-ray and electron spectra. It is deduced from the 365.5-keV gamma to K x-ray intensity ratio that more than 80% of the electron capture populates the level at 365.5 ± 0.2 keV above the ground state. This level apparently decays directly to ground by a delayed $M2$ transition, and is believed to be the same 14-microsecond isomer observed by others from a (γ, n) reaction on W^{182} .

A possible interpretation of the 365.5-keV transition in terms of the odd-neutron states of a spheroidal well is given.

DURING an investigation of neutron-deficient rhenium isotopes¹ produced by 48-MeV alpha particles on tantalum, a very prominent 365.5 ± 0.2 -keV gamma-ray transition was observed which decayed with an approximately 20-hour half-life. Since a 17-hour activity had been observed when bombarding with alpha-particle energies above 40 MeV in a stacked-tantalum-foil excitation function,² and since no isotope Re^{181} had been previously reported,³ we assigned the activity to the electron-capture decay of a new isotope, Re^{181} . The 365.5-keV gamma associated with this decay is then a transition in W^{181} .

To verify this assignment two experiments were done. The first consisted of two stacked-foil excitation functions obtained by bombarding a stack of 0.001-inch tantalum (99.98% pure) with a very low-intensity beam of alpha particles in the Berkeley 60-inch cyclotron.

In the first experiment the foils were counted directly in a sodium iodide counter with a 50-channel differential pulse-height analyzer. The intensity of the 365.5-keV gamma ray in each foil was then plotted against the mean energy of the alpha-particle beam in each foil to give a rough excitation function. The threshold energy was approximately 33 MeV, and the curve was still rising at the full energy of the cyclotron, ~ 48 MeV. This threshold and the shape of the curve clearly must correspond to a $\text{Ta}^{181}(\alpha, 4n)\text{Re}^{181}$ reaction. The excitation function is illustrated in Fig. 1.

The second stacked-foil bombardment was used to determine the half-life of the isotope. Much greater activities were produced, and carrier-free chemical separation to obtain pure rhenium was performed⁴ on the first foil. The decay of the 365.5-keV transition was followed in a double-focusing beta-ray spectrometer at

0.3% resolution and on the previously mentioned 50-channel gamma analyzer. The results obtained by the two methods were in good agreement. The half-life determined was 20 ± 2 hours. The L_{II} and L_{III} lines were observed in very, very weak intensity.

The second experiment was conducted several weeks after the second bombardment. Tungsten carrier was added to the previously separated pure rhenium activity. WO_3 was precipitated and the precipitate was gamma-analyzed. A single peak was observed at the energy of tantalum K x-rays. Since W^{181} , the only radioactive tungsten isotope in this region, has been observed to decay almost entirely to the ground state of Ta^{181} ,⁵ and since the long-lived rhenium isotopes exhibit com-

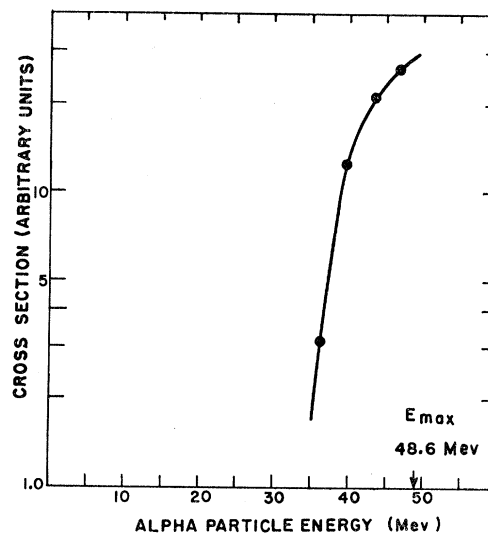


FIG. 1. Excitation function (in arbitrary units of σ) for the $\text{Ta}^{181}(\alpha, 4n)\text{Re}^{181}$ reaction. E_{\max} indicates the maximum alpha particle energy produced by the cyclotron. The function was determined from the relative intensity of the 365.5-keV gamma ray in a series of 0.001-inch foils.

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¹ Thulin, Rasmussen, Gallagher, Smith, and Hollander, *Phys. Rev.* **104**, 471 (1956).

² M. Sweeney and J. O. Rasmussen, Chemistry Division Quarterly Report, University of California Radiation Laboratory Report UCRL-2932, 1955 (unpublished).

³ Hollander, Perlman, and Seaborg, *Revs. Modern Phys.* **25**, 469 (1953).

⁴ Giles, Garrison, and Hamilton, *J. Chem. Phys.* **18**, 995 (1950).

⁵ Cork, Nester, LeBlanc, and Brice, *Phys. Rev.* **92**, 119 (1953); Bisi, Ferrani, and Zappa, *Nuovo cimento* **1**, 651 (1955), and **3**, 661 (1956); Derbrunner, Heer, Kündig, and Rüetschi, *Helv. Phys. Acta* **29**, 235 (1956).

plex gamma spectra,¹ we feel that this experiment further established the activity first observed as Re¹⁸¹. The half-life of the *K* x-ray peak is in agreement with the 120-day half-life reported for the W¹⁸¹, but we have not followed it for a long enough period to verify this value.

The Re¹⁸¹ as ordinarily produced is mixed with both Re¹⁸² isomers and the long-lived Re¹⁸³.⁵ By bombarding a stack of 0.001-inch tantalum foils we produced isotopes in different abundances in each foil. The different ratio of intensities of the various transitions in each foil allowed us to set a lower limit on the 365.5-keV to total electron-capture decay ratio. If we assume a total electron capture decay energy of ~ 1.0 MeV, which is in agreement with Coryell's beta decay-energy systematics,⁶ we calculate an L_I/K capture ratio of ~ 0.17 to the 365.5-keV state. Using our *K* x-ray to 365.5-keV gamma ratio of ~ 1.7 , we calculate that more than 80% of the electron capture goes to this state. To obtain this limit it was assumed that no decay occurs to this state from higher energy states. This limit is rendered somewhat uncertain by the presence of other weaker gamma transitions in Re¹⁸¹ decay. We calculate from the above intensity limits and decay energy estimate that the $\log ft$ value for decay to the 365.5-keV state is of the order of 6.

Dr. D. Strominger performed coincidence studies on the Re¹⁸¹, using fast-slow coincidence pulse-analysis apparatus. He observed no coincidences between the 365.5-keV gamma and *K* x-rays, and from this result it was concluded that the half-life of the state giving rise to this gamma ray is greater than 10^{-7} sec.⁷

Recent studies of short-lived isomers produced by betatron excitation of natural tungsten^{8,9} have revealed a 366-keV gamma transition with a half-life of 14.4×10^{-6} sec. The transition was reported⁹ to have a *K*-conversion coefficient of 0.30 ± 0.03 . These workers assigned the transition to W¹⁸¹. Our work confirms this assignment, since the isomer is formed in decay of Re¹⁸¹. Bureau and Hammer⁹ suggest that the multipolarity of the 366-keV transition, as determined by their absolute conversion coefficient, agrees with either a 28% *E1*-72% *M2* or 65% *M2*-35% *E3* mixture. From the very low intensity of the L_{II} and L_{III} conversion electrons relative to the L_I we can rule out the *M2*-*E3* assignment. Their mixing ratio was calculated by using Rose's¹⁰ theoretical conversion coefficients,

which are now generally considered to be too high because of the neglect of a correction for finite nuclear size. Using Sliv's¹¹ *K*-shell internal-conversion coefficients and their same conversion coefficient of 0.30, we recalculate the mixture to be 14% *E1*-86% *M2*. Furthermore, the experimental limits of error given by Bureau and Hammer do not eliminate a pure *M2* assignment.

Bureau and Hammer also advance an explanation of the isomeric W¹⁸¹ state in terms of nucleon states in a spheroidal well as calculated by Nilsson.¹² They suggest that the transition observed corresponds to a transition between two Nilsson odd-neutron states, $7/2- \rightarrow 9/2+$, the predominant *M2* character resulting from a high degree of cancellation of *E1* transition matrix elements for the Nilsson states in question. Although retardation from single-particle formula rates¹³ occurs generally for low-energy *E1* transitions, the retardation (granting *E1* admixture of 14% here) of about 10^{10} would be exceptionally large for *E1* transitions not *K*-forbidden, retardations of 10^4 - 10^6 being the general rule.¹⁴ These lifetime considerations strengthen our alternative assignment of pure *M2* character to the isomeric transition. The *M2* transition is retarded by a factor of about 700.

Consideration of the Nilsson diagram for neutron states also shows a nearby $5/2-$ state which would give rise to a pure *M2* transition to the $9/2+$ ground state in W¹⁸¹, and we favor this state assignment for the isomer.

Another test of the assignment of the isomeric state can perhaps be obtained from the Re¹⁸¹ studies by using the $\log ft$ of ~ 6 for electron capture to the 365.5-keV state. This test is a consequence of the asymptotic-quantum-number beta-selection rules proposed by Alaga.¹⁵ If we postulate that the $5/2+$ odd-proton ground state, proposed for Re¹⁸³,¹ Re¹⁸⁵,¹⁶ and Re¹⁸⁷,¹⁶ is also the ground state of Re¹⁸¹, then decay to the $7/2-$ state proposed by Bureau and Hammer should theoretically exhibit a larger $\log ft$ value than decay to the $5/2-$ state, since for electron capture to these states the $5/2+ \rightarrow 7/2-$ beta transition is first-forbidden, hindered, while the $5/2+ \rightarrow 5/2-$ transition is first-forbidden, unhindered. The estimated $\log ft$ value of ~ 6 is in better agreement with the latter.

⁶ C. D. Coryell, *Annual Review of Nuclear Science* (Annual Reviews, Inc., Stanford, 1953), Vol. 2, p. 305.

⁷ D. Strominger (unpublished data, 1956).

⁸ S. H. Vegors, Jr., and P. Axel, *Phys. Rev.* **101**, 1067 (1956); Stewart, Bureau, and Hammer, *Bull. Am. Phys. Soc. Ser. II*, **1**, 206 (1956); also private communication to I. Perlman by P. Axel, S. H. Vegors, Jr., and R. B. Duffield of a list of (γ, n) reactions (as of September, 1956) producing metastable states in excited nuclei.

⁹ A. J. Bureau and C. L. Hammer, *Phys. Rev.* **105**, 1006 (1957).

¹⁰ M. E. Rose (privately circulated tables). Also (with G. H. Goertzel) in *Beta- and Gamma-Ray Spectroscopy*, edited by

K. Siegbahn (Interscience Publishers, Inc., New York, 1955), Appendix IV, p. 905.

¹¹ L. A. Sliv (privately circulated tables).

¹² S. G. Nilsson, *Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd.* **29**, No. 16 (1955).

¹³ S. A. Moszkowski, in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955), Chap. XIII, p. 391.

¹⁴ D. Strominger and J. O. Rasmussen, *Nuclear Phys.* **3**, 197 (1957).

¹⁵ G. Alaga, *Phys. Rev.* **100**, 432 (1955).

¹⁶ B. R. Mottelson and S. G. Nilsson, *Phys. Rev.* **99**, 1615 (1955).

We have observed some additional weaker gamma transitions which may be associated with the decay of Re^{181} , but because of the complexity of the gamma spectra of the Re^{182} isomers with similar half-lives, the isotope assignment of the weaker transitions in our samples is somewhat uncertain.

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Energy and Angular Distributions of Neutrons from the Interaction of 14.1-Mev Neutrons with Zirconium*

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The distributions in energy and angle of the neutrons emitted from the interaction of 14.1-Mev neutrons with zirconium have been obtained by using nuclear emulsion plates as detectors. The energy distribution of the low-energy nonelastic neutrons is Maxwellian. The angular distribution of the low-energy nonelastic neutrons (0.5–4.0 Mev) is found to be isotropic; whereas, that of the high-energy nonelastic neutrons (4.0–12.0 Mev) is peaked in the forward direction. The significance of these results in terms of the statistical model of the nucleus is discussed.

I. INTRODUCTION

THE main object of this experiment is to test the validity of the compound-nucleus concept—and particularly the statistical model—by measuring the distributions in energy and angle of neutrons emitted in the nonelastic interaction of 14.1-Mev neutrons with natural zirconium. The differential cross sections at several angles for elastic scattering are also obtained in this experiment.

None of previous experiments agrees completely with any one of the present models of the nucleus. The results of this experiment indicate that the compound-nucleus model seems to apply to at least 80% of the nonelastic interactions. These results are quite similar to the results for Ta and Bi obtained by Rosen and Stewart.¹

II. EXPERIMENTAL PROCEDURE

The experiment was performed by measuring the recoil proton tracks in nuclear emulsions. The tracks were obtained by exposing 200-micron Ilford C-2 emulsion plates to neutrons emitted from zirconium due to its interaction with the 14.1-Mev neutron beam obtained from the reaction $\text{H}^3(d,n)\text{He}^4$ in the Cockcroft-Walton generator at the Los Alamos Scientific Laboratory. The zirconium scatterer is of natural isotopic abundance and is in the form of a cylinder $1\frac{1}{4}$ inches long and $1\frac{1}{4}$ inches in diameter. The scatterer was oriented so that its axis of symmetry coincided with the collimator

axis. The background was evaluated by making an irradiation with the scatterer removed. The iron collimator, which was developed by Rosen¹ and his group, was used. The plates were arranged around the periphery of a circle whose center coincided with the center of the scatterer. The details of tracks measurement and analysis are given by Rosen.²

III. ANALYSIS

The cross section $\sigma(E,\theta)$ for the emission of neutrons between energies E and $E+dE$ at angle θ is determined from:

$$F(E,\theta)dE = \frac{\bar{F} n_{Zr} \sigma(E,\theta) V_{Zr}}{\langle r^2 \rangle_{Av}} dE, \quad (1)$$

where $F(E,\theta)dE$ = neutron flux at the scattering angle θ , \bar{F} = average primary neutron flux on the scatterer allowing for attenuation, n_{Zr} = number of nuclei per cm^3 in the zirconium scatterer, V_{Zr} = volume of zirconium scatterer, and $\langle r^2 \rangle_{Av}$ = average for the square of the distance from the center of the scatterer to the center of the scanned area. Here, the average primary neutron flux on the scatterer with length x is given by

$$\bar{F} = \frac{F_0}{x} \int_0^x e^{-\sigma' n_{Zr} x} dx = F_0 \frac{1 - e^{-\sigma' n_{Zr} x}}{\sigma' n_{Zr} x}, \quad (2)$$

where σ' is equated to the total cross section³ in evaluat-

² L. Rosen, *Nucleonics* **11**, No. 7, 32, and No. 8, 38 (1953).

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† On leave from Yonsei University, Seoul, Korea.

¹ L. Rosen and L. Stewart, *Phys. Rev.* **107**, 824 (1957).

³ *Neutron Cross Sections*, compiled by D. J. Hughes and J. A. Harvey, Brookhaven National Laboratory Report BNL-325 (Superintendent of Documents, U. S. Government Printing Office, Washington, D. C., 1955).