Nuclear Spectroscopy of Ta^{181}†*

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The nuclear levels of Ta^{181} were investigated by a study of the β decay of Hf^{181} and the electron capture decay of W^{181}. Evidence for weak M-shell conversion lines of an ~6-kev transition in the Hf^{181} was found with a β spectrometer. An investigation of the W^{181} decay with this instrument revealed strong M-shell conversion lines corresponding to a 6.25±0.3-kev transition. With the aid of additional evidence, the conclusion is made that the 476-kev transition in the Hf^{181} decay occurs between the 482-kev level and a new level at 6 kev. On using an argon proportional counter, a 6-kev γ ray was also found in the W^{181} decay. The conversion coefficient of this transition was determined to be $\alpha_T=44\pm7$. This conversion coefficient and the M-subshell conversion ratios indicate that the 6-kev transition is of $E1$ multipolarity. The 6-kev level is assigned as the 9/2− [514] Nilsson intrinsic state. It is also concluded that the previously reported 152-kev transition in the W^{181} decay occurs between a new 11/2− ($K=9/2-$) rotational level at 158 kev and the 6-kev level. From a measurement of the tantalum L/K x-ray intensity ratio, the W^{181} decay energy is found to be 176_{-22}^{+44} kev. The branchings of this decay to the various Ta^{181} levels are as follows: 158 kev (0.11%), 136 kev (0.067%), 6.25 kev (~35%), and ground state (~65%). All findings and proposals are consistent with predictions of the unified model of the nucleus.

I. INTRODUCTION

THE success of the Bohr-Mottelson-Nilsson "unified model"[1,2] in interpreting nuclear spectra of deformed nuclei has led to many investigations of nuclei in the mass regions where the deformations are known to be large. One of the most frequently studied[3–9] deformed nuclei is Ta^{181}. This nucleus has been investigated in studies of the Hf^{181} β decay and W^{181} electron capture decay as well as in Coulomb excitation experiments. However, there are still some open problems concerned with the Ta^{181} levels.

Boehm and Marmier[3] found a weak 476-kev transition in the Hf^{181} β decay and suggested it was probably an $M2$ transition occurring between a $\frac{1}{2}-$ intrinsic level at 958 kev and the well-known 482-kev level. Debrunner *et al.*,[4] however, reported that a coincidence experiment excluded the 476-kev transition from being in cascade with the 482-kev transition. In a study of the W^{181} decay, Cork *et al.*[5] found 136- and 152-kev transitions. On the other hand, Bisi *et al.*[6] subsequently reported seeing no evidence for these transitions. From L/K x-ray intensity ratio measurements, these authors estimated the electron capture decay energy to be 92±10 kev. Confirming Cork's work, Debrunner *et al.*[7] found both the 136- and 152-kev transitions, and established that they were not in cascade. The 152-kev transition, which is not known from the Hf^{181} decay, was interpreted by these workers as being $E1+M2$.

The present work was undertaken to resolve some of these questions. The results lead to a consistent Ta^{181} level scheme which also is in good agreement with nuclear model predictions.

II. APPARATUS AND SOURCES

The apparatus and sources used in these measurements will now be described briefly. A more complete description has been given elsewhere.[10]

The low-energy electron spectra from the Hf^{181} and W^{181} decays were studied with a 180° magnetic β spectrometer[11] having a momentum resolution $\Delta p/p=\sim0.8\%$. The spectrometer detector was a Geiger counter with a 10–20-$\mu g/cm^2$ Formvar foil window. The homogeneous field ring focusing β-ray spectrometer[12] was used to supplement some of the 180° spectrometer measurements.

For the study of very low-energy γ rays, two gas-filled proportional counters were built.[13] These counters are $14\frac{1}{2}$ in. long and 4 in. in diameter, and have 1-in. diam side windows of 3-mg/cm² mica. One counter is filled with a mixture of 90% argon and 10% methane

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[1] A. Bohr, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **26**, 14 (1952); A. Bohr and B. R. Mottelson, *ibid.* **27**, 16 (1953); S. G. Nilsson, *ibid.* **29**, 16 (1955).

[2] B. R. Mottelson and S. G. Nilsson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Skr. **1**, 8 (1959).

[3] F. Boehm and P. Marmier, Phys. Rev. **103**, 342 (1956).

[4] P. Debrunner, E. Heer, W. Kündig, and R. Rüetschi, Helv. Phys. Acta **29**, 463 (1956).

[5] J. M. Cork, W. H. Nester, J. M. LeBlanc, and M. K. Brice, Phys. Rev. **92**, 119 (1953).

[6] A. Bisi, S. Terrani, and L. Zappa, Nuovo cimento **1**, 651 (1955).

[7] P. Debrunner, E. Heer, W. Kündig, and R. Rüetschi, Helv. Phys. Acta **29**, 235 (1956).

[8] D. Strominger, J. M. Hollander, and G. T. Seaborg, Revs. Modern Phys. **30**, 585 (1958).

[9] *Nuclear Data Sheets* (National Academy of Sciences, National Research Council, Washington, D. C.).

[10] A. H. Muir, Jr., Ph.D. thesis, California Institute of Technology, Pasadena, California, 1960 (unpublished).

[11] H. E. Henrikson, Norman Bridge Laboratory of Physics, California Institute of Technology Special Tech. Rept., 1956 (unpublished).

[12] J. W. M. DuMond, Ann. Phys. **2**, 283 (1957).

[13] A. H. Muir, Jr., California Institute of Technology internal report, 1959 (unpublished).

at 1 atm pressure, and the other with a mixture of 90% xenon and 10% methane at $\sim\frac{1}{3}$ atm pressure.

A standard 1-in. diam×1-in. long NaI(Tl) scintillation crystal spectrometer was employed for some of the γ-ray measurements. However, to measure the L/K x-ray intensity ratio from the W^{181} decay with good accuracy, special provisions were necessary because of the low energy of the L x rays ($\sim$9 kev) and because of the presence of scattering of both K and L x rays. Figure 1 shows the arrangement used for the L/K x-ray measurement. The thin square (2.3 cm×2.3 cm×0.55 cm) NaI scintillation crystal was freshly polished with methyl alcohol, lightly coated with mineral oil, and mounted directly on the face of the photomultiplier tube. Fresh polishing of the crystal face was necessary to prevent surface losses[10,14] of the low-energy L x rays. The crystal was covered by a 0.25-mil aluminum foil, while the source was sandwiched between 0.4-mil Mylar. With this covering, attenuation of the L x rays was very slight. Cylindrical Plexiglas spacers S of various lengths were used to support the source at different distances from the crystal. In order to reduce x-ray scattering and attenuation, a plastic bag attached to the counter body permitted the spectrometer to be surrounded by a helium atmosphere. The entire assembly was housed in a cave of $\frac{3}{8}$ in. brass, $\frac{1}{16}$ in. tin, and 2 in. lead.

A standard fast-slow triple-coincidence circuit[15] with resolving time $2\tau=4.4\times10^{-8}$ sec was employed to study the 476-kev transition in the Hf^{181} decay.

Sources

Three different samples of W^{181} were used in this study: (1) About 2 mg of WO_3 enriched in W^{180} (6.6% abundance) obtained from Oak Ridge National Laboratory was irradiated for about 30 days in a flux of $\sim3\times10^{14}$ neutrons/cm²-sec at the Materials Testing Reactor (MTR) at Idaho Falls. (2) About 3 mg of the same material was irradiated for about 50 days in a flux of $\sim5\times10^{14}$ neutrons/cm²-sec at the MTR. (3) About 2 mC of carrier-free W^{181}, accelerator produced by the reaction $Ta^{181}(p,n)W^{181}$, was procured from the Nuclear Science and Engineering Corporation, Pittsburgh, Pennsylvania. Samples (1) and (2) were vacuum evaporated onto sheets of aluminized mica and aluminum foil, respectively. Sources cut from these sheets were used for the β-spectrometer investigations. Sample (3) was deposited on a Plexiglas sheet for the proportional counter source. This same activity was redeposited, sandwiched between 0.4-mil Mylar, for the scintillation counter measurements. A source from sample (1) was also checked with the proportional counter.

A sample (from Oak Ridge) containing about 7 mg of HfO_2 enriched in Hf^{180} (93% abundance) was irradiated for about 10 days in a flux of $\sim2\times10^{14}$ neutrons/cm²-sec at the MTR. Approximately 5 mg of the activity was vacuum evaporated onto aluminized mica for β-spectrometer and proportional counter uses. The source employed in the coincidence measurement was also made from this sample.

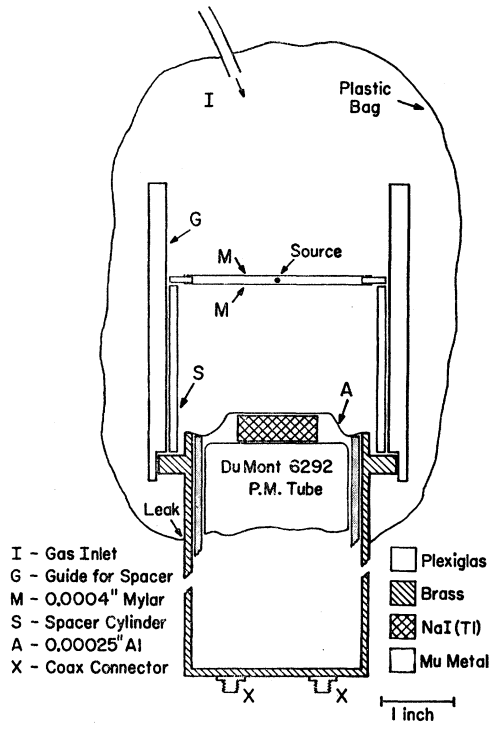

FIG. 1. Scintillation counter arrangement for measuring the L/K x-ray intensity ratio from the W^{181} decay.

III. EXPERIMENTAL RESULTS

A. Coincidence Experiment

Although Debrunner *et al.*[4] reported finding no coincidences between the 476- and 482-kev transitions in the Hf^{181} decay, this coincidence experiment has been repeated because the result is crucial for the decay scheme arguments. The single-channel pulse-height analyzers of the fast-slow coincidence circuit were set for an energy of about 480 kev with a 9% channel width. Coincidences were counted for a total run time of 300 min. The observed coincidence rate was 0.077 ±0.016 count/min, while the calculated random coincidence rate was 0.101±0.006 count/min. The expected true coincidence rate, if the two transitions were in cascade, could be determined from the 476-kev transition intensity[3] and the measured source strength. This rate was 4.1 counts/min. It is, therefore, concluded that the two transitions are not in cascade.

B. Low-Energy Electron Spectra

To check the possibility that the 476-kev transition is in parallel with the 482-kev line, a search was made

[14] D. E. Rehfuss and B. Crasemann, Phys. Rev. **114**, 1609 (1959).
[15] R. L. Garwin, Rev. Sci. Instr. **21**, 569 (1950); **24**, 618 (1953).

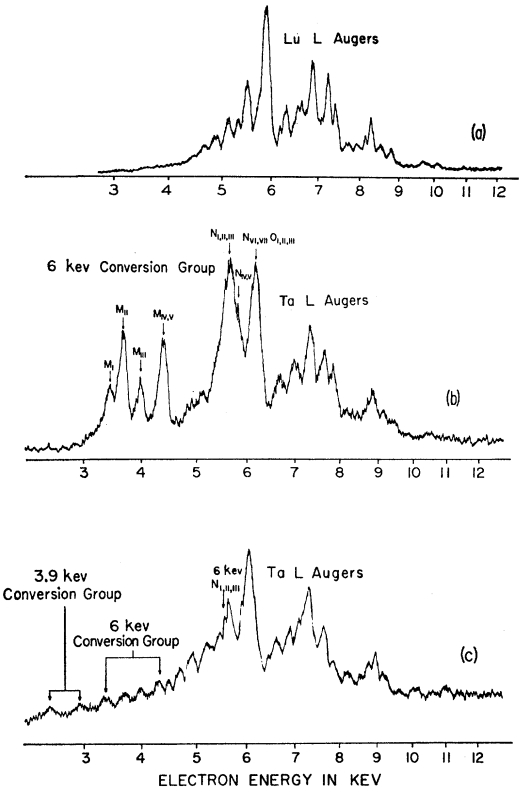

FIG. 2. 180° β-ray spectrometer curves from the decays of (a) Hf^{175}, (b) W^{181}, and (c) Hf^{181}. The predominant peaks between about 5–9 kev are L Auger electron groups. Low-energy conversion electron groups are present in (b) and (c). Curve (b) in particular exhibits pronounced M-shell conversion lines of a 6.25-kev transition. The energy region below 5 kev in curve (c) is a pictorial representation of the average of data from a large number of spectrometer runs. The line intensities derived from this region should be considered as upper limits.

for a difference transition of 6 kev in the Hf^{181} decay. The electron spectrum of an Hf^{181} source was carefully studied with the 180° β spectrometer. The spectrum below 12 kev is shown in Fig. 2(c). The prominent peaks are the Ta L Auger electrons. Conversion electrons corresponding to the previously reported[3] 3.9-kev transition appear at the left of the figure. The region in the figure marked "6-kev Conversion Group" is where the M-shell conversion lines of a 6-kev γ ray would be expected. Figure 2(a)[16] shows the low-energy electron spectrum of Lu^{175} excited by the electron capture decay of Hf^{175}. The prominent lines are the Lu L Auger electrons. No low-energy conversion lines have been found[16,8] in the Hf^{175} decay; hence, by comparing Figs. 2(a) and (c), it can be seen that the "bumps" at about 4 kev in the latter appear to be conversion lines.

A W^{181} source was also studied to see if the 6-kev transition might be excited in this decay as well. The results are shown in Fig. 2(b). Near 4 kev four very strong electron lines appear, which by comparison with (a) must be conversion lines. The four lines close to each other suggest an M-shell conversion group.[17] The energies of these lines, obtained by averaging the results of several runs, are 3.5, 3.8, 4.1, and 4.4 kev. These energies can be combined with M-shell binding energies[18] of Ta to yield values for the transition energy. A weighted average of the results gives $E_{\text{transition}}=6.25 \pm 0.3$ kev. The stated error takes into account the statistical error in the measurement and the calibration error of the instrument.

TABLE I. M-subshell conversion ratios for a 6.25-kev transition in Ta.

Multipolarity	Subshell conversion ratio $M_{\text{I}}/M_{\text{III}}$	$M_{\text{II}}/M_{\text{III}}$	$M_{\text{IV,V}}/M_{\text{III}}$
$E1$ (theoretical)	0.57	1.20	3.9
$M1$ (theoretical)	66.4	6.9	0.53
Observed	0.85	1.79	1.76

It has been shown[10] that the 6-kev transition in the tungsten decay cannot be due to the presence of any other tungsten isotopes (or daughters) which might be expected in the source material, and must, therefore, belong to Ta^{181}. (Subsequent proportional counter observation of 6-kev γ rays in accelerator-produced W^{181} confirms this conclusion.)

From curves such as those shown in Fig. 2, the 6-kev conversion intensity relative to the Auger intensity is obtained. Combining the results from several runs, the relative (M-, N-, and O-shell) internal conversion intensity in the W^{181} decay is[19] $I_{6\ MNO}/I_{L\ \text{Auger}} \cong 0.51 \pm 0.05$. By comparing curve (c) to curve (a), an estimate of the 6-kev M-shell conversion intensity in the Hf^{181} decay is obtained. The result, $I_{6\ M}/I_{L\ \text{Auger}} \leqq 0.07$, is given as an upper limit because of possible

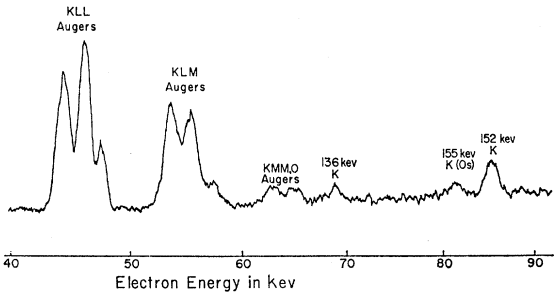

FIG. 3. β-spectrometer curve from a W^{181} source. The lines due to K Auger transitions and K conversion of the 136- and 152-kev γ rays are shown. A K conversion line of the 155-kev transition in Os^{188} appears as the result of a source impurity.

[16] E. N. Hatch, Ph.D. thesis, California Institute of Technology, Pasadena, California, 1956 (unpublished).

[17] The M_{IV} and M_{V} lines would not be resolved with this instrument.

[18] R. D. Hill, E. L. Church, and J. W. Mihelich, Rev. Sci. Instr. **23**, 523 (1952).

[19] Because of the uncertainty about a correction for the counter window attenuation, this result should be considered as a lower limit. Arguments can be made, however, which indicate that this attenuation ought to be negligible.

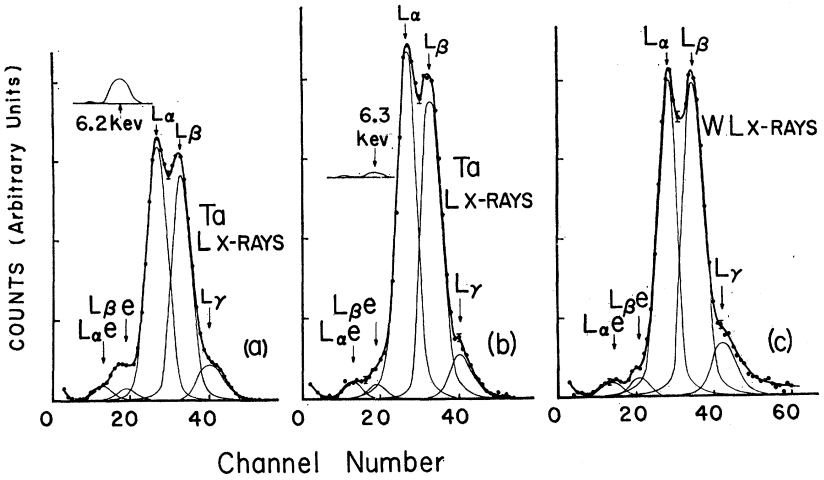

FIG. 4. Typical L x-ray spectra taken with the argon proportional counter using (a) W^{181}, (b) Hf^{181}, and (c) Ta^{182} as sources. The experimental data have been decomposed to exhibit a 6-kev transition in the decays of W^{181} and Hf^{181}. Gaussian shaped decompositions are drawn in fine line. The Ta^{182} spectrum, which has no 6-kev γ ray, is shown for comparison. The L x rays are labeled L_α, L_β, and L_γ, and the main x-ray escape peaks are labeled $L_\alpha e$ and $L_\beta e$.

contribution from the 3.9-kev transition and because of poor counting statistics.

At this point it may be noted that a calculation[10] has shown that the observed 6-kev intensity in the Hf^{181} decay is in agreement with what would be expected if the 6-kev transition were in cascade with the 476-kev transition. This result, along with the coincidence experiment, implies that a 476—6-kev cascade competes with the well-known 482-kev transition.

An indication of the multipolarity of the 6-kev transition is given by the M-subshell conversion line ratios of Fig. 2(b). Table I presents $E1$ and $M1$ theoretical[20] M-subshell conversion ratios for a 6.25-kev transition in Ta along with the observed ratios for the W^{181} decay. The agreement between the observed ratios and the theoretical $E1$ ratios is reasonably good. The $M1$ ratios are definitely not in agreement with experiment, and for higher multipolarities there is pronounced disagreement. Ratios calculated for multipolarity mixtures also fail to yield agreement with experiment. That there is not closer agreement with the $E1$ ratios can be understood[21] to a large extent in terms of the so-called anomalous conversion coefficients. Asaro *et al.*[22] have shown that for $E1$ transitions in heavy-element odd-A isotopes, L_{III} experimental coefficients usually agree with the theoretical ones, while L_I and especially L_{II} experimental coefficients *may* be considerably larger than the theoretical ones. Similar considerations apply to M-shell conversion coefficients, with $M_{IV,V}$ experimental coefficients tending to be low relative to the theoretical ones. The discrepancies between the theoretical $E1$ and the observed 6-kev coefficients (Table I) are all in the directions predicted by Asaro's observations. These workers[22] have related the conversion coefficient *anomaly* to the retardation of the γ-ray lifetime over the theoretical single proton lifetime estimate.[23] On using Asaro's technique[24] and his M-shell data, it has been concluded[10] that the 6-kev γ ray should be retarded by a factor of $\sim 2\times 10^5$, assuming all the discrepancy between theoretical and experimental coefficient ratios in Table I is due to anomalous conversion. Recently, Hauser[25] has measured the 6-kev transition half-life to be $(6.8\pm 0.4)\times 10^{-6}$ sec. This corresponds[26] to a γ-ray lifetime retardation of 5.2×10^5, in good agreement with the above estimate.

C. Electron Spectra at Higher Energy

A detailed study of the 136- and 152-kev transitions reported[5,7] in the W^{181} decay has been made with the 180° β spectrometer. Figure 3 is a portion of a spectrometer curve and shows the K Auger groups, as well as the weak 136- and 152-kev K conversion lines. A

TABLE II. Conversion line and Auger line relative intensities for the W^{181} decay.

Intensity ratio	Measured value
$I_{K\text{ Auger}}/I_{L\text{ Auger}}$	0.056 ±0.006
I_{152K}/I_{136K}	1.83 ±0.2
$I_{152K}/I_{K\text{ Auger}}$	0.0305±0.004
I_{152K}/I_{155K}[a]	2.74 ±0.35
$I_{152K}/I_{152,155L}$[b]	2.62 ±0.38

[a] The 155-kev transition is in Os^{188} and is present because of a W^{188} source impurity.
[b] The Os 155-kev L conversion lines are superimposed on the Ta 152-kev L conversion lines.

[20] These coefficients are obtained by extrapolating the values given by M. E. Rose, *Internal Conversion Coefficients* (North-Holland Publishing Company, Amsterdam, 1958).
[21] Interpolation and extrapolation of Rose's tabulated coefficients may also contribute to the discrepancy.
[22] F. Asaro, F. S. Stephens, J. M. Hollander, and I. Perlman, Phys. Rev. **117**, 492 (1960); F. Asaro, F. S. Stephens, J. M. Hollander, and I. Perlman, University of California Radiation Lab. Rept. UCRL-8786, June, 1959 (unpublished).
[23] S. A. Moszkowski, *Beta- and Gamma-Ray Spectroscopy*, edited by Kai Siegbahn (North-Holland Publishing Company, Amsterdam, 1955), Chap. 13.
[24] A log log plot of the *anomaly*, the relative deviation from the theoretical coefficients, vs the lifetime retardation for a number of $E1$ transitions is roughly linear.
[25] U. Hauser, Nuclear Phys. (to be published).
[26] The 6-kev transition conversion coefficient, $\alpha_6=44$ (derived in Sec. III G), is used in making this estimate.

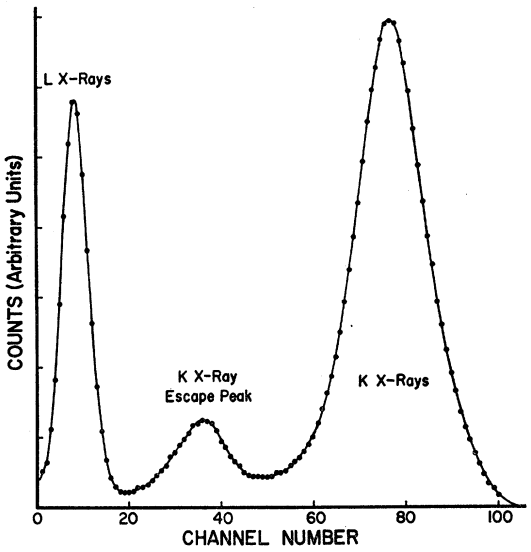

FIG. 5. Tantalum x-ray spectrum from the W^{181} decay observed with the thin window NaI scintillation crystal spectrometer.

weak K conversion line from the 155-kev transition in the Re^{188} decay also appears.[27] From this study, the exact energy of the 152-kev transition is found to be $E_{152}=152.5\pm0.3$ kev, in good agreement with Cork's[5] value, 152.5 kev. Table II summarizes various conversion line and Auger line intensities obtained with the 180° spectrometer. A W^{181} source was also studied with the ring-focusing β spectrometer. The K-conversion intensity ratio for the 152- and 136-kev transitions obtained with this instrument is $I_{152K}/I_{136K}=1.7\pm0.4$.

D. Proportional Counter Spectra

Proportional counters were used to search for the unconverted 6-kev γ ray. Figure 4 presents typical spectra taken with the argon proportional counter showing the decays of W^{181}, Hf^{181}, and Ta^{182}. The prominent peaks are the L x rays. To the left of the L x rays are two "bumps" that correspond to the escape peaks from the L_α and L_β x rays plus any low-energy (6 kev) γ-ray contribution. With the aid of measurements of the counter escape fraction and resolution as a function of energy, it has been possible to decompose the observed curves into Gaussian x-ray and escape peak components.[10] For each spectrum the difference between the sum of the decomposed curves and the observed curve gives the γ-ray contribution, which is shown in the inset. The W^{181} 6-kev γ-ray intensity was also determined by means of a differential absorber method and by means of a "normalized x-ray shape difference" method.[10] Weighted averages of all the relative intensity results, corrected for counter efficiency, are given in Table III.

[27] The source material had a W^{186} impurity, which through double neutron capture produced W^{188}. This isotope decays to Re^{188}, which in turn decays to Os^{188}. See reference 8.

Studies were also made using the xenon proportional counter, for which the L x-ray escape peak is no longer superimposed on the 6-kev line. Although the resolution of the xenon counter was not as good as that of the argon counter, the existence of the 6-kev γ ray could be confirmed.

E. Scintillation Spectrometer Measurements

If the W^{181} decay L/K x-ray intensity ratio is known, it is possible to determine the decay L/K capture ratio and thus the decay energy. Figure 5 shows a typical tantalum x-ray spectrum taken with the NaI counter of Fig. 1. Backscattering from the source mount was made negligible by having the source deposited on 0.4-mil Mylar. The apparent L/K x-ray ratio[28] depended on the source-crystal geometry, decreasing with increasing separation. This result was interpreted[10] as a scattering effect. The materials which surrounded the scintillation crystal had scattering coefficients[29] for the L x rays at least twice as great as those for the K x rays. In close geometry, scattering could increase the apparent relative L x-ray intensity. By measuring the L/K x-ray ratio in both air and helium as a function of the source-crystal separation, it was possible to extrapolate the data to a negligible scattering condition. The resultant x-ray intensity ratio is $I_{Lx}/I_{Kx}=0.264\pm0.013$.

Note added in proof. Quite recently a group at Livermore has measured this x-ray intensity ratio—see J. H. Zenger, C. D. Swift, H. Mark, and R. C. Jopson, Bull. Am. Phys. Soc. **6**, 72 (1961); R. C. Jopson, H. Mark, C. D. Swift, and J. H. Zenger, University of California Radiation Laboratory Rept. UCRL-6207, December, 1960 (Phys. Rev. to be published). Their value for the L/K x-ray intensity ratio is 0.312 ± 0.005. When allowance for detector efficiency is made, this corresponds to a ratio of about 0.30. Since this value is in disagreement with the result reported in the present paper, a remeasurement has been made. With a freshly polished NaI crystal ($2.3\times2.3\times0.55$ cm) and a source-crystal separation of 13.34 cm, a (corrected) ratio of 0.278 ±0.009 was obtained. The previous result for this geometry was 0.277 ± 0.014. Thus the extrapolated ratio is still 0.264. The discrepancy in the ratios of Zenger

TABLE III. Relative intensities from argon proportional counter measurements.

Intensity ratio	Value for W^{181} decay	Value for Hf^{181} decay
$I_{6\gamma}/I_{Lx}$[a]	0.046 ± 0.007	0.007 ± 0.003
$I_{L\beta}/I_{L\alpha}$	0.96	0.98
$I_{L\gamma}/I_{L\alpha}$	0.19	0.16

[a] I_{Lx} is the total L x-ray intensity, $I_{L\alpha}+I_{L\beta}+I_{L\gamma}$.

[28] Corrected for absorption and coincidence losses.

[29] G. W. Grodstein, *X-Ray Attenuation Coefficients from 10 kev to 100 Mev*, National Bureau of Standards Circular No. 583 (U. S. Government Printing Office, Washington, D. C., 1957).

et al. and the present measurements is probably related to differences in scintillation crystal preparation and scattering conditions at the detector. For example, roughing the surface of the polished crystal with emery paper gave considerably poorer resolution and an x-ray ratio about 10% *lower*.

In studying the very weak[7] 136- and 152-kev γ rays in the W^{181} decay with the standard scintillation spectrometer, it was necessary to use graded filters to attenuate the strong K x rays accompanying this decay. Figure 6 shows a typical observed spectrum. In the inset the background has been subtracted, and the experimental data have been fitted by two Gaussian curves of appropriate width and energy location. If corrections are made for counter efficiency and attenuation[29] in the absorbers, these data yield a value for the γ-ray relative intensity. The combined intensity result from five different runs with various filter combinations is $I_{152\gamma}/I_{136\gamma}=1.80\pm0.3$. The filtered spectra along with one unfiltered curve have also been used to determine the combined 136-, 152-kev γ intensity relative to the K x rays. A weighted average of the results gives $I_{136,152\gamma}/I_{Kx}=(1.2\pm0.3)\times10^{-3}$. The estimated errors allow for systematic as well as for statistical uncertainties.

F. L/K Capture Ratio and Decay Energy

Previously discussed results lead to the conclusion that Ta^{181} has a level at 6.25 kev above the ground state.[30] If for the moment it is assumed that all of the W^{181} capture decay occurs to the Ta^{181} ground state,[31] then the L/K capture ratio P_L/P_K can be related[6,10] to the L/K x-ray intensity ratio I_{Lx}/I_{Kx} by the formula

$$P_L/P_K=(1/\omega_{LL})[(I_{Lx}/I_{Kx})\omega_K-\omega_{LK}]. \quad (1)$$

Here ω_K is the K-shell x-ray fluorescence yield, ω_{LL} is the average L-shell fluorescence yield for vacancies produced directly by L capture, and ω_{LK} is the average L-shell fluorescence yield for vacancies produced through K capture. The K-shell yield, ω_K(Ta), is taken from the tables of Wapstra *et al.*,[32] while the L-shell yields must be computed[33] from the available data.

[30] Debrunner's upper limit on the W^{181} decay energy indicates that the 6-kev transition does not occur between the 482-kev level and a level at 476 kev.

[31] The weak branching to the higher energy levels may be neglected, while capture to the 6-kev level may be included with the ground state capture, since the energy difference is small and the wave functions for the two transitions are the same.

[32] A. H. Wapstra, G. J. Nijgh, and R. Van Lieshout, *Nuclear Spectroscopy Tables* (North-Holland Publishing Company, Amsterdam, 1959).

[33] For a process P,

$$\omega_{LP}=\sum_{i=1}^{3} u_{Pi}\omega_{Li},$$

where u_{Pi} is the chance that process P produces a vacancy in the L_i subshell, and ω_{Li} is the L_i-subshell partial yield. Table VII of Appendix A summarizes various partial yield measurements, and gives average values for ω_{Li}. Table VIII of Appendix A lists the values of u_{Pi} used in calculating ω_{LL} and ω_{LK}.

TABLE IV. X-ray fluorescence yields for Ta as excited in the W^{181} decay.

Fluorescence yield	Value
ω_K	0.943
ω_{LK}	0.1885
ω_{LL}	0.169

Table IV presents these yields. The L/K capture ratio obtained from Eq. (1) is $P_L/P_K=0.358\pm0.070$. The stated error only takes into account the uncertainty in the x-ray intensity measurement. P_L/P_K is *very* sensitive to the L-shell fluorescence yield values. Because of uncertainties associated with these yields, it is not practical to state a realistic error for the capture ratio. The value above is the best estimate that can be made on the basis of the presently available fluorescence yields.

Note added in proof. The Livermore group has also determined $P_L/P_K(=0.23\pm0.05)$ using a slightly different approach than the one given above. See note added in Sec. III E.

The orbital capture decay formulation of Brysk and Rose[34] may be used to compute the capture decay energy. Using their notation and assuming that the transitions are either allowed or first-forbidden, nonunique, the capture ratio is given by

$$\frac{P_L}{P_K}=\frac{q_{LI}^2}{q_K^2}\left(\frac{g_{LI}^2}{g_K^2}\right)\left(1+\frac{f_{LII}^2}{g_{LI}^2}\right). \quad (2)$$

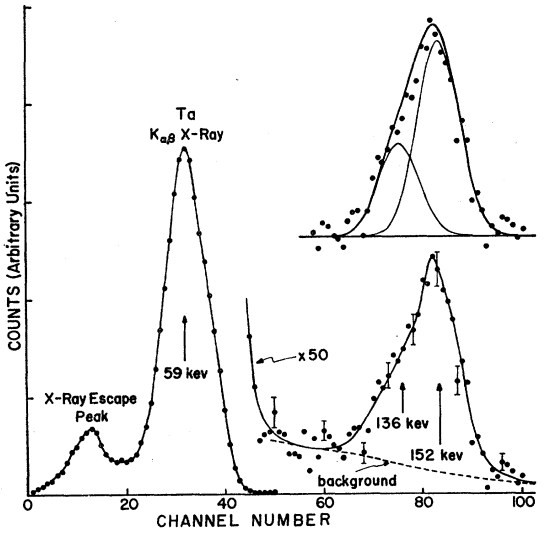

FIG. 6. NaI scintillation spectrum from the W^{181} source showing the combined peak due to the 136- and 152-kev γ-ray transitions. Filters of 0.018-in. Sn, 0.003-in. Mo, 0.005-in. Cu, and 0.003-in. Al have been used to attenuate the intense K x rays. The inset shows the experimental points (with background and scattering corrections) fitted with Gaussian curves.

[34] H. Brysk and M. E. Rose, Revs. Modern Phys. **30**, 1169 (1958).

The g's and the f's are radial wave functions evaluated at the nuclear surface, and the q's are the neutrino energies, $q_K=Q_{EC}-B_K$, etc. Q_{EC} is the decay energy and B_K is the K-shell binding energy. Using wave functions from reference 34 and binding energies from reference 32, Eq. (2) gives the value $Q_{EC}=176_{-22}^{+44}$ kev. The error limits take into account only the uncertainty in the x-ray intensity measurements.

This decay energy value is not in agreement with the value 92 kev (±10%) determined by Bisi *et al.*,[6] who used a similar method. Their L/K x-ray intensity ratio of 0.39±0.01 disagrees with the present result (0.264). The value $Q_{EC}=176$ kev is, however, in good accord with the decay energy arguments of Debrunner *et al.*,[7,35] who from x-ray–γ-ray coincidence experiments concluded that 160 kev$<Q_{EC}<$205 kev. Furthermore, the present decay energy value is in agreement with the $W^{181}-Ta^{181}$ mass difference, $Q=186$ kev, determined from the recent atomic mass tables of the Minnesota group.[36]

If, contrary to the previous assumptions, it is assumed that all of the capture occurs through a first-forbidden, unique transition ($\Delta I=2$, yes), then the above procedure would give a decay energy of about 422 kev, in contradiction with experimental evidence. This result and similar results for the higher orders of forbiddenness indicate the validity of the original assumption of allowed or first-forbidden, nonunique transitions.

G. 6-kev Conversion Coefficient

In evaluating the 6-kev conversion coefficient the weak branching to the 136 and 152 kev transitions may again be neglected. Assuming allowed or first-forbidden, nonunique, transitions, the L/K capture ratio is the same for both the 6-kev level and ground state branches.[34,37] The total $(M+N+O)$ shell internal conversion coefficient for the 6-kev transition may now be expressed in terms of the fluorescence and Auger[38] yields, and either (1) the 6-kev conversion and γ-ray intensities relative to the L Augers and L x rays, respectively, or (2) the 6-kev conversion and γ-ray intensities relative to the K Augers and K x rays, respectively. The results are (1) $\alpha_T=41.9$, and (2) $\alpha_T=45.0$ The fairly close agreement indicates an overall consistency. The final value adopted is $\alpha_T=44\pm7$. A multipolarity assignment of $E1$ is indicated by this result (see Table X of Appendix B).

H. W^{181} Decay Branchings

With the assumptions of Sec. III F, the L/K capture ratio and the ratio of $M+N+\cdots$ capture to L capture[32] are used to obtain the fractional K capture, $N_K/N=0.689$, and the fractional L capture, $N_L/N=0.247$. As above, two expressions may now be derived for N_0/N_6, the ratio of ground state to 6-kev state total capture. The value based on L-process intensities is $N_0/N_6=1.96$, while the K-process value is $N_0/N_6=1.82$. The average, $N_0/N_6=1.89$, is used to calculate the percentage branchings to the lower capture transitions, $N_6/N=35\pm10\%$ and $N_0/N=65\pm10\%$.

In the Discussion it is concluded that the 152-kev transition occurs between a level at 158 kev and the 6-kev level. To determine the branchings to the 136- and 158-kev levels, the following conversion coefficients[39] are used: $\alpha_K(136)=1.40$, $\alpha_T(136)=1.80$, $\alpha_K(152)=1.30$, and $\alpha_T(152)=1.58$. The ratio of capture branching between the two levels may be computed from the γ-ray intensity ratio and from the K conversion intensity ratio. The results are $(N_{158}/N_{136})_\gamma=1.66$ and $(N_{158}/N_{136})_K=1.82$. The average value, $N_{158}/N_{136}=1.7\pm0.2$, is used in calculating the percentage branchings.

The ratio of total upper level (158,136) to total lower level (6,0) capture, N_u/N_l, may be obtained from the γ-ray data and from the conversion line data. The results, $(N_u/N_l)_\gamma=2.04\times10^{-3}$ and $(N_u/N_l)_K=1.47\times10^{-3}$, give an average value, $N_u/N_l=1.8\times10^{-3}$, which is used to calculate the percentage branching to the two lower levels, $N_{6,0}/N_T=99.8\%$.

Table V summarizes all of the branching results in terms of percentages of the total W^{181} decay. A comparison of these results with those of Debrunner *et al.*,[7] shows that the present ratio N_{158}/N_{136} is in very close agreement with their value. The percentage values, N_{158} and N_{136}, agree with Debrunner's values within the stated error, but the agreement is not as close as in the case of the ratios.

I. Log *ft* Values

The often used nomographs of Moszkowski[40] for calculating *ft* values are not suitable for the present case because L capture is not taken into account and because the electron binding energies are neglected. Hoff and Rasmussen[41] give formulas that express f (for allowed transitions) in terms of the electron wave functions[34] and the capture neutrino energies. The neutrino energies are computed from the present decay energy value, 176 kev. The W^{181} half-life is known[8] to be 145 days. Partial half-lives can then be calculated using the branchings[42] of Table V.

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[35] P. Debrunner, E. Heer, W. Kündig, R. Rüetschi, and T. Lindquist, Helv. Phys. Acta **29**, 432 (1956).

[36] V. H. Bhanot, W. H. Johnson, and A. O. Nier, Phys. Rev. **120**, 235 (1960).

[37] The energy difference between ground state and 6-kev level capture is neglected.

[38] The Auger yields may be derived from data in Tables IV, VII, and VIII.

[39] See the discussion of conversion coefficients in Appendix B.

[40] S. A. Moszkowski, Phys. Rev. **82**, 35 (1951).

[41] R. W. Hoff and J. O. Rasmussen, Phys. Rev. **101**, 280 (1956).

[42] N_{158} and N_{136} are corrected for $(M+N+\cdots)$ shell capture. See Chap. 5.4 of reference 32.

TABLE V. Percentage branchings in the W^{181} decay.

Branch[a]	Value %	Estimated error % (±)
$N_{158,136}$	0.18	0.05
$N_{6,0}$	99.8	0.1
N_{158}	0.11	0.03
N_{136}	0.067	0.018
N_6	34.9	10
N_0	64.9	10

[a] The branch is designated by the energy of the level to which the transition occurs.

Table VI gives the results of the ft calculations. The errors for 0 and 6 kev take into account the uncertainty in both the decay energy and branching, while the remaining errors allow for the decay energy uncertainty only. The agreement between the independent evaluations of the logft values for K- and L-capture to the 6 kev and ground states is good.

IV. DISCUSSION

Although the present experiments alone allow several choices of Ta^{181} level arrangements and spins, a unique level scheme can be determined with the aid of data from previous experiments combined with nuclear model considerations.

The 6-kev transition has $E1$ multipolarity (with possible small $M2$ admixture). The Ta^{181} ground state is well known[2] to be the 7/2+ [404] state. The 6-kev level must have spin 5/2, 7/2, or 9/2 and negative parity. Observed decay branchings from the 9/2+ [624] W^{181} ground state[43] rule out spin 5/2. The 476-kev transition occurs between the 5/2+ [402] level at 482 kev and the 6-kev level. The conversion and branching data[3] for this transition and the 1.1×10^{-8} sec lifetime[8] for the 482-kev level require the 476-kev transition to be $M2$ with possible $E3$ admixture. This multipolarity choice then rules out spin 7/2−, and thus the 6-kev level must be assigned 9/2−. With this choice of spin and parity, the $M2$ transition would be classified as unhindered—in agreement with the observed lifetime. On the basis of Nilsson's[2] calculations, a 9/2− [514] intrinsic level is to be expected in Ta^{181}.

The 152-kev transition either occurs between a 152-kev level and the ground state or between a 158-kev level and the 6-kev level. If the latter case were true, the 158-kev level might be a first rotational (11/2−) level based on the 9/2− intrinsic level. The moment of inertia for such a rotational band has been estimated,[44] and a rotational level is expected in the neighborhood of 164 kev. The discussion of conversion coefficients in Appendix B indicates that the 152-kev transition should have multipolarity $M1$ $(+E2)$. Studies[3,9] of the Hf^{181} decay combined with nuclear model[1,2] and asymptotic selection rule[45] considerations give strong support[10] to the choice of a spin 11/2− level at 158 kev.

TABLE VI. Logft values for the W^{181} decay K- and L-capture transitions.

Transition to level at	$\log(ft)_K$	$\log(ft)_L$
0 kev	$6.70_{-0.2}{}^{+0.3}$	$6.70_{-0.1}{}^{+0.2}$
6 kev	$6.92_{-0.2}{}^{+0.4}$	$6.93_{-0.1}{}^{+0.3}$
136 kev	···	7.59±1
158 kev	···	6.04±2

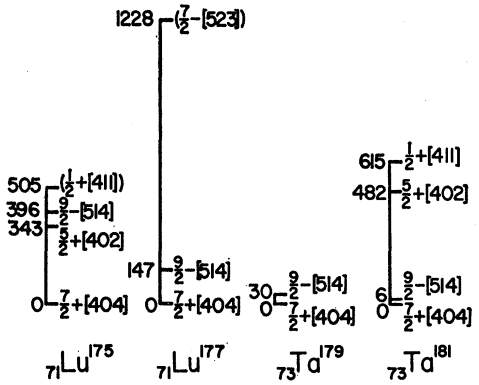

FIG. 7. Intrinsic levels of some odd-A, odd-proton nuclei near $A=181$. Each level is denoted by its energy, spin, parity, and asymptotic quantum numbers, $[Nn_z\Lambda]$. See B. R. Mottelson and S. G. Nilsson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Skr. **1**, 8 (1959). The trend of the 9/2− [514] level is of particular interest.

In agreement with the logft values of Table VI, the transitions to the $K=7/2+$ rotational band are classified[45] allowed, hindered, while the transitions to the 9/2− band are first-forbidden, unhindered. These classifications are consistent with the assumptions made in the previous section (Sec. III).

Some interesting exceptions to the orderly filling of the Nilsson levels have already been noted.[2] The tantalum odd-A isotopes are expected to have 5/2+ [402] ground states, but 7/2+ [404] are observed. Rhenium odd-A isotopes are expected to have 9/2− [514] ground states, but 5/2+ [402] are observed. The curious result is that the 9/2− level does not seem to occur as a ground state, although in Ta^{179} and Ta^{181} it is "almost" the ground state. Figure 7 is a schematic representation of the Nilsson intrinsic states of some odd-A, odd-proton nuclei near Ta^{181}. The diagrams are arranged in order of increasing A. The 9/2− [514] level starts at a relatively high excitation energy, 396 kev, in Lu^{175} and moves down toward the ground state with increasing A, until in Ta^{181} it is just 6 kev

[43] B. Harmatz, T. H. Handley, and J. W. Mihelich, Phys. Rev. **119**, 1345 (1960).

[44] The average ratio of the moment of inertia for the $K=9/2-$ rotational band to that of the ground state band for Lu^{175} and Re^{183} has been used in estimating the Ta^{181} 9/2− band moment from the ground state band moment.

[45] G. Alaga, Phys. Rev. **100**, 432 (1955); Nuclear Phys. **4**, 625 (1957).

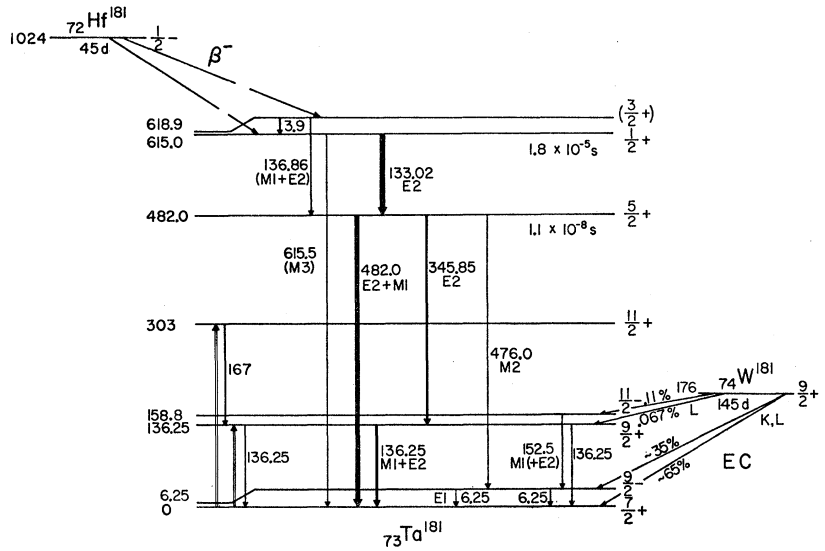

FIG. 8. Complete Ta^{181} level scheme showing the 9/2− 6.25-kev level. The 152-kev transition is placed between the new 11/2− 158.8-kev rotational level and the 6-kev level.

above the ground state. In view of this trend, information about the intrinsic levels in Ta^{183} and Ta^{185} might be most interesting.

The results of this work are summarized in a level scheme for Ta^{181} presented in Fig. 8. All experimental findings are consistent with the proposed decay scheme and with the unified model.[1,2]

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V. APPENDIX

A. Ta *L*-Shell Fluorescence Yields

In calculating the *L*-shell fluorescence yields of Table IV it is necessary to know the partial yields for the three *L* subshells. Unfortunately these yields are rather poorly known. The problem of experimentally measuring ω_{LI} and ω_{LII} is particularly difficult. The results of several measurements of the Ta *L* yields are given in Table VII, along with the average values which are used for the calculations. Errors in these yields may be as large as 10%. A more complete discussion of this subject is to be found in the book by Burhop.[46]

Table VIII lists the probabilities, u_{Pi}, that also enter into the fluorescence yield calculations. The $u_{Ki}(Z=73)$ are determined from the graphs of Fig. 7.52 in reference 32. Ratios of the $u_{Li}(Z=74)$ are obtained from graphs of electron radial wave functions evaluated at the nuclear surface given in reference 34. The u_{Li} are normalized by the condition $\sum u_{Li}=1$.

[46] E. H. S. Burhop, *The Auger Effect* (Cambridge University Press, New York, 1952).

TABLE VII. *L*-shell partial fluorescence yields for tantalum.

Source	ω_{LI}	ω_{LII}	ω_{LIII}
Laskar[a]	0.153	0.283	0.191
Kinsey[b]	0.17	0.31	(0.18)
Küstner and Arends[c]	(0.284)	(0.326)	0.191
Roos[d]	...	0.23	0.23
Average[e]	0.162	0.274	0.204

[a] W. Laskar, Ann. phys. **3**, 258 (1958). Laskar's values for ω_{Li} have been plotted and extrapolated to $Z=73$.
[b] B. B. Kinsey, Can. J. Research **26a**, 404 (1948).
[c] H. Küstner and E. Arends, Ann. Physik **22**, 443 (1935).
[d] This unpublished measurement by C. E. Roos has been cited by B. L. Robinson and R. W. Fink, Revs. Modern Phys. **32**, 117 (1960). These authors also give a value for $\omega_{LI}(=0.28)$ from Roos, but in a private communication Roos has stated that this value should have been labeled ω_{LT}, the total yield for all the *L* subshells together.
[e] The values in parentheses have not been included in the average. Kinsey's value for ω_{LIII} is rejected because, as he and other workers have noted, it is probably 10% too low. The ω_{LI} and ω_{LII} values of Küstner and Arends are rejected on the basis of arguments given in Chap. 7.5 of reference 32.

B. Internal Conversion Coefficients

The internal conversion coefficients of the 136- and 152-kev transitions were used in several calculations and the 6-kev theoretical coefficients were used for the multipolarity assignment. A brief discussion of these coefficient values will now be given.

136-kev Transition

This transition has been previously studied extensively and information relating to its conversion coefficients has been determined by several different means.

TABLE VIII. Probabilities for vacancy production in the tantalum *L* subshells.

Means of excitation	Vacancy probability for L_I	L_{II}	L_{III}
K hole filling (u_{Ki})	0.030	0.288	0.513
L capture process (u_{Li})	0.943	0.057	0

Experimental results on conversion coefficients, conversion ratios, and angular correlations (for Hf^{181} β decay and Ta^{181} Coulomb excitation) have been expressed[10] in terms of the percentage $E2$ in the 136-kev $M1+E2$ multipolarity mixture. The average percentage value, $16.1\pm10\%$,[47] and the theoretical[48,49] conversion coefficients give the following values for the K-shell and total $(K+L+M)$-shell coefficients: $\alpha_K(136)=1.40$ and $\alpha_T(136)=1.80$.

152-kev Transition

For this transition Debrunner *et al.*,[7] obtained $\alpha_K(152)=1.0\pm0.2$, but this result was based on $\alpha_K(136)=1.2$.[3] Their value, corrected for the new $\alpha_K(136)$ above, gives $\alpha_K(152)=1.17$. A value $\alpha_K(152)=1.42\pm0.2$ can be derived from $\alpha_K(136)$ and the K-conversion and γ-ray intensity ratios found in the present study. The value adopted for the calculations is the average, $\alpha_K(152)=1.30\pm0.15$.

This result allows the 152-kev transition to be assigned $M1$ $(+E2)$ or $E1+M2$. Table IX presents calculated conversion coefficients[48,49] for three possible multipolarity mixtures. Debrunner *et al.*,[7] preferred case 3, although their choice depended on nuclear model considerations. The K/L conversion ratio of Cork *et al.*,[5] $\alpha_K/\alpha_L=\sim8\pm2$, favors cases 1 or 2. K/L evidence[50] from the present study also suggests this. The 152-kev transition is therefore assigned as $M1$ $(+E2)$. This choice is completely in agreement with the decay systematics in the Discussion, whereas the other choice is not. Table IX is used to obtain $\alpha_T(152)=1.58$ from the experimental $\alpha_K(152)$.

TABLE IX. Conversion coefficients and possible multipolarity assignments of the 152-kev transition.

Case	Assumed α_K	Multipolarity mixture	Calculated α_L	α_T	α_K/α_L
1	1.17[a]	pure $M1$	0.166	1.41	7.0
2	1.15[b]	98% $M1$+ 2% $E2$	0.170	1.40	6.8
3	1.3	81% $E1$ +19% $M2$	0.342	1.77	3.8

[a] Theoretical coefficient for a pure $M1$ transition in Ta.
[b] Lower experimental limit for $\alpha_K(152)$ [1.30 −0.15].

TABLE X. M-shell conversion coefficients for $Z=73$.

Energy, k (m_0c^2)	$\alpha(E1)$	$\alpha(E2)$	$\alpha(M1)$	$\alpha(M2)$
0.20	0.0187	0.888	0.230	3.81
0.15	0.0403	3.34	0.528	13.0
0.10	0.126	24.3	1.72	78.5
0.05	0.878	732	13.2	2210
0.0122[a]	47.5	7.7×10^5	0.85×10^3	2.2×10^6
0.0122[b]	81	1.3×10^6	1.5×10^3	4.0×10^6

[a] 6.25 kev.
[b] The values above have been corrected to allow for contributions from the N and O shells. These values are thus α_T, the total conversion coefficient.

6-kev Transition

Theoretical ($E1$, $E2$, $M1$, and $M2$) M-shell conversion coefficients for tantalum obtained[51] from the tables of Rose[48] are presented in Table X. The values for $k=0.0122$ (6.25 kev) in the last row have been corrected[10] to allow for the contributions from the N and O shells, and thus represent total coefficients. All these coefficients should be used with caution for several reasons. The extreme extrapolation down to 6 kev may introduce errors of unknown magnitude and direction. Furthermore, no screening corrections were made in the calculations, and Rose[52] has estimated that the theoretical M-shell coefficients may be as much as a factor of two too large because of this.

[47] This percentage $E2$ value is in good agreement with an average value, $16\pm1.5\%$, published recently by E. M. Bernstein and R. Graetzer, Phys. Rev. **119**, 1321 (1960).
[48] M. E. Rose, *Internal Conversion Coefficients* (North-Holland Publishing Company, Amsterdam, 1958).
[49] L. A. Sliv and I. M. Band, Leningrad Physico-Technical Institute Report, 1956 [translation: Report 57 ICCK1, issued by Physics Department, University of Illinois, Urbana, Illinois (unpublished)].
[50] Overlapping of the Os^{188} 155-kev L-conversion lines with the 152-kev L lines made exact analysis difficult. The data in Table II suggest, however, that α_K/α_L (152) is considerably greater than 4.
[51] M-subshell coefficients for each energy were summed and this total was plotted against Z on semi-log paper. The interpolated coefficients for $Z=73$ were plotted on log log paper to extrapolate to 6.25-kev energy.
[52] M. E. Rose (private communication), reported in M. E. Bunker, B. J. Dropesky, J. D. Knight, J. W. Starner, and B. Warren, Phys. Rev. **116**, 143 (1959).