New neutron-rich isotope: ^{196}Os†

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A new neutron-rich isotope, ^{196}Os, was identified as the product of the ^{198}Pt(n,2pn) reaction. Chemically separated sources were used to characterize its β and γ radiations. The new isotope decays with a half-life of 34.9 ± 0.2 min. Ten γ rays have been assigned to this decay, the strongest being at 407.9 keV. A Q_{β^-} of 0.84 MeV was inferred from β-γ coincidence measurements. γ-ray energies and intensities as well as Q_{β^-} for the decay of the 52-sec ^{196}Ir daughter of ^{196}Os have been remeasured and are in agreement with the results of an earlier investigation. A partial decay scheme for ^{196}Os is proposed.

[RADIOACTIVITY ^{196}Os [from ^{198}Pt(n,2pn), medium energy neutron beam]; measured $T_{1/2}$; E_γ, I_γ, E_β, $\beta\gamma$ coin; deduced logft limits. ^{196}Ir measured E_γ, I_γ, E_β; deduced levels, J, π. Enriched targets, radiochemistry, Ge(Li), Si(Li), and plastic detectors.]

I. INTRODUCTION

Considerable progress has been made recently in the characterization of new neutron-rich nuclei which lie outside the mass region accessible from the fission of heavy elements. Instrumental techniques such as on-line isotopic identification of neutron-rich products from spallation or exotic heavy-ion rearrangement reactions have been used as an effective means for the study[1] of nuclei below the fission product region. Above the fission product region, several new neutron-rich isotopes[2-4] have been produced via the (p,3p) reaction. Unfortunately, protons with energies of ≈100 MeV which are required to make sources via the (p,3p) reaction also produce undesirably high yields of neutron deficient isotopes in the desired product element via (p,3pxn) reactions. It has been demonstrated recently that the analogous neutron-induced reaction (n, 2pn) offers very considerable improvement for the production of neutron-rich nuclei, both below[5] and above the fission region.[3] The Medium Energy Intense Neutron (MEIN) Facility[6] at Brookhaven National Laboratory was designed to take advantage of this improved selectivity in neutron-induced reactions. The MEIN facility produces an intense, energetic beam of neutrons by stopping the 200-MeV proton beam of the linac injector of the BNL Alternating Gradient Synchrotron in a water-cooled copper beam stop. Yields of neutron-deficient isotopes in the product element relative to the desired neutron-rich isotopes can be suppressed by factors of 10 to 100 through the use of these energetic neutrons instead of using protons. Characterization of the decays of new neutron-rich nuclei is thus made possible in spite of their generally short half-lives. Decay scheme information and atomic mass measurements from these studies are useful when applied as checks of current theoretical models for the behavior of nuclei far from stability.

Review of theoretical mass systematics and of the gross theory of β decay[7] indicates that ^{196}Os is expected to have a half-life sufficiently long (>10 min) to permit its detection by radiochemical means. A preliminary report on the discovery of ^{196}Os has appeared.[8] This article summarizes characterization of this new neutron-rich isotope.

II. EXPERIMENTAL TECHNIQUES AND RESULTS

Sources of ^{196}Os were produced via the (n, 2pn) reaction by irradiation in the MEIN facility of isotopically enriched (96%) ^{198}Pt with a flux of ≈10^{11} n/cm^2 sec in the energy range of 25–160 MeV. Targets in amounts of 20–300 mg (as $^{198}PtCl_4$ to facilitate rapid dissolution for subsequent radiochemical processing) were irradiated for periods up to one hour. The osmium fraction was separated in the following manner: (1) $^{198}PtCl_4$ target was dissolved in a minimum amount of warm H_2O, with small additions of warm 6 N HCl to hasten

dissolution; (2) the target solution was transferred to a distillation flask containing 10 ml of 12 N H_2SO_4 and ≈10-mg Os carrier held at 70 °C; (3) the temperature of the distillation flask was raised to ≈100 °C and, under slight negative pressure produced by a water aspirator, 30% H_2O_2 was cautiously added dropwise to start the distillation of OsO_4; (4) the OsO_4 distillate, mixed with air, was first drawn through two traps containing 6 N HCl at 80 °C which served to trap any radiochlorine compounds present in the distillate; (5) the air-distillate mixture was then drawn through two traps of 6 N NaOH held in an ice bath to catch the OsO_4; (6) after ≈15 min of distillation the first NaOH trap was neutralized with 6 N HCl to ≈pH 5 and H_2S was bubbled into the solution to precipitate OsS_4, which was washed twice with H_2O and ethanol before being filtered for source preparation. This procedure gave high chemical yields and was used routinely to make sources without detectable contamination from neighboring elements and with source strengths of ^{196}Os of up to ≈0.3 μCi. The ^{198}Pt target was recovered for additional irradiations by taking the target solution to dryness, allowing it to cool, then treating it with small volumes of fuming HNO_3 and concentrated HCl and heating to dryness to drive off the H_2SO_4. This treatment with HNO_3 and HCl was repeated a few times. The final heating to dryness was done at moderate temperature to obtain a readily soluble form of $^{198}PtCl_4$ for the next irradiation.

γ ray spectra were obtained using a 40-cm^3 Ge(Li) detector which had full width at half maximum (FWHM) resolution of 950 eV at 122 keV and 1.85 keV at 1332 keV. X-ray and low energy γ-ray spectra were obtained with a 10-mm^2×3-mm deep Si(Li) detector, which had FWHM resolution of 155 eV at 5.895 keV. Energy and detector efficiency (photopeak and total) calibrations of the Ge(Li) and Si(Li) detectors were determined with standardized IAEA and NBS γ-ray sources. The γ-ray spectra were analyzed by the INTRAL [9] computer code, and the CLSQ program [10] was used for decay-curve resolutions. Plastic detectors of 6- and 25-mm thicknesses were used to accumulate low energy β-ray spectra in the β-γ coincidence mode and higher energy β-ray spectra in the β singles mode, respectively. Their resolution was typically ≈20%. Energy calibration of the plastic detectors was made by using the conversion electron lines from ^{137}Cs and ^{207}Bi, and the β end-points from pure β emitters such as ^{90}Sr, ^{204}Tl and ^{210}Pb. β-ray spectra were analyzed by the Fermi-Kurie plot method.

A representative γ-ray spectrum obtained from the chemically separated osmium fraction is shown in Fig. 1. The broad energy distribution of the neutron flux resulted in the production of a number of other radioactive osmium isotopes in addition to ^{196}Os. Except for the very long-lived and very short-lived Os isotopes (i.e., ^{194}Os and ^{195}Os), the γ-ray spectra contained contributions from each of the known Os radioactivities (and eventually their daughter radioactivities) in the A = 180 to 193 mass range. These well-characterized nuclides, however, did not seriously interfere due to their relative yields, half-lives, or the complexity of their γ-ray spectra.

The presence of ^{196}Os was first noted by the occurrence of the 355.7- and 779.6-keV transitions from the daughter decay [11] of 52-sec ^{196}Ir in secular equilibrium with its ^{196}Os parent. Spectra from the most intense sources that could be prepared from the available target material were observed to contain all of the reported transitions [11] in the ^{196}Ir decay. These ^{196}Ir γ rays as well as 10 others assigned to the ^{196}Os decay exhibit a weighted average half-life of 34.9 ± 0.4 min. Comparison of the intensities of the γ rays assigned to ^{196}Os in Fig. 1 and corresponding energy regions in the ^{196}Ir γ-ray spectrum of Ref. 11 indicates that the γ rays of ^{196}Os, although weak, are not attributable to unreported, low intensity transitions in the ^{196}Ir decay. The secular equilibrium relationship between the 35-min ^{196}Os and its ^{196}Ir daughter was used along with the reported [11,12] 17.3% absolute γ-ray abundance for the 355.7-keV transition to calculate the absolute abundances of ^{196}Os γ-ray transitions. Table I lists these energies and absolute intensities for the ^{196}Os decay. Table II lists remeasured energies and intensities for γ rays of the ^{196}Ir decay. Agreement with the earlier study of ^{196}Ir by Jansen, Pauw, and Toeset [11] is very good.

While the observation of ^{196}Ir in secular equilibrium provides the strongest evidence for correct assignment of the new radioactivity to ^{196}Os, corroborative evidence of this assignment was obtained from x-ray spectra. The composite x rays, Ir $K\alpha_1$, 64.90 keV and Pt $K\alpha_2$, 65.12 keV, are produced solely from the decay of neutron-rich osmium nuclei and their daughter Ir activities. These 65-keV x-rays exhibited a two component decay curve with a 35.9 ± 1.2-min component and a longer-lived component of ≈45 h due to combined contributions from ^{191}Os and ^{193}Os.

β-ray spectrum accumulated in the singles mode from the Os source were dominated by contributions from the intense (82%) high-energy (E_β^{max} = 3.19 MeV) β-ray transitions from the ^{196}Ir daughter. Other Os isotopes in the source either decay by β^- emission with much lower end-point energies or by electron capture or isomeric transition,

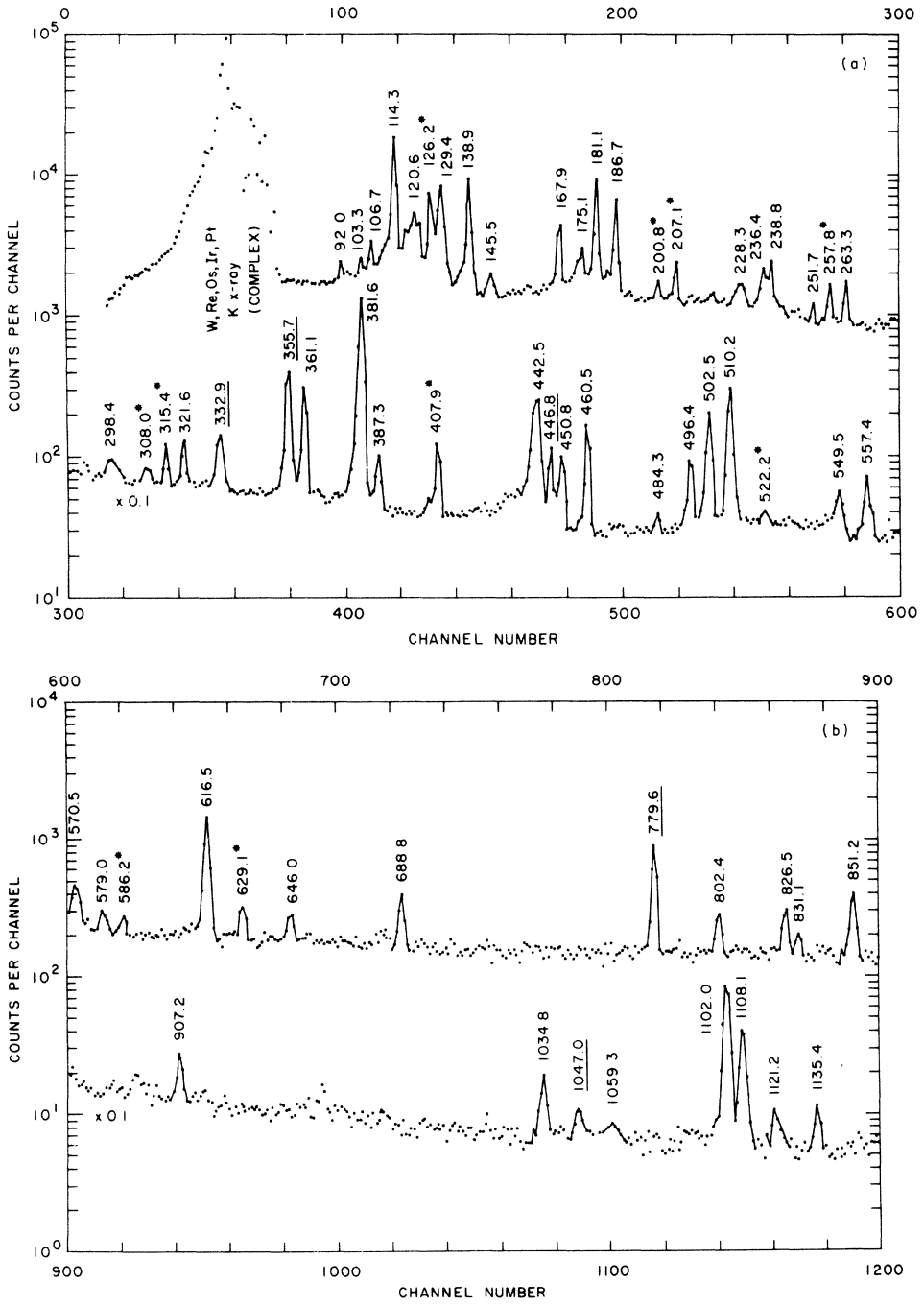

FIG. 1. γ-ray spectrum of the chemically separated osmium source containing ^{196}Os. Photopeak energies are in keV. Asterisks label transitions from the ^{196}Os decay. Underlined energies identify transitions from ^{196}Ir in secular equilibrium with its ^{196}Os parent.

neither of which contribute significantly to the β-ray spectrum. The decay curve that resulted from multiscaling the high-energy portion of the β spectrum ($E_\beta > 1.5$ MeV) exhibited a single component due to the ^{196}Os-^{196}Ir pair with essentially no background contributions. Least-squares fitting of this decay curve gave a half-life of 34.9 ± 0.2 min. Fermi-Kurie plot analysis of this β-ray spectrum yielded an end-point energy of 3.19 $\pm$ 0.10 MeV for the ^{196}Ir $\rightarrow$ ^{196}Pt ground state to ground state β^- transition, in excellent agreement with the earlier measurement of 3.20 MeV

TABLE I. γ-ray energies and absolute intensities in the decay of ^{196}Os.

E_γ	I_γ [a]
126.2 ± 0.2	5.3 ± 0.3
200.8 ± 0.3	0.56 ± 0.05
207.1 ± 0.2	2.4 ± 0.1
257.8 ± 0.2	2.3 ± 0.1
308.0 ± 0.4	0.43 ± 0.08
315.4 ± 0.2	2.5 ± 0.1
407.9 ± 0.2	5.9 ± 0.2
522.2 ± 0.3	0.78 ± 0.10
586.2 ± 0.2	0.59 ± 0.13
629.1 ± 0.4	1.6 ± 0.1

[a] Intensities are expressed as γ rays/100 β^-transitions.

TABLE II. γ-ray energies and absolute intensities in the decay of ^{196}Ir.

Present work		Jansen *et al.* [a]	
E_γ [b]	I_γ	E_γ	I_γ
332.9	3.7 ± 0.2	332.8 ± 0.3	4.0 ± 0.2
355.7	≡ 17.3 [c]	355.4 ± 0.3	≡ 17.3 [c]
446.8	5.5 ± 0.2	446.6 ± 0.3	4.2 ± 0.4
779.6	10.5 ± 0.4	779.4 ± 0.3	9.5 ± 0.3
1047.0	1.0 ± 0.1	1047.0 ± 0.7	0.9 ± 0.2
1228.6	0.33 ± 0.15	1228.6 ± 1.2	0.31 ± 0.07
1468.4	0.87 ± 0.24	1468.4 ± 0.8	0.76 ± 0.12
1564.2	0.70 ± 0.14	1564.2 ± 0.8	0.81 ± 0.17

[a] See Refs. 11 and 12.

[b] Energies are in keV, with uncertainties of ± 0.2 keV.

[c] Intensities are expressed as γ rays/100 β^- transitions (see text) and are computed from data of Refs. 11 and 12 with the assumption that the 355.4-keV transition is pure $E2$ ($\alpha = 0.061$).

of Jansen *et al.*[11]

A β-γ coincidence system with a Ge(Li) detector and a 6-mm thick plastic scintillator was used to record the β spectrum in coincidence with the 407.9-keV γ-ray transition (6% absolute abundance). β ray spectra were recorded both with the γ gate set on the 407.9-keV peak and with the γ gate set off the peak to assess the contributions of other nuclides to the measurement. A Fermi-Kurie plot of the β spectrum coincident with the 407.9-keV transition is shown in Fig. 2. Corrections for the Compton background based on the off-peak spectrum have been made. The end point of 435 ± 50 keV was determined by least-squares fitting of a line to the middle third of energy range and extension of this line to the abscissa. This procedure minimizes the effects of distortion due to multiple scattering at low energy and the occurrence of some counts beyond the true end point that result from the poor resolution (≈20%) of the plastic detector.

γ-γ coincidence measurements were not performed due to the inherently low abundance of the γ-ray transitions and the limitations in source intensity imposed by amount of available target material and neutron flux limitations. An attempt to observe conversion electrons from transitions in ^{196}Os decay superimposed on the intense β-ray spectrum of the ^{196}Ir daughter were not successful. Upper limits for the K-conversion coefficients for the transitions in the ^{196}Os decay are indicative of low ($\Delta l = 1, 2$) multipolarities.[13]

III. DISCUSSION

The following assumptions and experimental observations were used to construct a partial decay scheme for ^{196}Os: (1) β decay from the ^{196}Os ground state (0^+) will feed low spin, $I = 0$ or 1, states among the levels of ^{196}Ir via either allowed or first forbidden β^- transitions; (2) the low intensity of the ^{196}Os γ rays is indicative of an intense β-ray transition connecting the ^{196}Os ground state and the ^{196}Ir ground state,[11,12] $I^\pi = (0^-, 1^-)$, analogous to the intense β^- transition connecting the ^{196}Ir and ^{196}Pt ground states; (3) excited low-spin levels in ^{196}Ir, populated by the β decay of ^{196}Os, will decay by low multipole order transitions to other low-spin states in ^{196}Ir or directly to the ^{196}Ir ground state. Lacking γ-γ coincidence relationships, the level structure of ^{196}Ir was deduced from energy loops and differences which indicated the presence of cascade and crossover γ-ray transitions. γ-ray intensities were used to infer the ordering of levels. Intensity imbalances to and from a level were used to set limits for β-ray feeding, logft values, and conversion electron

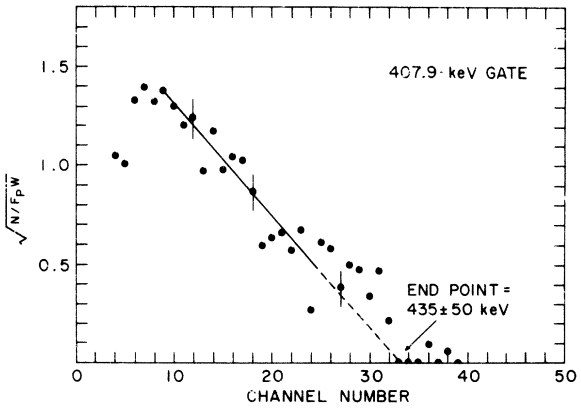

FIG. 2. Fermi-Kurie plot of the beta-ray spectrum coincident with the 407.9-keV γ-ray transition. The solid line is a least-squares fit to the middle third of the energy range. The dotted line extends this fitted line to the endpoint, for reasons discussed in the text.

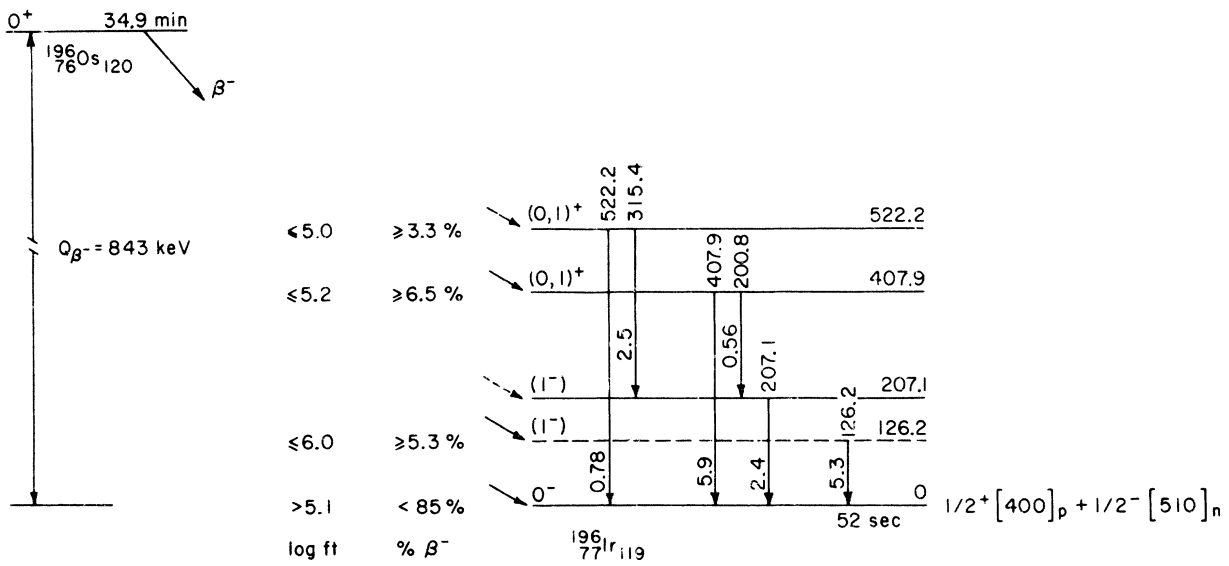

FIG. 3. Proposed decay scheme for ^{196}Os. All energies in keV. Numbers placed vertically along the transitions are absolute γ-ray intensities per 100 β^- decays.

coefficients and ultimately to deduce limiting transition multipolarities and spin-parity assignments for the levels.

A proposed partial decay scheme for ^{196}Os is shown in Fig. 3. The 200.8- and 207.1-keV transitions are placed in cascade, with the 407.9-keV γ ray assigned as the crossover transition. Similarly, the 522.2-keV transition represents the crossover of the 315.4- and 207.1-keV cascade γ rays. Transition intensity arguments require that the 200.8- and 315.4-keV transitions feed the 207.1-keV level. The transition at 126.2 keV is neither in any energy loops nor represented as an energy sum/difference. This transition is quite likely the strongest in the ^{196}Os decay; even with low multipole order, the 126.2-keV transition will be substantially converted.[13] This γ ray has therefore been placed tentatively as a ground state transition from a 126.2-keV level. It is somewhat surprising that the 257.8-keV transition does not feed or depopulate any of the other ^{196}Ir levels. This transition may originate from a low-spin level of sufficiently different character from the other states in ^{196}Ir to preclude the possibility of decay to or from these states, or more likely, the 257.8-keV γ ray is linked to the rest of the decay scheme by one or more unobserved (and probably low energy) transitions. The lines at 308.0 keV (0.43%), 586.2 keV (0.59%), and 629.1 keV (1.6%) were not placed.

The occurrence of β decays to the levels at 407.9 and 522.2 keV indicates that these states are of low spin (0 or 1). Observation of γ rays from these states to the ^{196}Ir ground state is further evidence of the low-spin character of the levels. If one or both of these states have nonzero spin, a γ ray of 114.3 keV could connect the two levels, although its abundance would, on energetic grounds, be low. No evidence for such a transition could be found in the decay curve analysis of the 114.3-keV line. This strong line decayed with the reported half-life of ^{183}Os and had the correct relative intensity in comparison with the other lines in that decay.[14] γ-rays to and from the 207.1-keV level indicate that this state is also of low spin. It is difficult to estimate the extent of β-decay feeding to this state since the 200.8- and 207.1-keV transitions will have substantial conversion coefficients even though they are both of low multipole order.

In the proposed decay scheme the logft limits for the various levels are based on the intensity imbalances through these levels. The 85% β branch to the ^{196}Ir ground state is the upper limit derived from transition abundances to the ground state as measured by γ-ray intensities *without* correction for internal conversion. Multipole order assignments for the ground state γ rays, particularly the low-energy ones, will affect the logft value for the ground-state decay and raises it closer to the corresponding β-ray transition connecting the ^{196}Ir and ^{196}Pt ground states (0^-, $1^- \rightarrow 0^+$, log$ft = 5.7$). Logft values for the 407.9- and 522.2-keV levels are characteristic of allowed, unhindered transitions, and on that basis these levels have been assigned positive parity. The 407.9- and 522.2-keV transitions are therefore most likely of $E1$ type.

TABLE III. Experimental and theoretical Q_{β^-} values for the ^{196}Os and ^{196}Ir decays.

		Theory [a]								
Isotope	Experiment (keV)	Myers	Groote Hilt Takahashi	Seeger Howard	Liran Zeldes	Bauer	Viola Swant Graber	Janecke Garvey Kelson	Comay Kelson	Janecke Eynon
^{196}Os	843 ± 50	400	470	1400	1080	•••	824	780	930	50
^{196}Ir	3190 ± 100	2660	2320	2000	3280	3050	3180	3140	3090	2660

[a] From Ref. 15, except for Viola, Swant, and Graber, Ref. 16.

The energy, 435 ± 50 keV, measured for the β ray in coincidence with 407.9-keV γ transition established a Q_{β^-} of 843 ± 50 keV for the ^{196}Os decay. This experimental value and the Q_{β^-} of 3190 keV for ^{196}Ir that was remeasured as part of the present work can be compared with several theoretical predictions, which are summarized in Table III. While the spread of the theoretical predictions[15,16] for the ^{196}Os Q_{β^-} is quite broad (50 to 1400 keV), several theories yield values close to the experimental value, with that of Viola, Swant, and Graber[16] being the closest. For the Q_{β^-} of ^{196}Ir, the relative spread is much narrower and values from the same group of theories again cluster about the experimental value.

IV. CONCLUSION

The present work, which was not highly sensitive to low energy and/or highly converted transitions cannot support a complete and definitive analysis of low-spin states in the ^{196}It level structure. It is possible, however, to gain some insight into the character of the ^{196}Ir level structure by the use of theoretical models.

^{196}Ir lies near the upper end of the deformed region. Its deformation and ground-state spin-parity can be understood from the character of single particle levels in the adjacent odd-A nuclei. The occurrence of ground state spin-parities of $\frac{3}{2}^+$ in the lighter odd Ir nuclei[17] point to a Nilsson assignment of $\frac{3}{2}^+[402]$ for the 77th proton in ^{196}Ir. High-spin isomers ($\frac{11}{2}^-$) in the same odd Ir nuclei indicate the proximity of the $\frac{11}{2}^-[505]$ orbital. First excited states of $\frac{1}{2}^+$ in ^{191}Ir and ^{193}Ir suggest the presence of the $\frac{1}{2}^+[400]$ orbital near the $\frac{3}{2}^+[402]$. Since the $\frac{1}{2}^+[400]$ and $\frac{3}{2}^+[402]$ orbitals parallel each other over a wide range of deformation,[18] the proton orbitals themselves cannot indicate the extent of deformation in ^{196}Ir. ^{197}Pt and ^{199}Hg, the odd mass isotones of ^{196}Ir, exhibit $\frac{1}{2}^-$ ground states and $\frac{13}{2}^+$ isomers. These states are attributable to Nilsson orbitals of $\frac{1}{2}^-[510]$ and $\frac{13}{2}^+[606]$, respectively, for the 119th neutron. These neutron assignments suggest a deformation of $\delta \approx 0.1$–0.15, a value consistent with the deformation inferred for other nuclei in this general mass region.[19] Coupling the $\frac{1}{2}^-[510]$ neutron orbital with the close lying proton orbitals of $\frac{1}{2}^+[400]$ or $\frac{3}{2}^+[402]$ yields a ground state spin-parity of 0^- or 1^- for ^{196}Ir. The absence of β rays from the ^{196}Ir ground state to the 2^+ first excited state in ^{196}Pt was used by Jansen *et al.*[11] to favor the 0^- assignment. The 1^- state formed from the alternative coupling of the odd nucleons in ^{196}Ir would then lie at low excitation and be fed in the decay of ^{196}Os. While neighboring odd-odd nuclei such as ^{194}Ir or ^{198}Au could, in principle, be used to estimate the strength of the n-p residual interaction in this mass region, the high level densities and complex decay patterns in these nuclei preclude an unambiguous estimate of this interaction strength. On strictly empirical grounds the 126.2- or 207.1-keV levels are the most likely candidates for the 1^- state. Combination of the high-spin Nilsson states yields a predicted spin-parity of 12^- for 1.4-h ^{196}Ir^m. While Jansen *et al.*[11] suggest a spin of 10 or 11, the 12^- assignment is also consistent with the β-decay pattern of this isomer.

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