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Walter M. Doyle and Richard Marrus

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ABSTRACT

The spins of several neutron-produced isotopes in the refractory region have been measured by the atomic-beam method. They are found to be Ta$^{183}$ ($I = 7/2$); W$^{185}$ ($I = 3/2$); W$^{187}$ ($I = 3/2$); Re$^{186}$ ($I = 1$); Re$^{188}$ ($I = 1$); Ir$^{192}$ ($I = 4$); and Ir$^{194}$ ($I = 1$). With the exception of Ir$^{192}$, these values are consistent with plausible state assignments based on the Nilsson model and with the coupling rules for odd-odd nuclei of Gallagher and Moszkowski.

A new method is described for the production of atomic beams of materials having high melting points and low vapor pressures.
INTRODUCTION

The neutron-enriched isotopes of the refractory elements \((71 \leq Z \leq 78)\) are especially relevant to nuclear theories, since they occur in a transition region between the extremes of highly deformed and spherical nuclear cores. Spin measurements in this region can serve as a crude test of deformations when compared with values obtained from the level orderings of Nilsson. In highly deformed odd-odd nuclei it has been shown that spins can be simply predicted from the quantum numbers of the orbitals of the last odd nucleons. Spin studies can therefore serve as a test of the validity of these coupling rules in the region of intermediate deformation.

In order to obtain spin measurements from a study of the Zeeman effect of atomic levels at low fields, we must know the electronic angular momentum \((J)\) and the spectroscopic splitting factor \((g_J)\) of the level. For the elements under investigation here, these quantities had been determined by optical spectroscopy. The numbers pertinent to our experiments are given in Table I. An interesting feature is the existence within thermal energies of levels arising from more than one configuration—one of which contains an unpaired s electron (Ir and W). This gives rise to the possibility of obtaining a hyperfine-anomaly measurement without having to measure the nuclear moment. We are currently investigating this possibility, and the spin measurements reported here constitute the first step in this program.
EXPERIMENTAL METHOD

The determination of spins by radioactive atomic beams is a subject that has been extensively reviewed, and the reader is referred to the literature for details. Our apparatus is of conventional flop-in design, although the problem of beam production is solved in a novel way. Ordinarily, atomic beams are produced by heating the desired material in an oven that acts as a suitable container at beam temperatures. The choice of suitable ovens for the refractories is severely limited by their high melting points. In the case of tungsten, for example, only one element—carbon—has a higher melting point, and only tungsten and carbon have higher melting points than tantalum. Finally, at high temperatures, there is a strong tendency for metals to interact with the common oven materials, and there are the additional problems, well known to atomic beamists, of creep and spritz. All these characteristic difficulties of the oven method led us to believe that it would be difficult, if not impossible, to find suitable oven materials for production of refractory beams.

All these problems can be neatly sidestepped, however. Our method is to use as a source 10-mil or 20-mil wire. The wire can be heated, either by passing a current through it or by electron bombardment. For reasons of beam stability, we prefer electron bombardment heating. The source mount is incorporated in an oven loader, as is shown in Fig. 1. The filament is placed above the source wire, so that the top of the wire is at the highest temperature. The requirement for this method to work is that the vapor pressure at or near the melting point be enough to sustain a sufficiently intense beam. Useful beams have been obtained with tantalum, tungsten, iridium, and rhenium. Only hafnium fails to meet the vapor-pressure requirement.

All of the isotopes used in these experiments were produced by neutron
irradiation at either the MTR in Arco, or the G. E. test reactor at Vallecitos. Most of the isotopes in this region have high capture cross sections, and the high specific activities important for beam production are obtainable. Bombardment time was chosen to enhance the production of the desired isotope in relation to undesired components that are simultaneously produced.

Our method of detection is to expose a platinum foil to the beam at a particular frequency and field setting and to count the deposited activity in a Geiger counter. Resonances are detected as an increase in counting rate. We have studied the collection efficiency for iridium on several materials: Au, Ag, Al, Ni, Ta, Sn, Cd, Cu, Mo, brass, and Q wax. To within a few percent the collection efficiency is the same for all. It seems very probable that the efficiency for collection is about 100% on these materials. Platinum was used in this work for reasons of convenience.

The determination of a spin (I) by the beam method is based on the determination of the transition frequencies between magnetic sublevels of a hyperfine state. In the Zeeman region, these frequencies are linearly related to the external magnetic field (H), and are given by

\[ \nu = \frac{g_J \hbar \nu}{h} \left[ F(F+1) + J(J+1) - I(I+1) \right], \]

where \( J \) is the electronic angular momentum of the level, \( g_J \) the spectroscopic splitting factor, and \( F \) the total angular momentum. A small term in the nuclear moment has been ignored.

RESULTS

Our investigations have resulted in spin assignments for seven nuclides in the refractory region. The results are listed in Table II along with the half-lives and the postulated Nilsson states of the last odd particles.
\( \text{W}^{185} \) and \( \text{W}^{187} \) are the only tungsten isotopes that can be produced in usable quantities. The large disparity in half-lives enabled us to obtain a preponderance of either isotope by varying the bombardment and decay periods. In each case a clear indication of the spin value, \( I = 3/2 \), was obtained as shown in the typical spin search, Fig. 1. The notation used in labeling transitions is "I(JF)," where "a" corresponds to \( F = F_{\text{max}} \), etc., as indicated in the energy-level diagram for \( I = 3/2 \) and \( J = 3 \), Fig. 2.

The samples used in the first rhenium runs consisted of about 80\% \( \text{Re}^{188} \) and 20\% \( \text{Re}^{186} \). Very intense resonances were observed for \( I = 1 \), and the decay curves indicated that the ratio of \( \text{Re}^{183} \) to \( \text{Re}^{186} \) was the same for the resonance exposures as for the total beam. Later, samples having a predominance of \( \text{Re}^{186} \) were used, and similar results were obtained (Fig. 3). The notation here is simply "I, F," since \( J = 5/2 \) for all points.

A typical iridium-194 search is shown in Fig. 4. The two observable electronic levels are characterized by the same angular momentum and are here distinguished by the notation "\( \text{exc} \)" for the excited configuration. Since the electronic \( g \) factors differ by only 3\%, the resonances are not resolved at this value of magnetic field. Sample decay curves are shown in Figs. 5 and 6. \( \text{Ir}^{192} \) has a very large cross section for thermal neutron capture (\( \sigma = 1000 \) barns), so that it comprised about 10\% of the radioactive sample. An \( I = 1 \) resonance exposure, however, exhibited only about 3\% \( \text{Ir}^{192} \), dictating the assignment of \( I = 1 \) to \( \text{Ir}^{194} \). Later runs made with a preponderance of \( \text{Ir}^{192} \) yielded indications for \( I = 4 \) only.

Our initial attempts to observe \( \text{Ta}^{183} \) resonances were focused on the \( 4F_{3/2} \) ground level of the configuration \( d^3 \). These proved unsuccessful, and we concluded that the ground-state electronic \( g \) factor (\( g_J = -0.447 \)) was too
small to allow sufficient deflection of the tantalum beam. By maintaining the operating temperature just below the melting point, we were able to sufficiently populate the two observable excited states, \(^{4}F_{5/2}\) \((g_J = -1.031)\) and \(^{4}F_{7/2}\) \((g_J = -1.218)\), to obtain reproducible results establishing the spin of Ta\(^{183}\) as \(I = 7/2\) (Fig. 7).

**DISCUSSION**

In this section we discuss plausible state assignments for the last odd particles on the basis of appropriate nuclear models. For the odd-odd nuclei we compare results with the predictions according to the coupling rules\(^2\) and review relevant experimental evidence.

**Tantalum-183:** The measured spin of 7/2 is consistent with the prediction based on \(\beta\) decay. The parity of the state inferred from \(\beta\) decay is positive, and the appropriate Nilsson state assignment\(^3\) is [404 7/2].

**Tungsten-185 and -187:** Dubey et al.\(^4\) have studied the \(\beta\) decay of these isotopes and have suggested the assignment of 3/2- and 1/2- for the spins and parities of W\(^{185}\) and W\(^{187}\), respectively. Their result for W\(^{187}\) disagrees with our directly determined value of 3/2. It seems reasonable to suggest that the ground level of the odd neutron is [512 3/2], with the deformation \(\delta \approx 0.22\) for W\(^{185}\), and the same ground level for W\(^{187}\), with \(\delta \approx 0.20\). These deformations are consistent with the deformations calculated by Mottelson and Nilsson.\(^5\)

**Rhenium-186 and -188:** The spins measured for these isotopes are consistent with results inferred from \(\beta\) decay. The \(\beta\)-decay measurements also show the parities of the ground levels to be negative. The ground levels of rhenium-185 and rhenium-187 are both known to be 5/2+, which is consistent with a state assignment of [402 5/2] for the odd proton. The tungsten measurements indicate that the odd neutron can be assigned to [512 3/2]. From the odd-odd
coupling rules a 1- ground state would be inferred, which is in agreement with the measurements.

Iridium-192 and 194: On the basis of β-decay studies, Johns and Nablo have suggested 4- as the ground state of Ir$^{192}$, and 1- or 2- for Ir$^{194}$. These are consistent with our results. The spins, electromagnetic moments, and parities of the ground levels of Ir$^{191}$ and Ir$^{193}$ indicate [402 3/2] for the 77th proton. The evidence for the assignment of the 115th neutron is ambiguous.

The ground levels of 76 Os$^{191}$ and 80 Hg$^{195}$ are 9/2- and 3/2-, respectively. Neither of these can be coupled with the proton spin in a way that is consistent with the spin and parity of Ir$^{192}$ and the odd-odd coupling rules. The Nilsson diagram shows that [510 1/2], [615 11/2], and [512 3/2] can all contain the 115th neutron, depending on the deformation, but no consistent fit to the spin and parity is possible with the assumed proton state.

The possibility of a fit based on the shell model has also been explored. The expected configurations of the protons and neutrons outside closed shells are $\pi(d_{3/2})$ and $\nu[(7i_{13/2})^8 (4p_{3/2})^1]$. Although these are consistent with the negative parity, any coupling yielding a spin of 4 would be an exceedingly complicated one.

The assignment for the 117th neutron in Ir$^{194}$ is more straightforward. The spins and parities of 78 Pt$^{195}$ and 80 Hg$^{197}$ are both 1/2-. The expected Nilsson state is [510 1/2]. When this combined with the assignment of the 77th proton, there is agreement with the spin, parity, and odd-odd coupling rules.
FOOTNOTES AND REFERENCES

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† Present address: Hughes Aircraft Company, Culver City, California.


Table I. Relevant electronic levels as determined from optical spectroscopy.  

<table>
<thead>
<tr>
<th>Element</th>
<th>Configuration</th>
<th>Term</th>
<th>J</th>
<th>Energy (cm⁻¹)</th>
<th>$\varepsilon_J$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tantalum</td>
<td>(5d)³(6s)²</td>
<td>$^4F$</td>
<td>3/2</td>
<td>0.00</td>
<td>-0.447</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>5/2</td>
<td>2010.10</td>
<td>-1.031</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>7/2</td>
<td>3962.92</td>
<td>-1.218</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>9/2</td>
<td>5621.04</td>
<td>-1.272</td>
</tr>
<tr>
<td>Tungsten</td>
<td>(5d)⁴(6s)²</td>
<td>$^5D$</td>
<td>0</td>
<td>0.00</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>1</td>
<td>1670.30</td>
<td>-1.51</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>2</td>
<td>3325.53</td>
<td>-1.48</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>3</td>
<td>4830.00</td>
<td>-1.50</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>4</td>
<td>6219.33</td>
<td>-1.49</td>
</tr>
<tr>
<td></td>
<td>(5d)⁵(6s)</td>
<td>$^7S$</td>
<td>3</td>
<td>2951.29</td>
<td>-1.98</td>
</tr>
<tr>
<td>Rhenium</td>
<td>(5d)⁵(6s)²</td>
<td>$^6S$</td>
<td>5/2</td>
<td>0.00</td>
<td>-1.95</td>
</tr>
<tr>
<td>Iridium</td>
<td>(5d)⁷(6s)²</td>
<td>$^4F$</td>
<td>9/2</td>
<td>0.00</td>
<td>-1.30</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>7/2</td>
<td>6323.91</td>
<td>-1.21</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>5/2</td>
<td>5784.62</td>
<td>-1.20</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>3/2</td>
<td>4078.94</td>
<td>-1.12</td>
</tr>
<tr>
<td></td>
<td>(5d)⁸(6s)</td>
<td>$^4F$</td>
<td>9/2</td>
<td>2834.98</td>
<td>-1.33</td>
</tr>
</tbody>
</table>

Table II. Summary of measured spins and inferred Nilsson state assignments.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Half-life</th>
<th>Nuclear spin</th>
<th>Nilsson state assignments</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tantalum-183</td>
<td>5 days</td>
<td>7/2</td>
<td>[404 7/2]</td>
</tr>
<tr>
<td>Tungsten-185</td>
<td>74 days</td>
<td>3/2</td>
<td>[512 3/2]</td>
</tr>
<tr>
<td>Tungsten-187</td>
<td>24 hours</td>
<td>3/2</td>
<td>[512 3/2]</td>
</tr>
<tr>
<td>Rhenium-186</td>
<td>91 hours</td>
<td>1</td>
<td>[402 5/2] [512 3/2]</td>
</tr>
<tr>
<td>Rhenium-188</td>
<td>17 hours</td>
<td>1</td>
<td>[402 5/2] [512 3/2]</td>
</tr>
<tr>
<td>Iridium-192</td>
<td>74 d.ys</td>
<td>4</td>
<td></td>
</tr>
<tr>
<td>Iridium-194</td>
<td>19 hours</td>
<td>1</td>
<td>[402 3/2] [510 1/2]</td>
</tr>
</tbody>
</table>
FIGURE CAPTIONS

Fig. 1. Apparatus used for producing refractory beams.

Fig. 2. $W^{185}$ spin search. $H = 4.20$ G.

Fig. 3. Hfs levels of $W^{185}$ and $W^{187}$ in the "$J = 3$" electronic states (schematic). $I = 3/2$.

Fig. 4. $Re^{186}$ spin search. $H = 2.82$ G.

Fig. 5. $Ir^{194}$ spin search. $H = 4.20$ G.

Fig. 6. $Ir^{194}$ full-beam decay.

Fig. 7. $Ir^{194}$ resonance decay. $I = 1$.

Fig. 8. $Ta^{183}$ spin search. $H = 2.82$ G.
Fig. 2
Fig. 3
Fig. 4
Fig. 5
Fig. 6
Fig. 7

- Count / min vs. Elapsed time (days)
- $T_{1/2} = 19\text{h} (\text{Ir}^{194})$
- $T_{1/2} = 74\text{days} (\text{Ir}^{192})$
Fig. 8
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