# Neutron Absorption and Scattering by Hafnium

L. M. BOLLINGER, S. P. HARRIS,*† C. T. HIBDON, AND C. O. MUEHLHAUSE‡
*Argonne National Laboratory, Lemont, Illinois*
(Received June 29, 1953)

Thermal activation, thermal absorption, resonance scattering, resonance transmission, and multiplicity of capture $\gamma$ rays have been measured for the stable isotopes of hafnium. Isotopic assignments of the various resonance levels are presented which are consistent with all the data observed. Where possible, the level spacing is estimated.

## INTRODUCTION

THE neutron cross-section study of a series of stable isotopes of the same atomic number provides a method for observing the changes in level density with both odd and evenness of particles and with atomic weights. A suitable example is hafnium, which has six stable isotopes: 174, 176, 177, 178, 179, and 180. One might expect to find a trend for the change of level density with mass number in the five isotopes of consecutive mass number as well as different behavior of the even-$Z$–even-$N$ (176, 178, and 180) when compared with the even-$Z$–odd-$N$ ones (177, 179).

The availability of hafnium isotopes from the Oak Ridge separation group[1] has made possible two investigations. The first, by Burson *et al.*,[2] covered the decay schemes resulting from neutron capture. This paper, the second, is concerned mainly with the low energy neutron cross-section behavior of hafnium. A series of preliminary experiments is first presented, and these are followed by the more definitive time-of-flight measurements.

## PRELIMINARY EXPERIMENTS

### A. Activation

On receipt of the enriched samples from Oak Ridge a quick search was first made for the isotope which, on thermal neutron absorption, gives rise to the 19-sec isomeric activity.[3] Known amounts of each isotope were bombarded in the pile flux of the Argonne heavy-water reactor. It was immediately obvious that $Hf^{178}$ was the initial nucleus[4] involved in the following reactions:

$$Hf^{178}+n\rightarrow Hf^{179*}+\gamma, \quad Hf^{179*}\rightarrow Hf^{179}+\gamma.$$

"Mattauch's rule" that no metastable states of even-$Z$–even-$N$ nuclei exist was obeyed. Later, however, an exception to this rule was found by Burson[2] and coworkers in their identification of the 5.6-hr hafnium activity[5] as belonging to a metastable isomer of $Hf^{180}$.

### B. Pile Oscillator Absorption

Insufficient amounts of the enriched isotopes of hafnium were available for determining thermal-absorption cross sections $<20$ barns. However, since the thermal-absorption cross section of normal hafnium[6] is $\sim$105 barns, one would expect to be able to identify one or more strongly absorbing isotopes by the pile oscillator technique.[7]

Known amounts of normal and 150 mg each of enriched hafnium oxides were oscillated in the central thimble of the Argonne heavy water reactor. These were compared with a boron standard ($\sigma_B=755$ b). An analysis of the data was made in which enrichment factors and approximate resonance contributions were taken into account. Three isotopes had detectable absorption cross sections for the method and amounts of material available. The results of these measurements were in satisfactory agreement with those of Pomerance:[6]

$$\sigma_{177}=350\pm50 \text{ barns},$$
$$\sigma_{178}=90\pm20 \text{ barns}, \quad \sigma_{179}=75\pm15 \text{ barns}.$$

Hafnium 177, 178, and 179, therefore, account for $\sim$100 barns of thermal absorption in normal hafnium. Another few barns result from 180 and 176. The low abundant $Hf^{174}$ has an absorption cross section estimated to be $\sim10^3$ barns on the basis of $\beta$ spectrographic film densities of the 70-day $Hf^{175}$ plates taken by Burson and group. It would contribute $\sim$2 barns to normal hafnium.

### C. Resonance Scattering

Thin resonance-scattering detectors of the enriched oxide samples were made by causing $\sim$50 mg to adhere to a 0.3-mil aluminum foil. The detectors thus formed were round and had an area $\sim$5 cm$^2$. These were placed in turn in the center of an annular $4\pi$ $BF_3$ proportional neutron counter.[8] A cadmium-filtered neutron beam from the Argonne heavy-water reactor was directed at the detector foils, and a neutron counting rate developed due to epi-thermal neutron scattering by the detector

* Now at Stewart-Warner Corporation, Chicago, Illinois.
† Portions of this work were presented in partial fulfillment of the requirements for the degree of Doctor of Philosophy at the Illinois Institute of Technology, Chicago, Illinois.
‡ Now at Brookhaven National Laboratory, Upton, New York.
[1] Acknowledgment is given Dr. C. P. Keim, who kindly provided the isotopically enriched samples used in this work.
[2] S. B. Burson *et al.*, Phys. Rev. **83**, 62 (1951).
[3] A. Flammersfeld, Naturwiss. **32**, 68 (1944).
[4] This result was reported privately and has appeared in various compilations.
[5] M. Goldhaber and R. D. Hill, Revs. Modern Phys. **24**, 179 (1952).
[6] H. Pomerance, Phys. Rev. **88**, 412 (1952).
[7] S. P. Harris *et al.*, Phys. Rev. **80**, 342 (1950).
[8] S. P. Harris *et al.*, Phys. Rev. **79**, 11 (1950).

TABLE I. Resonance-scattering cross sections in barns.

| Isotope | Oxide (mg/cm²) | $\sigma_{\text{epi-Cd}}-\sigma_p$ | $\Sigma_{rs}$ (corrected)[a] |
|---|---|---|---|
| 177 | 50 | ~40 | ~400±100 |
| 178 | 50 | ~20 | ~200± 50 |
| 179 | 50 | ~15 | ~150± 50 |

[a] H. Pomerance, Oak Ridge National Laboratory Report ORNL-1304, 1952 (unpublished), p. 13.

foils. Comparisons were made with both a blank aluminum foil and a graphite detector ($\bar{\sigma}_s=4.60$ b). In this manner average epi-cadmium scattering cross-sections, $\sigma_{\text{epi-Cd}}$, were obtained. Such a cross section has the following significance[7] with the particular counter employed:

$$\sigma_{\text{epi-Cd}}=\sigma_p+\frac{1}{8.4}\Sigma_{rs}, \tag{1}$$

where

$$\Sigma_{rs}=\int \sigma_{rs}d\epsilon/\epsilon \tag{2}$$

is the resonance-scattering integral, and

$$\sigma_p=\text{potential-scattering cross section.} \tag{3}$$

Therefore

$$\Sigma_{rs}=8.4(\sigma_{\text{epi-Cd}}-\sigma_p). \tag{4}$$

The resonance-scattering integral is a measure of the total resonance scattering occurring at the definite energy levels which possess a significant fraction of scattering. Equation (4) must also be slightly corrected for energy.[8]

This experiment was performed in order to ascertain which isotope possessed the significant scattering levels indicated by the authors in a previous paper.[8] An average potential scattering cross section of 23 barn for $HfO_2$ was subtracted from all epi-cadmium values. This had been previously determined by measuring the transmission cross-section for very thick filters of normal $HfO_2$. After analysis of the resulting data in terms of enrichment factors, the resonance integrals ($\Sigma_{rs}$) given in Table I were obtained. Again it may be seen that, as with thermal absorption, the principal isotopes having low-energy resonance levels are: $Hf^{177}$, $Hf^{178}$, and $Hf^{179}$. $Hf^{177}$ possessed the largest resonance scattering along with the largest thermal absorption. This result is similar to one found for tungsten, in that the principal resonance scatterer[8] ($W^{186}$) is also the principal thermal absorber.[7]

Further experiments were carried out to determine approximate energies of the levels having significant scattering. The above-mentioned detectors enriched in 177 and 178 were again placed in turn in the center of the annular scattering counter. Filters of enriched boron were placed in the neutron beam outside the counter, and boron transmission curves of the resonant scattered neutrons were obtained. Corrections were made for the effect of the boron on the neutrons scattered by potential scattering in hafnium by use of similar boron transmission curves for neutrons scattered by graphite, for which the scattering is all potential scattering. Again the data were analyzed in terms of enrichment factors, with the following results: significant resonance-scattering levels in $Hf^{177}$ and $Hf^{178}$ at ~6.7 ev and ~7.6 ev, respectively.

Since $Hf^{179}$ showed some resonance scattering, one would presume thermal energies to lie in the wing of a resonance level whose energy is <10 ev.[8] Similarly, one would expect the large thermal cross section of $Hf^{177}$ to be the result of absorption levels lower in energy than 5 ev.

### D. Capture γ-Ray Multiplicity

Following the method of one of the authors in a previous paper,[9] the neutron capture γ-ray multiplicity

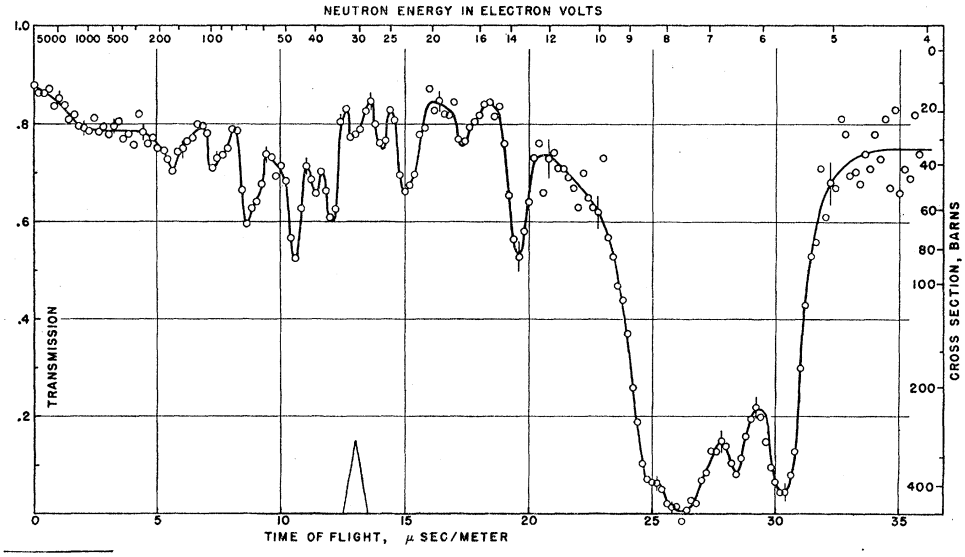


FIG. 1. The neutron transmission of a 2.45-g/cm² sample of normal $HfO_2$ in the energy range 4 to 5000 ev. The resolution used (full width at half maximum) was 0.5 μsec/meter for energies greater than 14.5 ev, and 0.6 μsec/meter for the remaining data.

[9] C. O. Muehlhause, Phys. Rev. **79**, 277 (1950).

($\bar{\nu}_\gamma$) was measured for $Hf^{177}$ and $Hf^{178}$. Thin foils of enriched material were placed in turn between two scintillation counters in coincidence which straddled a pile neutron beam. The ratio of coincidence to single counting rates was observed, and this was compared with normal hafnium that had been measured previously ($\bar{\nu}_\gamma = 3.8$).[9]

The observed multiplicities for $Hf^{177}$ and $Hf^{178}$ were $4.1 \pm 0.2$ and $3.5 \pm 0.2$, respectively. This is in qualitative agreement with the observations of the following section, namely: the level density of $Hf^{178}$ ($Hf^{177}+n$) is greater than the level density of $Hf^{179}$ ($Hf^{178}+n$).

## TIME-OF-FLIGHT MEASUREMENTS

The mechanical neutron velocity selector recently described by Selove[10] was used to investigate the transmission of hafnium oxide samples within the energy range 1 to 1000 ev. The results obtained for normal hafnium are given in Figs. 1 and 2, and those obtained for the isotopically enriched samples are given in Fig. 3. Data are not given for the enriched samples in the energy range 16 to 1000 ev because no structure was observed in that range. For the normal hafnium data, there are, in addition to the standard statistical errors indicated on the figures, possible systematic errors in the absolute level of the transmission curves, these errors being less than one percent for energies greater than 15 ev in Fig. 1 and less than three percent for the other data. The curves for the enriched samples have somewhat larger systematic errors. The transmission scales attached to the curves refer to the oxide sample, but the cross-section scales refer to the hafnium itself. The composition and thicknesses of the enriched samples that were used are listed in Table II.

In normal hafnium, transmission dips had previously been found[11] at 1.1, 2.2, 5.5, and 7.6 ev. The greater resolution used in the present work permits us to see that the pair of dips at 5.5 and 7.6 ev was caused by at least 5 resonances, at 5.6, 5.7, 6.5, 7.6, and 8.8 ev. Of these, the 6.5-, 7.6-, and 8.8-ev resonances are clearly seen in Fig. 3. An anomalous shape of the 5.5–5.7-ev transmission dip was obtained with 0.38-$\mu$sec/meter resolution (with a 20-meter neutron flight path), suggesting that it is caused by more than one resonance. Proof that this dip is caused by two resonances rests on the data obtained with the enriched samples and will be discussed in a later paragraph.

TABLE II. Composition of isotopically enriched hafnium oxide samples.

| Sample | Percentage of isotope 180 | 179 | 178 | 177 | 176 | 174 | Thickness[a] (g/cm²) |
|---|---|---|---|---|---|---|---|
| Normal | 35.3 | 13.8 | 27.1 | 18.4 | 5.2 | 0.2 | |
| 180 | 94.0 | 3.2 | 2.0 | 0.7 | 0.1 | 0.0 | 1.38 |
| 179 | 44.2 | 46.6 | 6.7 | 2.1 | 0.4 | 0.0 | 1.0 |
| 178 | 6.1 | 6.8 | 80.9 | 5.7 | 0.5 | 0.0 | 0.26 |
| 177 | 4.0 | 3.7 | 29.2 | 61.7 | 1.3 | 0.1 | 0.195 |
| 176 | 5.2 | 3.4 | 14.5 | 28.3 | 48.5 | 0.1 | 0.22 |

[a] The thicknesses listed above apply only to time-of-flight measurements. The enrichments apply to all samples including those used in pile-oscillator and scattering experiments.

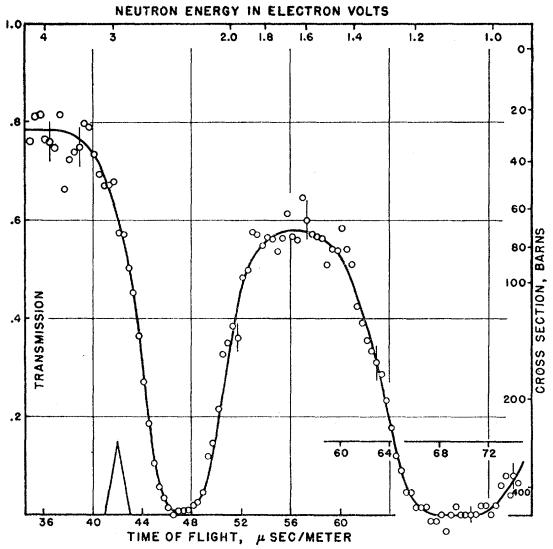


FIG. 2. The neutron transmission of normal $HfO_2$ in the energy range 1 to 4 ev with 1.1-$\mu$sec/meter resolution. The sample thickness is 2.45 g/cm².

A qualitative examination of the transmission curves obtained for the enriched samples seems to show that the 1.1-, 2.3-, 6.5-, 8.8-, and 13.6-ev dips are due to $Hf^{177}$, that the 7.6-ev dip is due to $Hf^{178}$, and that the 5.7-ev dip is due to $Hf^{179}$. A quantitative study of the isotopic assignments may be made by comparing the areas above the transmission dips obtained for the various samples. It has been shown[12] that, under favorable conditions, the area above a transmission dip provides a measure of $\sigma_0\Gamma^2$ for the resonance causing the dip. If a particular transmission dip is caused by a resonance in a single isotope, the values of $\sigma_0\Gamma^2$ determined for this resonance should be the same for all samples containing the isotope. In particular, the several values of $\sigma_0\Gamma^2$ that are calculated for each transmission dip for the hafnium samples having different abundances of the isotope in question will be equal if the correct isotopic assignment for the dip has been made and will differ markedly if a wrong assignment has been made.

Table III presents an area analysis of this kind for the hafnium resonances. The first column lists the samples for which the areas were measured. Each one of the other columns gives the values of $\sigma_0\Gamma^2$ that were obtained for a particular resonance. The number in parentheses below each value of $\sigma_0\Gamma^2$ is an estimate as to the factor by which that value may be in error because of uncertainties in area measurement. Further errors result from the fact that $\sigma_0\Gamma^2$ was calculated

[10] W. Selove, Rev. Sci. Instr. **23**, 350 (1952).
[11] P. A. Egelstaff and B. T. Taylor, Nature **167**, 896 (1951).
[12] W. W. Havens, Jr. and L. J. Rainwater, Phys. Rev. **83**, 1123 (1951).

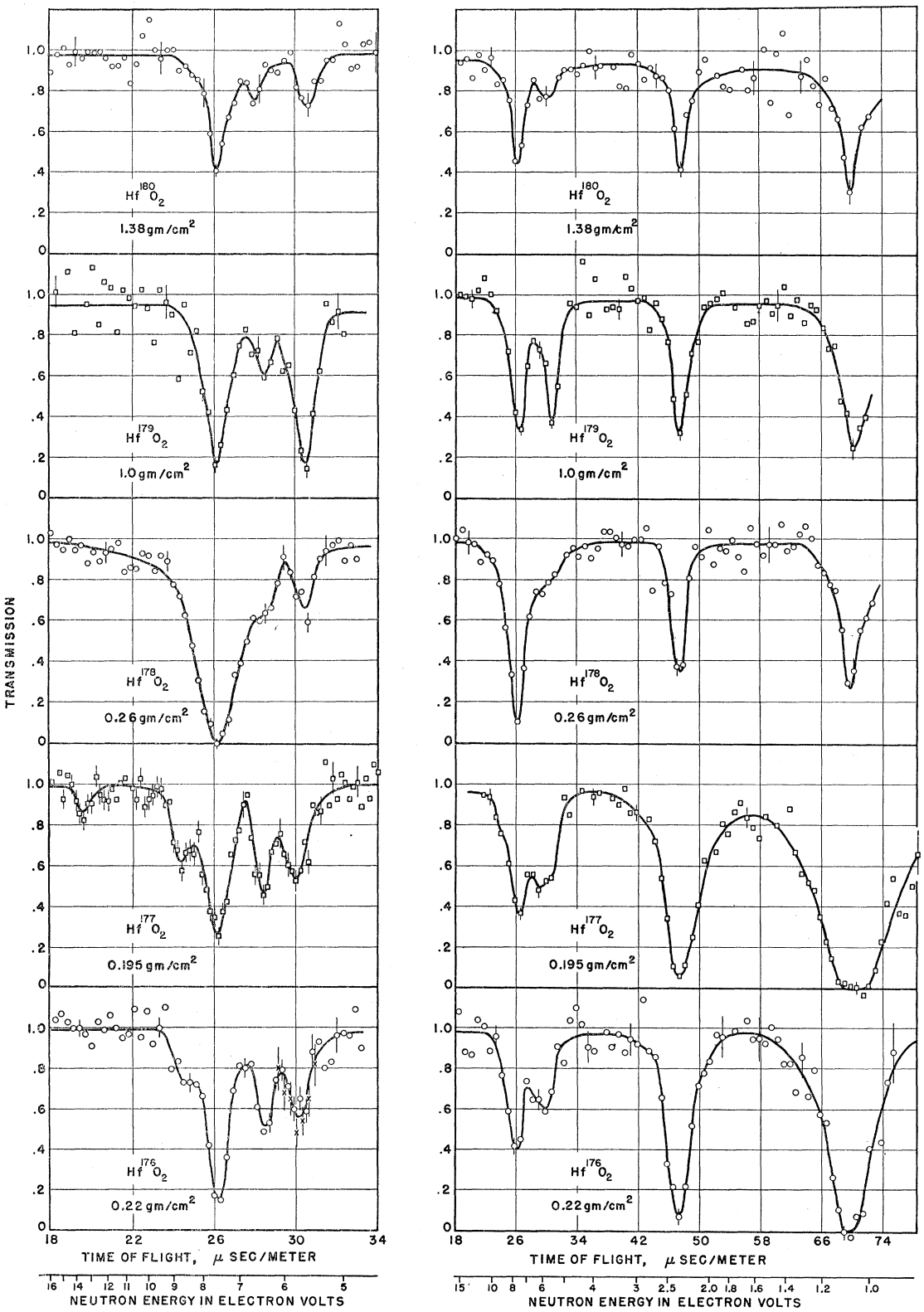


FIG. 3. The neutron transmission of isotopically enriched samples of $HfO_2$. The low energy data were obtained with 1.5-μsec/meter resolution and the higher energy data with 0.75-μsec/meter resolution. The isotopic composition of the samples is given in Table II.

under the assumption that we were dealing with thick samples; namely, that $n\sigma_0 \gg 1$, for which case $\sigma_0\Gamma^2 \rightarrow 4E_0^2A^2/n\pi$, where $A$ is the "relative area" defined in reference.[12] The thick sample assumption may not be valid for all the transmission dips that were used. In particular, it is not true for the measurements on the 1.1-ev resonance in the $Hf^{178}$, $Hf^{179}$, and $Hf^{180}$ samples. In the last row of the table is listed the isotope that was assumed to cause each resonance.

An examination of the results given in Table III for the 1.1-, 2.3-, 6.5-, 7.6-, and 13.6-ev transmission dips shows that the values of $\sigma_0\Gamma^2$ are equal within the errors listed; moreover, any other isotopic assignment fails to give satisfactory agreement. The isotopic assignment that was indicated by qualitative examination of the data is, therefore, confirmed for these resonances. For the 5.6–5.7-ev transmission dip, however, the values of $\sigma_0\Gamma^2$ that were calculated by assuming it to be due either to $Hf^{179}$ or to $Hf^{177}$ are clearly not equal. The data can be explained, however, by assuming that both $Hf^{177}$ and $Hf^{179}$ have resonances at about 5.7 ev. All the areas measured for the 5.6–5.7-ev transmission dip are in good agreement with a $\sigma_0\Gamma^2$ value of 52 barns $(ev)^2$ for the $Hf^{177}$ resonance and 25 barns $(ev)^2$ for the $Hf^{179}$ resonance.

Further support for the assumption that both $Hf^{177}$ and $Hf^{179}$ have resonances at about 5.7 ev is given by a close examination of the resonance energies observed. In the $Hf^{179}$ and $Hf^{180}$ samples, which contain very little $Hf^{177}$, the time of flight of the 5.6–5.7-ev transmission dip is 0.4 μsec/meter greater than is the corresponding time of flight for the $Hf^{177}$ and $Hf^{176}$ samples. We conclude, therefore, that of the 5.6–5.7-ev pair of resonances, the higher-energy one is due to $Hf^{177}$ and the lower-energy one is due to $Hf^{179}$.

TABLE III. Area analysis to determine the isotopes responsible for the resonances in hafnium. The values of $\sigma_0\Gamma^2$ are given in barns·$(ev)^2$. The factor by which each value may be in error is given in parentheses.

| Sample | Transmission dip (ev) 13.6 | 8.8 | 7.6 | 6.5 | (5.6–5.7) | | 2.3 | 1.1 |
|---|---|---|---|---|---|---|---|---|
| Normal 0.6 μsec/m | | 58 (2) | 1430 (1.3) | 106 (1.6) | 330 (1.5) | 250 (1.5) | | |
| Normal 1.0 μsec/m | | 20 (4) | 1420 (1.3) | 64 (1.8) | 240 (1.5) | 185 (1.5) | 310 (1.3) | 120 (1.5) |
| Normal 2.45 g/cm² | 31 (1.6) | | | | | | | |
| 180 | | | 850 (1.6) | 38 (3) | 32 (2) | 145 (2) | 240 (1.4) | 62 (1.5) |
| 179 | | | 950 (1.5) | 83 (3) | 40 (1.4) | 960 (1.4) | 200 (1.4) | 75 (1.5) |
| 178 | | | 1640 (1.3) | 69 (3) | 125 (1.8) | 145 (1.8) | 240 (1.5) | 54 (1.5) |
| 177 | 20 (2) | 59 (2) | 830 (1.4) | 52 (1.8) | 1100 (1.6) | 68 (1.6) | 290 (1.4) | 160 (1.5) |
| 176 | | 32 (3) | 1960 (1.3) | 88 (1.8) | 730 (1.8) | 88 (1.8) | 280 (1.3) | 120 (1.5) |
| Isotope assumed responsible | $Hf^{177}$ | $Hf^{177}$ | $Hf^{178}$ | $Hf^{177}$ | $Hf^{179}$ | $Hf^{177}$ | $Hf^{177}$ | $Hf^{177}$ |

TABLE IV. Summary of analysis of hafnium resonances.

| Energy (ev) | Hafnium isotope | $\sigma_0\Gamma^2$ [barn $(ev)^2$] | $\sigma_0$ (barns) | $\Gamma$ (ev) |
|---|---|---|---|---|
| 1.08±0.02 | 177 | 110± 25 | 55 000±15 000 | 0.045±0.010 |
| 2.34±0.05 | 177 | 280± 40 | >30 000 | <0.10 |
| 5.6 ±0.1 | 179 | 25± 10 | >1400 | <0.13 |
| 5.7 ±0.1 | 177 | 52± 15 | >2400 | <0.15 |
| 6.5 ±0.1 | 177 | 80± 30 | >7200 | <0.11 |
| 7.6 ±0.1 | 178 | 1400±300 | >20 000 | <0.26 |
| 8.8 ±0.2 | 177 | 55± 25 | >3600 | <0.12 |
| 13.6 ±0.4 | 177 | 28± 10 | >450 | <0.25 |

The results of the preceding paragraphs concerning the isotopic assignment of the hafnium resonances are summarized in Table IV. In addition, values of $\sigma_0$ and $\Gamma$, which have been corrected for lack of resolution, are given for the 1.1-ev resonance. Limits to the values of $\sigma_0$ and $\Gamma$ are given for the other resonances.

It is gratifying to note that the results from the time-of-flight measurements are in complete agreement with the conclusions of the previous sections. The large thermal absorber, $Hf^{177}$, possesses most of the levels observed, and these include the two of lowest energy. In addition, $Hf^{179}$ proved to have an observable resonance below 10 ev, as was expected on the basis of its thermal cross section. No levels were found in either $Hf^{176}$ or $Hf^{180}$. This, too, is in agreement with the pile oscillator measurements.

## CONCLUSION

It is evident both from the thermal cross sections and the resonance levels that the odd-$N$ isotopes have a smaller level separation $D^*$, than the even-$N$ isotopes.[8] In the case of $Hf^{177}$ it is further evident that the mean level separation is ~3 ev. Very little may be said of any of the even-$N$ isotopes except that their level separation is large and probably ~100 ev. An estimate of this in the case of $Hf^{178}$ may be made on the basis of the expression,[13] $D^* \sim 10^4\Gamma_n/E_0^{\frac{1}{2}}$. To do this, one uses the following:

$$\Sigma_s = \tfrac{1}{2}\pi\sigma_0\Gamma_n/E_0 = 200 \text{ b}, \tag{5}$$

$$\sigma_0\Gamma^2 = 1400 \text{ b}\cdot\text{ev}^2, \tag{6}$$

$$\sigma_0 = \frac{2.6\times10^6}{E_0}\cdot\frac{\Gamma_n}{\Gamma}, \tag{7}$$

$$E_0 = 7.6 \text{ ev}; \tag{8}$$

these relations are not complicated by an uncertain "$g$" factor ($g=1$ for even-$Z$—even-$N$). The above yields a value for $\Gamma_n$ of 0.023 ev, and this in turn yields $D^* \sim 85$ ev.

It may be difficult to observe additional levels in $Hf^{178}$ in the 100 to 1000-ev region with the present instrumental resolution. Such levels should be sharp (~0.2 ev) and furthermore should be masked by the presence of the dense levels of even a little $Hf^{177}$.

[13] Feshbach, Peaslee, and Weisskopf, Phys. Rev. **71**, 145 (1947).