# Search for the beta decay of $^{180}$Lu to $^{180}$Hf$^m$

S. E. Kellogg
*Nuclear Physics Laboratory, University of Washington, Seattle, Washington 98195*

E. B. Norman
*Nuclear Physics Laboratory, University of Washington, Seattle, Washington 98195*
*and Nuclear Science Division, Lawrence Berkeley Laboratory, University of California, Berkeley, California 94720*

(Received 5 August 1986)

The $t_{1/2}=5.7$ min $^{180}$Lu isotope was produced in the $^{180}$Hf(n,p) reaction and its subsequent $\beta$ decay back to $^{180}$Hf was studied with a Ge(Li) spectrometer. A radiochemical technique was used to measure the fractional population of the $I^\pi$, $K=(8^-,8)$ isomer in $^{180}$Hf to be $f_m=0.005\pm0.018\,\%$. The limit on $f_m$ is shown to be too small to account for the nucleosynthesis of $^{180}$Ta$^m$ in the $r$ process. The possible existence of a high-spin isomer in $^{180}$Lu and its astrophysical consequences are discussed.

## INTRODUCTION

Attempts have been made in recent years to explain the abundance of the naturally occurring high-spin isomer $^{180}$Ta$^m$ in terms of the slow ($s$) and/or rapid ($r$) neutron capture processes that account for the bulk of heavy element nucleosynthesis in stars.[1,2] Beer and Ward have suggested that $^{180}$Ta$^m$ may be produced through a weak $\beta$-decay branch of $^{180}$Hf$^m$, which is known to be populated in the $s$ process.[1] Figure 1 summarizes the salient features of this theory. An $r$-process contribution to the abundance of $^{180}$Ta$^m$ requires that (1) a fraction $f_m$ of the $\beta$ decays of the $I^\pi=(3^-,5^+)$ $^{180}$Lu leads to the population of the $I^\pi=8^-$ isomer in $^{180}$Hf, and (2) a fraction $f_\beta$ of the resulting $^{180}$Hf$^m$ population $\beta$ decays to the $I^\pi=9^-$ long-lived isomer in $^{180}$Ta. If atomic ionization effects are neglected, the solar abundance data compiled by Cameron[4] and the neutron capture cross-section work of Beer and Macklin[5] can be used to extract the $r$-process component of the observed solar abundance of $^{180}$Ta$^m$:

$$\left[\frac{N_r}{N_\odot}(^{180}\mathrm{Ta}^m)\right]=(1.2\pm0.2)\times10^4 f_m f_\beta\ . \tag{1}$$

Based on log$ft$ values for similar decays in neighboring nuclei, $f_\beta$ is expected to fall between 0.1% and the recently established experimental upper limit of 0.35%.[6,7] Numerically, therefore, a 100% $r$-process origin for $^{180}$Ta$^m$ requires $2\%<f_m\leq8\%$.

To date, $^{180}$Lu is the most neutron rich member observed on the $A=180$ isobar. Relatively little is known of its structure. The $t_{1/2}=5.7$ min ground state of this prolately deformed nucleus is most commonly given an $I^\pi K=(3^-,3)$ assignment,[8,9] though Ward and D'Auria argue for an $I^\pi,K=(5^+,5)$ assignment.[10] The subsequent $\beta$ transitions of $^{180}$Lu to states between 1.2 and 2.2 MeV of excitation in $^{180}$Hf have been the subject of a number of investigations.[8–12] These half dozen states in $^{180}$Hf are believed to have $K=2$, 3, or 4. All are seen to $\gamma$ cascade $\approx100\%$ of the time to the $2^+$ and $4^+$ components of the $K=0$ ground state rotational band. To an intensity of perhaps a percent, no strength has been observed to the well-studied $I^\pi,K=(8^-,8)$ isomer at 1142 keV in $^{180}$Hf. In fact, no $\gamma$ transition has been observed which feeds $^{180}$Hf$^m$, though it is readily produced in (n,$\gamma$) and (n,n') reactions.[13,14]

The present study uses a straightforward radiochemical separation technique following the production of $^{180}$Lu, and detects the subsequent $^{180}$Hf$^m$ decay. Earlier we presented a limit of $f_m\leq0.06\%$.[14] Eschner *et al.* have subsequently reported a measurement of $f_m=0.46\pm0.15\,\%$ using a multinucleon transfer technique.[15] We present here our final results, along with a possible explanation for the discrepancy between our measurement and that of Eschner *et al.*

## EXPERIMENT

Bulk samples of natural Hf metal with masses between 1 and 2 g were first activated with thermal neutrons at the University of Washington reactor to introduce the long-lived ($t_{1/2}=42$ day) isotope $^{181}$Hf as a radiochemical tracer. After a suitable cooldown period of 1–4 weeks, the "spiked" Hf samples were wrapped in Cd foils and inserted behind the water-cooled Be target/beam stop at the University of Washington cyclotron. The Hf samples were bombarded with fast neutrons generated by the $^9$Be(d,n) reaction using 22 MeV deuterons. Following an irradiation period of 5–10 min, the samples were expeditiously transported to a radiochemistry laboratory. The desired $^{180}$Lu, along with other rare earth activities, was extracted by dissolving the Hf sample in HF acid and precipitating out $LuF_3$ with the addition of Y as a carrier. The precipitation was accelerated by the use of a centrifuge, and a radiochemical "hafnium reduction factor" of $10^4$ was achieved in less than a $^{180}$Lu half-life (5 min). Geometrically similar samples containing about 1 ml of solution and precipitate were prepared. The precipitate was first counted with a 135 cm$^3$ Ge(Li) detector in a reproducible geometry to obtain the abundance of $^{180}$Lu.

 

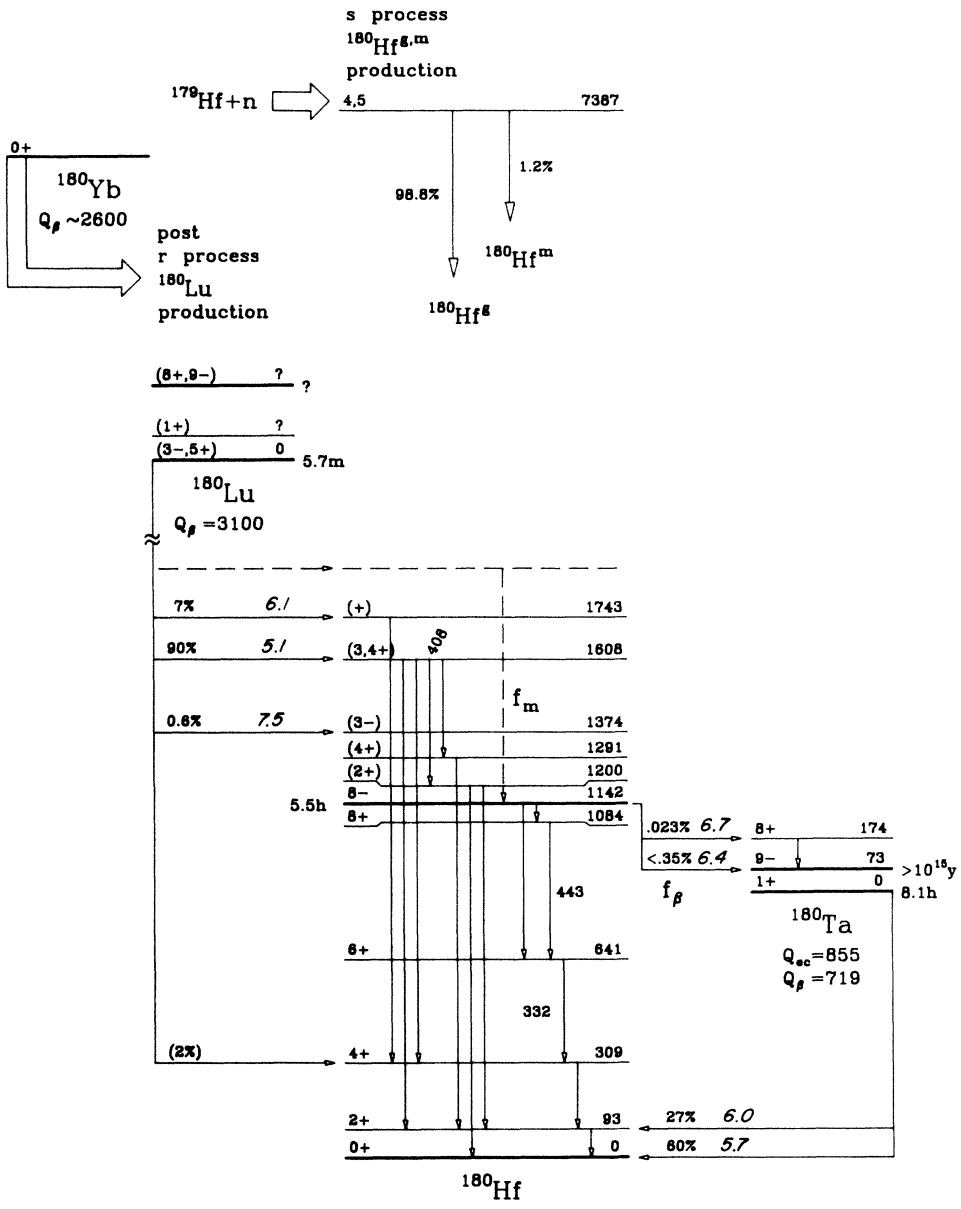


FIG. 1. A partial energy level scheme for $^{180}$Hf (Ref. 3) and the neutron-capture production paths to $^{180}$Ta$^m$ (Ref. 1). In the Beer and Ward model, $^{180}$Ta$^m$ can be produced in both the $s$ and $r$ processes by the population of $^{180}$Hf$^m$ followed by a weak $\beta$-decay branch, $f_\beta$. This experiment establishes a limit on $f_m$, the $r$-process fractional production of $^{180}$Ta$^m$ following the $\beta$ decay of $^{180}$Lu (dashed beta-gamma path). Shown are gamma transitions in the decay of $^{180}$Lu and $^{180}$Hf$^m$ with intensities greater than 2%. The three gamma rays specifically used in the determination of $f_m$ in this experiment are labeled. The levels in $^{180}$Lu are discussed in the text. All energies are in keV and the italicized numbers are log$ft$ values.

Several hours later, the solution was counted with the same detector to obtain the abundance of $^{180}$Hf$^m$ [produced directly in the $^{180}$Hf(n,n') reaction] and of the $^{181}$Hf tracer. Finally, the precipitate was recounted in 1 h time bins to determine the residual abundances of $^{180}$Hf$^m$ and $^{181}$Hf. The Ge(Li) detector was shielded for low-level counting throughout the experiment and its resolution was 1.5 keV [full width at half maximum (FWHM)] at 500 keV. Efficiency measurements were performed with standard sources in the experimental geometry.

Many activations were performed using Hf metal (containing 3% Zr) and "spectrographic-grade" Hf (156 ppm Zr). Typical $\gamma$ spectra between 275 and 525 keV at each stage of counting are shown in Fig. 2 for a Hf metal run. Table I lists the activities that were identified in the precipitate throughout the course of the experiment. Most of the activity between 5 and 10 h after the fast neutron bombardment proved to be from the Y and Sr products of Zr(n,p) and Zr(n,$\alpha$). While the spectrographic-grade runs were free of contaminating lines, the Hf reduction factor could not be pushed above $\approx$1000.

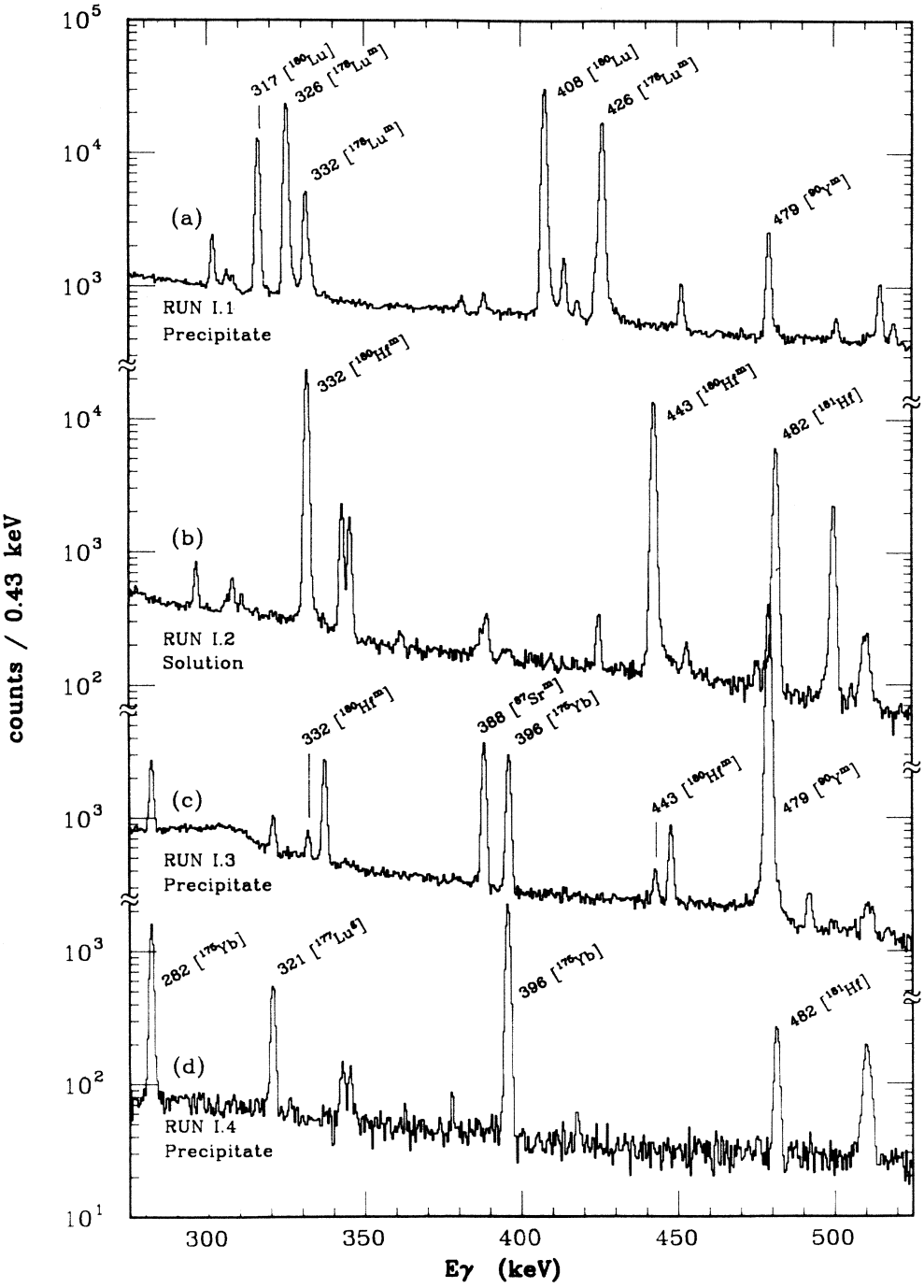


FIG. 2. Ge(Li) gamma-ray spectra following the radiochemical separation of lutetium (in precipitate form) from hafnium (in solution). The $^{180}$Lu, $^{180}$Hf$^{m}$, and $^{181}$Hf gamma rays used in this experiment are labeled as well as strong lines from other Lu, Yb, Y, and Sr activities. The chronologically ordered spectra (a)—(d) emphasize activities with vastly different half-lives. Counting times are listed in Table II. The counting geometries were identical for the solution (b) and the precipitate (a), (c), and (d).

## DERIVATION OF $f_m$ AND RESULTS

The final abundance of $^{180}$Hf$^{m}$ in the target has two sources. The first comes from prompt production reactions like inelastic scattering [$^{180}$Hf(n,n')] and thermal neutron capture [$^{179}$Hf(n,$\gamma$)]. The smaller second source comes from the fraction $f_m$ of the decay of $^{180}$Lu:

$$[N(^{180}\mathrm{Hf}^{m})]=[N(^{180}\mathrm{Hf(n,n')})]+f_m[N(^{180}\mathrm{Lu})]\ . \quad (2)$$

As described in the preceding section, the radiochemistry employed separates activities which are precipitated out of HF acid from those left in solution. The Hf reduction

TABLE I. Activities observed in the precipitate after fast neutron bombardment of natural Hf metal in run I. A Zr content of 3% is assumed. The ratio of $^{180}$Hf$^m$/$^{180}$Lu was determined in a separate activation to be $\approx 30$. Excitation energies for the isomers have been subtracted from the $Q$ values of Ref. 16.

| Observed activity | Primary reaction | $T_{1/2}$ | $Q$ value (MeV) | $\Delta I^\pi$ | Cross section relative to $^{180}$Hf$^m$ = 1000 |
|---|---|---|---|---|---|
| $^{87}$Sr$^m$ | $^{90}$Zr(n,$\alpha$) | 2.8 h | +1.362 | $\frac{1}{2}^-$ | 22 |
| $^{91}$Sr | $^{94}$Zr(n,$\alpha$) | 9.5 h | +2.067 | $\frac{5}{2}^+$ | 33 |
| $^{93}$Y$^g$ | $^{96}$Zr(n,$\alpha$) | 10.2 h | +0.170 | $\frac{1}{2}^-$ | 750 |
| $^{90}$Y$^m$ | $^{90}$Zr(n,p) | 3.19 h | −2.188 | $7^+$ | 280 |
| $^{91}$Y$^m$ | $^{91}$Zr(n,p) | 49.7 min | −1.318 | $2^+$ | 610 |
| $^{92}$Y | $^{92}$Zr(n,p) | 3.54 h | −2.841 | $2^-$ | 430 |
| $^{94}$Y | $^{94}$Zr(n,p) | 18.7 min | −4.22 | $2^-$ | 160 |
| $^{175}$Yb | $^{178}$Hf(n,$\alpha$) | 4.19 d | +7.905 | $\frac{7}{2}^-$ | 51 |
| $^{177}$Yb$^g$ | $^{180}$Hf(n,$\alpha$) | 1.9 h | +6.856 | $\frac{9}{2}^+, \frac{1}{2}^-$ | 16 |
| $^{176}$Lu$^m$ | $^{176}$Hf(n,p) | 3.68 h | −0.533 | $1^-$ | 63 |
| $^{177}$Lu$^g$ | $^{177}$Hf(n,p) | 6.71 d | +0.285 | $0^-$ | 20 |
| $^{177}$Lu$^m$ | $^{177}$Hf(n,p) | 160.9 d | −0.685 | $8^+$ | 3 |
| $^{178}$Lu$^g$ | $^{178}$Hf(n,p) | 28.4 min | −1.470 | $1^+$ | 56 |
| $^{178}$Lu$^m$ | $^{178}$Hf(n,p) | 23.0 min | −1.77 | $9^-$ | 16 |
| $^{179}$Lu | $^{179}$Hf(n,p) | 4.9 h | −0.570 | $1^+$ | 420 |
| $^{180}$Lu | $^{180}$Hf(n,p) | 5.7 min | −2.52 | $(3^-,5^+)$ | 33 |

factor $\xi$ is defined as the ratio of solution (sn) to precipitate (pt) abundance. A quantity $\eta$ is similarly defined to describe the effectiveness with which Lu is recovered in the precipitate:

$$\xi=\frac{[N_{sn}(^{181}\text{Hf})]}{[N_{pt}(^{181}\text{Hf})]},\quad \eta=\frac{[N_{pt}(^{180}\text{Lu})]}{[N_{sn}(^{180}\text{Lu})]}, \tag{3}$$

where continuity $[N]=[N_{pt}]+[N_{sn}]$ is assumed. A general figure of merit for the success of the radiochemistry in this experiment is $\eta\xi/(\eta+1)$. Typically, $\xi\approx 1000$–$40\,000$ and $\eta/(\eta+1)\approx 50$–$90\%$ were achievable. The quantities of $^{180}$Hf isomer which end up in the precipitate and solution can then be written as

$$\begin{aligned}[N_{pt}(^{180}\text{Hf}^m)]&=\frac{1}{\xi+1}[N(^{180}\text{Hf(n,n')})]\\&\quad+f_m[N_{pt}(^{180}\text{Lu})],\\ [N_{sn}(^{180}\text{Hf}^m)]&=\frac{\xi}{\xi+1}[N(^{180}\text{Hf(n,n')})]\\&\quad+f_m\frac{1}{\eta}[N_{pt}(^{180}\text{Lu})].\end{aligned} \tag{4}$$

Combining these two expressions to eliminate $[N(^{180}\text{Hf(n,n')})]$ and then substituting the ratio of tracer activities in (3) for $\xi$, $f_m$ is given by

$$f_m=(1+\delta)\left\{\frac{[N_{pt}(^{181}\text{Hf})]}{[N_{pt}(^{180}\text{Lu})]}\right\}\left\{\frac{[N_{pt}(^{180}\text{Hf}^m)]}{[N_{pt}(^{181}\text{Hf})]}-\frac{[N_{sn}(^{180}\text{Hf}^m)]}{[N_{sn}(^{181}\text{Hf})]}\right\}, \tag{5}$$

where $1+\delta=(1-1/\eta\xi)^{-1}$. $\delta$ is of the order $10^{-4}$ and is subsequently neglected. The five bracketed abundances were measured by integrating photopeak areas in their most favorable time windows $(t_{sta}, t_{sta}+t_{dur})$ and extrapolating back to an initial time $t_0$. The initial time was taken to be the last step in the radiochemical separation. Because $f_m$ is expressed in terms of ratios, only relative $\gamma$ line intensities and Ge(Li) detector efficiencies were needed. Furthermore, these only affect the scaling of the final result. The 408- and 482-keV $\gamma$ lines in $^{180}$Lu and $^{181}$Hf were used as the primary lutetium and tracer reference lines. The 332- and 443-keV $\gamma$ lines in $^{180}$Hf$^m$ were used separately in the analysis. For the 443-keV line, then,

$$f_m=\left[\frac{i_\gamma\epsilon_p\epsilon_s(408)}{i_\gamma\epsilon_p\epsilon_s(443)}\right]\left\{\frac{[FN_{pt}(482)]}{[FN_{pt}(408)]}\right\}\times\left\{\frac{[FN_{pt}(443)]}{[FN_{pt}(482)]}-\frac{[FN_{sn}(443)]}{[FN_{sn}(482)]}\right\}, \tag{6}$$

where

$$F=\{(e^{-\lambda t_{sta}})(1-e^{-\lambda t_{dur}})\}^{-1}$$

is the inverse of the fraction of total decays which occur in the time window $(t_{sta}, t_{sta}+t_{dur})$, $i_\gamma$ is the absolute gamma line intensity, $\epsilon_p$ is the measured photopeak efficiency of the Ge(Li) detector, and $\epsilon_s$ is the correction for $\gamma$-cascade summing out of the photopeak.

Table II lists the photopeak areas for three independent runs, each analyzed for both the 332- and 443-keV $\gamma$ lines. The limitation on the Hf metal runs (I and II) is purely statistical, while the limitation on the spectrographic-grade run (III) is probably systematic since it is difficult to extract photopeak areas with an accuracy better than

TABLE II. Efficiencies and run by run photopeak areas used in three independent activations to determine $f_m$. The 332- and 443-keV $^{180}Hf^m$ $\gamma$ lines are treated separately. Only $\approx 1$ out of 50 ml of solution (sn) was counted to match the geometry used in the precipitate (pt) runs.

| Activity | | | $^{180}Lu^g$ | $^{180}Hf^m$ | $^{180}Hf^m$ | $^{181}Hf$ |
|---|---|---|---|---|---|---|
| $t_{1/2}$ | | | 5.7 min | 5.518 h | 5.518 h | 42.4 d |
| $E_\gamma$ (keV) | | | 408.1 | 332.3 | 443.2 | 482.0 |
| $i_\gamma$ | | | 0.50 | 0.944 | 0.821 | 0.810 |
| $\epsilon_p(E_\gamma)$ | | | 0.031 | 0.038 | 0.029 | |
| $\epsilon_s(E_\gamma)$ | | | 0.91 | 0.77 | 0.77 | |
| Run (pt/sn) | $t_{sta}$ (min) | $t_{dur}$ (min) | Photopeak areas (counts) | | | |
| I (Hf metal) | | | | | | |
| 1. pt | 1.6 | 4.7 | 110270±330 | | | |
| 2. sn | 226 | 18 | | 76260±280 | 50270±230 | 22620±150 |
| 3. pt | 364 | 420 | | 973±67 | 635±48 | |
| 4. pt | 7940 | 850 | | | | 1015±37 |
| II (Hf metal) | | | | | | |
| 1. pt | 1.6 | 13.6 | 112300±350 | | | |
| 2. sn | 101 | 63 | | 186860±450 | 121770±360 | 99230±320 |
| 3. pt | 302 | 661 | | 280±32 | 251±35 | |
| 4. pt | 5902 | 392 | | | | 249±20 |
| III (Spectr.-grd. Hf[a]) | | | | | | |
| 1. pt | 1.8 | 6.1 | 78940±290 | | | |
| 2. sn | 129 | 63 | | 103204±330 | 67554±310 | 36090±270 |
| 3. pt | 201 | 654 | | 21610±160 | 13800±120 | 14840±130 |

[a]Spectr.-grd. denotes spectrographic grade.

1%. Table III gives the six determinations of $f_m$. We combine only the four values of $f_m$ from the Hf metal runs to obtain $f_m = 0.005 \pm 0.018\%$. To this is added a scaling uncertainty of 10%, mostly due to the uncertainty in $i_\gamma(408)$ to obtain a firm upper limit (67% confidence level) on $f_m$ of 0.026%.

## DISCUSSION

The measurement by Eschner *et al.* of $f_m = 0.46 \pm 0.15\%$ (Ref. 15) is clearly incompatible with the null result reported here. There are two likely explanations for this discrepancy. (1) An error in experimental technique was made by one of the groups, or (2) a high-spin, short-lived isomer of $^{180}Lu$ exists to which the experiment of Eschner *et al.* was more sensitive. It will be helpful to compare the technique of Eschner *et al.* with our own. It will also prove worthwhile to explore the second explanation for its nuclear and astrophysical consequences. In the following text, superscripts "$g$" and "$m$" are used to differentiate between quantities which refer to the ground state ($t_{1/2} = 5.7$ min) and the proposed short-lived isomeric state in $^{180}Lu$.

Both experiments are similar in their use of a Ge(Li) detector to measure the activities of $^{180}Lu$, $^{180}Hf^m$, and a "tracer" Hf isotope, but differ in the production reaction used and in the method employed to subtract the directly produced $^{180}Hf^m$. In the experiment of Eschner *et al.*, a beam of 8.6 MeV/nucleon $^{136}Xe$ was directed onto a W target/ion source. $^{180}Lu$ (and other rare-earth reaction products) diffused out of their target on a timescale of 10 sec and was then run through a mass spectrometer to a catcher foil for on-line and later off-line counting. Discrimination against the promptly produced $^{180}Hf^m$ relied on (1) different Lu and Hf diffusion time constants, (2) different Lu and Hf ionization efficiencies, and (3) relative production cross sections for $^{180}Hf^m$ and $^{177}Hf^m$, which played the role of the "tracer."[17] Eschner *et al.* reported a fast release time constant for lutetium, $\alpha_{Lu} = 5^{+5}_{-2} \times 10^{-2}$ sec$^{-1}$, which corresponds to a half-life against release of between 7 and 23 sec. $\alpha_{Hf} = (7.4 \pm 1.0) \times 10^{-4}$ sec$^{-1}$ was much slower.

TABLE III. The fractional population $f_m$ of $^{180}Hf^m$ following the $\beta$ decay of $^{180}Lu$. The final experimental results are derived from formula (6) and the data in Table II for the 332 and 443 keV lines in each of three runs. While the values from run III (which used spectrographic-grade Hf) are not included in the statistically weighted average, they are nevertheless consistent with the null result obtained from runs I and II.

| Run | $f_m(332)$ (%) | $f_m(443)$ (%) |
|---|---|---|
| I | $+0.019 \pm 0.040$ | $+0.014 \pm 0.042$ |
| II | $-0.025 \pm 0.027$ | $+0.042 \pm 0.038$ |
| III | $+0.20 \pm 0.14$ | $-0.06 \pm 0.15$ |

Statistically weighted average of runs I and II: $f_m = 0.005 \pm 0.18\%$

Both groups have proffered the possible existence of a high-spin, short-lived isomer in $^{180}$Lu to explain the discrepancy in the measurement of $f_m$.[14,15] With this assumption, the 5.7 min ground state need contribute nothing to the population of $^{180}$Hf$^m$ ($f_m^g=0$), consistent with our results, while the proposed isomer must have a high spin to allow $f_m^m>0.5\%$, consistent with the measurement of Eschner *et al.* The isomeric production fraction

$$P_m=\frac{\sigma_m}{\sigma_g+\sigma_m} \tag{7}$$

is undoubtedly different in the two experiments, and is probably greater for the multinucleon transfer reaction. In fact, however the (n,p) reaction readily populates high spin states. In the $^{178}$Hf(n,p) reaction, for example, the isomeric production fraction for the $9^-$ isomeric state is $P_m=0.22$, as shown in Table I.

The proposed isomer of $^{180}$Lu should have a half-life in the range from 1 to 100 sec, long enough to be recovered by the diffusion method of Eschner *et al.* but short enough so that it has transmuted into $^{180}$Lu$^g$ and $^{180}$Hf$^m$ during the 5 min radiochemical separation in our experiment. This is possible if $f_m^m$ competes not against $\beta$ transitions that lead to the ground state of $^{180}$Hf, but rather with $\gamma$ transitions that feed the 5.7 min $^{180}$Lu$^g$. It is reasonable then to express $f_m^m$ as a ratio of partial $\beta$ and $\gamma$ transition rates:

$$f_m^m=\frac{\lambda_\beta^m}{\lambda_\gamma^m+\lambda_\beta^m}\ . \tag{8}$$

If the proposed $^{180}$Lu isomer decays most of the time by low energy, heavily converted electromagnetic transitions, it may have escaped direct detection and gone unnoticed in previous experiments. Indeed, the shape of the on-line build-up curve for $^{180}$Lu displayed in Fig. 1 of Ref. 15 is identical for the 10 sec release half-life described by Eschner *et al.*, as it would be for the in-feeding of $^{180}$Lu$^g$ from a 10 sec half-life isomer.

Figure 3 pictorially summarizes the results from the two experiments. It displays the limits on the product of $P_m$ and $f_m^m$ as a function of the half-life of the proposed isomer of $^{180}$Lu. Our experiment excludes half-lives greater than about 100 sec, while that of Eschner *et al.* implies that the half-life must be greater than about 1 sec. The shaded region is allowed by both experiments. The dashed line in Fig. 3 represents a solution for $f_m^m$ assuming that the production fraction $P_m=0.5$ and the partial $\beta$ half-life $t_{1/2}^{m,\beta}=42$ min. Such a half-life corresponds to a log$ft$ of 6.4 for a direct $\beta$ transition to $^{180}$Hf$^m$, which is a reasonable estimate for the strength of an allowed hindered transition.[18] The dashed line passes through the shaded region for isomeric half-lives between 20 and 40 sec. Finally, the data points at 5.7 min represent the measurements of Eschner *et al.* and ourselves for $f_m^g$ with the assumption that the proposed isomer of $^{180}$Lu does not exist (or $P_m=0$).

There are theoretical reasons to believe that a low-lying, high-spin isomer could exist in $^{180}$Lu. By coupling the known odd proton and neutron quasiparticle states near their respective Fermi surfaces, it is possible to construct

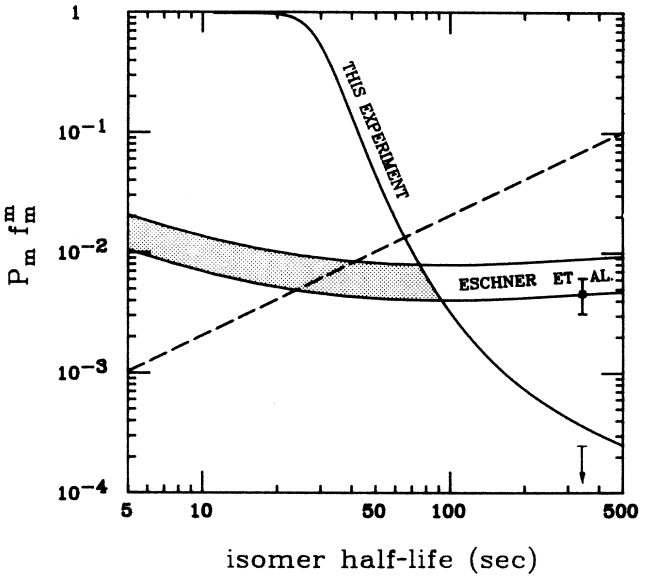


FIG. 3. Interpretation of $f_m$ in terms of a proposed high-spin isomer of $^{180}$Lu. The ordinate is the product of the $^{180}$Lu isomer's production factor, $P_m$, and its $\beta$-decay fraction that populates $^{180}$Hf$^m$, $f_m^m$. The result of this experiment (shown as the lower data point at 5.7 min) also establishes an upper limit of about 100 sec on the half-life of such an isomer. We have solved the equations of Ref. 15 including a source term for a $^{180}$Lu isomer to generate curves for the upper and lower limits implied by the positive value reported by Eschner *et al.* (upper data point). The shaded region is in agreement with both experimental measurements. The dashed line is described in the text.

both low-spin ($I=1$–$5$) and high-spin ($I=7$–$10$) states. The region of isotopes around $^{180}$Lu is filled with systematic examples of such low- and high-spin $K$ isomers. In terms of the unified model, the $t_{1/2}=5.7$ min ground state of $^{180}$Lu is most probably the $I^\pi,K=(3^-,3)$ coupling of the odd proton $\frac{7}{2}^+[404]_\pi$ and the odd neutron $\frac{1}{2}^-[510]_\nu$ quasiparticles.[8,9] Ward and D'Auria prefer an $I^\pi,K=(5^+,5)$ assignment, coupling $\frac{9}{2}^-[514]_\pi$ with $\frac{1}{2}^-[510]_\nu$.[10] These states are seen within 20 keV of each other in $^{182}$Ta. The $\frac{7}{2}^+[404]_\pi$ and $\frac{9}{2}^+[624]_\nu$ states can couple with their particle spins aligned to form the low-spin $1^+$ state observed as the ground states of $^{176}$Lu, $^{178}$Lu, and $^{180}$Ta. With their particle spins antialigned, these same orbitals couple to form an $8^+$ state seen at 174 keV of excitation in $^{180}$Ta.[18] Finally, the $\frac{9}{2}^-[514]_\pi$ and $\frac{9}{2}^+[624]_\nu$ orbitals can couple to form the high-spin $9^-$ isomers seen at 73 keV of excitation in $^{180}$Ta and at about 300 keV in $^{178}$Lu. Hoff has recently calculated that the $8^+$ and $9^-$ states should lie within 200 keV of the $3^-$ ground state of $^{180}$Lu.[19] Hoff also has placed the $I^\pi=5^+$ state approximately 50 keV below these prospective $^{180}$Lu isomer candidates.

If the $9^-$ state is the lower of the two high-spin states in $^{180}$Lu, and is thus the isomer, the $M4$ transition to the $5^+$ state will be much slower than the allowed $\beta$ transition

to the $8^-$ isomer in $^{180}$Hf. Using the observed log$ft$ of 6.4 for the analogous decay of $^{180}$Hf$^m$, a partial half-life of 40 min is calculated. Such an isomer should have been easily produced and observed previously. Suppose, on the other hand, that the $8^+$ state plays the role of the $^{180}$Lu isomer. The single particle estimate for the $M3$ transition to the $5^+$ level yields a partial half-life of about 10 sec. The $\beta$-branching ratio for the high-spin isomer is then on the order of 1%, consistent with the measurement of Eschner *et al.* and with our null result. Other $M3$ transitions in the rare earths occur a factor of 10—100 times slower than the single-particle rate,[20] but this factor can be recovered by increasing the energy difference between the $5^+$ and $8^+$ states. We note that the prospective $8^+$ isomer cannot lie any lower than about 150 keV in energy since it is the spin antialigned member of a Gallagher-Moszkowski doublet with the $1^+$ state.[21] If the $t_{1/2}=5.7$ min activity of $^{180}$Lu is the $K^\pi=3^-$ state, then the $1^+$ state must lie above it.

Beyond the circumstantial evidence discussed here, there is no direct evidence supporting the existence of a high-spin isomer in $^{180}$Lu. Takahashi *et al.* reported a $2.5\pm0.5$ min $\beta$ activity with a $3.3\pm0.1$ MeV endpoint.[12] This was interpreted as the $\beta$ decay from a *low-spin* isomer ($1^+$?) directly to the ground state of $^{180}$Hf, but it has not been confirmed by subsequent $\beta$ and $\gamma$ experiments. Zychor *et al.* specifically searched for evidence of high-spin Lu isomers in a $\gamma$-ray singles experiment following the bombardment of $^{nat}$W with $^{136}$Xe and reported null results for half-lives greater than 2 min.[17] The following paper of Lesko *et al.*[22] describes an investigation tailored to search for isomers of $^{180}$Lu with the properties described here. Their null results for half-lives greater than 10 sec lessens the likelihood that an isomer can be invoked to explain the discrepancy between the present results and those of Eschner *et al.*[15]

## ASTROPHYSICAL IMPLICATIONS

In the absence of a high-spin isomer of $^{180}$Lu, our measured limit on $f_m$ clearly excludes an $r$-process origin for $^{180}$Ta$^m$. The existence of an isomer would change this conclusion. Little is known about the site of the $r$ process which accounts for the nucleosynthesis of half of the elements heavier than iron and all of the actinides.[23] To achieve the high density of neutrons necessary to create the very neutron-rich progenitor matter probably involves a thermal transient to peak temperatures of $(1-3)\times10^9$ K. The post $r$ process is characterized by a rapidly falling temperature during which time nuclei $\beta$ decay back to the valley of $\beta$ stability. $^{180}$Lu is probably reached within the first minute when the temperature remains high. Thermal equilibration of the high-spin isomeric and low-spin ground states will be mediated by higher lying states of intermediate spin via processes which include photoabsorption, Coulomb excitation, and positron annihilation. As the temperature continues to fall, this communication link eventually will be broken, freezing out the isomeric population. $^{180}$Lu$^m$ would still be hindered from making direct isomeric transitions to the ground state because $K$- and $L$-shell ionization would block the internal conversion channel. (For example, $\alpha_L=10^5$ for a 50-keV $M3$ transition.) Thus the $\beta$-decay branch of $^{180}$Lu$^m$ would be significantly enhanced. ($f_m^m\approx100\%$.) Finally, the continuing drop in temperature increases the survivability of the high-spin isomers of $^{180}$Hf and $^{180}$Ta against the same processes responsible for the thermalization of $^{180}$Lu.[24]

The $\beta$-decay yields of the isomeric and ground states of $^{180}$Hf, and consequently the $r$-process production of $^{180}$Ta$^m$, are sensitive to the time-integrated thermodynamics of the post $r$ process. Therefore it may be possible to combine future detailed nuclear structure data for $^{180}$Lu with the observed solar abundance of $^{180}$Ta$^m$ to place constraints on the time-temperature history of the post $r$ process. Ultimately, such constraints may aid in the determination of the $r$-process sites itself.

## CONCLUSION

We have measured the fractional population of $^{180}$Hf$^m$ following the $\beta$ decay of $^{180}$Lu to be $f_m=0.005\pm0.018$ %, in disagreement with the positive value reported by Eschner *et al.* We believe that our measurement, which employs a simpler technique and yields a null result with a much smaller statistical uncertainty, is the correct value for the 5.7 min activity of $^{180}$Lu. Astrophysically, $f_m$ is too small to account for the observed abundance of $^{180}$Ta$^m$ by straightforward $r$-process production. We have shown that the experimental discrepancy can be reconciled if the multinucleon transfer reaction of Eschner *et al.* populated a high-spin, short-lived isomer in $^{180}$Lu. Further, the existence of such an isomer may substantially enhance the resultant nucleosynthesis of $^{180}$Ta$^m$. With more complete nuclear structure information, it may be possible to establish constraints on the post-$r$-process astrophysical environment. Clearly more work is called for in the understanding of the doubly odd $A=180$ nuclei and in the pinpointing of the $r$-process site before the origin and fate of $^{180}$Ta$^m$ is known.

## ACKNOWLEDGMENTS

We wish to thank Pat Miller and Deloss Fry of the University of Washington Nuclear Engineering Department for our many thermal neutron irradiations. We also wish to thank the Cyclotron staff at the Nuclear Physics Lab, especially Peter Wiest, Georgia Rohrbaugh, and Joel Schoen for delivering intense deuteron beams above the machine ratings. We are indebted to Eric Adelberger and Wick Haxton for reading the manuscript and helpful comments. This work was funded in part by the Department of Energy, Division of Nuclear Physics at the University of Washington under Contract No. DE-AC06-81ER40048 and at Lawrence Berkeley Laboratory under Contract No. DE-AC03-76SF00098.

[1]H. Beer and R. A. Ward, Nature (London) **291**, 308 (1981).
[2]K. Yokoi and K. Takahashi, Nature (London) **305**, 198 (1983).
[3]*Table of Isotopes,* 7th ed., edited by C. M. Lederer and V. S. Shirley (Wiley, New York, 1978).
[4]A. G. W. Cameron, in *Essays in Nuclear Astrophysics,* edited by C. A. Barnes, D. D. Clayton, and D. N. Schramm (Cambridge University Press, Cambridge, 1982), p. 23.
[5]H. Beer and R. L. Macklin, Phys. Rev. C **26**, 1404 (1982).
[6]C. J. Gallagher, M. Jørgenson, and O. Skilbreid, Nucl. Phys. **33**, 285 (1962).
[7]S. E. Kellogg and E. B. Norman (to be published).
[8]D. L. Swindle, T. E. Ward, and P. K. Kuroda, Phys. Rev. C **3**, 259 (1971).
[9]S. C. Gujrathi and J. M. D'Auria, Nucl. Phys. **A161**, 410 (1971).
[10]T. E. Ward and J. M. D'Auria, Phys. Rev. C **10**, 1227 (1974).
[11]A. H. W. Aten, Jr. and A. G. Funke-Klopper, Physica **26**, 79 (1960).
[12]K. Takahashi, T. Kuroyanagi, H. Yuta, K. Kotajima, K. Nagatani, and H. Morinaga, J. Phys. Soc. Jpn. **16**, 1664 (1961).
[13]D. L. Bushnell, D. J. Buss, and R. K. Smither, Phys. Rev. C **10**, 2483 (1974).
[14]S. E. Kellogg and E. B. Norman, Bull. Am. Phys. Soc. **28**, 989 (1983); in Proceedings of the Second Workshop on Nuclear Astrophysics, Ringberg Castle, Tegernsee, West Germany, Max-Planck-Institut für Astrophysik Report No. 90, 1983, p. 92.
[15]W. Eschner, W.-D. Schmidt-Ott, K. L. Gippert, E. Runte, H. Beer, G. Walter, R. Kirchner, O. Klepper, E. Roeckl, and D. Schardt, Z. Phys. A **317**, 281 (1984).
[16]N. B. Gove and A. H. Wapstra, Nucl. Data Tables **11**, 127 (1972).
[17]I. Zychor, K. Rykaczewski, H. Ahrens, H. Folger, W. Kurcewicz, K. Summerer, N. Kaffrel, and N. Trautmann, Radiochim. Acta **33**, 1 (1983).
[18]S. E. Kellogg and E. B. Norman, Phys. Rev. C **31**, 1505 (1985).
[19]R. W. Hoff, private communication.
[20]K. E. G. Lobner and S. G. Malmskog, Nucl. Phys. **80**, 505 (1966).
[21]C. J. Gallagher and S. A. Moszkowski, Phys. Rev. **111**, 1282 (1958).
[22]K. T. Lesko, E. B. Norman, D. M. Moltz, R. M. Larimer, S. G. Crane, and S. E. Kellogg, Phys. Rev. C **34**, 2256 (1986), the following paper.
[23]D. N. Schramm, in *Essays in Nuclear Astrophysics,* Ref. 4, p. 325.
[24]E. B. Norman, S. E. Kellogg, T. Bertram, S. Gil, and P. Wong, Astrophys. J. **281**, 360 (1984).

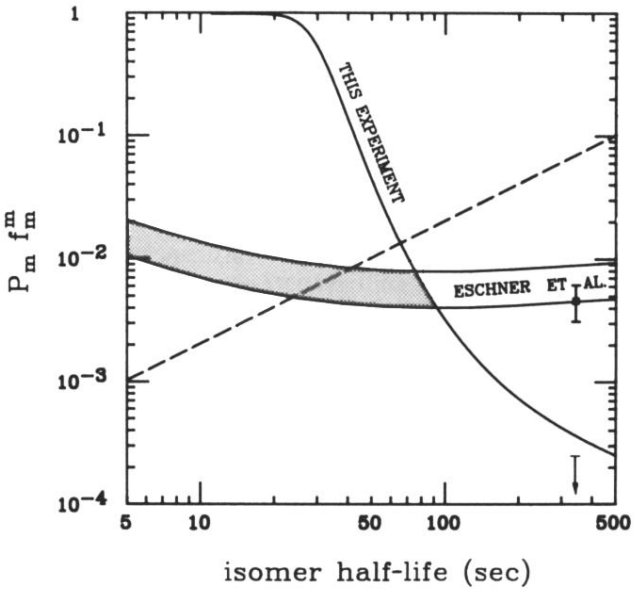


FIG. 3. Interpretation of $f_m$ in terms of a proposed high-spin isomer of $^{180}$Lu. The ordinate is the product of the $^{180}$Lu isomer's production factor, $P_m$, and its $\beta$-decay fraction that populates $^{180}$Hf$^m$, $f_m^m$. The result of this experiment (shown as the lower data point at 5.7 min) also establishes an upper limit of about 100 sec on the half-life of such an isomer. We have solved the equations of Ref. 15 including a source term for a $^{180}$Lu isomer to generate curves for the upper and lower limits implied by the positive value reported by Eschner *et al.* (upper data point). The shaded region is in agreement with both experimental measurements. The dashed line is described in the text.