Search for low-energy induced depletion of $^{178}\text{Hf}^{m2}$ at the SPring-8 synchrotron

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**A B S T R A C T**

Electromagnetic transitions within nuclei reflect specific aspects of nuclear structure. This is particularly true for metastable excited states, or isomers, like $^{178}\text{Hf}^{m2}$ ($T_{1/2} = 31$ years, excitation energy 2446 keV). The interaction of external radiation with isomers can be used to study atomic and nuclear properties and, perhaps, to induce a release of the stored energy. Some experiments indicated that low-energy photons near the $L_3$ edge (9.561 keV) of hafnium could cause this to occur for $^{178}\text{Hf}^{m2}$, but the lack of a viable physical model and null experiments by other groups have left these claims in doubt. The present work describes a new experiment to examine this process by closely duplicating the irradiation conditions in positive studies, but using a more advanced multi-detector $\gamma$ array. No support for an induced depletion of $^{178}\text{Hf}^{m2}$ by low-energy photons was obtained, with an upper limit for the integral cross section that is eight orders-of-magnitude below the reported value.

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1. Introduction

Long-lived nuclear isomers can store large amounts of energy for significant periods. The existence of these metastable excited states motivated the development of nuclear shell and collective rotational models [1], and particularly durable isomers may be used in practical applications [2]. Ultimately, the stability of isomers results from inhibitions against their decay [3]. For high-spin isomers like $^{180}\text{Ta}^{m}$, this is expressed in the multipole nature of possible decay transitions: decay of the $I_g^m = 9^{-}$ isomer to the $I_g^p = 1^+$ $^{180}\text{Ta}$ ground state would require the emission of magnetic 2$^+$-pole radiation, with a correspondingly small transition rate. In fact, decay of $^{180}\text{Ta}^{m}$, with $T_{1/2} > 7.1 \times 10^{15}$ years, has never been observed [4]. Just as electromagnetic transitions from isomers to lower-lying levels reflect specific aspects of nuclear structure, so also do transitions that feed isomers from higher-lying states [5].

Electromagnetic transitions may well hold the key to the realization of new applications for isomers. The core concept would then require the absorption by an isomeric nucleus of a real [6] or virtual photon (from electron shells [7] or Coulomb excitation [8]) to reach a higher-lying intermediate state (or depletion level). The intermediate state must then have a decay branch that leads to the ground state. This process would deplete some portion of the isomeric population, releasing stored excitation energy from the nucleus (more would be released if the ground state was short-lived). Induced isomer depletion was first demonstrated for $^{180}\text{Ta}^{m}$, with eight energies identified at which real photons (within bremsstrahlung) initiated the process [9]. Some of the depletion levels reached by photon absorption were subsequently correlated with specific nuclear states [10] as observed by in-beam $\gamma$ spectroscopy. Induced depletion of $^{68}\text{Cu}^{m}$ by Coulomb excitation has now been demonstrated [11], as well as that of $^{177}\text{Lu}^{m}$ and $^{178}\text{Hf}^{m2}$ via neutron scattering [12].

The $I_g^m = 1^+$ isomer $^{178}\text{Hf}^{m2}$, with $T_{1/2} = 31$ years, has been the subject of considerable study since its discovery in 1968 [13].
In addition to general interest in its high spin and yrast nature, it was recognized more than a decade ago [14] that this isomer was very attractive for induced depletion. Its high excitation energy of 2.446 MeV corresponds to more than a gigajoule/gram and systems developed for photoactivation of isomers in neighboring nuclides, plus the induced depletion of $^{180}\text{Ta}$, had hinted that depletion levels might exist within a few hundred keV of $^{178}\text{Hf}^{m2}$. Direct examination of this possibility was initiated in 1996 at CNRSM Orsay using in-beam $\gamma$ spectroscopy and particle scattering, and based on the suggestion in Ref. [15]. The results were apparently inconclusive and remain unpublished. Thereafter, efforts concentrated on the use of photons to investigate this process.

A first indication for “triggering” of this isomer via bremsstrahlung irradiation was reported in Ref. [16]. The lowest bremsstrahlung endpoint used in that experiment was 70 keV, indicating that the intermediate state must be within this energy above the isomer. However, no known nuclear level within this range has an angular momentum permitting an electromagnetic transition of $\Delta I < 4$. Also, the quoted integral cross section for isomer depletion, $10^{-21}$ cm$^2$/keV, was quickly recognized to be too large to be initiated by direct nuclear absorption of irradiating photons [17,18]. A subsequent experiment was performed in which monochromatic synchrotron radiation (SR) of high intensity was used to examine the claimed effect [19]. Evidence was presented there of induced depletion of $^{178}\text{Hf}^{m2}$ by incident photons having energies above, but close to 9.561 keV, the $L_3$ edge of hafnium. The integral cross section was again very large and the NEET process was invoked [19] as an explanation, with atomic photoionization by incident photons and excitation of the nucleus by a virtual photon initiating $\gamma$ cascade passing through the 4 s structure. The data were obtained during irradiations of much longer than 4 s duration to the incoming beam direction (see Fig. 1(a) and Fig. 8 of [25]) in order to irradiate as much of the hafnium deposit as possible. Calibrated ionization chambers were placed up-beam and down-beam from the sample to determine the flux before ($I_B$) and after ($I_C$) passage of incident photons through the sample. Fig. 1(b) shows $I_C$ in a raster spatial scan of the target through the fixed beam, with a 1 mm × 1 mm beam spot and an energy of 9.561 keV. The hafnium deposit is identified by its greater opacity to these photons within the gap in the Al frame as viewed by the beam. The indicated positions for larger beam spots of 0.65 mm (vertical) × 5 mm (horizontal) were defined for ON (deposit) and OFF (deposit) irradiations of the sample. Fig. 1(c) shows an energy scan giving $I_C/I_B$ for ON and OFF beam spots when the sample was held stationary and the beam energy was changed. The dip in ON transmission just above the $L_3$ edge is due to a “white line” resulting from electronic excitation to bound $d$ states in hafnium just below the continuum. The amount of hafnium irradiated with the ON position was determined from the shift in slope at the $L_3$ edge, including the small attenuation by the inclined Be disks. This amount was compared with a geometric calculation based on the transmitted intensity variations in Fig. 1(b). The average of the two methods gave the fraction of irradiated hafnium to be $f = 0.60 \pm 0.15$. The hafnium deposit was optically-thin, providing a maximum attenuation of monochromatic SR at about 10% above the $L_3$ edge.

The YSU miniball detector array [25] surrounding the sample was made of one 65% relative efficiency p-type HPGe detector and six 76.2 mm (diameter) × 63.5 mm (length) BGO scintillators. The center of the sample was 8.5 cm from the Ge detector and 4 cm from any BGO scintillator. The detectors possessed Al encapsulation of about 1 mm that strongly attenuated scattered SR and there was no evidence of pile up in the $\gamma$ spectra. Energy and time data were recorded from every detector, with the system trigger generated by the Ge detector. Reports claiming depletion near the $L_3$ edge utilized smaller volume Ge detectors to obtain singles (non-coincidence) data [16,19,23] or doubles spectra [26]. Those detectors were placed within about 2 cm of a sample, raising the possibility of coincidence summing effects. Timing data [23] motivated the suggestion that the induced emission was prompt, leading to emission at 213.2 keV without any delay from the cascade passing through the $4\ s\ m_1$ isomer (see Fig. 2), although earlier works [16] claimed the induced cascade did proceed through the $m_1$ isomer. For either case, the YSU miniball provided a high-sensitivity test since singles Ge spectra and higher-fold data structures could be extracted from the data. The data were obtained here during irradiations of much longer than 4 s duration so evidence of induced depletion should appear even for gamma rays delayed by a cascade through the $4\ s\ m_1$ isomer. Detection of a prompt effect would be even more evident in doubles or higher-fold data, with multiple gamma rays being emitted within the resolving time of the detector system [25]. Figs. 3(a), (b) show sections of the Ge singles spectrum obtained from an ON irradiation at 9.5676 keV, where the greatest effect was reported to occur [23]. Full singles spectra were obtained for all ON and OFF irradiations.
Fig. 1. (a) Depiction of the inclined target as seen from the beam direction. (b) Raster spatial scan showing measured transmission of SR through the sample. (c) Fixed-position energy scans showing measured transmission of SR as a function of energy through the hafnium deposit.

Fig. 2. Partial energy level diagram (see [27]) for $^{178}$Hf, showing natural decay transitions (energies in keV) from the $m_2$ isomer. The dotted transition is heavily converted. The gray band represents an intermediate state excited from the isomer and leading to either slow (S) cascades through the $m_1$ isomer or prompt (P) cascades bypassing the $m_1$ isomer.

A normalization procedure was utilized prior to quantitatively comparing $\gamma$ emissions during different irradiations. This was needed because (1) the irradiations were of slightly different lengths; (2) the discrimination level varied for the Ge signal, which initiated event recording, over the two-week experimental period; and (3) the efficiency of the Ge detector (and BGOs) varied slightly between ON and OFF sample positions. An intrinsic calibration was performed by normalizing the peak area of each $^{178}$Hf line to the peak area of a nearby line from $^{172}$Hf decay. The regular decay of $^{172}$Hf activity, $T_1/2 = 1.87$ years, was included in this normalization. Peak areas were determined using Fitzpeaks [28] to obtain Gaussian fits via a linear least squares algorithm. Backgrounds were examined manually to ensure consistent fitting of the same line in every spectrum (note that Regions of Interest to obtain gross or integral counts were apparently used in some experiments from which the effect was claimed [19,23]). Figs. 4(a)-(d) show peak areas for lines at 213.4 keV and 325.6 keV that appear in $^{178}$Hf decay, normalized to nearby lines from $^{172}$Hf decay. Emission of these $^{178}$Hf$^{m_2}$ gamma rays was reported (both lines in [23], 213 keV in [19]) to be enhanced when samples were irradiated with photons having energies greater than the L3 edge at 9.561 keV. Figs. 4(e)-(f) show normalized peak areas for 495 keV, which was reported to enhance under bremsstrahlung irradiation [16]. The error bars in Figs. 4(a)-(f) show statistical error; systematic errors in the fits are estimated to only contribute up to 1% additional error. Gamma emission for the peaks of Figs. 4(a)-(f) showed no enhancement for irradiations with the sample in either ON or OFF positions. Statistical $t$ tests were performed to determine if significant differences existed for individual normalized peak areas within each ON or OFF set for irradiations below...
the L₃ edge compared with those obtained above the L₂ edge (the no-effect and effect regions seen in Fig. 6 of [23], respectively). No statistically significant differences in the means were found in the present work at the 95% confidence level. In addition, t tests were performed for other lines occurring in the spontaneous decay of $^{178}$Hf$^{m2}$, including at 426.4 keV, which was also reported [23] to enhance under irradiation above the L₃ edge, and at 216.7 keV. No statistically significant enhancements were seen for lines from the natural decay of $^{178}$Hf$^{m2}$ and no new lines were detected.

The integral cross section for an induced depletion may be found by examining the amount of any excess γ emission compared to the amount of natural emission by

$$ICS = \Delta C / \left[ \int \tau C_{nat}(d\Phi / dE) \right]$$

where $C_{nat}$ is the normalized area of a $^{178}$Hf$^{m2}$ γ-ray peak resulting from irradiations in the reference region below the L₃ edge. The ΔC is the excess number of counts in the same normalized γ-ray peak resulting from irradiations in the region above the L₃ edge. The fraction of hafnium irradiated may contribute to an induced effect, while all the hafnium contributes to the natural decay, and $\tau$ is the natural-decay lifetime of $^{178}$Hf$^{m2}$. The depletion effect was claimed to occur over a range of incident energies greater than the L₃ edge, with the largest excess emission being near 9.5676 keV [23, Fig. 6], so mean values in the ranges above and below the L₃ edge were used instead of values from individual irradiations. In the absence of any statistically-significant difference between the means, the standard deviation of the difference in means was substituted for ΔC. The most restrictive upper limit in singles spectra from ON irradiations was 6.58 $\times$ 10⁻²⁷ cm² keV, obtained from the 213.4 keV line (due to its large area and that of the 181 keV peak to which it was normalized). This may be compared with the upper limit of 3 $\times$ 10⁻²⁷ cm² keV from the full measurement of [21] and the claimed magnitude of the effect on the order of 10⁻₂² cm² keV [19,23].

The possibility of prompt induced γ emission was further investigated by extracting multi-fold coincidence spectra from the miniball data, obtained during the same irradiations. Fig. 3(c) shows a spectrum obtained during an ON irradiation at 9.5676 keV. The detected BGO fold $F = 2$, so this data corresponds to γ triples; the summed γ energy detected by any two BGO scintillators is used for one axis [25], resulting in the bi-dimensional display in the figure. Various three-fold coincidences are possible in the natural decay of $^{178}$Hf$^{m2}$, giving peaks on loci of constant total energy as shown in the figure. Three-fold coincidences from $^{172}$Yb gamma rays, following $^{172}$Hf decay, are also seen with lesser intensity due to the reduced probabilities for those cascades and reduced detector efficiencies at higher γ energies. Prompt induced γ emission, in cascades bypassing the 4 s $^{178}$Hf$^{m1}$ isomer, would correspond to a high physical multiplicity, and thus a high detected fold and a high summed energy. Full energy peaks from such events would lie in the indicated region in Fig. 3(c). No ON or OFF data extracted for any coincidence fold showed peaks within the prompt sum energy region.

A quantitative analysis considered the ratio between counts in high- and low-fold coincidence data. Calculations indicate [25, Fig. 17] that a significant part of all events should correspond to $F > 2$, even up to $F = 5$, when the physical multiplicity is 7 or 8 as expected for prompt induced cascades. Natural-decay events with a physical multiplicity of 3 would have the highest probability for detection with $F = 1$, with no events producing $F > 2$. A comparison between the total counts for high detected fold, $F > 2$, and the total counts for low detected fold, $F < 2$, would then represent the yield of prompt vs. natural decay events.

Data subsets for all $F$ were extracted for each ON irradiation and the total number of counts was determined whose sum energies were less than 2530 keV. The counts so determined, $C(F)$, contained only events resulting from either natural or induced decay and not background at higher summed energy. These counts,
from full-energy peaks or Compton continua, are related to the $\gamma$ cascade and the detector system by

$$C(F) = YAT \epsilon_G e_{\text{BGO}}^F g_1,$$

(2)

where $A$ is taken as the natural $^{178}$Hf$^{m2}$ activity for low-fold events or $f$ times the natural activity for high-fold (prompt depletion) events, and $T$ is the data acquisition time. The combinatorial factor is given by

$$g_1 = (M_p!/(M_p - F - 1)!)(n!/(n - F)!),$$

(3)

where $n = 6$ is the number of BGO detectors in the array and $\epsilon_G$ and $e_{\text{BGO}}$ are the total (not peak) efficiencies for the individual Ge and BGO detectors, respectively. The $Y$ represents the yield of a given cascade of physical multiplicity $M_p$. For natural decay of $^{178}$Hf$^{m2}$, two separate cascades with $M_p \sim 3$ exist, one above and one below the 4 $s$ m1 isomer, so that $Y = 2$. For prompt induced depletion, $M_p \sim 7$ and the corresponding yield would then be that of the searched-for induced effect, with

$$Y_{\text{ind}} = \text{ICS}(d\Phi/dE).$$

(4)

Figs. 4(g), (h) show the ratio $C(5)/C(1)$, where $C(1)$ is used since that BGO fold has the highest probability for detection of natural-decay events. A statistical t test indicated no significant difference between means for this ratio below and above the L3 edge of hafnium. In the absence of any meaningful difference between the means, one standard deviation of the difference was again taken as a measure of the maximum induced effect. This gave an upper limit of $4.39 \times 10^{−30}$ cm$^2$ keV for induced depletion of $^{178}$Hf$^{m2}$ by photons above the L3 edge. Nor do data from folds of $F = 3$ or 4 show statistically-significant differences between means below and within the claimed effect region, with less restrictive upper limits on the ICS.

Data were examined for coincidences between the 213 keV gamma rays from $^{178}$Hf$^{m2}$ decay and the region near 129 keV. No peak was in evidence near 129 keV in any spectrum, but background in that region did occur in coincidence with gamma rays at 213 keV, 325 keV and 426 keV from the $^{178}$Hf ground state band. This is expected in natural decay, since Compton events from 325 keV and 426 keV gamma rays would contribute to the background near 129 keV, and 213 keV is in coincidence with 325 keV and 426 keV emissions as seen in Fig. 2.

3. Discussion

It is useful to compare the upper limits measured here with estimates based on established physical models of electromagnetic transitions. Nuclei of $^{178}$Hf exhibit an axially-symmetric deformed shape, so that rotational excitations occur in addition to intrinsic (shell model) states, as seen in Fig. 2. For deformed nuclei, the quantum number $K$ is defined to describe the projection of the angular momentum vector on the symmetry axis [3]. While a strict selection rule based on $K$ does not exist for electromagnetic transitions, large changes of $K$ can suppress transition rates. This is expressed by the $K$ hindrance, $F_K = (f_{\text{red}})^K$, where $f_{\text{red}}$ is the empirically measured reduced hindrance per degree of $K$ hindrance, $\nu = \Delta K - \Delta l$. The m2 isomer is an intrinsic state, with $F_K = 16^\nu$, $K_m = 16$, and its dominant decay transition, to the $I^π = 13^−$, $K = 8$, level at 2443.3 keV (in the m1 band, see Fig. 2), has been found to have $F_K = (66)^5$ [29].

Direct nuclear photoabsorption to initiate depletion would require an intermediate state located near (2446 + 9.57) keV and that should be reached by a low-multipolarity transition to suppress the magnitude of the claimed effect. Transitions with $E1$, $M1$ or $E2$ are considered here, so that a proposed intermediate state would have $I^π = 15^−$, 15$^+$, 14$^+$, or $I^π = 17^−$, 17$^+$, 18$^+$. No levels are known [5,27] within 27 keV of the required excitation energy, so the intermediate state should be a previously undiscovered intrinsic state, with $l_i = K_i$. A transition from the isomer to this intermediate state would be subject to no $K$ hindrance, since $\nu = 0$.

An intermediate state must also have a decay branch via a low-multipolarity transition to another level besides the $m2$ isomer. Again, $E1$, $M1$ and $E2$ are considered, as well as the availability of $\nu = 0$ below (2446 + 9.57) keV with suitable angular momenta. The same $I^π = 13^−$ level at 2443.3 keV is the most likely candidate for decay of the proposed intermediate state, with its spin now restricted to $I^π = 15^−$ or $14^+$. In both cases, a transition to the $13^−$ state will be five-fold $K$ forbidden, leading to a strong $K$ hindrance. It is assumed that the reduced hindrance for this transition is similar in magnitude to that measured in the natural decay of the $m2$ isomer, $f_{\text{red}} = 66$.

Integral cross sections were estimated for proposed intermediate states according to

$$\text{ICS} = 2\pi^2 \chi^2 \frac{g_{\gamma}}{\Gamma_{\gamma}} \frac{m_{I-m}}{m_{I}},$$

(5)

where $\chi = \hbar c/E_\gamma$, with $E_\gamma$ being the transition energy between isomer and intermediate state, and $m_{I-m}$ being the corresponding $\gamma$ width. The total width of the intermediate state to any lower level is $\Gamma = \Gamma_{\gamma} + \Gamma_{\gamma}^{\text{I-I}}$, and its full width to the $13^−$ state which then cascades to the ground state is $\Gamma_{\gamma}$. The standard spin factor for excitation of the intermediate state from the isomer is $g_{\gamma} = (2l_i + 1)/(2l_{\text{m1}} + 1)$.

Weisskopf single-particle estimates [30] and $K$ hindrances, as discussed above, were used to obtain partial $\gamma$ widths for transitions from a proposed intermediate state to the isomer or to the $I^π = 13^−$ level. The total widths were then determined from the respective partial $\gamma$ widths by $\Gamma = \Gamma_{\gamma}^- (1 + \alpha)$ where $\alpha$ is the conversion coefficient for the transition. Values of $\alpha$ for the transitions were obtained from Ref. [31]. The resulting estimates for different intermediate state spin are given in Table 1. In the complete absence of $K$ hindrance, the values of the estimated cross sections in Table 1 increase by a factor of $(66)^5$, but reach no more than $8.78 \times 10^{−29}$ even in this unlikely case. With or without $K$ hindrance, the integral cross sections for proposed intermediate state properties are many orders-of-magnitude less than those reported in Refs. [16,19,23], but are in agreement with the upper limits established by the present experiment.

Detailed analyses were performed elsewhere [32] to consider the possibility of the claimed effect being due to NEET, initiated with photoionization by a real photon. Similar assumptions were made regarding proposed nuclear intermediate states and the interaction between the nucleus and the atomic shells was calculated. The integral cross sections were found to be less than $10^{−31}$ cm$^2$ keV$^2$. Again, this is in agreement with the present results. The “resonance conversion” process described in Ref. [33], while interesting, also fails to support claims of induced depletion of $^{178}$Hf$^{m2}$.

| $I^π$ | Absorption multipolarity | Decay multipolarity | ICS [cm$^2$ keV] |
|-------|--------------------------|---------------------|------------------|
| 15$^−$ | E1                       | E2                  | $4.81 \times 10^{−39}$ |
| 14$^+$ | E2                       | E1                  | $7.01 \times 10^{−38}$ |

Table 1 Estimated integral cross section for depletion of $^{178}$Hf$^{m2}$ by real photons, via assumed intermediate states.
4. Conclusions

A search for induced depletion of $^{178}\text{Hf}^{m2}$ has been conducted at the SPring-8 synchrotron, using monochromatic photons near the $L_3$ edge of hafnium. A multi-detector array was used to obtain both singles and higher-fold coincidence data to provide improved measurements of $\gamma$ radiation emitted from an isomeric sample under irradiation. The data show no evidence in support of the claimed effect and upper limits were established that agree with theoretical estimates. It appears that there is no theoretical or experiment basis for claims of low-energy depletion of the $^{178}\text{Hf}^{m2}$ isomer. Notwithstanding this null result, further examinations of the interaction of externally-produced photons, or other radiation, with isomers may indeed lead to the identification of interesting physics.

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