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γ rays emitted in the decay of 31-yr ^{178}Hf^{m2}

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The spontaneous decay of the $K^{\pi}=16^{+}$, 31-yr ^{178}Hf^{m2} isomer has been investigated with a 15-kBq source placed at the center of a 20-element γ-ray spectrometer. High-multipolarity $M4$ and $E5$ transitions, which represent the first definitive observation of direct γ-ray emission from the isomer, have been identified, together with other low-intensity transitions. Branching ratios for these other transitions have elucidated the spin dependence of the mixing between the two known $K^{\pi}=8^{-}$ bands. The $M4$ and $E5$ γ-ray decays are the first strongly K-forbidden transitions to be identified with such high multipolarities, and demonstrate a consistent extension of K-hindrance systematics, with an inhibition factor of approximately 100 per degree of K forbiddenness. Some unplaced transitions are also reported.

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One of the most remarkable isomers known in the $A \approx 180$ region of deformed atomic nuclei [1] is the second metastable state of ^{178}Hf, commonly referred to as ^{178}Hf^{m2}. This state, which lies 2.4 MeV above the ground state of ^{178}Hf, has angular momentum and parity $I^{\pi}=K^{\pi}=16^{+}$ (K is the angular-momentum projection on the body-fixed symmetry axis) and a half-life of 31 yr [2–5]. Its four-quasiparticle structure is based on a broken neutron (ν) pair and a broken proton (π) pair, each contributing $I^{\pi}=K^{\pi}=8^{-}$. The exceptionally long lifetime of the isomer arises not only because of its high K value and the associated hindrance caused by the K-selection rule, but also because it lies lower in excitation energy than any other states of spin 14 or higher. This yrast trap is forced, therefore, to decay by transitions with both high multipolarity and a large change in K.

On account of its long half-life and high excitation energy, the ^{178}Hf^{m2} isomer has attracted considerable interest and experimental investigation. For example, enriched samples have been studied by laser hyperfine spectroscopy [6], and there is evidence that the isomer can be Coulomb excited in a multistep process from the $I^{\pi}=K^{\pi}=0^{+}$ ground state [7]. A more controversial observation is the stimulated decay of the isomer, induced by x rays and synchrotron radiation, which has been reported by Collins *et al.* [8–11], but refuted by Ahmad *et al.* [12,13].

The high-spin level structure of ^{178}Hf has been studied extensively [3–5,14], but basic knowledge gaps remain. Surprisingly, radiations emitted directly from the isomer itself are not yet well established. The problem arises, in essence, because 99.9% of the isomer decay proceeds through a highly converted, 13-keV $E3$ transition ($16^{+}\rightarrow 13^{-}$) which has not so far been directly detected. The suggestion by Khoo and Løvhøiden [4] that the isomer is at 2447.4 keV, deduced from its population from higher-energy states, is in conflict with the singles conversion-electron data of van Klinken *et al.* [5] for a 0.1%, $M4$ decay branch ($16^{+}\rightarrow 12^{-}$) which implies an excitation energy of 2446.0 keV. Although the 1.4-keV difference in proposed isomer energies is small, it is well outside statistical uncertainties, and represents a basic difficulty with the current understanding of the isomer and its decay modes.

In the present γ-γ-coincidence study, the $M4$ γ-ray transition has been clearly identified, confirming the interpretation of van Klinken *et al.* [5]. We also establish the competing $E5$ γ-ray transition ($16^{+}\rightarrow 11^{-}$) and additional low-intensity transitions that had not previously been placed in the ^{178}Hf^{m2} decay sequence. The results extend the general understanding of high-multipolarity, K-forbidden decays, and shed new light on the nature of the band mixing between members of the two $K^{\pi}=8^{-}$ bands, which are populated in the isomer decay. Although the latter states are well known from in-beam γ-ray spectroscopic studies [4,14], significant additional band mixing information is now obtained.

The radioactive source material was extracted from a tantalum target, irradiated in 1980 with an intense proton beam at Los Alamos National Laboratory. Hafnium isotopes were chemically separated [15] and shipped to SRS Technologies, Alabama, where a 15-kBq source of ^{178}Hf^{m2} ($T_{1/2}=31$ yr) was prepared. On delivery for the present measurement, the source also contained ^{172}Hf ($T_{1/2}=1.9$ yr, in secular equilibrium with its daughter ^{172}Lu, $T_{1/2}=6.7$ days) with a decay rate approximately equal to that of ^{178}Hf^{m2}. The source was placed at the center of the 8π spectrometer [16], an array of 20 Compton-suppressed n-type HPGe detectors situated at TRIUMF-ISAC. The absolute full-energy-peak efficiency was 4% at 426 keV, and at that energy the final full width at half maximum energy resolution was 1.6 keV, after gain

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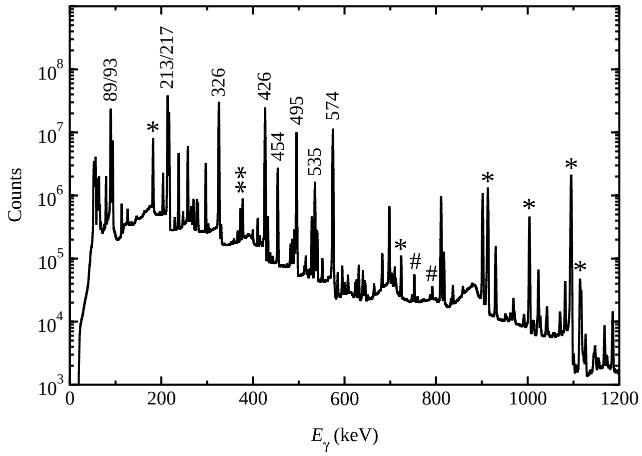

FIG. 1. Total γ-γ projection, from the data acquired with copper absorbers. The energy dispersion is 0.33 keV/channel. The most intense transitions in ^{178}Hf are labeled with their energy in keV. Sum peaks (#) and some of the most intense transitions from the ϵ decay of ^{172}Lu (*) are also indicated.

matching all the germanium detectors. The icosahedral geometry of the 8π spectrometer leads to minimal angular correlation effects [17].

When searching for extremely weak decay branches, many instrumental effects have to be taken into account, such as germanium x-ray escape events, peak summing, and scattering between detectors on opposites sides of the detector array. As part of this investigation, copper and lead absorbers, of thickness 0.5 mm and 1.0 mm, respectively, were placed (separately) in front of the germanium detectors for portions of the data taking. Data were acquired over a total period of 42 days as follows: 8 days with no absorbers, five days with the lead absorbers, and 29 days with the copper absorbers. Event pairs were sorted into γ-γ coincidence matrices, with a $\leqslant$20-ns time-difference requirement. Other timing conditions were also investigated. Using the RADWARE [18] software package, most of the analysis was performed with the data from the 29-day measurement. The corresponding total coincidence spectrum is shown in Fig. 1. The data taken with the lead absorbers, and without absorbers, were used independently to verify some of the results, and to obtain optimum data where low-energy peak summing otherwise limited the sensitivity. A major part of the data analysis was concerned with the identification of previously unreported γ-ray transitions in the complex decay scheme following the electron-capture (ϵ) decay of ^{172}Lu to ^{172}Yb [19]. More than 100 transitions have been newly placed in this decay (most being completely new to ^{172}Yb) and the details of these will be the subject of a separate report.

The emphasis of the present work was on the identification and characterization (in terms of energy and intensity) of low-intensity γ-ray transitions. Given the associated inherent limitations in counting statistics, the energy and efficiency calibrations were not themselves needed to very high accuracy. These calibrations were obtained internally from the well-known, intense decay transitions of ^{178}Hf^{m2} [5,20] and ^{172}Lu [19] giving, in the present work, energies and relative efficiencies to accuracies of ± 0.1 keV and 1.7%, respectively.

The sensitivity limit of the present measurement is at about the 0.001% level, i.e., 1 in 10^5 parent decays of ^{178}Hf^{m2} or ^{172}Lu. This is not only a matter of counting statistics. Of the various instrumental effects, the scattering between germanium γ-ray detectors, across the 8π spectrometer, is perhaps the most serious limitation. The use of a mass-separated sample of ^{178}Hf^{m2} could further improve the sensitivity. Within the obtained sensitivity limit, exotic α and β decays of ^{178}Hf^{m2}, whose possibility was discussed by van Klinken *et al.* [5], could not be detected, nor could any direct γ-ray decay into the ground-state band be identified.

After exhaustive investigation of the γ-ray coincidence relationships, only one additional source of activity was positively identified, close to the limit of sensitivity of the measurement. This is a product of the natural ^{232}Th decay series, namely the decay of ^{208}Tl to ^{208}Pb, with (583−2615)-keV coincidences [21]. In addition to this, also close to the limit of sensitivity, the following γ-ray energies were found to be in mutual coincidence: 154.4, 212.6, and 958.6 keV; further, 90.7- and 868.0-keV transitions were in coincidence with each other and with the 154.4- and 212.6-keV transitions, i.e., they appear to be in parallel with the 958.6-keV transition. While a 90.6-keV transition is well known in the ϵ decay of ^{172}Lu [19], the other four energies are clearly distinct from transition energies that can reasonably be associated with the decay of ^{178}Hf^{m2} and ^{172}Hf in the present source, though the 212.6-keV transition is close in energy to the intense 213.4-keV transition in ^{178}Hf. The origin of these five transitions is, at present, undetermined. However, their apparently long-lived ($\geqslant$1 year) parentage, and association with hafnium chemical extraction, makes their origin of great potential interest.

The partial level scheme of ^{178}Hf deduced from the present work, showing only those states involved in the decay of the $K^\pi = 16^+$ isomer (together with the $K^\pi = 8_2^-$ bandhead), is shown in Fig. 2. Transition energies and relative intensities, normalized to the 326-keV transition, are summarized in Table I. The ^{178}Hf^{m2} isomer was previously known to decay predominantly by an $E3$ conversion-electron transition of energy close to 13 keV [3–5], and evidence for a $M4$, 309.5-keV transition, from detection of conversion electrons in singles mode, was also reported [5]. In the present work, the clear observation of γ rays from the direct deexcitation of the isomer is reported for the first time, confirming the 309.5-keV, $M4$, $16^+ \rightarrow 12^-$ transition and adding the 587.0-keV, $E5$, $16^+ \rightarrow 11^-$ transition. These are illustrated in Fig. 3. In addition, the low-intensity 231-, 343- and 601-keV γ-ray transitions, which link members of the two $K^\pi = 8^-$ bands, have been observed in the decay path of ^{178}Hf^{m2}. These are illustrated in Fig. 4. The latter two transitions are known from in-beam studies [4,14], whereas the 231-keV $E2$ transition was previously reported [22] based on singles counting with a ^{178}Hf^{m2} source. These transitions have now been definitively established in the decay of the 31-yr isomer, from γ-γ coincidences.

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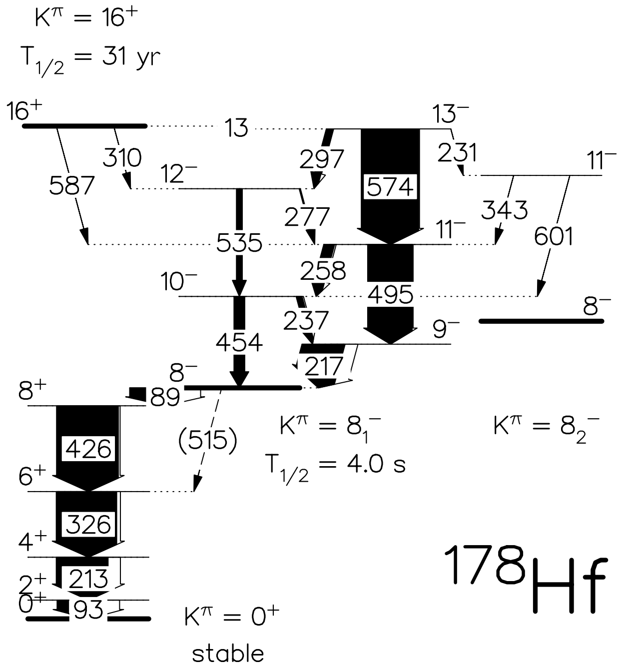

FIG. 2. Decay scheme of ^{178}Hf^{m2} deduced from the present work. The arrow widths represent the relative transition intensities, the unfilled parts corresponding to internal conversion. The dashed 515-keV, $8^- \rightarrow 6^+$ transition has not been established. The highly converted 13-keV transition is inferred from γ-ray coincidence relationships. Only the 11^- member of the 8_2^- band is identified in the present work, though other band members are known from previous studies [4,14].

The expected 515-keV, $8^- \rightarrow 6^+$ transition from the 4-s isomer could not be established in this work. However, with the lead absorbers in front of the germanium detectors, the sum peak from 89- and 426-keV coincidences was highly attenuated, enabling a stringent limit to be placed on the 515-keV γ-ray intensity (see Table I).

Part of the analysis involved searching for the 129.4-, 210.3-, and 546.2-keV transitions, which Collins *et al.* [9,10] reported as being associated with the stimulated decay of ^{178}Hf^{m2}, in the presence of a similar proportion of ^{172}Hf. In the present work, these transitions could not be found in the corresponding spontaneous decay. However, transitions at the nearby energies of 130.4 and 544.8 keV have been newly identified as being part of the ϵ decay of ^{172}Lu. It is not apparent how these might have influenced the earlier interpretation [9,10].

It is well known in ^{178}Hf that two $K^{\pi}=8^-$ configurations, $\pi\{7/2[404],9/2[514]\}$, and $\nu\{7/2[514],9/2[624]\}$, mix together. (These are also the four quasiparticles that account for the structure of the $K^{\pi}=16^+$ isomer.) Earlier work [3] established that the 8_1^-, 4-s isomer consists of 37(2)% π configuration and 63(2)% ν configuration. Also [3,5] there is evidence that the mixing matrix element V decreases with spin. Emery *et al.* [23] have shown that the conversion coefficients and branching ratios within the 8_1^- band can be understood with the specific spin dependence $V_I = V_{I=8}\exp\{-b[I(I+1)-K(K+1)]\}$, with $V_{I=8}=159.2$ keV, $b=0.001\,79$, and $K=8$.

TABLE I. γ-ray energy, relative γ-ray intensity, initial and final spin and parity, multipole order λ, and reduced hindrance f_ν, for transitions in the decay path of ^{178}Hf^{m2}. Uncertainties are given in parentheses.

E_γ (keV)	I_γ (%)	$I_i^\pi \rightarrow I_f^\pi$	λ	f_ν
12.7		$16^+ \rightarrow 13^-$	$E3$	66(1)
88.8(1)	68.7(12)	$8^- \rightarrow 8^+$	$E1$	79(1)
93.2(1)	19.0(3)	$2^+ \rightarrow 0^+$		
213.4(1)	85.7(15)	$4^+ \rightarrow 2^+$		
216.7(1)	69.5(14)	$9^- \rightarrow 8^-$		
230.8(1)	0.0060(10)	$13^- \rightarrow 11^-$		
237.4(1)	9.6(2)	$10^- \rightarrow 9^-$		
257.6(1)	17.5(4)	$11^- \rightarrow 10^-$		
277.4(1)	1.8(1)	$12^- \rightarrow 11^-$		
296.8(1)	10.9(2)	$13^- \rightarrow 12^-$		
309.5(1)	0.015(1)	$16^+ \rightarrow 12^-$	$M4$	72(2)
325.5(1)	100.0(17)	$6^+ \rightarrow 4^+$		
343.3(1)	0.0018(3)	$11^- \rightarrow 11^-$		
426.3(1)	102.2(18)	$8^+ \rightarrow 6^+$		
454.0(1)	17.4(4)	$10^- \rightarrow 8^-$		
495.0(1)	73.6(14)	$11^- \rightarrow 9^-$		
515.1	<0.0008	$8^- \rightarrow 6^+$	$M2$	>160
535.1(1)	9.5(2)	$12^- \rightarrow 10^-$		
574.3(1)	94.9(18)	$13^- \rightarrow 11^-$		
587.0(1)	0.0062(5)	$16^+ \rightarrow 11^-$	$E5$	165(5)
601.1(1)	0.0026(3)	$11^- \rightarrow 10^-$		

It is now possible from the observed intensity of the 231-keV, $13^- \rightarrow 11^-$, $E2$ transition, relative to the 574-keV, $E2$ transition, to obtain an independent estimate of the mixing strength between the two 8^- structures. The determination of the relevant γ-ray intensities from the present coincidence data requires knowledge of other decay branches from members of the 8_2^- band, and for this purpose the intensities of Mullins *et al.* [14,24] have been used. (The intraband transitions account for about 10% of the decay intensity from the $I=11$ member of the 8_2^- band.) It is notable that the 231-keV transition is the only stretched $E2$ transition established here between the two 8^- bands. While, in principle, the known $I \rightarrow I-1$ and $I \rightarrow I$ interband transitions can also be used to quantify the band mixing, the additional complication of $M1/E2$ admixtures, and the consequent need to quantify the magnetic and electric moments, limits the utility of that approach. Therefore, the identification of an interband $I \rightarrow I-2$, $E2$ transition can be considered to be especially valuable.

Applying standard two-band-mixing formulas (see, for example, Ref. [25]) together with the specified [23] spin-dependent mixing matrix element, the predicted $13^- \rightarrow 11^-$, $B(E2)$ ratio is 0.0054 compared with the present experimental value of 0.0057(9). The excellent agreement provides strong support for the interpretation of Emery *et al.* [23]. The other predicted $B(E2)$ ratios and the corresponding experimental limits are given in Table II. Also in the table are the $B(E2)$ ratios obtained by Karamian *et al.* [22] from singles γ-ray intensities, with evident disagreement for the $B(E2)$

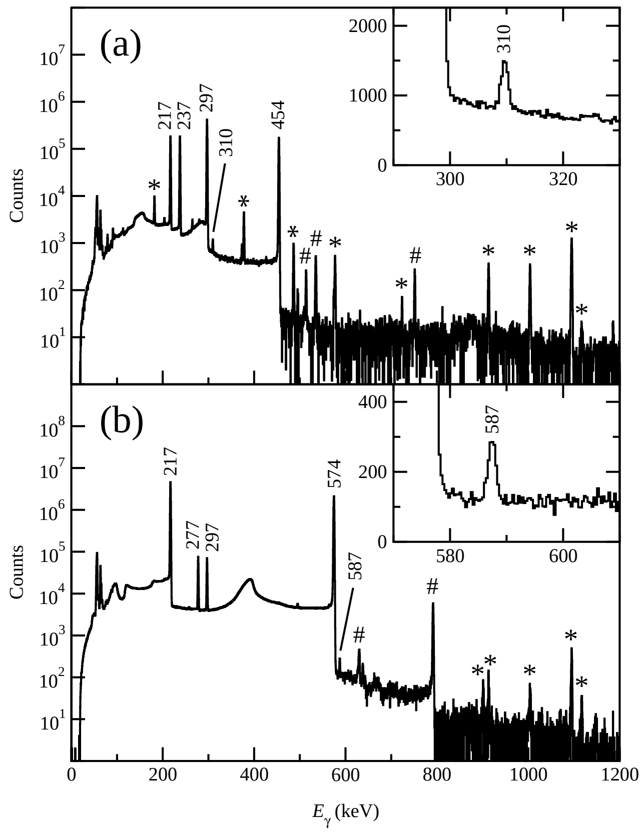

FIG. 3. Background-subtracted γ-ray coincidence spectra produced by gating on (a) the 535-keV transition and (b) the 495-keV transition. Transitions in ^{178}Hf are labeled with their energy in keV. Sum peaks (#) and contaminants from the ϵ decay of ^{172}Lu (*) are also indicated. The insets show the direct decays from ^{178}Hf^{m2}, on a linear scale.

ratios from the $I=10$ and 11 levels. This disagreement presumably results from the poor peak-to-background ratio in singles counting.

The $M4$ and $E5$ transitions identified in the decay of ^{178}Hf^{m2} are the only known examples of such high-multipolarity transitions that are strongly K forbidden. The transitions go from the $K=16$ isomer to members of a $K=8$ band ($\Delta K=8$) and the degree of forbiddenness $\Delta K-\lambda$ is $\nu=4$ and 3, for the $M4$ and $E5$ transitions, respectively, where λ is the multipole order. If K were a strictly conserved quantity, then the transitions would be forbidden by angular-momentum vector coupling rules. However, K mixing leads to transitions that are hindered, rather than forbidden. A measure of the goodness of the K quantum number is the hindrance per degree of K forbiddenness (or reduced hindrance). This can be defined as $f_\nu=(F_W)^{1/\nu}$, where $F_W=T^{\gamma}_{1/2}/T^{W}_{1/2}$ is the hindrance factor, $T^{\gamma}_{1/2}$ is the partial γ-ray half-life, and $T^{W}_{1/2}$ is the corresponding Weisskopf single-particle estimate [21].

In a general analysis of K-forbidden transitions, Löbner [26] concluded that for each degree of K forbiddenness, transitions are typically retarded by a factor of 100, i.e., $f_\nu\approx100$. In the present work, these systematics can now be extended to $\lambda=5$, with consistent results. The numerical values for ^{178}Hf, including the decay of the $K^\pi=8^-$ isomer, are given in Table I. For the 12.7-keV $E3$ transition, $f_\nu=66$ was evaluated using a conversion coefficient $\alpha=1.4\times10^7$ [27]. The $M2$, $8^-\rightarrow6^+$ decay from the $K^\pi=8^-$ isomer has $f_\nu>160$. The partial half-life of $>5\times10^5$ s for this 515-keV, γ-ray transition is greater than for either of the corresponding transitions in ^{180}Hf (501 keV, 1.4×10^5 s) or ^{182}Hf (507 keV, 1.7×10^4 s). Although the $M2/E3$ admixture is unknown for the ^{178}Hf case, the high reduced-hindrance limit is remarkable.

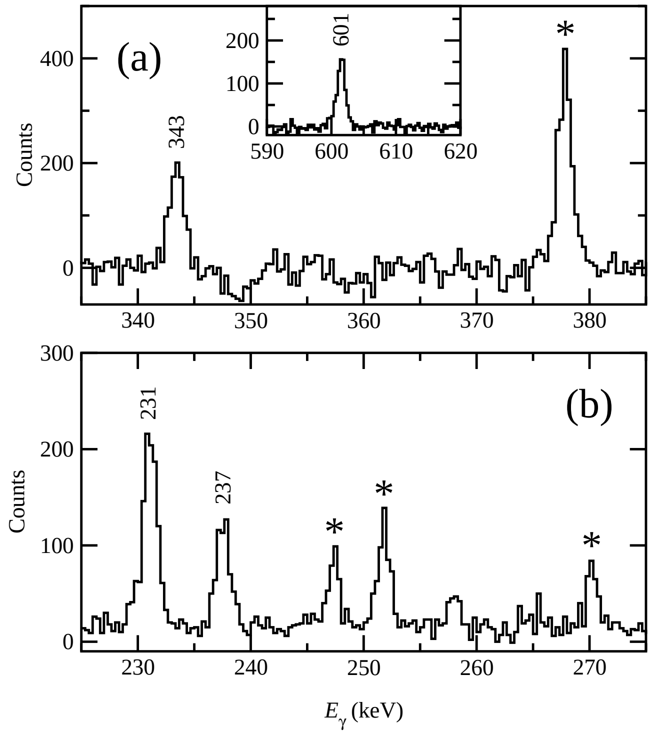

FIG. 4. Background-subtracted γ-ray coincidence spectra produced by gating on (a) the 231-keV transition and (b) the 601-keV transition. The inset shows a higher-energy portion of the spectrum gated on the 231-keV transition. Peaks associated with ^{178}Hf are labeled with their energy in keV, and impurities from the ϵ decay of ^{172}Lu (*) are indicated.

It is also striking that the Weisskopf hindrance factors F_W themselves vary over more than six orders of magnitude for the ^{178}Hf isomeric decays (discounting the $M2$ limit), while the reduced-hindrance values span the relatively narrow

TABLE II. Initial angular momentum, spin-dependent mixing matrix element, and ratio of out-of-band to in-band $E2$ strengths for $I\rightarrow I-2$ decays from $K^\pi=8^-_1$ band members in ^{178}Hf.

I	V (keV)	$R[B(E2)]$ Calc.	Expt.	Ref. [22]
10	148.73	0.0111	<0.02	0.24(9)
11	142.99	0.0096	<0.02	0.21(2)
12	136.98	0.0076	<0.02	<0.034
13	130.75	0.0054	0.0057(9)	0.0049(8)

range of $f_\nu = 115 \pm 50$. These high reduced-hindrance values for the $^{178}Hf^{m2}$ decay transitions contrast with transitions from some other four-quasiparticle isomers in the same mass region [28], such as the $f_\nu = 2$ value for the $E2$ decay of a $K^\pi = 16^+$, 6-ns isomer in ^{182}Os [29]. For the purpose of building an understanding of K-mixing processes, it is important to be able to establish any hindrance-factor consistency, such as for given isomer decays with competing branches. The five $E2$ branches having $f_\nu \approx 5$, identified in the decay of the $K^\pi = 14^-$, 4-μs isomer in ^{174}Hf [30], provide another example. Thus, the picture emerges that isomers in a given nuclide may have reasonably well-defined decay patterns, but apparently similar isomers in different nuclides can have widely different reduced hindrances. This needs to be the focus of models to describe K-forbidden transition rates.

To summarize, the decay of $^{178}Hf^{m2}$ has been studied by coincidence γ-ray spectroscopy, with a transition sensitivity limit of about ten parts per million. New information on the structure of two interacting $K^\pi = 8^-$ rotational bands has been interpreted using a spin-dependent matrix element. The reduced-hindrance values for decays directly from $^{178}Hf^{m2}$ show good consistency, with $f_\nu \approx 100$.

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