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Evolution of E2 strength of the rare earth isotopes ^{174,176,178,180}Hf

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Mean lifetimes of yrast states of the isotopes ^{174,176,178,180}Hf have been measured using fastelectronic scintillation timing. Excited states of 174,176,178 Hf were populated via β decay, while ¹⁸⁰Hf was populated via Coulomb excitation. The lifetimes of the 2^+_1 and 4^+_1 states of all isotopes and the lifetimes of the 6_1^+ states of 174,178 Hf were measured, using the slope and the centroid shift methods. The mean lifetime, $\tau(4_1^+) = 85(13)$ ps, of 178 Hf has been determined for the first time. In addition, the mean lifetimes of the 2_1^- and the 3_1^- states of 176 Hf have been determined. Systematic uncertainties on the evolution of data as a function of neutron number were reduced by using the same setup for all the isotopes of interest. The data are in agreement with other recent lifetime measurements where available and shows a shift of the maximum of collectivity for the Hf isotopic chain from neutron mid-shell at N=104 to N=100.

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INTRODUCTION I.

An important observable regarding the shape of eveneven nuclei is the E2 transition strength from the ground state to the first 2^+ state, which is a measure of nuclear quadrupole collectivity. Along an isotopic chain a gradual evolution of the $B(E2; 2_1^+ \rightarrow 0_{gs}^+)$ is expected. Well-deformed nuclei, e.g., nuclei in the rare-earth region around the mass number $A \approx 170$, with a large quadrupole deformation, i.e. β deformation values of about 0.2 - 0.4 [1–14], typically exhibit large B(E2) values in the order of 100 W.u. or more, while close to magic numbers single-particle excitations predominate and the B(E2) strength is at a minimum ($\approx 1 - 10$ W.u.). In a naive valence-shell picture the E2 transition strength should increase towards, and maximize at mid-shell, increasing with the number of valence nucleons (holes). The expected maximum of collectivity at mid-shell can also be shown in the SU(3) limit of the interacting boson model (IBM) [15, 16].

It was pointed out by Zhang et al. [17], based on the available experimental data at that time, that B(E2)values and g factors [18, 19] of the first excited states of even-even nuclei around A = 170 do not maximize, but instead saturate near mid-shell, as can be seen in Figure 1 (a) and (b). This has been explained qualitatively by an overlap of the proton and neutron wave functions and a reduction of the proton-neutron interaction strength near mid-shell. Furthermore, it was pointed out in Refs. [16, 20] that the evolution of E2 strengths of the tungsten and hafnium isotopes exhibits an irregularity, which can be clearly identified in the differential of B(E2) values defined as $\delta B(E2) = B(E2)_N - B(E2)_{N-2}$. A smooth trend of the differential is observed for heavier isotopes (osmium to lead), but oscillations or sharp drops were seen for the data on hafnium and tungsten isotopic chains. These oscillations may hint at nuclear structural anomalies or may simply point to incorrect experimental data (since the same $B(E2)_N$ value occurs in $\delta B(E2)_N$ and $\delta B(E2)_{N+2}$ [16]). Missing data and large uncertainties of experimental data provide motivation for new experiments in this region of the nuclear chart to clarify the situation [16, 21–24], and indeed, discrepancies from literature values of the lifetime of the first excited 2^+ states of the hafnium and tungsten isotopes in the order of up to 20% were found. B(E2) observables directly relate to the β degree of freedom. As such an effective β value is obtained from $\beta^2 \propto B(E2; 0_1^+ \rightarrow 2_1^+)$, and the $B_{4/2} = B(E2; 4_1^+ \rightarrow 2_1^+)/B(E2; 2_1^+ \rightarrow 0_{gs}^+)$ ratio quantifies the softness of the nuclear potential in β [25–27]. Unfortunately, data on the $B(E2; 4^+_1 \rightarrow 2^+_1)$ value were missing for ¹⁷⁸Hf which complicates the analysis of the evolution of quadrupole collectivity in the isotopes under investigation.

In the current work, we present E2 transition strengths

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FIG. 1. (a) Systematics of g factors of first excited 2_1^+ states of even-even nuclei around neutron number N = 104 (marked with grey-dashed line) of Er, Yb, Hf, W and Os isotopes. The g factor values seem to saturate around (or slightly below) N = 104 apart from the Hf isotopic chain, which only has three data points. Data have been taken from [19]. (b) Systematics of $B(E2; 2_1^+ \to 0_{gs}^+)$ values of even-even Er, Yb, Hf, W and Os isotopes around neutron number N = 104 (marked with vertical grey dashed line) from literature data [1–14, 28]. The values of different isotopes are slightly shifted for a better visibility.

of the yrast bands of the isotopes 174,176,178,180 Hf extracted from a measurement campaign, using fast electronic scintillation timing (FEST) at the IFIN-HH in Bucharest. Systematic uncertainties on their evolution across this sequence of nuclide were reduced by using the same setup for all isotopes of interest. Moreover, mean lifetimes of the non-yrast states (3_1^- and 2_1^- states of 176 Hf) have been determined and will be presented.

II. EXPERIMENT AND ANALYSIS

Excited states of ¹⁸⁰Hf were populated via Coulomb excitation induced by an ¹⁶O primary beam at 55 MeV. just at the Cline-criterion for safe Coulomb excitation [29] and well below the Coulomb barrier at 88 MeV. The oxygen beam, delivered by the Bucharest FN Tandem accelerator, impinged on a $12 \,\mathrm{mg/cm^2}$ thick $^{180}\mathrm{Hf}$ target. The enrichment of the target was 93.9% of $^{180}\mathrm{Hf}$ with small contaminations from other stable Hf isotopes: 179 Hf (1.6%), 178 Hf (2.8%), 177 Hf (1.3%) and 0.4% of other isotopes. The other Hf isotopes 174,176,178 Hf were excited via β -decay following the fusion-evaporation reactions 171 Yb $({}^{6}$ Li,3n $){}^{174}$ Ta, 172 Yb $({}^{7}$ Li,3n $){}^{176}$ Ta and $^{174}\mathrm{Yb}(^{7}\mathrm{Li},\!3\mathrm{n})^{178}\mathrm{Ta}$ at a beam energy of 30 MeV with target thicknesses of $^{172}\mathrm{Yb}:$ $2.3\,\mathrm{mg/cm^2},$ $^{171}\mathrm{Yb}:$ $3\,\mathrm{mg/cm^2}$ and 174 Yb: 2.5 mg/cm^2 . The beam was cycled between ON (one hour) and OFF (one hour) to take in- and offbeam data. De-excitation γ rays were detected using the ROSPHERE detector array [30] in a configuration with 11 LaBr₃ and 14 HPGe detectors. The LaBr₃ and HPGe detectors were arranged in five rings around the target chamber.

The master-trigger condition for the experiment was set on either two or more coincident γ rays in the LaBr₃ detectors or two or more coincident γ rays in the HPGe detectors. In addition, for the ¹⁸⁰Hf data, for two hours the trigger conditions were set to HPGe singles, in order to be able to perform a Coulomb-excitation analysis of ¹⁸⁰Hf. The energy and the efficiency calibrations in the range between 121 keV and 1408 keV were done using a ¹⁵²Eu source, which was also used to determine the energy-dependent time walk of the experimental setup. The lifetime of an excited nuclear state can be extracted using the fast-timing method, by measuring the time difference between two signals coming from a populating and a depopulating transition. A detailed description can be found in [31–33]. The measured time difference $\Delta t_{\rm m}$ between two signals does not only contain the effective lifetime $\tau_{\rm eff}$, i.e. the sum of the lifetimes that lie between the γ -ray transitions, but also the energy-dependent time walk [31]:

$$\Delta t_{\rm m} = \tau_{\rm eff} + t_{\rm tw,1}(E_{\gamma,1}) + t_{\rm tw,2}(E_{\gamma,2}).$$
(1)

To determine the lifetime, the energy-dependent time walk of each detector was determined using an ^{152}Eu source with its well-known γ lines from the γ decay of 152 Gd and 152 Sm ranging from 244 keV to 1299 keV, as described in [33–35], by fitting a polynomial function to the time response using the full energy peaks of the europium source. After applying the corrections the data was sorted into $E_{\gamma, \text{LaBr}_3}$ - $E_{\gamma, \text{LaBr}_3}$ - Δt cubes. Timedifference spectra between two transitions were extracted from these cubes by selecting the coincidence areas of the transitions (e.g. populating and depopulating transition of the nuclear state of interest) in the energy-energy plane (see Figure 2). To exclude contaminants of other transitions on the time-difference spectrum, the LaBr₃ gates were compared with $E_{\gamma,\text{Ge}}$ - $E_{\gamma,\text{Ge}}$ matrices and only pairs of coincidences, where a clean selection was guaranteed, were used. The resulting time difference was corrected for random coincidences and the Compton background



FIG. 2. Section of the E_{γ,LaBr_3} - E_{γ,LaBr_3} matrix obtained after projection of the E_{γ,LaBr_3} - E_{γ,LaBr_3} - Δt cube. Marked are the coincidence areas of the γ -ray pair 93 and 213 keV, corresponding to the transitions $2^+_1 \rightarrow 0^+_{\text{gs}}$ and $4^+_1 \rightarrow 2^+_1$ of ¹⁷⁸Hf. They are not distinguishable from the γ -ray pair $8^-_1 \rightarrow 8^+_1$ (88 keV) and $4^+_1 \rightarrow 2^+_1$ (213 keV) as mentioned in the text.

below the full energy peaks in the energy spectrum (see also Figure 3). This was done by selecting an area around the 2-D peak in the energy-energy plane.

The resulting delayed time distribution $D_{\lambda}(t)$, without any background contributions, is a convoluted function of the prompt response of the detection system P(t') and the exponential decay of the nuclear state of interest [36]:

$$D_{\lambda}(t) = n\lambda \int_{-\infty}^{t} P(t')e^{-\lambda(t-t')}dt', \qquad (2)$$

with the transition rate $\lambda = 1/\tau$ and the normalization n. If the lifetime of the nuclear state of interest is larger than the full width at half maximum (FWHM) of the system's response, a simple fit of the exponential decay of the time distribution gives the lifetime of the nuclear state. In the case that the lifetime is small in comparison to the FWHM of the system's response other methods, e.g. the centroid shift method [31, 32], have to be used to determine the lifetime of the nuclear state. The centroid of the time distribution is defined as [37]:

$$C(D_{\lambda}) = \langle t \rangle = \frac{\int t D_{\lambda}(t) dt}{\int D_{\lambda}(t) dt},$$
(3)

resulting for the delayed case (start condition on the feeding transition) in

$$\tau = C_d(D_\lambda) - C_d(P),\tag{4}$$

with the centroid of the delayed time distribution $C_d(D_\lambda)$ and of the system's prompt response $C_d(P)$. When the gates are switched also C(D) and C(P) switch places in the equation. After the Time-Walk correction $C_d(P)$ is equal to the centroid $C_a(P)$ of the anti-delayed time distribution. It follows for the centroid difference [38]:

$$\Delta C = C_d(D) - C_a(D) \tag{5}$$

$$= C_d(P) + \tau - (C_a(P) - \tau) \tag{6}$$

$$= 2\tau.$$
 (7)

Lifetimes of the long-lived 2_1^+ states of 174,176,178,180 Hf and of the 2_1^- state of 176 Hf, in the range of ns, were determined using the slope method using γ - γ coincidences within the LaBr₃ detectors. Energy gates were set on the $4_1^+ \rightarrow 2_1^+$ and $2_1^+ \rightarrow 0_{gs}^+$ transitions in the case of the 2_1^+ state, and on the $2_2^- \rightarrow 2_1^-$ and $2_1^- \rightarrow 2_1^+$ transitions for the 2_1^- state. The most reliable region for the fit was determined by moving time gates with different widths across the time-difference spectrum. Only the regions without fluctuations, i.e. contributions from the prompt peak or the background, were used for the fits. The time-difference spectra, including the fits are shown in Figure 4 and Figure 7(a).

The lifetimes of short-lived excited states, in this work 4_1^+ , 6_1^+ , 3_1^- states, were determined by the centroid shift method. Figure 5 and Figure 7 (b) show the delayed and anti-delayed time distributions for the short-lived yrast states of 174,176,178,180 Hf and for $\tau(3_1^-)$ of 176 Hf. The used decay cascades are shown next to the time distributions. In the case of 176 Hf smaller gate widths were taken because of the higher transition density [see Figure 3 (b)], resulting in lower statistics of the time distributions and therefore larger uncertainties of the determined lifetimes.

III. RESULTS

A. ¹⁷⁴**Hf**

Figure 3 shows the obtained γ -ray spectra of the HPGe and LaBr₃ detectors. Transitions relevant for the determination of the investigated lifetimes are marked with triangles.

The decay of ¹⁷⁴Ta to ¹⁷⁴Hf via electron capture populates excited positive-parity states [39]. The yrast band is populated up to the 6_1^+ state and transitions feeding and depopulating these states can be identified in the energy spectrum [see Figure 3 (a)]. The partial level scheme of ¹⁷⁴Hf, including the investigated states, is depicted in Figure 6 (a). Lifetimes of the 2_1^+ , 4_1^+ and 6_1^+ states were extracted from the data. The pair of $6^+_1 \rightarrow 4^+_1$ and $4_1^+ \rightarrow 2_1^+$ coincident transitions has not been used for the determination of the lifetime of the 4^+_1 state as for the other investigated isotopes, because the sum energy of both transitions $[E(6_1^+ \rightarrow 4_1^+) + E(4_1^+ \rightarrow 2_1^+) = 517 \text{ keV}]$ is close to 511 keV. As a result, Compton scattered events from 511 keV annihilation γ rays contaminate the coincidence area of the $6_1^+ \rightarrow 4_1^+$ and $4_1^+ \rightarrow 2_1^+$ transitions. Hence, the pair of the $4_3^+ \rightarrow 4_1^+$ and $4_1^+ \rightarrow 2_1^+$ transitions has been used instead to determine the lifetime of the 4_1^+ state. The obtained results are given in Table I.



FIG. 3. Partial energy spectra of all LaBr₃ (red) and HPGe detectors (black) for (a) 174 Hf, (b) 176 Hf, (c) 178 Hf and (d) 180 Hf. Transitions used for the determination of the lifetimes are marked by blue triangles.



FIG. 4. Logarithmic plots of the time-difference spectra for the decays of 2_1^+ states of the isotopes 174,176,178,180 Hf from (a)-(d). Final linear fits are indicated by the dashed black lines.



FIG. 5. Logarithmic plots of the time-difference spectra for the decays of short lived (range of ps) yrast states of the isotopes ^{180,178,176,174} Hf. The delayed time spectra are shown in black (solid lines) and the anti-delayed spectra in orange (dashed lines). (a) Shows the time-difference spectra for the $6_1^+ \rightarrow 4_1^+ \rightarrow 2_1^+$ cascade of ¹⁸⁰ Hf, (b) for the $6_1^+ \rightarrow 4_1^+ \rightarrow 2_1^+$ cascade of ¹⁷⁸ Hf, (c) for the $8_1^+ \rightarrow 6_1^+ \rightarrow 4_1^+ \rightarrow 4_1^+$ cascade of ¹⁷⁸ Hf, (d) for the $3_1^- \rightarrow 4_1^+ \rightarrow 2_1^+$ cascade of ¹⁷⁶ Hf, (e) for the $4_3^+ \rightarrow 4_1^+ \rightarrow 2_1^+$ cascade of ¹⁷⁴ Hf and (f) for the $4_2^+ \rightarrow 6_1^+ \rightarrow 4_1^+ \rightarrow 4_1^+$ cascade of ¹⁷⁴ Hf.



FIG. 6. Partial level schemes of the investigated Hf isotopes, $^{174-180}$ Hf (a)-(d). Shown are the states and transitions used for fast-timing lifetime measurement. The determined mean lifetimes are given in yellow. In the case of 180 Hf the transition widths correspond to the observed γ -ray intensity.

TABLE I. Measured mean lifetimes of excited states of 174,176,178,180 Hf. Adopted literature values were taken from Ref.[10–13] and from Ref.[24]. Values obtained from the Coulomb-excitation analysis are indicated with CLX. Newly obtained lifetimes and transition strengths are marked by an asterisk^{*}. Two different cascades can be used to determine $\tau(3_1^-)$ of 176 Hf, resulting in an adopted value of $\tau(3_1^-) = 25(12)$ ps. Weighted average value of $\tau(2_1^+)$, $\tau(4_1^+)$ and $\tau(6_1^+)$ of 174,176 Hf have been taken from [24] and this work. $\tau_{\text{weighted}}(4_1^+)$ of 180 Hf has been taken from the CLX calculation and the fast-timing measurement. $B(E2; \lambda \to \lambda - 2)$ have been determined from τ_{weighted} and $\tau_{\text{exp.}}$ with the internal conversion coefficients taken from BrIccFO [40].

| isotopes | J_n^p | gate-1 | gate-2 | $\tau_{\rm exp.}$ in ps | $	au_{ m lit.}$ in ps | $	au_{	ext{weighted}}$ in ps | α [40] | $\begin{array}{c} B(E2)\\ \text{in W.u.} \end{array}$ |
|-------------------|---|--|--|--|--|---|--|---|
| ¹⁷⁴ Hf | $2^+_1 \\ 4^+_1 \\ 6^+_1$ | $\begin{array}{c} 4^+_1 \to 2^+_1 \\ 4^+_3 \to 4^+_1 \\ 4^+_2 \to 6^+_1 \end{array}$ | $\begin{array}{c} 2^+_1 \to 0^+_1 \\ 4^+_1 \to 2^+_1 \\ 6^+_1 \to 4^+_1 \end{array}$ | $1920(30) \\95(11) \\23(11)$ | $1847(58) [24] \\111(7) [24] \\23(7) [24]$ | $1905(30) \\ 106(7) \\ 23(6)$ | $5.12(8) \\ 0.258(4) \\ 0.071(1)$ | $194(4) \\ 282(16) \\ 198^{+69}_{-41}$ |
| ¹⁷⁶ Hf | $\begin{array}{c} 2^+_1 \\ 4^+_1 \\ 2^1 \\ 3^1 \\ 3^1 \\ 3^1 \end{array}$ | $\begin{array}{c} 4^+_1 \to 2^+_1 \\ 3^1 \to 4^+_1 \\ 2^2 \to 2^1 \\ 2^2 \to 3^1 \\ (2,3)^- \to 3^1 \end{array}$ | $\begin{array}{c} 2^+_1 \to 0^+_1 \\ 4^+_1 \to 2^+_1 \\ 2^1 \to 2^+_1 \\ 3^1 \to 4^+_1 \\ 3^1 \to 4^+_1 \end{array}$ | $2100(60) \\ 133(24) \\ 6280(160) \\ 26(18)^* \\ 25(16)^*$ | 2121(87) [24] 130(9) [24] 6720(250) [11] | $2107(49) \\ 130(8) \\ 6407(200) \\ 25(12)^*$ | 5.77(9) 0.279(4) - - | 181(5) 250(16) - - - |
| ¹⁷⁸ Hf | $2^+_1 \\ 4^+_1 \\ 6^+_1$ | $\begin{array}{c} 4^+_1 \to 2^+_1 \\ 6^+_1 \to 4^+_1 \\ 8^+_1 \to 6^+_1 \end{array}$ | $\begin{array}{c} 2^+_1 \to 0^+_1 \\ 4^+_1 \to 2^+_1 \\ 6^+_1 \to 4^+_1 \end{array}$ | $2119(13) \\ 85(13)^* \\ 12(12)$ | $2155(33) [12] \\ - \\ 16(1) [12]$ | $2124(12) \\ 85(13)^* \\ 16(1)$ | $\begin{array}{c} 4.66(7) \\ 0.232(4) \\ 0.062(1) \end{array}$ | $163(3) \\ 296^{+53*}_{-39} \\ 221(14)$ |
| ¹⁸⁰ Hf | $2^+_1 \\ 4^+_1 \\ 4^+_1 \\ 6^+_1$ | $\begin{array}{c} 4_1^+ \to 2_1^+ \\ 6_1^+ \to 4_1^+ \end{array} \\ \text{CLX} \end{array}$ | $\begin{array}{c} 2_1^+ \rightarrow 0_1^+ \\ 4_1^+ \rightarrow 2_1^+ \end{array}$ | $2198(16) \\ 100(23) \\ 103(12) \\ 12(2)$ | 2191(14) [13] 102(14) [13] 14(1) [13] | $2194(11) \\ 102(11) \\ 14(1)$ | $4.63(7) \\ 0.225(4) \\ 0.059(1)$ | $155(2) \\ 234^{+28}_{-23} \\ 225(16)$ |

B. ¹⁷⁶**Hf**

The decay of ¹⁷⁶Ta to ¹⁷⁶Hf via electron capture results in a complicated decay scheme with many transitions [41, 42], in comparison to the decay of ¹⁷⁸Hf, as can be seen from Figure 3 (b). The part of the level scheme relevant for this work is shown in Figure 6 (b). Apart from the yrast band, also a low-lying negative-parity K = 2band of ¹⁷⁶Hf is strongly populated via this decay. The mean lifetimes of the 2^-_1 state and for the first time of the 3^-_1 state have been determined (see Figure 7). For the determination of $\tau(3^-_1)$ two possible populating transitions have been used, i.e. the transitions at 611 keV $[(2,3)^- \rightarrow 3^-_1]$ and at 645 keV $(2^-_2 \rightarrow 3^-_1)$, resulting in a more precise value of $\tau(3^-_1) = 25(12)$ ps. The extracted mean lifetime of the 2^-_1 state is lower than the adopted value given in [11] but in agreement with other measurements, e.g. [43].

Since many transitions can be seen, the gates for the determination of the mean lifetimes have to be carefully selected in the LaBr₃ detectors. All selected energy gates were cross checked within the energy spectra of the HPGe detectors. Table I summarizes the extracted mean lifetimes of 176 Hf.

C. ¹⁷⁸**Hf**

Excited states of $^{178}{\rm Hf}$ were populated via $\beta{\rm -decay}$ from $^{178}{\rm Ta}.$ Primarily the 8^- isomeric state at an en-

ergy of 1147 keV was populated, which decays through the emission of γ rays through the yeast band. The obtained γ -ray spectrum is shown in Figure 3 (c). The decay transition of the 8^-_1 state to the 8^+_1 state (88.9 keV) and the transition $2^+_1 \rightarrow 0^+_{\rm gs}$ (93.2 keV) are very close in energy. For this reason, these transitions can not be distinguished within the $LaBr_3$ detectors [see Figure 3 (c)] and it is not possible to set additional energy gates in the HPGe detectors since both transitions are from the same decay cascade. Selecting the region marked in Figure 2 results in a time-difference spectrum which is a superposition of two time-difference distributions, on one hand a distribution gated on the $2_1^+ \rightarrow 0_{gs}^+$ and $4_1^+ \rightarrow 2_1^+$ transitions and on the other hand a distribution gated on the $8_1^- \rightarrow 8_1^+$ and $4_1^+ \rightarrow 2_1^+$ transitions. The latter corresponds to the effective mean lifetime of the 4_1^+ , 6_1^+ and the 8_1^+ states combined (all on the order of ps). However, the time-difference distribution is a superposition of a delayed and an anti-delayed distribution, since the ordering of the gates is reversed for the two cases [the 88 keV transition is above the 213 keV transition and the 93 keV transition is below 213 keV, cf. Figure 6 (c)]. Due to the fact that the tail, stemming from the effective mean lifetime of the 4_1^+ , 6_1^+ and the 8_1^+ states, is located on the other side of the time difference distribution, it is possible to identify the exponential decay of the long lived 2_1^+ state and to extract its mean lifetime [see Figure 4 (c)]. In addition to $\tau(2_1^+)$ the mean lifetimes of the 4_1^+ (for the first time) and 6_1^+ states have been determined by using the centroid-shift method. The results



FIG. 7. (a) Logarithmic plots of the time-difference spectrum for the decay of 2_1^- state of the isotopes ¹⁷⁶Hf. Final linear fit is indicated by the dashed black line. (b) Delayed (black) and anti-delayed (orange) time distributions of the cascade $(2/3)^- \rightarrow 3_1^- \rightarrow 4_1^+$.

are presented in Table I.

D. ¹⁸⁰**Hf**

Mean lifetimes of ¹⁸⁰Hf have been determined via FEST and Coulomb excitation, with the computer code CLX [44], based on the original code by Winther and De Boer[45]. Since the ¹⁸⁰Hf target had been also used in a previous fusion-evaporation experiment, there was still activated material in the target and many transitions from other isotopes, e.g. $^{181}\mathrm{Ta},\ ^{182}\mathrm{W}$ and ¹⁸⁴W can be identified in the HPGe energy spectrum [see Figure 3(d)]. Moreover, for a determination of the mean lifetime of the first 2^+ state the contribution from the 178 Hf content of the target has to be taken into account. The transition energies of the $2_1^+ \rightarrow 0_{gs}^+$ and $4_1^+ \rightarrow 2_1^+$ transitions are very close in energy (93.2 keV vs. 93.3 keV and 213.4 keV vs. 215.3 keV) in ¹⁷⁸Hf and ¹⁸⁰Hf. They cannot be separated in the LaBr₃ detectors, resulting in a superposition in the time-difference spectrum. However, with the measured mean lifetimes of 178 Hf and the observed intensity of the $6_1^+ \rightarrow 4_1^+$ transition of ¹⁷⁸Hf at 332 keV, the ¹⁷⁸Hf content can be subtracted. The determined mean lifetimes together with the transitions used are listed in Table I. In addition to the mean lifetimes determined by the fast-timing measurement the values obtained by the Coulomb-excitation analysis are shown.

The Coulomb-excitation yields were calculated from the efficiency-corrected γ -ray intensities, with the ¹⁷⁸Hf γ -ray transition intensities subtracted, normalized to the $2_1^+ \rightarrow 0_{gs}^+$ transition. The yields of the excited states, relative to that of the 2_1^+ state, are proportional to the relative Coulomb-excitation cross sections. The matrix elements of the $4_1^+ \rightarrow 2_1^+$ and $6_1^+ \rightarrow 4_1^+$ transitions were fitted to reproduce the relative Coulomb-excitation yields with the multiple Coulomb-excitation code CLX [44]. The energy loss of 18 MeV of the beam in the target was taken into account. Uncertainties for the Coulomb-excitation analysis were determined via variation of the matrix elements within the range of the calculated Coulomb-excitation yields. Both values of $\tau(4_1^+)$ agree within their uncertainties and a weighted average of these independent evaluations can be extracted: $\tau(4_1^+) = 102(11)$ ps.

The determined mean lifetimes are directly related to the transition strength

$$\frac{1}{\tau} = 8\pi \frac{\lambda + 1}{\lambda [(2\lambda + 1)!!]^2} \left(\frac{E_{\gamma}}{\hbar c}\right)^{2\lambda + 1} \\ \times B(\sigma\lambda; J_i \to J_f) \times (1 + \alpha), \tag{8}$$

with the electron conversion coefficient α , the multipolarity λ , the energy of the γ rays E_{γ} and the radiation character σ . Since the decay between the states of the yrast band in even-even nuclei is dominated by E2 radiation, i.e. it is the only possible γ decay in the case of the $2_1^+ \rightarrow 0_{gs}^+$ transition, the B(E2) values can be extracted from the determined mean lifetimes. Since a similar measurement method was used in [24] and the lifetimes are in good agreement with each other, weighted average values of the lifetimes of this work and [24] are given in Table I. The conversion coefficients for the determination of the B(E2) values were taken from BrIccFO [40]. In the case of large uncertainties in $\tau_{weighted}$, asymmetric uncertainties are given for the B(E2) values.

IV. DISCUSSION

A. Quadrupole Collectivity

The new B(E2) values do not show a saturation at mid-shell, but a maximum shifted towards a lower neutron number, as can be seen in Figure 8 (a). Depicted are $B(E2; 2_1^+ \rightarrow 0_{gs}^+)$ strengths over the product $N_p N_n$



FIG. 8. (a) $B(E2; 2_1^+ \to 0_{\rm gs}^+)$ strengths in W.u. plotted over the product proton valence particles (holes) and of neutron valence particles (holes) $N_p N_n$ of the Er, Yb, Hf and W isotopic chains. The gray dashed lines are plotted to guide the eyes. Data taken from [1–14, 24] and this work. (b) "Quadrupole" B(E2) values, $B(E2)_Q$, vs. $N_p N_n$ obtained by removing the β_4 hexadecapole deformations as described in [46]. β_2 values are taken from [28] and the β_4 values from [47]. (c) β_4 hexadecapole deformations vs. the neutron number. Data taken from [47].

[48] of numbers of proton valence particles or holes with respect to the major shell closures at Z=50 and Z=82and of neutron valence particles or holes with respect to the major neutron shell closures at N=82 and N=126. The turning point of each graph for the different isotopic chains is at neutron mid-shell, since the number of valence particles (holes) maximize at mid-shell. Approaching mid-shell from lower neutron numbers, members of different isotopic chains show the same $N_p N_n$ dependence as indicated by the dotted line in Figure 8(a). Their collectivity, i.e. the B(E2) strength, increases with the number of valence particles (holes) [48]. However, generally the B(E2) strengths show an early drop before mid-shell is reached. In addition, an asymmetry is visible for the isotopic chains from Yb to W. That is, values past the inflection point are significantly lower, while again a continuous trend with respect to $N_p N_n$ emerges [see the dashed line in Figure 8 (a)]. In the case of the Er isotopic chain B(E2) data is not available at and beyond mid-shell. The pre-mid-shell maximum of the B(E2) strengths gets more pronounced for higher proton numbers. Zamfir *et al.* [46] proposed that the different trends of the B(E2) values versus N_pN_n are due to the influence of the hexadecapole deformation parameter β_4 on the quadrupole moment Q. The expansion of the quadrupole moment shows the dependence of Q from β_4 :

$$Q \propto \beta_2 (1 + 0.36\beta_2 + 0.97\beta_4 + \dots). \tag{9}$$

The B(E2) strength is proportional to Q^2 and therefore depends on β_4 . The influence of β_4 can be removed by defining a "pure quadrupole" B(E2) value [46]:

$$B(E2; 2_1^+ \to 0_{\rm gs}^+)_Q = B(E2; 2_1^+ \to 0_{\rm gs}^+) \left(\frac{Q(\beta_4 = 0)}{Q(\beta_4 \neq 0)}\right)^2.$$
(10)

The $B(E2)_Q$ values are shown in Figure 8 (b). The deformation parameters were taken from [28] or if not experimentally available from [47]. Note that β_4 changes slope from near-constant around $\beta_4 = 0$ to negative values at N = 98 in the Hf (Z = 72) isotopic chain [cf. Figure 8(c)]. Figure 8(b) shows the effect of the hexadecapole correction: the B(E2) values of the low-N branches of the curves increase while the values of the high-N branches decrease. That means, the pre-mature reversion of B(E2) values is lifted. Although numerous indications for hexadeapole collectivity in the high-N, high-Z region of this major shell exist, a fully microscopic explanation of the early B(E2) maximum has not been provided to date. Nevertheless we examine the systematic behavior of another observable in the following, which has been discussed in conjunction with the evolution of collectivity, e.g. the energy ratios $R_{4/2}$ [49]. The average proton-neutron interaction of the last proton with the last neutron of the nucleus [48] is the double difference of binding energies, introduced by [50]:

$$\delta V_{pn}(Z,N) = \frac{1}{4} \{ [BE(Z,N) - BE(Z,N-2)] - [BE(Z-2,N) - BE(Z-2,N-2)] \},$$
(11)

with the binding energy BE(Z, N). In general [51], δV_{pn} depends on the trend of the occupation of neutron and proton orbits from high *j*-low *n* to low *j*-high *n* orbits within a major shell. δV_{pn} is large for smaller values of δn and δj , hence, it should be large for regions where the overlap of neutron and proton orbits is large. Figure 9 shows the δV_{pn} obtained with the binding energies from [52, 53] in a *N*-*Z* chart. δV_{pn} is in general small in the area above proton mid-shell and below neutron mid-shell or above proton and neutron mid-shell. Figure 9 (b) shows δV_{pn} of the Hf and W isotopic chains, calculated from available experimental data [52, 53]. δV_{pn} is nearly constant before and after the neutron mid-shell, but shows



FIG. 9. (a) Depicted is the double difference δV_{pn} [20] in a N-Z chart. N=104 is marked by the dotted dashed line. The areas marked in blue is shown in (b). The dotted dashed lines are fits with a constant to the Hf data before and after mid-shell, illustrating the sudden increase of δV_{pn} at N=104 and the constant behavior before and after N=104. The vertical grey dashed line marks the neutron mid-shell N=104. Binding energies taken from [52, 53].

a sudden increase at N=104. The increase is in accordance with the above mentioned pattern. Potentially, the sudden increase of the δV_{pn} could already be reflected at N<104, if neutrons already scatter early into the corresponding orbitals.

Examining E2 observables within the ground-state bands of the Hf isotopes, firstly, we measured the $B(E2; 4_1^+ \rightarrow 2_1^+)$ value for the first time, although, with large relative uncertainty. Nevertheless, the resulting value of the $B_{4/2}$ ratio fits well into the systematics shown in Figure 10 (b). All $B_{4/2}$ values of the Hf isotopes are distributed around the rotational limit of $B_{4/2,\text{rot}} = 1.44$. Inspection of the B(E2) rates of higherlying states, however, as shown in Figure 10(a) reveal significant and seemingly robust deviations from the rotational limit. In comparison to the rigid rotor value, the determined $B(E2; 6_1^+ \rightarrow 4_1^+)$ transition strength of ¹⁷⁴Hf is unexpectedly smaller than the corresponding $B(E2; 4_1^+ \rightarrow 2_1^+)$ value. For the case of ¹⁷⁶Hf a similar trend cannot be excluded, because of the large uncertainty of the $B(E2; 6_1^+ \rightarrow 4_1^+)$ value. Unfortunately no data are available for the higher lying states of the yrast band of these two isotopes. Similar results were obtained in Ref. [24] and in other rare-earth isotopes, e.g. 162 Yb [54] and a too low, with respect to the rigid rotor limit, $B(E2; 6_1^+ \to 4_1^+)$ value seems to be a re-occurring and so far unexplained feature in the rare-earth region, while other B(E2) values, e.g. in ^{178,180}Hf are in good



FIG. 10. (a) Normalized $B(E2; J \rightarrow J - 2)$ transition strengths are plotted over J for the isotopes 174,176,178,180 Hf. 178,180 Hf are close to the rigid rotor limit. The $B(E2; 6^+_1 \rightarrow 4^+_1)$ strength is significantly lower than the $B(E2; 4^+_1 \rightarrow 2^+_1)$ strength of 174 Hf. The isotopes are slightly shifted for a better visibility. (b) Ratio $B_{4/2}$ of the B(E2)transition strengths $B(E2; 4^+_1 \rightarrow 2^+_1)$ and $B(E2; 2^+_1 \rightarrow 0^+_{gs})$. Shown in the red box are the averaged values from this work and [12, 13, 24] and outside the box the known literature data taken from [5–9]. The dotted line represents the value obtained in the rigid rotor limit and in the vibrator limit.

agreement with the rigid-rotor limit. Therefore, again, a change in rotational structure past N = 100 is seen in the Hf isotopes.

B. $K^{\pi} = 2^{-}$ band of ¹⁷⁶Hf

In ¹⁷⁶Hf, in addition, the low-lying negative parity band was observed. The 2_1^- and 3_1^- states have been assigned as members of a $K^{\pi} = 2^-$ band by [41, 42]. The expected *E*1 decay transitions from this band to the yrast band are first forbidden transitions ($\Delta K = 2$) according to the Alaga rules [55] and from conversionelectron data a large admixture of *M*2 and *E*3 has been confirmed for the decay transitions of the 2_1^- state [41]. The decay transitions of the 3_1^- state were assigned to have *E*1 character. Unfortunately, the decay of the $3_1^$ state to the 2_1^- state at 65 keV was not observed or is obscured by x-ray transitions. The 3_1^- state is short lived with a mean lifetime of $\tau(3_1^-) = 25(12)$ ps. The extracted mean lifetime of the 2_1^- state is slightly below the adopted literature value [11], but in good agreement with other measurements [43].

Under the assumption that the transition width of the

transition $3_1^- \rightarrow 2_1^-$ is negligible, an upper limit for the E1 transition strength of the two other decay transitions of the 3_1^- state can be determined. Even from this upper limit the suppression of the transition rates is visible. The E1 strength is three orders of magnitude smaller than other E1 strength in this nucleus. An overview over the E1 strength distribution in ¹⁷⁶Hf was given by [56]. From the new weighted average value of $\tau(2_1^-)$ the transition strengths of its decay $2_1^- \rightarrow 2_1^+$ can be determined with the known multipole mixing ratios from [43] and the branching ratios from [11]. The transition strengths are given in Table II.

TABLE II. The $B(E\lambda)$ strengths of ¹⁷⁶Hf were determined with the known branching ratios taken from [11], the conversion coefficients taken from [40] and the multipole-mixing ratios of the $2_1^- \rightarrow 2_1^+$ transition taken from [43].

| | $B(M/E\lambda) \ { m in} \ \mu_N^2 { m b}^{2\lambda-2}/e^2 { m b}^\lambda$ | $\begin{array}{c} B(E\lambda)\\ \text{in W.u.} \end{array}$ |
|--|--|---|
| $\begin{array}{c} B(E1;2^1 \to 2^+_1) \\ B(M2;2^1 \to 2^+_1) \\ B(E3;2^1 \to 2^+_1) \end{array}$ | $\begin{array}{c} 4(2) \times 10^{-10} \\ 10(3) \times 10^{-3} \\ 6(2) \times 10^{-2} \end{array}$ | $2(1) \times 10^{-8} 2(1) \times 10^{-2} 30(12)$ |
| $B(E1; 3_1^- \to 4_1^+) B(E1; 3_1^- \to 2_1^+)$ | $\begin{array}{c} 8^{+6}_{-3} \times 10^{-8} \\ 9^{+9}_{-3} \times 10^{-8} \end{array}$ | $\begin{array}{c} 4^{+3}_{-2} \times 10^{-6} \\ 4^{+5}_{-1} \times 10^{-6} \end{array}$ |

V. SUMMARY

In this work several lifetimes of yrast states of the hafnium isotopes with A = 174, 176, 178 and 180 have been determined using fast-electronic scintillation timing and a Coulomb-excitation analysis. The mean lifetimes of the 2_1^+ and 4_1^+ states of all isotopes and the lifetimes of the 6_1^+ states of 174,178,180 Hf have been measured and are in good agreement with other recent results [24]. The mean lifetime, $\tau(4_1^+) = 85(13)$ ps, of 178 Hf has been determined for the first time. From these lifetimes the B(E2) transition strengths between the yrast states have been calculated and the $B_{4/2}$ ratios have been extracted.

In addition, the mean lifetimes of the 2^-_1 and the 3^-_1 states of $^{176}{\rm Hf}$ have been determined.

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