

First On-Line Laser Spectroscopy of Radioisotopes of a Refractory Element

J. M. G. Levins,¹ D. M. Benton,² J. Billowes,¹ P. Campbell,¹ T. G. Cooper,¹ P. Dendooven,³ D. E. Evans,²
D. H. Forest,² I. S. Grant,¹ J. A. R. Griffith,² J. Huikari,³ A. Jokinen,³ K. Peräjärvi,³ G. Tungate,²
G. Yeandle,¹ and J. Äystö³

¹*Schuster Laboratory, University of Manchester, Manchester M13 9PL, United Kingdom*

²*School of Physics and Astronomy, University of Birmingham, Edgbaston, Birmingham B15 2TT, United Kingdom*

³*Department of Physics, Accelerator Laboratory, University of Jyväskylä, PL 35, Jyväskylä SF-40351, Finland*

(Received 9 October 1998)

The first fully on-line isotope shift measurement of a radioactive refractory element is reported. Collinear laser-induced fluorescence measurements were made on the radioactive isotopes $^{170,172,173,174}\text{Hf}$ produced with a flux of $2-3 \times 10^3$ ions per second from an ion-guide fed isotope separator. The method may be applied to all elements and isomers with lifetimes as short as 1 ms. The systematics of the new charge radii measurements are well reproduced by theory, with the maximum deformation in the chain occurring significantly below the midshell. [S0031-9007(99)08734-7]

PACS numbers: 21.10.Ft, 21.10.Ky, 27.70.+q, 42.62.Fi

The sensitivity of laser spectroscopy gives it a unique role in the investigation of mean-square charge radii and ground state moments of rare nuclear species [1]. Studies of short-lived radioactive nuclei have been performed at on-line isotope separators with great success for over twenty years [2]. The majority of the work has used the collinear atom- or ion-laser beam technique. Access to many nuclear systems has, however, been severely hindered by the slow release times and restrictive chemical selectivities of conventional catcher-ionizer sources. Low vapor pressures, complex atomic structures, and high transition frequencies can add further difficulty in these cases. "Semi-on-line" measurements have been performed on Pt isotopes, produced as daughters of the decay of Hg beams, most successfully using resonance ionization synchronized with pulsed laser desorption [3]. Further off-line spectroscopy on radioisotopes of Hf and on stable isotopes of Hf, Ta, and W has been achieved, using ion trap techniques [4,5] and ion-sputtering atomic beam sources [6], but no isotope shift measurements have previously been made on isotopes in the Pt and Zr regions produced directly in nuclear reactions. As a result, these areas of particular nuclear structural interest have largely been left unexplored. In this Letter we describe a method with universal applicability, where an ion-guide takes the place of a conventional ion source. We present new data on the nuclear deformation and the odd-even staggering of the charge radius in the $^{170,172,173,174}\text{Hf}$ radionuclides. Although the half-lives are greater than 16 h for these isotopes, the speed of the ion extraction allows measurements on ions with half-lives as short as a millisecond; transmission efficiencies as high as 10% have been achieved for such isotopes [7]. Furthermore, the photon-tagged ion detection used in this work gives sensitivity to isotopes with low production rates.

Efficient secondary production of all chemical species can be obtained at ion-guide isotope separators [7]. Developed at the University of Jyväskylä, Finland, these

offer a fast, ~ 1 ms, and chemically independent method for producing singly charged ionic species from nuclear reaction products. The low emittance of such devices, required to achieve sufficient sensitivity for collinear laser spectroscopy, has been demonstrated in studies on the neutron-rich barium isotopes using proton-induced uranium fission [8]. A full description of the Jyväskylä IGISOL ion-guide isotope separator can be found in Ref. [7]. In the ion-guide technique, recoiling products from nuclear reactions in a target foil are stopped and thermalized in helium gas (~ 200 mbar). For this work (p, xn) reactions were favored because of the low recoil velocities and low proton-induced ionization of the buffer gas. The ions are then carried in the buffer gas and ejected through a small hole in the guide. With rapid gas flow, up to 10% of the thermalized products remain singly charged upon entering the jet expansion region. The ions are skimmed from the buffer gas by a shaped negative potential electrode and injected into the high vacuum of a conventional mass separator. The skimmer electrode is crucial in determining both the ion-guide efficiency and the energy spread of the extracted beam. An increase in extraction efficiency with skimmer voltage (operated at up to -500 V) is achieved at the cost of a large ion beam energy spread. However, a sharp local maximum in extraction efficiency occurs when the skimmer electrode is operated close to -10 V, giving up to 30% of the optimum efficiency and a low ion beam energy spread (~ 5 eV). This mode of guide operation, in which thermalized ions are drifted rather than electrostatically extracted from the source, was used exclusively during on-line laser investigations.

At the IGISOL facility, the extracted ion beam was accelerated to 40 keV and mass analyzed by a 55° dipole magnet. The selected beam was transported 15 m to the laser-ion beam interaction region, where a narrow (5 mm^2), low divergence (1 mrad half-angle) waist was

formed. An 18 mm length of the region where the ion beam and counterpropagating laser beam overlap was imaged onto a 16-fold segmented Hamamatsu R5900P-03-L16 photomultiplier. The interaction region was electrically insulated from the rest of the beam line and held at a controlled potential. Adjusting this potential allowed the ion beam to be Doppler tuned into resonance with the locked laser frequency. Downstream of the imaged region, the ions were deflected onto a microchannel plate. Only those photons detected in correct time coincidence with an ion were accepted [9]. Positional information on the origin of the fluorescent ion was used to narrow the photon-ion coincidence time window to 20(1) ns, a level corresponding to the optical resolution limit of the apparatus. The correction for time-of-flight differences across the imaged region provided a fivefold increase in sensitivity, as compared with uncorrected coincidence detection. An ionic resonance transition at 301 nm was studied in this work and a spatial resolution for the imaged ultraviolet light of 2–3 mm was achieved using a quartet of $f/0.5$ fused-silica lenses. The UV light was produced by intracavity frequency doubling with LiIO_3 in a Spectra Physics 380D dye laser. A UV power density of 0.7 mW mm^{-2} was required to provide optimum signal to background. Laser stabilization giving a linewidth of $\sim 3 \text{ MHz}$ was achieved by the Spectra Physics Stabilok system and active locking of the cavity to an absorption line of molecular iodine at $16\,579.2883 \text{ cm}^{-1}$.

Although the IGISOL technique provides a relatively “cold” method of production (with the bulk of the ion guide held typically at 300 K), two phenomena, associated with far higher temperatures, were observed. First, careful conditioning of the ion guide was required to reduce the hydrocarbon complexes and other contaminants observed in the produced beam. The level of principal contaminants, such as water, was reduced by baking *in situ*, but the level of others, presumably those from the hot target, could be reduced only by on-line conditioning for periods of the order of several days. Second, substantial metastable ion populations were observed from the ion guide. Many low lying metastable states exist in the hafnium ion, originating from d^2s , ds^2 , and d^3 electronic configurations. Under off-line conditions, resonance fluorescence originating from levels $\sim 3 \text{ eV}$ above the ground state was readily detected from a few pA of Hf^+ . The distribution of the population is nonthermal and sensitively dependent on the buffer gas pressure. The on-line pressure, 220 mbar, was selected to optimize total production efficiency with respect to contaminant levels (the contaminant levels increase at lower pressure due to the reduction in gas cooling of the guide). Optimum ion beam purity and intensity are the principal requirements for the coincidence technique and a loss of population from the ground state of $\sim 90\%$ was tolerated. Attempts to reduce the metastable populations using diatomic gases, N_2 and H_2 , and partial pressures of Ne (to quench possible col-

lisional excitation mechanisms involving metastable He atoms) were unsuccessful.

Prior to on-line running, the IGISOL system was optimized with naturally occurring ^{174}Hf produced from a glow discharge ion source [8]. Optimization of the reaction channel, and absolute IGISOL output, was then achieved on-line using reactions on a natural tantalum target. The $^{181}\text{Ta}(p, 2n)^{180m}\text{W}$ reaction provided a swift method of optimization using γ counting of collected activity. Investigation of the near-stable neutron-deficient hafnium isotopes used the $^{175}\text{Lu}(p, xn)^{176-x}\text{Hf}$ reaction (the ion-guide construction allows for two targets to be installed and for the guide to be rotated between them). The long half-lives of the hafnium isotopes prevented γ -counted yield calibration. With $5 \mu\text{A}$ of 20 MeV protons a beam flux of $3 \times 10^3 \text{ s}^{-1}$ was observed for ^{174}Hf , as estimated from the change in the $A = 170\text{--}180$ background beams between the Lu and Ta targets.

Proton beam energies of 20, 30, 40, and 55 MeV were used during the investigation. At 30 MeV isotopes of $A = 172, 173,$ and 174 were simultaneously produced, allowing their resonance positions to be measured under identical IGISOL conditions. A sample spectrum of the $ds^2 \ ^2D_{3/2}\text{--}dsp \ ^2D_{5/2}$ transition in ^{173}Hf is displayed in Fig. 1, clearly showing the two strongest components of the three-member multiplet. Evidence of the $A = 171$ structure was apparent at 55 MeV, with a large number of real counts forming a substantial peak in the timing spectrum. Insufficient statistics were obtained for a determination of hyperfine parameters or structure centroid. The $A = 171$ system is complicated by the production of a 30 s isomer, the existence of which was determined during yield tests at the IGISOL prior to this experiment [10].

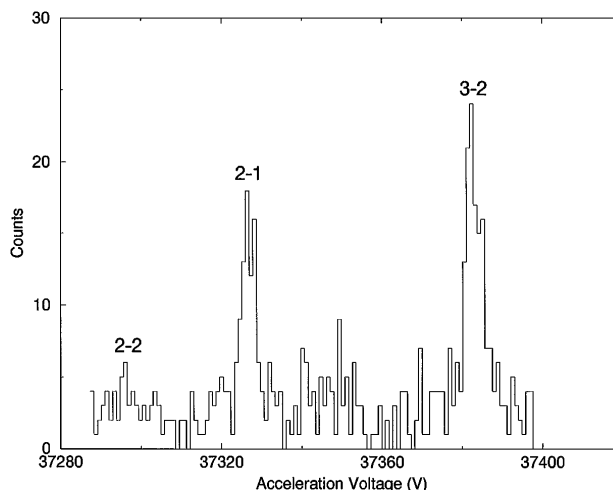


FIG. 1. Resonance fluorescence spectrum for the $ds^2 \ ^2D_{3/2}\text{--}dsp \ ^2D_{5/2}$ transition in ^{173}Hf . The isotope was produced in the $^{175}\text{Lu}(p, 3n)^{173}\text{Hf}$ reaction. The total angular momentum, F , of the upper and lower levels of the transition is indicated by each peak. The acceleration required to bring the ions to resonance with the frequency doubled laser is shown.

The resonance position of $A = 174$ was observed to change by ~ 3 V when the beam energy was initially changed from 20 to 30 MeV, presumably related to different plasma conditions within the guide. Following changes to higher beam energies, after ion-guide conditioning, no further drifting of resonance position with beam energy was observed. At fixed proton beam energy, the resonance position was observed to drift by ≤ 1 V in 10 h. The resonance positions recorded at 30 MeV primary beam energy provided resonance positions for the $A = 172, 173,$ and 174 isotopes with low systematic uncertainty and only the $A = 170$ hafnium result has been assigned the full ± 3 V error. An overall systematic uncertainty on the calibration of the separator voltage of 0.1% has been included in the isotope shift extraction.

The measured isotope shifts are shown in Table I. The magnetic moment of the $I^\pi = 1/2^-$ ground state of ^{173}Hf was determined to be $+0.502(7)\mu_N$ using hyperfine parameters scaled from $^{177,179}\text{Hf}$ and assuming no hyperfine anomaly (see Table II). This value is consistent with the systematics of neighboring spin-half isotones, $^{171}\text{Yb}(\frac{1}{2}^-) = +0.493\,67(1)\mu_N$, $^{169}\text{Er}(\frac{1}{2}^-) = +0.4850(2)\mu_N$ [15].

The isotope shift is, to an excellent approximation, composed of two linear terms, the mass shift and the field shift [1]. The field shift is directly proportional to the change in nuclear mean-square charge radius, $\delta\langle r^2 \rangle$, between the isotopes. Calibration of the atomic factors required for $\delta\langle r^2 \rangle$ evaluation must be attempted either by direct calculation or with reference to other optical and nonoptical data. Relative values of the field shift, and thus relative values of $\delta\langle r^2 \rangle$, are only weakly dependent on the atomic parameters and can often be extracted with high accuracy [1]. Evaluations of relative field shifts between the stable Hf isotopes are consistent for all reports [5,11,13,14,16,17] (on the whole afforded by identical assumptions on the purity of the chosen atomic reference line [11]). Reports of absolute values of $\delta\langle r^2 \rangle$ are how-

ever inconsistent. Two detailed evaluations of key atomic parameters (primarily, the $6s$ electron density at the nucleus, $|\psi(0)|_{6s}^2$) are available [11,14] and the $\delta\langle r^2 \rangle$ reports differ by $\sim 35\%$. The $\delta\langle r^2 \rangle$ evaluation in this work, presented in Table I, is compatible with, and follows, the former evaluation in Zimmermann *et al.* [11]. The alternative calibration, Aufmuth *et al.* [14], results in significantly lower absolute values of $\delta\langle r^2 \rangle$ which are in disagreement with nonoptical measurements [18,19] and invoke a $|\psi(0)|_{6s}^2$ substantially different from that which may be evaluated from the observed hyperfine splittings. Such discrepancy is common to other transition elements [1] and the level of complexity in the atomic structure of open d -shell elements may well preclude the use of semiempirical methods in $\delta\langle r^2 \rangle$ evaluation.

The results for the radioactive isotopes are complemented by measurements on natural Hf, evaluated by our collaboration in preparatory work at the Daresbury On-line Isotope Separator [20]. These measurements improve the statistical accuracy of the $A = 174, 177,$ and 179 results. The $\delta\langle r^2 \rangle$ calibration given is normalized to $\delta\langle r^2 \rangle^{178,180} = 0.098\text{ fm}^2$, with $0.098(13)\text{ fm}^2$ being the final result of the evaluation by Zimmermann *et al.* [11]. The assigned error includes all experimental uncertainty and the uncertainty arising from the allowance of a standard mass shift error in the reference line. No allowance is made for the absolute calibration of the results reported.

In Fig. 2 the measured $\delta\langle r^2 \rangle$ are plotted and compared with a droplet model estimate [21] that includes the predicted quadrupole, β_2 , deformation corrections from a global model [22]. The optical $\delta\langle r^2 \rangle$ are positioned relative to the experimental deformation of ^{178}Hf , $\beta_2 = 0.265(2)$, as evaluated from nuclear lifetime measurements [23]. Presentations made relative to the measured deformations of other isotopes bear close resemblance to one another with the predicted deformations of Ref. [22] appearing systematically higher than the experimental values (albeit at a level less than 5% of β_2).

A number of interesting features are apparent in Fig. 2. For the even hafnium isotopes, accepting the possible calibration error and offset of the experimental data, excellent agreement between theory and measurement is observed. The trend in the charge radius closely corresponds to that of the predicted β_2 ; the peaking of the deformation beyond the midshell, near $A = 173$, appears common to both the theoretical and measured deformation. Indeed the

TABLE I. Isotope shifts (IS) for the 301.29 nm hafnium ionic transition and evaluated charge radii. The charge radii are normalized to $\delta\langle r^2 \rangle^{178,180} = 0.098\text{ fm}^2$ after [11]. A 10% systematic normalization error is not included.

Mass A	IS (MHz) $\nu^{178} - \nu^A$	$\delta\langle r^2 \rangle^{178A}$ (fm ²)		Ref.
		This work	Other	
170	-10 180(30)	-0.466(7)		
172	-6632(10)	-0.302(5)	-0.313(13)	[4]
			-0.295(19)	[5]
173	-5017(7)	-0.228(4)		
174	-3704.3(31)	-0.168(3)		
175			-0.178(11)	[12]
176	-1868.1(20)	-0.084(2)		
177	-1337.4(18)	-0.061(1)		
178m2			-0.076(12)	[13]
179	790.8(19)	0.036(1)		
180	2149.2(15)	0.098(2)		
	2144(18)			[14]

TABLE II. Magnetic dipole and electric quadrupole hyperfine parameters, given in MHz.

	$^{173}\text{Hf}(\frac{1}{2}^-)$	$^{177}\text{Hf}(\frac{7}{2}^-)$	$^{179}\text{Hf}(\frac{9}{2}^+)$
$A(^2D_{3/2})$	+139(2)	+31.4(3)	-19.6(3)
$B(^2D_{3/2})$...	+1708(3)	+1937(3)
$A(^2D_{5/2})$	+262(4)	+58.8(2)	-37.3(2)
$B(^2D_{5/2})$...	+829(4)	+931(5)

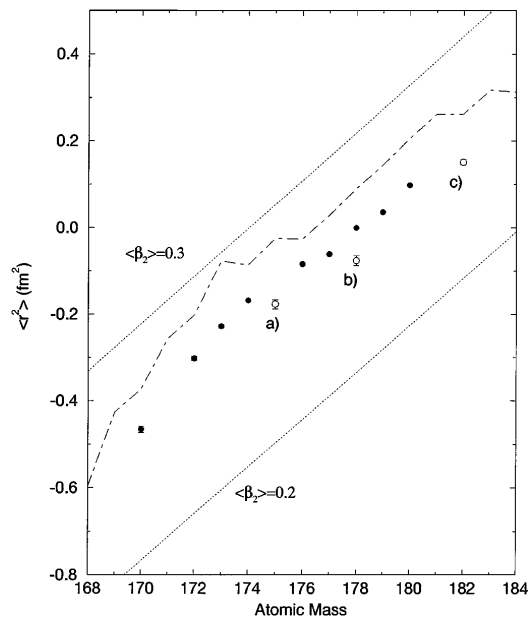


FIG. 2. The mean-square charge radii in the hafnium isotope chain, normalized to ^{178}Hf . The data include radioactive measurements reported for (a) $A = 175$ [12], (b) $A = 178^{m2}(16^+)$ [13], and (c) $A = 182$ [17], all others being from this work. The droplet model calculation [21] including β_2 deformation [22] (dot-dashed line) is offset from optical measurements which are taken relative to the experimental deformation of ^{178}Hf ground state [23]. The data are shown bounded by $\beta_2 = 0.30$ and $\beta_2 = 0.20$ isodeformation lines.

agreement here far exceeds that of other regions. This may be attributed to the large static deformations and the well-defined symmetry axis of the hafnium nuclei, which allows the relation between deformation and $\delta\langle r^2 \rangle$ to be reliably made by classical geometry. The agreement for the even- N isotopes extends to other measures of nuclear deformation, in particular, nuclear lifetime measurements [23].

The mean-square charge radius is, however, also modified by higher-order effects such as hexadecapole deformation, nonaxial shapes, and dynamical effects. Reasonable estimates may be formed for many of these effects, suggesting that they are relatively small. However, no method exists for quantifying the role of pairing or the nuclear rigidity. The effects of changes in "rigidity" parameters are clear in the course of the odd- N nuclear radii. The charge radii of these nuclei imply nuclear deformations that, although reasonably consistent with the intrinsic deformations inferred from their spectroscopic quadrupole moments (afforded only by large errors in the case of $A = 175$ [12]), are significantly lower than the droplet model prediction. In the model, no account has been made for the odd-even staggering (OES) of the charge radius, which is seen in all isotope chains [2]. The

absence of OES at $A = 173$ strongly suggests that static deformation changes dominate in determining the course of the charge radius. The persistent overestimation of the odd- N charge radii however clearly implies that a sizable stagger still underlies these changes.

Investigation of the OES and the general trend of $\delta\langle r^2 \rangle$ in the hafnium chain requires further experimental measurements. The striking negative isomer shift for the four-quasiparticle $^{178m2}\text{Hf } 16^+$ isomer [13] suggests that good quantitative data on the role of nucleon pairing will come from the extension of these measurements to high- K isomeric systems (abundant in the near-stability region).

Two of the authors (J.M.G.L., D.H.F.) acknowledge financial support provided by the University of Manchester and the University of Birmingham. This work has been financially supported by the U.K. Engineering & Physical Sciences Research Council. Further support has been provided by the Access to Large Scale Facility program under the Training and Mobility of Researchers program of the European Union.

- [1] W.H. King, *Isotope Shifts in Atomic Spectra* (Plenum Press, London, 1984).
- [2] E.W. Otten, *Treatise on Heavy-Ion Science* (Plenum Press, New York, 1988), Vol. 8, p. 517.
- [3] U. Krönert *et al.*, Nucl. Instrum. Methods Phys. Res., Sect. A **300**, 522 (1991).
- [4] J. Rink *et al.*, Z. Phys. A **342**, 487 (1992).
- [5] W.Z. Zhao *et al.*, Hyperfine Interact. **108**, 483 (1997).
- [6] M. Wakasugi *et al.*, Rev. Sci. Instrum. **64**, 3487 (1993).
- [7] H. Penttillä *et al.*, Nucl. Instrum. Methods Phys. Res., Sect. B **126**, 213 (1997).
- [8] J. Billowes *et al.*, Nucl. Instrum. Methods Phys. Res., Sect. B **126**, 416 (1997).
- [9] D.A. Eastham *et al.*, Opt. Commun. **60**, 293 (1986).
- [10] P. Campbell *et al.*, J. Phys. G **23**, 1141 (1997).
- [11] D. Zimmermann *et al.*, Phys. Rev. A **50**, 1112 (1994).
- [12] W.G. Jin *et al.*, Phys. Rev. C **55**, 1545 (1997).
- [13] N. Boos *et al.*, Phys. Rev. Lett. **72**, 2689 (1994).
- [14] P. Aufmuth *et al.*, Z. Phys. D **18**, 107 (1991).
- [15] P. Raghavan, At. Data Nucl. Data Tables **42**, 189 (1989).
- [16] J. Schecker *et al.*, Phys. Rev. A **46**, 3730 (1992).
- [17] A. Anastassov *et al.*, Z. Phys. A **348**, 177 (1994).
- [18] S.K. Bhattacharjee, F. Boehm, and P.L. Lee, Phys. Rev. **188**, 1919 (1969).
- [19] Y. Tanaka *et al.*, Phys. Rev. C **30**, 350 (1984).
- [20] T.G. Cooper, Ph.D. thesis, University of Manchester, United Kingdom, 1995.
- [21] W.D. Myers and K.H. Schmidt, Nucl. Phys. **A410**, 61 (1983).
- [22] P. Möller *et al.*, At. Data Nucl. Data Tables **59**, 185 (1995).
- [23] S. Raman *et al.*, At. Data Nucl. Data Tables **36**, 1 (1987).