# Schottky mass measurements of heavy neutron-rich nuclides in the element range 70 < Z < 79 at the ESR

D. Shubina, <sup>1, 2, 3</sup> R.B. Cakirli, <sup>1, 4</sup> Yu.A. Litvinov, <sup>1, 3</sup> K. Blaum, <sup>1</sup> C. Brandau, <sup>3, 5</sup> F. Bosch, <sup>3</sup> J.J. Carroll, <sup>6</sup> R.F. Casten, <sup>7</sup> D.M. Cullen, <sup>8</sup> I.J. Cullen, <sup>9</sup> A.Y. Deo, <sup>9</sup> B. Detwiler, <sup>10</sup> C. Dimopoulou, <sup>3</sup> F. Farinon, <sup>3</sup> H. Geissel, <sup>3, 11</sup> E. Haettner, <sup>11</sup> M. Heil, <sup>3</sup> R.S. Kempley, <sup>9</sup> C. Kozhuharov, <sup>3</sup> R. Knöbel, <sup>3</sup> J. Kurcewicz, <sup>3</sup> N. Kuzminchuk, <sup>11</sup> S.A. Litvinov, <sup>3</sup> Z. Liu, <sup>12</sup> R. Mao, <sup>13</sup> C. Nociforo, <sup>3</sup> F. Nolden, <sup>3</sup> Z. Patyk, <sup>14</sup> W.R. Plass, <sup>11</sup> A. Prochazka, <sup>3</sup> M.W. Reed, <sup>9, 15</sup> M.S. Sanjari, <sup>3, 16</sup> C. Scheidenberger, <sup>3, 11</sup> M. Steck, <sup>3</sup> Th. Stöhlker, <sup>3, 17, 18</sup> B. Sun, <sup>11, 19</sup> T.P.D. Swan, <sup>9</sup> G. Trees, <sup>10</sup> P.M. Walker, <sup>9, 20</sup> H. Weick, <sup>3</sup> N. Winckler, <sup>1, 3</sup> M. Winkler, <sup>3</sup> P.J. Woods, <sup>12</sup> T. Yamaguchi, <sup>21</sup> and C. Zhou<sup>7</sup>

<sup>1</sup>Max-Planck-Institut für Kernphysik, Saupfercheckweg 1, 69117 Heidelberg, Germany <sup>2</sup>Fakultät für Physik und Astronomie, Universität Heidelberg, Philosophenweg 12, 69120 Heidelberg, Germany <sup>3</sup>GSI Helmholtzzentrum für Schwerionenforschung, Planckstraße 1, 64291 Darmstadt, Germany <sup>4</sup>Department of Physics, University of Istanbul, Istanbul, Turkey <sup>5</sup>ExtreMe Matter Institute EMMI, GSI Helmholtzzentrum für Schwerionenforschung, 64291 Darmstadt, Germany <sup>6</sup>US Army Research Laboratory, 2800 Powder Mill Road, Adelphi MD, USA <sup>7</sup>Wright Nuclear Structure Laboratory, Yale University, New Haven, Connecticut 06520, USA <sup>8</sup>Schuster Laboratory, University of Manchester, Manchester M13 9PL, United Kingdom <sup>9</sup>Department of Physics, University of Surrey, Guildford, Surrey GU2 7XH, United Kingdom <sup>10</sup> Youngstown State University, One University Plaza, Youngstown, Ohio 44555, USA <sup>11</sup>II Physikalisches Institut, Justus-Liebig-Universität Gießen, 35392 Gießen, Germany <sup>12</sup>School of Physics and Astronomy, University of Edinburgh, Edinburgh EH9 3JZ, United Kingdom <sup>13</sup>Institute of Modern Physics, Chinese Academy of Sciences, Lanzhou 730000, People's Republic of China <sup>14</sup>National Centre for Nuclear Research, PL-00681 Warsaw, Poland <sup>15</sup>Department of Nuclear Physics, R.S.P.E., Australian National University, Canberra ACT 0200, Australia <sup>16</sup>Goethe-Universität Frankfurt, 60438 Frankfurt, Germany  $^{17}$ Friedrich-Schiller-Universität Jena, 07737 Jena, Germany <sup>18</sup>Helmholtz-Institut Jena, 07743 Jena, Germany <sup>19</sup>School of Physics and Nuclear Energy Engineering, Beihang University, 100191 Beijing, PRC <sup>20</sup>CERN, CH-1211 Geneva 23, Switzerland <sup>21</sup>Graduate School of Science and Engineering, Saitama University, Saitama 338-8570, Japan

Storage-ring mass spectrometry was applied to neutron-rich  $^{197}$ Au projectile fragments. Masses of  $^{181,183}$ Lu,  $^{185,186}$ Hf,  $^{187,188}$ Ta,  $^{191}$ W, and  $^{192,193}$ Re nuclei were measured for the first time. The uncertainty of previously known masses of  $^{189,190}$ W and  $^{195}$ Os nuclei was improved. Observed irregularities on the smooth two-neutron separation energies for Hf and W isotopes are linked to the collectivity phenomena in the corresponding nuclei.

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#### I. INTRODUCTION

The binding energy of the nucleus, or its mass, contains information about interactions between its constituent protons and neutrons. Precision mass data as well as separation energies extracted from them can unveil much about the underlying nuclear structure [1, 2]. For instance, magic numbers can easily be seen from separation energies which reveal a huge drop (depending on the size of the closed shell), or a jump or a gap (depending on the way the separation energy is constructed) after a magic number [3]. In addition, separation energies can reflect collective effects as we will discuss below. Such data are therefore valuable in understanding the underlying shell structure in nuclei and the evolution of collective effects. and are therefore essential to provide the basis for the development and testing of a comprehensive theory of nuclei. In turn, a reliable nuclear theory is of utmost importance for calculating the properties of unknown nuclei, which are needed, for example, in the modelling of the rapid neutron capture process (r-process) of nucleosynthesis in stars [4]. Therefore, new data on neutronrich heavy nuclei are essential.

However, such data are scarce mainly due to the complexity of producing these exotic nuclei. This becomes evident if one glances at the chart of nuclides (see e.g. Ref. [5]) where for elements  $Z \sim 70-80$  the number of observed neutron-rich nuclides is very limited. Very recently, several tens of new isotopes were discovered in projectile fragmentation of uranium beams, though no spectroscopic information could yet be obtained for these nuclei [6]. Their production rates are tiny, which requires very efficient measurement techniques. One such technique for mass measurements is storage-ring mass spectrometry [7].

In this paper we report on direct mass measurements of neutron-rich nuclei in the element range from lutetium to osmium. Masses for nine nuclei were measured for the first time, and for three nuclei the mass uncertainty was improved. It is known that nuclear collective effects can be seen in the behavior of nucleon separation energies [8]. Here, we investigate the relation between rather subtle effects in two-neutron separation energies,  $S_{2n}$ , and changes in both collectivity and neutron number. Observed irregularities in the smooth two-neutron separation energies for Hf and W isotopes are linked to changes in collective observables. The importance of the number of valence nucleons is discussed in the context of collective contributions to binding energies calculated with the IBA model [9].

## II. EXPERIMENT

The experiment was conducted at GSI Helmholtzzentrum für Schwerionenforschung in Darmstadt. Heavy neutron-rich nuclei of interest were produced in projectile fragmentation of <sup>197</sup>Au primary beams. The experiment described here was part of a larger experimental campaign, some results of which are described in Refs. [10–13]. The <sup>197</sup>Au beams were accelerated to the energy of 11.4 MeV/u in the linear accelerator UNILAC and then injected into the heavy ion synchrotron SIS-18 [14], where they were further accelerated to an energy of 469.35 MeV/u. The  $^{197}$ Au $^{65+}$  beams were fast extracted (within about 1  $\mu$ s) and focused on a production target located at the entrance of the fragment separator FRS [15, 16]. As target we used 1036 mg/cm<sup>2</sup> thick <sup>9</sup>Be with a 221 mg/cm<sup>2</sup> Nb backing for more efficient electron stripping. The reaction products emerged from the target as highly-charged ions having mostly 0, 1, or 2 bound electrons. The nuclides of interest were transported through the FRS, being operated as a pure magnetic rigidity  $(B\rho)$  analyzer [16], and injected into the cooler-storage ring ESR [17]. The transmission through the FRS and the injection into the ESR were optimized with the primary beam, and the magnetic setting of FRS-ESR was fixed at  $B\rho = 7.9$  Tm throughout the entire experiment. All ion species within the acceptance of the FRS-ESR of about  $\pm 0.2\%$  were injected and stored. Only 25% of the ESR acceptance is filled at the injection. We note, that in contrast to the settings described in Refs. [10, 11], in this experiment no energy-loss degraders were employed in the FRS.

The relationship between relative revolution frequencies (f), relative mass-over-charge ratios (m/q) and velocities (v) of the particles stored in a ring is given by [7, 18-20]:

$$\frac{\Delta f}{f} = -\alpha_p \frac{\Delta_{\frac{m}{q}}}{\frac{m}{q}} + (1 - \alpha_p \gamma^2) \frac{\Delta v}{v}, \tag{1}$$

where  $\gamma$  is the relativistic Lorentz factor,  $\alpha_p$  is the momentum compaction factor, which characterizes the relative variation of the orbit length of stored particles per relative variation of their magnetic rigidity (for more details see Refs. [7, 18–20]). For the ESR,  $\alpha_p$  is nearly constant for the entire revolution frequency acceptance and

is  $\alpha_p \approx 0.179$ . From Eq. (1) it becomes obvious that the revolution frequency is a measure of the mass-over-charge ratios of the stored ions provided that the second term on the right hand side, containing the velocity spread  $(\Delta v/v)$ , can be eliminated. The latter is achieved by applying electron cooling [21]. For this purpose the stored ions are merged over a length of about 2.5 m with a continuous beam of electrons in the electron cooler device. The mean circumference of the ESR is 108.4 m and at our energies the ions circulate with a frequency of about 2 MHz passing the electron cooler at each revolution. The energy of the electrons is very accurately defined by the applied acceleration potential. Within a few seconds the mean velocity of the ions becomes equal to the mean velocity of the electrons. The velocity spread of the stored ions, which is  $\Delta v/v \approx 4 \cdot 10^{-3}$  at the injection, is thereby reduced to  $\Delta v/v \approx 10^{-7}$  [21].

The Schottky mass spectrometry (SMS) technique has been applied to the electron cooled ions [7, 19]. In this technique, every stored highly-charged ion at each revolution in the ESR induces mirror charges on a couple of parallel electrostatic copper plates, the Schottky pick-up installed inside the ring aperture. The noise from the pick-up, which is dominated by the thermal noise, is amplified by a broad-band low-noise amplifier [22]. In the present experiment, we analyzed the noise power at about 60 MHz, corresponding to the  $30^{th}$  harmonic of the revolution frequency of the stored ions. The pick-up signal was down-mixed using a ~60 MHz reference frequency from an external frequency generator. The acceptance of the ESR at the  $30^{th}$  harmonic corresponds to about 320 kHz [23, 24]. Therefore, to cover the entire ESR acceptance we digitized the signal with a sampling frequency of 640 kHz using a commercial 16-bit ADC [25]. A Fourier transform of the digitized data yielded the noisepower density spectrum, or the Schottky frequency spectrum [7, 24].

New ions were injected every few minutes. At the injection into the ESR, the previously stored ions were removed. Several Schottky frequency spectra were created for each injection. The parameters of the Fourier transform algorithm were optimized offline. A frequency resolution of 4.77 Hz/channel was chosen, which corresponds to the time resolution of 0.21 s per spectrum. Furthermore, every 50 consecutive Schottky spectra were averaged to enhance the signal-to-noise ratio. Thus, Schottky spectra integrated over 10 s were produced. The latter means that several independent subsequent frequency spectra were obtained for each injection of the ions into the ESR.

The electron cooling forces the ions to the same mean velocity thus filling the entire acceptance of the ESR of  $\Delta B \rho/B \rho \sim \pm 1.5\%$  (see Ref. [19]). Since  $B \rho = m v \gamma/q$ , by changing the velocity of the electrons in different injections, ions with different m/q can be studied. In the present experiment we varied the electron cooler voltage in the range from 204 kV to 218 kV. On average eight injections were recorded for each cooler setting. In order

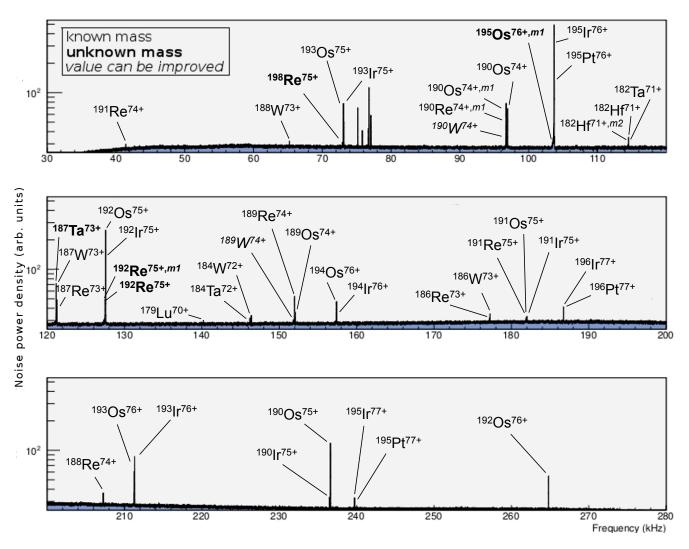


FIG. 1: Example of a Schottky frequency spectrum with the corresponding isotope identification. This spectrum is combined from eight independent injections acquired for the electron cooler voltage  $U_c = 209$  kV. Nuclides with known and previously unknown masses are indicated by different fonts (see legend).

to facilitate the assignment of the revolution frequencies with the corresponding isotope identification, all spectra within each cooler setting were combined together. In this case the maximum number of ion species present in each setting can be used for the identification. The latter is done based on Eq. (1). As a starting point for the identification we used the frequency of the stored <sup>197</sup>Au<sup>76+</sup> primary ions. An example of the combined Schottky frequency spectrum for an electron cooler voltage of  $U_c = 209 \text{ kV}$  is illustrated in Fig. 1. Fig. 2 shows a zoom on a quadruplet of lines of A = 190 isobars present in ground and/or isomeric states. The latter are indicated with a label m. The peak finding and the isotope identification were done automatically with a dedicated ROOT-based [26] software [27]. The nuclides observed in this experiment are illustrated on the chart of nuclides in Fig. 3 together with the nuclides in the ground and isomeric states identified in the other part of this experiment (see Refs. [10–13]).

## III. DATA ANALYSIS AND RESULTS

In order to determine the unknown mass-over-charge ratios, the Schottky frequency spectra have to be calibrated [19, 24]. For this purpose we selected the nuclides which were identified in our spectra and for which masses are known experimentally according to the Atomic-Mass Evaluation 2003 (AME) [28]. We note that an update of the AME was made available in 2011 [29], which however contains no new information in the mass region studied here. The data of the present work were already included in the latest AME published very recently [30]. Furthermore we required that the reference masses were obtained by more than one independent measurement technique and that there must exist no other ionic species with

TABLE I: Nuclides with accurately known masses used as references to calibrate Schottky frequency spectra. Listed are the proton (Z) and mass (A) numbers, the number of experimental settings  $(N_{set})$  in which this reference mass was observed, literature mass excess values from the Atomic-Mass Evaluation [28]  $(ME_{AME})$  as well as the re-determined mass excess values (ME) (see text) with the corresponding  $\sigma_{stat}$  uncertainty and its difference to the literature value  $(\delta = ME - ME_{AME})$ . Note that the systematic uncertainty of  $\sigma_{syst} = 38$  keV (see text) is not added here.

$\overline{z}$	A	$N_{set}$	$ME_{ m AME}$	ME	δ
		000	(keV)	(keV)	(keV)
72	181	1	-47412(2)	-47412(40)	0(40)
73	181	1	-48442(2)	-48383(40)	59(40)
73	182	3	-46433(2)	-46466(29)	-32(29)
73	183	3	-45296(2)	-45276(16)	20(16)
73	185	6	-41396(14)	-41350(14)	46(20)
74	184	5	-45707(1)	-45663(17)	44(17)
74	186	7	-42510(2)	-42493(12)	17(13)
74	187	8	-39905(2)	-39863(8)	41(8)
75	189	9	-37978(8)	-38063(10)	-85(13)
75	191	9	-34349(10)	-34364(3)	-15(11)
76	188	7	-41136(1)	-41115(12)	21(12)
76	190	7	-38706(2)	-38637(15)	69(15)
76	192	7	-35881(3)	-35833(8)	48(8)
76	193	7	-33393(3)	-33329(8)	63(8)
77	191	1	-36706(2)	-36650(71)	56(71)
78	194	4	-34763(1)	-34779(24)	-16(24)
78	196	9	-32647(1)	-32655(4)	-7(4)
79	196	5	-31140(3)	-31126(4)	14(5)

a close mass-to-charge ratio which could simultaneously be stored in the ESR. Also the peaks corresponding to long-lived isomeric states (we observed  $^{182}\mathrm{Hf^{m1}},\,^{186}\mathrm{W^{m2}},\,^{190}\mathrm{Os^{m1}}$  and  $^{190}\mathrm{Ir^{m2}}$  isomers) were not used for calibration. The list of reference masses is given in Table I.

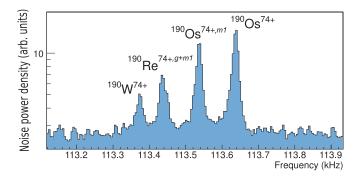


FIG. 2: A zoom of the Schottky frequency spectrum illustrated in Fig. 1 on a quadruplet of A=190 isobars. The peaks corresponding to nuclides in isomeric states are labeled with m.

The momentum compaction factor  $\alpha_p$ , although nearly constant, is a complicated function of the revolution frequency f. If  $\alpha_p$  were exactly constant, then the m/qwould linearly depend on f. Fig. 4 (top) shows an example of a linear fit through the calibration m/q-values for one of the measured spectra. The residuals of the fit (bottom panel of the same figure) clearly show that the calibration function is more complicated than a low-order polynomial function. Polynomials of up to 4th order (5 free coefficients) were employed, but different compared to the analyses performed in Refs. [19, 24, 31], the quality of the fits was found unacceptable. A possible reason for the latter is the small number of reference masses in individual 10-s Schottky spectra. Furthermore, due to time variations of the storage ring and electron cooler parameters, such as, e.g., their magnetic fields, it is not possible to establish a universal calibration curve. Therefore, we employed an analysis procedure in which we used linear splines to approximate the calibration curve in each individual spectrum.

Changes of the electron cooler voltage were done in steps of 0.5 kV so that for adjacent cooler settings the measured frequency spectra have a significant overlap. Furthermore, the same nuclide can be present in different charge states, which allows for a redundant analysis. The nuclides whose masses have been measured for the first time or which mass accuracy was improved in this work are listed in Table II.

Since the calibration curve is not known exactly and is approximated with linear splines, and since the number of calibration points in each spectrum is small, there is inevitably a systematic error introduced by the analysis method. Ways to estimate systematic uncertainty have been described in our previous works [19, 24, 31]. For this purpose in the present work, we re-determined the mass of each reference nuclide. This was done consecutively by setting each of the references as "no"-reference and obtaining its mass from the remaining 17 references. The re-determined mass excess values are listed in Table I along with their literature values [28]. The systematic error  $\sigma_{syst}$  has been obtained from solving the following equation:

$$\sum_{i=1}^{N_{ref}} \frac{(ME_{AME}^{(i)} - ME^{(i)})^2}{\sigma_{AME(i)}^2 + \sigma_{(i)}^2 + \sigma_{syst}^2} = N_{ref},$$
 (2)

where  $N_{ref}=18$  is the number of reference nuclides,  $ME^{(i)}$  ( $\sigma_{(i)}$ ) and  $ME^{(i)}_{AME}$  ( $\sigma_{AME(i)}$ ) are the re-calculated and literature mass excess values (statistical uncertainties) of the *i*-th reference nuclide, respectively. The systematic uncertainty of the present analysis amounts to  $\sigma_{syst}=38$  keV. The final uncertainties listed in Table II were obtained from a quadratic sum of the systematic and statistical uncertainties.

We note, that in contrast to Ref. [31] we do not observe any significant systematic dependence of the recalculated mass values versus their proton number and correspondingly do not reduce the systematic errors. A

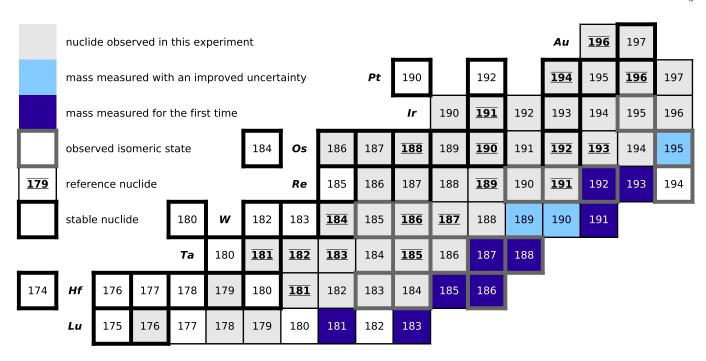
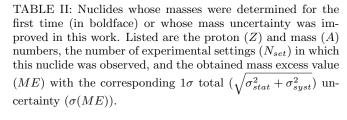


FIG. 3: (Color online) A part of the chart of nuclides indicating the nuclides measured in this work as well as the nuclides in the ground and isomeric states identified in the other part of this experiment devoted to the search for new K-isomers in this region [10, 11].

dedicated study should be performed to investigate the origin of this inconsistency.



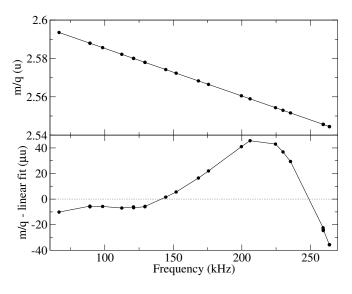


FIG. 4: Top: Mass-over-charge ratio m/q as a function of revolution frequency. The solid line illustrates a straight line fit through the calibration m/q-values. Bottom: The same as top but with the subtracted linear fit.

$\mathbf{Z}$	A	$N_{set}$	ME	$\sigma(ME)$
			(keV)	(keV)
<b>71</b> 1	181	1	-44797	126
<b>71</b> 1	183	1	-39716	80
<b>72</b> 1	185	1	-38320	64
<b>72</b> 1	186	1	-36424	51
<b>73</b> 1	187	2	-36896	56
<b>73</b> 1	188	<b>2</b>	-33612	55
74	189	5	-35618	40
74	190	7	-34388	41
<b>74</b> 1	191	1	-31176	<b>42</b>
<b>75</b> 1	192	1	-31589	71
<b>75</b> 1	193	7	-30232	39
76	195	1	-29512	56

#### IV. DISCUSSION

The new masses allow us to obtain interesting information on nuclear structure. Fig. 5 (left, middle) shows two-neutron separation energies  $(S_{2n})$  as a function of neutron number for Z=66-78 in the  $A\sim180$  region. Fig. 5 (left) is for even proton numbers while Fig. 5 (middle) is for odd proton numbers. The new  $S_{2n}$  values, for  $^{181,183}$ Lu,  $^{185,186}$ Hf,  $^{187,188}$ Ta,  $^{191}$ W,  $^{192,193}$ Re, calculated from the masses measured in this study, are marked in red color. For known masses whose values were improved in this experiment,  $^{189,190}$ W and  $^{195}$ Os, the literature  $S_{2n}$  values are illustrated with black color.

By inspecting Fig. 5 (left), one can notice, that in the  $S_{2n}$  values of the even-Z nuclei a flattening in Yb, Hf and W is seen at almost the last neutron numbers experimentally known. The flattening in  $S_{2n}(W)$ , using the improved W masses, is confirmed and  $S_{2n}$  at N=117 continues with the same behavior. In contrast to Hf and W, the new  $S_{2n}(Os)$  point, which is a bit lower at N=119 than in previous measurements, shows no flattening.

Fig. 5 (middle) with the new measured masses does not show similar effects in  $S_{2n}$  as in Fig. 5 (left). However, there is a small change in slope (a more rapid fall-off) at N=110 compared to the lower-N trend in  $S_{2n}(_{71}\text{Lu})$ ,  $S_{2n}(_{73}\text{Ta})$  and  $S_{2n}(_{75}\text{Re})$  (we also see this drop for  $_{72}\text{Hf}$  and  $_{74}\text{W}$  in Fig. 5 (left)), and maybe at N=115 in  $S_{2n}(_{73}\text{Ta})$  as well. It is highly desirable to have more odd-Z mass measurements in this region for a comparison with even-Z where more data are available. Thus, we concentrate below on discussing even-Z nuclei.

Fig. 5 (right) shows the energy of the first excited  $2^+$  states against neutron number for Z=70-78 in the  $A\sim 180$  region. This important, simple, observable has a high energy (can be a few MeV) at magic numbers where nuclei are spherical and very low energies (less than  $100~{\rm keV}$  for well deformed heavy nuclei) near mid-shell. The  $E(2_1^+)$  values usually decrease smoothly between the beginning and middle of a shell except when there is a sudden change in structure. Since  $N\sim 104$  is mid-shell for the nuclei illustrated in Fig. 5,  $E(2_1^+)$  has a minimum at or close to N=104. After the mid-shell, the energy increases towards the N=126 magic number.

Let us now focus on the W-Pt nuclei in Fig. 5 and compare the behavior of  $E(2_1^+)$  and  $S_{2n}$ . In particular, we look at  $S_{2n}$  in isotopes where  $E(2_1^+)$  changes rapidly, indicating a sudden change in structure. In W,  $E(2_1^+)$  increases from N=114 to 116 by a considerably larger amount compared to the other W isotopes (see Ref. [33]). This jump signals a structural change from approximately constant deformation to decreasing deformation at N=116 (after N=114). Note that this neutron number is exactly where  $S_{2n}$  exhibits flattening.

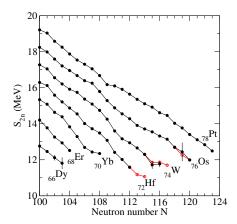
Similar to W, Os at N=120 (after N=118) has a jump in  $E(2_1^+)$ . However,  $S_{2n}(^{196}\mathrm{Os})$  does not reveal an obvious change at the same neutron number. At first glance, this seems inconsistent with the interpretation explained for W above but, in fact, there might be an ex-

planation for this different behavior which would provide additional insight into the relation of binding to collectivity.

Reference [8] showed the structural sensitivity of calculated collective contributions to binding. In addition Ref. [8] stressed that collective binding is very sensitive to the number of valence nucleons. Calculated collective contributions to binding using the IBA-1 model for boson numbers  $N_B = 5$  (left) and  $N_B = 16$  (right) are illustrated in the symmetry triangle [8] in Fig. 6. The three corners of the triangle describe three dynamical symmetries, U(5) (vibrator), SU(3) (rotor) and O(6) ( $\gamma$ -soft) (for more details, see Ref. [34]). The color code in Fig. 6 changes from yellow to red when the collective effects increase. Needless to say, nuclei have more valence particles (so boson numbers) around the SU(3) corner than the U(5) (and also O(6)) corner. One sees that the collective binding energy (B.E.) rapidly increases for nuclei with axial deformation, that is, near SU(3). Note that the triangles are presented for fixed boson numbers. In both, the color scale is kept the same to point out that the collective B.E.s are larger in  $N_B=16$  than 5. As shown in Fig. 4 of Ref. [8], the collective binding energies vary approximately as the square of the number of valence nucleons in the context of IBA calculations. Therefore, for a lower number of valence nucleons, Fig. 6 shows similar trends for both  $N_B = 5$  and  $N_B = 16$ , but the overall binding is considerably less (compare (left) and (right) of Fig. 6). We now suggest that the behavior of  $E(2_1^+)$  and  $S_{2n}$  in Fig. 5 can be understood in terms of this dual dependence of binding on collectivity and valence nucleon

If  $^{190}$ W and  $^{196}$ Os are mapped in the symmetry triangle,  $^{190}$ W will likely be closer to the SU(3) corner than  $^{196}$ Os [35]. One of the ways to understand this is from the P-factor [36], defined as  $P=N_p\cdot N_n/(N_p+N_n)$ , where  $N_p$  denotes the number of valence protons (proton holes) and  $N_n$  the number of valence neutrons (neutron holes). Thus P is a quantity that can provide a guide to structure. For example, the onset of deformation in heavy nuclei corresponds to the P-factor around 4 and 5. Generally, if P is larger than 3, collective effects increase. That is, nuclei become deformed and approach closer to the SU(3) corner.

Fig. 7 shows color-coded values for the P-factor for the Z=50-82, N=82-126 region, and indicates the P-factors for the nuclei relevant to this discussion. <sup>190</sup>W has 8 valence protons and 10 valence neutrons while <sup>196</sup>Os has  $N_p=6$  and  $N_n=6$ . Correspondingly, these nuclei have P-factors of 4.4 and 3, respectively. The greater collectivity of <sup>190</sup>W compared to <sup>196</sup>Os suggested by their P-factors is reflected in its lower  $2_1^+$  energies as seen in Fig. 5 (right). Thus, for two reasons – both greater collectivity and more valence nucleons – the collective binding should be much greater in <sup>190</sup>W than in <sup>196</sup>Os and changes in binding energies ( $S_{2n-coll}$  values) should be on a larger scale. We suggest that this accounts for the fact that we see a flattening in <sup>190</sup>W clearly but not in



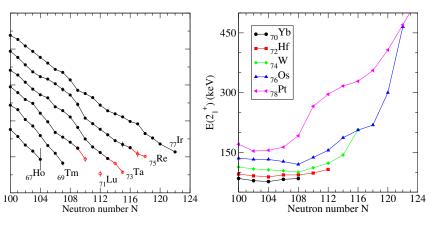


FIG. 5: (Color online) Data for the  $A \sim 180$  region, two-neutron separation energies [28] as a function of neutron number from N = 100 to N = 124 for even-Z (left) from Dy to Pt and odd-Z (middle) from Ho to Re. The new  $S_{2n}$  values obtained from this work are shown in red color while the literature  $S_{2n}$  values are shown in black color. Right: Energies of the first excited  $2^+$  states [32] for the same even-even nuclei as in the left panel.

 $^{196}$ Os in Fig. 5 (left). Obviously, it is very important to have new data, both masses and spectroscopic information, on even more neutron rich W isotopes although such experiments are difficult. Even the mass of  $^{192}$ W alone would be telling since the trend in  $E(2_1^+)$  is quite clear already.

Existing data on Pt nicely illustrate and support these ideas. Fig. 5 (right) shows two jumps in  $E(2_1^+)$  for Pt, around  $N \sim 110$  and  $N \sim 118-120$ . Looking at  $S_{2n}$  for Pt, there is a kink near  $N \sim 110$  but a smooth behavior near  $N \sim 118$ . For <sup>188</sup>Pt,  $N_p$  is 4 and  $N_n$  is 16 so the P-factor is 3.2. This isotope, with 20 valence nucleons, is relatively collective and once again one sees an anomaly in  $S_{2n}$  as well. In contrast, for <sup>198</sup>Pt<sub>120</sub> with only 10 valence nucleons, and a P-factor of only 2.4, the lower collectivity (seen in the much higher  $2^+$  energy) and the lower number of valence nucleons are such that  $S_{2n}$  shows no anomaly, but rather a nearly straight behavior.

This qualitative interpretation is supported by collective model calculations. A thorough and detailed study of this or any transition region requires a very careful and

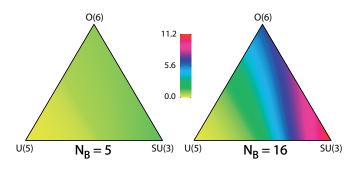


FIG. 6: (Color online) The symmetry triangle of the IBA showing the three dynamical symmetries at the vertices. The colors indicate calculated collective contributions in MeV to binding energies for  $N_B=5$  (left) and  $N_B=16$  (right). A similar triangle for  $N_B=16$  was presented in Ref. [8].

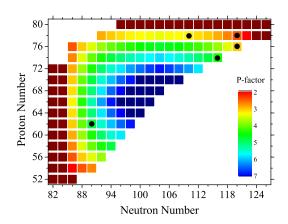


FIG. 7: (Color online) P-factor values illustrated with a color code for even-even nuclei in the Z=50-82 and N=82-126 shells. Black points marked are for the key nuclei discussed, namely,  $^{190}$ W,  $^{196}$ Os,  $^{188}$ Pt,  $^{198}$ Pt, and  $^{152}$ Sm.

systematic assessment of all the data on energies, transition rates, and binding energies, the choice for the specific terms to include in the Hamiltonian and the optimum approach to fitting the data. We are undertaking such a study and will present the results in a future publication [37]. Nevertheless, it is useful to present an example of the model results here to validate the ideas presented above. To this end, we have carried out a schematic set of IBA calculations using the Hamiltonian [38, 39]

$$H = \epsilon \hat{n}_d - \kappa Q \cdot Q \tag{3}$$

where Q is a quadrupolar operator

$$Q = (s^{\dagger} \tilde{d} + d^{\dagger} s) + \chi (d^{\dagger} \tilde{d})^{(2)}. \tag{4}$$

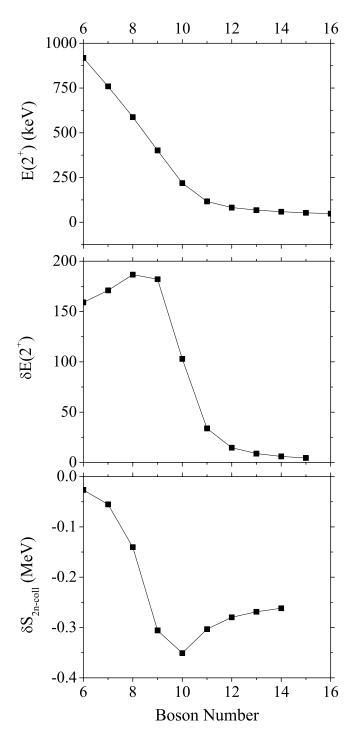


FIG. 8: Calculated  $E(2_1^+)$  (top),  $\delta E(2_1^+)$  (middle) and  $\delta S_{2n-coll}$  (bottom) values from IBA calculations as a function of boson number  $N_B$ . The points for  $\delta E(2_1^+)$  and  $\delta S_{2n-coll}$  correspond to a set of schematic IBA calculations in which  $\kappa$ , and  $\chi$  are constant (at 0.02 and -1.32, respectively) while  $\epsilon$ , and  $N_B$  vary in a smooth way to simulate a spherical-to-deformed transition region. The following equations are used for  $\delta E(2_1^+)$  and  $\delta S_{2n-coll}$ :  $\delta E(2_1^+)(Z,N) = [E(2_1^+)(Z,N) - E(2_1^+)(Z,N) - S_{2n-coll}(Z,N+2)]$ , respectively.

The first term in Eq. (3) drives nuclei spherical while the  $Q \cdot Q$  term induces collectivity and deformation. Therefore a spherical-deformed transition region involves a systematic change in the ratio of  $\epsilon$  to  $\kappa$ . No generality is lost by keeping  $\kappa$  constant (at 0.02 MeV). We follow a trajectory along the bottom axis of the triangle corresponding to  $\chi = -1.3228$ . Fig. 8 illustrates the results, for  $N_B = 6 - 16$  showing  $E(2_1^+)$  and the differentials of  $E(2_1^+)$  (for  $N_B=6-15$ ) and for the collective contributions to  $S_{2n}$ ,  $S_{2n-coll}$ , (for  $N_B = 6 - 14$ ). There is a clear change in structure at  $N_B \sim 10$  which is seen in a change in trend of  $E(2_1^+)$ . Between  $N_B = 10$  and 11,  $R_{4/2}$  changes from 2.60 to 3.13. This corresponds to a maximum in the normalized differential of  $E(2_1^+)$ . Confirming our association of structural changes with kinks in  $S_{2n}$ , the differential of the collective part of  $S_{2n}$  also shows an extremum at exactly the same point. These ideas will be expanded in our future publication [37].

Besides the experimental examples of a correlation of  $E(2_1^+)$  energies and  $S_{2n}$  values discussed in the context of Fig. 5, our interpretation can easily be illustrated with the Sm isotopes around N = 90. As is well known, there is a sudden onset of deformation for the rare earth nuclei from N=88 to 90. This effect is clear from various observables. One example is seen in Fig. 9 which shows the experimental  $E(2_1^+)$  energies (top) and  $S_{2n}$  (bottom) as a function of neutron number. Note that we plot these against decreasing neutron number so that the deformed nuclei are on the left and spherical ones on the right to make the comparison with Fig. 5 easier. Note also that the overall trend in  $S_{2n}$  is opposite from that in Fig. 5 since  $S_{2n}$  values decrease with increasing neutron number (going to the left in Fig. 9 (bottom) which simply reflects the filling of the shell model orbits. The noticeable deviation occurs near  $N \sim 90$  where there is a distinct flattening. To correlate the trends in these two observables in the N=90 region, one can use the same interpretation as above for W at N = 116, namely, if there is a visible change at neutron number N in  $E(2_1^+)$  and there are many valence nucleons, we expect to see a change in the behavior of  $S_{2n}$ . In Fig. 9 (top), the  $E(2_1^+)$  change occurs at  $N \sim 90$ . The isotope <sup>62</sup>Sm at N = 90 has  $N_p = 12$  and  $N_n = 8$  so it has 10 bosons and its P-factor is 4.8 (see Fig. 7). One therefore expects to see a change in  $S_{2n}$ . Fig. 9 (bottom) confirms this expectation. The clear structural change at N = 90 shown in  $E(2_1^+)$  is correlated with a larger binding compared to the general trend in  $S_{2n}$  as a function of N.

Similar correlations can be seen in some other nuclei as well. Further details will be discussed in Ref. [37]. However, here, it is worth mentioning two more examples marked in Fig. 5. The case of Yb-isotopes is interesting. Yb at N=107 starts to change slope in  $S_{2n}$  and a flattening occurs at N=108. The P-factor is  $\sim 7$ . With the interpretation above, one would expect to see a change in  $E(2_1^+)$  after N=106, at N=108. However, there is no sudden change in  $E(2_1^+)$  in  $^{178}$ Yb. To understand Yb around  $N \sim 108$  better, it might therefore be useful

to have additional  $S_{2n}$  values (mass measurements) and also more spectroscopic results for the neutron-rich Yb isotopes.

Hf at N = 114 has a P-factor 5.4 and one sees a flattening in  $S_{2n}$ . The corresponding  $2^+$  energies, however, are not known. Thus, similarly as in Yb, we need more spectroscopic results for Hf.

To summarize, we observed a correlation between the behavior of  $S_{2n}$  obtained from our measured masses with the spectroscopic data for  $E(2_1^+)$ , which could be related to nuclear collectivity and valence nucleon number.

#### V. CONCLUSION

Direct mass measurements of neutron-rich <sup>197</sup>Au projectile fragments at the cooler-storage ring ESR yielded new mass data. Masses of nine nuclides were obtained for the first time and for three nuclei the mass uncertainty was improved.

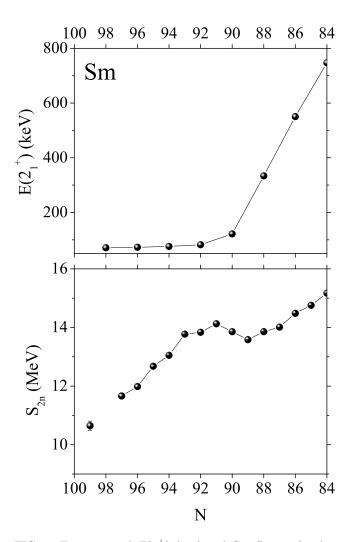


FIG. 9: Experimental  $E(2_1^+)$  (top) and  $S_{2n}$  (bottom) values against neutron number for  ${}_{62}\mathrm{Sm}$  [29, 32].

With the new masses, two-neutron separation energies,  $S_{2n}$ , are investigated. We showed that changes in structure, as indicated by changes in the collective observable  $E(2_1^+)$ , are reflected in  $S_{2n}$  values in nuclei such as <sup>190</sup>W and  $^{188}$ Pt, which have large P-factors, are collective, and have large valence nucleon numbers. For nuclei with similar changes in  $E(2_1^+)$ , such as <sup>196</sup>Os, and <sup>198</sup>Pt, which have lower collectivity and P-factors, and fewer valence nucleons, the sensitivity of collective binding to structure is greatly reduced and smooth trends in  $S_{2n}$  are observed. In Hf, there are new  $S_{2n}$  values at N=113, 114 where we see a flattening but there is no spectroscopic data at N = 114. To confirm the ideas discussed in this paper and also in Ref. [37], it would be useful to measure the  $E(2_1^+)$  for Hf at N=114. Similarly, mass and spectroscopic measurements are suggested for nuclei such as Yb with  $N \sim 108$ . To conclude, these new data illustrate subtle changes in structure and the correlation with  $E(2_1^+)$  reveals a valuable way to correlate changes in structure in terms of both masses and spectroscopic observables. Of course, to quantitatively test these ideas requires a systematic collective model study of the massstructure relationship in this region. Such a project has been initiated [37] and we illustrated some of the results here.

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