# **Penning trap mass measurements on nobelium isotopes**

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The Penning trap mass spectrometer SHIPTRAP at GSI Darmstadt allows accurate mass measurements of radionuclides, produced in fusion-evaporation reactions and separated by the velocity filter SHIP from the primary beam. Recently, the masses of the three nobelium isotopes  $252-254$ No were determined. These are the first direct mass measurements of transuranium elements, which provide new anchor points in this region. The heavy nuclides were produced in cold-fusion reactions by irradiating a PbS target with a <sup>48</sup>Ca beam, resulting in production rates of the nuclei of interest of about one atom per second. In combination with data from decay spectroscopy our results are used to perform a new atomic-mass evaluation in this region.

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## **I. INTRODUCTION**

The mass of a nuclide is a fundamental property that provides information about the nuclear binding energy. It is thus crucial for nuclear structure studies and allows tests of theoretical nuclear models. Compared to an indirect mass determination via decay energies, a method in which the mass can be directly addressed is favorable since it is independent of a detailed knowledge of nuclear level schemes. Penning traps allow such a direct mass determination by measuring the cyclotron frequency of the ion of interest stored in a homogeneous magnetic field. Since frequency measurements can be carried out with a very high precision, the Penning trap method is the most accurate one in mass spectrometry [\[1\]](#page-8-0). Thus, Penning traps are nowadays widely used for mass measurements on short-lived nuclides [\[2\]](#page-8-0). However, although measurements with very low production rates have been performed, such as 0.3 particles*/*s in the case of the discovery of  $2^{29}$ Rn [\[3\]](#page-8-0), up to now this technique has not been applied to transuranium elements where the yields are in general even much lower.

The existence of the elements in the superheavy region was primarily thought to be impossible since the fission barrier calculated according to macroscopic models disappears for fission parameters  $Z^2/A$  larger than about 40. Therefore, nuclei having a proton number larger than  $Z \approx 100$  were expected to decay immediately after creation by prompt fission [\[4\]](#page-8-0). However, it was found later that the shell structure of the nuclei creates a modification of the binding energy leading to a notable fission barrier stabilizing nuclei with *Z >* 100 [\[5,6\]](#page-8-0). In the region of the nobelium isotopes addressed in this paper this gives rise to nuclear deformations. To get a deeper understanding of the stabilizing mechanisms in these superheavy elements it is essential to measure the subtle contribution of shell effects (i.e., the binding energies of these nuclei). This can be achieved by performing high-precision mass measurements.

Previous mass determinations on elements heavier than uranium were based solely on measurements of *α*-decay energies. Superheavy nuclides have their masses determined via *α*-decay chains down to known masses. The uncertainty of the mass is then determined by the uncertainties of the  $\alpha$ -decay energies linking the decay chain members. Additionally, in the case of odd-odd nuclides or nuclides with odd *A* the mother nuclide often decays into excited states of the daughter nuclide. Therefore, a detailed knowledge of the nuclear level schemes of all nuclides in the decay chain is required for these

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<span id="page-1-0"></span>procedures. An accurate determination of the excitation energy is experimentally challenging, in particular for low-lying excited states that decay into the ground state by internal conversion. It is therefore of utmost importance to obtain unambiguous data from direct mass measurements.

An additional demand for mass determinations in this region comes from the fact that some decay chains end by spontaneous fission and are not linked to any known nuclide. This problem can be solved by directly addressing the end-point nuclides using Penning trap mass spectrometry.

The Penning trap system SHIPTRAP [\[7\]](#page-8-0) installed behind the velocity filter SHIP [\[8\]](#page-8-0) at GSI allowed us so far to determine the masses of more than 60 radionuclides, many of them for the first time [\[9–12\]](#page-8-0). Recently, direct mass measurements were extended to the three nobelium isotopes  $252-254$  No ( $Z =$ 102), which are the first direct mass measurements above uranium  $(Z = 92)$ . The results of these investigations are reported in Ref. [\[13\]](#page-8-0). In this publication we describe the work in more detail and compare our results with previous measurements.

In Sec. II, Sec. III, and Sec. [IV](#page-2-0) the production of the nobelium isotopes, the experimental setup, and the measurement procedure are explained. In Sec. [V](#page-2-0) and Sec. [VI](#page-3-0) the results of the mass measurements are presented and compared with the results of previous measurements. The influence of our mass values on heavier nuclides is discussed in Sec. [VII.](#page-7-0) An outlook is given in Sec. [VIII.](#page-7-0)

### **II. PRODUCTION OF THE NOBELIUM ISOTOPES**

The nobelium isotopes  $252-254$ No were produced by a  $48$ Ca beam reacting with different lead targets consisting of one of the enriched isotopes  $206Pb$ ,  $207Pb$ , and  $208Pb$  in the molecule PbS. The target thickness was 0.5 mg*/*cm2. Eight target segments were mounted on a wheel rotating with a frequency of 1125 turns per minute. The beam was delivered in 5.5-ms pulses at a repetition rate of 50 Hz. The reaction products were separated from the primary beam by the velocity filter SHIP [\[8\]](#page-8-0). The highest possible cross sections were always obtained in fusion-evaporation reactions via the 2*n* channel, which were in all three cases about a factor of 10 larger than the cross sections of the other reaction channels. The

TABLE I. Half-lives of ground and longest lived isomeric states, and production cross sections of the nobelium isotopes investigated in this work.

<b>Isotope</b>	$T_{1/2}$ (gs) [22]	$T_{1/2}$ (isomer) [24–27]	Cross section
$^{252}$ No	$2.44(4)$ s	$110(10)$ ms	$400$ nb
$^{253}$ No	$1.62(15)$ min	$715(30) \mu s$	$1 \mu b$
$254$ No	$51(10)$ s	$266(2)$ ms	1.8 $\mu$ b

highest production rates were achieved by using the enriched target with the best-suited enriched lead isotope. The optimum projectile energy was 4.55 MeV*/*u in all three cases, which results in an excitation energy of the compound nucleus of about 22 MeV.

The <sup>48</sup>Ca beam had an average intensity of about  $6 \times$  $10^{12}$  particles per second. The production cross sections ranged from 1.8  $\mu$ b for the isotope <sup>254</sup>No to 400 nb for the isotope  $^{252}$ No (see Table I). The latter corresponds to about 0.4 particles per second entering the gas cell of SHIPTRAP. The total efficiency of SHIPTRAP was on the order of a few percent, resulting in about one detected particle every minute.

### **III. THE SHIPTRAP SETUP**

Figure 1 shows the SHIPTRAP setup [\[7\]](#page-8-0) used for this experiment. It consists of a gas cell, an RFQ cooler and buncher, and a double Penning trap system.

The nobelium ions coming from SHIP with a kinetic energy of around 22 MeV were stopped in the gas cell filled with ultrahigh purity He (1 ppm) at a pressure of 50–60 mbar [\[14\]](#page-8-0). The gas cell is separated from the  $10^{-6}$  mbar vacuum of the transfer beam line by a titanium entrance window with a thickness of about 2 mg/cm<sup>2</sup> and an open diameter of 60 mm. An additional set of mylar degraders (whose thickness could be varied between 0.5 and 3.0  $\mu$ m) is installed in the beam line in front of the entrance window to stop most ions in the center of the gas cell. Inside the gas cell an electrode system consisting of a DC voltage cage and an RF funnel is mounted to generate an electric field that drags the ions toward the exit nozzle within a few milliseconds [\[15\]](#page-8-0). Upon arriving



FIG. 1. Schematic overview of the SHIPTRAP setup. It consists of a gas cell to stop the high energetic reaction products, an RFQ cooler and buncher to cool and accumulate the ions, and a double Penning trap system to perform high-precision mass measurements.

<span id="page-2-0"></span>at the nozzle the ions were ejected from the gas cell by the helium gas flow.

Subsequently, the ions were transported to the RFQ cooler and buncher [\[16\]](#page-8-0) made of four segmented rods. Here, the ions were cooled in collisions with helium gas atoms at a pressure of about  $5 \times 10^{-3}$  mbar and were accumulated in a potential minimum. The cooling time was about 3 ms [\[16\]](#page-8-0).

After a typical storage time of 1–2 s in the buncher the ions were ejected in pulses with a width of about 500 ns and transferred to the double Penning trap system, which is placed in a superconducting solenoid magnet providing a field strength of 7 T. In the first trap isobaric separation with a mass resolving power of up to 100 000 is achieved by applying buffer-gas cooling [\[17\]](#page-8-0). In the second trap the cyclotron frequency of the ions is determined with the time-of-flight ion cyclotron resonance (TOF-ICR) detection method [\[18\]](#page-8-0). While one sample of ions was cooled and investigated in the Penning traps, another sample was accumulated and cooled in the RFQ cooler and buncher.

### **IV. MASS MEASUREMENT PROCEDURE AND ANALYSIS**

The trapped ions were excited at a frequency *ν*<sub>exc</sub> near the cyclotron frequency  $v_c = qB/(2\pi m)$  for the duration *T*exc. After the ejection, the time of flight from the trap to a Channeltron detector was measured. This measurement cycle was repeated several times while stepping the excitation frequency  $v_{\text{exc}}$  across  $v_c$  from one cycle to the next.

The time of flight was then plotted as a function of the excitation frequency *ν*<sub>exc</sub>, resulting in a resonance curve (see Fig. 2). The cyclotron frequency *νc* was determined by a fit of the expected line shape to the experimental data points [\[19\]](#page-8-0). Resonances of a reference ion of well-known mass were taken at times*t*<sup>1</sup> and *t*2, before and after the measurement of the ion of interest, respectively. The reference frequency *ν*<sub>c,ref</sub> at the time of the measurement of the ion of interest was obtained from linear interpolation between the two frequencies *ν*c*,*ref*,*<sup>1</sup> and *ν*c*,*ref*,*2. Since in all cases the average number of ions injected into the Penning trap was much less than 1 count per cycle and



FIG. 2. Time-of-flight ion cyclotron resonance of  $254No^{2+}$  taken during the beam time in August 2008. The excitation time was  $T_{\text{exc}} =$ 900 ms. The solid line is a fit of the expected line shape to the experimental data points. The cyclotron frequency was determined to be  $v_c = 846665.281(51)$  Hz.

the efficiency of the channeltron was determined to be close to 100% a count-rate class analysis as described in Ref. [\[20\]](#page-8-0) was not applied.

The mass was then determined from the frequency ratio  $r = v_{c,ref}/v_c$  of the nuclide of interest to the reference ion according to [\[20\]](#page-8-0)

$$
m_{\text{atom}} = \frac{z}{z_{\text{ref}}} r(m_{\text{atom,ref}} - z_{\text{ref}} m_e) + z m_e, \tag{1}
$$

where  $m_e$  is the electron mass.  $m_{\text{atom}}$  and  $m_{\text{atom,ref}}$  denote the atomic masses, *νc* and *ν*c*,*ref are the cyclotron frequencies, and  $z$  and  $z_{ref}$  are the charge states of the ion under investigation and of the reference ion, respectively. In the present measurement doubly charged nobelium ions were investigated  $(z = 2)$ , which led to a decrease of the relative uncertainty. Furthermore, the observation of doubly charged ions indicates that the helium gas in the gas cell and the purification trap is of high purity.

The uncertainty resulting from nonlinear changes of the magnetic field  $\epsilon_B$  is added quadratically to the statistical uncertainty *δr*<sub>stat</sub> obtained from each frequency ratio *r*:

$$
\delta r_{\rm res} = \sqrt{\delta r_{\rm stat}^2 + [(t_2 - t_1)\epsilon_B r]^2}.
$$
 (2)

The final frequency ratio  $\bar{r}$  is the weighted mean of the frequency ratios  $r_i$  of all individual measurements. The uncertainty of the final frequency ratio  $\bar{r}$  is increased quadratically by the relative systematic uncertainty  $\epsilon_{syst}$ :

$$
\delta \overline{r}_{\text{total}} = \sqrt{\delta \overline{r}_{\text{res}}^2 + (\epsilon_{\text{syst}} \overline{r})^2}.
$$
 (3)

The relative systematic uncertainty  $\epsilon_{syst}$  and the dispersion of the magnetic field  $\epsilon_B$  have been determined in calibration measurements by reference ions with well-known masses to be  $\epsilon_{syst} = 4.5 \times 10^{-8}$  and  $\epsilon_B = 1.3 \times 10^{-9}/h$  [\[7,21\]](#page-8-0).

 $133^{\circ}$ Cs<sup>+</sup>, which is known with a mass uncertainty of only 22 eV [\[22\]](#page-8-0), was used as a reference ion since its *A/q* ratio is close to that of the doubly charged nobelium ions. This minimizes possible mass-dependent frequency shifts [\[23\]](#page-8-0).

## **V. SHIPTRAP RESULTS**

As a first step toward direct mass measurements on superheavy nuclides, the masses of the three nobelium isotopes <sup>252</sup>*,*253*,*254No were determined in August 2008 [\[13\]](#page-8-0).

The half-lives of the ground states of all nobelium isotopes addressed here are rather long (see Table [I\)](#page-1-0) and allow long excitation times, resulting in a measurement cycle on the order of two to three seconds. The half-lives of the known isomeric states [\[24–27\]](#page-8-0) are short compared to the cycle time (Table [I\)](#page-1-0). One can thus assume that all isomers already decayed before entering the measurement trap and did not affect the measurement. Owing to the narrow excitation functions  $(\approx 6 \text{ MeV})$  for specific evaporation channels in the considered reactions, atoms of neighboring isotopes are produced with more than an order of magnitude lower rates at the given bombarding energy. Isobaric contamination can be excluded as well, since the proton evaporation probability is more than one order of magnitude lower than the one for the neutron evaporation.

<span id="page-3-0"></span>TABLE II. Mean frequency ratios  $\bar{r}$  (column 2) of the nobelium isotopes investigated; the absolute statistical uncertainty  $\delta \overline{r}_{stat}$  and the absolute total uncertainty  $\delta \overline{r}_{total}$  are listed in columns 3 and 4, respectively.  $133Cs^{+}$  was used as the reference ion.

<i>I</i> sotope	r	$\delta \overline{r}_{\rm stat}$	$\delta \overline{r}_{\text{total}}$
$252$ No	0.948 376 768	$1.18 \times 10^{-7}$	$1.26 \times 10^{-7}$
253 <sub>N<sub>0</sub></sub>	0.952 144 941	$2.8 \times 10^{-8}$	$5.1 \times 10^{-8}$
$254$ No	0.955 908 553	$3.9 \times 10^{-8}$	$5.8 \times 10^{-8}$

For the isotope 252No, which was produced in the reaction  $^{206}Pb(^{48}Ca, 2n)^{252}No$ , three resonances were taken, two with  $T_{\text{exc}} = 200$  ms and one with  $T_{\text{exc}} = 900$  ms. Since the production cross section for 252No was as low as about 400 nb, only one ion in a time interval of five to six minutes was detected by the channeltron detector, which has a detection efficiency of nearly 100% [\[28\]](#page-8-0). The values obtained from the three resonances agree with each other (see Fig. 3 top). The mean frequency ratio was determined to be  $\bar{r}$  = 0.948 376 768(126), corresponding to a mass excess of  $ME =$ 828 50(31) keV $/c^2$ . The statistical and total uncertainties for the individual isotopes are shown in Table  $II$ .

<sup>253</sup>No was produced in the reaction <sup>207</sup>Pb( $^{48}Ca, 2n$ )<sup>253</sup>No. One resonance with  $T_{\text{exc}} = 200 \text{ ms}$  and four resonances with  $T_{\text{exc}} = 900$  ms were recorded. The values obtained from the five individual resonances are in excellent agreement with each other (Fig. 3 middle). The combined frequency ratio is  $\bar{r} = 0.952144941(51)$ , which results in a mass excess of ME = 843 56(13) keV/c<sup>2</sup>.

 $ME = 843\,56(13) \text{ keV}/c^2.$ <br><sup>254</sup>No was produced in the reaction <sup>208</sup>Pb(<sup>48</sup>Ca, 2*n*)<sup>254</sup>No with the highest yield of the three isotopes investigated. The resonances were taken with a count rate of about one to two ions per minute. Four resonances were taken for this isotope, one with  $T_{\text{exc}} = 200$  ms and three with  $T_{\text{exc}} = 900$  ms. Figure [2](#page-2-0) shows a resonance of doubly charged <sup>254</sup>No taken with  $T_{\rm exc} = 900$  ms as an example. Again, the values obtained from the four resonances are in excellent agreement with each other (see Fig. 3 bottom). The resulting frequency ratio was found to be  $\bar{r} = 0.955908553(58)$  and the corresponding mass excess of <sup>254</sup>No is ME = 847 33(14) keV/ $c^2$ .

## **VI. COMPARISON OF THE SHIPTRAP RESULTS WITH PREVIOUS DATA**

Prior to our experiments, all nuclear mass values above  $Z = 92$  had been derived from indirect mass determinations based on  $Q_{\alpha}$  values from decay spectroscopy. In the case of an  $\alpha$  decay the mass difference between mother and daughter nuclide can be written as

$$
M_{\text{mother}} - M_{\text{daughter}} = \frac{Q_{\alpha}}{c^2} + M(^4\text{He}). \tag{4}
$$

In first approximation the  $Q_{\alpha}$  value can be written as

$$
Q_{\alpha} = \left(1 + \frac{M_{\alpha}}{M_{\text{daughter}}}\right)E_{\alpha} + E_{\text{exc}},\tag{5}
$$



FIG. 3. Results for the mass excesses of the individual measurements of the nobelium isotopes  $252-254$  No. The solid lines mark the mean values and the gray area their 1*σ* uncertainty.

where  $E_\alpha$  denotes the observed energy of the  $\alpha$  particles and *E*exc a possible excitation energy of a state in the daughter nucleus, after the mother decays by  $\alpha$  emission. From this formula one can indirectly determine the mass of the mother nuclide by measuring  $E_{\alpha}$  if the excitation energy  $E_{\text{exc}}$  is known.

In the Atomic-Mass Evaluation (AME) 2003  $[22]$  <sup>252</sup>No and <sup>254</sup>No are listed as secondary nuclides, whereas <sup>253</sup>No was derived from systematic trends. Their masses were determined from  $\alpha$ -decay chains ending at a primary nucleus, <sup>232</sup>U in the case of  $^{252}$ No,  $^{241}$ Cm in the case of  $^{253}$ No, and  $^{238}$ Pu in the case of 254No. Primary nuclides are defined by the fact that their masses are determined from at least two experiments with different methods. Such nuclides are labeled in the AME with "degree 1." Nuclides whose mass values are determined from only one link to another nuclide are called secondary nuclides. These nuclides are labeled with degrees starting from "degree 2" if there is one direct link to a primary nucleus (for

<span id="page-4-0"></span>

FIG. 4. (Color online) Scheme of evaluated masses before (top) and after (bottom) implementing the SHIPTRAP data into the Atomic-Mass Evaluation. The so-called degree of a specific nuclide (see text) is represented by different colors.

example in the case of  $236$ Pu in Fig. 4 top). The degree is increasing with the distance to the related primary nuclide in the respective data chain. Generally speaking, the degree of a nuclide is  $n + 1$  if its mass is determined using a link to a nuclide with degree *n*. The degrees of the nuclides of the three decay chains, which are relevant for the work reported here, <span id="page-5-0"></span>are indicated in Fig. [4](#page-4-0) by a color code. In such a data chain the nuclide of lower degree is influencing all the nuclides of higher degree by 100%. The uncertainty of the mass determination of a nuclide within a chain is increasing with its degree since it includes the uncertainties of all corresponding lower degree nuclides determining it. In addition, the mass uncertainty of a nuclide becomes larger if its level scheme or the level schemes of the involved nuclides with lower degrees are unknown. To obtain more accurate data it is desirable to get experimental input from independent methods.

Figure [4](#page-4-0) (top) shows the situation before implementing the SHIPTRAP data into the AME network. The masses of the nuclides in the three decay chains were based on the primary nuclides  $^{232}$ U,  $^{241}$ Cm, and  $^{238}$ Pu, whose masses were known with a rather small uncertainty. Thus, the uncertainties of the masses of the higher degree nuclides were mainly limited by the uncertainties of the  $Q_{\alpha}$  values of the linking *α* decays. In all three cases the mass uncertainties of the nuclides up to californium were small (around 2–3 keV). Starting from fermium the mass uncertainty increased owing to higher uncertainties in the  $Q_{\alpha}$  values. This also affected the mass uncertainties of the nobelium isotopes, even though in the first two decay chains the uncertainties of the  $Q_{\alpha}$  values of the decay from nobelium to fermium are smaller than the ones of the decay from fermium to californium. Based on all spectroscopic data measured until 2009, the masses of the nobelium isotopes were known with an uncertainty of about 10 to 20 keV.

Figure [4](#page-4-0) (bottom) shows the three decay chains after adding the SHIPTRAP data. All values are summarized in Table III, where, additionally, the contribution of the SHIPTRAP data to the new weighted mean values is given in percent. One can see that the SHIPTRAP results confirm all values derived from  $Q_{\alpha}$  measurements. Furthermore, in all three cases, there is a remarkable influence resulting in the new intermediate AME data from the link of  $^{133}Cs$  to the nobelium isotopes and its decay products.

The mass value of the nobelium isotope  $252$ No was previously only based on an *α*-decay chain ending in 232U (Fig. [4](#page-4-0) left). Since this chain consists of even-even nuclei the most intensive *α* transitions always occur between the ground state of the mother nuclide and the ground state of the daughter nuclide. The most critical connections with respect to the mass uncertainty of  $^{252}$ No are the decays from  $^{252}$ No to

TABLE III. Mass excess (ME) values and influence of the new nobelium data on the ME values for the three *α*-decay chains ending at  $^{232}$ U,  $^{241}$ Cm, and  $^{238}$ Pu (see Fig. [4\)](#page-4-0). AME2003: ME values for the nuclides as published in Ref. [\[22\]](#page-8-0). Before SHIPTRAP: ME values before the SHIPTRAP data evaluation. SHIPTRAP: SHIPTRAP results on <sup>252–254</sup>No. After SHIPTRAP: AME values after implementing the SHIPTRAP data. Contribution: contribution of the SHIPTRAP values to the new AME value. The sign # indicates those mass data for which the value is estimated from systematic trends.

Nuclide	AME2003 ME (keV/ $c^2$ )	<b>Before SHIPTRAP</b> ME (keV/ $c^2$ )	<b>SHIPTRAP</b> ME (keV/ $c^2$ )	<b>After SHIPTRAP</b> ME (keV/ $c^2$ )	Contribution (percentage)
$232$ [J	34610.7(2.2)	34603.7(2.0)		34603.7(1.9)	0.3
$^{236}Pu$	42902.7(2.2)	42895.7(2.0)		42895.7(1.9)	0.3
$240$ Cm	51725.4(2.3)	51718.4(2.1)		51718.4(2.0)	0.4
244Cf	61479.2(2.9)	61472.2(2.7)		61472.2(2.7)	0.7
$^{248}\mathrm{Fm}$	71906(12)	71892.3(9.7)		71891.0(9.3)	8.4
252 <sub>NO</sub>	82881(13)	82866(11)	82850(31)	82864(11)	11
$^{256}\mathrm{Rf}$	94236(24)	94221(23)		94219(23)	11
$^{260}\mathrm{Sg}$	106580(40)	106568(38)		106566(38)	11
$264$ Hs	119600(40)	119584(43)		119582(43)	11
$^{268}\mathrm{Ds}$	133940#(500#)	133289#(303#)		133287#(303#)	11
$241$ Cm	53703.4(2.2)	53705.1(2.2)		53705.2(2.1)	1.5
$245$ Cf	63386.9(2.9)	63388.6(2.8)		63388.6(2.8)	2.6
$^{249}\mathrm{Fm}$	73620#(100#)	73523(12)		73521.0(8.8)	46
$^{253}\mathrm{No}$	84470#(100#)	84360(13)	84356(13)	84357.8(8.8)	52
$^{257}\mathrm{Rf}$	95930#(100#)	95829(20)		95827(18)	52
$^{261}Sg$	108160#(130#)	108056#(75#)		108054#(74#)	52
$^{265}$ Hs	121170#(140#)	121068#(90#)		121065#(89#)	52
$^{269}$ Ds	135180#(140#)	135077#(92#)		135075#(92#)	52
$^{238}Pu$	46164.7(1.8)	46166.2(1.8)		46166.3(1.8)	0.7
$242$ Cm	54805.2(1.8)	54806.7(1.8)		54806.73(1.8)	0.7
246Cf	64091.7(2.1)	64093.2(2.1)		64093.3(2.1)	0.9
$^{250}$ Fm	74074(12)	74074.1(9.0)		74077.7(8.2)	16
$^{254}\mathrm{No}$	84724(18)	84711(17)	84733(14)	84725(11)	60
${}^{258}\mathrm{Rf}$	96400#(200#)	96329(35)		96342(32)	60
${}^{262}{\rm Sg}$	108420#(280#)	108354#(203#)		108367#(203#)	60
$^{266}\mathrm{Hs}$	121190#(280#)	121114#(204#)		121128#(204#)	60
$^{270}\mathrm{Ds}$	134810#(290#)	134735#(210#)		134749#(210#)	60

 $^{248}$ Fm and from  $^{248}$ Fm to  $^{244}$ Cf since, compared to the other decays in this chain, they have relatively large uncertainties of 5.5 and 9.3 keV, respectively.

The  $Q_{\alpha}$  value of the decay from <sup>252</sup>No to <sup>248</sup>Fm is presently based on two measurements [\[29,30\]](#page-8-0), resulting in an average value of 8548.7(5.5) keV. In case of the decay from  $^{248}$ Fm to <sup>244</sup>Cf the  $Q_{\alpha}$  value was determined to be 7995.2(9.3) keV [\[30–32\]](#page-8-0). The uncertainties of the  $Q_\alpha$  values of the other decays down to 232U are much smaller and have values of 1.8 keV for the decay from  $^{244}$ Cf to  $^{240}$ Cm, 0.6 keV for the decay from <sup>240</sup>Cm to <sup>236</sup>Pu, and only 0.08 keV for the decay from <sup>236</sup>Pu to  $232$ U

The mass excess of the isotope  $232U$ , which is the end point of the  $\alpha$ -decay chain, is known with an uncertainty of 1.96 keV. It was determined by a measurement of the energies of the  $\alpha$  decay to <sup>228</sup>Th for 75% and the <sup>234</sup>U( $p, t$ )<sup>232</sup>U–  $184W(p, t)$ <sup>182</sup>W differential reaction for 25% [\[33,34\]](#page-8-0). This uncertainty is mainly determining the uncertainties of the mass excesses of <sup>236</sup>Pu (1.96 keV), <sup>240</sup>Cm (2.04 keV), and <sup>244</sup>Cf (2.7 keV), respectively. The uncertainties of the mass excesses of  $^{248}$ Fm (9.7 keV) and  $^{252}$ No (11.2 keV), however, are mainly determined by the rather large uncertainties of the respective *Qα* values.

The SHIPTRAP value with an uncertainty of 31 keV is in agreement with the value based on  $Q_{\alpha}$  measurements. The contribution to the mass value of nobelium itself is 11% and 8.4% to the mass value of its daughter 248Fm. The influence on the lighter decay products is negligible since they are determined more strongly by the mass of  $^{232}$ U, which has a much smaller uncertainty and the linking  $Q_{\alpha}$  values have a high precision. The SHIPTRAP measurement reduced the mass excess uncertainty of <sup>252</sup>No slightly from 11.2 to 10.5 keV.

The AME 2003 mass value of the isotope <sup>253</sup>No was based on an  $\alpha$ -decay chain down to <sup>241</sup>Cm. The members of this decay chain are nuclides with even *Z* and odd *N*, which means that the ground state to ground state *α* decay does not represent a favored transition and thus often has only a small intensity. Thus, the excitation energies *E*exc of the excited states that are populated in the decay have to be known in each case to draw any conclusions on the  $Q_{\alpha}$  values. Detailed decay schemes can be constructed from  $\alpha$ - $\gamma$  coincidences. However, this requires high statistics and a clear separation of the  $\alpha$  line of interest from those of neighboring isotopes that might be produced as well. Moreover, many states in this region decay by internal conversion and not by emission of *γ* rays.

The decay of  $253$ No to  $249$ Fm has been studied at SHIP [\[30,35\]](#page-8-0). Figure 5 shows the decay scheme that has been constructed based on the most recent results. The favored *α* decay was assigned to the transition from the 9*/*2<sup>−</sup> [734] ground state of <sup>253</sup>No into the 9/2<sup>-</sup> [734] state of <sup>249</sup>Fm with  $E_\alpha = 8004(5)$  keV. This latter state decays via three transitions: one into the  $7/2^+$  [624] ground state ( $E_\gamma$  = 279.7 keV), one into a  $9/2^+$  state ( $E_\gamma = 222.0$  keV), and one into an  $11/2^+$  state ( $E_\gamma = 151.4$  keV). A  $Q_\alpha$  value of 8413(11) keV can be deduced from the decay into the ground state. From this and an additional measurement [\[36\]](#page-8-0) a weighted mean value of  $Q_{\alpha} = 8412.2(4.4)$  keV has been derived.

Similarly, in the case of the decay from  $249$ Fm to  $245$ Cf, a mass determination is difficult to make because of a



FIG. 5. Simplified decay scheme of 253No summarizing our present knowledge [\[30,35,37\]](#page-8-0). The energies are given in keV. The numbers in brackets indicate the Nilsson levels. Details are given in the text.

complicated decay scheme involving several excited states (see Fig. 5). Here, the  $\alpha$  decay from the  $7/2^+$  [624] ground state of <sup>249</sup>Fm into the 7/2<sup>+</sup> [624] state of <sup>245</sup>Cf (7520 keV) is followed by an  $E2$  transition into the  $1/2^+$  [631] ground state of 245Cf with an energy of 55(10) keV. The latter value was estimated on the basis of the energy difference of the *α* lines in Ref. [\[37\]](#page-8-0) and in Ref. [\[30\]](#page-8-0). In Ref. [\[37\]](#page-8-0) the He-jet technique was used. In this an undisturbed  $\alpha$  line was measured, whereas in Ref. [\[30\]](#page-8-0) the nuclei were implanted into a silicon detector and thus the  $\alpha$  energy was influenced by a contribution of conversion electrons from the transition of the  $7/2^+$  [624] state into the ground state. The resulting value of  $Q_\alpha$  = 7709.4(11.6) keV is thus not very precise.

The  $Q_{\alpha}$  value of the decay from <sup>245</sup>Cf to <sup>241</sup>Cm is known with an uncertainty of 1.8 keV. The mass excess of  $241$ Cm, which is the next primary nuclide in this decay chain, was measured with an uncertainty of 2.15 keV [\[38\]](#page-8-0). Based on these two values the mass excess of  $245$ Cf can be determined with an uncertainty of 2.8 keV. The uncertainty of the mass excess of  $249$ Fm is determined to be 11.9 keV, owing to the rather large uncertainty of the  $Q_{\alpha}$  value of the decay from  $249$ Fm to  $245$ Cf. This decay also limits the uncertainty of the mass excess of 253No, which is 12.7 keV.

The mass value of the isotope  $253$ No was measured with an uncertainty of 13 keV by SHIPTRAP, which is comparable to the uncertainty of the most recent AME value. The contribution of the SHIPTRAP measurement to the new AME value is thus 52% and the uncertainty of the <sup>253</sup>No mass excess is reduced to 8.8 keV. In addition, the mass value of  $249$ Fm is influenced by the new measurement by 46%, which leads to a reduction of its mass excess uncertainty from 12 to 8.8 keV. Also the *Qα* value of the decay from  $249$ Fm to  $245$ Cf, which was so far the weakest link in the decay chain, is reduced from 12 to 8.7 keV. More important is that it confirms the decay scheme suggested by [\[35\]](#page-8-0), where an excitation energy of 55(10) keV was assumed for an excited state in  $245$ Cf. The lighter nuclides are only slightly affected by the implementation of the SHIPTRAP value.

<span id="page-7-0"></span>The mass of  $254$ No was based on an  $\alpha$ -decay chain consisting of even-even nuclides, so that only ground state to ground state transitions are involved. The decay chain ends at the isotope 238Pu.

The  $Q_{\alpha}$  value of the decay from <sup>254</sup>No to <sup>250</sup>Fm has been measured several times [\[39–41\]](#page-8-0) and results in a weighted mean value of  $Q_\alpha = 8212.2(14.8)$  keV. The measurements of the  $Q_\alpha$ value of the decay from  $250$ Fm to  $246$ Cf result in a weighted mean value of  $Q_{\alpha} = 7556.0(8.7)$  keV [\[29–31,37\]](#page-8-0). The further  $Q_{\alpha}$  values of this decay chain are known with an uncertainty of 1.0 keV for the decay from 246Cf to 242Cm and 0.08 keV for the decay from <sup>242</sup>Cm to <sup>238</sup>Pu, respectively.

The mass excess of  $^{238}$ Pu was measured with an uncertainty of 1.81 keV. Based on this measurement the mass-excess uncertainty of  $242$ Cm is 1.81 keV and the one of  $246$ Cf is 2.1 keV. Like in the previous two decay chains the mass excess uncertainties of the fermium and nobelium isotopes are limited by the uncertainty of the  $Q_{\alpha}$  measurements: The uncertainty of the mass excess of  $^{250}$ Fm is 8.9 keV and that of  $^{254}$ No is 17.3 keV.

The SHIPTRAP mass value of <sup>254</sup>No has an uncertainty of 14 keV and thus mainly determines the new AME value with a contribution of 60%. The mass-excess uncertainty of 254No is reduced to 11 keV and the uncertainty of the corresponding  $Q_{\alpha}$  value is reduced from 15 to 11 keV. The contribution to the mass excess of the daughter nuclide  $^{250}$ Fm is 16%. The influence on the lighter nuclides can be neglected.

The implementation of the new mass values of SHIPTRAP determined from the mass ratio to  $^{133}Cs$ , which is a primary nuclide, results in a connection to two different primary nuclides for each of the three nobelium isotopes. Now all nuclides of the data chains are influenced by each other and a loop of mutual dependence is achieved. Therefore, not only the nobelium isotopes but all other lighter nuclides in the decay chains become primary nuclides as well. Moreover, the direct mass measurements with SHIPTRAP confirm the mass values obtained from decay spectroscopy and thus the assigned nuclear level schemes, in particular for the decay chain involving 253No.

### **VII. INFLUENCE ON HEAVIER NUCLIDES**

The mass values of the higher *Z* nuclides in the three decay chains are affected similarly to the respective nobelium isotopes (i.e., their mass-excess values shift correspondingly). In the case of 252No the mass excesses were deduced from  $Q_{\alpha}$  values up to the hassium isotope <sup>264</sup>Hs [\[42\]](#page-8-0). However, the mass-excess uncertainties remain almost unchanged for these nuclides.

The implementation of the new mass-excess value of  $^{253}$ No leads to a slight reduction of the uncertainty of its mother nuclide <sup>257</sup>Rf from 20 to 18 keV. Since the decay spectrum of  $^{261}$ Sg has also been unambiguously identified [\[43\]](#page-8-0), its mass excess can also be determined with an uncertainty in the order of 20 keV. This value, however, has not been implemented into the AME yet and is thus not shown in Table [III.](#page-5-0) The mass excesses of the nuclides 265Hs and 269Ds are estimated from systematic trends.

The  $Q_{\alpha}$  value for the decay from <sup>258</sup>Rf to <sup>254</sup>No is known with an uncertainty of 30 keV [\[44\]](#page-8-0). This is the dominating contribution to the mass-excess uncertainty of 258Rf, which was reduced from 35 to 32 keV by implementing the new SHIPTRAP data. The three higher *Z* nuclides <sup>262</sup>Sg, <sup>266</sup>Hs, and  $^{270}$ Ds have been observed in the  $\alpha$ -decay chain of the latter. For the nuclide  $^{262}$ Sg, however, so far only spontaneous fission has been observed. An  $\alpha$ -branching ratio of 22% has been estimated [\[45\]](#page-8-0), but the  $\alpha$  branch has not yet been observed. Thus, the decay chain to the nuclide <sup>258</sup>Rf is not yet established. When this gap gets closed the masses of the three heavier nuclides will be connected to our nobelium result so that a firm anchor point at  $Z = 110$  will be obtained for improved predictions of the properties of superheavy nuclides.

### **VIII. CONCLUSIONS AND OUTLOOK**

The first direct mass measurements on transuranium elements result in unambiguous ground-state mass values independent from the knowledge of any nuclear level scheme. These results are valuable for the interpretation of the often complicated decay schemes of nuclides in this region. The SHIPTRAP results confirm the previously determined masses for all three nobelium isotopes and demonstrate that the  $\alpha$ -decay schemes were all correct. In particular the values are consistent with the level structure assignments within the decay scheme of  $253$ No. In addition, most of the AME 2003 mass values determined from decay energies have been slightly improved.

The new data contribute to the AME values not only for the nobelium isotopes but also for other nuclides connected to them via  $\alpha$ -decay chains. In this way the SHIPTRAP results have an impact on mass excesses and  $Q_{\alpha}$  values between fermium and hassium. All nuclides in the three decay chains up to the element nobelium are now primary nuclides. The area of nuclides with well-known mass values has been extended for the first time to the region of transuranium elements and thus moved a step closer to the expected island of stability [\[6,46,47\]](#page-8-0).

In addition, ground-state masses of nuclei that are very short-lived and thus not accessible for direct mass measurements in ion traps can be determined with the help of *α*-decay links. An experiment to measure the  $\alpha$  decay of <sup>262</sup>Sg is presently being prepared at SHIP. The  $\alpha$ -decay energy of <sup>262</sup>Sg will determine the masses of the decay chain members up to  $^{270}$ Ds.

It is planned to extend direct mass measurements with SHIPTRAP to higher *Z* nuclides. A major challenge is to fix the end points of decay chains of superheavy elements that are presently not linked to the region of known nuclides. This will be crucial for the identification and unambiguous assignment of the heaviest elements [\[48\]](#page-8-0).

The ongoing technical improvements include an increase of the efficiency of SHIPTRAP by a new cryogenic gas cell [\[15\]](#page-8-0). An optimized geometry of the new gas cell along with the operation temperature of about 40 K will lead to a higher stopping efficiency. Furthermore, a very high purity of the buffer gas can be achieved more easily as most impurities will freeze out.

<span id="page-8-0"></span>Another improvement will address an increase of the sensitivity and efficiency of SHIPTRAP for mass measurements of nuclides with production rates well below one particle per second. To this end a nondestructive detection scheme based on the measurement of induced image currents will be implemented at SHIPTRAP. This narrow-band Fourier transform ion cyclotron resonance (FT-ICR) technique [49] is expected to allow the measurement of the cyclotron frequency and thus a mass measurement with a single trapped ion. Such a system is about to be implemented and tested in the TRIGA-TRAP setup [50].

With these improvements, it will be possible to address further key nuclides on the way to the island of stability. In the near future SHIPTRAP will provide more anchor points to fix

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further decay chains, especially those consisting of nuclides with odd nucleon number. These will help to benchmark different nuclear models in the heavy and superheavy region of the nuclear chart.

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