# **Direct high-precision mass measurements on <sup>241</sup>***,***243Am, 244Pu, and 249Cf**

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The absolute masses of four long-lived transuranium nuclides,  $^{241,243}$ Am,  $^{244}$ Pu, and  $^{249}$ Cf, in the vicinity of the deformed  $N = 152$  neutron shell closure have been measured directly with the Penning-trap mass spectrometer TRIGA-TRAP. Our measurements confirm the AME2012 mass values of  $241.243$ Am and  $244$ Pu within one standard deviation, which were indirectly determined, by decay spectroscopy studies. In the case of the <sup>249</sup>Cf mass, a discrepancy of more than three standard deviations has been observed, affecting absolute masses even in the superheavy element region. The implementation of the mass values into the AME2012 network yields a reduced mass uncertainty for 84 nuclides, particularly for <sup>244</sup>Pu and its strongly correlated  $\alpha$  decay chains.

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## **I. INTRODUCTION**

Superheavy nuclides which have been observed even up to element number  $Z = 118$  [\[1\]](#page-6-0) owe their existence purely to quantum-mechanical shell effects. As the Coulomb repulsion rapidly increases with Z, these shell effects become more significant in heavy nuclei as they can enhance nuclear lifetimes tremendously near the shell closures. The next spherical shell closure beyond the doubly magic nuclei 208Pb is predicted to give rise to an "island of stability" of superheavy nuclei [\[2\]](#page-6-0). Indications for its existence come from the observation of isotopes with half-lives of seconds or even longer in the region of  $Z = 112-114$  [\[3\]](#page-6-0). However, its location in the nuclear landscape has been predicted at different proton numbers depending on the underlying theoretical models, some favoring  $Z = 114$ , while others prefer  $Z = 120$  or 126 [\[4–6\]](#page-6-0).

The experimental mass data of nuclides beyond uranium and information on their nuclear structure have been entirely relying on a network of nuclear transitions anchored to several uranium isotopes [\[7,8\]](#page-6-0). This network was predominantly established by decay energy measurements, affected by insufficient information on the level schemes and decay paths.

Penning traps became excellent tools for direct mass measurements. In particular nuclear structure studies benefited from high-precision mass measurements of radioactive nuclides in the neutron-rich and deficient region as well as in the region of the heaviest elements  $[10,11]$ . The SHIPTRAP facility at GSI Darmstadt was pioneering these efforts, which resulted in a total of six direct measurements of nuclides of nobelium ( $Z = 102$ ) and lawrencium ( $Z = 103$ ) [\[12](#page-6-0)[–14\]](#page-7-0). Thereby, new anchor points were established in the decay chains reducing the influence of nuclear transitions on the masses of superheavy nuclides. Certainly, more experimental data is needed to pin down the exact location of the island of stability.

A detailed network linking the masses of all nuclides was created in the Atomic-Mass Evaluation, the most recent one being the AME2012 [\[7,8\]](#page-6-0). Isolated parts of such networks, which are only connected to the nuclear chart via one nuclear transition, are vulnerable to measurement errors, in particular in regions where only scarce experimental data are available. This illustrates the substantial demand for further direct mass measurements of transuranium elements in order to ensure the accuracy of masses from the decay chains up to the heaviest elements. Furthermore, this approach on the determination of atomic masses and decay energies provides an independent test of the strength of the nuclear shell structure in this region, and can provide further insight into the deformed shell closures identified at  $N = 152$  (94  $\le Z \le 106$ ) and  $N = 162$  (Z > 105), which are currently under investigation [\[15\]](#page-7-0).

Penning-trap mass spectrometry provides the possibility to directly measure the energy difference between mother

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<span id="page-1-0"></span>

FIG. 1. Sketch of the TRIGA-TRAP setup [\[9\]](#page-6-0). Ions are produced in the laser ablation ion source and captured in the Penning traps. The mass measurement is carried out using the ToF-ICR technique where the flight time of the ions to an MCP detector outside the magnetic field is measured.

and daughter ground states, independent of sample specific parameters, thus offering a complementary, accurate approach on the determination of the strength of nuclear shell closures. Such a shell closure, for example, manifests as sudden drop in the two-neutron separation energy  $S_{2n}$ , for the determination of which the experimental masses of several nuclides around the shell closure need to be known precisely, in particular since shell effects in the heaviest elements are rather weak in comparison to the neutron shell closure at  $N = 126$  around  $208\overline{Pb}$ .

Within this work, the masses of four nuclides,  $241,243$  Am, <sup>244</sup>Pu, and <sup>249</sup>Cf, in the vicinity of the deformed  $N = 152$ neutron shell closure, were measured directly at TRIGA-TRAP [\[9\]](#page-6-0) and thereby connected to the atomic-mass standard  $12$ C. This paves the way for a detailed mapping of the evolution of this deformed shell closure as function of Z. While its presence is well established around nobelium [\[14\]](#page-7-0), evaluations of two-neutron separation energies suggest the shell closure to be less pronounced at lower proton numbers. Furthermore, measurements of the nuclear moment of inertia have revealed a weak shell closure for  ${}^{248}_{96}$ Cm<sub>152</sub> [\[16\]](#page-7-0) but none for the isotone  $^{250}_{98}$ Cf<sub>152</sub> [\[17\]](#page-7-0). A measurement of the size of the shell gap is best possible through further direct mass measurements, in particular as our mass measurement of  $249$ Cf indicates large discrepancies in the present mass values based on recent  $α$ -decay studies. They were performed off-line at mass uncertainties of 0.7–2 keV/ $c^2$  requiring a meticulous energy calibration of the spectrometer. However, such measurements could be affected by source parameters like variations in thickness and location of the source and the energy uncertainty of the used standard [\[18\]](#page-7-0).

## **II. EXPERIMENTAL PROCEDURE**

As part of the TRIGA-SPEC experiment [\[9\]](#page-6-0) the double Penning-trap mass spectrometer TRIGA-TRAP is dedicated to high-precision mass measurements. The experimental setup is displayed in Fig. 1 showing all key parts of TRIGA-TRAP

that are relevant for the present work. Ion production takes place in an upgraded version of a former laser ablation ion source described in [\[19\]](#page-7-0), using a frequency-doubled Nd:YAG laser for ablation at a wavelength of 532 nm and a pulse width of 5 ns carrying at maximum 50 mJ energy. In this newly developed version, the ion production is carried out inside a miniature radiofrequency quadrupole structure with two end-cap electrodes (mini RFQ) of about 4 cm length (see Fig. 2). At first, the ions are confined axially by a DC potential applied to the front and rear electrodes and radially by a superposition of DC- and RF-potentials applied to the nonsegmented rods. Within 5 ms of storage inside the mini RFQ, the ions' energy spread is reduced by collisions with helium atoms at typical gas pressures of about 10−<sup>3</sup> mbar at



FIG. 2. (Color online) 3D sketch of the laser ablation ion source including the mini RFQ for precooling the ions.

<span id="page-2-0"></span>

FIG. 3. Time-of-flight spectrum of carbon cluster ions from the laser ablation ion source to MCP 1 (see Fig. [1\)](#page-1-0). The DC-potential at the rods was chosen to favor the transmission of heavy clusters.

room temperature.<sup>1</sup> This increases the number of ions accepted of the setup, as the produced ion pulse has a large geometric and energetic distribution. Hence, samples with less material are required for mass measurements. By switching the DC potential of the front electrode within 100 ns, the ion bunch is extracted by an electrode in Pierce geometry [\[20\]](#page-7-0) and guided towards the Penning traps. For on-line operation radioactive fission products are ionized on a high-voltage platform. After mass selection, bunching, and cooling they are slowed down in a pulsed drift tube prior to delivery to TRIGA-TRAP [\[9](#page-6-0)[,21\]](#page-7-0).

At TRIGA-TRAP carbon cluster ions are used as mass references being never more than six mass units different from the masses of the ions of interest. To visualize the carbon cluster  ${}^{12}C_n^+$  (8 < n < 31) distribution of an ion bunch created from a SigradurTMplate, its time-of-flight distribution to a microchannel plate detector (MCP) about 2 m downstream in front of the superconducting magnet (MCP 1) is recorded. With the old concept described in [\[19\]](#page-7-0) a distinction of ions with different  $q/m$  ratio based on their flight time was not possible. The cooling in the mini RFQ yields a resolution as shown in Fig. 3, where each peak corresponds to a cluster with a different number of  ${}^{12}C$  atoms. For the selection of heavy cluster ions, the mini RFQ is operated as a mass filter. A potential difference between neighboring rods is introduced while opposing rods are kept at the same potential. Thus, the storage of light cluster ions is suppressed. The heaviest ion which is clearly distinguishable in the time-of-fight spectrum is  ${}^{12}C_{27}^+$ , whereas even heavier cluster ions are concealed in the continuous part of the spectrum at 80  $\mu$ s and above.

For the production of long-lived transuranium ions, a droplet of 10  $\mu$ l nitric acid containing not more than 10<sup>15</sup> atoms of the isotope under investigation was placed on the Sigradur backing and evaporated to dryness. This resulted in the creation of rings in which the majority of the solved material was distributed as shown in radiographic images [\[22\]](#page-7-0) of an unused as well as a used but different target in Fig. 4. The blue circle



FIG. 4. (Color online) Radiographic images of two different targets with  $10^{15}$  atoms <sup>249</sup>Cf per spot, (a) an unused and (b) a used one. The color is a measure of the radioactivity, ranging from high activity (red) to low activity (blue). The dashed circle in (b) represents the laser accessible region when the target is rotated.

represents the Sigradur plate, the dashed line indicates the laser path on the rotatable target. Due to several small apertures, which the laser beam has to pass, only a small fraction of the radioactive material is accessible.  $α$ -decay measurements showed that more than 90% of the isotopes of interest remained on the target. The number of radioactive atoms on the target was always determined by radiometric techniques. For future use, investigations on several backing materials, in particular with superhydrophobic surfaces, successfully demonstrated an increased yield [\[23\]](#page-7-0) and open a way to improve target preparation as well as ion production at yet reduced sample amounts.

The efficiency of the sample usage in the mass spectrometers was greatly improved by the mini RFQ, since ions from the laser ablation plume were captured and cooled *in situ*, in contrast to the previous approach. As an additional benefit of the cooling, the axial emittance of the ions is sufficiently reduced to separate a different carbon cluster specimen by using a beam gate. All ions of one species were captured in the purification trap due to their lower energy spread.

The Penning traps are located inside the 7-T superconducting magnet. In a Penning trap, a superposition of the strong homogeneous magnetic field  $\vec{B}$  and a weak electrostatic quadrupole field  $\vec{E}$  confines the ions in three dimensions. The ion motion is characterized by three eigenmotions: axial, magnetron, and modified cyclotron motion with their eigenfrequencies  $v_z$ ,  $v_$ , and  $v_+$ , respectively [\[24\]](#page-7-0). Inside the Purification trap, one specimen is selected from the entire ensemble by means of mass selective buffer gas cooling [\[25\]](#page-7-0). Subsequently, the cooled ions are transferred through a differential pumping barrier with an inner diameter of 1.5 mm to the precision Penning trap.

The mass measurement is carried out via a determination of the free cyclotron frequency of the ion:

$$
\nu_c = \frac{1}{2\pi} \frac{q}{m_{\text{ion}}} B \tag{1}
$$

with charge-to-mass ratio  $q/m$  oscillating in a magnetic field with strength  $B$ , by employing the time-of-flight ioncyclotron-resonance technique (TOF-ICR) [\[26,27\]](#page-7-0). To this end the conversion from the magnetron into the reduced cyclotron motion is probed with a quadrupolar rf field around

<sup>&</sup>lt;sup>1</sup>The helium with 99.999% purity is led into the mini RFO through a small nozzle with a needle valve.

<span id="page-3-0"></span>

FIG. 5. (Color online) Time-of-flight resonance of  $^{243}Am^{16}O^{+}$ recorded with the Ramsey excitation profile applying two 200 ms long pulses and 1600 ms waiting time in between.

the side-band  $v_{\text{rf}} = v_+ + v_- = v_c$  by the determination of the time of flight of the ions to MCP 2 outside the magnetic field. The time of flight recorded as function of the excitation frequency results in a minimum for resonant excitation at  $v_c$ . At TRIGA-TRAP, instead of continuous RF excitation of the ion motion, the so-called Ramsey technique is applied using a two-pulse excitation with a long waiting time in between for a gain in precision [\[28,29\]](#page-7-0).

However, the measurement of  $v_c$  of the isotopes of interest is not sufficient since in Eq. [\(1\)](#page-2-0) the magnetic field strength remains unknown. Thus, the same measurement is performed with a reference ion prior and subsequent to the measurement of the ion of interest. From these two frequencies, the magnetic field at the time of the measurement of the ion of interest is interpolated. A carbon cluster ion  ${}^{12}C_n^+$  being an integer multiple of the atomic mass standard with similar  $q/m$  is chosen as reference for minimizing mass dependent systematic shifts [\[30\]](#page-7-0). The mass of the neutral atom is then derived as

$$
m = \frac{v_{\rm c,ref}}{v_{\rm c}} (m_{\rm ref} - m_{\rm e}) + m_{\rm e},\tag{2}
$$

where  $v_{c,ref}$  and  $(m_{ref} - m_e)$  denote cyclotron frequency and mass of the reference ion and  $m<sub>e</sub>$  the electron mass. Within this work the masses of four long-lived transuranium nuclides,  $241,243$  Am,  $244$  Pu, and  $249$  Cf have been determined. As in previous experimental work it was observed that the singly charged monoxide ions yield the highest production rates

being thus used for the mass measurements [\[31\]](#page-7-0). The atomic mass is calculated by simply subtracting the mass of  $^{16}O$ [\[8\]](#page-6-0) from the oxide mass, since the chemical binding energy on the order of eV can be neglected at the present limit of precision. As the samples were isotopically pure and all molecular contaminations in a cyclotron frequency range of  $\pm$ 3 Hz could be ruled out due to chemical reasons no disturbing contaminants were present.

The cyclotron frequencies were measured by the TOF-ICR technique employing a Ramsey excitation profile with two 200 ms long pulses and 1600 ms waiting time in between. A complete description of the measurement procedure is given in  $[32]$ . For each isotope  $10^{15}$  atoms placed on the Sigradur target were sufficient to perform an entire mass measurement, consisting of 15 to 23 recorded cyclotron resonances of the ion of interest, each containing at least 500 ion events. A time-of-flight resonance of  $^{243}$ Am<sup>16</sup>O<sup>+</sup> is shown in Fig. 5. The laser power for the creation of the ions was optimized to keep the count rate at about one detected ion per cycle.

Effects such as a trap misalignment in the magnetic field may cause small frequency shifts, still allowing cyclotron frequency measurements via the sidebands for small tilting angles [\[33\]](#page-7-0). They result in a shift in the frequency ratio  $r = \frac{v_{\text{c,ref}}}{v_{\text{c}}}$  and therefore in the calculated mass of the ion of interest, depending on the mass difference to the reference ion. Although carbon cluster ions provide a reference having a mass close to every ion of interest, this mass shift can be noticeable. In turn, carbon cluster ions are well suited for the investigation of this effect. As all nuclides studied within this work lie within a very limited mass range, this was done locally. All three possible cyclotron frequency ratios between the carbon clusters  ${}^{12}C_{21;22;23}^+$  were recorded and compared. The determined mean frequency ratios  $r$  and their statistical uncertainties are shown in Table I. The mass excess is defined as  $ME = (m - A \times u)c^2$  and is essentially zero for carbon clusters as their chemical binding energy [\[34\]](#page-7-0) is negligible at the present precision level of TRIGA-TRAP. Two of the mass excesses deviate slightly from the theoretical value. The weighted mean of the deviation from the theoretical value,  $\Delta r$ , being referred to as mass-dependent shift, was found to be in agreement with zero:

$$
\epsilon_m = -1.5(2.3) \times 10^{-10} / \mathbf{u}.\tag{3}
$$

Nevertheless, it is still considered in the evaluation process as is the uncertainty originating from nonlinear magnetic field fluctuations discussed in [\[32\]](#page-7-0). Systematic effects due to

TABLE I. Calibration measurements of carbon cluster ions. The deviation  $\Delta r$  of the measured cyclotron frequency ratio  $r$  from the theoretical value was determined. For clarity the deviations from the mass excess  $\Delta ME$ , uncorrected and corrected for the mass dependent shift from Eq. (3), which in theory is zero, are given.

<b>Ion</b>	Reference	r	$\Delta r/10^{-9}$	ΔME (keV)	$\triangle ME$ (keV) corrected
${}^{12}C_{22}^+$	${}^{12}C_{21}^+$	1.0476191447(56)	6.6(5.6)	$-1.6(1.3)$	$-2.0(1.5)$
${}^{12}C_{23}^+$	${}^{12}C_{22}^+$	1.0454546446(64)	$-4.7(6.4)$	1.2(1.6)	0.7(1.7)
${}^{12}C_{23}^+$	${}^{12}C_{21}^+$	1.0952383114(73)	$-8.9(7.3)$	2.1(1.7)	1.2(2.2)

temperature fluctuations of the setup were eliminated by the implementation of stabilization systems prior to this work. The carbon cluster  ${}^{12}C_{22}^+$  was chosen as mass reference due to its mass being close to that of the investigated ion species. During the experiment the ion species were swapped between ion-ofinterest and reference ion by rotating the target so that the laser hits either the radioactive material or the bare carbon surface.

## **III. RESULTS**

The measurements reported here uniquely link the masses of some transuranium nuclides to the atomic-mass standard. The masses in our studied region are so far determined by α decay chains, connected via  $β^{\pm}$  decays or nuclear reactions, being ultimately linked to an absolute measurement of uranium. They form a complex network being tested for self-consistency by least-squares analyses, which are still vulnerable to single wrong measurements.

The results obtained in the recent measurement campaign are presented in Table II. Average frequency ratios  $r$  are listed with purely statistical uncertainties. The mass-excess values result from  $r$  corrected for the mass-dependent shift from Eq. [\(3\)](#page-3-0) and are presented in chronological order of their measurement. The final TRIGA-TRAP values corrected for the mass-dependent shift from Eq. [\(3\)](#page-3-0) are displayed in Fig. 6 and compared to the AME2012 values [\[8\]](#page-6-0). The dashed line marks the AME2012 values and the solid black lines the respective 1  $\sigma$  uncertainty. In the following the elements are discussed individually.

# **A. <sup>241</sup>***,***243Am**

The two americium isotopes are rather long-lived and consequently only moderately radioactive and served as ideal candidates for performance tests of the mini RFQ. Compared to the old laser ion source design, an ion production efficiency gain of more than one order of magnitude was observed. In total, the flight times of 8738 ion events distributed over 15 timeof-flight resonances for  $^{241}Am^{16}O^+$  and of 9789 ion events contained in 21 resonances for  $243$ Am<sup>16</sup>O<sup>+</sup> were recorded. From this data corrected mass excesses of  $ME_{TT}(^{241}Am)$  = 52936.9(1.8) keV and  $ME_{TT}(^{243}Am) = 57176.2(1.4)$  keV were calculated, corrected for the mass-dependent shift. Both are in excellent agreement with the values published in the AME2012 and prove the TRIGA-TRAP experiment performance to be suitable for direct high-precision mass



FIG. 6. Illustration of the results from Tables [I](#page-3-0) and II. Both are corrected by the mass dependent shift from Eq. [\(3\)](#page-3-0). The solid lines mark the  $1\sigma$  error of the AME2012 values.

measurements of long-lived transuranium isotopes even below the  $10^{-8}$  level.

#### **B. 244Pu**

The uncertainty of the mass excess of  $244$ Pu as given in the AME2012 is 5 keV. This originates from its connection to the remaining network of nuclides, the nuclear reaction  $^{244}Pu(d,t)^{243}Pu$ , dominating the uncertainty of several nuclides in the  $\alpha$  decay chains of <sup>244</sup>Pu and <sup>249</sup>Cm. A measurement series with 26 frequency ratios containing 16299 ion events yielded the corrected mass excess  $ME_{TT} = 59806.2(1.8)$  keV, which reduces the uncertainty by a factor of 2.8.

# **C. 249Cf**

The fourth investigated nuclide,  $249Cf$ , is of special interest as its neutron number is closest to the deformed shell closure  $N = 152$ . From the 24 recorded resonances with 13818 ion events, with a corrected mass excess value of  $ME_{TT} = 69718.1(1.3)$  keV was obtained. This shows a significant deviation of 7.9 keV, i.e., more than  $3\sigma$  from the AME2012 value. This is surprising, as the masses of <sup>252–255</sup>No and <sup>255,256</sup>Lr (situated near or at the  $N = 152$ shell closure) as contained in the AME predating the SHIPTRAP measurements were confirmed by the latter [\[12](#page-6-0)[–14\]](#page-7-0). Since a connection between <sup>249</sup>Cf and <sup>241</sup>Am, whose mass was confirmed within this work, is established via two α and one β decay, it must be assumed that at least one energy of the transitions <sup>249</sup>Cf( $\alpha$ )<sup>245</sup>Cm, <sup>245</sup>Cm( $\alpha$ )<sup>241</sup>Pu, or  $^{241}Pu(B^-)^{241}$  Am might be wrong. This appears most unlikely

TABLE II. Results of the mass measurements in the transuranium region. The ion chosen for the mass measurement is given, together with the frequency ratio to the reference ion  ${}^{12}C_{22}^+$  and the relative uncertainty  $\delta r/r$ . The mass excesses refer to the atom measured at TRIGA-TRAP and the value given in the AME2012 [\[8\]](#page-6-0) with their differences to the AME2012  $\Delta ME^{atom} = ME^{atom}_{TT} - ME^{atom}_{AME2012}$ .

Ion	to ${}^{12}C_{22}^+$	$\delta r/r$ $/ 10^{-9}$	$ME_{TT}^{atom}$ (keV)	$ME_{AME2012}^{atom}$ (keV)	$\triangle ME^{atom}$ (keV)
$^{241}$ Am <sup>16</sup> O <sup>+</sup>	0.9736807966(72)	7.4	52936.9(1.8)	52936.2(1.8)	0.7(2.5)
$^{243}$ Am <sup>16</sup> O <sup>+</sup>	0.9812738087(56)	5.7	57176.2(1.4)	57176.3(2.3)	$-0.1(2.7)$
$^{244}$ Pu <sup>16</sup> O <sup>+</sup>	0.9850723904(72)	7.3	59806.2(1.8)	59807(5)	$-0.8(5.3)$
$^{249}Cf^{16}O^+$	1.0040521297(52)	5.2	69718.1(1.3)	69726.0(2.2)	$-7.9(2.5)$

<span id="page-5-0"></span>

FIG. 7. Links between <sup>241</sup>Am and <sup>249</sup>Cf used in the AME2012. The total decay energies, given in keV, are calculated in [\[8\]](#page-6-0) from the original energy of the  $\alpha$  or  $\beta$  particle extracted from the references [\[35–44\]](#page-7-0) considering the recoil of the nucleus.

for the last one, which was measured independently by four groups [\[35–38\]](#page-7-0) with all results agreeing with each other. Additionally, the nuclides  $^{245}$ Cm,  $^{245}$ Am,  $^{249}$ Bk, and  $^{249}$ Cf are interconnected in a loop resulting in an interdependence of their masses. This small part of the network is shown in Fig. 7 with the experimental data adopted from [\[35–44\]](#page-7-0).

## **IV. DISCUSSION**

Due to the vast amount of data interconnecting the masses of nuclides, these are overdetermined in most cases.

The evaluation is performed using a least-squares fit to all experimental data. For the implementation of our data into the AME the procedure given in [\[7\]](#page-6-0) and in particular the one for Penning traps described in [\[45\]](#page-7-0) is applied. The implementation of the mass of 249Cf is not straightforward. Indeed, the reduced  $\chi^2$  increases drastically if the newly measured mass is implemented directly. However, for the decay  $^{249}Bk(\alpha)^{245}Am$  two different energies are reported indicating an inconsistency. Remarkably, Baranov *et al.* [\[40\]](#page-7-0) measured a 5.7 keV higher  $\alpha$ -particle energy than Ahmad *et al.* [\[42\]](#page-7-0). The latter repeated this measurement recently,



FIG. 8. (Color online) Relative changes in the mass uncertainties between the AME2012 with and without the implementation of the recent TRIGA-TRAP mass measurement data. No change in the uncertainty is represented by the value 1. Due to the decoupling procedure the uncertainty of <sup>245</sup>Cm is increased while the mass uncertainty of 84 nuclides got decreased.

<span id="page-6-0"></span>reproducing their formerly obtained value [\[18\]](#page-7-0). Ahmad *et al.* in [\[18\]](#page-7-0) also address ambiguous energy calibrations in the latest data treatment by Baranov *et al.* [\[46\]](#page-7-0). Furthermore, this discrepancy questions the energy measurements of the other two α decays, having been carried out by Baranov *et al.* as well. Thereby, the absolute masses of the four nuclides within this loop have to be questioned. As the nuclei in the loop are end-points of decay chains originating in the region of superheavy elements, even masses of remotely situated nuclei are affected. For completeness it should be mentioned that the energy of the decay  $2^{245}Am(\beta^-)^{245}Cm$  has a comparably large uncertainty while the energy of the <sup>249</sup>Bk( $\beta$ <sup>-</sup>)<sup>249</sup>Cf decay is confirmed by two independent measurements and is reported with higher precision.

Since the implementation of the new mass data into the AME2012 requires to consider this discrepancy, the uncertainty of the three questionable  $\alpha$  decay energies was increased to 6 keV. This effective decoupling of  $249$ Bk and  $249$ Cf caused a shift of the absolute masses of the 20 nuclides in the aforementioned  $\alpha$  decay chains, including <sup>245</sup>Am and 245Cm, while the remaining nuclides are unaffected. In addition, the recent mass measurement also had an effect on the uncertainty of further nuclides as depicted in Fig. [8,](#page-5-0) where the relative changes of the mass uncertainties between the AME2012 including and excluding the latest TRIGA-TRAP data are displayed. In total, the mass values of 84 nuclides were improved by our measurements reported here. Only the uncertainty of  $245$ Cm became larger by a factor of 1.7 due to the decoupling procedure. The largest improvement has been achieved for all nuclides in the  $\alpha$  decay chain of <sup>244</sup>Pu as well as their strongly correlated neighbors.

### **V. CONCLUSION**

In this publication we reported on direct mass measurements of the transuranium nuclides <sup>241,243</sup> Am, <sup>244</sup> Pu, and <sup>249</sup> Cf and their impact on the Atomic-Mass Evaluation 2012. The measurements performed here used only  $10^{15}$  atoms target material demonstrating a significant increase in the efficiency of the TRIGA-TRAP ion source as well as the opportunities for further mass measurements on even smaller sample sizes. Since the direct mass measurements were performed in the vicinity of the deformed shell closure  $N = 152$ , they influence the mass of the nuclides at the shell closure significantly. While the mass measurement of 244Pu decreased the uncertainty of several nuclides by more than a factor of two, the deviation of the 249Cf mass urges for further mass measurements in this region. In particular its closer vicinity has to be subjected to investigation in order to resolve the present discrepancies. A direct mass measurement of  $245$ Cm, for instance, would be required to clarify whether the energies of  $^{249}Cf(\alpha)^{245}Cm$ and  $2^{45}$ Cm( $\alpha$ )<sup>241</sup>Pu were determined correctly and thereby to support either of the two published  $^{249}Bk(\alpha)^{245}Am$  energies. Further measurements are necessary to test the selfconsistency of the loop of  $^{249}$ Cf,  $^{249}$ Bk,  $^{245}$ Cm, and  $^{245}$ Am, as it is only connected to the nuclide chart via  $^{245}$ Cm( $\alpha$ )<sup>241</sup>Pu.

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