Regular Article - Experimental Physics



High-precision mass measurement of doubly magic ²⁰⁸Pb

Kathrin Kromer^{1,a}, Chunhai Lyu¹, Menno Door¹, Pavel Filianin¹, Zoltán Harman¹, Jost Herkenhoff¹, Wenjia Huang², Christoph H. Keitel¹, Daniel Lange^{1,3}, Yuri N. Novikov^{4,5}, Christoph Schweiger¹, Sergey Eliseev¹, Klaus Blaum¹

¹ Max-Planck-Institut für Kernphysik, 69117 Heidelberg, Germany

² Advanced Energy Science and Technology Guangdong Laboratory, Huizhou 516007, China

³ Ruprecht-Karls-Universität Heidelberg, 69117 Heidelberg, Germany

⁴ Department of Physics, St Petersburg State University, Saint Petersburg 198504, Russia

⁵ NRC "Kurchatov Institute" Petersburg Nuclear Physics Institute, Gatchina 188300, Russia

Received: 7 June 2022 / Accepted: 12 October 2022 / Published online: 25 October 2022 © The Author(s) 2022, corrected publication 2022 Communicated by Navin Alahari

Abstract The absolute atomic mass of ²⁰⁸Pb has been determined with a fractional uncertainty of 7×10^{-11} by measuring the cyclotron-frequency ratio R of $^{208}\text{Pb}^{41+}$ to ¹³²Xe²⁶⁺ with the high-precision Penning-trap mass spectrometer PENTATRAP and computing the binding energies E_{Pb} and E_{Xe} of the missing 41 and 26 atomic electrons, respectively, with the *ab initio* fully relativistic multiconfiguration Dirac-Hartree-Fock (MCDHF) method. R has been measured with a relative precision of 9×10^{-12} . $E_{\rm Pb}$ and E_{Xe} have been computed with an uncertainty of 9.1 eV and 2.1 eV, respectively, yielding 207.976650571(14) u $(u = 9.3149410242(28) \times 10^8 \text{ eV/c}^2)$ for the ²⁰⁸Pb neutral atomic mass. This result agrees within 1.2σ with that from the Atomic-Mass Evaluation (AME) 2020, while improving the precision by almost two orders of magnitude. The new mass value directly improves the mass precision of 14 nuclides in the region of Z = 81-84 and is the most precise mass value with A > 200. Thus, the measurement establishes a new region of reference mass values which can be used e.g. for precision mass determination of transuranium nuclides, including the superheavies.

1 Introduction

Heavy and superheavy nuclides beyond the doubly magic nucleus of 208 Pb can only exist due to nuclear shell effects holding them together by counteracting the rapidly increasing Coulomb repulsion with growing proton number Z [1]. Insight into these quantum-mechanical nuclear structure effects can be derived from the masses of such nuclides. In

addition to some direct heavy mass measurements [2-5], a network of nuclear transitions and relative mass measurements, i.e. the Atomic-Mass Evaluation (AME), provides mass values for most heavy and superheavy nuclides by tracing them back to a few well-known masses of uranium isotopes [6]. However, no nuclide beyond Z = 70 can be found whose mass is known to a relative precision of better than 2×10^{-9} to act as a precise reference point for these heavy elements. This directly limits the achievable precision in the heavier mass regions and can possibly lead to tensions or shifts of the relative measured masses due to their referencing to only one reference point. The limitations by mass dependent shifts can be reduced significantly once there is a reference mass with similar mass known to high precision [7]. The need for new anchor points for the AME arose during recent mass measurements with TRIGA-TRAP [5,8] at the research reactor TRIGA in Mainz, specifically, an improved absolute mass of ²⁰⁸Pb [9]. Measuring this mass will also directly improve the masses of several Pb isotopes and other nuclides in this mass region [6].

In addition to the impact as a mass reference for other mass measurements, the mass of 208 Pb will soon be needed when the magnetic moment, or the *g*-factor, of the bound electron of hydrogen-like 208 Pb is planned to be determined by the Penning-trap experiments Alphatrap at the MPIK in Heidelberg [10] and Artemis at GSI Darmstadt [11]. This measurement could be the most stringent test of bound-state quantum electrodynamics in strong fields. The error of the mass of the nucleus, however, enters the error budget and therefore needs to be known to high precision [12]. With the results of this paper, the error of the mass of 208 Pb will be negligible in future *g*-factor determinations.

^a e-mail: kathrin.kromer@mpi-hd.mpg.de (corresponding author)

Based on the accurate absolute mass of 132 Xe [13, 14], in this paper, we present a determination of the absolute atomic mass of 208 Pb with a fractional uncertainty of 7×10^{-11} . This is the result of measuring the cyclotron-frequency ratio of 208 Pb⁴¹⁺ and 132 Xe²⁶⁺ with the high-precision Penningtrap mass spectrometer PENTATRAP [15, 16] in combination with a computation of the binding energies of the missing 41 and 26 atomic electrons, respectively, using the *ab initio* fully relativistic multi-configuration Dirac–Hartree–Fock (MCDHF) method. The masses of 132 Xe²⁶⁺ and 208 Pb⁴¹⁺ can be related to their neutral counterparts via

$$m\left(^{132}\mathrm{Xe}^{26+}\right) = m\left(^{132}\mathrm{Xe}\right) - 26m_e + E_{\mathrm{Xe}},\tag{1}$$

$$m\left(^{208}\mathrm{Pb}^{41+}\right) = m\left(^{208}\mathrm{Pb}\right) - 41m_e + E_{\mathrm{Pb}},\tag{2}$$

with $m_e = 5.48579909065(16) \times 10^{-4}$ u being the electron rest mass [17] and $m (^{132}\text{Xe}) = 131.904155086(10)$ u being the mass of a neutral ¹³²Xe atom [13,14], each has a relative accuracy of 2.9×10^{-11} and 7.6×10^{-11} , respectively. E_{Xe} and E_{Pb} are the binding-energy differences that represent the energies required to ionize the outermost 26 and 41 electrons, respectively, from neutral Xe and Pb atoms. With the mass ratio

$$R = \frac{m \left({}^{208} \text{Pb}^{41+}\right)}{m \left({}^{132} \text{Xe}^{26+}\right)} \tag{3}$$

being experimentally measured, one can improve the accuracy of the absolute mass of 208 Pb via

$$m \left({}^{208}\text{Pb} \right)$$

= $R \left[m \left({}^{132}\text{Xe} \right) + 26m_e - E_{\text{Xe}} \right]$
+ $41m_e - E_{\text{Pb}}$, (4)

based on the theoretically calculated E_{Xe} and E_{Pb} . By improving the mass of ²⁰⁸Pb the masses of other Pb isotopes and nearby elements can be improved accordingly since they are linked via decays of which the energy has been measured.

2 Experimental and theoretical methods

If one introduces a charged particle into a magnetic field *B*, it will describe a free space cyclotron motion with the frequency $\omega_c = \frac{q}{m}B$, with q/m being the charge-to-mass ratio. The working principle of a Penning trap is based on a strong homogeneous magnetic field in combination with an electrostatic quadrupole potential. While the electrostatic potential prevents the ion from escaping in axial direction, forcing it onto an oscillatory axial motion with frequency ω_z , the magnetic field forces the ion in radial direction onto a circular orbit with a modified cyclotron frequency ω_+ . The

cross product of the two fields in the Lorentz equation leads to an additional slow drift around the trap center called magnetron motion with frequency ω_{-} . When comparing these three Penning-trap eigenfrequencies to the movement of a free charged particle in a purely magnetic field, it holds [18]:

$$\omega_c = \sqrt{\omega_+^2 + \omega_z^2 + \omega_-^2} \,. \tag{5}$$

From this equation we can see that the determination of eigenfrequencies of an ion in a Penning trap can be used to determine its mass, if the magnetic field inside the trap is known. However, a determination of a magnetic field of $B \approx 7 \text{ T}$ inside a volume of just a few 10 μ m³ to sufficient precision is not possible. Therefore, a relative measurement is chosen at PENTATRAP, using a reference ion and a sequential measurement scheme to determine mass ratios [15]. Highly charged ions are used due to the advantage that with higher q/m the modified cyclotron frequency increases and can therefore be measured to a higher relative precision. For each mass determination a reference nuclide and charge states have to be chosen that form a q/m doublet with the nuclide of interest in order to largely suppress systematic effects in the cyclotronfrequency ratio determination [15, 16]. The advantage being, that with q/m doublets the same trapping voltage can be used to match the axial frequency to the detection tank circuit's resonance frequency. Using the same trapping voltage reduces systematic shifts due to trap anharmonicities. In addition, the absolute mass of the reference nuclide has to be known better than the aimed uncertainty of the mass of the nuclide of interest. More technical restrictions are posed by the production of the reference ion, limited by binding energies and the availability of probe material. For these reasons, the near q/m doublet ²⁰⁸Pb⁴¹⁺ (q/m = 0.197138 e/u) and ¹³²Xe²⁶⁺ $(q/m = 0.197\,113\,e/u)$ [13,14] was chosen. The ¹³²Xe²⁶⁺ ion was created from a gaseous natural source inside a commercial Dresden electron beam ion trap (DREEBIT) [19,20]. The DREEBIT is connected to a beamline with a large bender magnet for q/m selection, see Fig. 1a upper beamline. The ²⁰⁸Pb⁴¹⁺ ion was produced in a Heidelberg Compact electron beam ion trap (compact EBIT) [21] equipped with an in-trap laser-desorption target of monoisotopic ²⁰⁸Pb [22]. After ion breeding, the q/m selection was achieved using the time-offlight separation technique with fast high-voltage switches recently developed at the MPIK [23], supplying the voltages to a Bradbury-Nielson gate [24], see Fig. 1a lower beamline. Once the ions were selected and decelerated by two pulsed drift tubes, they were consecutively trapped in the first of PENTATRAP's five traps and transported down to their individual traps.

Due to the five stacked Penning traps available, see Fig. 1b, a simultaneous measurement in two traps is possible, increasing the measurement speed by higher statistics and offering up the opportunity for cross checks between the traps

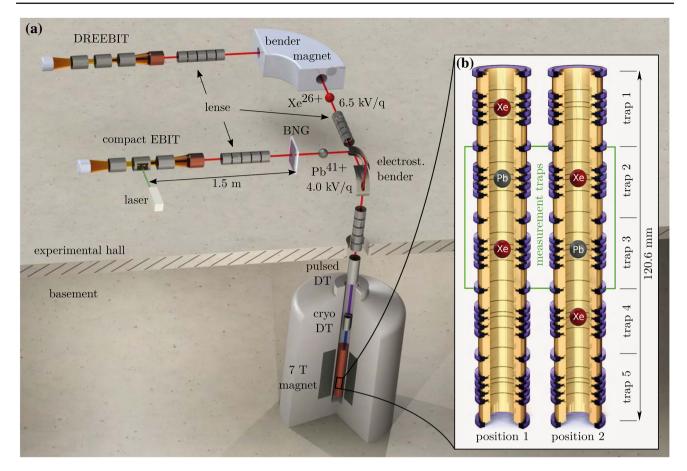


Fig. 1 a Schematic illustration of the ion production section, the two beamlines, and the combined deceleration region ending in the trap chamber. The ion bunches with their respective energies are shown after they have been q/m selected by the bender magnet or the Bradbury-Nielson gate (BNG). **b** Schematic drawing of the Penning-trap tower

with two measurement ion configurations. The ions are moved from position 1–2 or back every \approx 15 min. The frequency measurements are carried out in traps 2 and 3. Traps 1 and 4 are utilized as storage traps and trap 5 is currently not in use but is planned to be used to monitor magnetic field fluctuations in the future

and several analysis methods. Out of the other three traps, two are needed for ion storage and one trap is planned for monitoring, however, currently not in use.

The ion's frequencies depend on the magnetic field and the electrostatic potential. All environmental influences on these quantities need to be stabilized over the duration of the measurement. For this, the PENTATRAP laboratory is temperature-stabilized to $\delta T < 50$ mK/h and the height of the liquid helium level $z_{\rm lHe}$ used for cooling the superconducting magnet, Penning traps, and the detection system is stabilized to $\delta z_{\rm lHe} < 1$ mm/h along with the pressure of helium gas inside the magnet's bore to $\delta p < 10 \,\mu$ bar/h [15].

We employ the Fourier-transform ion-cyclotron-resonance detection technique [25] using cryogenic tank circuits connected to the Penning traps to pick up the small image current induced in the trap electrodes by the ion. The largest frequency ω_+ , and therefore the frequency with the highest contribution to the overall error, is measured using the phase-sensitive pulse and phase (PnP) method [26,27]. This method, described in more detail below, sets an initial phase of the reduced cyclotron frequency, then the motion is left decoupled for a variable phase accumulation time t_{acc} during which the phase can evolve freely, before reading out the final phase ϕ_{meas} . The other two frequencies ω_z and ω_- are measured with the Fast-Fourier-Transform (FFT) dip and the double-dip technique, respectively [28].

The measurements of 208 Pb⁴¹⁺ versus 132 Xe²⁶⁺ were carried out with the measurement scheme shown in Fig. 2. After a rough estimate of all three frequencies of both ions in both positions, shown in Fig. 1b, using the dip and double-dip technique, the measurement run starts with an *N* determination, with *N* being the integer number of full turns of the reduced cyclotron motion during the phase accumulation time. This preparatory measurement is necessary before the actual PnP measurement because the cyclotron phase of the ion increases linearly with time with the increment of the frequency ω_+ and will thereby pass a full turn of 2π many times during the phase accumulation time. The inte-

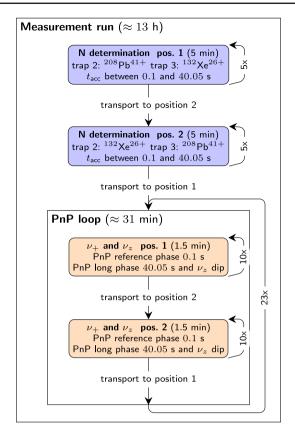


Fig. 2 The figure depicts a flowchart of the measurement scheme and the relevant measurement times during the lead mass campaign. In blue at the top one can see the initial N determination followed by the main measurement PnP loop in orange with the simultaneous measurement of axial frequency and modified cyclotron phase. The ions are frequently swapped in order to minimize the magnetic field drift between position 1 and 2

ger N needs to be known in order to determine the modified cyclotron frequency through the total accumulated phase

$$\phi(t_{\rm acc}) = t_{\rm acc} \times \omega_+ = 2\pi \times N + \phi_{\rm meas} , \qquad (6)$$

with ϕ_{meas} being the measured phase at the end of the accumulation time t_{acc} . The N determination utilizes 9 different phase accumulation times between 0.1 and 40.05 s and finds an ω_+ for which each N for all accumulation times is integer.

A constant phase offset, unavoidable due to the phase readout, is cancelled out by subtracting a short reference phase with an accumulation time of 0.1 s from each long measurement phase. After concluding the *N* determination in both traps for lead and xenon, the actual PnP loop is started, see Fig. 2 lower half. Here, a starting phase is imprinted on the modified cyclotron motion by an ω_+ dipole excitation pulse. The phase can then evolve freely for t_{acc} before the final modified cyclotron phase is imprinted on the axial motion by a coupling π -pulse, where it can be detected as an axial phase [26,27]. All excitation and coupling pulses are shaped with a Tukey window [29] in order to avoid systematic phase shifts during the phase readout. During the phase evolution time of the PnP sequence, an FFT axial-dip measurement is performed. This simultaneous phase determination and dip detection leads to a reduction of systematic effects associated with the temporal variation of the trap potential and the magnetic field, since they cancel out when calculating the free cyclotron frequency, using the invariance theorem in Eq. (5). After repeating the measurement of the two ions in trap 2 and 3 ten times, the ions are swapped. If ${}^{132}Xe^{26+}$ was in the trap, then ²⁰⁸Pb⁴¹⁺ is swapped in and vice versa. This is repeated for around 12 hours before restarting the whole measurement scheme again with the N determination. The magnetron frequency, being a factor $\approx 1,600$ smaller than the modified cyclotron frequency, does not need to be measured repeatedly since the double-dip determination during the preparation phase is sufficiently precise. After one measurement run a relative statistical uncertainty of $\approx 10^{-11}$ is reached.

To determine a neutral mass of ²⁰⁸Pb from the ions' free cyclotron frequency ratio we need to include the mass of the missing electrons in combination with their binding energies (in the following we always refer to the absolute binding energies). We employ the *ab initio* fully relativistic multiconfiguration Dirac-Hartree-Fock (MCDHF) method [30–32] to compute the binding energies E_{Xe} and $E_{\rm Pb}$ of the outermost 26 and 41 electrons, respectively, in neutral Xe and Pb atoms. First, for the case of Xe, the ionization energy of the outermost 8 electrons has been experimentally determined to be of 424.7(7) eV [33]. Thus, one only needs to calculate the binding-energy difference between the ground states of Xe^{26+} ([Ar] $3d^{10-1}S_0$) and Xe^{8+} ([Kr]4 d^{10} ¹ S_0). Similarly, since the ionization energy of the outermost 4 electrons in neutral Pb has been measured to be 96.71904(61) eV [33], only the corresponding binding-energy difference between the ground states of Pb^{41+} ([Kr]4 $d^{5} {}^{4}P_{5/2}$) and Pb^{4+} ([Xe]4 $f^{14}5d^{10} {}^{1}S_{0}$) needs to be determined theoretically. In the following, we use $E_{\rm Xe^{08-26}}$ and $E_{\rm Pb^{04-41}}$ to represent these two terms.

Within the MCDHF scheme, the many-electron atomic state function (ASF) is constructed as a linear combination of configuration state functions (CSFs) with common total angular momentum (*J*), magnetic (*M*), and parity (*P*) quantum numbers: $|\Gamma PJM\rangle = \sum_k c_k |\gamma_k PJM\rangle$. The CSFs $|\gamma_k PJM\rangle$ are given as *jj*-coupled Slater determinants of one-electron orbitals, and γ_k summarizes all the information needed to fully define the CSF, i.e. the orbital occupation and coupling of single-electron angular momenta. Γ collectively denotes all the γ_k included in the representation of the ASF. The set of CSF basis is generated by the GRASP2018 code [32] via single and double (SD) excitation of electrons from the reference configurations to high-lying virtual orbitals. After solving the self-consistent MCDHF equations for the radial wavefunctions and the mixing coefficients c_k , the relativistic configuration interaction (RCI) method is applied to account for the corrections arising from the quantum electrodynamic terms and Breit interactions. We systematically expand the size of the basis set by adding and optimizing virtual orbitals layer by layer up to n = 10 (*n* is the principle quantum number), with the final correlation energies being derived by extrapolating to $n = \infty$.

As the ground states of Xe^{26+} and Xe^{8+} are both in closedshell configurations, the CSF basis sets used for the calculations can be generated by allowing SD excitations from all the core electrons starting with the 1*s* orbitals. This gives a contribution from the SD electron correlation energy of 25.6 eV. The contributions from the Breit interactions and QED effects are 4.0 and 0.5 eV, respectively.

For the calculation of Pb^{41+} , however, due to its open $4d^5$ configuration, the number of CSFs for J = 5/2 easily grows above 4 million even for the SD exchanges of the $4s^24p^64d^5$ electrons and becomes not tractable. Therefore, to access $E_{Ph^{04-41}}$, one needs to construct an ion chain in the calculation to reduce the errors. We first calculate the binding-energy difference between Pb⁴⁺ and Pb²²⁺ via SD excitations from core electrons down to the 3p subshell, and then calculate the binding-energy difference between Pb^{22+} and Pb³⁶⁺ by allowing SD excitations of all the core electrons. Finally, the connection between Pb^{36+} and Pb^{41+} is bridged over Pb^{42+} via SD excitations from the 4s orbitals. In total, the SD electron correlation effects contribute 58.1 eV to $E_{Pb}04-41$. The Breit interactions and QED terms give rise to corrections of 9.3 and -0.6 eV, respectively, to the binding energy difference.

Until now, only the SD correlation energies are included in the calculations. Considering that the uncertainties in Breit and QED terms are small, the neglected higher-order correlations will account for the systematic errors. To estimate these errors, we make use of the accurate ionization data of the outermost 8 and 4 electrons in Xe and Pb, respectively. The estimations are based on three observations. First, as a selfconsistent theory, the MCDHF always approaches the real ground-state energy from above. Thus, the MCDHF binding energy of a given ionic ground state is always smaller than its real value. Second, for a given element, the contributions from higher-order correlation terms scale with the number of bound electrons. Therefore, the differences between the MCDHF binding energy and its real value is more likely to be smaller in highly charged ions. Lastly, within the same isoelectronic sequence, the contributions from higher-order correlations are always smaller for highly charged ions. This is because, perturbatively, in the denominator of each perturbation term, the energy differences between atomic states in highly charged ions are much larger than those in lower charged ions. As a result, for closed-shell ions, the calculated binding-energy differences based on the SD excitations are always smaller than their real values, but their deviations become narrower when the charge states become larger.

For the case of Xe, we find that the calculated bindingenergy difference between Xe⁸⁺ and Xe is 3.5 eV smaller than the experimentally measured value of 424.7(7) eV, with the single-electron ionization energies of Xe, Xe⁷⁺, and Xe^{8+} being 0.32 eV, 0.22 eV and 0.10 eV, respectively. smaller than their experimental measurements. Though the deviations of the single-electron ionization energies for some open-shell ions between Xe^{8+} and Xe^{26+} may be larger than the 0.22 eV deviation of that in Xe⁷⁺, one can still conservatively expect that the average deviation of the 16 electrons will not be larger than 0.22 eV. This indicates that the systematic shift of $E_{\chi_e^{08-26}}$ shall be within 4.0 eV. To cover the range between 0 to 4.0 eV, one can add a systematic correction of 2.0(2.0) eV, with both systematic shift and uncertainty being 2.0 eV. This leads to $E_{Xe^{08-26}} = 8546.5(2.0)$ eV and $E_{\rm Xe} = 8971.2(2.1) \, {\rm eV}.$

With a similar procedure, the ionization energy of Pb²⁺ and Pb³⁺ are found to be 1.26 and 0.95 eV smaller than their experimental measured values when SD excitation from the 3*p* subshells are considered. This indicates an average deviation of < 1.0 eV for the single-electron ionization energies for Pb⁴⁺ to Pb²²⁺, and a systematic correction of 9.0(9.0) eV to the binding energy of the corresponding 18 electrons. For the ions between Pb²²⁺ and Pb⁴¹⁺, since they are close to the isoelectronic systems of Xe ions, one can conservatively assume a < 0.22 eV average deviation of the corresponding single-electron ionization energies. After the summation, we obtain $E_{Pb^{04-41}} = 28633.9(9.1)$ eV and $E_{Pb} = 28730.6(9.1)$ eV.

3 Results

After calculating the free cyclotron frequencies during each PnP loop, the interpolation method [34] is applied to calculate the frequency ratios, see Fig. 3. This method uses two consecutive cyclotron frequencies of one trap in position 1 and interpolates them to the time the cyclotron frequency of the position 2 in the same trap was measured. Then the frequency ratio of the interpolated value of position 1 and the matching value of position 2 can be formed cancelling out in first order the magnetic field drift over time. The linear drift of the magnetic field is $\Delta B/B = -2.3 \times 10^{-10}$ /h. The impact of the non-linear drifts of the B-field was thoroughly investigated and found insignificant on the level of the achieved statistical uncertainty. The final measured ion frequency ratio is

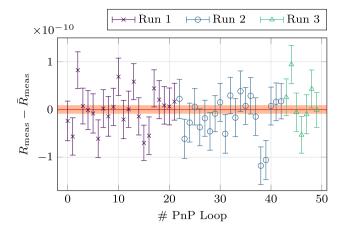


Fig. 3 The plot shows the frequency ratios for the different PnP loops. Each color represents a different measurement run. The 1σ error band of the averaged ratio \bar{R}_{meas} is visualized in red

Table 1 Systematic shifts and their uncertainties on the measured modified-cyclotron frequency ratio R_{meas} . For more details see text

Effect	Correction to R_{meas} (10 ⁻¹²)	Uncertainty (10^{-12})
Image charge shift	185	9
Relativistic shift	0	4
Total	185	10

$$R_{\text{meas}} - 1 = \frac{\nu (^{208} \text{Pb}^{41+})}{\nu (^{132} \text{Xe}^{26+})} - 1$$

= 1.252 194 24(9) × 10⁻⁴, (7)

with a relative statistical uncertainty of 9×10^{-12} .

The measured ratio is then corrected for known systematic shifts and their respective uncertainties. An overview of the relevant systematic effects and their size is listed in Table 1.

The largest systematic shift comes from the image charge shift (ICS) [35]. The highly charged ions induce an oscillating image charge in the trap electrodes. While this is necessary for detection, it causes a shift of the ions' frequencies by generating a counteracting electric field. The image charge shift depends strongly on the mass difference of the ions and on the radius of the trap, the latter being in the case of PENTATRAP 5.000(5) mm. The ICS was determined to be $R_{\text{meas}} - \tilde{R} = \Delta(R_{\text{meas}})_{\text{ICS}} = 1.85(9) \times 10^{-10}$, with \tilde{R} being the corrected ratio. In addition to this, the relativistic shift due to relativistic mass increase [36] leads to another systematic uncertainty related to the size of the excited radii during the PnP measurement scheme: $\Delta(R_{\text{meas}})_{\text{rel}} = 0(4) \times 10^{-12}$.

All other known systematic effects, due to e.g. trap potential anharmonicity, are on the order of 10^{-13} and below and are therefore neglected. Thus, the final ω_c -ratio is R - 1 =

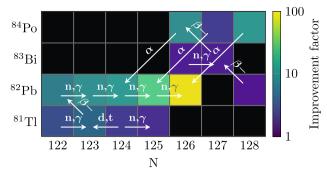


Fig. 4 This colormap depicts a cutout of the nuclide chart. The color corresponds to the improvement factor (with 1 being no improvement) in mass precision after including the new mass value of ²⁰⁸Pb in the AME [6]. The labeled arrows show the relevant connections for the mass determination of the different nuclides via different forms of decays from which the energy is known

 $1.252\,192\,39(9)(10)(13) \times 10^{-4}$, where the number in the first, second, and third brackets indicate the statistical, systematic, and total uncertainty, respectively.

Combining the binding energies of the missing electrons calculated by theory, the experimentally determined mass ratio, and the mass excess of the reference isotope of 132 Xe [13,14] as listed in the AME2020, the mass excess of 208 Pb is determined to be -21749.855(13) keV, which amounts to a neutral atomic mass of 207.976650571(14) u. The new value improves the mass uncertainty of neutral 208 Pb by a factor of 88 to a relative uncertainty of $\delta m/m = 7 \times 10^{-11}$ and shifts the mass excess value by -1.4(1.1) keV.

4 Discussion and conclusion

In addition to the improvement of precision of the mass of ²⁰⁸Pb itself, our measurement also improves the masses of a series of lead isotopes, connected by (n, γ) reactions. So far, their mass precision was limited by the precision of ²⁰⁸Pb, but is now limited by the precision of the energy of the (n, γ) reactions. Furthermore, Fig. 4 shows the improvement in precision of, in total, 14 neighbouring nuclides' masses connected to the mass of ²⁰⁸Pb via different decays, e.g. α decay, for which the energy is well known. Since the new value of the mass excess of ²⁰⁸Pb is shifted downward, all these connected nuclides will be shifted down by 1.4 keV. Table 2 lists the new mass values for all nuclides which were significantly improved. With the reported measurement we have established a new region in the nuclear chart with reference masses for experiments on heavy and superheavy nuclides. When measuring masses around m = 200 u the error due to the reference mass will be as low as a few 10^{-10} and therefore negligible for mass determinations on radionuclides.

Furthermore, with the new mass precision of 208 Pb of 7 × 10^{-11} the *g*-factor of the bound electron can be determined

el.

T1

T1

Pb

Tl

Pb

Tl

Ph

Pb

Pb

Bi

Pb

Bi

Po

Ро

Po

 $T_{1/2}$ [37]

3.783(12) y

 $1.4(6) \times 10^{17} \text{ y}$

 $1.70(9) \times 10^7 \text{ y}$

 $2.01(8) \times 10^{19}$ y

22.20(22) y

5.012(5) d

0.516(3) s

294.3(8) ns

138.376(2) d

4.202(11) min

Stable

Stable

Stable

Stable

Stable

А

203

204

204

205

205

206

206

207

208

209

210

210

210

211

212

Ζ

81

81

82

81

82

81

82

82

82

83

82

83

84

84

84

Table 2 New mass values of affected nuclides, wh

204,974,427.3

204,974,481.7

205,976,110.1

205,974,465,2

206,975,896.8

207,976,652.0

208,980,398.6

209,984,188.4

209,984,120,2

209,982,873.7

210,986,653.2

211,988,868.0

(1.3)

(1.2)

(1.4)

(1.2)

(1.2)

(1.2)

(1.5)

(1.6)

(1.5)

(1.2)

(1.3)

(1.2)

hen including the new mass value of ²⁰⁸ Pb in the AME2020 [6]						
AME2020 mass (μu)	Improved mass (μ u)		Improvement factor		
202,972,344.1	(1.3)	202,972,342.7	(0.4)	3.2		
203,973,863.4	(1.2)	203,973,862.01	(0.26)	4.8		
203,973,043.5	(1.2)	203,973,042.09	(0.18)	7.0		

(0.6)

(0.13)

(0.7)

(0.12)

(0.06)

(0.014)

(0.8)

(1.0)

(0.8)

(0.14)

(0.6)

(0.12)

2.4

9.2

2.1

10.6

21.6

88.0

1.8

1.6

1.8

8.8

2.4

10.1

204,974,425.9

204,974,480.26

205,976,108.7

205.974.463.79

206,975,895.39

207,976,650.571

208,980,397.2

209,984,118.9

209,982,872.27

210,986,651.7

211,988,866.55

209,984,187

to the same level of precision. It therefore allows to carry
out the experiments on ²⁰⁸ Pb ⁸¹⁺ at Alphatrap and Artemis
without having a large mass dependent error limiting their
g-factor determination.

Acknowledgements This work is supported by the Max-Planck-Gesellschaft and the International Max-Planck Research School for Precision Tests of Fundamental Symmetries, the German Research Foundation (DFG) Collaborative Research Centre "SFB 1225 (ISO-QUANT)". The project received funding from the European Research Council (ERC) under the European Union's Horizon 2020 research and innovation programme under Grant agreement number 832848 - FunI. Furthermore, we acknowledge funding and support by the Max Planck, RIKEN, PTB Center for Time, Constants and Fundamental Symmetries.

Funding Information Open Access funding enabled and organized by Projekt DEAL.

Data Availability Statement This manuscript has no associated data or the data will not be deposited. [Authors' comment: The datasets analysed in this study are available from the corresponding author.]

Open Access This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons licence, and indicate if changes were made. The images or other third party material in this article are included in the article's Creative Commons licence, unless indicated otherwise in a credit line to the material. If material is not included in the article's Creative Commons licence and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this licence, visit http://creativecomm ons.org/licenses/by/4.0/.

References

- 1. Y.T. Oganessian, V.K. Utyonkov, Y.V. Lobanov, F.S. Abdullin, A.N. Polyakov, R.N. Sagaidak, I.V. Shirokovsky, Y.S. Tsyganov, A.A. Voinov, G.G. Gulbekian, S.L. Bogomolov, B.N. Gikal, A.N. Mezentsev, S. Iliev, V.G. Subbotin, A.M. Sukhov, K. Subotic, V.I. Zagrebaev, G.K. Vostokin, M.G. Itkis, K.J. Moody, J.B. Patin, D.A. Shaughnessy, M.A. Stoyer, N.J. Stoyer, P.A. Wilk, J.M. Kenneally, J.H. Landrum, J.F. Wild, R.W. Lougheed, Phys. Rev. C 74(4), 044602 (2006). https://doi.org/10.1103/PhysRevC.74.044602
- 2. M. Block, D. Ackermann, K. Blaum, C. Droese, M. Dworschak, S. Eliseev, T. Fleckenstein, E. Haettner, F. Herfurth, F.P. Heßberger, S. Hofmann, J. Ketelaer, J. Ketter, H.J. Kluge, G. Marx, M. Mazzocco, Y.N. Novikov, W.R. Plaß, A. Popeko, S. Rahaman, D. Rodríguez, C. Scheidenberger, L. Schweikhard, P.G. Thirolf, G.K. Vorobyev, C. Weber, Nature 463(7282), 785 (2010). https://doi.org/10.1038/ nature08774
- 3. M. Dworschak, M. Block, D. Ackermann, G. Audi, K. Blaum, C. Droese, S. Eliseev, T. Fleckenstein, E. Haettner, F. Herfurth, F.P. Heßberger, S. Hofmann, J. Ketelaer, J. Ketter, H.J. Kluge, G. Marx, M. Mazzocco, Y.N. Novikov, W.R. Plaß, A. Popeko, S. Rahaman, D. Rodríguez, C. Scheidenberger, L. Schweikhard, P.G. Thirolf, G.K. Vorobyev, M. Wang, C. Weber, Phys. Rev. C 81(6), 064312 (2010). https://doi.org/10.1103/PhysRevC.81.064312
- 4. E.M. Ramirez, D. Ackermann, K. Blaum, M. Block, C. Droese, C.E. Düllmann, M. Dworschak, M. Eibach, S. Eliseev, E. Haettner, F. Herfurth, F.P. Heßberger, S. Hofmann, J. Ketelaer, G. Marx, M. Mazzocco, D. Nesterenko, Y.N. Novikov, W.R. Plaß, D. Rodríguez, C. Scheidenberger, L. Schweikhard, P.G. Thirolf, C. Weber, Science 337(6099), 1207 (2012). https://doi.org/10.1126/ science.1225636
- 5. M. Eibach, T. Beyer, K. Blaum, M. Block, C.E. Düllmann, K. Eberhardt, J. Grund, S. Nagy, H. Nitsche, W. Nörtershäuser, D. Renisch, K.P. Rykaczewski, F. Schneider, C. Smorra, J. Vieten, M. Wang, K. Wendt, Phys. Rev. C 89(6), 064318 (2014). https://doi. org/10.1103/PhysRevC.89.064318
- 6. M. Wang, W.J. Huang, F.G. Kondev, G. Audi, S. Naimi, Chin. Phys. C 45(3), 030003 (2021). https://doi.org/10.1088/1674-1137/ abddaf

- A. Kellerbauer, K. Blaum, G. Bollen, F. Herfurth, H.J. Kluge, M. Kuckein, E. Sauvan, C. Scheidenberger, L. Schweikhard, Eur. Phys. J. D 22(1), 53 (2003). https://doi.org/10.1140/epjd/e2002-00222-0
- J. Ketelaer, J. Krämer, D. Beck, K. Blaum, M. Block, K. Eberhardt, G. Eitel, R. Ferrer, C. Geppert, S. George, F. Herfurth, J. Ketter, S. Nagy, D. Neidherr, R. Neugart, W. Nörtershäuser, J. Repp, C. Smorra, N. Trautmann, C. Weber, Nuclear instruments and methods in physics research section a: accelerators. Spectromet. Detect. Assoc. Equip. **594**(2), 162 (2008). https://doi.org/10.1016/j.nima. 2008.06.023
- J. Grund, M. Asai, K. Blaum, M. Block, S. Chenmarev, C.E. Düllmann, K. Eberhardt, S. Lohse, Y. Nagame, S. Nagy, P. Naubereit, J.J.W. van de Laar, F. Schneider, T.K. Sato, N. Sato, D. Simonovski, K. Tsukada, K. Wendt, Nuclear instruments and methods in physics research section a: accelerators. Spectromet. Detect. Assoc. Equip. 972, 164013 (2020). https://doi.org/10.1016/j.nima.2020.164013
- S. Sturm, F. Köhler, J. Zatorski, A. Wagner, Z. Harman, G. Werth, W. Quint, C.H. Keitel, K. Blaum, Nature **506**(7489), 467 (2014). https://doi.org/10.1038/nature13026
- M. Vogel, M.S. Ebrahimi, Z. Guo, A. Khodaparast, G. Birkl, W. Quint, Annalen der Physik **531**(5), 1800211 (2019). https://doi. org/10.1002/andp.201800211
- F. Köhler, K. Blaum, M. Block, S. Chenmarev, S. Eliseev, D.A. Glazov, M. Goncharov, J. Hou, A. Kracke, D.A. Nesterenko, Y.N. Novikov, W. Quint, E. Minaya Ramirez, V.M. Shabaev, S. Sturm, A.V. Volotka, G. Werth, Nat. Commun. 7(1), 10246 (2016). https:// doi.org/10.1038/ncomms10246
- M. Redshaw, B.J. Mount, E.G. Myers, Phys. Rev. A 79(1), 012506 (2009)
- A. Rischka, H. Cakir, M. Door, P. Filianin, Z. Harman, W.J. Huang, P. Indelicato, C.H. Keitel, C.M. König, K. Kromer, M. Müller, Y.N. Novikov, R.X. Schüssler, C. Schweiger, S. Eliseev, K. Blaum, Phys. Rev. Lett. **124**(11), 113001 (2020). https://doi.org/10.1103/ PhysRevLett.124.113001
- J. Repp, C. Böhm, J.R. Crespo López-Urrutia, A. Dörr, S. Eliseev, S. George, M. Goncharov, Y.N. Novikov, C. Roux, S. Sturm, S. Ulmer, K. Blaum, Appl. Phys. B 107(4), 983 (2012). https:// doi.org/10.1007/s00340-011-4823-6
- C. Roux, C. Böhm, A. Dörr, S. Eliseev, S. George, M. Goncharov, Y.N. Novikov, J. Repp, S. Sturm, S. Ulmer, K. Blaum, Appl. Phys. B 107(4), 997 (2012). https://doi.org/10.1007/s00340-011-4825-4
- J. Zatorski, B. Sikora, S.G. Karshenboim, S. Sturm, F. Köhler-Langes, K. Blaum, C.H. Keitel, Z. Harman, Phys. Rev. A 96(1), 012502 (2017). https://doi.org/10.1103/PhysRevA.96.012502
- L.S. Brown, G. Gabrielse, Phys. Rev. A 25(4), 2423 (1982). https:// doi.org/10.1103/PhysRevA.25.2423

- V.P. Ovsyannikov, G. Zschornack, Rev. Sci. Instrum. 70(6), 2646 (1999). https://doi.org/10.1063/1.1149822
- 20. Startseite » DREEBIT GmbH DREEBIT Company | DE. https:// www.dreebit.com/de/
- P. Micke, S. Kühn, L. Buchauer, J.R. Harries, T.M. Bücking, K. Blaum, A. Cieluch, A. Egl, D. Hollain, S. Kraemer, T. Pfeifer, P.O. Schmidt, R.X. Schüssler, C. Schweiger, T. Stöhlker, S. Sturm, R.N. Wolf, S. Bernitt, J.R. Crespo López-Urrutia, Rev. Sci. Instrum. 89(6), 063109 (2018). https://doi.org/10.1063/1.5026961
- C. Schweiger, C.M. König, J.R. Crespo López-Urrutia, M. Door, H. Dorrer, C.E. Düllmann, S. Eliseev, P. Filianin, W. Huang, K. Kromer, P. Micke, M. Müller, D. Renisch, A. Rischka, R.X. Schüssler, K. Blaum, Rev. Sci. Instrum. **90**(12), 123201 (2019). https://doi.org/10.1063/1.5128331
- 23. C. Schweiger, et al., in preparation (2021)
- N.E. Bradbury, R.A. Nielsen, Phys. Rev. 49(5), 388 (1936). https:// doi.org/10.1103/PhysRev.49.388
- A.G. Marshall, C.L. Hendrickson, G.S. Jackson, Mass Spectrom. Rev. 17(1), 1 (1998). https://doi.org/10.1002/ (SICI)1098-2787(1998)17:1<1::AID-MAS1>3.0.CO;2-K
- E.A. Cornell, R.M. Weisskoff, K.R. Boyce, R.W. Flanagan, G.P. Lafyatis, D.E. Pritchard, Phys. Rev. Lett. 63(16), 1674 (1989). https://doi.org/10.1103/PhysRevLett.63.1674
- E.A. Cornell, R.M. Weisskoff, K.R. Boyce, D.E. Pritchard, Phys. Rev. A 41(1), 312 (1990). https://doi.org/10.1103/PhysRevA.41. 312
- X. Feng, M. Charlton, M. Holzscheiter, R.A. Lewis, Y. Yamazaki, J. Appl. Phys. 79(1), 8 (1996). https://doi.org/10.1063/1.360947
- F. Harris, Proc. IEEE 66(1), 51 (1978). https://doi.org/10.1109/ PROC.1978.10837
- 30. I.P. Grant, Adv. Phys. 19, 747 (1970)
- 31. J.P. Desclaux, D.F. Mayers, F. O'Brien, J. Phys. B 4, 631 (1971)
- C.F. Fischer, G. Gaigalas, P. Jönsson, J. Bieroń, Comput. Phys. Commun. 237, 184 (2019)
- 33. A. Kramida, Yu. Ralchenko, J. Reader, and NIST ASD Team. NIST Atomic Spectra Database (ver. 5.8), [Online]. Available: https:// physics.nist.gov/asd [2021, August 11]. National Institute of Standards and Technology, Gaithersburg, MD (2020)
- 34. M. Door, et al., in preparation (2021)
- M. Schuh, F. Heiße, T. Eronen, J. Ketter, F. Köhler-Langes, S. Rau, T. Segal, W. Quint, S. Sturm, K. Blaum, Phys. Rev. A 100(2), 023411 (2019). https://doi.org/10.1103/PhysRevA.100.023411
- L.S. Brown, G. Gabrielse, Rev. Mod. Phys. 58(1), 233 (1986). https://doi.org/10.1103/RevModPhys.58.233
- 37. IAEA Nuclear Data Services. https://www-nds.iaea.org/