

Mass-Difference Measurements on Heavy Nuclides with an eV/c² Accuracy in the PENTATRAP Spectrometer

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First ever measurements of the ratios of free cyclotron frequencies of heavy, highly charged ions with $Z > 50$ with relative uncertainties close to 10^{-11} are presented. Such accurate measurements have become realistic due to the construction of the novel cryogenic multi-Penning-trap mass spectrometer PENTATRAP. Based on the measured frequency ratios, the mass differences of five pairs of stable xenon isotopes, ranging from ^{126}Xe to ^{134}Xe , have been determined. Moreover, the first direct measurement of an electron binding energy in a heavy highly charged ion, namely of the 37th atomic electron in xenon, with an uncertainty of a few eV is demonstrated. The obtained value agrees with the calculated one using two independent, different implementations of the multiconfiguration Dirac-Hartree-Fock method. PENTATRAP opens the door to future measurements of electron binding energies in highly charged heavy ions for more stringent tests of bound-state quantum electrodynamics in strong electromagnetic fields and for an investigation of the manifestation of light dark matter in isotopic chains of certain chemical elements.

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Many areas of fundamental physics require the knowledge of mass differences or mass ratios of a variety of nuclides with very low uncertainty [1,2]. Notable examples are, e.g., neutrino physics [3], a test of special relativity [4] and bound-state quantum electrodynamics (QED) [5], ion clocks [6], and the search for dark matter via high-resolution isotope shift measurements [7–9]. In order to satisfy these requirements, the novel experiment PENTATRAP [10,11] has been set up at the Max Planck Institute for Nuclear Physics in Heidelberg. This is the first experiment that finally offers a realistic opportunity to pursue a mass-ratio measurement on heavy ($A > 100$) highly charged ions with a fractional uncertainty below 10^{-11} .

PENTATRAP is based on high-precision Penning-trap mass spectrometry—nowadays the only technique which enables mass-ratio measurements with such low uncertainties [2]. This arises from the fact that, in the Penning trap,

one converts the determination of the mass m of an ion with charge q into a determination of the free cyclotron frequency $\nu_c = (1/2\pi)(q/m)B$ of the ion stored in a combination of a strong uniform magnetic field B and a weak harmonic electrostatic potential well. In practice, one measures the frequencies ν_- , ν_z , ν_+ of three independent trap eigenmotions (magnetron, axial, and cyclotron motions, respectively) which an ion undergoes in the Penning trap and applies the invariance theorem $\nu_c^2 = \nu_-^2 + \nu_z^2 + \nu_+^2$ [12] to determine the free cyclotron frequency.

In this Letter, we present the first mass measurements carried out with PENTATRAP on stable isotopes of xenon. The mass differences of five pairs, namely ^{134}Xe - ^{132}Xe , ^{132}Xe - ^{131}Xe , ^{131}Xe - ^{129}Xe , ^{129}Xe - ^{128}Xe , and ^{128}Xe - ^{126}Xe , have been determined with relative uncertainties of a few 10^{-11} by measuring the ratios of free cyclotron frequencies of the corresponding xenon isotopes in a charge state of $17+$. The stable xenon isotopes were chosen for a first operation of PENTATRAP for three reasons. First, some of the stable xenon isotopes belong to a group of just a few nuclides for which mass ratios have been determined with sub 10^{-10} relative uncertainty [13]. In fact, the only other setup which can reach relative uncertainties of a few 10^{-11} is the Florida State University (FSU) trap [14], but

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only on low charged ion species. e.g., the mass differences of ^{134}Xe - ^{132}Xe , ^{132}Xe - ^{131}Xe , and ^{131}Xe - ^{129}Xe have been determined with the FSU trap [14] and, thus, can be considered a suitable candidate for reference measurements for PENTATRAP. Second, xenon has many (virtually) stable isotopes and, thus, in this respect, is similar to isotopic chains proposed in [7–9] for the search for dark matter. Third, the addressed xenon isotopes span a mass range of eight atomic mass units forming a reference mass comb for on-line Penning-trap experiments in this mass region and, thus, serve as a backbone of precision mass measurements for the atomic mass evaluation (AME) [13].

To demonstrate the capability of PENTATRAP to perform measurements of mass ratios with a fractional uncertainty of close to 10^{-11} , we also determined the binding energy of the 37th electron in Xe by measuring the ratio of the free cyclotron frequencies of $^{131}\text{Xe}^{17+}$ and $^{131}\text{Xe}^{18+}$ ions. The obtained value is compared with the even more precisely known theoretical prediction published in this Letter. This “proof-of-principle” measurement opens the door to future measurements of electron binding energies in very highly charged heavy ions (e.g., in hydrogenlike xenon ions) with an uncertainty of an eV required to perform stringent tests of bound-state QED in strong electromagnetic fields [15]. Moreover, measuring the binding energy of many—or even all—shell electrons

benchmarks atomic structure theory in a challenging and previously inaccessible way.

The design of the PENTATRAP experiment rests upon several basic requirements that are prerequisite to high-precision mass measurements: (1) the use of highly charged ions to increase the sensitivity of the experiment since the cyclotron frequency ν_c scales with the charge state q , (2) the long storage of a single ion in an ultrahigh vacuum in a very small volume by cooling the ion’s surrounding environment to the temperature of liquid helium, and (3) the application of the fast phase-sensitive frequency-measurement techniques pulse and phase [16] and pulse and amplify [17].

Highly charged ions of xenon are produced with a room-temperature Dresden-EBIT ion source [18]. With a typical kinetic energy of 7 keV/ q , they are ejected as 1- μs long bunches from the electron beam ion trap (EBIT) and sent through a 90-degree sector dipole magnet, see Fig. 1. The sector magnet serves as a q/m separator with a resolving power of about 400 allowing us to have a highly purified beam for many cases of our interest [see Fig. 1(a)]. Prior to the injection of the chosen ions into the 7-T magnetic field of the mass spectrometer, the kinetic energy of the ions is lowered in a room-temperature pulsed drift tube from a few keV/ q to about 200 eV/ q . The final deceleration of the ions to nearly zero energy required for their capture in the

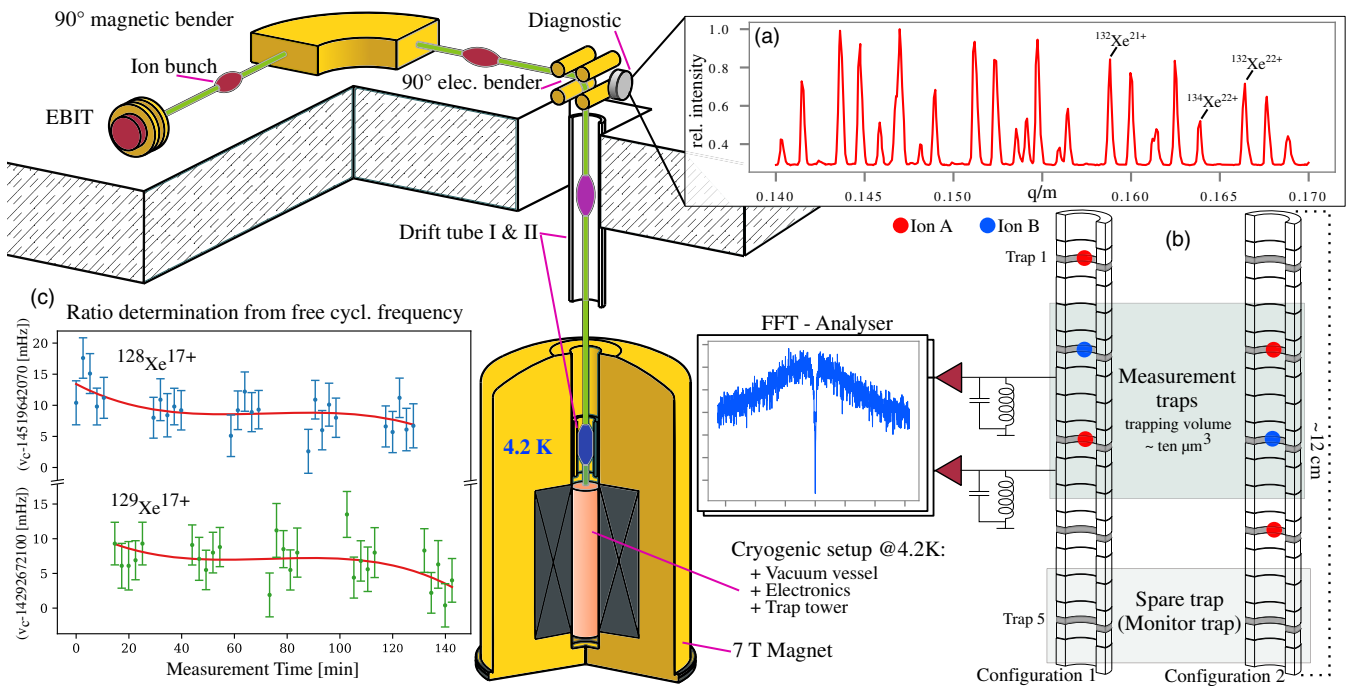


FIG. 1. Schematic overview of the PENTATRAP setup and the measurement procedure. The setup contains an external ion source (EBIT) and a beam line in which the bunched ion beam is guided to the trap tower in the 7 T magnet. (a) On the upper right, a mass spectrum is shown which was recorded by scanning the magnetic bender. (b) The two trap towers represent the two different configurations in which the ions are stored during the measurement cycle. (c) On the lower left, a typical two-hour measurement of the free cyclotron frequencies of $^{129}\text{Xe}^{17+}$ (green) and $^{128}\text{Xe}^{17+}$ (blue) ions is shown. The red line is a third-order simultaneous polynomial fit to the free cyclotron frequencies. For details see text.

traps takes place in a second cryogenic pulsed drift tube, which is situated right above trap 1. The ions are captured in trap 1 by a reflecting potential well produced by the lowermost trap electrode and the restored high potential of the cryogenic pulsed drift tube. Adiabatic transportation of the ions to the other traps is realized by adiabatic changes of the potentials of the trap electrodes. The vacuum chamber that houses the traps and the associated cryogenic frequency-measurement electronics is cooled down to the temperature of liquid helium, 4.2 K. The mass spectrometer is located in a room with temperature stabilization to a few 10 mK/day. Together with a stabilization of the helium pressure and level in the magnet bore around the traps and active shielding of the trap region from stray magnetic fields, this results in a very stable magnetic field with a relative time variation better than $\leq 1 \times 10^{-10}/\text{h}$.

Prior to the measurement of the ion trap frequencies in one of the measurement traps, the amplitudes of the ion motions are reduced to a few μm via the resistive cooling technique [12]. The axial motion is cooled via its direct coupling to an LC circuit which has a quality factor of 4000 (trap 2) or 9000 (trap 3) and a temperature below 10 K. The magnetron and cyclotron motions are cooled via their side-band coupling to the axial motion. The resistive cooling technique is also used to measure the frequencies of the trapped ion. In this case, it is named “single-dip” method if the axial frequency is measured, and “double-dip” method in the case of the measurement of the magnetron or cyclotron frequencies [19]. The technique of choice for the measurement of the cyclotron frequency is the pulse and phase technique due to its higher precision compared to the double-dip method [16]. It can be split up into two steps: (1) a measurement of its frequencies with moderate accuracy using the dip techniques in order to identify the ion species, and (2) a main measurement of the ion frequencies with high accuracy.

During the main measurement procedure, the cyclotron and the axial frequencies are measured simultaneously. This measurement scheme, employed for the first time in our Letter, substantially reduces the uncertainty in the determination of the free cyclotron frequency. After a single ion is prepared in the measurement trap, its cyclotron motion is excited via a 5 ms-long dipole radio frequency (rf) pulse to a radius of approximately 10 μm in order to fix the initial cyclotron-motion phase. During the phase-evolution time, the cyclotron motion freely evolves and accumulates a certain phase. This waiting time is used to measure the axial frequency. In order to measure the accumulated phase afterwards, the energy of the cyclotron motion is transferred to the axial motion via a 30 ms-long quadrupole π pulse. This imprints the accumulated phase of the cyclotron motion to the phase of the axial motion, which is in turn measured with the Fourier-transform ion cyclotron resonance technique. Such a measurement is performed for two phase-evolution times, usually $t_1 = 0.1$ s and $t_2 = 40$ s

resulting in a measurement of two accumulated phases ϕ_1 and ϕ_2 . This yields the cyclotron frequency $\nu_+ = [\phi_2 - \phi_1 + 2\pi n]/[2\pi(t_2 - t_1)]$, where n is the number of full cyclotron revolutions the ion performs in the time interval $t_2 - t_1$. Since the magnetron frequency is small compared to the other ion frequencies, it is sufficient to measure it once a day during the preparatory step.

A unique feature of the PENTATRAP mass spectrometer is its multitrap configuration which consists of five identical cylindrical traps [11] [see Fig. 1(b)]. At the moment, two traps, trap 2, and trap 3, are used for measurements of the ion free cyclotron frequency, whereas the remaining three traps serve as ion storage containers. A measurement of the ratio of free cyclotron frequencies of two ions is performed with the following procedure. First, three ions of two species are loaded into trap 1, trap 2, and trap 3 [measurement configuration 1 in Fig. 1(b)]. The ions in trap 1 and trap 3 are identical. The free cyclotron frequencies of the ions in trap 2 and trap 3 are measured for approximately 15 min. Afterwards, the ion species in the measurement traps are swapped by adiabatically transporting them into the neighboring traps [measurement configuration 2 in Fig. 1(b)] with subsequent measurements of their free cyclotron frequencies. This measurement procedure is then repeated. Such a multitrap configuration significantly reduces the uncertainty in the mass-ratio determination.

The data analysis scheme of choice is called the “polynomial” method, which has already been successfully employed in several experiments [20–22]. It is based on the reasonable assumption that the magnetic field drift and, hence, the free cyclotron frequency changes in time can be approximated by a polynomial of low order. Thus, the free cyclotron frequency drift of ion 1 and ion 2 is approximated by two polynomials that differ only by a coefficient of proportionality $R = [\nu_c(\text{ion1})]/[\nu_c(\text{ion2})]$. In the reported measurements on xenon isotopes, we divided the measurement period into approximately two-hour intervals and obtained R for each interval from the simultaneous fit of two polynomials to the corresponding free-cyclotron-frequency values [see Fig. 1(c)]. The interval duration is chosen so that, on one hand, the number of the frequency measurement points exceeds the number of free parameters of the polynomial, on the other hand, the interval must be as short as possible to be able to approximate the free-cyclotron-frequency drift with a polynomial of a low order. Already, a two-hour measurement allows one to reach an uncertainty of 4×10^{-11} in the determination of the free-cyclotron-frequency ratio of xenon ions in the 17+ charge state.

The mass differences of the five pairs of stable xenon isotopes were determined by measuring the free-cyclotron-frequency ratios of the corresponding xenon ions in a charge state of 17+. The results of the measurement are summarized in Table I. Moreover, we also determined the binding energy of the 37th electron in xenon by measuring the ratio of the free cyclotron frequencies of

TABLE I. Results of the measurements on the five stable xenon isotopes obtained in this Letter. The first and second columns list the addressed xenon ion pairs and their free cyclotron frequency ratios, respectively. The mass differences of the neutral states of these pairs determined on the basis of the measured free cyclotron frequency ratios are presented in the third column. The frequency ratios and the mass differences are given with their statistical and systematic uncertainties placed in the first and second round brackets, respectively. The mass differences evaluated in the AME2016 [13] are presented in the fourth column with their total uncertainties. The last column lists the so-called “improvement of accuracy” factor that demonstrates the increase of precision in the mass-difference determination achieved in this Letter compared to the AME2016 uncertainties.

Ion pair	Frequency ratio	Mass difference / u (this Letter)	Mass difference / u (AME2016 [13])	Improvement of accuracy
$^{134}\text{Xe}^{17+}/^{132}\text{Xe}^{17+}$	1.015 172 982 205(19)(8)	2.001 237 945 4(25)(11)	2.001 237 947(12)	4
$^{132}\text{Xe}^{17+}/^{131}\text{Xe}^{17+}$	1.007 632 569 193(13)(6)	0.999 070 956 6(17)(8)	0.999 070 951(11)	6
$^{131}\text{Xe}^{17+}/^{129}\text{Xe}^{17+}$	1.015 518 803 388(9)(8)	2.000 303 273 5(12)(10)	2.000 303 277(11)	7
$^{129}\text{Xe}^{17+}/^{128}\text{Xe}^{17+}$	1.007 828 736 895(10)(6)	1.001 250 105 6(13)(8)	1.001 249 9(11)	740
$^{128}\text{Xe}^{17+}/^{126}\text{Xe}^{17+}$	1.015 880 167 834(18)(8)	1.999 233 328 2(23)(10)	1.999 234(4)	1700

$^{131}\text{Xe}^{18+}$ and $^{131}\text{Xe}^{17+}$ ions: $\nu_c(^{131}\text{Xe}^{18+})/\nu_c(^{131}\text{Xe}^{17+}) = 1.058\,827\,929\,585(17)(30)$. Current experimental literature values for electron binding energies in highly charged ions of Xe-isotopes are either not accurate enough or do not exist. Thus, we compare our experimental result with two theory values obtained from two independent groups, to be detailed in what follows.

The ground state of the $^{131}\text{Xe}^{18+}$ ion is a simple Kr-like configuration $[\text{Ar}]3d^{10}4s^24p^6$. The additional electron in Rb-like $^{131}\text{Xe}^{17+}$ is in the $4d_{3/2}$ state, i.e., the valence shell is filled according to Coulomb ordering, instead of the usual Madelung ordering.

Because of the high nuclear charge, relativistic corrections such as the Breit correction to the electron-electron interaction are rather significant on the individual energy levels. We apply the multiconfiguration Dirac-Hartree-Fock (MCDHF) method [23,24] to account for such relativistic correlation effects. Within this scheme, the many-electron atomic state function is given as a linear superposition of configuration state functions (CSFs) sharing 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 constructed as jj -coupled N -particle Slater determinants of one-electron wave functions, and γ_k is a multi-index that includes all the information needed to fully describe the CSF, i.e., orbital occupation and coupling of one-electron angular momenta. Γ collectively denotes all the γ_k included in the representation of the Kr- or Rb-like ground state. Using two different implementations of the MCDHF method [25,26], we systematically expand the active space of virtual orbitals to monitor the convergence of the calculations and to assess their uncertainties.

The binding energy difference for these two Xe ions is dominated by the Dirac-Hartree-Fock term. Electron correlation effects contribute tens of eVs for both ions, however, since the $4d_{3/2}$ outermost valence electron polarizes the $3d$, $4s$, and $4p$ subshells, their contribution is more complicated to account for in the case of the Rb-like ion.

Nevertheless, correlation terms largely cancel in the energy difference.

In highly charged ions, typically QED corrections such as the self-energy are of relevance. The self-energy corrections have been calculated in two different ways: the newly proposed effective operator method from Ref. [27] has been compared to computing the Lamb shift of the $4d_{3/2}$ electron (see [28]) employing an effective radial potential which accounts for the screening by the core electrons. However, the QED corrections amounting to approx. 21 meV are not observable at the current level of precision; the same thing can be said about the mass shift contributions. One of the calculations employing the code in Ref. [25] uses full relaxation of all spectroscopic orbitals, and single and double excitations from all these orbitals to the free single-electron states up to $5d$, with a result of 432.4(3.0) eV. In the other computation, employing [26], we generate the set of CSFs with excitations from the $3s-4d$ states up to $10h$, with the virtual orbitals optimized layer by layer, arriving to the value 435.1(1.0) eV.

The measured value of 432.4(1.3)(3.4) agrees within one-sigma uncertainty with the theoretical values. This measurement and theory comparison can be considered a proof-of-principle experiment for higher charge states where, e.g., stringent QED tests could be performed by measuring the binding energy of the remaining electrons in few-electron ions.

In order to determine the mass difference of neutral xenon isotope pairs from the measurement of the ratio of the free cyclotron frequencies of corresponding xenon isotope ions, we calculated the total binding energy of the 17 missing electrons to be 2982(5) eV. The obtained values of the mass differences for all five xenon pairs agree within 1 sigma with the values evaluated in the atomic mass evaluation (AME2016) [13]. The uncertainty of the mass differences of three pairs, ^{134}Xe - ^{132}Xe , ^{132}Xe - ^{131}Xe , ^{131}Xe - ^{129}Xe [14], were decreased by at least a factor of 4 (see Fig. 2 and Table I). Whereas the mass differences of

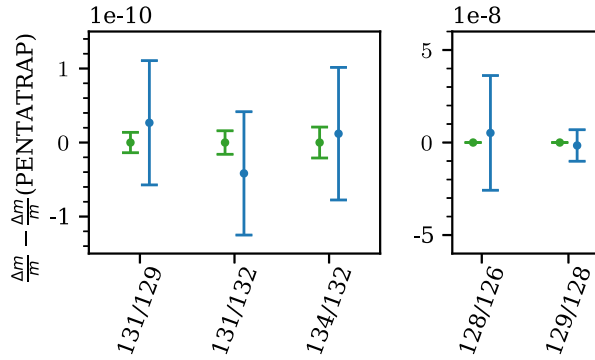


FIG. 2. Comparison of the relative mass differences $\Delta m/m$ of five stable xenon pairs determined in this Letter with the evaluated AME2016 values [13]. The green and blue error bars are the PENTATRAP and AME2016 uncertainties, respectively. The zero point of the Y axis corresponds to the mass difference of the corresponding xenon pair determined in this Letter.

two pairs, ^{129}Xe - ^{128}Xe and ^{128}Xe - ^{126}Xe , were determined by at least 2 orders of magnitude more precisely.

In the case of the mass differences, the statistical and systematic uncertainties are similar in size, whereas the total uncertainty of the determination of the binding energy of the 37th electron in xenon is dominated by the systematic uncertainty. This is due to the fact that the difference of the m/q ratio in the case of ^{131}Xe ions in the 17+ and 18+ charge states is by a factor of at least 3.5 larger than that of the different xenon isotopes in the same 17+ charge state. The total systematic uncertainty in the frequency ratio determination of $^{131}\text{Xe}^{18+}/^{131}\text{Xe}^{17+}$ is equal to 3×10^{-11} and is fully dominated by the impact of the linear gradient of the magnetic field along the trap axis on the values of the trap frequencies. The total systematic uncertainty in the frequency ratio determination of the xenon isotopes in the 17+ charge state does not exceed 10^{-11} and is a result of (1) the nonharmonicity of the trap electrostatic potential and of (2) the nonuniformity of the magnetic field.

To conclude, we presented first ever measurements of the ratios of free cyclotron frequencies of heavy highly charged ions with relative uncertainties close to 1×10^{-11} . Based on the measured frequency ratios, we determined the mass differences of five pairs of stable xenon isotopes, ^{134}Xe - ^{132}Xe , ^{132}Xe - ^{131}Xe , ^{131}Xe - ^{129}Xe , ^{129}Xe - ^{128}Xe , and ^{128}Xe - ^{126}Xe , with high precision, thus, having demonstrated the ability of PENTATRAP to perform high-precision measurements on an isotope chain of the same element. Such measurements on isotope chains such as Ca, Sr, and Yb are of paramount importance to laser-spectroscopic experiments for dark matter searches [7–9] and, thus, belong to planned measurements of the highest priority for PENTATRAP.

Furthermore, the binding energy of the 37th atomic electron in xenon of 432.4(1.3)(3.4) eV was determined.

This proof-of-principle measurement finally renders a realistic way to stringently test QED in strong electromagnetic fields by comparing determinations of electron binding energies with ≤ 1 eV uncertainty in very highly charged heavy ions. With the current uncertainty of PENTATRAP, the measurement of the binding energy of the 11th atomic electron would be sufficient for a QED test.

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- [1] K. Blaum, *Phys. Rep.* **425**, 1 (2006).
- [2] E. Myers, *Atoms* **7**, 37 (2019).
- [3] S. Eliseev, Y.N. Novikov, and K. Blaum, *Ann. Phys. (Amsterdam)* **525**, 707 (2013).
- [4] S. Rainville, J. K. Thompson, E. G. Myers, J. M. Brown, M. S. Dewey, E. G. Kessler Jr, R. D. Deslattes, H. G. Börner, M. Jentschel, P. Mutti, and D. E. Pritchard, *Nature (London)* **438**, 1096 (2005).
- [5] F. Köhler, K. Blaum, M. Block, S. Chenmarev, S. Eliseev, D. A. Glazov, M. Goncharov, J. Hou, A. Kracke, D. A. Nesterenko *et al.*, *Nat. Commun.* **7**, 10246 (2016).
- [6] M. G. Kozlov, M. S. Safronova, J. R. C. López-Urrutia, and P. O. Schmidt, *Rev. Mod. Phys.* **90**, 045005 (2018).
- [7] V. V. Flambaum, A. J. Geddes, and A. V. Viatkina, *Phys. Rev. A* **97**, 032510 (2018).
- [8] D. Antypas, O. Tretiak, A. Garcon, R. Ozeri, G. Perez, and D. Budker, *Phys. Rev. Lett.* **123**, 141102 (2019).
- [9] T. Manovitz, R. Shaniv, Y. Shapira, R. Ozeri, and N. Akerman, *Phys. Rev. Lett.* **123**, 203001 (2019).
- [10] J. Repp, C. Böhm, J. R. C. López-Urrutia, A. Dörr, S. Eliseev, S. George, M. Goncharov, Y. N. Novikov, C. Roux, S. Sturm *et al.*, *Appl. Phys. B* **107**, 983 (2012).

- [11] C. Roux, C. Böhm, A. Dörr, S. Eliseev, S. George, M. Goncharov, Y.N. Novikov, J. Repp, S. Sturm, S. Ulmer *et al.*, *Appl. Phys. B* **107**, 997 (2012).
- [12] L. S. Brown and G. Gabrielse, *Rev. Mod. Phys.* **58**, 233 (1986).
- [13] M. Wang, G. Audi, F. G. Kondev, W. J. Huang, S. Naimi, and X. Xu, *Chin. Phys. C* **41**, 030003 (2017).
- [14] M. Höcker, R. Rana, and E. G. Myers, *Phys. Rev. A* **88**, 052502 (2013).
- [15] H. Häffner, N. Hermanspahn, P. Indelicato, H. Kluge, E. Lindroth, V. Natarajan, W. Quint, S. Stahl, J. Verdú, and G. Werth, *Hyperfine Interact.* **127**, 271 (2000).
- [16] E. A. Cornell, R. M. Weisskoff, K. R. Boyce, and D. E. Pritchard, *Phys. Rev. A* **41**, 312 (1990).
- [17] S. Sturm, A. Wagner, B. Schabinger, and K. Blaum, *Phys. Rev. Lett.* **107**, 143003 (2011).
- [18] G. Zschornack, F. Grossmann, U. Kentsch, V. Ovsyannikov, E. Ritter, M. Schmidt, A. Thorn, and F. Ullmann, *Rev. Sci. Instrum.* **81**, 02A512 (2010).
- [19] X. Feng, M. Charlton, M. Holzscheiter, R. Lewis, and Y. Yamazaki, *J. Appl. Phys.* **79**, 8 (1996).
- [20] W. Shi, M. Redshaw, and E. G. Myers, *Phys. Rev. A* **72**, 022510 (2005).
- [21] S. Eliseev, K. Blaum, M. Block, S. Chenmarev, H. Dörrer, C. E. Düllmann, C. Enss, P. E. Filianin, L. Gastaldo, M. Goncharov, U. Köster, F. Lautenschläger, Yu. N. Novikov, A. Rischka, R. X. Schüssler, L. Schweikhard, and A. Türlér, *Phys. Rev. Lett.* **115**, 062501 (2015).
- [22] J. Karthein, D. Atanasov, K. Blaum, M. Breitenfeldt, V. Bondar, S. George, L. Hayen, D. Lunney, V. Manea, M. Mougéot, D. Neidherr, L. Schweikhard, N. Severijns, A. Welker, F. Wienholtz, R. N. Wolf, and K. Zuber, *Phys. Rev. C* **100**, 015502 (2019).
- [23] I. P. Grant, *Adv. Phys.* **19**, 747 (1970).
- [24] J. P. Desclaux, D. F. Mayers, and F. O'Brien, *J. Phys. B* **4**, 631 (1971).
- [25] P. Indelicato and J.-P. Desclaux, MCDPFGME: A Multiconfiguration Dirac-Fock and General Matrix Elements Program (2005), <http://dirac.spectro.jussieu.fr/mcdf>.
- [26] C. F. Fischer, G. Gaigalas, P. Jönsson, and J. Bieroń, *Comput. Phys. Commun.* **237**, 184 (2019).
- [27] V. Shabaev, I. Tupitsyn, and V. Yerokhin, *Comput. Phys. Commun.* **189**, 175 (2015).
- [28] V. A. Yerokhin and V. M. Shabaev, *Phys. Rev. A* **60**, 800 (1999).