PHYSICAL REVIEW A **86**, 050502(R) (2012)

Atomic masses of strontium and ytterbium

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(Received 30 September 2012; published 26 November 2012)

The atomic masses of the three most abundant isotopes of strontium, ^{86,87,88}Sr, and of the six most abundant isotopes of ytterbium, ^{170,171,172,173,174,176}Yb, have been obtained from measurements of cyclotron frequency ratios of pairs of ions simultaneously trapped in a Penning trap. Our results, with one standard deviation uncertainty, are $M(^{86}\mathrm{Sr}) = 85.909\,260\,730\,9\ (91)$ u, $M(^{87}\mathrm{Sr}) = 86.908\,877\,497\,0\ (91)$ u, $M(^{88}\mathrm{Sr}) = 87.905\,612\,257\,1\ (97)$ u, and $M(^{170}\mathrm{Yb}) = 169.934\,767\,241\ (18)$ u, $M(^{171}\mathrm{Yb}) = 170.936\,331\,514\ (19)$ u, $M(^{172}\mathrm{Yb}) = 171.936\,386\,655\ (18)$ u, $M(^{173}\mathrm{Yb}) = 172.938\,216\,213\ (18)$ u, $M(^{174}\mathrm{Yb}) = 173.938\,867\,539\ (18)$ u, $M(^{176}\mathrm{Yb}) = 175.942\,574\,702\ (22)$ u. These results have application to photon-recoil determinations of the fine-structure constant.

DOI: 10.1103/PhysRevA.86.050502 PACS number(s): 32.10.Bi, 06.20.Jr, 21.10.Dr, 07.75.+h

The current most precise value for the fine-structure constant $\alpha \equiv e^2/(4\pi\varepsilon_0\hbar c)$, with relative precision of 0.25 parts per 10^9 (ppb), is obtained by combining theory [1] and experiment [2] for the anomalous magnetic moment of the electron. This provides strong motivation for measurements of α using alternate techniques: By inserting an independent value for α into the anomaly theory (an extensive calculation which includes quantum electrodynamics contributions up to tenth order, as well as hadronic and electroweak contributions), the comparison with experiment (also the result of many unique developments) searches for physics beyond the standard model [3]. Such an alternate technique is the so-called "photon-recoil" method, which makes use of the relation $\alpha = [(2R_\infty/c)(h/m_a)(m_a/m_e)]^{1/2}$. Here R_∞ is the Rydberg constant, known to 5×10^{-12} from hydrogen spectroscopy [4]; h/m_a is the ratio of Planck's constant to the mass of a laser-excitable test atom and is determined by measuring its recoil velocity after absorbing or emitting a photon using atom interferometric techniques [5,6]; and m_a/m_e is the ratio of the mass of the test atom to that of the electron, which is most easily determined as a ratio of atomic masses.

Usually the precision-limiting link in the chain is the photon-recoil determination of h/m_a. Currently the most precise value is for ^{87}Rb with a fractional precision of 1.2 ppb [7]. When combined with the atomic mass of ^{87}Rb, measured to below 0.15 ppb [8,9], and of the electron (0.42 ppb) [4,10], this leads to the second most precise value for α with fractional precision 0.66 ppb. While the results for h/m_a for ^{87}Rb and also for ^{133}Cs [5,11] can be improved, there are now prospects for competitive h/m_a measurements on atoms with two valence electrons, in particular, Sr and Yb. Compared to the alkali metals, these atoms have the advantages of a spin-singlet ground state insensitive to magnetic fields; several laser-accessible excited states with different lifetimes, which facilitates laser cooling by providing transitions with different line widths; and a range of stable isotopes with different quantum statistics and collisional properties, which aids in investigating systematics. Several isotopes of Sr and Yb have now been used in studies of gases of ultracold bosons, fermions, and their mixtures (see, e.g. [12,13]), and in optical lattice clocks [14] and atom interferometers (e.g., [15,16]). Specifically, work is now in progress to measure h/m_a for several Yb isotopes using contrast atom interferometry starting with Bose-Einstein condensates [17], and related work on Sr isotopes can be foreseen. Since current values for atomic masses of stable isotopes of Sr and Yb have quoted uncertainties of ~13 ppb [18,19], improved measurements are required to match the anticipated sub-ppb values of h/m_a. Here we report Penning trap measurements of the atomic masses of the three most abundant isotopes of strontium and of the six most abundant isotopes of ytterbium with fractional precision below 0.2 ppb.

Cyclotron frequency ratios. The atomic masses were determined from cyclotron frequency ratios (CFRs) of pairs of ions simultaneously trapped in an 8.5 T Penning ion trap. In this Penning trap [20–22] we detect only the axial motion of an ion, which we do via the image current induced in a high-Q (30 000) superconducting circuit connected to one end-cap of the trap, coupled to a dc-SQUID. We used our two-ion technique [23,24] in which the cyclotron frequency, given by $f_c = (1/2\pi)qB/m$, of one ion at the center of the trap is measured using the "pulse-and-phase" method [25,26], while the other ion is temporarily parked in a large radius cyclotron orbit [27]. In the pulse-and-phase method the trap-modified cyclotron frequency f_{ct} is determined from the phase of the pulse-excited cyclotron motion, after free evolution for a period of up to 60 s, the phase being "read out" by phase-coherent transfer of the cyclotron action to the axial mode, using a pulse at the axial-to-cyclotron coupling frequency. The axial frequency f_z is also directly measured, and the magnetron frequency f_m is determined from the approximate expression $f_m = (f_z^2/2f_{ct})[1 + (9/4)\sin^2\theta_{\mathrm{mag}}]$, where θ_{mag} is obtained from separate measurements of f_m using the "avoided crossing technique" [26]. [In a Penning trap with a cylindrically symmetric electrostatic potential, θ_{mag} is the tilt angle between the symmetry axis and the magnetic field direction. More generally, it parametrizes the effects of both trap tilt and elliptical distortions of the potential; see Eq. (16) of Ref. [28].] The "true" cyclotron frequency of the inner ion f_c that it would have in the magnetic

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TABLE I. Average cyclotron frequency (i.e., inverse mass) ratios and applied systematic corrections for each Sr^{2+}/Kr^{2+} ion pair. N is the number of runs included in the average. Δ_{trap}, Δ_{ii}, and Δ_{fz} are the estimated systematic corrections in ppt, with estimated uncertainty in parentheses, due to trap field imperfections and image charges, ion-ion interaction, and shifts in f_z due to ion-detector interaction and differential voltage drift, respectively. σ_{syst} and σ_{stat} are the total systematic and statistical uncertainties (in ppt) for each average ratio. $\langle R\rangle$ is the average ratio after applying systematic corrections with σ_{syst} and σ_{stat} combined in quadrature in parentheses. The result for $^{86}Kr^{2+}/^{84}Kr^{2+}$ is also included.

Ion pair	N	Δ_{trap}	Δ_{ii}	Δ_{fz}	σ_{syst}	σ_{stat}	$\langle R\rangle$
$^{86}Sr^{2+}/^{84}Kr^{2+}$	5	5(25)	3(10)	6(25)	36	49	0.976 745 365 137(61)
$^{86}Kr^{2+}/^{86}Sr^{2+}$	8	1(6)	−1(10)	−3(30)	31	63	0.999 984 286 726(70)
$^{87}Sr^{2+}/^{84}Kr^{2+}$	4	6(38)	5(10)	9(30)	48	44	0.965 510 800 825(65)
$^{87}Sr^{2+}/^{86}Kr^{2+}$	7	3(13)	2(11)	5(27)	30	49	0.988 513 492 824(57)
$^{88}Sr^{2+}/^{84}Kr^{2+}$	3	8(50)	8(11)	7(24)	56	51	0.954 563 033 516(76)
$^{88}Sr^{2+}/^{86}Kr^{2+}$	6	4(26)	4(11)	4(37)	45	47	0.977 304 901 803(65)
$^{86}Kr^{2+}/^{84}Kr^{2+}$	8	5(24)	4(10)	8(24)	34	33	0.976 730 017 222(48)

field without the quadratic electrostatic potential, is then obtained using the Brown-Gabrielse invariance theorem, $f_c^2 = f_{ct}^2 + f_z^2 + f_m^2$ [28]. The ions are then repeatedly interchanged, typically giving 8–10 groups of measurements of f_c on each ion in a 10-h run. To obtain the average CFR the interleaved measurements of f_c versus time for the two ions are then fitted with similar polynomials, thus partly allowing for variation in the magnetic field.

For Sr we measured CFRs of each of $^{86,87,88}Sr^{2+}$ against both of $^{84,86}Kr^{2+}$; for Yb we measured $^{170,171,172,173,174,176}Yb^{4+}$ against one or both of $^{129,132}Xe^{3+}$. These Kr and Xe isotopes have been previously measured by us [22] relative to lighter ions that can be referenced to the ^{12}C mass standard. These choices of charge states and reference ions resulted in comparisons between ions of mass-to-charge (m/q) ratio in a relatively narrow range from 42 to 44, which reduces most systematic errors. This m/q ratio is well matched to our Penning trap with an axial detection frequency of 213 kHz. Single Sr^{2+} and Yb^{4+} ions were produced directly in the trap by electron beam ionization of atoms in metal vapor emitted from a miniature oven 2 m above the trap, followed by our usual procedures that eliminate all but a single ion of the desired isotope and charge state. The Kr^{2+} or Xe^{3+} reference ion was then made, and more easily, from a small quantity of injected gas (more than 99% isotopically enriched), and additional unwanted ions removed, with the Sr^{2+} or Yb^{4+} ion in a large cyclotron orbit. With the natural Sr and Yb samples available to us, producing single ions of the remaining stable isotopes, ^{84}Sr and ^{168}Yb, with abundances of 0.56% and 0.13%, respectively, would have been very difficult and was not attempted.

For every ion pair we took data at two values of the cyclotron radius of the inner ion ρ_{ci} (nominally 75 and 150 μm for Sr^{2+}/Kr^{2+}, and 50 and 100 μm for Yb^{4+}/Xe^{3+}), and two values of the parking radius of the outer ion ρ_{ck} (nominally 1.65 and 2.2 mm for all ions). To provide additional consistency checks and checks of systematic errors we also measured $^{84}Kr^{2+}$ against $^{84}Kr^{3+}$, $^{86}Kr^{2+}$ against $^{84}Kr^{2+}$, and, with $\rho_{ci} \geqslant 150\,\mu$m, $^{88}Sr^{2+}$ against $^{12}C^{16}O_2^{+}$. For Yb-Xe we also took data for the ratio $^{172}Yb^{4+}/^{132}Xe^{3+}$ with ρ_{ck} reduced to 1.1 mm, and also with mismatched ρ_{ck} for the Yb^{4+} and Xe^{3+}. In addition to these CFR measurements, and with only one ion in the trap, we periodically carried out measurements of the parameters C_4, C_6, and B_2 that characterize trap field imperfections [29], by measuring f_z as a function of magnetron and cyclotron radii. With the $^{172}Yb^{4+}$ and $^{132}Xe^{3+}$ pair we also verified the predicted $1/\rho_{ck}{}^3$ dependence of the shift to f_z of the inner ion due to the outer ion [23], and used the measured shift to check our calibration of ρ_{ck} versus product of drive pulse duration and voltage.

We base our mass results on only those runs with the smaller ρ_{ci} and larger ρ_{ck} since these runs have the smallest systematic shifts due to trap imperfections and ion-ion interaction. The average CFRs obtained from a weighted average over these runs, for each ion pair, are given in Table I for Sr and Table II for Yb. These tables also summarize estimates of the main systematic shifts to the average CFRs and their uncertainties.

In Tables I and II, under Δ_{trap} we include the effects of amplitude-dependent shifts to the ion mode frequencies due to trap imperfections and special relativity [30]. These are calculated from our measured values of the field imperfection parameters C_4, C_6, and B_2, and estimates of the amplitudes of the various motions of the ions. We take into account the frequency dependence of the transfer functions for the various drives, and the effects of small detunings between the drive frequencies and the ion mode frequencies. In all cases, because of the small ρ_{ci} and the partial cancellation between the ions in a pair with similar m/q, these shifts are less than 10 parts per 10^{12} (ppt). Under Δ_{trap} we also include the shift due to image charges induced in the trap electrodes using the result calculated by Porto [31]. Because of the different charge states, image charges result in CFR shifts of ∼30 ppt for Yb^{4+}/Xe^{3+}, while for Sr^{2+}/Kr^{2+} the effect is negligible. In the uncertainty of Δ_{trap} we also include the effects of the uncertainty in θ_{mag}, which we measure to be 0.54(3) degrees, and of nonidentical equilibrium positions of the two ions, combined with a gradient in the magnetic field. Systematic differences in the equilibrium positions of the ions occur due to differing contact potentials and charge patches on the electrodes combined with the m/q dependence of the trap voltage for a fixed f_z. The combined uncertainty due to these non-amplitude-dependent effects, which are proportional to the difference in m/q between ions in a pair, is estimated to be <11 ppt/u.

TABLE II. Average cyclotron frequency (i.e., inverse mass-to-charge) ratios and applied systematic corrections for each Yb^{4+}/Xe^{3+} ion pair (the column headings are as in Table I).

Ion pair	N	Δ_{trap}	Δ_{ii}	Δ_{fz}	σ_{syst}	σ_{stat}	$\langle R \rangle$
$^{129}Xe^{3+}/^{170}Yb^{4+}$	6	−29(10)	−2(7)	−102(51)	51	48	0.988 722 497 571(70)
$^{132}Xe^{3+}/^{170}Yb^{4+}$	6	−26(30)	1(7)	−104(49)	58	50	0.966 239 613 795(76)
$^{129}Xe^{3+}/^{171}Yb^{4+}$	3	−29(6)	−2(7)	−111(45)	45	55	0.994 549 921 376(71)
$^{132}Xe^{3+}/^{172}Yb^{4+}$	6	−27(22)	−1(7)	−93(50)	54	48	0.977 620 857 204(73)
$^{173}Yb^{4+}/^{129}Xe^{3+}$	3	31(8)	5(7)	117(47)	48	59	0.993 840 640 888(75)
$^{132}Xe^{3+}/^{173}Yb^{4+}$	3	−28(15)	−1(8)	−97(47)	50	68	0.983 317 277 812(84)
$^{174}Yb^{4+}/^{129}Xe^{3+}$	7	31(13)	5(8)	113(46)	49	35	0.988 123 111 853(61)
$^{132}Xe^{3+}/^{174}Yb^{4+}$	5	−29(11)	−2(8)	−126(64)	65	70	0.989 006 998 775(95)
$^{176}Yb^{4+}/^{129}Xe^{3+}$	5	33(24)	7(8)	132(59)	66	61	0.976 869 814 901(90)
$^{176}Yb^{4+}/^{132}Xe^{3+}$	4	31(5)	9(9)	120(68)	73	138	0.999 600 046 860(156)

Under Δ_{ii} we give the shifts to the average CFRs due to the Coulomb interaction with the outer ion, estimated according to the prescription given in [23]. Due to the small ρ_{ci} and large ρ_{ck} these shifts are <10 ppt, and in fact somewhat smaller for Yb^{4+}/Xe^{3+} than for Sr^{2+}/Kr^{2+}, despite the imbalance in charge states. This is because, in the limit of zero motion for the inner ion, the time-averaged Coulomb interaction with the outer ion shifts both f_{ct} and f_z of the inner ion by amounts, proportional to $1/{\rho_{ck}}^3$, that compensate each other when inserted into the invariance theorem. Hence, only effects that are higher order in the axial and radial amplitudes of the inner ion, corresponding to a modification of the electrostatic imperfection parameters C_4, C_6, etc, produce a shift to f_c. Hence, to lowest order, the residual shifts to f_c obtained using the invariance theorem, vary as ${\rho_{ci}}^2/{\rho_{ck}}^5$. We also include second-order ion-ion interaction effects, in particular, the shifts due to induced motion of the outer ion, resonantly back acting on the inner ion. These are negligible for all ratios except the mass doublets $^{86}Sr^{2+}/^{86}Kr^{2+}$ and $^{176}Yb^{4+}/^{132}Xe^{3+}$.

Under Δ_{fz} we include the estimated shift due to the interaction between the inner ion's axial motion and the high-Q detection circuit [20,21]. In our ratio measurements, in order to increase the damping time of the axial motion, we adjust the trap voltage so the ion's f_z is tuned 15 Hz below the resonance of the detection circuit. The ion-detector interaction results in a small, charge-dependent decrease in f_z. For Yb^{4+}/Xe^{3+} this produces shifts to the CFRs of ∼100 ppt. This is the largest systematic shift in Tables I and II. Based on previous measurements with Xe^{5+} ions [32], its uncertainty is conservatively estimated at 30 ppt. In the uncertainty for Δ_{fz} we also allow for the fact that, in our pulse-and-phase technique, f_{ct} and f_z are not measured simultaneously, and so the CFR can be affected by drifts in the trap voltages that are different for the two ions.

Under σ_{syst} we give the estimated total systematic uncertainty. Since we have allowed for correlations between certain systematics this is not exactly equal to the quadratic sum of the listed contributions. Under σ_{stat} we give the statistical uncertainty. This is determined from the internal uncertainties of the runs used in each average CFR, as provided by the routine that fits the f_c versus time data for the two ions. However, where the scatter of the results of the different runs about the weighted mean gave a reduced chi-squared value greater than 1, we increased the statistical errors by the square root of the reduced chi-squared value. This increased the statistical errors for $^{87}Sr^{2+}/^{86}Kr^{2+}$, $^{86}Sr^{2+}/^{84}Kr^{2+}$, $^{86}Sr^{2+}/^{86}Kr^{2+}$, $^{172}Yb^{4+}/^{132}Xe^{3+}$, $^{176}Yb^{4+}/^{129}Xe^{3+}$, and $^{176}Yb^{4+}/^{132}Xe^{3+}$.

In fact the greatest run-to-run scatter occurred for the two close doublet ratios $^{86}Sr^{2+}/^{86}Kr^{2+}$ and $^{176}Yb^{4+}/^{132}Xe^{3+}$, with anomalously large reduced chi-squared values of 2.08 and 4.11, respectively. Since we do not fully understand this increased scatter, for these two ratios we have less confidence in our uncertainty estimates. However, with the increased statistical error applied, these ratios have only a small effect on the resulting atomic masses.

To test the estimates of amplitude-dependent shifts in Tables I and II we applied the same model to correct the 47 additional runs obtained with larger inner and smaller outer ion radii. Averaging over all corrected ratios for

TABLE III. Atomic mass differences corresponding to the ratios given in Table I. The statistical, systematic, and total uncertainties are shown in parentheses.

Mass difference	Result (u)
$^{86}Sr - {}^{84}Kr$	1.997 762 999 2 (43)(31)(53)
$^{86}Kr - {}^{86}Sr$	0.001 349 896 5 (54)(27)(60)
$^{87}Sr - {}^{84}Kr$	2.997 379 769 9 (40)(43)(59)
$^{87}Sr - {}^{86}Kr$	0.998 266 865 9 (43)(26)(50)
$^{88}Sr - {}^{84}Kr$	3.994 114 530 6 (47)(52)(70)
$^{88}Sr - {}^{86}Kr$	1.995 001 625 6 (42)(40)(58)
$^{86}Kr - {}^{84}Kr$	1.999 112 899 9 (29)(30)(42)

TABLE IV. Atomic mass differences corresponding to the ratios given in Table II.

Mass difference	Result (u)
$4(^{129}Xe) - 3(^{170}Yb)$	5.814 821 686 (25)(27)(36)
$4(^{132}Xe) - 3(^{170}Yb)$	17.812 318 642 (26)(31)(40)
$4(^{129}Xe) - 3(^{171}Yb)$	2.810 128 892 (28)(23)(37)
$4(^{132}Xe) - 3(^{172}Yb)$	11.807 460 378 (25)(29)(38)
$3(^{173}Yb) - 4(^{129}Xe)$	3.195 525 184 (31)(25)(39)
$4(^{132}Xe) - 3(^{173}Yb)$	8.801 971 687 (36)(26)(45)
$3(^{174}Yb) - 4(^{129}Xe)$	6.197 479 237 (19)(26)(32)
$4(^{132}Xe) - 3(^{174}Yb)$	5.800 017 795 (37)(34)(50)
$3(^{176}Yb) - 4(^{129}Xe)$	12.208 600 696 (33)(35)(49)
$3(^{176}Yb) - 4(^{132}Xe)$	0.211 103 718 (73)(38)(82)

TABLE V. Final atomic masses (in u) of ^{86,87,88}Sr and ^{170,171,172,173,174,176}Yb compared with results of the AME2003 [18], and with preliminary results of the AME2013 [19]. In parentheses we give our propagated statistical and systematic uncertainties, the uncertainty due to the reference masses, and the total uncertainty, respectively.

Atom	This work	AME2003 [18]	Audi and Meng [19]
^{86}Sr	85.909 260 730 9 (35)(29)(79)(91)	85.909 260 2(12)	85.909 260 6(12)
^{87}Sr	86.908 877 497 0 (30)(34)(79)(91)	86.908 877 1(12)	86.908 877 5(12)
^{88}Sr	87.905 612 257 1 (31)(45)(80)(97)	87.905 612 1(12)	87.905 612 5(12)
^{170}Yb	169.934 767 241 (6)(10)(14)(18)	169.934 761 8(26)	169.934 767 6(23)
^{171}Yb	170.936 331 514 (9)(8)(15)(19)	170.936 325 8(26)	170.936 331 5(22)
^{172}Yb	171.936 386 655 (9)(10)(13)(18)	171.936 381 5(26)	171.936 387 1(22)
^{173}Yb	172.938 216 213 (8)(9)(14)(18)	172.938 210 8(26)	172.938 216 4(22)
^{174}Yb	173.938 867 539 (6)(10)(14)(18)	173.938 862 1(26)	173.938 867 7(22)
^{176}Yb	175.942 574 702 (11)(12)(15)(22)	175.942 571 7(28)	175.942 577 7(25)

Sr^{2+}/Kr^{2+} we obtain $[R(150,2200) - R(75,2200)] = -30(42)$ ppt and $[R(150,1650) - R(150,2200)] = 46(61)$ ppt, and for Yb^{4+}/Xe^{3+} $[R(100,2200) - R(50,2200)] = 12(40)$ ppt, $[R(50,1650) - R(50,2200)] = -52(52)$ ppt, where we indicate the nominal ρ_{ci} and ρ_{ck} in microns. Given that we expect amplitude-dependent systematics to vary as ${\rho_{ci}}^2$ or higher powers of the inner ion radius and as $1/{\rho_{ck}}^5$ or higher inverse powers of the outer ion radius, this is a good test of these systematics. The two close doublet ratios *did* show additional shifts that depended on ρ_{ci} and ρ_{ck}, but within our estimated uncertainties. The additional measurements for $^{88}Sr^{2+}/^{12}C^{16}O_2{}^+$ at larger ρ_{ci}, and $^{172}Yb^{4+}/^{132}Xe^{3+}$ at smaller, and imbalanced ρ_{ck} also gave results consistent with our model and error estimates. As a test that we have not underestimated shifts that depend only on the difference in m/q between the ions, our measurement of the CFR for $^{84}Kr^{2+}/^{84}Kr^{3+}$ gave 0.666 662 308 605 with a statistical uncertainty of 29 ppt, to be compared with 0.666 662 308 524, obtained using the predicted value allowing for electron mass and ionization energies. The difference is well within the above estimate of 11 ppt per unit difference in m/q. Also, the result for $^{84}Kr^{2+}/^{86}Kr^{2+}$ in Table I is in good agreement with our previous results [9,22].

Mass difference equations and final masses. We first convert the CFRs into mass differences between neutral atoms, accounting for the mass and ionization energies of the missing electrons [4,33,34]. The mass differences corresponding to the ratios in Tables I and II are given in Tables III and IV.

These mass differences are intended for use in global least-squares atomic mass evaluations. However, here, for simplicity, we obtain strontium and ytterbium atomic masses using these mass difference equations and our previous measurements of ^{84,86}Kr and ^{129,132}Xe from [22]. Where more than one ratio was measured for a Sr or Yb isotope we take the $1/\sigma^2$ weighted average, linearly propagating the systematic uncertainty and the uncertainties in the reference masses. In Table V, our final atomic masses are presented and compared to values in the latest published global atomic mass evaluation, the AME2003 [18], and preliminary results of the upcoming evaluation, the AME2013 [19].

As Table V shows, for Sr we are in good agreement with the results of the AME2003, while for Yb there is a systematic 2σ discrepancy of $\sim 5.5(2.6) \times 10^{-6}$ u. However, this discrepancy is removed in the preliminary results of the AME2013. These previous Sr and Yb atomic masses were derived from a least-squares adjustment making use of nuclear reaction data and non-Penning trap mass spectrometry. Our new results, which are limited by the precision of the ^{84,86}Kr and ^{129,132}Xe references, should be sufficiently precise to meet the requirements of the first generation of photon-recoil experiments on two-electron atoms. Higher precision will require dedicated measurements that directly relate the required isotope masses to the carbon mass standard.

Acknowledgments. We acknowledge early contributions to this work from Brianna Mount and assistance with the manuscript from Austin Erickson and Bridget Wesson. This work was supported by the NSF AMO program under PHY-0968889. M.H. also acknowledges support from the Max Planck Society.

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