Atomic masses of ^{6}Li, ^{23}Na, ^{39,41}K, ^{85,87}Rb, and ^{133}Cs

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The atomic masses of the alkali-metal isotopes ^{6}Li, ^{23}Na, ^{39,41}K, ^{85,87}Rb, and ^{133}Cs have been obtained from measurements of cyclotron frequency ratios of pairs of ions simultaneously trapped in a Penning trap. The results, with one standard deviation uncertainty, are: $M(^{6}\mathrm{Li}) = 6.015\,122\,887\,4(16)$ u, $M(^{23}\mathrm{Na}) = 22.989\,769\,282\,8(26)$ u, $M(^{39}\mathrm{K}) = 38.963\,706\,485\,6(52)$ u, $M(^{41}\mathrm{K}) = 40.961\,825\,257\,4(48)$ u, $M(^{85}\mathrm{Rb}) = 84.911\,789\,739(9)$ u, $M(^{87}\mathrm{Rb}) = 86.909\,180\,535(10)$ u, and $M(^{133}\mathrm{Cs}) = 132.905\,451\,963(13)$ u. Our mass of ^{6}Li yields an improved neutron separation energy for ^{7}Li of 7251.1014(45) keV.

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I. INTRODUCTION

We report atomic-mass measurements of the alkali-metal isotopes ^{6}Li, ^{23}Na, ^{39}K, ^{41}K, ^{85}Rb, ^{87}Rb, and ^{133}Cs with fractional uncertainties from 0.3 to 0.1 parts per billion (ppb), obtained by measuring ratios of cyclotron frequencies of pairs of ions simultaneously trapped in a cryogenic Penning trap. The alkali metals have great importance in physics, since their electronic structure facilitates many applications including atomic clocks, tests of fundamental physics, and the production and manipulation of ultracold atoms. As an immediate motivation for precise alkali-metal atomic masses, there are photon-recoil experiments, intensively developed for both ^{133}Cs [1,2] and ^{87}Rb [3,4], which measure h/m_a, where h is Planck's constant and m_a is the mass of the atom in Système International units. Making use of the expression,

$$\alpha^2 = \left(\frac{2R_\infty}{c}\right)\left(\frac{m_a}{m_e}\right)\left(\frac{h}{m_a}\right), \tag{1}$$

where R_∞ is the Rydberg constant, c is the velocity of light, and m_e is the mass of the electron, these experiments yield the current second and third most precise values for the fine structure constant $\alpha = e^2/(4\pi\varepsilon_0\hbar c)$, with uncertainties of 4.5 ppb for ^{87}Rb [4] and 8 ppb for ^{133}Cs [1,5], with further improvements in precision expected. Hence, precise alkali-metal atomic masses, in conjunction with the atomic mass of the electron, known to 0.42 ppb [6–8], are needed to obtain the ratio m_a/m_e. (The Rydberg constant is known to 6.6×10^{-12} from precision hydrogen spectroscopy [8].)

Although they do not yield the most precise value of α, these photon-recoil determinations are especially important because they enable the current most precise method—which obtains α to 0.37 ppb by combining complex theory [9] and experiment [10] for the magnetic moment of the electron—to instead be used as a precise test of quantum electrodynamics and to search for physics beyond the standard model [11]. Although the atomic masses of ^{23}Na, ^{85,87}Rb, and ^{133}Cs have previously been measured to $\leqslant$0.2 ppb [12], additional measurements at the same level of precision provide useful checks. Additional reasons for measuring the atomic masses of the alkali metals precisely are that they make convenient references for mass spectrometers used to measure masses of short-lived isotopes [13], and that they provide reference points for the global evaluation of atomic masses [14].

II. METHOD

We measured the ratios of cyclotron frequencies of alkali-metal ions to reference ions of known atomic mass using an 8.5-tesla cryogenic orthogonally compensated [15], single-ion Penning trap. This Penning trap, originally developed at the Massachusetts Institute of Technology (MIT), uses image-charge detection of the ion's axial mode (at frequency $f_z = 213$ kHz) using a high-Q superconducting resonant inductor and a direct current superconducting quantum interference device and addresses the trap-modified-cyclotron and magnetron modes by coupling to the axial mode using rf drives. The trap-modified-cyclotron frequency f_{ct} is measured using the pulse-and-phase (PNP) technique, which measures the accumulated phase following pulsed excitation of the trap-cyclotron mode [16]. The magnetron frequency f_m is calculated from f_{ct} and f_z and the trap-tilt angle [15], which is obtained using less-frequent measurements of f_m by the avoided-crossing technique [17]. The true cyclotron frequency $f_c = qB/2\pi m$ is then derived from f_z, f_{ct}, and f_m using the Brown-Gabrielse invariance theorem [18].

Single ions were created inside the trap by electron-beam ionization of gas or vapor introduced through a small hole in the upper end cap of the trap. With the exception of ^{6}Li, for which we made our own source consisting of a niobium tube containing ^{6}Li (>90% enrichment), the alkali-metal vapors were produced using commercial alkali-metal dispensers [19], with natural isotopic abundances (^{23}Na 100%, ^{39}K 93.3%, ^{41}K 6.7%, ^{85}Rb 72%, ^{87}Rb 28%, ^{133}Cs 100%). All unwanted ions were removed from the trap by selectively exciting their axial motion and then lowering the potential on the lower end cap until they combined with it.

To reduce the effects of variation in the magnetic field, we simultaneously trapped the alkali-metal ion and its reference, and alternated them between the center of the trap, where the cyclotron frequency was measured, and a large 1.5–2.5-mm radius cyclotron orbit, which served to temporarily park the

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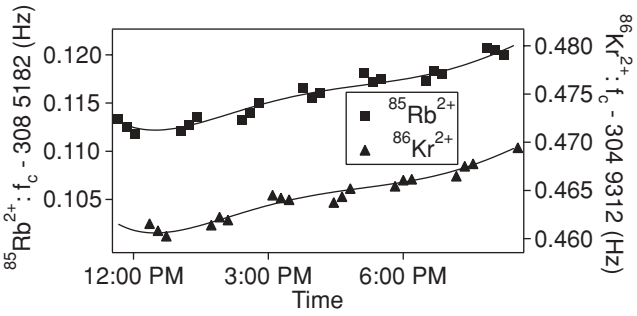

FIG. 1. An example of cyclotron frequency ratio data. The cyclotron frequency of one ion at the center of the trap is measured three times, while the other ion remains in a large cyclotron orbit. The ions are then interchanged, and the process is repeated. The vertical scales give the cyclotron frequencies of the two ions in hertz, and the horizontal scale is the time during the day. The cyclotron frequency ratio is obtained from a simultaneous fit of similar polynomials to both ions' cyclotron frequencies. The ratio obtained from this particular run had a statistical uncertainty of 7×10^{-11}.

other ion [20,21]. In each case, the ion to be measured and its reference ion had the same charge state, 1^+,2^+, or 3^+, and similar mass. A single cyclotron frequency ratio measurement was obtained from a data run lasting up to 15 h, limited by the ion lifetime or the need to refill a liquid-nitrogen dewar. An example of data from a single run is given in Fig. 1. In addition to the measurements of cyclotron frequency ratios, we periodically performed a series of additional measurements to estimate systematic corrections and errors. These included measurements of f_z as a function of the magnetron radius, cyclotron radius, and axial amplitude, to determine the field imperfection parameters C_4,C_6, and B_2, etc. [15], and especially the voltage setting of the compensation electrode to make $C_4 = 0$. These parameters determine the amplitude-dependent shifts for the mode frequencies. In order to study ion-ion interaction effects, we also carried out ratio measurements in which we systematically varied the cyclotron radius of the outer parked ion and the cyclotron radius of the inner ion. Further details of our general procedures can be found in Refs. [22–24].

III. CYCLOTRON FREQUENCY RATIO MEASUREMENTS

The cyclotron frequency ratio measurements were carried out at various times over a 14-month period in the order ^{85}Rb,^{87}Rb,^{39}K,^{41}K,^{133}Cs,^{23}Na and ^{6}Li. The average ratios from which the alkali-metal masses were obtained are given in Table I. Table I also lists estimates, with uncertainties, of the main systematic shifts for the average cyclotron frequency ratios. These result from differential shifts to f_z and f_{ct} for the two ions and are mainly due to imperfections in the electric and magnetic trapping fields combined with significant axial and cyclotron mode amplitudes, Coulomb interaction between the two ions, and shifts to f_z due to ion-detector interaction and differential voltage drift [24]. We label these $\Delta_{\rm trap}$,$\Delta_{\rm ii}$, and $\Delta_{\rm fz}$, respectively. Besides contributions that depend on differences in m/q between the ions, these shifts and their uncertainties also allow for run-specific systematics, such as possible differences in the amplitudes and detunings of the various rf drives, and settings of the compensation voltages, etc. In addition to the 47 runs used to obtain the alkali-metal ratio data in Table I, an additional 49 runs were used as checks. These included 23 with previously measured masses such as $^{12}C\ ^{16}O_2{}^+/^{40}Ar^+$, as well as runs in which the cyclotron radii for the inner and outer ions were systematically varied. In general, the statistical uncertainties in Table I were determined from the fitting errors for the individual runs. However, for the three ratios $^{23}Na^+/^{12}C_2{}^+$,$^{40}Ar^+/^{41}K^+$, and $^{84}Kr^{2+}/^{85}Rb^{2+}$, where it happened that the scatter of repeated runs was larger than that expected from the fitting uncertainties (i.e., the χ^2 was larger than 1), this error was increased by the square root of χ^2. So, in effect, the quoted statistical uncertainty for these ratios was based on the observed scatter. This resulted in an increase by factors of 1.9, 1.3, and 1.2 for these three ratios, respectively.

TABLE I. Average cyclotron frequency (i.e., inverse mass) ratios and systematic corrections for each ion pair. N is the number of runs included in the average. $\Delta_{\rm trap}$,$\Delta_{\rm ii}$, and $\Delta_{\rm fz}$ are the estimated systematic corrections in parts per trillion (ppt), with estimated uncertainty in parentheses, due to trap field imperfections, ion-ion interaction, and shifts in f_z due to ion-detector interaction and differential voltage drift, respectively. $\sigma_{\rm syst}$ is the total systematic error, and $\sigma_{\rm stat}$ is the statistical error (in ppt) for each average ratio. $\langle R\rangle$ is the average ratio after applying systematic corrections, with statistical and systematic uncertainties combined in quadrature, in parentheses.

Ion pair	N	$\Delta_{\rm trap}$	$\Delta_{\rm ii}$	$\Delta_{\rm fz}$	$\sigma_{\rm syst}$	$\sigma_{\rm stat}$	$\langle R\rangle$
$^{6}Li_2{}^+/^{12}C^+$	7	3(55)	−2(20)	−1(33)	67	249	0.997 485 741 614(258)
$^{12}C_2{}^+/^{23}Na^+$	9	−118(56)	−32(53)	−1(1)	77	74	0.957 906 091 266(107)
$^{40}Ar^+/^{39}K^+$	7	−170(95)	14(31)	−9(17)	101	33	0.975 009 239 187(106)
$^{41}K^+/^{40}Ar^+$	7	145(75)	−14(32)	−11(5)	82	52	0.975 600 318 068(97)
$^{40}ArH^+/^{41}K^+$	1	1(11)	0(1)	6(5)	12	147	0.999 795 387 703(148)
$^{86}Kr^{3+}/^{85}Rb^{3+}$	1	−40(18)	7(1)	−37(19)	26	78	0.988 373 496 338(82)
$^{86}Kr^{2+}/^{85}Rb^{2+}$	2	46(37)	−15(11)	106(27)	47	50	0.988 373 570 567(69)
$^{85}Rb^{2+}/^{84}Kr^{2+}$	4	1(52)	−14(15)	37(27)	60	47	0.988 219 481 271(76)
$^{87}Rb^{2+}/^{86}Kr^{2+}$	3	−36(39)	18(8)	74(28)	49	49	0.988 510 045 784(69)
$^{133}Cs^{3+}/^{132}Xe^{3+}$	3	−8(31)	2(3)	12(15)	35	70	0.992 466 003 022(78)
$^{133}Cs^{3+}/^{129}Xe^{3+}$	3	30(27)	−9(7)	31(27)	39	70	0.969 897 994 594(80)

Some further details on the specific measurements are given in the following.

A. ^{85,87}Rb and ^{133}Cs

The measurements of ^{85,87}Rb and ^{133}Cs were enabled by our previous measurements of the masses of ^{84,86}Kr and ^{129,132}Xe against singly charged reference ions using single-ion techniques [23]. Here, we measured either the 2^+ or 3^+ alkali-metal ion against an ^{84,86}Kr or ^{129,132}Xe ion in the same charge state. For these heavy atoms, it is necessary to use higher-charge states to increase the cyclotron frequency, and, hence, to reduce the sensitivity of the ratio to systematic shifts in the axial frequency, since this varies as $(f_z/f_{ct})^2$ and f_z is fixed by the detection circuit. It is also advantageous for reducing the phase-evolution time required for measuring f_{ct} to a given precision. But furthermore, because of the increased signal size, it enables the use of smaller cyclotron radii and axial amplitudes for the ion being measured, which reduces the systematic shifts Δ_{trap} and Δ_{ii}—in fact, the estimated uncertainties for both these shifts were smaller for these multiply charged pairs than for the singly charged nondoublet pairs. As an overall test of systematics when measuring multiply charged nondoublets with our two-ion technique, we measured the ratios $^{84}Kr^{2+}/^{86}Kr^{2+}$ and $^{129}Xe^{3+}/^{132}Xe^{3+}$, with the results 0.976 730 017 167(56)(42)(70) and 0.977 260 673 537(31)(55)(63), respectively, where the systematic, statistical, and total errors are given in parentheses. These are in excellent agreement with our previous results obtained with only one ion in the trap [23].

B. ^{39,41}K

For ^{39,41}K, we primarily used the reference ^{40}Ar [14,25]. We investigated systematic shifts using the more severe non-mass-doublet ratios $^{12}C^{16}O_2{}^{+}/^{40}Ar^{+}$ and $^{40}Ar^{+}/^{18}O_2{}^{+}$ [24]. We also performed one ratio measurement run each for the ratio $^{41}K^{+}/^{40}ArH^{+}$ and the additional test ratio $^{40}ArH^{+}/^{40}Ar^{+}$. ($^{40}ArH^{+}$ ions were made serendipitously on two occasions from single $^{40}Ar^{+}$ ions already in the trap via collisions with background hydrogen gas.) A correction of 92(2) ppt has been made to allow for the polarizability shift of the cyclotron frequency of $^{40}ArH^{+}$ in our 8.5-tesla magnetic field [26]. Hence, the $^{40}ArH^{+}/^{41}K^{+}$ ratio in Table I corresponds to the inverse mass ratio.

C. ^{23}Na

We obtained the mass of ^{23}Na from the ratio $^{12}C_2{}^{+}/^{23}Na^{+}$. The combination of the large fractional difference in m/q and the need to use larger amplitudes because of the lower mass resulted in the largest systematic shifts due to ion-ion interactions. We carefully investigated these shifts by taking a series of ratio measurements where we systematically varied the parking radius of the outer ion and the cyclotron radius (and, hence, axial amplitude) of the inner ion used in the PNP measurement. Additional tests of our model were made by taking ratio data with various cyclotron radii for the worse nondoublet $^{23}Na^{+}/^{20}Ne^{+}$ [27].

D. ^{6}Li

Here, we measured the mass doublet $^{6}Li_2{}^{+}/^{12}C^{+}$. Implementing our techniques for such light ions was difficult since the energy in the ion's axial mode is proportional to mf_z^2, and we use a relatively low detection frequency. A significant increase in the amplitudes was prevented because of increased anharmonicity, which increases phase measurement noise. Further, producing and isolating a single $^{6}Li_2{}^{+}$ was challenging due to the small (<1%) dimer population in the lithium vapor [28]. Nevertheless, we were able to isolate single $^{6}Li_2{}^{+}$ ions on eight occasions. Because a mass doublet was being measured, the uncertainties were dominated by statistics. Attempts at isolating a single $^{7}Li_2{}^{+}$ ion, both using a commercial alkali-metal vapor source and our own source, were unsuccessful due to an overwhelming signal from background $^{14}N^{+}$ ions.

TABLE II. Atomic mass differences and derived alkali-metal atomic masses corresponding to the ratios given in Table I. For the mass differences, the systematic, statistical, and total uncertainties are shown in parentheses. For the alkali-metal masses, we give the total uncertainty, which includes the propagated uncertainty of the references.

Ion pair	Mass difference	Result (u)	Atom	Mass (u)
$^{6}Li_2{}^{+}/^{12}C^{+}$	$2(^{6}Li) - {}^{12}C$	0.030 245 774 8(7)(30)(31)	^{6}Li	6.015 122 887 4(16)
$^{12}C_2{}^{+}/^{23}Na^{+}$	$2(^{12}C) - {}^{23}Na$	1.010 230 717 2(19)(18)(26)	^{23}Na	22.989 769 282 8(26)
$^{40}Ar^{+}/^{39}K^{+}$	$^{40}Ar - {}^{39}K$	0.998 676 636 9(41)(13)(43)	^{39}K	38.963 706 485 6(52)
$^{41}K^{+}/^{40}Ar^{+}$	$^{41}K - {}^{40}Ar$	0.999 442 134 8(33)(21)(39)	^{41}K	40.961 825 257 4(48)
$^{40}ArH^{+}/^{41}K^{+}$	$^{40}Ar + {}^{1}H - {}^{41}K$	0.008 382 900 5(5)(60)(61)	^{41}K	40.961 825 254 1(68)
$^{86}Kr^{3+}/^{85}Rb^{3+}$	$^{86}Kr - {}^{85}Rb$	0.998 820 891 7(23)(67)(71)	^{85}Rb	84.911 789 736(10)
$^{86}Kr^{2+}/^{85}Rb^{2+}$	$^{86}Kr - {}^{85}Rb$	0.998 820 889 1(41)(43)(59)	^{85}Rb	84.911 789 739(10)
$^{85}Rb^{2+}/^{84}Kr^{2+}$	$^{85}Rb - {}^{84}Kr$	1.000 292 012 1(51)(39)(64)	^{85}Rb	84.911 789 743(10)
$^{87}Rb^{2+}/^{86}Kr^{2+}$	$^{87}Rb - {}^{86}Kr$	0.998 569 906 8(42)(42)(59)	^{87}Rb	86.909 180 535(10)
$^{133}Cs^{3+}/^{132}Xe^{3+}$	$^{133}Cs - {}^{132}Xe$	1.001 296 880 3(46)(93)(103)	^{133}Cs	132.905 451 966(15)
$^{133}Cs^{3+}/^{129}Xe^{3+}$	$^{133}Cs - {}^{129}Xe$	4.000 671 100 7(50)(90)(103)	^{133}Cs	132.905 451 959(14)
$^{86}Kr^{2+}/^{84}Kr^{2+}$	$^{86}Kr - {}^{84}Kr$	1.999 112 904 6(48)(36)(60)		
$^{132}Xe^{3+}/^{129}Xe^{3+}$	$^{132}Xe - {}^{129}Xe$	2.999 374 222 9(40)(73)(83)		

TABLE III. Final atomic masses (in u) of ^{6}Li,^{23}Na,^{39,41}K,^{85,87}Rb, and ^{133}Cs compared with results of the AME2003 [14] and other recent Penning trap measurements.

Atom	This paper	AME2003	Other recent results
^{6}Li	6.015 122 887 4(16)	6.015 122 795(16)	6.015 122 889(26) [33] 6.015 122 890(40) [34]
^{23}Na	22.989 769 282 8(26)	22.989 769 280 9(29)	
^{39}K	38.963 706 485 6(52)	38.963 706 68(20)	38.963 706 52(17) [35]
^{41}K	40.961 825 257 4(48)	40.961 825 76(21)	
^{85}Rb	84.911 789 739(9)	84.911 789 738(12)	
^{87}Rb	86.909 180 535(10)	86.909 180 527(13)	
^{133}Cs	132.905 451 963(13)	132.905 451 933(24)	

IV. ATOMIC MASS DIFFERENCE EQUATIONS AND MASSES

We first convert the cyclotron frequency ratios into mass differences between neutral atoms. To do this, we account for the mass of the missing electron and the ionization and chemical binding energies, which we obtain from Refs. [29–32]. The mass differences corresponding to the ratios in Table I are given in the third column of Table II. Because the ratios $^{84}Kr^{2+}/^{86}Kr^{2+}$ and $^{129}Xe^{3+}/^{132}Xe^{3+}$ that we measured in this paper are more precise than those given in Ref. [23], we have also included the corresponding mass differences in Table II.

These mass differences are the primary results of this paper and are intended for use in global least-squares mass evaluations. However, we also obtain preliminary alkali-metal atomic masses by simply treating the other atomic masses as known references, with uncorrelated uncertainties. We used the masses of ^{1}H and ^{40}Ar as given in the Atomic Mass Evaluation (AME2003) [14], and the masses of ^{84,86}Kr and ^{129,132}Xe from Ref. [23]. For ^{41}K,^{85}Rb, and ^{133}Cs, where more than one ratio was measured, we take the weighted average, linearly propagating the systematic uncertainty and the uncertainties in the reference masses. Our final atomic masses, compared to values in the AME2003 and other, more recent Penning trap measurements [33–35], are presented in Table III.

V. DISCUSSION AND CONCLUSION

Our mass for ^{6}Li is higher than the AME2003 result, which was mainly derived from a Penning trap measurement using image-current detection [36], by 15(3) ppb. Hence, we confirm the recent results from Penning traps using time-of-flight techniques [33,34] but with an order-of-magnitude improvement in precision. Combining our new mass for ^{6}Li with the 0.63-ppb mass for ^{7}Li [34] and the neutron mass from Ref. [8] yields an improved neutron separation energy of ^{7}Li [Q value for the ^{6}Li$(n,\gamma)^{7}$Li reaction] of 7251.1014(45) keV. This is in good agreement with the value of 7251.10(4) keV reported in Ref. [34] and 7251.02(9) of Ref. [37] and deviates significantly from the AME2003 [14] value of 7249.97(8) keV.

Our masses of ^{23}Na,^{85}Rb, and ^{87}Rb are in excellent agreement with, and our mass for ^{133}Cs is only one standard deviation higher than, the masses obtained at MIT [12], on which the values in the AME2003 are based. Although our main improvement is to halve the uncertainty for ^{133}Cs, our measurements provide an important cross-check, particularly because of the large run-to-run variation for Rb and Cs data observed in Ref. [12]. Although we used the same Penning trap apparatus, there are several important differences between the two sets of measurements. These include differences in the shimming of the superconducting magnet and in the precise positioning of the Penning trap, the use of a different detection circuit with a higher resonant frequency (213 kHz versus 160 kHz), and a trap with different electrostatic-field imperfections due to different charge patches on the electrodes. Furthermore, compared to the techniques used in Ref. [12], where ions, sometimes with different charge states, were compared by repeatedly making them inside the trap, we used reference ions of the same charge state and similar mass, and, by simultaneously trapping both ions, measured both cyclotron frequencies under more similar conditions. In general, we also used different reference ions, used smaller mode amplitudes, and took more data for each ion, facilitated by the lower daytime magnetic-field noise in our laboratory. Conversely, the good agreement between our results and the MIT results provides a check of our previous measurements of the masses of ^{84,86}Kr and ^{129,132}Xe.

Our measurements of the masses of ^{39}K and ^{41}K, respectively, are 5(5) and 12(5) ppb lower than the results in the AME2003, which are partially based on nuclear reaction data. For ^{39}K, they are in good agreement with recent time-of-flight Penning trap measurements reported in Ref. [35]. In both cases, our results reduce uncertainties by over a factor of 30.

In conclusion, including the result for ^{7}Li obtained by the Stockholm group [34], the masses of all the stable alkali metals have now been measured to better than 1 ppb. This implies that, for any alkali metal, if a sufficiently accurate value of h/m_a could be measured, a value for α could be derived that has precision comparable to that currently obtained from the electron g factor.

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[27] From this $^{23}Na^{+}/^{20}Ne^{+}$ data, we can obtain the mass difference $M[^{23}\mathrm{Na}] - M[^{20}\mathrm{Ne}] = 2.997\,329\,1147(137)(9)(137)$ u, with systematic, statistical, and total uncertainties in parentheses, and, using the mass of ^{20}Ne in Ref. [14], $M[^{23}\mathrm{Na}] = 22.989\,769\,290(14)$u, which agrees with the more precise result from $^{12}C_2{}^{+}/^{23}Na^{+}$. However, because the uncertainty is large and mainly systematic, we do not use this value for determining our final ^{23}Na mass.

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