High-Accuracy Mass Determination of Neutron-Rich Rubidium and Strontium Isotopes

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Abstract

The Penning trap mass spectrometer ISOLTRAP, installed at the on-line isotope separator ISOLDE at CERN, has been used to measure atomic masses of ^{88,89,90m,91,92,93,94}Rb and ^{91,92,93,94,95}Sr. Using a resolving power of $R \approx 1$ million a mass accuracy of typically 10 keV was achieved for all nuclides. Discrepancies with older data are analyzed and discussed, leading to corrections to those data. Together with the present ISOLTRAP data these corrected data have been used in the 1995 mass adjustment.

Key words: ATOMIC MASSES ^{88,89,90,91,92,93,94}Rb – ^{91,92,93,94,95}Sr; Measured masses. On-line mass spectrometry. Penning trap. Least-squares adjustment of data.

1 Introduction

Precision mass measurements of unstable neutron-rich nuclei contribute to a better understanding of the evolution of nuclear binding as one departs from the line of beta stability towards more and more exotic systems. Direct mass measurements in long isotopic chains are of particular importance for a stringent test of model predictions of masses, which for neutron-rich isotopes are of great importance for the modelling of the nuclear astrophysical r-process. Furthermore accurate data in such chains can give a firm anchorage for mass links to other isotopes by Q-values and they can test the reliability of using β-decay chains for the determination of masses very far away from stability. The measurements on neutron-rich strontium and rubidium isotopes reported here are the most precise measurements in this region. They were carried out with the Penning trap mass spectrometer ISOLTRAP installed at the on-line mass separator facilty ISOLDE at CERN. These were the first mass measurements performed after the move of ISOLTRAP from the shut-down ISOLDE-II facility to the new PS-Booster-ISOLDE. The experimental results presented in this paper have already been included in the 1995 mass evaluation.

2 Experimental setup and procedures

A very detailed description of the spectrometer as it was used for the measurements reported here is given in [1]. It should be noted that in the meantime the spectrometer has been considerably improved, as discussed in [2,3].

The version of the ISOLTRAP setup used in this work consisted of two main sections, the heart of each being a Penning trap. The task of the first Penning trap, situated in a 0.7 T electromagnet was the preparation of cooled low-energy ion bunches. The second part, a high-precision Penning trap in a 6T superconducting magnet, was employed for the actual mass measurement via a cyclotron frequency determination.

A mass measurement started with the collection of radioactive ions delivered by the ISOLDE on-line mass separator. The ions were implanted into a rhenium foil placed at the entrance of the first Penning trap. Then the foil was

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turned and heated by a pulsed current. In this way the radioactive atoms were released and surface-ionized. The resulting ions were trapped and cooled by collisions with a buffer gas. Simultaneously the ion motion was centered in the trap by a mass selective cooling technique based on radiofrequency excitation [4,5]. The ions were then ejected from the trap and transferred to the second trap. Here the ion pulse was captured in flight. Possible isobaric contaminations were removed by applying a strong dipole electric RF field at the (reduced) cyclotron frequency of the unwanted ions. Subsequently the motion of the ions of interest was driven by an azimuthal quadrupole RF field at an excitation frequency close to their cyclotron frequency. For the detection of the cyclotron resonance a time-of-flight technique was used. For this, the ions were ejected axially out of the trap and allowed to drift through the inhomogeneous part of the magnetic field where radial energy is converted into axial energy. Finally, their time of flight from the trap to the detector was observed. This cycle was repeated with different excitation frequencies, scanning the expected resonance frequency.

The determination of the mean time of flight as a function of the applied frequency yields a resonance curve with a minimum at the ions cyclotron frequency

$$\nu_c = \frac{1}{2\pi}\frac{q}{m} \cdot B \tag{1}$$

where q/m is the charge to mass ratio of the ions and B the magnetic field strength. An example of such a cyclotron resonance is depicted in Fig. 1 for the case of ^{95}Sr. A Gaussian is fitted to the resonance curve. The width of the resonance is $\Delta\nu_c(FWHM) \approx 1.0\,\mathrm{Hz}$, corresponding to $\Delta m(FWHM) \approx 92\,\mathrm{keV}$. With a cyclotron frequency of $\nu_c \approx 1$ MHz this width leads to a resolving power of $R \approx 1$ million. The statistical error for the center frequency is typically of the order of $\delta\nu_c = 0.03\,\mathrm{Hz}$ for 10^4 detected ions, corresponding to $\delta\nu_c/\nu_c \approx 5 \cdot 10^{-8}$. A fit by the true theoretical line shape having small side bands [4] instead by the Gaussian does not change the value of the center frequency of the resonance but improves slightly the statistical accuracy. In view of the larger estimated systematic error of $1 \cdot 10^{-7}$ (see below), a fit by a Gaussian is regarded as sufficient.

The magnetic field has to be known to the same accuracy or better in order to use eq. (1) to convert the measured frequencies into mass values. It can be determined by frequently measuring the cyclotron frequency of a reference nuclide. For the measurements reported here ^{85}Rb was used as reference. In this way the uncertainty due to a possible drift of the magnetic field of the superconducting magnet was kept well below $1 \cdot 10^{-7}$.

Finally, for each nuclide under investigation the ratio of the cyclotron frequen-

cies $r = \nu_{ref}/\nu$, and its statistical error $\sigma_{r,stat}$, is calculated. To account for the sum of systematic errors due to an incomplete correction of magnetic field drifts and possible mass dependent effects that arise in an on-line experiment, an estimated systematic error of $1 \cdot 10^{-7}$ is added quadratically to the error of the averaged frequency ratio [1].

The frequency ratio r and its total error σ_r is the final direct result of an ISOLTRAP mass comparison. It can be converted into an atomic mass value m by multiplying the frequency ratio with the atomic mass of the reference nuclide m_{ref} and taking into account the electron mass m_e by

$$m = r \cdot (m_{ref} - m_e) + m_e \tag{2}$$

with the statistical uncertainty

$$\sigma_m^2 = (r \cdot \sigma_{m,ref})^2 + ((m_{ref} - m_e) \cdot \sigma_r)^2 \quad . \tag{3}$$

Given the low ion velocity in the trap no relativistic correction is necessary. The contribution of the binding energy of the electron is small compared to the total error of the present measurements and therefore neglected.

3 Measurements

The measurements were carried out at the PS-Booster ISOLDE facility. Radioactive ions were produced by 1 GeV protons impinging on a heated niobium foil target. A tungsten surface ionizer was used to ionize the radioactive alkali and earth alkali isotopes produced in the nuclear reaction. The mass separated 60-keV beam was then transported to the ISOLTRAP setup and implanted in the collection foil.

In the cases of ^{88,89,90m,91}Rb and ^{91,92,93,94,95}Sr a collection time of T_{coll}= 9 s to 24 s was sufficient to obtain between 1 and 5 resonance curves, each taking typically 90 s. Only in the cases of the shorter-lived isotopes ^{92,93,94}Rb one single collection could not yield a full resonance curve. Therefore the following procedure was chosen: after one collection lasting about three half-lives, the cycle was repeated several times with the same RF excitation frequency. Then a new collection was made to perform the next series of cycles with a slightly different frequency. After the measurements for one nuclide were achieved, the collection foil was heated for typically 5 to 10 min at the maximum electrical current of $I = 20\ A$ to clean it from the residual ions.

The reference ^{85}Rb was delivered from a test ions source integrated in ISOLTRAP. A cyclotron frequency measurement of this nuclide was performed every 1.5

to 2 h in order to monitor possible slow drifts of the magnetic field caused by temperature fluctuations in the laboratory.

3.1 Isobars and Isomers

Penning trap mass measurements require rather clean ion samples for the cyclotron frequency determination. The reason is that Coulomb interaction between unlike ion species can cause undesired frequency shifts [6]. Therefore, during the measurements, care was taken to remove isobaric contaminations present in the ISOLDE ion beam. For the measurements on rubidium isotopes the ionizer of the ISOLDE ion source was kept at a low temperature in order to suppress the ionization of strontium atoms. In the case of the measurements on strontium isotopes their longer half-life compared to the rubidium isobars was used to let the latter decay away before the actual measurement was started. Furthermore, residual contaminating ions were removed in the precision trap by strong dipole excitation of their cyclotron frequency. In order to trace the presence of unremoved isobars and of long-lived isomers all resonances were carefully checked with respect to their expected linewidth and lineshape. The few cases which raised doubts about the purity of the ion sample showed up were excluded from our results.

In the case of ^{90}Rb very reliable shapes of the cyclotron resonances were observed but a first on-line analysis resulted in a mass of the nucleus which was 95(13) keV less bound than the known literature value. Half-life measurements proved that the measured species corresponded more closely to the isomer ^{90m}Rb (E=106.9 keV, $T_{1/2}$=258 s) than to the ground state ($T_{1/2}$=153 s). Therefore the measured mass was assigned to this isomeric state.

3.2 Test of mass-dependent systematic errors

A possible source for systematic errors in mass measurement with Penning traps are calibration errors. They can for example be due to imperfections of the Penning trap. Since the data reported here are the first measurements of ISOLTRAP after its move to the PS-Booster ISOLDE a calibration check was performed. For this the resonance frequencies of ^{133}Cs and ^{226}Ra were measured during the run. These nuclides were chosen for two reasons: their masses were quite precisely known and measuring the frequency ratio of two nuclides with a large mass difference represents a sensitive test for mass dependent systematic errors. The mass excess of ^{133}Cs derived from the measured frequencies is $ME^{exp}(^{133}Cs)$ = –88085(13) keV to be compared to the value given in the 1995 mass tables $ME^{AME95}(^{133}Cs)$ = –88075.7(3.0) keV or to the recent high precision value based on a Penning trap measurement at

MIT [7], $\mathrm{ME}^{\mathrm{MIT}}(^{133}\mathrm{Cs})$ = –88070.960(0.022) keV [6]. For a mass difference of 93 u, this result corresponds to a deviation of $\Delta m/m \sim 7 \cdot 10^{-8}$, respectively $\Delta m/m \sim 1 \cdot 10^{-7}$. The largest mass difference in the present mass doublets is only 10 u. Hence, a possible mass-dependent systematic error is well within the estimate of $\delta m/m = 1 \cdot 10^{-7}$ for the overall systematic error of ISOLTRAP. Recent systematic measurements with ISOLTRAP on carbon-cluster ions of largely different size confirm these findings [9].

4 Frequency ratios

Table 1 compiles the ratios of the cyclotron frequencies of the investigated and the reference nuclides. The two uncertainty values given are the statistical error and the total error which includes the estimated systematic error of $1 \cdot 10^{-7}$. It can be seen that the statistical uncertainty is close to $1 \cdot 10^{-8}$, therefore the total error is fully determined by the conservative estimate for the systematic error.

5 Mass values

From the frequency ratios given in Table 1, mass values given in column 3 of Table 2 are readily derived by use of eq. (2), using for the reference ^{85}Rb a mass value based on work at MIT [7]. The MIT result expressed as mass excess is $\mathrm{ME}^{\mathrm{MIT}}(^{85}\mathrm{Rb}) = -82167.33(0.01)\,\mathrm{keV}$ (see footnote on ^{133}Cs). This value agrees very well but is more than an order of magnitude more precise than the 1995 mass table value $\mathrm{ME}^{\mathrm{AME95}}(^{85}\mathrm{Rb}) = -82167.7(2.3)\,\mathrm{keV}$ [10]. In the cases of ^{90}Rb the ground state mass has been derived from the measured frequency ratio for ^{90m}Rb and the known excitation energy of this isomer. For comparison the results of the 1993 mass evaluation [11], in which Penning trap data had not yet been available, are given in column 4 of Table 2. The ISOLTRAP data have been used for the most recent mass update of 1995 [10]. These values are listed in column 5.

It is interesting to compare the directly obtained Penning trap mass values with the 1993 evaluation [11]. The upper parts of Figures 2 and 3 show the difference between the 1993 mass values (points with error bars) and the Penning trap data (zero line with error band) for the rubidium and strontium isotopic chains. In 8 of the 12 cases the 1993 adjusted values are not within

[6] This value has been obtained by a mass evaluation where the result from [7] was included. A energy mass conversion factor of 931494.0090(0.0071) keV/u was used [8].

the present experimental uncertainties. The origin of these contradictions is expected to be older erroneous input data of the 1993 mass table. This was investigated as discussed in the next section.

5.1 Mass adjustments with the ISOLTRAP data

As in previous works the ISOLTRAP data were used as input data for a global mass adjustments similar to the one performed by Audi and Wapstra for preparing the mass tables. None of the data presented here were included in the 1993 mass evaluation, all of them have been taken into account in the 1995 update.

In order to be used as input data for the mass tables the measured frequency ratios have to be recast into linear mass relations following a procedure outlined in detail in [12]. The result are mass differences as given in Table 3. The adjusted values given in column 3 are identical to those in the 1995 mass evaluation. The only exception is the value for ^{90}Rb. At the time of completion of the 1995 mass adjustment it was still not excluded that in the case of ^{90m}Rb measurement a weak ground state mixture was present. Therefore, the error was increased from 9 keV to 14 keV. In the meantime the off-line analysis was carefully repeated and gave no indication for any contamination. For this reason, the original error of 9 keV should be used in any future mass evaluation.

In the course of a first intermediate mass adjustment with the ISOLTRAP data a number of discrepancies were found. All publications and data, which contribute to the masses of one of the concerned nuclei were carefully studied. Several doubtful data were identified [13] and corrected in collaboration with the authors of the mass tables:

- The Q_β-values [14,15] of the ^{91}Rb decay were re-evaluated. It was taken into account that the measured end point energy does not correspond to a pure feeding of the ground state of ^{91}Sr, but to a mixed decay [16] of less than 6% to the ground state and 24% to the 93.64 keV level. Our measurement of ^{91}Sr and ^{91}Rb masses allows to derive the average mixture of the lines in the β-decay to ^{91}Sr to be at an excitation energy of 39 ± 11 keV in fair agreement with the reported feedings in [16].
- The Q_β-values for the ^{94}Rb decay in [17] and [18] was based on an incomplete decay scheme. In the case of [17] it was possible to correct the Q_β-value from 10304 keV to 10322 keV [13]. In [18] however no correction could be applied. In this case an additional error of 100 keV was added to the original one, resulting in a new total error of 140 keV.
- Due to inconsistencies in error handling a number of uncertainties of Q_β-

values had to be corrected in [17]. The applied corrections are summarized in table 4.

- The consistency factor CF [11] of mass doublet measurements [19] performed with a prism mass spectrometer in St. Petersburg was increased from 1.5 to 4. Hints were found [10,13] that these measurements suffer from a calibration error.
- The consistency factor CF of mass triplet measurements performed at ISOLDE [20,21] was increased from 1.5 to 2.5. The correlation in the systematic errors was underestimated for long chains of measurements.

The result of the 1995 mass evaluation which takes these corrections into account is given in column 5 of table 2 and visualized in the lower parts of Figures 2 and 3. The consistency of the data has been significantly improved but is still not perfect. This indicates that other erroneous data have still to be identified and that more precise measurements in this region are highly desirable.

6 Conclusions

The measurements reported here were the first measurements carried out with the ISOLTRAP spectrometer at the new ISOLDE facility after its move to the PS-Booster accelerator at CERN. Due to their high accuracy of better than 10 keV, these results have considerably improved our knowledge of masses of neutron-rich nuclides in the midshell region between Z=28 and Z=50. In the meantime many more measurements have been carried out with ISOLTRAP [22,12,23–27] and the spectrometer has been subject of improvements with respect to its applicability and sensitivity [2,3]. A new highlight of measurements in the rubidium isotopic chain was the determination of the mass of ^{74}Rb which has a halflife of only $T_{1/2} = 65$ ms [27]. Furthermore, the masses of ^{76,77}Sr were determined recently [28]. Including the earlier measurements on proton-rich rubidium isotopes [29], ISOLTRAP has determined accurate masses in two long isotopic chains, reaching from ^{74}Rb to ^{94}Rb and from ^{76}Sr to ^{95}Sr.

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Figure captions

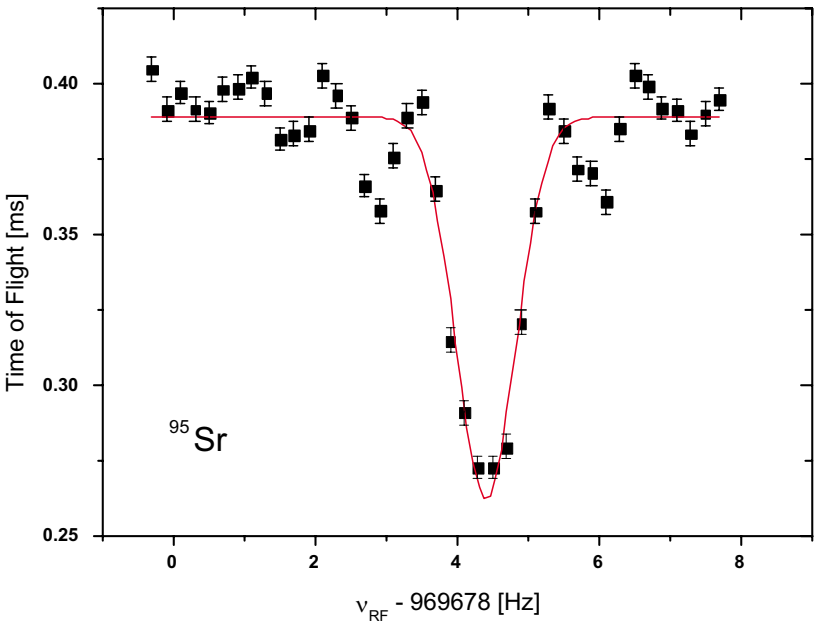

Figure 1: Cyclotron resonance curve for ^{95}Sr. The solid curve shows a fit by a Gaussian (see text).

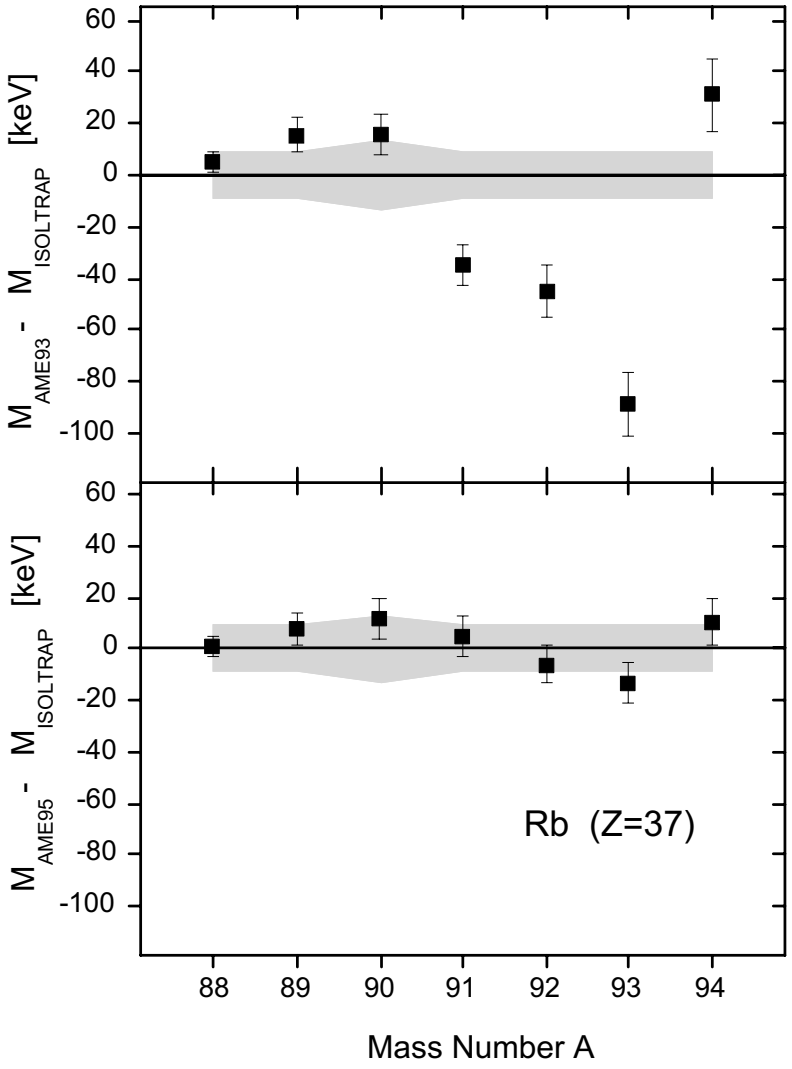

Figure 2: Difference between the mass values of the 1993 (top) and the 1995 (bottom) mass adjustments (data points with errors bars) and the ISOLTRAP results (shaded error band) for rubidium isotopes.

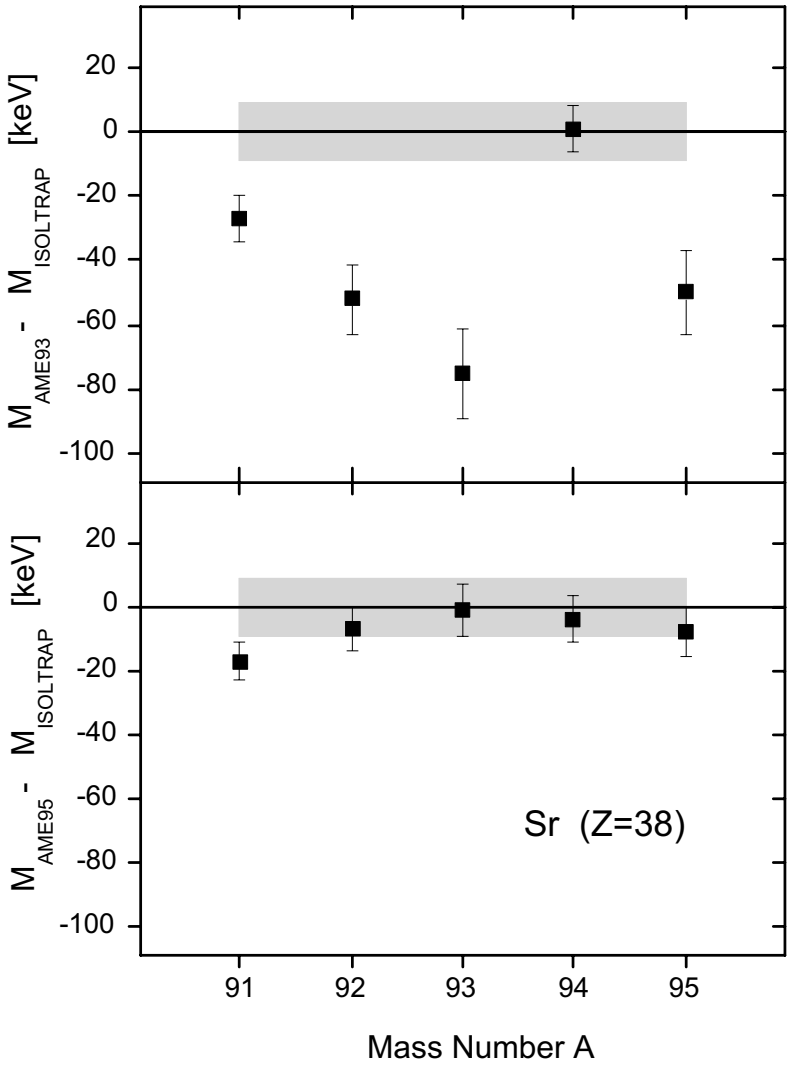

Figure 3: Difference between the mass values of the 1993 (top) and the 1995 (bottom) mass adjustments (data points with errors bars) and the ISOLTRAP results (shaded error band) for strontium isotopes.

Table 1: Cyclotron frequency ratios $r = \nu_{ref}/\nu$ for Rb and Sr ions relative to the cyclotron frequency of ^{85}Rb. The investigated nuclides are listed in the first column. In column 2 the measured frequency ratios are given. The uncertainties in column 3 are the statistical ones. The total error given in column 4 includes the maximum estimated systematic error of $1 \cdot 10^{-7}$.

Nuclide	*Frequency Ratio* $r = \nu_{ref}/\nu$		
^{88}Rb	1.035 325 454	(10)	(104)
^{89}Rb	1.047 113 687	(16)	(105)
^{90m}Rb	1.058 921 783	(14)	(107)
^{91}Rb	1.070 717 843	(34)	(112)
^{92}Rb	1.082 532 565	(19)	(110)
^{93}Rb	1.094 336 825	(25)	(112)
^{94}Rb	1.106 165 066	(19)	(112)
^{91}Sr	1.070 643 641	(31)	(112)
^{92}Sr	1.082 430 179	(13)	(109)
^{93}Sr	1.094 242 341	(27)	(113)
^{94}Sr	1.106 035 126	(18)	(112)
^{95}Sr	1.117 859 282	(14)	(113)

Table 2: Mass excesses as determined by ISOLTRAP. Column 3 lists mass values obtained by combining the frequency ratios given in Table 1 with the value ME^{MIT} = –82167.33(0.01) keV (based on [7]) for the mass excess of the reference ^{85}Rb. In the case of ^{90}Rb where the isomer was studied, the known isomeric energy of 106.90 keV was subtracted. The data are compared to the mass values listed in the 1993 mass tables (without any Penning trap data) [11] and those of 1995 (with all Penning trap data) [10]. The half-lives are taken from Ref. [30]. The last column gives the sum of influences *IF* of the Penning trap measurements in the determination of the mass value of the corresponding nuclide in the 1995 atomic mass evaluation [10]. It should be noted that in a new evaluation that includes the MIT result for ^{85}Rb, the influence of ISOLTRAP for this isotope would no longer be significant.

Isotope	$T_{1/2}$	Mass Excess [keV]				IF [%]
	(from [30])	this exp.	1993 adj.[11]		1995 adj.[10]	[10]
^{88}Rb	17.78 m	–82607 (8)	–82602	(4)	–82606 (4)	21
^{89}Rb	15.15 m	–81719 (8)	–81703	(7)	–81711 (6)	41
^{90}Rb	158 s	–79366 (8)	–79351	(8)	–79355 (8)	35
^{91}Rb	58.4 s	–77752 (9)	–77788	(8)	–77748 (8)	70
^{92}Rb	4.492 s	–74769 (9)	–74814	(10)	–74775 (7)	56
^{93}Rb	5.84 s	–72613 (9)	–72702	(12)	–72626 (8)	61
^{94}Rb	2.702 s	–68561 (9)	–68530	(14)	–68551 (9)	77
^{91}Sr	9.63 h	–83621 (9)	–83649	(7)	–83639 (6)	44
^{92}Sr	2.71 h	–82867 (9)	–82920	(11)	–82875 (7)	58
^{93}Sr	7.423 m	–80086 (9)	–80162	(14)	–80088 (8)	64
^{94}Sr	75.3 s	–78838 (9)	–78837	(7)	–78842 (7)	58
^{95}Sr	23.90 s	–75109 (9)	–75159	(13)	–75117 (8)	62
^{85}Rb	stable	reference	–82164.8	(2.5)	–82167.7 (2.3)	15

Table 3: Results of the conversion from cyclotron frequency ratios given in Table 1 into linear relations for mass adjustment. The equations representing the ISOLTRAP data are listed in the first column, where the symbol for the nuclides refer to their mass. The second column gives the experimental values for these relations in μu. Their adjusted values in the least squares fit in the 1995 Atomic Mass Evaluation [10] are given in the third column. In column 4 the relative deviations v/s between measured and adjusted values are given as their differences v divided by the uncertainty s of the experimental value. The last column gives the significance S [31] of the present data in the adjustment. The largest part of this significance influences the investigated nuclide; the rest, summing up to 15%, do influence the final value of the reference mass ^{85}Rb (see Table 2). The distribution of the v/s is a normal one with 2 out of 12 beyond 1σ and only one beyond 2σ.

linear relation	exp. value [μu]	adj. value [μu]	v/s	S
^{88}Rb$-1.035\cdot{}^{85}$Rb	2615 (9)	2617 (4)	0.2	24%
^{89}Rb$-1.047\cdot{}^{85}$Rb	4628 (9)	4636 (6)	0.9	43%
^{90}Rb$-1.059\cdot{}^{85}$Rb	8211 (9)	8224 (8)[a]	0.9[a]	37%[a]
^{91}Rb$-1.071\cdot{}^{85}$Rb	11003 (10)	11008 (8)	0.5	71%
^{92}Rb$-1.082\cdot{}^{85}$Rb	15176 (9)	15169 (7)	-0.7	57%
^{93}Rb$-1.094\cdot{}^{85}$Rb	18549 (10)	18535 (8)	-1.4	62%
^{94}Rb$-1.106\cdot{}^{85}$Rb	23958 (10)	23968 (9)	1.0	78%
^{91}Sr $-1.071\cdot{}^{85}$Rb	4702 (9)	4683 (6)	-2.1	47%
^{92}Sr $-1.082\cdot{}^{85}$Rb	6482 (9)	6474 (7)	-0.9	59%
^{93}Sr $-1.094\cdot{}^{85}$Rb	10526 (10)	10525 (8)	-0.1	65%
^{94}Sr $-1.106\cdot{}^{85}$Rb	12924 (10)	12921 (8)	-0.3	60%
^{95}Sr $-1.118\cdot{}^{85}$Rb	17987 (10)	17978 (8)	-0.9	65%

a)Adjusted value, significance and deviation v/s for ^{90}Rb as in the 1995 adjustment where an uncertainty of 14 keV was considered. With this uncertainty now reduced to 9 keV (see section 5.1), the ^{90}Rb significance is strongly increased, and its v/s decreased.

Table 4: Corrections applied to Q_β data in [13] and in the 1995 mass adjustment. Column 1 give the concerned β-decays. The published Q-values with their former uncertainties are listed in column 2. The new assigned values and uncertainties are given in column 3 (bold type highlights changes).

reaction	Q_β (keV)	Ref.	corrected Q_β (keV)
$^{88}Rb(\beta^-)^{88}Sr$	5318(4)	[17]	5318(**9**)
$^{89}Rb(\beta^-)^{89}Sr$	4510(8)	[17]	4510(**9**)
$^{91}Rb(\beta^-)^{91}Sr$	5857(8)	[17]	5857(**13**)
$^{91}Sr(\beta^-)^{91}Y$	2704(3)	[17]	2704(**8**)
$^{94}Rb(\beta^-)^{94}Sr$	10304(30)	[17]	**10322(100)**
$^{94}Rb(\beta^-)^{94}Sr$	10353(100)	[18]	10353(**140**)
$^{94}Sr(\beta^-)^{94}Y$	3512(5)	[17]	3512(**10**)
$^{94}Y(\beta^-)^{94}Zr$	4920(5)	[17]	4920(**9**)
$^{95}Y(\beta^-)^{95}Zr$	4445(5)	[17]	4445(**9**)
$^{96}Sr(\beta^-)^{96}Y$	5413(20)	[17]	5413(**22**)