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High precision mass measurements of intermediate-mass neutron-deficient nuclei via MRTOF-MS

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Precision mass measurements of ^{63}Cu , $^{64-66}\text{Zn}$, $^{65-67}\text{Ga}$, $^{65-67}\text{Ge}$, ^{67}As , $^{79,81}\text{Br}$, ^{79}Kr , $^{80,81}\text{Rb}$, and $^{79,80}\text{Sr}$ were performed with a multireflection time-of-flight mass spectrograph. The masses of these nuclides were determined by the single reference method using isobaric references. In order to obtain precise results, time-of-flight drift compensations were performed and a phenomenological fit function was employed. Consequently, in the case of ^{65}Ga , a mass uncertainty of 2.1 keV, corresponding to a relative precision of $\delta m/m = 3.5 \times 10^{-8}$, was obtained and the mass value is in excellent agreement with the 2016 Atomic Mass Evaluation.

KEYWORDS: Mass measurement, MRTOF, rp-process

1. Introduction

Nuclear masses of proton-rich nuclei along the $N = Z$ line are crucial in determining the reaction flow of the rapid proton-capture (rp -) process. Mass uncertainties need to be less than roughly 10 keV to significantly reduce the uncertainties of rp -process calculations [1]. Half-lives of key nuclei in the rp -process span from $\sim 10 - 100$ ms. Multireflection time-of-flight mass spectrograph (MRTOF-MS) [2] has an advantage in the mass measurements of these short-lived nuclei since measurement time can be less than 10 ms. However there are no on-line MRTOF-MS measurements satisfying the required precision so far. In the present study, we demonstrate high-precision MRTOF-MS measurements of the intermediate-mass neutron-deficient nuclei.

2. Experiment

The experiment was performed by using the superheavy element mass facility at RIKEN [3], consisting of the MRTOF-MS and the gas-filled recoil ion separator GARIS-II [4], which has been developed to enable mass measurements of superheavy elements (SHE) produced at RIKEN. A beam of $^{36}\text{Ar}^{10+}$ at 3.30 MeV/nucleon with a maximum intensity of 3 pμA, was provided by the RIKEN linear accelerator RILAC, and used to produce the intermediate-mass, neutron-deficient nuclei via the fusion-evaporation reactions of $^{\text{nat}}\text{S}(^{36}\text{Ar},\text{X})$ and $^{\text{nat}}\text{Ti}(^{36}\text{Ar},\text{X})$. Further details of the experimental setup and conditions are provided in [5].

3. Analysis

3.1 Single reference method

The observed time-of-flight (TOF) value t_{obs} of an ion which travels in the MRTOF-MS device with ℓ laps is given by

$$t_{\text{obs}} = (a + b \cdot \ell) \sqrt{m} + t_0, \quad (1)$$

where m is the ion mass, t_0 represents an additional systematic offset, and a and b are constants related to the single non-reflection flight path and the length of flight path between consecutive reflections from one of the mirrors, respectively. The time offset t_0 originates from the delay between ion ejection and the TDC start time. It has been directly measured to be 40(10) ns.

The single reference method [6] was used to determine the masses of observed nuclei. If t_{X} and t_{ref} are the measured TOF of nuclide X and the reference ion, respectively, then we can define a parameter ρ^2 :

$$\rho^2 = \frac{m_{\text{X}}}{m_{\text{ref}}} = \left(\frac{t_{\text{X}} - t_0}{t_{\text{ref}} - t_0} \right)^2, \quad (2)$$

where m_{X} and m_{ref} are the masses of nuclide X and the reference ion, respectively. In the present measurements, only singly-charged ions were measured. Thus, the atomic mass of nuclide X, M_{X} , is given by

$$M_{\text{X}} = \rho^2 (M_{\text{ref}} - m_{\text{e}}) + m_{\text{e}}, \quad (3)$$

where M_{ref} and m_{e} are the atomic mass of the reference nuclide and the electron rest mass, respectively. The 2016 Atomic Mass Evaluation (AME16) values [7, 8] were used for the mass references.

Excluding low statistics species, the least squares fitting routine of the ROOT package [9] was used to determine ρ . For the low statistics cases the maximum likelihood method was used.

3.2 Drift compensation

Under the actual experimental conditions, TOF values varied as a function of time due to thermal expansion of the MRTOF-MS device and minor instabilities in the HV power supply system for the mirror electrodes [2]. These TOF drifts could be compensated by use of isobaric reference species. Before performing such a correction, we first calculated the ‘‘standard TOF’’ of the reference. Equation 1 can be rewritten as a sum of the ℓ dependent and independent terms:

$$t_{\text{obs}} = \eta \cdot \ell + \zeta. \quad (4)$$

Parameters η and ζ were determined by a peak fit of raw TOF data of the isobaric reference with different ℓ , and the standard TOF for each ℓ were deduced. The raw data were then divided by equal sweeps (measurement times) into i subsets. For ions in each subset i the corrected TOF $t_{\text{corr},i}$ was calculated with the following relation:

$$t_{\text{corr},i} = t_{\text{raw},i} \left(\frac{t_{\text{std}}}{t_{\text{std}} + t_{\text{drift},i}} \right), \quad (5)$$

where $t_{\text{raw},i}$ is the uncorrected TOF of each ion in subset i , while t_{std} and $t_{\text{drift},i}$ are the standard TOF for the isobaric reference ion and the deviation of the peak center of the isobaric reference species within the i^{th} subset, respectively. After applying corrections to each subset, the data were recombined for precision peak fitting. This correction method was applied to all the data presented here. The peak width improvements by drift compensation in the present experiment are discussed in [5].

3.3 Fit function

A phenomenological fit function was developed through the study of high-statistics offline measurements of $^{85}\text{Rb}^+$ ions [5]. A typical TOF spectrum of $^{85}\text{Rb}^+$ ions is shown in Fig. 1. In order to reproduce the shape of main peak, we employed the function:

$$f_p(\tau) = \begin{cases} \mu \exp[-\tau^2/\nu] & (\text{for } \tau < t_{s1}) \\ \xi \exp[\eta/\tau^2] & (\text{for } t_{s1} \leq \tau < t_{s2}) \\ (\kappa/\sigma) \exp[-\tau^2/(2\sigma^2)] & (\text{for } t_{s2} \leq \tau < t_{s3}) \\ (\kappa/\sigma) \exp[t_{s3}(t_{s3} - 2\tau)/(2\sigma^2)] & (\text{for } \tau \geq t_{s3}), \end{cases} \quad (6)$$

where the characteristic times t_{s_i} indicate the available range of each sub-function. The variable τ is defined as: $\tau \equiv t - t_c$, where t_c is the peak center. Thus, f_p has seven independent parameters, excluding the characteristic times t_{s_i} . The number of parameters can be analytically reduced to three by imposing the continuity condition at each time t_{s_i} ; the remaining independent parameters are then t_c, κ , and σ .

In addition to the primary peaks, we observed ‘‘bump’’ structures neighboring the intense peaks in sufficiently high-statistics TOF spectra. These bumps maintain a constant intensity and position relative to the primary peak and are deduced to result from the following process: secondary electrons are emitted from the surface of the MCP ion detector, they are accelerated to nearby surfaces and produce tertiary ions which accelerate back to the MCP. Hence, these bumps are not actual events and depend only on the intensities of parent peaks. The bump shapes are modeled with the following Gaussian function:

$$f_b(\tau) = (\kappa_b/\sigma_b) \exp[-(\tau - t_b)^2/(2\sigma_b^2)]. \quad (7)$$

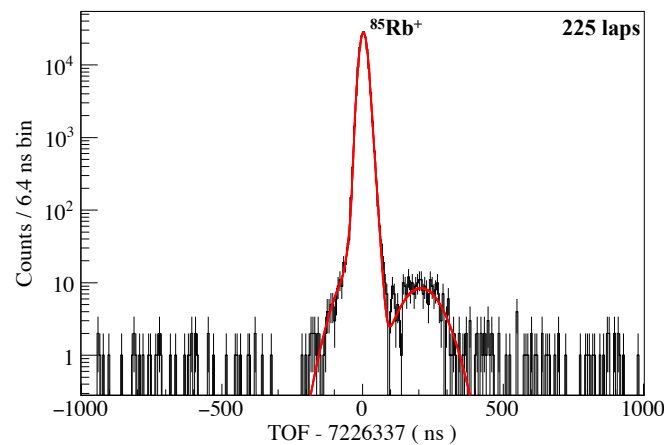


Fig. 1. Typical time-of-flight spectrum of $^{85}\text{Rb}^+$ ions. Such spectra were used to determine the fit function, indicated by a red line, based on an exponential-Gaussian hybrid function.

All independent parameters of the fitting function including the switching times were obtained as an average of the off-line measurements of the $^{85}\text{Rb}^+$ ions with two different number of laps. In the fitting algorithm, t_{s1} and t_{s2} were determined by scaling the σ parameter of mass reference peaks relative to that of $^{85}\text{Rb}^+$:

$$t_{si} = t_{si,85\text{Rb}} \times (\sigma/\sigma_{85\text{Rb}}) \quad (i = 1, 2). \quad (8)$$

The t_{s3} values were determined as independent parameters from fits to the mass reference nuclide peaks, then subsequently fixed for each isobaric species of interest. The bump height parameter κ_b was calculated assuming a constant relative intensity,

$$\kappa_b = \kappa_{b,85\text{Rb}} \times (\kappa/\kappa_{85\text{Rb}}). \quad (9)$$

The other two bump parameters, σ_b and t_b , were fixed using the $^{85}\text{Rb}^+$ fit results.

For the species of interest, the only free parameters in the fitting function are the peak center t_c and the peak height κ since the σ parameters were determined from the mass reference nuclide peaks. In the fitting algorithm, to enhance the mass precision, the τ parameter was treated as a function of t , t_{ref} , and ρ , where ρ is the TOF ratio from Eq. 2. Then the fitting function F for N peaks is described by

$$F(t, t_{\text{ref}}, \rho_1, \dots, \rho_N, \kappa_1, \dots, \kappa_N) = \sum_{i=1}^N \{f_p(t, t_{\text{ref}}, \rho_i, \kappa_i) + f_b(t, t_{\text{ref}}, \rho_i, \kappa_i)\}. \quad (10)$$

If the i^{th} peak corresponds to the mass reference, ρ_i is always unity; the total number of fitting parameters is $2 \times N$.

3.4 Error evaluation

The systematic mass ratio error $\delta(\rho^2)_{\text{sys}}$ consists of terms originating from the drift compensation and the constant time offset t_0 :

$$\delta(\rho^2)_{\text{sys}} = 2\rho \sqrt{\left(\frac{\partial\rho}{\partial t_{\text{drift}}}\right)^2 \delta t_{\text{drift}}^2 + \left(\frac{\partial\rho}{\partial t_0}\right)^2 \delta t_0^2}. \quad (11)$$

The TOFs of both nuclide X and the mass reference nuclide are compensated following Eq. 5 and with the same reference TOF data set $\{t_{\text{drift},i}\}$ as used to evaluate ρ values. The uncertainties of drift compensation of each TOF value must be treated in the same manner. Thus the influence of such uncertainties on δt_{drift} can be found to be strongly correlated: a coefficient of δt_{drift} can be written as

$$\frac{\partial\rho}{\partial t_{\text{drift}}} = \left(\frac{\partial\rho}{\partial t_X}\right) \frac{dt_X}{dt_{\text{drift}}} + \left(\frac{\partial\rho}{\partial t_{\text{ref}}}\right) \frac{dt_{\text{ref}}}{dt_{\text{drift}}}. \quad (12)$$

By substituting the above equation in Eq. 11, one can calculate

$$\frac{\delta(\rho^2)_{\text{sys}}}{\rho^2} \approx 2 \sqrt{\left(1 - \frac{1}{\rho}\right)^2 \left(\frac{t_0}{t_{\text{ref}}}\right)^2 \left[\left(\frac{\delta t_0}{t_0}\right)^2 + \left(\frac{\delta t_{\text{drift}}}{t_{\text{ref}}}\right)^2\right]}. \quad (13)$$

If we introduce the variable $\Delta m = m_X - m_{\text{ref}}$, the TOF ratio ρ can be rewritten as a function of Δm : $\rho = \sqrt{1 + \Delta m/m_{\text{ref}}}$. Equation 13 is plotted in Fig. 2 and the systematic error could be suppressed down to $(\delta\rho^2)_{\text{sys}}/\rho^2 \leq 10^{-9}$ (green area of Fig. 2) by employing an isobaric reference. This value is negligibly small compared to the fitting error, thus the uncertainties of the mass values are dominated by the fitting errors and the mass uncertainties of the reference nuclides.

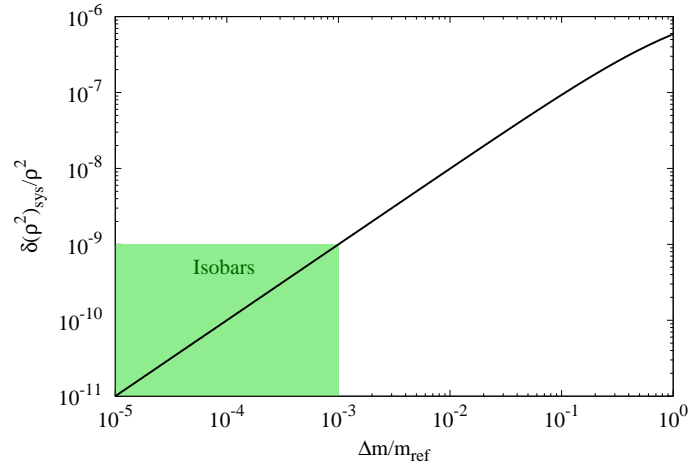


Fig. 2. The systematic error as a function of relative mass difference. In this figure, $\delta t_0 = 10$ ns and $\delta t_{\text{drift}} = 0.5$ ns are assumed. See the texts for details.

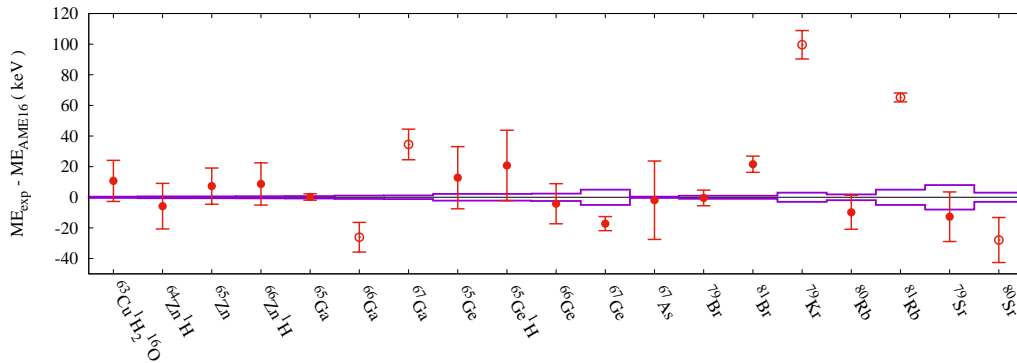


Fig. 3. Differences between the present measurement results and the AME16 values. Purple lines represent error band of the AME16 values. The open symbols indicate data derived from spectral peaks insufficiently separated from adjacent peaks.

4. Results

The results from [5] are summarized again in Fig 3. There are some inconsistencies with the 2016 Atomic Mass Evaluation (AME16) values. Our previous framework further includes a careful discussion of all inconsistencies with the AME16 values seen in Fig 3, and two reliability requirements were imposed on the mass values: (i) there are no contaminations from unresolved isomers, (ii) there are no undue influences of intense neighboring peaks [5]. Among the masses that were inconsistent with AME16, two were found to meet the reliability criteria, and we propose new mass excess values: $ME(^{67}\text{Ge}) = -62675.2(46)$ keV and $ME(^{81}\text{Br}) = -77955.4(53)$ keV. These mass values were previously deduced through indirect measurements [10–12].

The relative mass precisions in the present study span the range of $\delta m/m = 4.1 \times 10^{-7}$ to 3.5×10^{-8} . In the most precise measurement, that of ^{65}Ga , a mass uncertainty of 2.1 keV was obtained. The mass reference of the $A/q = 65$ series is ^{65}Cu ($\delta m = 0.6$ keV in AME16) and a total event number of ^{65}Ga is 2×10^4 in all three measurements with different number of laps ℓ . This result shows that mass measurements satisfying the requirement to impact on rp -process calculations, $\delta m \lesssim 10$ keV, can be

achieved with the MRTOF-MS, given sufficient statistics.

In Section 3.4 the systematic errors originating from the drift compensation and the constant time offset t_0 were investigated. The correctness of our result from ^{65}Ga indicates that these systematic errors are negligibly small, at the level of 10^{-8} , by employing the isobaric reference.

5. Summary

Precision mass measurements of ^{63}Cu , $^{64-66}\text{Zn}$, $^{65-67}\text{Ga}$, $^{65-67}\text{Ge}$, ^{67}As , $^{79,81}\text{Br}$, ^{79}Kr , $^{80,81}\text{Rb}$, and $^{79,80}\text{Sr}$ performed by using an MRTOF-MS coupled with GARIS-II have been reported in [5]. The masses of these nuclides have been determined by the single reference method using isobaric references. In order to obtain precise results, compensation of TOF drifts originating from the instabilities in the experimental system were performed. A phenomenological fit function was constructed from high-statistics off-line data of ^{85}Rb and used to reproduce complex shape TOF peaks in low statistics region. Consequently, for ^{65}Ga , a mass uncertainty of 2.1 keV, corresponding to a relative precision of $\delta m/m = 3.5 \times 10^{-8}$, was obtained and the mass value is in excellent agreement with the AME16 value.

The present works extends the study while focusing on the relative mass difference dependency of systematic error resulting from both of the constant time offset and the drift compensation. The result of ^{65}Ga supports the our evaluation to the systematic error: when employing the isobaric mass reference, its contribution will be suppressed to negligibly small at the level of 10^{-8} .

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References

- [1] H. Schatz, *Int. J. Mass Spectrom.* **251**, 293 (2006) & **349–350**, 181 (2013).
- [2] P. Schury, M. Wada, Y. Ito, F. Arai, S. Naimi, T. Sonoda, H. Wollnik, V.A. Shchepunov, C. Smorra, and C. Yuan, *Nucl. Instrum. Methods Phys. Res., Sect. B* **335**, 39 (2014).
- [3] P. Schury, M. Wada, Y. Ito, D. Kaji, F. Arai, M. MacCormick, I. Murray, H. Haba, S. Jeong, S. Kimura, H. Koura, H. Miyatake, K. Morimoto, K. Morita, A. Ozawa, M. Rosenbusch, M. Reponen, P.-A. Söderström, A. Takamine, T. Tanaka, and H. Wollnik, *Phys. Rev. C* **95**, 011305(R) (2017).
- [4] D. Kaji, K. Morimoto, N. Sato, A. Yoneda, and K. Morita, *Nucl. Instrum. Methods Phys. Res., Sect. B* **317**, 311 (2013).
- [5] S. Kimura, Y. Ito, D. Kaji, P. Schury, M. Wada, H. Haba, T. Hashimoto, Y. Hirayama, M. MacCormick, H. Miyatake, J. Y. Moon, K. Morimoto, M. Mukai, I. Murray, A. Ozawa, M. Rosenbusch, H. Schatz, A. Takamine, T. Tanaka, Y. X. Watanabe, and H. Wollnik, *Int. J. Mass Spectrom.* **430** 134 (2018).
- [6] Y. Ito, P. Schury, M. Wada, S. Naimi, T. Sonoda, H. Mita, F. Arai, A. Takamine, K. Okada, A. Ozawa, and H. Wollnik, *Phys. Rev. C* **88**, 011306(R) (2013).
- [7] W. Huang, G. Audi, M. Wang, F. Kondev, S. Naimi, X. Xu, The ame2016 atomic mass evaluation (i). evaluation of input data; and adjustment procedures, *Chin. Phys. C* **41**, 030002 (2017).
- [8] M. Wang, G. Audi, F. Kondev, W. Huang, S. Naimi, X. Xu, The ame2016 atomic mass evaluation (ii). tables, graphs and references, *Chin. Phys. C* **41**, 030003 (2017).
- [9] R. Brun and F. Rademakers, *Nucl. Instrum. Methods Phys. Res., Sect. A* **389**, 81 (1997).
- [10] M.J. Murphy, C.N. Davids, E.B. Norman, R.C. Pardo, *Phys. Rev. C* **17** 1574 (1978).
- [11] A.M. Al-Naser, A.H. Behbehani, P.A. Butler, L.L. Green, A.N. James, C.J. Lister, P.J. Nolan, N.R.F. Rammo, J.F. Sharpey-Schafer, H.M. Sheppard, L.H. Zybert, R. Zybert, *J. Phys. G: Nucl. Phys.* **5** (3) 423 (1979).
- [12] R.C. Waddell, E.N. Jensen, *Phys. Rev.* **102** 816 (1956).