# Nuclear masses weigh heavily with us

As a self-organizing quantum system, the nucleus makes every effort to reduce its mass. As a natural consequence, the nuclear mass contains all the information on mechanisms that stabilize the nucleus. These mechanisms can be traced back using the nuclear mass data.

In 1919, Francis William Aston performed the first successful mass measurement and discovered many isotopes using his mass spectrograph. At that time, the mass resolution was approximately 1%. The subsequent century has witnessed incredible developments in techniques used for measuring nuclear masses. The most established and famous techniques of precision mass measurements are the Penning trap method and the Schottky method using a storage ring.

Unfortunately, neither of the methods can be applied to mass measurements of rare nuclei produced at the RI Beam Factory (RIBF). This is solely because of their short lifetimes of $\gg$1 s. In the long measurement time (>1 s) of the Penning trap and the Schottky methods, the short-lived nuclei decay to their daughters, which prevents us from measuring their masses. The main physics cases in nuclear mass measurements at RIBF involve the magicity and the $r$-process nucleosynthesis, both of which require a mass precision of 100 keV, or $\delta$m/m $\sim 10^{-6}$. Since the statics expected for rare nuclei are as small as $\sim 10^2$, a mass resolving power of $10^5$ or better is mandatory in their studies.

The unprecedentedly fast mass-measurement technique with a mass resolving power as high as $m/\Delta m > 10^5$ has been highly desired, but accomplishing it is a big challenge for nuclear physicists.

Scientists at RIKEN, KEK, and CNS have tackled this problem and have recently succeeded in measuring masses of rare nuclei: The first one is the TOF-$B\rho$ mass measurement using the long OEDO beamline coupled with the SHARAQ spectrometer. The measurement combining a flight length of 105 m (one path) and the high-resolution magnetic system with a controlled correction of higher-order aberrations enables an extremely short measurement time of 0.0005 ms with a reasonable resolving power of $>10^4$ and a high efficiency of $\sim$100%.

The second is an isochronous mass measurement using the Rare RI Ring (R3). R3 is the world's first cyclotron-type storage ring with an individual injection capability. The setup leads to a 0.7-ms measurement time with a resolving power $> 3 \times 10^5$.

The third is the multireflection time-of-flight (MRTOF) mass spectrograph combined with a gas catcher system. This versatile and "portable" device can be placed at any beamline to accept a wide variety of nuclei, whose masses are measured in several tens of ms (including the time needed to stop the ions) with a high resolving power of $10^6$.

The combined use of the abovementioned three techniques with different characteristics, shown in Fig. 1, opens a broad spectrum of research opportunities at RIBF.

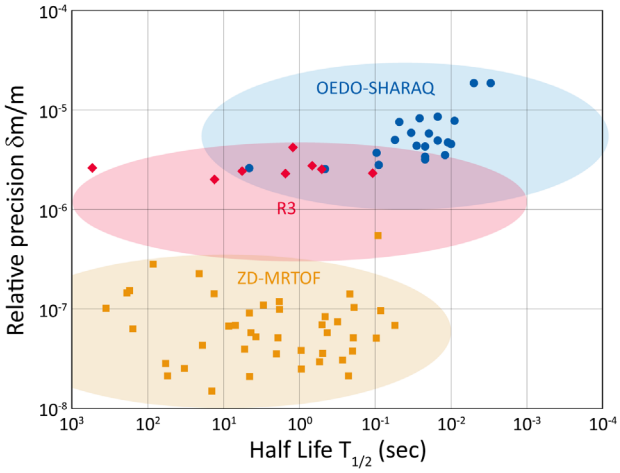


Fig. 1. Mass precision and nuclear half-life regions covered by the three mass measurement methods.

## TOF-$B\rho$ mass measurement using OEDO-SHARAQ

The OEDO-SHARAQ system, initiated collaboratively by CNS, the University, of Tokyo and RIKEN Nishina Center is promoting the program of direct mass measurements of radioactive nuclei far from the $\beta$ stability line. In 2009, the OEDO-SHARAQ program started with the installation of the SHARAQ spectrometer[1] and a dispersion-matched High-Resolution beamline[2] (renamed by OEDO beamline in 2017) into the downstream of the BigRIPS separator[3] at the RIBF facility.

Atomic masses of very short-lived nuclei have been determined so far by various time-of-flight (TOF) methods. In our system, the TOF magnetic-rigidity (TOF-$B\rho$) method, developed at GANIL,[4] has been adopted, and further sophistication in its ion optics and detector performance has been implemented for the measurements. The principle of mass determination using the TOF-$B\rho$ method can be simply expressed by the following relation:

$$\frac{m}{q} = \frac{B\rho}{c}\sqrt{\left(\frac{ct}{L}\right)^2 - 1}, \quad (1)$$

where $m$, $q$, $c$, $t$, and $L$ are the nuclear mass, atomic

charge, speed of light in vacuum, TOF, and the flight-path length, respectively. This relation corresponds to the Lorentz force in a magnetic field, and indicates that the mass-to-charge ratio ($m/q$) is determined by TOF as long as the flight-path length and the magnet field of the beamline are fixed. However, because the flight-path length is indeed dependent on the incident condition of the short-lived nuclei, we perform an event-by-event correction of the flight-path length using the trajectory information obtained at the starting and final foci of the beamline. The essential point that has led to the success of our direct mass measurement program has been the high-resolution performance of the system.

The detector setup for the mass measurements is shown in Fig. 2. The CVD diamond detectors were installed at F3 and S2 to measure the TOF. The CVD diamond detector was developed at CNS to achieve high-resolution timing and high-rate capability.[5] The low-pressure multiwire drift chambers (LP-MWDCs)[6] were used for beam tracking at F3 and S2, and provided beam-trajectory information that is mandatory for accurate corrections of the flight-path lengths. A delay-line parallel-plate avalanche counter (DL-PPAC) installed at S0 measured the $B\rho$ values of the beam particles. The $m/q$ values of the individual short-lived nuclei were determined based on the combination of these measured quantities on an event-by-event basis. Additionally, a silicon solid-state detector (SSD) was used to identify the atomic numbers. The delayed $\gamma$-ray detection system was placed at the end of the beamline to search for the unknown isomeric states in the radioactive nuclei.

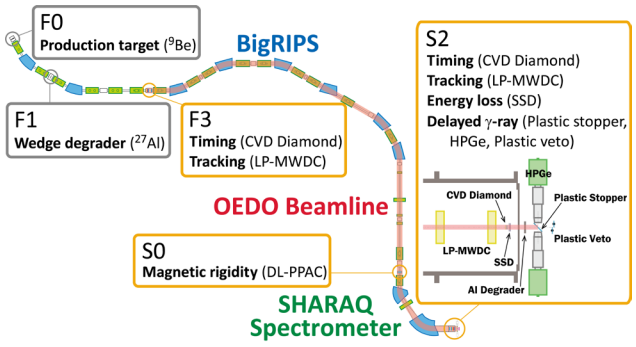


Fig. 2. Experimental detector setup in the OEDO-SHARAQ system for the direct mass measurements.

An advantage of the SHARAQ mass measurements is its accessibility to very short-lived isotopes, whose lifetimes are less than 1 ms, with its high transmission efficiency, because the TOF of the system is as short as ~500 ns. With this capability, the system is expected to enable the measurement of nuclear masses on the dripline.

We have successfully measured the atomic masses of very neutron-rich Ca and Ti isotopes towards the dripline by a single setting of the BigRIPS separator and the SHARAQ spectrometer. These new masses are covered with the region connecting the emergence of magicities at $N = 32$ and 34 and the island-of-inversion at $N = 40$. The work has clearly exhibited the sudden emergence of magicity in $^{54}$Ca.[7] Moreover, the results for Ti isotopes reveal an onset of the Jahn-Teller stability around $^{62}$Ti,[8] indicating shell quenching at $N = 40$ based on the comparison with the theoretical mass predictions.[9]

These experiments have demonstrated that our experimental technique has capabilities to efficiently pin down the mass irregularities and provide essential information on its onset mechanism based on mass evolution. Recently, the first TOF-$B\rho$ mass measurements after the construction of the OEDO system were performed to access the mass region of two-proton radioactivity.[10] The TOF-$B\rho$ method excels in extending the mass data toward the nuclear drip lines. We will extend our challenge toward the $^{78}$Ni and $^{100}$Sn regions in the near future.

## Isochronous mass measurement with the rare RI ring

In the decade after the R3 was built, we have successfully conducted its commissioning using a primary beam, validation of the mass measurement method using unstable nuclei with known masses, and precise mass measurements. Recently, the mass of $^{123}$Pd, one of the nuclei near the second peak of the $r$-process, was successfully determined.[11] The impact of the measured mass on the heavy element synthesis was investigated by inputting the new mass value into the $r$-process network simulations from which its effect on the probability of neutron capture and neutron emission after beta decay were evaluated. The compositions observed in the solar system can be reproduced with the new mass value.

Figure 3 illustrates the mass measurement scheme of the R3. The mass measurements for such nuclei have been achieved by our bold move to connect a cyclotron and a storage ring, which have been known to be essentially incompatible, taking the advantage of the "long" fragment separator. The key point of this scheme is to know, prior to injection, that the nucleus of interest is produced in the first section of the BigRIPS and to inject it into R3. The masses are then determined event-by-event by performing isochronous mass spectrometry (IMS).[12,13]

In RIBF, various unstable nuclei are produced by the in-flight fission of the uranium beam or the projectile fragmentation of heavy-ion beams such as xenon, krypton, *etc.* Their extremely low production rate makes the timing of production completely random.

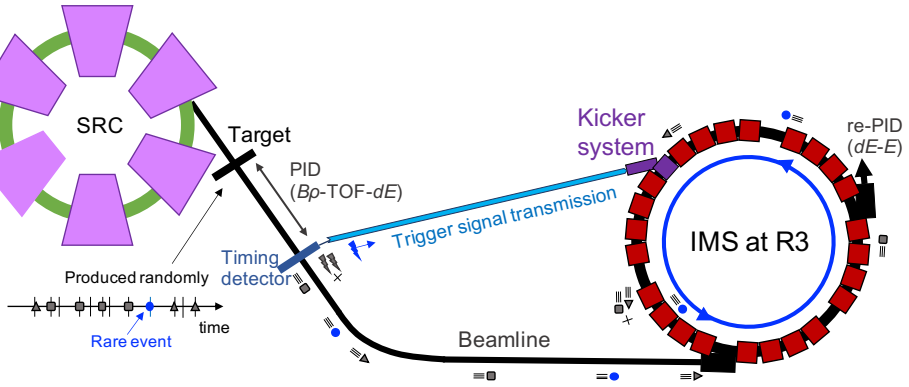


Fig. 3. Conceptual design and method for measuring the mass of short-lived rare nuclei using R3.

The species of the produced nuclei can be identified on an event-by-event basis using the $B\rho$-TOF-$dE$ method in the F0–F3 section of BigRIPS.[14] Subsequently, the identified isotope passes though the timing detector placed at F3. Using the timing and energy-loss information from the detector, the signal to trigger the R3 kicker system is generated only for the isotopes of interest. Synchronization of the kicker excitation to the arrival of the isotope beam is the key technology enabling the unique combination of a cyclotron and a storage ring. It should also be noted that the RF signal from the accelerator is utilized to efficiently reduce the trigger rate by filtering the signals corresponding to unnecessary events. Using this setup, we have realized the isotope-selectable self-triggered injection, called the individual injection, for the first time.

The injected isotope is circulated in R3 for certain turns (typically ∼2000) and extracted after ∼1 ms. The revolution time is obtained by dividing the flight-time between injection and extraction by the number of turns. Further, the extracted events are identified once again for verification (re-PID). The ratio of the revolution times of the nucleus of interest to a reference nucleus, whose mass is well known, is use to accurately determine the mass. It should be noted here that isochronism of R3 holds only for a single isotope, the reference nucleus in many cases and the revolution time of the nucleus of interest has momentum dependence. Thus a small correction is introduced using the velocity, $\beta$, or $B\rho$ determined by the measurements prior to injection.

The main factor crucial for the precision of the derived mass is the degree of isochronism, including the magnetic field fluctuation. Isochronism is defined by the spread of the revolution time, $dT/T$, in the acceptance of the ring. A standard deviation of $dT/T \sim 2.8 \times 10^{-6}$ for a high degree of isochronism has been achieved at R3 for the full momentum acceptance ($dp/p \sim \pm 0.3\%$). This leads to a high mass resolving power of $3.5 \times 10^6$. We have established a scheme that can determine masses of the order of $10^{-6}$ with a measurement time of less than 1 ms. Further efforts are ongoing to increase the mass resolving power and injection efficiency by upgrading the kicker system,[15,16] and by fine-tuning the ion optics. In the near future, it is expected to be able to derive the masses of nuclei in a region that can only be achieved with the R3, *i.e.*, the nuclei having half-lives of <10 ms, and extremely rare that one can be extracted in a day.

## MRTOF mass measurement

Three multi-reflection time-of-flight mass spectrographs (MRTOF-MS) are in operation at RIBF. One of them is called ZD-MRTOF, which is located behind the ZeroDegree spectrometer (ZDS), combined with an RF-carpet-type cryogenic He gas catcher (RFGC)[17,18] in the SLOWRI project (Fig. 4). The fast (>100 MeV/nucleon) RIs provided from BigRIPS are stopped in the He gas and extracted as slow (<10 eV) RI ions from the RFGC using RF ion carpets. They are then guided into an ion trap, accumulated, cooled, and injected into the mass spectrograph (Fig. 5).[19] The ions are reflected back and forth between the ion mirrors typically for 600 revolutions corresponding to ≈10 ms at a maximum kinetic energy of 2.5 keV. Currently, the maximum mass resolving power has reached to $10^6$.

Since the location of the ZD-MRTOF is just in front of the beam dump of ZDS, mass measurements have been conducted symbiotically by re-using the RIs from other experiments carried out upstream without extra costs. For example, during the in-beam $\gamma$-ray experiments (HiCARI campaign) in winter 2020, more than 70 masses on RIs were measured with the ZD-MRTOF system (Fig. 6). Among them, three isotope masses have been measured for the first time and mass uncertainties of 11 isotope have been significantly improved from the previous ones.

For Ti and V isotopes, the nonexistence of the $N = 34$ empirical two neutron shell-gaps has been revealed experimentally with the new precision achieved by our equipment.[20]

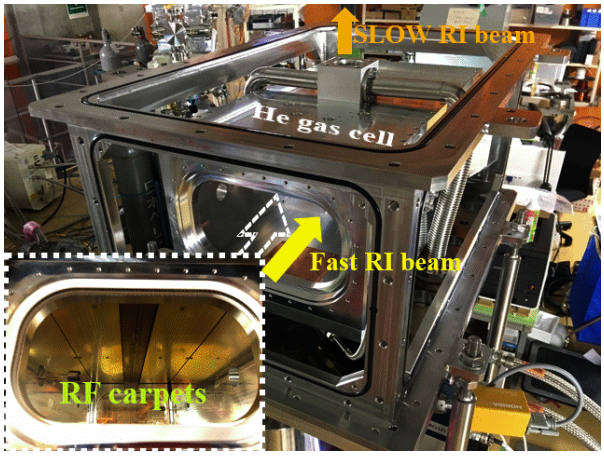


Fig. 4. Photographs of an RF-carpet-type He gas catcher (RFGC) behind ZDS of BigRIPS.

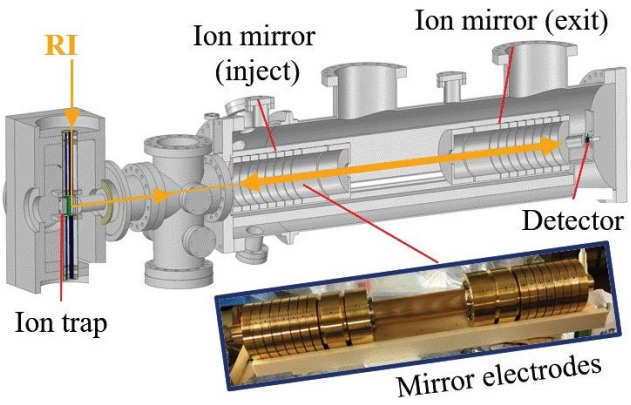


Fig. 5. Schematic view of MRTOF-MS (top) and a photograph of the ion mirrors behind ZDS of BigRIPS (bottom).

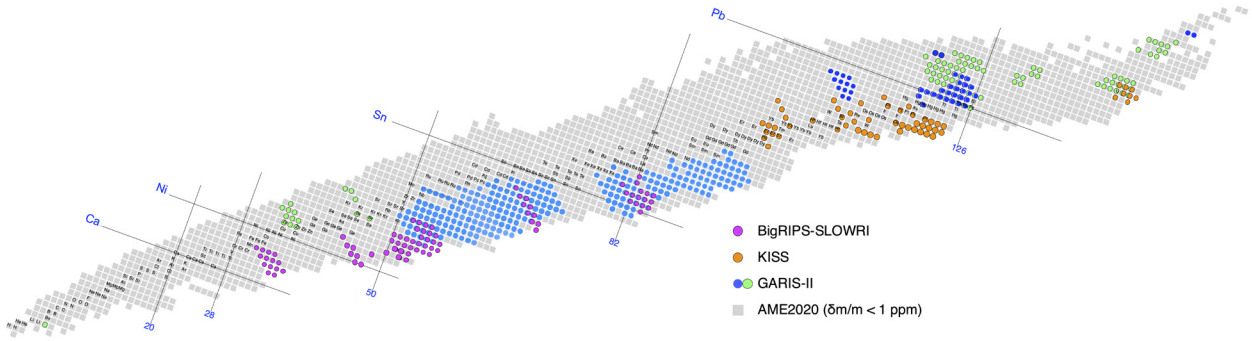


Fig. 6. Nuclear mass measurements performed with the three MRTOF-MS.

The second MRTOF is located downstream of the beamline at the KEK isotope separation system (KISS), which employs a mass spectrograph and a particle identification detector for laser spectroscopy. In-gas-cell laser resonant ionization spectroscopies for Os and Pt isotopes have been conducted with the KISS-MRTOF.[21,22] In addition, a new isotope of $^{241}$U has also been discovered by a precise mass measurement using KISS-MRTOF.[23]

The third MRTOF, SHE-Mass, is located behind GARIS-II at E6. It has been used for mass measurements of super-heavy nuclei such as $^{257}$Db.[24] In addition, a 9 MBq $^{252}$Cf fission source has been installed just in front of the He gas catcher. Even off-line, mass measurements on the fission fragments have continued and several first mass measurements have been performed.[25]

We have a plan to increase this versatile and portable instrumentation at RIBF; an MRTOF-MS is being installed behind GARIS-III, and a plan is underway to install an MRTOF-MS with a large He gas catcher behind SD4 at BigRIPS, which will lead to more opportunities to study unexplored nuclear species.

References

1) T. Uesaka *et al.*, Prog. Theor. Exp. Phys. **2012**, 03C007 (2012).
2) S. Michimasa *et al.*, Nucl. Instrum. Methods Phys. Res. B **317**, 305 (2013).
3) T. Kubo, Nucl. Instrum. Methods Phys. Res. B **204**, 97 (2003).
4) N. Orr *et al.*, Phys. Lett. B **258**, 29 (1991).
5) S. Michimasa *et al.*, Proc. Sci. **INPC2016**, 106 (2017).
6) H. Miya *et al.*, Nucl. Instrum. Methods Phys. Res. B **317**, 701 (2013).
7) S. Michimasa *et al.*, Phys. Rev. Lett. **121**, 022506 (2018).
8) S. Michimasa *et al.*, Phys. Rev. Lett. **125**, 122501 (2020).
9) N.-N. Ma *et al.*, Chin. Phys. C **43**, 044105 (2019).
10) S. Hanai *et al.*, in this report.
11) H. F. Li *et al.*, Phys. Rev. Lett. **128**, 152701 (2022).
12) H. Wollnik, Nucl. Instrum. Methods Phys. Res. A **258**, 289 (1987).
13) J. Trotscher *et al.*, Nucl. Instrum. Methods Phys. Res. B **70**, 455 (1992).
14) N. Fukuda *et al.*, Nucl. Instrum. Methods Phys. Res. B **317**, 323 (2013).
15) Y. Yamaguchi *et al.*, RIKEN Accel. Prog. Rep. **54**, 100 (2021).
16) Y. Yamaguchi *et al.*, in this report.

17) A. Takamine *et al.*, in this report.
18) J. M. Yap *et al.*, in this report.
19) M. Rosenbush *et al.*, in this report; Nucl. Instrum. Methods Phys. Res. A **1047**, 167824 (2023).
20) S. Iimura *et al.*, in this report; Phys. Rev. Lett. **130**, 012501 (2023).
21) H. Choi *et al.*, Phys. Rev. C **102**, 034309 (2020).
22) Y. Hirayama *et al.*, Phys. Rev. C **106**, 034326 (2022).
23) T. Niwase *et al.*, in this report; Phys. Lett. **130**, 132502 (2023).
24) P. Schury *et al.*, Phys. Rev. C **104**, L021304 (2021).
25) S. Kimura *et al.*, RIKEN Accel. Prog. Rep. **55**, 8 (2022).