First application of mass measurement with the Rare-RI Ring reveals the solar r-process abundance trend at $A = 122$ and $A = 123$

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The Rare-RI Ring (R3) is a recently commissioned cyclotron-like storage ring mass spectrometer dedicated to mass measurements of exotic nuclei far from stability at Radioactive Isotope Beam Factory (RIBF) in RIKEN. The first application of mass measurement using the R3 mass spectrometer at RIBF is reported. Rare isotopes produced at RIBF, ^{127}Sn, ^{126}In, ^{125}Cd, ^{124}Ag, ^{123}Pd, were injected in R3. Masses of ^{126}In, ^{125}Cd, and ^{123}Pd were measured and the mass uncertainty of ^{123}Pd was improved. The impact of the new ^{123}Pd result on the solar r-process abundances in a neutron star merger event is investigated by performing reaction network calculations of 20 trajectories with varying electron fraction Y_e. It is found that the neutron capture cross section on ^{123}Pd increases by a factor of 2.2 and β-delayed neutron emission probability, P_{1n}, of ^{123}Rh increases by 14%. The neutron capture cross section on ^{122}Pd decreases by a factor of 2.6 leading to pileup of material at $A = 122$, thus reproducing the trend of the solar r-process abundances. Furthermore, the nuclear deformation predicted to reach its maximum before $N = 82$ in the Pd isotopic chain is examined. The new mass measurement shows no evidence of such large deformation, though, experimental uncertainty should be further improved to draw a definitive conclusion. This is the first reported measurement with a new storage ring mass spectrometery technique realized at a heavy-ion cyclotron and employing individual injection of the pre-identified rare nuclei. The latter is essential for the future mass measurements of the rarest isotopes produced at RIBF.

The discovery of the historical GW170817 event of binary neutron stars merger and the subsequent kilonova AT2017go [1] for the GW170817 [2] was a major milestone toward revealing the secret of the synthesis of heavy elements via the rapid neutron capture process (r-process)[3]. The recent identification of strontium in the kilonova radiation gave a strong evidence of the production of r-process elements [4]. However, modeling of the accretion disk formed in supernova-triggered collapse of rapidly rotating massive stars or collapsars, showed that r-process elements could be also produced in considerable amounts [5]. The presence of r-process heavy elements was also observed in the dwarf galaxy Reticulum II [6], where the accretion disk of collapsars might be the main source of production. Recent studies suggest that heavy elements might be synthesized in three different sites based on observations of low metallicity stars [7], characterized by three types of patterns, a weak r-process, a strong solar-type r-process, and an actinide boosted r-process. To model the formation of heavy chemical elements under different astrophysical conditions, a large and diverse amount of nuclear data is needed, especially for neutron-rich nuclei that live for a fraction of a second. Nuclear masses are important ingredients since they reflect the neutron separation energies, which are required

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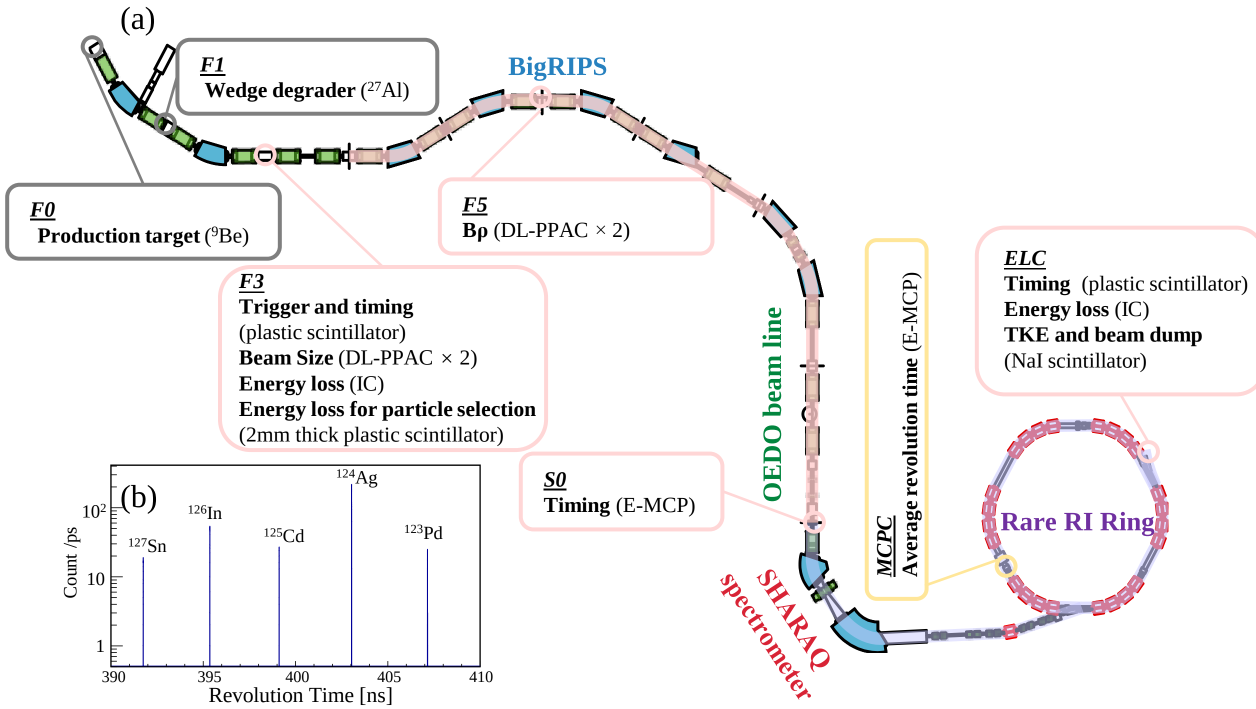

FIG. 1. (a) Configuration of the detectors installed in the beam line and Rare RI Ring (R3). (b) The revolution time spectrum for the 5 nuclei.

for the determination of neutron capture rates and photodissociation rates [3, 8, 9]. A vast number of neutron-rich nuclei involved in the r-process can now be produced in the laboratory at rare isotope facilities and their properties measured with high precision. However, all nuclei needed for modelling the r-process will not be accessible even at the new-generation radioactive-ion beam facilities. A robust model based on accurate properties of neutron-rich nuclei is thus essential to reveal the astrophysical conditions in which heavy elements could be produced. Such a model will help quantify the production rates in various sites, which will result in more accurate galactic chemical evolution calculations capable of reproducing the r-process elements' chemical abundances [10, 11].

This Letter reports precision mass measurements of neutron-rich nuclei produced at the Radioactive Isotope Beam Factory (RIBF) and their implication in the production of r-process elements with atomic mass number A=122 and A=123. Mass measurements of nuclei with neutron number N=77 were performed for the first time with a new type of mass spectrometer, namely the Rare-RI Ring (R3), recently commissioned at the RIBF/RIKEN facility [12]. We examine the implication of the ^{123}Pd mass on the abundance calculation for a neutron star merger event. These first mass measurements at RIBF of neutron-rich isotopes in a remote region of the nuclear chart open a door to reaching r-process nuclei at N=82 and beyond.

In the experiment, the secondary beam was produced by in-flight-fission of the 345 MeV/u ^{238}U beam provided by the Superconducting Ring Cyclotron (SRC) impinged on the 6 mm thick beryllium target which was placed upstream of the BigRIPS separator at F0 focal plane (see Fig. 1). The secondary fragments of interest were separated by the first stage of the BigRIPS as described in [13]. For this purpose, a 5 mm wedge-shaped degrader was introduced at the F1 focal plane of the BigRIPS. The magnetic rigidity Bρ and the transmission efficiency were optimized for the reference particle ^{124}Ag. The momentum selection was done by setting the slits at F1 to $\pm$2 mm, corresponding to the R3 momentum acceptance of $\pm$0.3%. The injection kicker magnets system placed inside the R3 is limited to a repetition rate of 100 Hz. Therefore, to accept the quasi-continuous beam from the SRC, the individual self-injected trigger technique was developed for injecting pre-identified particles of interest [14]. The particle identification (PID) was achieved by the ΔE-TOF method in the beam line, where ΔE is the energy loss measured by the ionization chamber (IC) placed at F3 and TOF is the time-of-flight measured by the plastic scintillator at F3 and the E-MCP detector [15] at S0 of the SHARAQ spectrometer. Also a 2-mm thick plastic scintillator was placed after the IC at F3 to get a rough ΔE information needed for removing contaminations [16]. Two position monitors PPACs (Parallel Plate Avalanche Counter) were installed at F3 to monitor the beam size and two double PPACs were installed at F5 which is a dispersive focal plane to measure Bρ of every individual particle prior to its injection into the R3.

The particle circulates in the R3 for about 1800 revolutions before it is ejected from the ring. The total TOF in the R3 was measured by the E-MCP detector at S0 and a plastic scintillator detector placed at ELC after the ejection from the R3. Another IC was installed at ELC, where an additional PID was performed. Finally, particles were stopped in the NaI scintillator detector placed behind the IC at ELC.

The mass-to-charge ratio (m/q) of the particle of interest with a revolution time T is determined relative to a reference particle with m_0/q_0 and T_0 by using the following formula [12, 17]:

$$\frac{m}{q} = \frac{m_0}{q_0}\frac{T}{T_0}\sqrt{\frac{1-\beta^2}{1-\left(\frac{T}{T_0}\beta^2\right)}}, \tag{1}$$

where β is the velocity of the particle of interest relative to the speed of the light in vacuum. Revolution time spectrum of all injected nuclei is shown in Fig. 1 (b) (details of determination of the revolution time in R3 can be found in [18]). Since the isochronous condition of the ring is optimized for the reference particle, T_0 is independent of the momentum. To determine the mass, the velocity β needs to be determined event-by-event from the time-of-flight along the beamline from F3 to S0 (TOF_{3S0}) by using the following equation,

$$\beta = \frac{Length_{3S0}}{(TOF_{3S0} + TOF_{offset})}. \tag{2}$$

The average path length from F3 to S0 ($Length_{3S0}$) and the TOF_{offset} caused by the electronics and the energy loss in the detectors on the beamline, are determined via Eq.(2) by using known masses of ^{124}Ag and ^{127}Sn. The parameters that could reproduce the known m/q values are $Length_{3S0} = 84.859(2)$ m and $TOF_{offset} = 325.47(1)$ ns. The mass is then determined for each event via Eq.(1). Additional systematic uncertainties, σ_{sys}, due to the determination of parameters such as $Length_{3S0}$, TOF_{offset} and T_0 were estimated and reported in Table I. Details of data analysis method can be found in references [12, 18]. The full data analysis method as well as the details of estimating the systematic uncertainties will be reported in a subsequent publication. The mass excess values determined for all nuclei are listed in Table I. Comparison with literature values from the recent Atomic Mass Evaluation, AME2020 [19], are plotted in Fig. 2. As shown in Table I, the uncertainties are dominated by the mass uncertainty of the reference particle ^{124}Ag at 250 keV. The choice of ^{124}Ag as a reference instead of ^{125}Cd, which has lower uncertainty, is mainly due to the presence of a long-lived isomeric state at 186 keV in the latter that is difficult to separate with R3. The mass precision was therefore scarified for higher accuracy. However, if the mass of ^{124}Ag is remeasured with higher precision, the uncertainties of all other masses will be reduced.

In Fig. 3, the two neutron separation energies (S_{2n}) are shown with the updated value for the most neutron-rich Pd isotope at N=77. Nuclear shape deformation before the magic number N=82 was predicted by several models [20–23]. The deformation in this mass region is believed to affect the r-process abundances before the rise of the A=130 peak. Failure to produce enough material in the A=120 region by several models was thought to be due to the shell quenching at N=82 [23]. However, better description of the deformation in recent nuclear models led to more accurate reproduction of the r-process abundances before A=130 [24]. The increase in S_{2n} values can be a signature of such deformation. As can be seen in Fig. 3, the FRDM predicts that nuclear deformation reaches its maximum at the Pd isotopic chain. The new S_{2n} value of ^{123}Pd shows a smooth decrease following the trend of the mass surface. Due to still relatively large uncertainty of our mass value, the presence of the deformation cannot be excluded. Based on the estimation of our systematic uncertainties, the mass uncertainty could be reduced to about 100 keV if the mass of ^{124}Ag is remeasured with a precision of less than 30 keV. It should be noted that the FRDM overestimates the size of the deformation in the Cd isotopic chain, especially when approaching N=82. Based on experimental masses of Cd isotopes, the deformation in the Ag and Pd isotopic chains might not be as large as that predicted by the FRDM.

We simulate the impact of the mass measurement of ^{123}Pd in the astrophysical r-process by employing the

TABLE I. Mass excess from literature and the mass excess of nuclei measured in this work are shown in the second and third column, respectively. Total uncertainties are shown as well as the contribution from the reference mass uncertainty σ_{m_0} and the statistical uncertainty σ_{stat}. The systematic uncertainty σ_{sys} is estimated from the uncertainty of T_0 and the fit parameters $Length_{3S0}$ and TOF_{offset} of Eq.(2).

Nucleus	ME_{AME20} [keV]	ME_{R3} [keV]	σ_{total} [keV]	σ_{m_0} [keV]	σ_{stat} [keV]	σ_{sys} [keV]
^{126}In	-77809(4)	-77707	269	254	65	62
^{125}Cd	-73348.1(29)	-73237	320	252	192	40
^{123}Pd	-60430(790)	-60282	265	248	86	40

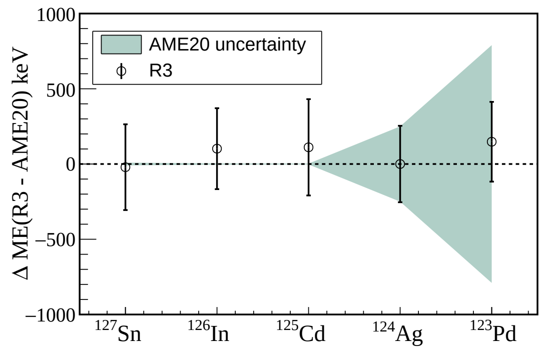

FIG. 2. Mass excess values of nuclei measured at R3 compared to literature values from AME2020 [19]

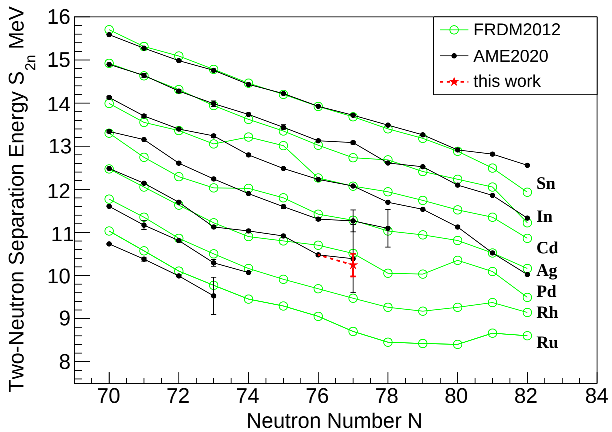

FIG. 3. The two neutron separation energy values (S_{2n}) plotted as a function of neutron number from Ru to Sn isotopic chains taken from AME2020 [19]. The S_{2n} value derived from our new mass value of ^{123}Pd is shown in red, while predictions of the FRDM are shown by the green line [20].

Portable Routines for Integrated nucleoSynthesis Modeling (PRISM) reaction network [25, 26]. The baseline nuclear physics properties are simulated with FRDM2012 [20, 27–30]. The mass of ^{123}Pd in the baseline model is also taken from the FRDM2012. Changes to the mass propagate to cross sections and branching ratios in neighboring nuclei as in [9]. We find that the changes in the capture cross sections, and β-delayed neutron probabilities (discussed below) are significant in contrast to other works which do not include these effects [31].

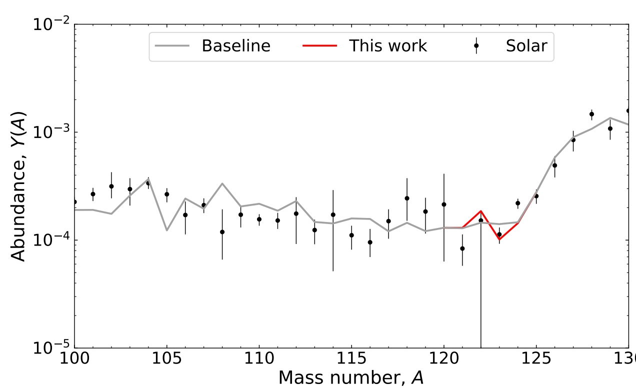

FIG. 4. The local impact (red) of the ^{123}Pd mass measurement when simulated in the r-process. The baseline calculation is shown in grey and the solar r-process residuals in black [32].

Since there are uncertainties in the astrophysical conditions that could produce nuclei in the mass $A \sim 120$ range, we simulate a set of 20 trajectories with varying neutron-richness from electron fraction of $Y_e = 0.15$ to $Y_e = 0.35$ chosen using a procedure similar to that described in [33]. Figure 4 shows the impact of ^{123}Pd when combining all trajectories together in a weighted sum that best matches the solar r-process residuals. We find that the neutron capture cross section for ^{122}Pd decreases by a factor of 2.6 and for ^{123}Pd increases by a factor of 2.2, while the P_{1n} value, probability for the β-delayed neutron emission, of ^{123}Rh increases by 14% with the updated mass, resulting in an effective pileup of material along the $A = 122$ isobar relative to the baseline. Some conditions enhance this effect, notably $Y_e = 0.28$ ($F = 0.61$), $Y_e = 0.29$ ($F = 1.88$), $Y_e = 0.30$ ($F = 1.41$), $Y_e = 0.31$ ($F = 0.99$), $Y_e = 0.32$ ($F = 0.61$) with F defined as in [34]. As a conclusion, for these conditions there is a larger flow through the ^{123}Pd nucleus, see also the discussion in [35]. The average impact factor is $\langle F \rangle = 0.247$, indicating a local change in the abundances, in line with the prediction of sensitivity studies [34].

In summary, the first application of mass measurements performed by the Rare-RI Ring at the RIBF facility is reported. The most neutron-rich nuclei below the doubly magic nucleus ^{132}Sn were studied, proving the feasibility for mass measurements of r-process nuclei at N=82. The present uncertainty of our measurement can be reduced if the reference mass of ^{124}Ag is remeasured with higher precision, which will result in a firm conclusion about the presence of nuclear deformation in the Pd isotopic chain. We performed calculations to estimate the impact of the ^{123}Pd mass measured in the r-process. We found if our new mass value is used instead of the FRDM value the solar r-process abundances at A=122 and A=123 are modified, resulting in a better reproduction of the trend in the abundance at these masses. This indicates that the r-process calculations are very sensitive to masses in this region since a change of ^{123}Pd mass by just 478keV causes a sizeable effect. This finding highlight the need for high precision mass measurements to address the r-process in this mass region.

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