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科学研究費助成事業　　研究成果報告書

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研究課題名（和文）An advanced in-trap deflector for background-free nuclear mass measurements

研究課題名（英文）An advanced in-trap deflector for background-free nuclear mass measurements

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交付決定額（研究期間全体）：（直接経費）　　3,100,000円

研究成果の概要（和文）：本研究では、理研RIBFのオンライン多重反射飛行時間型質量分析計のための新しいデフレクタ(in-MRTOF deflector: IMD)を開発した。これを使用することで、不純物イオンを全て除去し、目的の複数質量のイオンのみを検出できる。IMDを3つのMRTOFシステムに設置し、その各々でオンライン精密質量測定に成功した。BigRIPS実験において73-75Niの質量を同時に測定し、GARIS-II実験では257,258Dbの超重元素の質量を測定した。さらにIMDを利用し、ヘリウムガス中でのイオン荷電分布の元素依存性を初めて系統的に測定できるようになった。

研究成果の学術的意義や社会的意義
In this research project, a new tool was developed to obtain backgound-free spectra in an MRTOF mass spectrometer for radioactive ions. Mass measurements of exotic nuclei are a fundamental tool for the study of nuclear properties far away from stability, astrophysics, and fundamental interactions.

研究成果の概要（英文）：In this research project, a new in-MRTOF deflector (IMD) for the on-line multi-reflection time-of-flight mass spectrographs (MRTOF-MS) at RIKEN/RIBF has been developed. The new deflection system has been successfully installed, tested, and applied in on-line nuclear mass measurements at the BigRIPS separator.
Using the new IMD, several chosen ion-mass units could be transmitted to the TOF detector, while all other ions were ejected. This new system allowed for overwhelmingly clean ion spectra in the experiments RIBF199 and RIBF202 (see RIKEN RIBF experiments), where the masses of 73Ni - 75Ni have been measured simultaneously. IMD systems have been installed in three MRTOF systems and allowed for successful online mass measurements. At the GARIS-II separator, the superheavy element masses for 257,258Db have been measured using the IMD. Furthermore, the IMD allowed for the first systematic measurements of the electrical charge state formed for different chemical elements in a helium gas.

研究分野：Nuclear Physics and Mass Spectrometry

キーワード：Nuclear Structure　Nuclear Astrophysics　Ion Traps　Ion Manipulation　MRTOF-MS　Isotope Separation　Background Reduction

１．研究開始当初の背景

The nuclear mass is one of the most fundamental observables of atomic nuclei and crucial ingredient to study exotic isotopes for nuclear structure investigations and astrophysics. For the investigation of exotic nuclei by the nuclear mass, high-precision technologies are the preferred tool as they allow for accurate and precise mass measurements with a relative precision of ***δm/m* < 10-7**, where ***δm*** is the mass uncertainty of the measurement and ***m*** is the mass of the atom or nucleus. Such precision can presently only be reached with methods performed at low ion energies. The multi-reflection time-of-flight mass spectrograph (MRTOF-MS) is nowadays a widely-used tool for fast, efficient, and precise atomic mass measurements, and is used in many radioactive-isotope (RI) beam facilities worldwide.

However, a common impediment to perform MRTOF mass measurements comes from the fact that ions produced by in-flight fission or fusion-evaporation reactions (like the case at RIKEN/RIBF) must be stopped in a gas cell to make them accessible at low-energies. Chemical reactions in the gas and ionization of residual stable molecules becomes a critical issue if the RI of interest are produced at low rates. Additionally, exotic ions are typically delivered in a "cocktail beam" with the least exotic species having the greatest yields. Such unwanted ions create an unwieldy background that complicates analyses. Worse, their existence can cast doubt on the identification of low-yield species. It has been shown [Y. Toker et al., J. Instrum. 4, P09001 (2009)] that a pulsed deflector in the drift region of an MRTOF device can rapidly and fully suppress such contaminants. While the ions are circulating in an MRTOF device, the deflector is pulsed in a specific pattern which allows to select/keep the ions of interest while ejecting all contaminants (see Fig. １). This research project intended to develop and assemble a novel in-MRTOF deflector (IMD) system to eject unwanted ions dependent on their mass to succeed mass measurements in challenging on-line experiments at the RIBF facility.

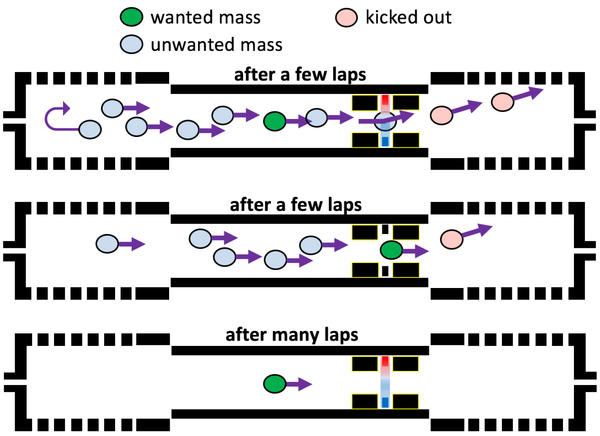

Figure 1: Principle of an in-MRTOF deflector. Top: In-trap deflector is activated and deflects unwanted ions. Center: deactivation for wanted ions. Bottom: Ions of interest are isolated and the spectrum is back-ground free.

２．研究の目的

The goal of the study was to develop a pulsed electric ion-deflector and install the unit into the drift-tube section of the ZD MRTOF system at RIBF and other MRTOF systems. Furthermore: to test the performance and apply the new technique in on-line beam times to allow for atomic mass measurements of exotic isotopes.

(1) Developing a mechanical design for the IMD: The deflector system had to fulfil the condition that the electric field must be strong in a very limited region, while negligibly weak in other regions of the MRTOF mass spectrograph to not disturb the ions of interest. Thus, a low voltage was anticipated while still allowing for a full suppression of unwanted ions. Furthermore, the new deflector system had to integrate into the existing system without changing the stack of electrostatic mirrors.

(2) Goal of the electronics and control system: The IMD required a reliable and fast electronic pulse-generator delivering the square-wave voltage pulse to the deflector electrode. This pulser had to guarantee an extremely low residual voltage when being in the off-state to not disturb ion of interest.

The timing- and control system providing electronic trigger signals to the pulser had to be accurate in time and programmable to provide a timing sequence which avoids any disturbance of the ions of interest.

３．研究の方法

The new in-MRTOF deflection system has been constructed, installed, and applied at three existing MRTOF systems: the ZD MRTOF system at the ZeroDegree spectrometer (BigRIPS facility), the SHE Mass MRTOF system located at the GARIS-II separator, and the MRTOF-MS at the KISS (KEK Isotope Separator System) facility.

(1) The new mechanical design was supported by simulations of the ions' trajectory using the state-of-the-art software SIMION. The length of the deflector electrode was chosen so that a voltage between 20 V and 30 V guarantees for an ejection of unwanted ions. Upon the simulations, the design (see Fig. 2) has been chosen to have an asymmetry (deflector electrode placed out of the axial center) to provide a longer path for ions inside the central drift section in order to ensure a contact with the electrode walls with less than 30 V applied.

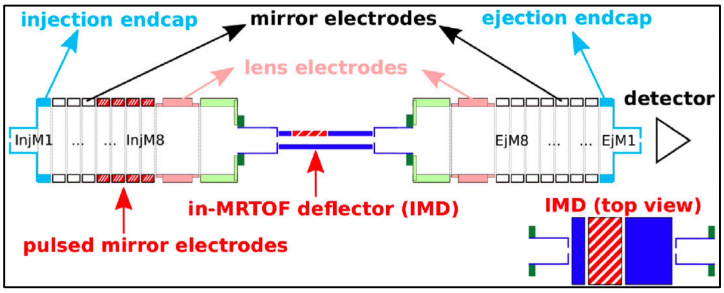

Figure 2: Scetch of the ZD MRTOF-MS with the new IMD shown with side view and top view.

The distance of the of the plates is 1 cm while the width was chosen with 10 cm to shield the region from the vacuum-chamber potential outside of the electrode stack. For the deflection electrode, an axial length of 5 cm was chosen to efficiently eject ions during- or after their passage. All parts were precision machined by an external company to exactly match the geometric details of the existing structure. The whole IMD integrates as a part of the former cylindrical drift tube and had to be embedded into the mechanical mounting structure. All metal parts except the deflection electrode by itself was electrically connected to the drift-tube structure while the deflection electrode was mounted using a ceramic block as holder and insulator (see Fig. 3).

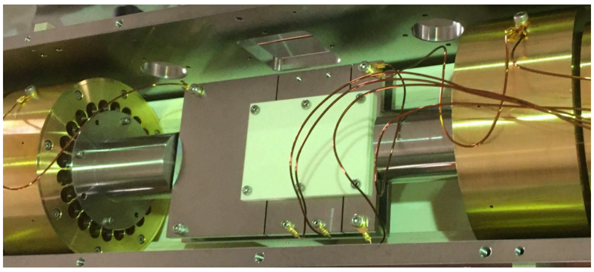

Figure 3: Photo of the center of the ZD MRTOF-MS after installation of the new IMD section.

(2) In order to drive the IMD, an electric pulsing system has been developed which is able to provide square-wave pulses in a voltage range between 15 V and 45 V. The trigger signal is provided by a TTL logic trigger from an existing timing FPGA system. Using fast transistor switches, a duration for the change between on-state and off-state of less than 20 ns was achieved. The entire circuit is supplied by insulating DC-DC converters, so that the electric potential of the central MRTOF section can be freely chosen for reasons of ion optics. Power supplies to provide the deflection voltage have been installed on-board and are integrated into the circuit (see Fig. 4).

Crucial parameters for the timing of the pulses have been measured initially using alkali ions (e.g. the flight duration to the IMD and MRTOF lap time). Upon the known parameters, the ion-mass dependent timing structure of the deflection pulses has been calculated by a routine using the software LabVIEW and subsequently transmitted to the FPGA memory providing the TTL signals during the measurement cycle. Complex patterns can now be calculated to protect not only one single ion mass, but several mass units at the same time while all other ions with unwanted mass units are eliminated.

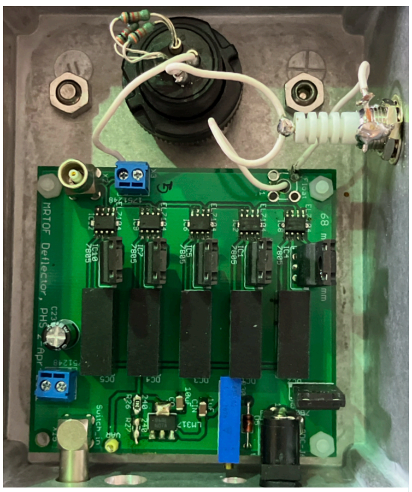

Figure 4: Voltage-pulse circuit for the IMD. Power supplies and transistor switches are integrated.

４．研究成果

The IMD project was successful by all means and it denotes the most crucial modification of MRTOF systems at RIBF in the recent years. The new IMD unit has been used in on-line experiments and became an invaluable standard device in our MRTOF systems. First tests have been performed using molecules ionized in a helium-filled ion catcher by an alpha-emitter source irradiating contaminations inside the helium gas. The molecules have been extracted and forwarded to the MRTOF-MS equipped with the new deflector system. Using repetitive pulses synchronized to not affect the ions of interest, only a single mass unit could be transmitted to the TOF detector while all other mass units have been removed from the flight path by interaction with the activated deflection field. Additional programming of the pulsing sequence allowed to include multiple mass units (e.g. A=74, A=75, and A=76) simultaneously while excluding all other ions from reaching the detector. The most important results will be explained in the following.

<u>**First systematic studies about the ion charge states produced in a cryogenic He gas:**</u>
Incoming ions from in-flight fission or fusion-evaporation production enter the gas cell as highly-charged ions and obtain electrons from the helium gas wherein they are stopped.

There is only scarce knowledge about the electric charge state which certain chemical elements will obtain after collecting electrons from a clean helium gas. This is a major challenge for online experiments as the identification of the most abundant charge state to be used for mass measurements costs expensive time at the accelerator.

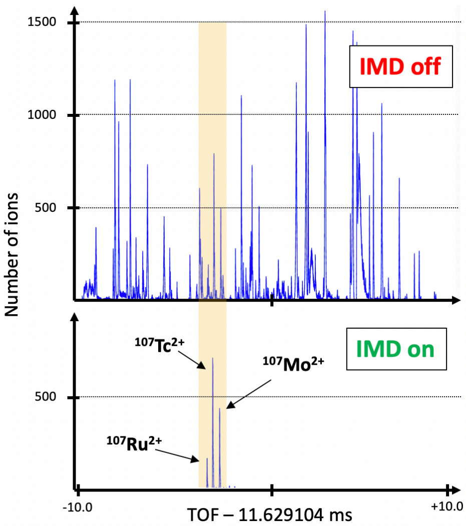

Figure 5: Comparison of spectra with IMD on/off for A = 107 fission products from the gas cell.

The first attempt for a systematic study was done by producing high-energy radionuclides from a ^{246}Cm fission source and capture the products in the gas cell of the ZD MRTOF-MS. Ions have been extracted and mass separated by the IMD unit to transmit and identify the chemical species of up to three isotopic mass units simultaneously (see next section). Without using the IMD, such measurements were extremely ambiguous and uncertain as many signals from other mass units were present (see Fig. 5). As the number of laps performed for unwanted mass units is unknown, a clear identification and ion counting can become impossible in the case that the peaks are overlapping.

As a result of the study, a first systematic relation between the relative abundance of electric charge states and the ionization energies (1+, 2+ states) has been obtained for many chemical elements **[A. Takamine et al., in preparation]**. The study shows a common trend depending on the 2nd and 3rd ionization potential of the element (see Fig. 6). In future online experiments, this information is invaluable to shorten the preparation for mass measurements, i.e. the time needed to search for the most abundant charge state.

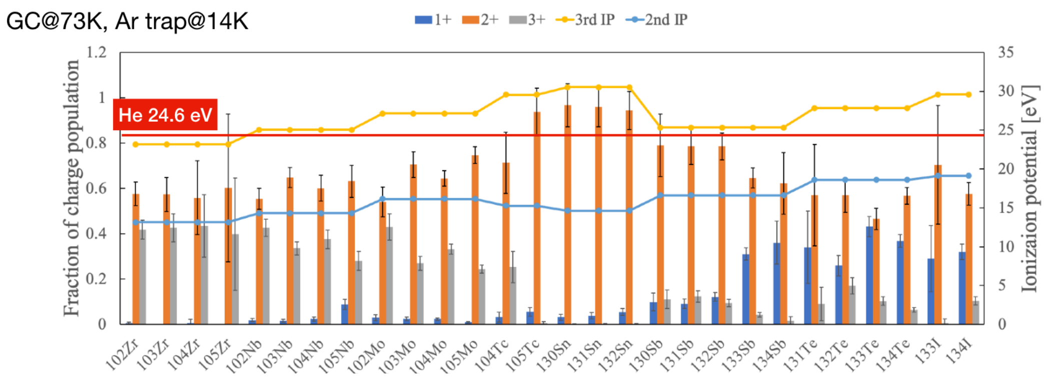

Figure 6: fraction of charge-state population for elements between Zr and I, including different isotopes of the same element. A relation between ionization potential and fraction was confirmed.

Mass measurements of neutron-rich Ni isotopes delivered at BigRIPS:

In 2021, online beam time for the measurement of neutron-rich Ni isotopes was allocated. The isotopes of interest have been produced using a ^{238}U beam with 345MeV/nucleon impinging on a Be target (experiment RIBF202, see RIKEN RIBF experiments or contact marco.rosenbusch@riken.jp). The Ni isotopes have been slowed down using SUS and Al beam-energy degraders, and have been stopped in the He gas cell at the ZD MRTOF-MS. Upon extraction and transport of ions in the ion guides and trap system, a high number of contaminating ions complicated the measurements (stable molecules and other products delivered with the beam, similar to the top of Fig. 5). The IMD unit was used to select the mass numbers of interest.

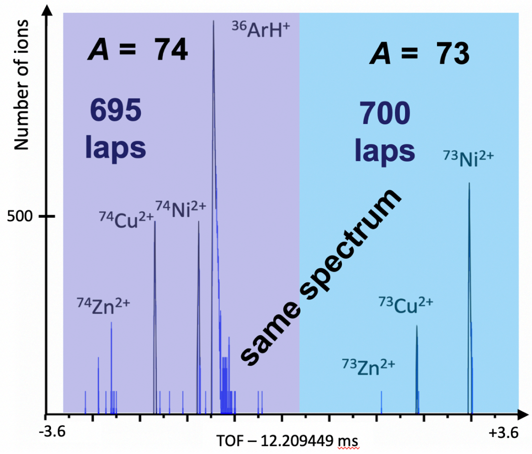

Figure 7: Time-of-flight measurement of Ni isotopes. Two isobaric chains, A = 74 and A= 75, have been measured simultaneously while being free of any other contaminants.

For the first time in an online experiment the new mode of selectively protecting more than one mass unit has been applied. A logic NOR condition was employed to pre-calculate the timing pattern of the trigger signals. The NOR condition guaranteed the off-state of the IMD if any of the wanted ions is crossing the critical region. In this way, the isotopes ^{73,74}Ni have been measured at the same time, and also ^{74,75}Ni (see Fig. 7), where the two latter represent the present cutting edge of known Ni masses. The new IMD lead to overwhelmingly clean ion spectra and allowed for unambiguous, accurate, and precise mass measurements. The new mass results will be published soon **[W. Xian, M. Rosenbusch et al., in preparation]**.

Extended mass measurements of ^{257,278}Db:

After the first mass measurement success of the superheavy element ^{257}Db at the SHE Mass MRTOF system located at the GARIS-II separator **[P. Schury et al., PRC 104, L021304 (2021)]**, a subsequent online measurement has been performed to obtain more data for ^{257}Db, and also ^{258}Db for the first time. Both isotopes have been obtained as doubly-charged ions from the gas cell. The production cross section was low and the measurement took 46 hours, where only a few events per day were detected. Even with clean conditions in the gas cell, such long measurement times are generally accompanied with contaminant background molecules in the spectrum. The installation and application of an IMD in this system allowed for clean spectra lasting for a long measurement time. With success, the two isotope masses have been measured at the same time while contaminants were excluded. Together with decay-spectroscopic correlation of the events, the identification of both species was unambiguous and the count statistics for both cases could be extended **[P. Schury et al., in preparation]**. The new results are preliminary published in **[RIKEN Accel. Prog. Rep. 55, 6 (2022)]** and **[RIKEN Accel. Prog. Rep. 55, 7 (2022)]**, and will be published after finishing the remaining available beam time.

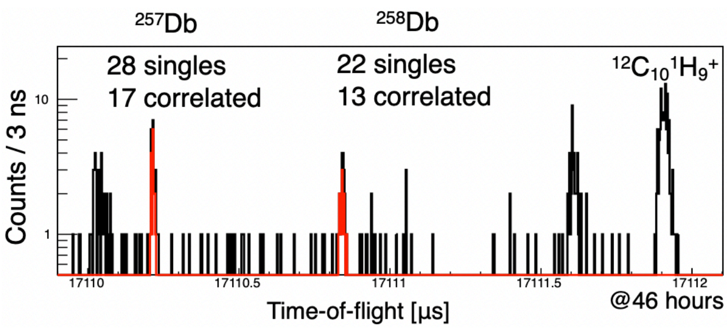

Figure 8: IMD-purified spectra for the mass measurement of the superheavy elements ^{257}Db and ^{258}Db.

５．主な発表論文等

〔雑誌論文〕　計5件（うち査読付論文　5件／うち国際共著　2件／うちオープンアクセス　3件）

１．著者名 Iimura S.、Rosenbusch M. et al.	４．巻 130
２．論文標題 Study of the N=34 Shell Gap for Ti and V by the First High-Precision Multireflection Time-of-Flight Mass Measurements at BigRIPS-SLOWRI	５．発行年 2023年
３．雑誌名 Physical Review Letters	６．最初と最後の頁 12501
掲載論文のDOI（デジタルオブジェクト識別子） 10.1103/PhysRevLett.130.012501	査読の有無 有
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 該当する

１．著者名 Rosenbusch M.、Wada M.、Chen S.、Takamine A.、Iimura S.、Hou D.、Xian W.、Yan S.、Schury P.、Hirayama Y.、Ito Y.、Ishiyama H.、Kimura S.、Kojima T.、Lee J.、Liu J.、Michimasa S.、Miyatake H.、Moon J.Y.、Mukai M.、Naimi S.、Nishimura S.、Niwase T.、Sonoda T.、Watanabe Y.X.、Wollnik H.	４．巻 1047
２．論文標題 The new MRTOF mass spectrograph following the ZeroDegree spectrometer at RIKEN’s RIBF facility	５．発行年 2023年
３．雑誌名 Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment	６．最初と最後の頁 167824～167824
掲載論文のDOI（デジタルオブジェクト識別子） 10.1016/j.nima.2022.167824	査読の有無 有
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 該当する

１．著者名 Schury P., Niwase T., Wada M., et al.	４．巻 55
２．論文標題 Improved mass measurements on 257Db by decay-correlated mass spectroscopy	５．発行年 2022年
３．雑誌名 Riken Accelerator Progress Report	６．最初と最後の頁 6
掲載論文のDOI（デジタルオブジェクト識別子） なし	査読の有無 有
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１．著者名 Schury P., Niwase T., Wada M., et al.	４．巻 55
２．論文標題 Initial mass measurements on 258Db by decay-correlated mass spectroscopy	５．発行年 2022年
３．雑誌名 Riken Accelerator Progress Report	６．最初と最後の頁 7
掲載論文のDOI（デジタルオブジェクト識別子） なし	査読の有無 有
オープンアクセス オープンアクセスとしている（また、その予定である）	国際共著 －

1．著者名 Rosenbusch M., Wada M., Takamine A., et al.	4．巻 55
2．論文標題 New developments and progress of the ZD-MRTOF system in 2021	5．発行年 2022年
3．雑誌名 Riken Accelerator Progress Report	6．最初と最後の頁 86
掲載論文のDOI（デジタルオブジェクト識別子） なし	査読の有無 有
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〔学会発表〕　計8件（うち招待講演　3件／うち国際学会　3件）

1．発表者名 Marco Rosenbusch
2．発表標題 Recent atomic mass measurements of radioactive species using the new ZD-MRTOF system at BigRIPS/RIKEN
3．学会等名 JPS spring meeting 2023
4．発表年 2023年

1．発表者名 Marco Rosenbusch
2．発表標題 Exploring exotic nuclei by high-precision MRTOF mass measurements: The new ion catcher and mass spectrograph at RIKEN's RIBF facility
3．学会等名 RIKEN seminar（招待講演）
4．発表年 2023年

1．発表者名 Marco Rosenbusch
2．発表標題 Exploring exotic nuclei by high-precision MRTOF mass measurements: The new ion catcher and mass spectrograph at RIKEN's RIBF facility
3．学会等名 Trapped Charged Particles（国際学会）
4．発表年 2022年

１．発表者名 Marco Rosenbusch
２．発表標題 New high-precision nuclear mass studies by the first MRTOF mass spectrometer at the BigRIPS facility
３．学会等名 TCHoU workshop, Tsukuba University（招待講演）
４．発表年 2022年

１．発表者名 Marco Rosenbusch
２．発表標題 High-precision mass measurements for astrophysics and nuclear structure studies using the new ZD-MRTOF system
３．学会等名 UKAKUREN workshop, Osaka University（招待講演）（国際学会）
４．発表年 2022年

１．発表者名 Marco Rosenbusch
２．発表標題 New mass measurements of exotic nuclides by the first MRTOF setup at BigRIPS/RIKEN
３．学会等名 Japanese Physical Society spring meeting 2020
４．発表年 2021年

１．発表者名 Marco Rosenbusch
２．発表標題 The new MRTOF Mass Spectrograph at the ZeroDegree spectrometer
３．学会等名 Japanese Physical Society autumn meeting 2020
４．発表年 2021年

1．発表者名 Marco Rosenbusch
2．発表標題 The new MRTOF Mass Spectrograph at the ZeroDegree spectrometer
3．学会等名 16th International Symposium on Nuclei in the Cosmos（国際学会）
4．発表年 2021年

〔図書〕　計0件

〔産業財産権〕

〔その他〕

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6．研究組織

	氏名 （ローマ字氏名） （研究者番号）	所属研究機関・部局・職 （機関番号）	備考

7．科研費を使用して開催した国際研究集会

〔国際研究集会〕　計1件

国際研究集会	開催年
16th International Symposium on Nuclei in the Cosmos	2021年～2021年

8．本研究に関連して実施した国際共同研究の実施状況

共同研究相手国	相手方研究機関