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Review

Unveiling nuclear isomers through multiple-reflection time-of-flight mass spectrometry

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Abstract Nuclear isomers, the excited meta-stable states of nuclei, offer profound insights into nuclear structure. This article reviews the intersection of nuclear isomer research with mass spectrometry methodologies, particularly focusing on novel capabilities of the multiple-reflection time-of-flight mass spectrometry (MR-TOF-MS) technique. Through a comprehensive examination of established methods for isomer identification and characterization, alongside the technical principles underlying MR-TOF-MS, this review discusses the pivotal role of mass spectrometry in advancing our understanding of nuclear isomers. The operational principles and recent developments in MR-TOF-MS technology are explained and exemplified through case studies from prominent research facilities. Furthermore, this work discusses ongoing efforts to enhance sensitivity, resolution, and measurement capabilities in MR-TOF-MS, promising continued advancements in nuclear physics research and applications.

1 Introduction

Nuclear isomers represent a captivating facet of nuclear structure. They are nuclei in excited meta-stable states with identical atomic and mass numbers to the ground states [1]. The lifetime of these isomeric states varies from nanoseconds to years. One isomeric state is even stable, ^{180m}Ta [2]; this is especially noteworthy as the ground state has a half-life of about 8 h. A very prominent application of nuclear isomers is nuclear clocks [3]. Thus, they offer insights into nuclear structure dynamics and the potential for applications in diverse fields; for an in-depth discussion on the subject of nuclear isomers, please see Ref.[4].

Mass spectrometry is a powerful technique employed to identify and quantify species based on their mass-to-charge ratios. With advancements in instrumentation and methodology, mass spectrometry has evolved into a versatile tool capable of probing the intricate details of nuclear structure, including detecting and characterizing nuclear isomers.

The established methods to measure and identify isomeric states are (i) identification by decay information (energy and time), (ii) kinematics of the productions, (iii) measurement of properties, which can be mass (excitation energy) or laser spectroscopy (spin). The method (i) has yielded the most information on nuclear isomers; one can access short-lived isomers and get detailed structure information from the gamma radiation emitted by the isomer; that strength is also a weakness of this method if the isomer is long-lived (no decay information) or not decaying by gamma radiation. The range from 1 to 40 keV in the γ spectroscopy is X-ray dominated and challenging for this method [5]. The identification typically needs several particles detected in coincidence. Therefore, a longer half-life means more random coincidences and, thus, a large background. This method mainly applies to gamma-decaying isomers with half-lives shorter than one millisecond. Method (ii) is only used for extremely short-lived cases inaccessible by other methods [6]. A key advantage of method (iii) is its universal identification of the isomer by mass or laser spectroscopy, not relying on decay properties. Thus, this method can study and identify isomeric states independently of knowledge of their properties (e.g., decay mode). Mass spectrometry has the advantage of far superior sensitivity down to single ions per hour with little to no background [7] compared to ions per second

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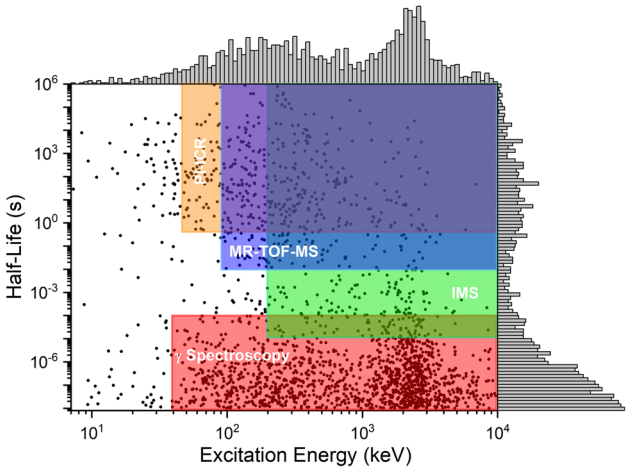

Fig. 1 Distribution of known isomers in excitation energy vs. half-life plot. The raw data are obtained from the Atlas of isomers-second edition [1]. The colored boxes represent the capabilities of the detection methods for isomers with different excitation energies and half-lives

for laser spectroscopy [8]. Scanning methods, such as laser spectroscopy, require prior knowledge of the properties of the isomeric state to be studied.

The interplay between nuclear isomers and mass spectrometry presents a unique way of exploring fundamental principles of nuclear physics. The central mass spectrometry methods in the past were Penning traps and storage rings. In Penning traps, a new technology was implemented in recent years that boosts the mass-resolving power and reduces the measurement time, the so-called Phase Imaging Ion Cyclotron Resonance (PI-ICR) [9]. This allows the mass-resolving powers of 2,000,000 in about 0.5 s of measurement time. With this new method, several isomers have been discovered in recent years [10–12]. In storage rings, two methods are applied: the isochronous mass spectrometry at IMP-Lanzhou [13, 14] and ESR, GSI [15]. This allows high-speed measurements in the 10 μs time scale with mass-resolving powers of a few hundred thousand, leading to several isomer discoveries [16, 17]. The other method is Schottky mass spectrometry [18], with a mass-resolving power of 700,000 and a measurement time of several seconds. This method has been used for several isomer discoveries and studies [19, 20]. In recent years, work on further improving this method is ongoing, from double detectors or position-sensitive detectors [21–23] to a combination of both methods [24], promising resolving powers of close to one million at a measurement time of a few tens of milliseconds [25].

A further improvement in sensitivity and measurement speed is highly desirable to make mass spectrometry even more attractive for isomer studies. This has been achieved recently with the invention of multiple-reflection time-of-flight mass spectrometry (MR-TOF-MS) with superior mass-resolving power, sensitivity, and speed [26]. MR-TOF-MS, like Penning traps, when coupled to a gas-filled stopping cell [27], are compatible with all production methods for nuclear isomers. In contrast, storage ring mass spectrometry only works for relativistic ions, i.e., not for exotic nuclei production methods at the Coulomb barrier (fusion evaporation, multinucleon transfer, etc.). That is highly relevant for investigations of nuclear isomers as some low-energy production methods, like multinucleon transfer reactions, are especially powerful for isomer production [28]. In facilities that had a Penning trap and upgraded recently with the MR-TOF-MS[29, 30], it has been demonstrated that MR-TOF-MS exhibits sensitivity at least an order of magnitude higher. This heightened sensitivity is evidenced by its consistent measurement of isotopes situated one or two neutrons further away from stability compared to those measured by Penning traps for the same experiment [11, 31, 32].

Figure 1 shows a map of experimentally known isomers in excitation energy vs. half-life plot. The color-coded boxes represent the capabilities of the detection methods of isochronous mass spectrometry (IMS), PI-ICR, MR-TOF-MS, and γ-spectroscopy, as discussed above.

We observe several distinct features, one of which is the scarcity of millisecond isomers, aligning with the absence of an established experimental method for that half-life regime that is applicable to all production mechanisms. It is important to note that IMS is not suitable for nuclear reactions at the Coulomb barrier. However, MR-TOF-MS can precisely access this millisecond region, presenting significant discovery potential. The accomplishments of MR-TOF-MS in this context and its future prospects for nuclear isomer studies will be explored in this paper.

2 Technical

MR-TOF-MS is a powerful technique for determining ions' mass-to-charge ratio with high accuracy and resolution; it has found widespread applications in various fields [26, 33]. MR-TOF-MS operates based on time-of-flight principles, where ions are accelerated by an electric field and then travel through a flight tube toward a detector.

The ion path is folded to increase the flight distance and, thereby, the mass-resolving power by orders of magnitude. This is realized by two electrostatic mirrors between which the ions fly back and forth for up to thousands of reflections. The time that ions travel to the detector is directly proportional to their mass-to-charge ratio, allowing for accurate mass determination. For the highest mass-resolving powers, the reflection must be isochronous, and in general, all ion optical aberrations must be minimized [34]. This also requires injecting a cold ion sample in the time-of-flight analyzer, typically achieved with dedicated linear RF traps, generating ion bunches of less than five nanoseconds width [33, 35, 36]. Figure 2 shows the conceptual idea of an MR-TOF-MS.

As discussed in the introduction, MR-TOF-MS offers several advantages over traditional mass spectrometry techniques. Its ability to provide high-resolution, accurate mass measurements with the highest sensitivity, fast measurement times, and applicability to all production methods for short-lived nuclei makes it an invaluable tool in nuclear physics and the studies of nuclear isomers. Therefore, it has been implemented in many major research laboratories worldwide. The systems operate at facilities using relativistic ion beams for exotic nuclei production, including the ISOL facilities ISOLTRAP [30] and TITAN [37–39] and in-flight facilities FRS Ion Catcher [35, 40, 41] and RIBF [42]. Also, MR-TOF-MS is used for species produced in reactions at the coulomb barrier, including fusion [43] and multinucleon transfer reactions [44]. Moreover, they are used for spontaneous fission products, where, besides mass measurements [45], isobar separation [46] and fission yield [47] measurements are also done.

Figure 3 shows a histogram of the mass-resolving power needed to separate the ground and isomeric states of all known isomers [1] with a half-life longer than ten milliseconds. To be used for general nuclear isomer studies, only systems capable of resolving the majority of known isomers are considered. As seen in Fig. 3, a mass-resolving power larger than 500,000 is needed. These systems achieving this are marked in the figure and are discussed in the next section. The mass-resolving power expected from a next-generation MR-TOF-MS is expected to be higher than a million [48], allowing us to measure more than 90% of all known nuclear isomers.

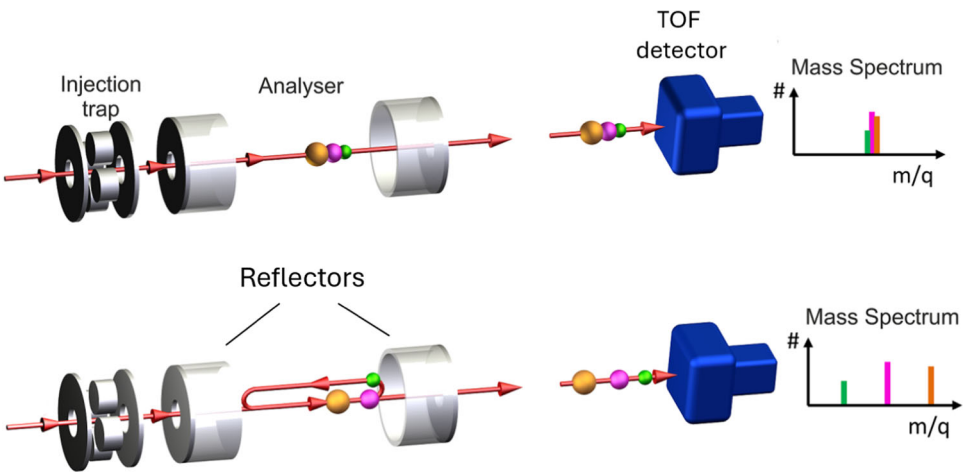

Fig. 2 Top: the linear time-of-flight (Linear-TOF) analyzer with limited ion path length and low mass-resolving power. Bottom: the MR-TOF-MS with maximized ion path length and high mass-resolving power

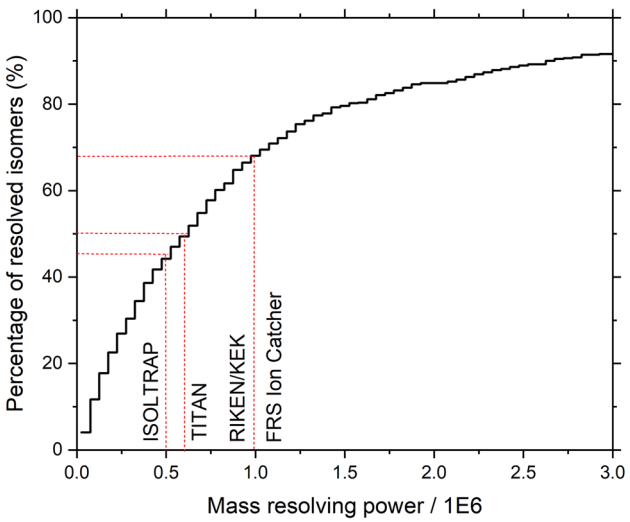

Fig. 3 The mass-resolving power needed for resolving isomers with half-lives longer than 10 ms from the ground state according to the atlas of isomers-second edition [1]. Different systems are marked in the figure RIKEN/KEK [49], ISOLTRAP [50], TITAN [51], and FRS-IC [52]. Future systems are expected to have mass-resolving powers up to three million [48]

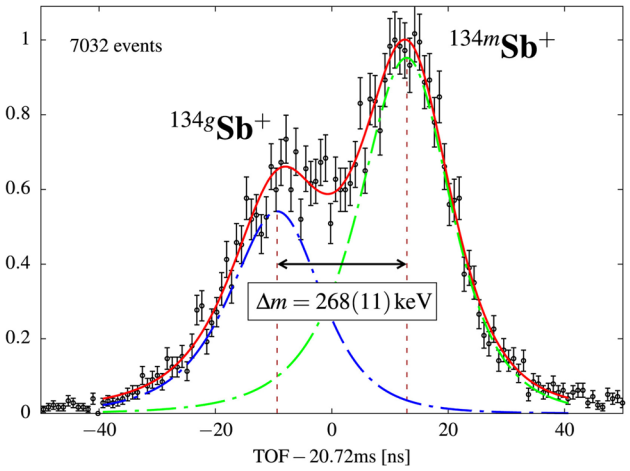

Fig. 4 Simultaneous ground and isomeric state mass measurement of ^{134g,m}Sb by achieving recently developed high mass-resolving power at RIKEN's MR-TOF-MS. Source: reprinted figure with permission from [49]

3 MR-TOF-MS for isomer studies

3.1 RIKEN

At RIKEN, Wako, Japan, several MR-TOF-MS are actively utilized for various purposes. These systems play a crucial role in studying nuclei produced through different processes, including in-flight fragmentation and fission [53], fusion reactions [54], and multinucleon transfer reactions [44]. Despite their utility, there have been instances where the mass-resolving power of these systems proved insufficient to distinguish low-lying isomers, such as ^{58m}Mn [55] or isotopes like ^{111}Pd, ^{112}Rh, and ^{111}Mo [56]. To address this challenge, the research group at RIKEN has continuously enhanced the mass-resolving power of their MR-TOF-MS systems, achieving an impressive milestone of one million mass-resolving power in offline measurements in a short measurement time of about 20 milliseconds [49]. This significant advancement has been particularly beneficial in resolving isomeric states, exemplified by their successful demonstration with the isomeric state in ^{134}Sb (refer to Fig. 4).

Moreover, alongside improvements in mass-resolving power, the group has pioneered the development of a novel time-of-flight detector known as alpha-TOF [57]. This innovative detector integrates time-of-flight measurement with the measurement of decay energy, opening up new avenues of research. The alpha-TOF system has already been deployed in initial experiments for super-heavy element research [58], and its potential, along with recent findings in this research domain, is extensively discussed in Ref. [59]. This development holds promise for extending the reach to even lower lying isomers, provided that the decay properties of these isomers allow for such investigations.

3.2 ISOLTRAP

The ISOLTRAP experiment, situated at the ISOLDE facility at CERN (Geneva, Switzerland), represents a pioneering effort in nuclear physics to produce and study the properties of exotic nuclei. By combining the capabilities of both MR-TOF-MS [30] and Penning trap setups [60, 61], the ISOLTRAP experiment achieves remarkable precision in its measurements [62].

The experiment relies on the so-called isotope separator on line, ISOL method, where a high energy proton beam is directed at a target. The reaction products are stopped within the target, defuse out, and are ionized by various methods. This methodology allows for the isolation and subsequent study of specific short-lived (sub-seconds) isotopes.

The MR-TOF-MS functions both as a standalone mass spectrometer and a beam purification apparatus for downstream analysis in the Penning trap. Recent advancements in the mass-resolving power of the MR-TOF-MS have opened up new avenues of exploration, facilitating groundbreaking discoveries such as the identification of an isomeric state within ^{99}In [50]. This finding, depicted in Fig. 5, underscores the importance of technological advancements in pushing the boundaries of nuclear science.

ISOLTRAP has provided the detailed properties of the isomer in ^{79}Zn, thereby enriching our understanding of the nuclear landscape surrounding the double magic nucleus ^{78}Ni [63].

Moreover, the precision afforded by the MR-TOF-MS extends beyond direct mass measurements of isomeric states, serving as a versatile tool for other research endeavors. Notably, the system's low-background detection capabilities enable MR-TOF-MS-assisted laser spectroscopy of neutron-deficient gold isotopes, revealing the abrupt deformations observed in these nuclei [64].

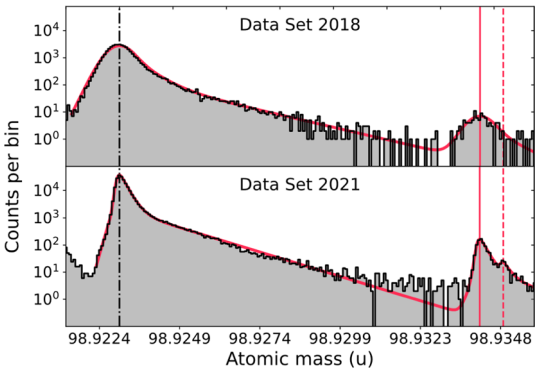

Fig. 5 The discovery of ^{99m}In isomer with recent upgrade in ISOLTRAP's MR-TOF-MS mass-resolving power in 2021 [50]. The issue was not resolved in the 2018 campaign, where the measurement was performed with a lower mass resolution. The red solid and dashed lines show the ground and isomeric state of ^{99g,m}In, respectively. Source: reprinted figure with permission from [50]

3.3 TITAN

Situated within the TRIUMF facility in Vancouver, Canada, the TITAN (TRIUMF's Ion Trap for Atomic and Nuclear science) facility [29] occupies a pivotal position at ISAC (Isotope Separator and Accelerator Complex) [65], boasting the highest beam power on target among ISOL (Isotope Separation On-Line) facilities worldwide. TITAN comprises four distinct ion traps for conducting mass and in-trap decay spectroscopy analyses on exotic nuclei.

First, the ions from ISAC are processed with the RFQ Cooler and Buncher [66], adept at preparing a thermalized bunch of ions while efficiently distributing them downstream to three other traps: the MR-TOF-MS [37], the Electron Beam Ion Trap (EBIT) [67], and the Measurement Penning Trap (MPET) [68].

The MR-TOF-MS facilitates direct mass determination and beam purification, both as its own isobar separator and for the downstream traps. The first (and second overall) identification and excitation energy determination of an isomeric state at TITAN was done for ^{84m}Rb [38]. Subsequently, the MR-TOF-MS has been instrumental in discovering and measuring numerous other isomeric states across various regions of the nuclear chart.

Notably, measurements of ten isomeric states within the $^{127-133}$In isotopes have expanded our understanding of nuclear structure[69]. Furthermore, the discovery of the isomer ^{69m}Fe represents a notable milestone, achieving a mass-resolving power of 600,000 [51]. The MR-TOF-MS has also been pivotal in elucidating nuclear structure phenomena, such as the origin of the stability of $h_{11/2}$ isomeric states and its excitation energy within the $N = 81$ isotonic chain [70].

Figure 6 illustrates this unique nuclear structure phenomenon, showcasing isomeric states within the $N = 81$ isotonic chain with equivalent excitation energies. Through the examination of ^{151m}Yb's excitation energy, the persistence of this effect at the proton dripline has been established, complemented by accurate modeling of the underlying nuclear structure via mean-field calculations, as depicted in Fig. 6. Furthermore, this investigation led to the discovery of a new isotope, ^{150}Yb, utilizing the MR-TOF-MS as its own isobar separator [71]. This feat was made possible by implementing mass selective retrapping [72, 73], significantly enhancing background suppression by more than four orders of magnitude and enabling the exploration of exotic isotopes one or two neutrons closer to the driplines. This methodological advancement proves invaluable at ISOL facilities like ISAC, where many experiments are background limited.

The unique amalgamation of resolving power, sensitivity, and high production yields renders TITAN an exceptionally captivating experiment for probing exotic isomeric states and advancing our understanding of nuclear physics.

3.4 FRS Ion Catcher

The FRS Ion Catcher (FRS-IC) [41] at the GSI facility in Darmstadt, Germany, represents remarkable mass-resolving power (1,000,000) and high-precision mass measurement capabilities ($1.7 \cdot 10^{-8}$) [52]. This system, which integrates an MR-TOF-MS [35] with a cryogenic stopping cell (CSC) [74, 75], serves as a versatile platform for studying exotic nuclei generated through various mechanisms. One primary function of the FRS-IC is the investigation of exotic nuclei produced via in-flight fragmentation and fission occurring within the FRS target [52, 76]. In addition, the system facilitates the study of spontaneous fission products from a ^{252}Cf source installed inside CSC [45], as well as isotopes produced via MNT reactions by positioning a secondary target inside the CSC

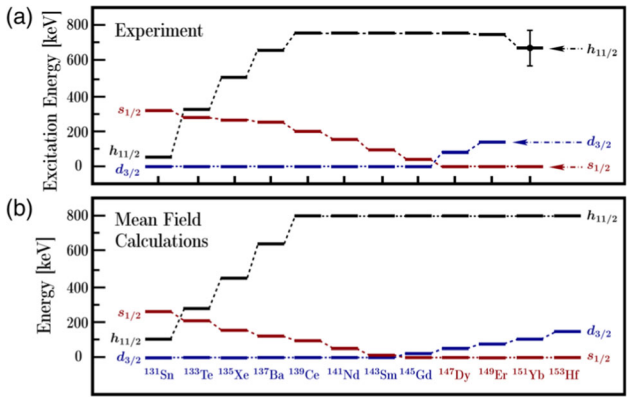

Fig. 6 **a** The measured excitation energies of isomers in the even-Z $N = 81$ isotones ranging from Sn to Yb. The determination for ^{151m}Yb was done with an MR-TOF-MS. The consistent excitation energy of $h_{11/2}$ from Ce to Yb is noteworthy. **b** Corresponding mean-field calculations utilizing a universal parametrization of the Woods–Saxon Hamiltonian. Across the range from Nd ($Z = 60$) to Hf ($Z = 72$), the filled proton levels exhibit substantial degeneration, resulting in the stability of the excitation energy. Source: reprinted figure with permission from [70]

[77]. A pioneering achievement was the first identification and excitation energy determination of an isomeric state (^{211m}Po isomeric state at 1572 keV) with an MR-TOF-MS [78]. The MR-TOF-MS also plays a crucial role in identifying and characterizing isomeric states of nuclei near the neutron-proton ($N = Z$) line. The studies include the first discovery of an isomeric state in ^{97m}Ag [79]as well as precise measurements of excitation energies for ^{101m}In [79] and ^{94m}Rh [80] isomers. The high beam energy at the FRS makes the system uniquely suitable for heavy isotopes.

The MR-TOF-MS with fast measurement cycles is also used for isomeric fission yield measurements from ^{252}Cf source [47]. The method allows for simultaneous measurement of isomeric and ground states and, thus, the yield ratio determination in the fission mechanism.

In addition, the MR-TOF-MS of the FRS Ion Catcher serves as a highly efficient separator, owing to its exceptional mass-resolving power and the implementation of a fast-switching Bradbury–Nielsen Gate (BNG) [40]. This capability was demonstrated through the spatial separation and isolation of an isomeric beam, exemplified by the isolation of ^{211m}Po [78], see Fig. 7. The identification of isolated isotopes is achieved through alpha spectroscopy, utilizing a Si-detector positioned behind the BNG. Thus, the FRS-IC system can produce isomerically clean beams with high-rate capability and short half-lives from various production methods. This enables background-free precision studies and presents a substantial discovery potential for new isomers and related phenomena in nuclear physics research.

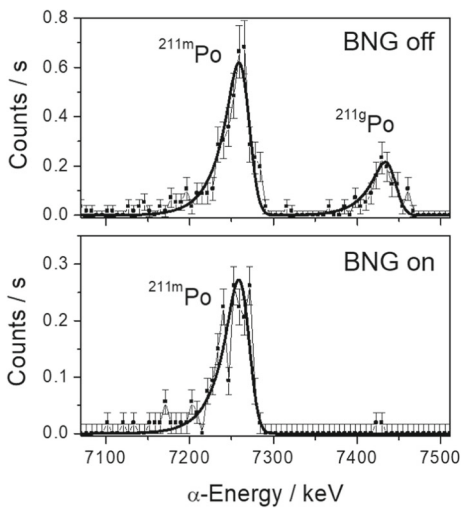

Fig. 7 First spatial separation of isomeric beam with MR-TOF-MS. Alpha spectroscopy identifies ^{211}Po in its ground and isomeric state. Using the fast-switching BNG gate coupled to FRS-IC's MR-TOF-MS, the separation of ground and isomeric state is achieved [78]

4 Summary and outlook

Mass spectrometry has evolved into a versatile tool for probing nuclear structure intricacies, including isomer detection and characterization. It offers advantages over other methods for identifying isomeric states, such as decay information analysis, kinematic properties examination, and property measurement like laser spectroscopy. Mass spectrometry, particularly MR-TOF-MS, presents significant advantages in sensitivity, speed, and applicability to all isomer production methods.

Several research facilities worldwide utilize MR-TOF-MS for isomer studies. At RIKEN (Japan), ISOLTRAP (CERN, Switzerland), TITAN (TRIUMF, Canada), and FRS Ion Catcher (GSI, Germany), MR-TOF-MS systems are employed for high-precision mass measurements, isomer detection, and nuclear structure studies. These facilities have achieved breakthroughs in resolving power, sensitivity, and measurement time, enabling the discovery and precise characterization of various isomeric states and contributing significantly to nuclear physics research. This emphasizes the importance of MR-TOF-MS in advancing our understanding of nuclear isomers and highlights ongoing efforts to enhance sensitivity and measurement capabilities, promising further breakthroughs in this field.

Future investigations of isomers with MR-TOF-MS include studies of fission isomers [81], the search for new exotic phenomena [47], decay studies of isomerically clean beams, and the discovery of many isomeric states, see Fig. 1.

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