



First spatial separation of a heavy ion isomeric beam with a multiple-reflection time-of-flight mass spectrometer



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ABSTRACT

²¹¹Po ions in the ground and isomeric states were produced via ²³⁸U projectile fragmentation at 1000 MeV/u. The ²¹¹Po ions were spatially separated in flight from the primary beam and other reaction products by the fragment separator FRS. The ions were energy-bunched, slowed-down and thermalized in a gas-filled cryogenic stopping cell (CSC). They were then extracted from the CSC and injected into a high-resolution multiple-reflection time-of-flight mass spectrometer (MR-TOF-MS). The excitation energy of the isomer and, for the first time, the isomeric-to-ground state ratio were determined from the measured mass spectrum. In the subsequent experimental step, the isomers were spatially separated from the ions in the ground state by an ion deflector and finally collected with a silicon detector for decay spectroscopy. This pioneering experimental result opens up unique perspectives for isomer-resolved studies. With this versatile experimental method new isomers with half-lives longer than a few milliseconds can be discovered and their decay properties can be measured with highest sensitivity and selectivity. These experiments can be extended to studies with isomeric beams in nuclear reactions.

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1. Introduction

Isomers are excited metastable states of nuclei with relatively long half-lives, due to hindrance in their decay modes [1]. The interest in isomers ranges from nuclear structure [2] and nuclear astrophysics [3] to their possible use for energy storage devices [1].

So far, experiments with isomers mostly have been performed using gamma-ray spectroscopy techniques [4–7]. Isomers with half-lives in the μ s range and below are especially well suited for coincidence measurements in gamma-ray spectroscopy. In complement to this, long-lived isomers can be well investigated by high-resolution mass spectrometry. Furthermore, high-accuracy mass

measurements rely on the fact that ions in the ground and isomeric states are clearly identified and resolved in the mass spectrum, otherwise the results can be erratic. This requirement is a permanent strong motivation to increase the experimental mass resolving power.

The first resolution of ground and isomeric states in a mass measurement was achieved with the ISOLTRAP Penning trap [8]. At LEBIT, the first discovery of an isomer with such a device was achieved [9] and a measurement of an isomeric-to-ground state ratio was performed at JYFLTRAP [10]. By use of ions in higher charge-states in a Penning trap, the resolution of ground and isomeric states could be increased at the TITAN facility [11]. Isomers have also been observed and resolved from their ground state in mass measurements of projectile fragments stored in the Experimental Storage Ring ESR, using Schottky Mass Spectrometry (SMS) [12] and Isochronous Mass Spectrometry (IMS) [13], and

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their decay has been studied. In storage-ring experiments, the ions are mainly fully ionized or carry only a few electrons, whereas in traps the measured ions are typically singly or doubly charged. Therefore, storage ring and ion trap experiments are highly complementary, because atomic charge states can have a strong effect on the nuclear decay properties of isomers, see [14] and references therein.

Experimental methods employing pure isomeric beams can reveal new nuclear properties. Such experiments can be isomer-resolved decay spectroscopy or investigations via nuclear reactions with isomers. There are different ways to produce pure isomeric beams. The first nuclear reactions with a pure isomeric beam were performed at ISOLDE by selection of the isomer with laser resonance ionization and subsequent post-acceleration [15]. Using a novel excitation method for ion motion in a Penning trap, isomerically clean ion samples could be separated [16] and gamma-ray spectroscopy of pure nuclear states was performed at JYFLTRAP [17]. It was also shown that pure isomeric beams may be provided using storage rings [18,19]. However, these methods are limited to isomers with half-lives of one second or even longer. For the special case, where the half-life of the isomer is significantly longer than the corresponding ground state, such as in ^{212}Po , where the isomeric state has a half-life of 45 s, but the half-life of the ground state is 300 ns only, one does not need an experimental method for separation; one can simply wait until the ions in the ground state have decayed.

Multiple-reflection time-of-flight mass spectrometers (MR-TOF-MS) [20,21] have recently been developed into powerful tools for research with exotic nuclei [22–24]. They are mass spectrometers, and in combination with a Bradbury–Nielsen Gate (BNG) [25,26], a fast electrostatic ion deflector, they can also be employed as high-resolution spatial mass separators [27]. Because of their high mass resolving power, measurement speed, sensitivity and broadband characteristics, they have recently been proposed for efficient experiments with isomers [21]. In this work, such pioneering measurements have been exemplarily performed with ^{211}Po ions for the first time.

2. Experiment and data analysis

The experiment was performed with the FRS Ion Catcher setup [23] at GSI. ^{211}Po nuclei in ground and isomeric states were produced with the FRS [28] via projectile fragmentation of a ^{238}U primary beam at an energy of 1000 MeV/u in a beryllium target with an areal density of 1.629 g/cm², that was followed by a 0.223 g/cm² Nb backing. The fully ionized polonium ions were separated in flight from the primary beam and other reaction products and were energy-bunched using two-fold magnetic rigidity analysis and a 4.063 g/cm² Al monoenergetic degrader, located at the central focal plane of the FRS (Fig. 1, upper panel). The isotope identification of the ions was performed using the particle detectors of the FRS. After further slowing-down in a homogenous degrader at the final focal plane, the ions were injected into a gas-filled cryogenic stopping cell (CSC) [29–31]. In the CSC, they were thermalized as singly-charged ions in helium gas at a pressure of 95 mbar and a temperature of 86 K, corresponding to an areal density of 5.6 mg/cm². They were guided to the exit side of the CSC by electric fields, radially focussed onto an extraction nozzle by an RF carpet and extracted with the gas flow into an RFQ quadrupole beam line [23].

From there, the ions were transmitted to the MR-TOF-MS [27, 32,33] (Fig. 1(a)–(c)). The MR-TOF-MS is a powerful and universal mass spectrometer with single-ion sensitivity as well as a spatial mass separator. Mass resolving powers (FWHM) as high as 600,000 and a mass accuracy of 10^{-7} have been achieved off-line [32].

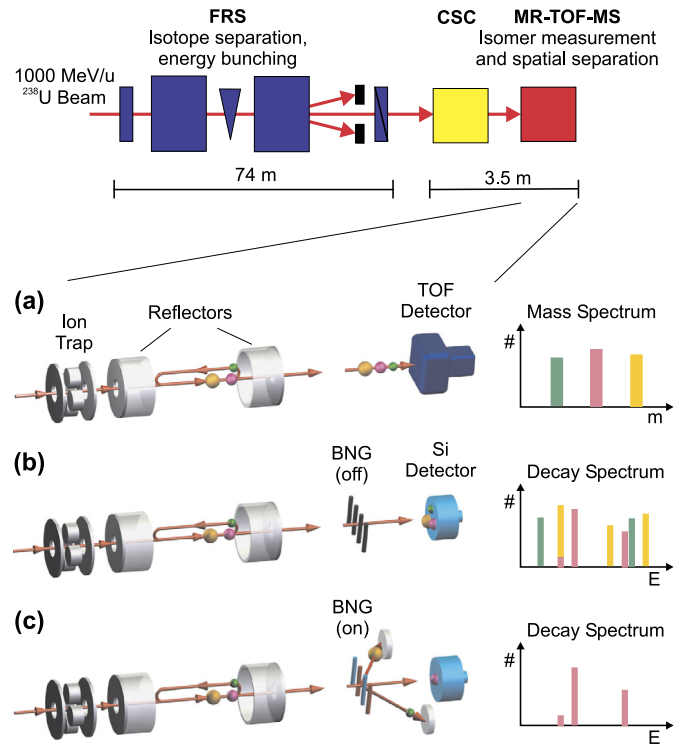


Fig. 1. Schematic drawing of the experimental setup (upper panel). The spatial isotope separation of ions and their energy bunching and slowing down were performed with the FRS before the ions were injected into the gas-filled cryogenic stopping cell (CSC). (a) Mass measurement of the extracted ions with the MR-TOF-MS. The time-of-flight is measured with a TOF detector after multiple reflections between two electrostatic reflectors. (b) Without voltages on the BNG, all ions are transmitted to the Si decay detector behind the gate. (c) With proper timing of the voltages applied on the BNG, only the selected isomer ions are transmitted and their decay is measured.

In the present experiment, a maximum mass resolving power of 370,000 has been obtained for ions with a mass of 133 u after 430 turns in the analyzer, corresponding to a time-of-flight of only 15.3 ms. In the MR-TOF-MS, the ions were accumulated, cooled by collisions with a helium buffer gas in an RF trap system and injected as bunches into the time-of-flight analyzer with a kinetic energy of 1.3 keV. The analyzer is formed by two electrostatic reflectors, between which the ions traveled for 192 turns, corresponding to a time-of-flight of 8.7 ms and a total path length of about 300 m. During their storage in the analyzer, the ions were dispersed in time according to their mass-to-charge ratios. After ejection from the analyzer, the ions alternatively (i) impinged on a TOF detector (isochronous secondary electron multiplier) for measurement of their time-of-flight and hence their mass-to-charge ratio (mass measurement mode) or (ii) passed through the BNG, in which unwanted ions can be deflected, and only the ions of interest were transmitted (mass separator mode). These latter ions were implanted into a Si detector mounted behind the BNG. In the Si detector their alpha decay pattern was measured. The TOF detector and the BNG with the Si detector are mounted on a remote-controlled platform, which can be moved such that either the TOF detector or the BNG and the Si detector are in the beam line.

A mass spectrum of ^{211g}Po and ^{211m}Po was recorded by the TOF detector for a duration of 500 s. The detector signal was digitized by a time-to-digital converter. Next, the BNG and the Si detector were moved into the ion beam. Energy spectra from the alpha decay of ^{211g}Po and ^{211m}Po ions were acquired by the Si detector with the BNG voltages turned off for transmission of all ions, as well with the BNG voltages turned on for a duration chosen such

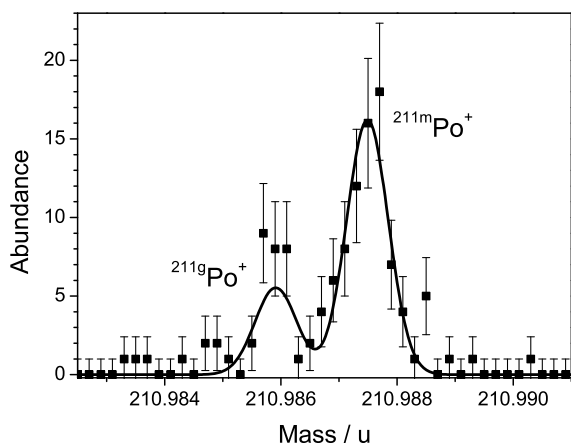


Fig. 2. Mass spectrum of ^{211}Po ions in the ground and isomeric states measured with the MR-TOF-MS. The curve is a least squares fit of two Gaussian functions to the data. The excitation energy determined from the spectrum amounts to (1472 ± 120) keV.

that the ^{211}gPo ions were deflected and only the ^{211}mPo ions were allowed to reach the Si detector. The acquisition time amounted to 300 s each. The Si detector signals were amplified and digitized by an analog-to-digital converter.

The time-of-flight spectra acquired in the mass measurement mode were calibrated with ^{211}Pb ions obtained by the consecutive decay from a ^{223}Ra source mounted in the CSC. The calibration spectrum was acquired before the measurement of ^{211}Po . Since the mass distributions of ^{211}mPo and ^{211}Pb ions would overlap, the transport of ions from the ^{223}Ra source was disabled for the duration of the measurement of ^{211}Po by applying a negative voltage relative to the surrounding electrodes of the source. In this mode, the amount of ^{211}Pb ions extracted from the cell is only about 10% of the ^{211}Po ions. The energy spectra from the alpha decays were calibrated with measurements of the emitted alpha particles from ^{211}Pb and ^{219}Rn from the same source.

3. Results and discussion

Fig. 2 shows the mass spectrum obtained with the MR-TOF-MS in the mass measurement mode. Two clearly resolved mass peaks are observed. The mass resolving power (FWHM) amounts to about 250,000. In total, there are 123 ions recorded in the mass spectrum. The two mass peaks show tails, which are due to the particular tuning of the voltages of the MR-TOF-MS. The measured data were fitted with two Gaussian functions. Based on the mass excess determined from both distributions, the ions can be identified in comparison with the literature values of the mass excess of the ground state [34] and the excitation energy of the isomeric state [35] as $^{211}\text{gPo}^+$ ($9/2^+$, half-life $T_{1/2} = 516$ ms) and $^{211}\text{mPo}^+$ ($25/2^+$, $T_{1/2} = 25.2$ s). The exact results for the measured mass excess values in the present experiment and the respective uncertainties require a more extensive data analysis procedure and will be the subject of a forthcoming publication. However, the excitation energy, i.e., the mass difference, of the isomeric state could be determined for the first time using mass spectrometry; it is (1472 ± 120) keV, which is in good agreement with the literature value of (1462 ± 5) keV [35]. The literature value is derived from the measured alpha decay energies of both states to the daughter ^{207}Pb . Two more isomeric states are known in ^{211}Po nuclei ($31/2^-$, $T_{1/2} = 243$ ns and $43/2^+$, $T_{1/2} = 2.8$ μs) [35]. However, these states do not survive the extraction time from the CSC and the measurement time in the MR-TOF-MS and are therefore not observed in our mass spectrum. From the mass spectrum, the ra-

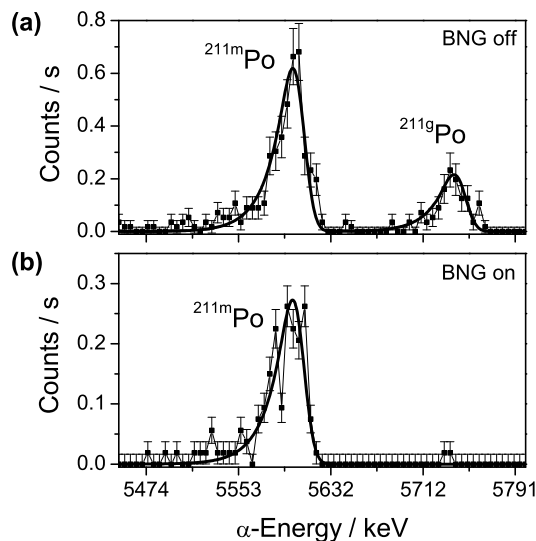


Fig. 3. Spatial separation of ground and isomeric states of ^{211}Po by use of the BNG in the MR-TOF-MS. Energy spectra of the main alpha decay branches are shown, which were measured with the Si detector behind the gate with the voltages on the BNG off (a) and with the voltages on the BNG pulsed for transmission of the isomeric state only (b).

tio of ions in the isomeric state with respect to the ground state has been determined. It amounts to (2.5 ± 0.8) . This value includes a correction for decay losses during separation, thermalization and measurement by a factor of 0.84. The isomeric ratio is specific to the production mechanism. The isomeric ratio for the $43/2^+$ state in ^{211}Po produced by projectile fragmentation at 1000 MeV/u was measured recently using gamma ray spectroscopy [36]. However, because of its long lifetime, the $25/2^+$ state could not be measured.

In the mass separator mode, the energy spectra from the alpha decay of ^{211}gPo and ^{211}mPo were measured with the Si detector behind the BNG. **Fig. 3** shows the section of the spectra with the two dominant alpha-decay peaks with (a) the BNG turned off, i.e. with transmission of all ions, and with (b) the BNG voltages turned on for a period chosen such that the ^{211}gPo ions were deflected. The measured data were fitted with a Gaussian convoluted with an exponential tail on the slope of the low-energy side [37]. The alpha particle energies in the spectrum (a) amount to (7269 ± 15) keV and (7442 ± 15) keV for ^{211}mPo and ^{211}gPo , respectively. These values are in good agreement with the literature values of (7275 ± 15) keV and (7450.3 ± 0.5) keV [38], respectively. The measurement of the alpha-particle energies serves as an independent identification of the ions, in addition to the identification with the FRS detectors and by use of mass spectrometry with the MR-TOF-MS. The spectrum (b) shows that with the BNG operated as an ion deflector, ions in the isomeric state could be spatially separated and transmitted as a beam of pure isomers to the Si detector. In this experiment, a pure isomeric beam was achieved for the first time using an MR-TOF-MS. Only two counts of ground state ions were transmitted and detected in this mode. In the future, this can be completely avoided by an optimization of the MR-TOF-MS settings in order to improve the peak shape or by a further increase of the mass resolving power. Even at the present performance level, a suppression ratio of at least four orders of magnitude can be achieved, albeit at larger binding energy differences [32]. Therefore, even weakly populated states can be investigated and separated. Note, that for a given excitation energy the required mass resolving power increases linearly with mass. Furthermore, the time needed to obtain a required mass resolving power also increases linearly with mass for most high-resolution

mass spectrometers. For MR-TOF-MS, the required time-of-flight only increases with the square-root of the mass. Thus, the successful separation of an isomer in the mass region beyond 200 u within a time-of-flight of 8.7 ms only is an important achievement.

4. Perspectives

These pioneering measurements demonstrate the potential of MR-TOF-MS for isomer research. MR-TOF-MS are fast mass spectrometers with mass resolving powers in excess of several 10^5 , i.e. the capability to resolve essentially all nuclear isobars as well as isomers with excitation energies of several hundred keV or more. As non-scanning mass spectrometers, they can perform measurements, which cover several mass units, and during which all ion species are measured simultaneously. They can therefore very efficiently be used to search for hitherto unknown isomeric states with life times down to a few milliseconds. The discovery of isomers, identification, determination of the excitation energy and the determination of the isomeric ratio can result from the same measurement. Furthermore, measurements with MR-TOF-MS are complementary to gamma ray spectroscopy experiments, which are limited to half-lives below one millisecond without loss of the correlated particle identification.

Further research potential arises from the use of MR-TOF-MS in combination with a BNG as isomer separators, which supply pure isomeric beams. At variance to laser-ionization methods, the MR-TOF-MS mass separator can be employed universally, i.e. is applicable to all elements. The reach is limited only due to the cycle time of a few milliseconds. Experiments that are envisaged include decay measurements of isomerically separated ions, e.g. to investigate also weakly populated decay channels of isomers. The search for new fission isomers [39,40] and a direct determination of the excitation energy of known fission isomeric states could also be performed. So far, only one fission isomer with a half-life of several milliseconds has been discovered, i.e. in ^{242}Am with a half-life of 14 ms; this is accessible by the MR-TOF-MS technique. Furthermore, the post-acceleration of isomeric beams separated with an MR-TOF-MS and reactions with isomeric beams are now on the horizon.

In the present experiment, projectile fragmentation at relativistic energies was used, which gives access to all nuclides up to the heaviest projectiles. A major advantage of in-flight separation at high velocities is the unambiguous particle identification and spatial isotopic separation. By employing fusion and transfer reactions, isomers almost up to the heaviest known elements can be produced. At these low energies, direct particle identification can be performed by the MR-TOF-MS only. Furthermore, MR-TOF-MS can be applied at ISOL or IGISOL facilities as well. Thus, all nuclides on the nuclear chart can be accessed. Efficient large-scale measurements of isomers are within reach, as performed recently by SMS with electron-cooled beams at the FRS-ESR facility at GSI [41]. However with the MR-TOF-MS technique the half-life limits will be lower by three orders of magnitude than presently with SMS.

5. Summary

Ions of the nuclide ^{211}Po in the ground and isomeric states were produced by projectile fragmentation at 1000 MeV/u and separated in flight with the FRS. ^{211g}Po and ^{211m}Po ions were resolved in the mass spectrum measured with the MR-TOF-MS. The excitation energy of the isomer and the isomeric-to-ground state ratio were determined by mass spectrometry. The measured value of the excitation energy is in good agreement with the literature value. The isomeric-to-ground state ratio has been determined in

this experiment for the first time. The separation of a pure isomeric beam using an MR-TOF-MS represents a novel experimental achievement. These pioneering measurements demonstrate the great potential of the MR-TOF-MS combined with an ion deflector (BNG) for research with isomers, including large-scale searches for new isomers and decay and reaction experiments with pure isomer beams.

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