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Full Length Article

A laser ablation carbon cluster ion source for the FRS Ion Catcher

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ABSTRACT

A Laser Ablation Carbon Cluster Ion source (LACCI) has been developed and commissioned at the FRS Ion Catcher at GSI. It is the first laser ablation ion source for long-term measurements with a multiple-reflection time-of-flight mass spectrometer (MR-TOF-MS). This is enabled by two key developments: (i) a two-dimensional movable target platform that ensures the long-term stable production of ions and (ii) a mass filter for the isolation of the ions of interest. LACCI is capable of providing closely-spaced reference ions in the mass range up to ∼300 u for accurate mass measurements of exotic nuclei (relative mass uncertainty ∼ 10^{-8}) and systematic studies of the mass uncertainties with the multiple-reflection time-of-flight mass spectrometer. Investigations of the laser energy and repetition rate and long-term measurements have been carried out with carbon targets (Sigradur®, Fullerene) and metallic targets (silver, gold, copper, tungsten, and erbium). In a test experiment, LACCI has delivered a stable ion current for more than 20 h at a laser repetition rate of 100 Hz, corresponding to an operation for one week at 10 Hz. It thus fully satisfies the requirements for long-term accurate mass measurements of exotic nuclei with an MR-TOF-MS. This enables the access of exotic ions, detected with rates as low as 0.1 per hour at the FRS Ion Catcher.

1. Introduction

Masses of exotic nuclei [1] are key properties for understanding the evolution of nuclear structure [2] and stellar nucleosynthesis [3,4]. Multiple-Reflection Time-Of-Flight Mass Spectrometers (MR-TOF-MS) have been developed to accurately determine masses of short-lived exotic nuclei [5]; they are used at accelerator facilities worldwide [5–9]. At the FRS Ion Catcher (FRS-IC) [10] at the GSI Helmholtz Centre for Heavy Ion Research in Darmstadt, Germany, high-accuracy experiments with exotic nuclei can be performed, which have been produced by projectile fragmentation or fission at relativistic energies and separated at the fragment separator FRS [11]. The nuclei are stopped and thermalized in a Cryogenic Stopping Cell (CSC), extracted and transported by a Radio-Frequency Quadrupole (RFQ) beamline to the MR-TOF-MS [5,8], where their masses can be measured at a mass resolving power up to ∼1,000,000 (FWHM) and a relative mass uncertainty down to a few 10^{-8} [12].

For high-precision mass measurements of exotic nuclei across the chart of the nuclides with an MR-TOF-MS, the best reference ion is an isobar to the ion of interest since isobars experience almost the same conditions, e.g., switching electric fields in the trap and the analyzer of the MR-TOF-MS, such that systematic errors are minimized. Details on calculating the uncertainties and their origin are presented in [13]. Therefore, reference ions at every mass number up to a mass of about 300 u are desirable. Carbon clusters, enriched with ^{13}C, are well suited for this purpose [14]. The atomic mass unit u is 1/12 of ^{12}C. Therefore, carbon cluster ions have a negligible mass uncertainty, giving them an inherent advantage as reference ions. Carbon clusters are well studied and understood [15–17], including their structure and abundance as well as their dissociation energies, which are in the order of a few eV [15] only. This corresponds to a relative mass uncertainty in the 10^{-10} range, which is still two orders of magnitude beyond the highest yet achieved relative accuracy of an MR-TOF-MS [12] and can be neglected for most applications. In addition, carbon clusters can be easily produced by laser ablation over most of the relevant mass range for mass measurements in nuclear physics.

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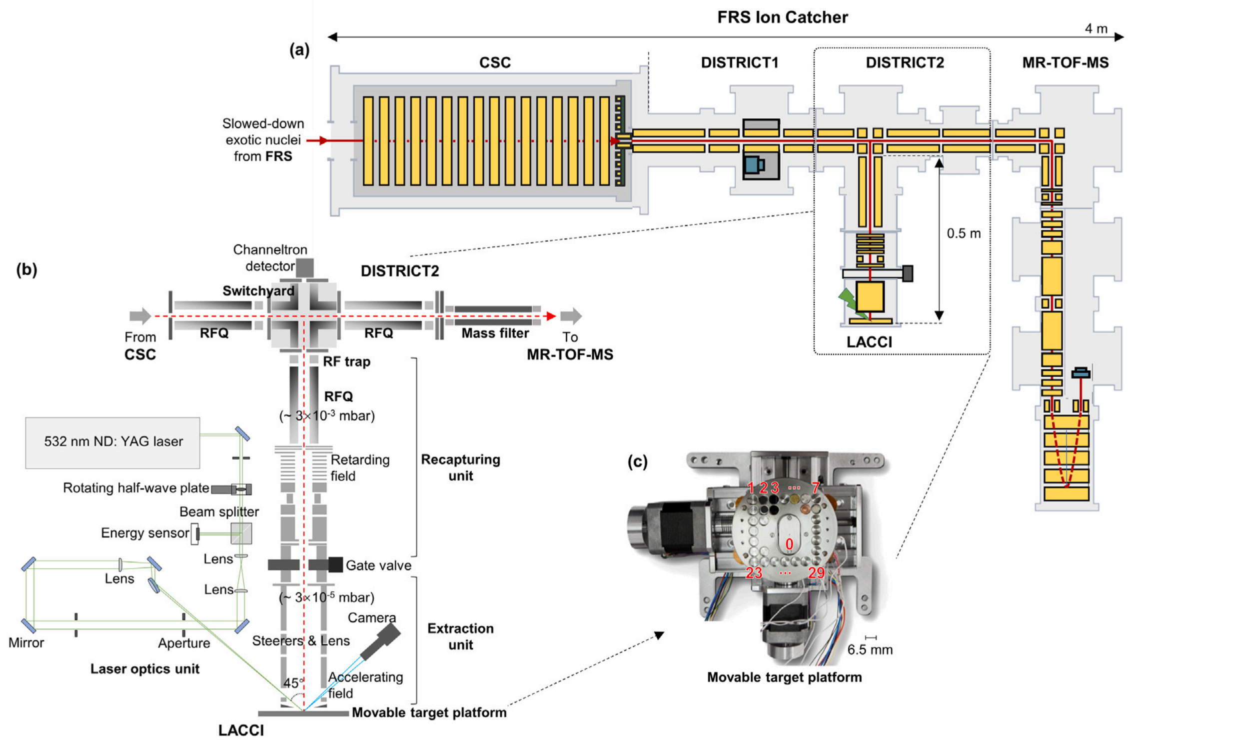

Fig. 1. (a) Schematic view of the FRS Ion Catcher setup. The secondary ion beam produced and separated at the fragment separator FRS is slowed down and thermalized in the CSC. The thermalized ions are extracted from the CSC and transmitted through the RFQ beamline (DISTRICT1 and DISTRICT2) to the MR-TOF-MS. (b) Detailed schematic view of LACCI and DISTRICT2, including the laser optics, the ion optics, the RFQ beamline, and a dedicated RFQ mass filter. The ions of interest from the CSC can be merged with the reference ions produced in LACCI via the RFQ switchyard. The mass region of interest is selected in the mass filter and then transmitted to the MR-TOF-MS. (c) Photograph of the 2D movable target platform with 29 target holders marked by numbers. The spot marked as 0 is a small hole with a diameter of 0.5 mm, behind which a thermal ion source providing surface-ionized Cs ions is located.

Laser ablation is a versatile and powerful technique to produce ions from various solid target materials. Laser ablation carbon cluster ion sources have been used for calibration purposes in combination with Penning trap mass spectrometers for over 20 years. The first ion source of this type, explicitly used for the calibration of mass measurements of nuclei, was developed and commissioned at ISOLTRAP/CERN [18, 19]. Since then, laser ablation carbon cluster ion sources have been developed for various Penning trap systems [20–24]. They also enable systematic studies of the mass measurement uncertainties of the device, e.g. the quantification of mass-dependent uncertainties.

However, these ion sources are unsuitable for calibration of MR-TOF-MS because an MR-TOF-MS is operated at cycle frequencies that are two orders of magnitude larger than that of the Penning trap. To provide carbon cluster ions for calibration purposes in an MR-TOF-MS, specific requirements have to be fulfilled: (i) The repetition rate of the ion production cycle, and thus the laser repetition rate, should match the speed of the MR-TOF-MS (cycle frequency 20 to 100 Hz). (ii) A long-term stable operation (∼weeks) has to be ensured even at the highest repetition rate. This is a challenge since the laser creates holes in the target material, making it harder to extract ions and decreasing the ion rate over time. This can be avoided by continuously moving the target relative to the laser so that the laser does not repeatedly hit the same spot. A one-dimensional movement is employed in existing laser ablation carbon cluster ion sources. However, the repetition rate and operation time required for MR-TOF-MS make a two-dimensional movement necessary to use the full target area. (iii) Space-charge effects in the ion trap and the analyzer of the MR-TOF-MS are known to deteriorate the accuracy of mass measurement. For high-accuracy mass measurements, there should not be more than a few (isobaric) ions per mass number per cycle and no more than 100 ions simultaneously in the ion trap [8]. Therefore, only the reference ions of interest should be transmitted to the MR-TOF-MS, and other highly abundant carbon cluster ions should be removed in a mass filter in front of the MR-TOF-MS. The Laser Ablation Carbon Cluster Ion source (LACCI) presented in this paper fully satisfies, for the first time, all these requirements for high-accuracy mass measurements with an MR-TOF-MS with carbon cluster ions as reference ions.

2. Setup

LACCI has been installed in a new beamline module DISTRICT2 (Diagnostics, Ion Sources, TRansport, Isolation, Cooling and Trapping module), which is behind the already existing DISTRICT1 between the CSC and the MR-TOF-MS at the FRS-IC, as shown in Fig. 1(a). In addition to LACCI, DISTRICT2 comprises an extension of the RFQ beamline, an RFQ switchyard [25], and an RFQ mass filter [26]. Integrating the new ion source requires minimal space and intervention to the existing setup. LACCI itself consists of a laser, the laser optics unit, a two-dimensional (2D) movable target platform, an ion extraction unit including a pair of electrostatic steerers, a recapturing unit, an RFQ, and an RF trap located at the end of the RFQ, as shown in Fig. 1(b). They are mounted perpendicular to the already existing beamline. The extraction unit and the recapturing unit can be separated by a gate valve.

A frequency-doubled Nd:YAG laser with a wavelength of 532 nm and a pulse length of 5–10 ns is used. The repetition rate is adjustable or pulse on demand from 1 to 100 Hz, which is ideally suited for the requirements of the MR-TOF-MS. The laser energy can be adjusted from 0 to 500 μJ by rotating a half-wave plate and a polarizing cube beam splitter (Thorlabs: CCM1-PBS25-532). The laser beam is separated in the s- and p-polarized components by reflecting the s-polarized component at the dielectric beam splitter to a laser energy sensor (Coherent Inc.: J-25MB-LE) while allowing the p-polarized component to pass further towards the target of LACCI. The sensor monitors

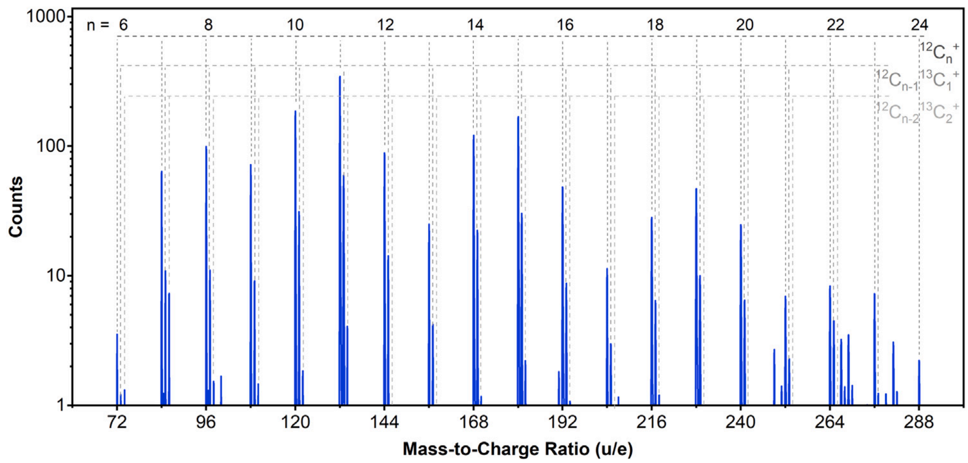

Fig. 2. Mass spectrum of carbon cluster ions produced by LACCI on the Sigradur® target and measured by the MR-TOF-MS. Carbon clusters up to $^{12}C_{24}^+$ with a mass of 288 u were detected and identified. The spectrum is dominated by the carbon cluster ions produced by laser ablation. Unidentified background peaks on the one percent level exist in the spectrum and can come from other means of ionization, such as vacuum gauges. These background ions do not pose a problem in high-precision mass measurements, as they can readily be resolved in the mass spectrum.

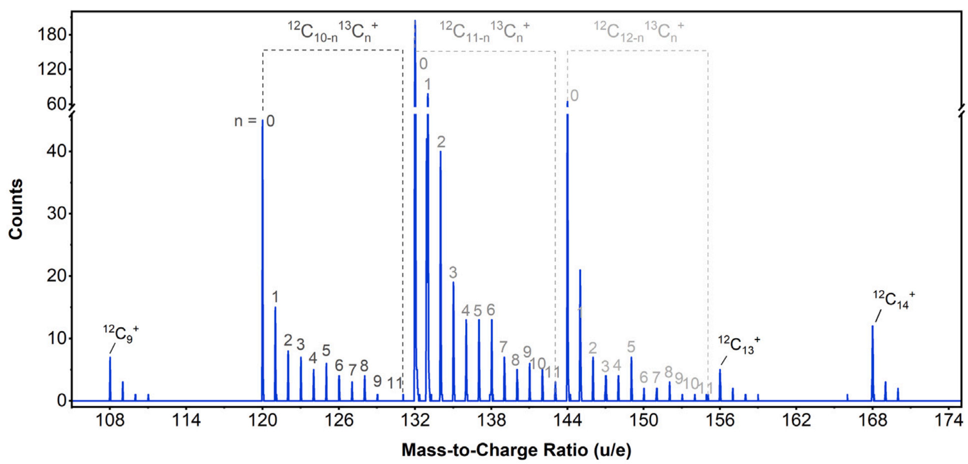

Fig. 3. Mass spectrum of carbon cluster ions produced on the ^{13}C enriched fullerene target. The carbon clusters produced from the enriched target material cover a large mass range, where a reference ion with a well-known mass can be found at quasi-every mass number. The spectrum is dominated by the carbon cluster ions produced by laser ablation. Unidentified background peaks on the one percent level exist in the spectrum and can come from other means of ionization, such as vacuum gauges. These background ions do not pose a problem in high-precision mass measurements, as they can readily be resolved in the mass spectrum.

the energy, and the actual energy reaching the target plate can be calculated accordingly. The laser beam is focused on the target through a series of optical components. The size of the laser beam spot on the target is a few tens of micrometers, which is used to improve the ionization efficiency of different samples. Fig. 1(c) shows a target holder with a 75 mm diameter placed on the 2D movable platform; it includes 29 target sample slots with a diameter of 6.5 mm each. The whole target surface is available for ablation and can be scanned in an automated way with a 2D movement using a scan speed of up to 120 mm/min and step size down to 80 μm. In addition, fast switching (∼1 s) between different targets is possible. In addition, a thermal ion source is mounted on the target platform to provide cesium ions for beamline tuning independent from the laser.

During laser operation, the target is moved continuously, such that the laser spot moves across the target, covering the whole surface in a 2D scan as depicted schematically in the inset of Fig. 8. In this way, the laser spot is prevented from constantly hitting the same spot and thus creating deep holes in the target, out of which ions can be extracted only with reduced efficiency. Hence, the operation time of the ion source without target change can be increased tremendously. An in-house software solution allows one to choose the form of the movement between a horizontal movement (black arrows in the inset of Fig. 8), a vertical movement (gray arrows in the inset of Fig. 8), or a combination with adjustable raster size and speed. In addition, a user-defined sequence of different targets can be chosen. The position of the target platform, the laser spot on the target, and laser energy can be monitored by a camera (Mindvision MV-U38130M) equipped with a filter, which is installed facing the target from the opposite side compared to the incoming laser. The filter is installed in front of the zoom lenses to prevent laser damage to the camera. In addition, the movable target platform is equipped with two pairs of end switches, which allow a reliable initialization of the position.

A Pierce electrode [27] extracts the ions generated by laser ionization and the ions are accelerated by a potential difference of 3 kV. The ion optical design allows the ions produced by LACCI with a large space and velocity distribution to be focused on the recapturing unit. The phase space after the ion production in a laser ablation ion source was characterized in similar systems, e.g., in [28,29]. The ion optical design is also optimized to tolerate the vast amount of neutral particles produced in the laser ablation. Subsequently, the ions are focused and steered into the entrance of the recapturing unit, where they are decelerated, cooled in a buffer-gas-filled RFQ, and stored in a dedicated RF trap. Next, they are guided into the RFQ beamline between the CSC and the MR-TOF-MS via the RFQ switchyard. Here, the ion beam from LACCI can be merged with the ions from the CSC. After the switchyard, the ions pass through a dedicated RFQ mass filter, where the mass range can be restricted. This combination of LACCI with a mass filter is one of the key developments to provide carbon clusters with the necessary rate of the reference ions in the mass range of interest and simultaneously transmit only the ions of interest to avoid space-charge effects in the MR-TOF-MS. Then, the ions are transported to the MR-TOF-MS, where high-accuracy mass measurements are performed. A gate valve is mounted between the extraction unit and the recapturing unit. This allows quick replacement of samples and maintenance without breaking the vacuum of the RFQ beamline.

3. Results and discussion

3.1. Mass range

An investigation of the mass range of ions delivered by LACCI was carried out using a Sigradur® Glassy Carbon target to prove that the entire mass range of the chart of the nuclides can be covered. Sigradur® Glassy Carbon is an ultra-pure form of carbon, nearly without any contaminants inside, that combines properties from ceramic and graphite. The transport in the trap system of MR-TOF-MS is mass-dependent. Therefore, the RF frequency and the timing of the transport between the traps of the trap system were changed during the test to adjust the transmitted mass range. Thus, the entire mass range provided by LACCI could be investigated. Fig. 2 shows the accumulated mass spectrum of carbon cluster ions collected for 1000 s at a laser repetition rate of 20 Hz. The investigated distribution of carbon cluster ions ranges from ${}^{12}C_7{}^+$ at a mass of 84 u to ${}^{12}C_{24}{}^+$ at a mass of 288 u. Moreover ${}^{12}C^{13}_{n-1}C^+_1$ and ${}^{12}C^{13}_{n-2}C^+_2$ containing one or two ${}^{13}C$ atoms are also observed. The abundance pattern of the clusters agrees with earlier publications [30,31]. By adjusting the low mass cut-off (LMCO) of the RFQs in the FRS-IC beamline, low-mass ions could also be transported. In previous commissioning measurements, it was shown that cluster ions as small as ${}^{12}C_3{}^+$ can be produced as well [32]. The other ion sources of the MR-TOF-MS can also produce reference ions of lower masses, e.g., by electron impact ionization. Therefore, the mass region below 70 u was not the focus in the development of LACCI. Only singly charged carbon cluster ions were investigated in the measurement. The literature shows that multiply-charged carbon cluster ions can also be produced [33], but the dominant ion production channel is the singly-charged channel. In the MR-TOF-MS, multiply-charged cluster ions could also be used for calibration. The results show that LACCI can provide reference ions for the full mass range of the MR-TOF-MS for mass measurements of nuclides.

Another aspect of the mass range covered by LACCI is the density of available mass lines. This was investigated using a fullerene target enriched with ${}^{13}C$ (∼5%). The mass range of the MR-TOF-MS remained the same, with the center at 133 u. Fig. 3 shows the accumulated mass spectrum of carbon cluster ions acquired for 263 s at a laser repetition rate of 20 Hz. At each mass line, ions in the range from ${}^{12}C_{10}{}^+$ at a mass of 120 u to ${}^{12}C^{13}_{10}C_3{}^+$ at a mass of 159 u were identified.

In the discussed spectra (Figs. 2 and 3), it can be seen that in the laser ablation process, a large number of ions over a broad mass range are produced. For the work with exotic ions, avoiding space-charge effects in the MR-TOF-MS and a deterioration of the mass measurement accuracy is important. Due to this, one of the requirements for the system was to provide a tool to select only the mass region of interest for the carbon cluster ions. The mass filter [26] located at the end of DISTRICT2 fulfills this task. Using the mass filter to restrict the mass range has been tested with carbon cluster ions of ${}^{12}C_{10}{}^+$–${}^{12}C_{16}{}^+$ generated using a Sigradur® target. Only the ions of interest are allowed to pass through the mass filter and enter the trap system of the MR-TOF-MS. Fig. 4(a) shows a broadband spectrum of carbon clusters transmitted to the MR-TOF-MS when the mass filter was set to the full

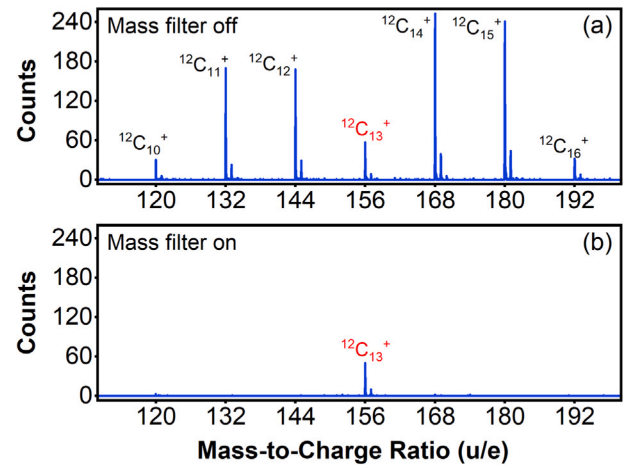

Fig. 4. Mass spectra illustrating the operation of the RFQ mass filter to transmit only the mass region of interest. The top panel (a) shows a broadband spectrum of carbon clusters, which were transmitted to the MR-TOF-MS when the mass filter was set to the full transmission of all ions. For the mass spectrum which is shown in the bottom panel (b), the mass filter was set to transmit only the mass range 151–162 u. Therefore, only the ions ${}^{12}C_{13}{}^+$ and ${}^{12}C^{13}_{12}C_1{}^+$ were transmitted and detected by the MR-TOF-MS.

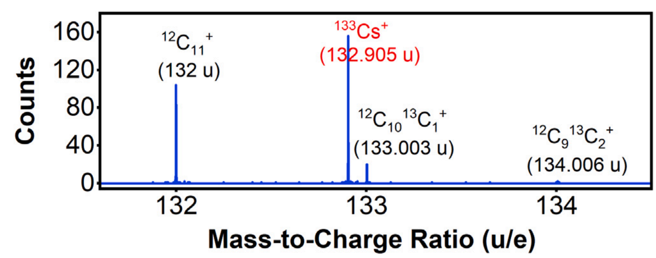

Fig. 5. Mass spectrum of ions, which have been produced by two different ion sources, viz. carbon cluster ions (${}^{12}C_{11}{}^+$, ${}^{12}C^{13}_{10}C_1{}^+$ and ${}^{12}C^{13}_9C_2{}^+$) produced by LACCI on a Sigradur® target and ${}^{133}Cs^+$ ions from a thermal ion source that was moved into the beamline close to the CSC in DISTRICT1. The ions have been merged in the switchyard and measured simultaneously by the MR-TOF-MS.

transmission of all ions. For the mass spectrum shown in Fig. 4(b), the mass filter was set to transmit only the mass range 151–162 u. It can be seen that as the range selected by the mass filter narrows, the intensity of the reference ion ${}^{12}C_{13}{}^+$ remains unchanged. This selection of the mass range of interest is one of the key steps to fulfill the requirements of an MR-TOF-MS for a carbon cluster calibration ion source.

An alternative pre-selection of the carbon clusters is possible in the ion extraction unit of LACCI by operating the pair of electrostatic steerers in a pulsed mode. The ion extraction unit of LACCI acts as a short linear time-of-flight mass spectrometer, in which the laser pulse starts the ions' flight and carbon clusters ions of different mass-to-charge ratios pass the electrostatic steer at different times. Thus, the steerers can deflect unwanted carbon cluster ions, and only the cluster ions in the mass region of interest can be transmitted. This concept was successfully used in the system's initial commissioning. The method offers only a reduced mass resolving power compared to the RF mass filter but can be easily applied to other existing laser ablation sources.

A critical feature of DISTRICT2 is to merge ions of interest from the CSC and LACCI in the RF switchyard. This was demonstrated, see Fig. 5, by merging the ions of interest and reference ions in the same spectrum. Here, ${}^{133}Cs^+$ ions were produced via surface-ionization by a thermal ion source located in DISTRICT1 close to the CSC and served as ions of interest, while carbon cluster ions produced by LACCI with a Sigradur® target were used as the reference ions.

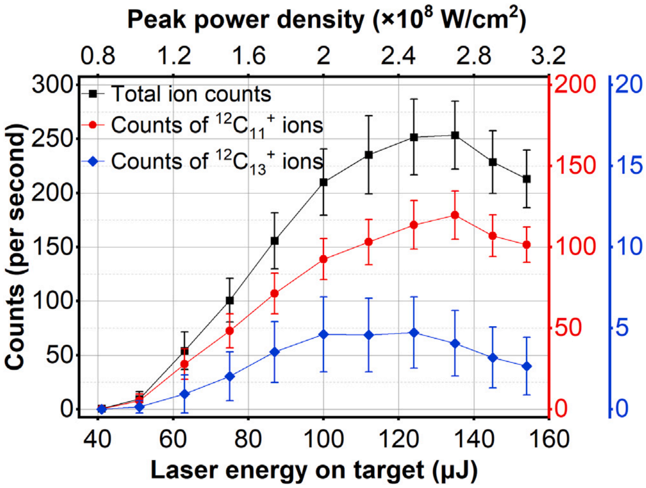

Fig. 6. Ion counts per second (total ion counts per second and ion counts gated on the mass lines of $^{12}C_{11}^+$ and $^{12}C_{13}^+$, respectively) produced by LACCI and measured in the MR-TOF-MS, as a function of the laser energy and peak power density. Please note the different color-coded abundance scales corresponding to the respective clusters. The laser energy impacts the total ion counts, but the ratio of the abundances is not affected by laser energy. The ion rate of the different cluster ions can be adjusted using the laser energy by up to two orders of magnitude, depending on the required reference ion rate of the experiment. The ion counts per second are average values covering 200 s; the error bars reflect the standard deviation.

3.2. Dynamic range

The laser energy is an important parameter of the laser ionization process, directly affecting the ionization efficiency. To investigate the dynamic range of LACCI, the dependence of the ion rate on the laser energy was investigated for ions produced on a Sigradur® target at a laser repetition rate of 20 Hz. The mass spectrum, similar to the spectrum shown in Fig. 2, shows peaks for ions of $^{12}C_9^+$, $^{12}C_{10}^+$, $^{12}C_{11}^+$, $^{12}C_{12}^+$, $^{12}C_{13}^+$, $^{12}C_{14}^+$, $^{12}C_{15}^+$ and the corresponding ions of $^{12}C^{13}_{n-1}C_1^+$ and $^{12}C^{13}_{n-2}C_2^+$. Fig. 6 shows the total ion counts accumulated for one second and averaged over 200 s, the rate of $^{12}C_{11}^+$ ions, and the rate of $^{12}C_{13}^+$ ions as a function of the laser energy. The laser energy was adjusted by changing the rotation angle of the half-wave plate in front of the beam splitter, which is explained in Section 2. The laser peak power density was calculated using a laser spot size (80 μm), a laser pulse width (10 ns), and laser energy (variable). The laser energy was increased from 41 μJ to 154 μJ, resulting in a peak power density to have increased from 8×10^7 W/cm^2 to 3×10^8 W/cm^2. The ion count rate increases up to 135 μJ, followed by a decreasing trend afterward. This decrease is caused by breaking the carbon–carbon bond at high power densities, causing an increase in fragmentation and a decrease in medium-mass cluster production efficiency. In addition, the translational temperature of the ions increases with laser power, leading to a lower transmission efficiency of the ion into the RFQ. These results show that the maximal ion rate of all carbon cluster ions is achieved for a laser energy of about 125 μJ. In addition, the results show that laser energy does not affect the abundance ratio of the carbon clusters within the investigated cluster range; only the laser power affects the total yield. The fragmentation mechanism of the carbon clusters has not been investigated. The required count rate for calibration is much lower than the maximum rate reached. Thus, the laser energy can adjust the ion rate of the carbon cluster of interest for a specific experiment. The results of the laser energy investigation show a dynamic range of two orders of magnitude, on the lower end limited by the ablation threshold and on the upper side by breaking the carbon–carbon bond at high power densities. This dynamic range offers the necessary flexibility to suit the experimental requirement.

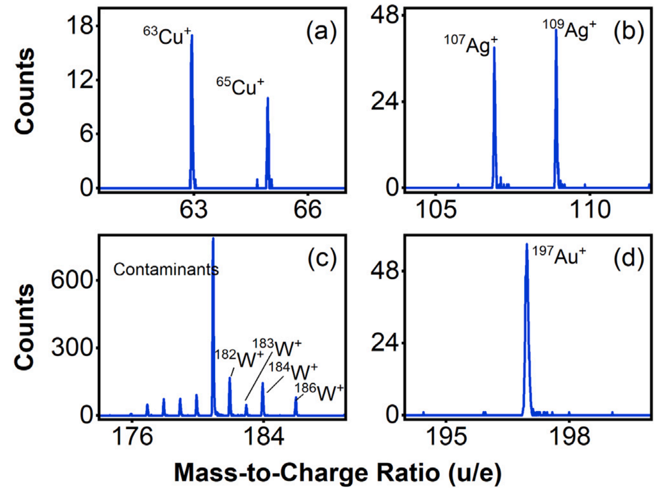

Fig. 7. Mass spectrum of different metal ions produced by LACCI, copper (a), silver (b), tungsten (c) and gold (d).

The laser used in LACCI has an adjustable repetition rate of 1–100 Hz. If we compare the total ion counts at 20 Hz and 100 Hz, an increase in the measured ion rate of a factor of 5 would be expected. An evaluation of the laser repetition rate dependency was carried out using a Sigradur® target with laser energy of 124 μJ. Since the speed of the target is currently limited to 120 mm/min, the laser repetition rate is set to 20 Hz and 100 Hz with the 2D scan speed of the target of 24 mm/min and 120 mm/min, respectively. This configuration ensures identical overlapping bombardment points. By increasing the repetition rate 5 times, the total ion counts of carbon clusters increased from about 100 cps to about 375 cps, an increase of 3.75. In those points, the ion production and release efficiency from the target is reduced. Since the speed of the target changing direction is currently limited by the control software to 120 mm/min, an update to the software can solve this problem. Assuming a spot size of 80 μm, a velocity of 480 mm/min would be needed at a laser repetition rate of 100 Hz to avoid an overlap in the bombarding points.

3.3. Metal samples

LACCI generates carbon cluster ions as reference ions for MR-TOF-MS and serves as an offline source in combination with MR-TOF-MS for measuring other ions from other targets, e.g., metals. Fig. 7 shows the mass spectra of copper, silver, tungsten, and gold ions produced in laser ablation with LACCI. The contaminants in the tungsten spectrum can be correlated with cross-contamination from targets in its neighborhood. Such metal ions may be suitable reference ions in special cases where the ion of interest is an isobar to one of the isotopes of the metal targets. Also, the metal targets may enable further applications in the future, making LACCI into a universal tool, for example, for cluster research [34–36].

3.4. Long-term measurements

The main design goal of LACCI was to provide reference ions during accelerator beam experiments and to ensure high accuracy of the mass measurements of the exotic nuclei using these reference ions. Typically, beam time at FRS-IC lasts for several days, which requires a stable operation of LACCI concerning count rate and reliability over this time period.

An initial test of the long-term stability of LACCI was performed before DISTRICT2 and LACCI were integrated into the FRS-IC beamline. The results of the measurements are shown in Fig. 8 for erbium ions

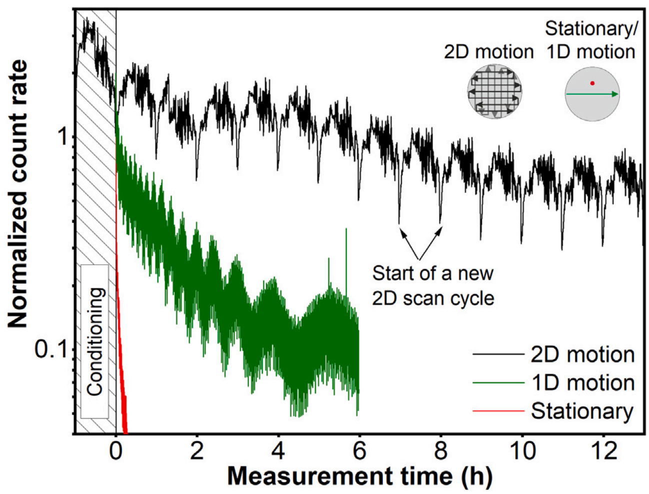

Fig. 8. Long-term measurement of the count rate for erbium ions produced in laser ablation by LACCI with a repetition rate of 100 Hz. The count rate versus time with a 2D motion of the movable target is compared to a stationary target and to 1D motion of the target, respectively. The latter is the established technique for similar laser ion sources. The data are normalized to the count rate obtained after the target surface has been conditioned by completing one movement cycle of the laser over the target.

at a laser repetition rate of 100 Hz. The ions were detected with the channeltron detector at the RFQ switchyard without He gas in the RFQ. This allowed us to use the beam path for a time-of-flight measurement. In Fig. 8, the ion count rate for a 2D motion of the movable target is compared to the count rate with a stationary target and to the count rate for a 1D motion of the target. The 2D scan cycle shows a much better stability of the count rate over a long time period compared to the stationary target and to the 1D motion of the target. The latter is the established state-of-the-art technique in similar laser ablation ion sources. During the 2D scan cycle, 4.42×10^6 laser pulses hit the target. In the measurement, it was seen that the first layer of the target has different properties compared to the following ones in terms of abundance pattern, including additional adducts, since the first layer was exposed to its surroundings for a longer time, leading to, e.g., oxidation. To compare the deterioration of the measurement's main target structure, the first motion cycle was disregarded, and the count rate in Fig. 8 was normalized to the counts observed in the second cycle.

A long-term stability measurement of the whole system was performed using the MR-TOF-MS after fully integrated into the FRS-IC. The laser repetition rate was set to 100 Hz, and the laser energy was set to 57 μJ. A continuous 21.5-hour measurement was conducted using a Sigradur® target. The test results, shown in Fig. 9, indicate that after 21.5 h, the count rate of ${}^{12}C_{11}{}^{+}$ decreased from about 600 counts per 5 s to about 300 counts per 5 s, corresponding to a decrease of the count rate by a factor of only two. The equidistant fluctuations seen in the ion count rate are mainly caused by reversing the movement of the 2D scan and by starting a new 2D scan cycle. Each 2D scan cycle takes about 57 min, which comprises a 28.5 min horizontal scan and a corresponding 28.5 min vertical scan at a scan speed of 20 mm/min and a step size of 80 μm. The red line indicates the average over 14.25 min, two of which correspond to the time needed to perform a 2D scan over the whole target in one direction, either horizontal or vertical. On the right side of Fig. 9, photographs of the target at different times during the measurement are shown. It can be seen that after prolonged scanning, visible lithographic marks appear on the surface, indicating the high reliability of the 2D movement target and laser focusing at a level of several tens of micrometers, which is less than the step size of the 2D movement of the target. During the whole operation of LACCI, no deteriorating effects of the electrical and mechanical system caused by the deposition of neutral laser ablation products onto electrodes and other sensitive surfaces were observed.

The measurement time of 21.5 h at a repetition rate of 100 Hz is equivalent to a measurement time of about 90 days at a repetition rate of 1 Hz. During this measurement, 7.74×10^6 laser pulses hit the target. This constitutes a much longer, more effective, and more stable operation time than possible with existing laser ablation ion sources at accelerator facilities. For a cycle frequency of the MR-TOF-MS of 10 to 20 Hz, the requirement of one week of beamtime is readily fulfilled by using only one Sigradur® target. However, higher cycle frequencies up to 100 Hz can be accommodated by changing between different targets several times during the experiment, which is possible without opening the system by simply moving the target platform. The long-term test of the stability of the laser ablation in combination with the 2D movable target platform was the last missing piece for the long-term operation of the whole system since the reliability of the other components, as the electrostatic ion optics or the RFQ beamline was shown in previous experiments to be stable over month [37]. Thus, LACCI ensures a stable long-term operation at high repetition rates for the first time, perfectly matching the requirements for providing reference ions for an MR-TOF-MS.

4. Summary and outlook

A long-term stable laser ablation carbon cluster ion source (LACCI) has been developed and commissioned at the FRS-IC. It is the first laser ablation ion source for long-term measurements with an MR-TOF-MS. LACCI provides carbon cluster ions as a reference for mass measurements of exotic nuclei. The reference ions from LACCI and the ions from the CSC can be merged into one ion beam in the RFQ switchyard, guided to the MR-TOF-MS, and measured simultaneously. A mass filter can isolate the ions to the mass region of interest, thus avoiding space-charge effects in the MR-TOF-MS. Combining a laser ablation carbon cluster ion source with a dedicated mass filter is one of the key steps to make LACCI suitable for operation as a calibration ion source at an MR-TOF-MS.

To enhance the performance of LACCI, advanced technical solutions have been implemented: (i) A 2D movable platform with several laser targets allows using different materials such as carbon targets (glassy carbon (Sigradur®) and ${}^{13}C$ enriched fullerene) and metal targets (silver, gold, copper, tungsten, and erbium). A continuous 2D motion during the laser ablation process prevents the laser beam from constantly hitting the same target spot and, therefore, a reduction of the extraction efficiency of the ions. It also allows a change of targets without opening the system. (ii) A target monitoring system and a laser spot and energy monitoring laser optics system facilitate easy tuning of the laser beam and its position on the target. (iii) An ion source region in a vacuum, separated by a gate valve from a dedicated re-capture unit, from which the ions are injected into a gas-filled RFQ for ion cooling and highly efficient ion transmission. This makes the system maintenance-friendly and tolerant to the huge amount of neutrals that the laser ablation process can produce.

Carbon clusters up to C_{24}^{+} with a mass of 288 u were produced by LACCI and identified in the MR-TOF-MS, thus covering almost the whole chart of the nuclides with reference ions. Using a target with ${}^{13}C$ enriched fullerenes, reference ions at every mass number can be produced over a large mass range. The dependency of the ion production on the laser energy and the repetition rate was investigated; the ion rate can be varied over a large abundance range to match the specific requirements of the experiment. LACCI was shown to provide reference ions at a repetition rate of 100 Hz for 21.5 h with a single Sigradur® target.

LACCI thus offers a large variety of reference ions with well-known masses at stable rates; reference ions with similar mass numbers to the ion of interest can be selected during online mass-measurement

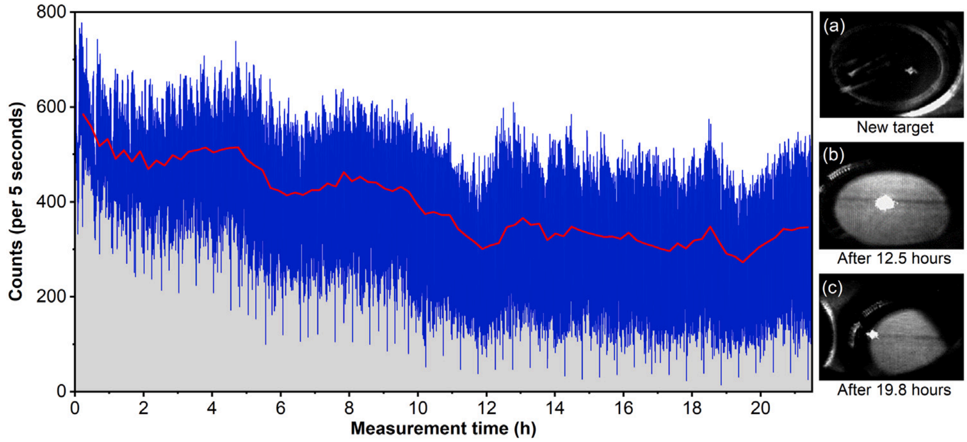

Fig. 9. Measurement of the long-term stability of the count rate of $^{12}C_{11}{}^{+}$ produced by laser ablation from a Sigradur® target in LACCI with a repetition rate of 100 Hz. The data were accumulated every 5 s. Fluctuations were mainly caused by reversing the movement of the 2D scan from horizontal to vertical or by starting a new 2D scan cycle from its original start position. A 2D scan comprises a 28.5 min horizontal scan and a corresponding 28.5 min vertical scan. The average over half of this period of 14.25 min is shown as a red line. On the right side, photographs of the target at different times during the measurement are shown: (a) new target, (b) after 12.5 h in the center of the target, (c) after 19.8 h on the edge of the target. The laser spot is the bright point, in the later pictures after starting the go several times over the target grooves get visible.

campaigns to minimize systematic mass-dependent uncertainties. Furthermore, systematic studies of the mass accuracy of the MR-TOF-MS will be performed by carbon cluster cross-reference measurements in the near future. The new ion source will, therefore, help achieve a higher mass measurement accuracy all over the chart of nuclei and, with this, enable mass measurements of short-lived exotic nuclei, which are currently not accessible. Those mass measurements will be conducted at the FRS at the GSI Helmholtz Centre for Heavy Ion Research in Darmstadt, Germany, and in the future at the Super-FRS at the Facility for Antiproton and Ion Research (FAIR), in Darmstadt, Germany. They will be performed at the forefront of nuclear physics research and help understand the origin of the chemical elements in the universe and the structure of exotic nuclei.

CRediT authorship contribution statement

Jiajun Yu: Writing – original draft, Visualization, Investigation, Formal analysis, Data curation. **Christine Hornung:** Writing – original draft, Supervision, Resources, Methodology, Investigation, Formal analysis. **Timo Dickel:** Writing – review & editing, Supervision, Methodology, Investigation, Conceptualization. **Wolfgang R. Plaß:** Writing – review & editing, Supervision, Methodology, Investigation, Conceptualization. **Daler Amanbayev:** Writing – review & editing, Validation, Investigation. **Julian Bergmann:** Validation, Software, Investigation. **Zhuang Ge:** Validation, Investigation. **Florian Greiner:** Validation, Investigation. **Hans Geissel:** Validation, Conceptualization. **Lizzy Gröf:** Validation, Investigation. **Gabriella Kripko-Koncz:** Writing – review & editing, Validation, Investigation. **Meetika Narang:** Validation, Investigation. **Ann-Kathrin Rink:** Validation, Investigation. **Christoph Scheidenberger:** Writing – review & editing, Validation, Supervision, Methodology, Funding acquisition, Conceptualization. **Jianwei Zhao:** Validation, Investigation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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References

[1] T. Yamaguchi, H. Koura, Y.A. Litvinov, M. Wang, Masses of exotic nuclei, Prog. Part. Nucl. Phys. 120 (2021) 103882.

[2] A.N. Bohr, B.R. Mottelson, Nuclear Structure (in 2 Volumes), World Scientific Publishing Company, 1998.

[3] H. Schatz, Nuclear masses in astrophysics, Int. J. Mass Spectrom. 349 (2013) 181–186.

[4] J. Clark, G. Savard, M. Mumpower, A. Kankainen, Precise mass measurements of radioactive nuclides for astrophysics, Eur. Phys. J. A 59 (9) (2023) 204.

[5] W.R. Plaß, T. Dickel, C. Scheidenberger, Multiple-reflection time-of-flight mass spectrometry, Int. J. Mass Spectrom. 349 (2013) 134–144.

[6] R.N. Wolf, F. Wienholtz, D. Atanasov, D. Beck, K. Blaum, C. Borgmann, F. Herfurth, M. Kowalska, S. Kreim, Y.A. Litvinov, et al., ISOLTRAP's multi-reflection time-of-flight mass separator/spectrometer, Int. J. Mass Spectrom. 349 (2013) 123–133.

[7] P. Schury, M. Wada, Y. Ito, F. Arai, S. Naimi, T. Sonoda, H. Wollnik, V.A. Shchepunov, C. Smorra, C. Yuan, A high-resolution multi-reflection time-of-flight mass spectrograph for precision mass measurements at RIKEN/SLOWRI, Nucl. Instrum. Methods Phys. Res. B 335 (2014) 39–53.

[8] T. Dickel, W.R. Plaß, A. Becker, U. Czok, H. Geissel, E. Haettner, C. Jesch, W. Kinsel, M. Petrick, C. Scheidenberger, A. Simon, M.I. Yavor, A high-performance multiple-reflection time-of-flight mass spectrometer and isobar separator for the research with exotic nuclei, Nucl. Instrum. Methods Phys. Res. A 777 (2015) 172–188.

[9] T.Y. Hirsh, N. Paul, M. Burkey, A. Aprahamian, F. Buchinger, S. Caldwell, J.A. Clark, A.F. Levand, L.L. Ying, S.T. Marley, et al., First operation and mass separation with the CARIBU MR-TOF, Nucl. Instrum. Methods Phys. Res. B 376 (2016) 229–232.

[10] W.R. Plaß, T. Dickel, S. Purushothaman, P. Dendooven, H. Geissel, J. Ebert, E. Haettner, C. Jesch, M. Ranjan, M.P. Reiter, et al., The FRS ion catcher–A facility for high-precision experiments with stopped projectile and fission fragments, Nucl. Instrum. Methods Phys. Res. B 317 (2013) 457–462.

[11] H. Geissel, K. Beckert, F. Bosch, H. Eickhoff, B. Franczak, B. Franzke, M. Jung, O. Klepper, R. Moshammer, G. Münzenberg, et al., First storage and cooling of secondary heavy-ion beams at relativistic energies, Phys. Rev. Lett. 68 (23) (1992) 3412.

[12] I. Mardor, S.A. San Andrés, T. Dickel, D. Amanbayev, S. Beck, J. Bergmann, H. Geissel, L. Gröf, E. Haettner, C. Hornung, et al., Mass measurements of As, Se, and Br nuclei, and their implication on the proton-neutron interaction strength toward the N=Z line, Phys. Rev. C 103 (3) (2021) 034319.

[13] S. Ayet San Andrés, et al., Multiple-reflection time-of-flight mass spectrometry for low-intensity short-lived ion beams and their low-lying isomeric states, Phys. Rev. C 99 (2019) 064313.

[14] Z. Wang, Aufbau und Charakterisierung einer Laserablationsionenquelle mit Kohlenstoffclustern als Referenz für Flugzeitmassenspektrometrie von exotischen Kernen (Ph.D. thesis), Universität Gießen, 2006.

[15] A. Van Orden, R.J. Saykally, Small carbon clusters: spectroscopy, structure, and energetics, Chem. Rev. 98 (6) (1998) 2313–2358.

[16] L. Bloomfield, M. Geusic, R. Freeman, W. Brown, Negative and positive cluster ions of carbon and silicon, Chem. Phys. Lett. 121 (1–2) (1985) 33–37.

[17] W. Brown, R. Freeman, K. Raghavachari, M. Schlüter, Covalent group IV atomic clusters, Science 235 (1987) 860–865.

[18] K. Blaum, G. Bollen, F. Herfurth, A. Kellerbauer, H.-J. Kluge, M. Kuckein, E. Sauvan, C. Scheidenberger, L. Schweikhard, Carbon clusters for absolute mass measurements at ISOLTRAP, Eur. Phys. J. A 15 (2002) 245–248.

[19] C. Scheidenberger, G. Bollen, F. Herfurth, A. Kellerbauer, H.-J. Kluge, M. Koizumi, S. Schwarz, L. Schweikhard, Production and trapping of carbon clusters for absolute mass measurements at ISOLTRAP, Nuclear Phys. A 701 (1–4) (2002) 574–578.

[20] V.V. Elomaa, T. Eronen, U. Hager, A. Jokinen, T. Kessler, I. Moore, S. Rahaman, C. Weber, J. Äystö, Development of a carbon-cluster ion source for JYFLTRAP, Nucl. Instrum. Methods Phys. Res. B 266 (19–20) (2008) 4425–4428.

[21] C. Izzo, G. Bonen, S. Bustabad, M. Eibach, K. Gulyuz, D.J. Morrissey, M. Redshaw, R. Ringle, R. Sandler, S. Schwarz, A.A. Valverde, A laser ablation source for offline ion production at LEBIT, Nucl. Instrum. Methods Phys. Res. B 376 (2016) 60–63.

[22] J. Lommen, Reference Masses for Precision Mass Spectrometry: Design and Implementation of a Pierce Geometry to the Cluster Ion Source at ISOLTRAP (Bachelor thesis), University of Heidelberg, 2011.

[23] C. Smorra, K. Blaum, K. Eberhardt, M. Eibach, J. Ketelaer, J. Ketter, K. Knuth, S. Nagy, A carbon-cluster laser ion source for TRIGA-TRAP, J. Phys. B: At. Mol. Opt. Phys. 42 (15) (2009) 154028.

[24] M. Sameed, D. Maxwell, N. Madsen, Ion generation and loading of a penning trap using pulsed laser ablation, New J. Phys. 22 (1) (2020) 013009.

[25] W.R. Plaß, T. Dickel, S.A. San Andrés, J. Ebert, F. Greiner, C. Hornung, C. Jesch, J. Lang, W. Lippert, T. Majoros, D. Short, H. Geissel, E. Haettner, M.P. Reiter, A.K. Rink, C. Scheidenberger, M.I. Yavor, High-performance multiple-reflection time-of-flight mass spectrometers for research with exotic nuclei and for analytical mass spectrometry, Phys. Scr. T166 (2015) 014069.

[26] E. Haettner, W.R. Plass, U. Czok, T. Dickel, H. Geissel, W. Kinsel, M. Petrick, T. Schäfer, C. Scheidenberger, A versatile triple radiofrequency quadrupole system for cooling, mass separation and bunching of exotic nuclei, Nucl. Instrum. Methods Phys. Res. A 880 (2018) 138–151.

[27] J.R. Pierce, Rectilinear electron flow in beams, J. Appl. Phys. 11 (8) (1940) 548–554.

[28] D. Davies, A High-Power Laser Ablation Ion Source for Penning Trap Studies of Rare Isotopes (Ph.D. thesis), Michigan State University, 2006.

[29] C.S. Aké, H. Sobral, P. Ramos-Alvarez, C. Lemen, M. Villagrán-Muniz, Ion kinetic energy control in cross-beam pulsed laser ablation on graphite targets, J. Phys. Conf. Ser. 59 (1) (2007) 728.

[30] S.B.H. Bach, J.R. Eyler, Determination of carbon cluster ionization potentials via charge transfer reactions, J. Chem. Phys. 92 (1) (1990) 358–363.

[31] S.K. Lai, I. Setiyawati, T.W. Yen, Y.H. Tang, Studying lowest energy structures of carbon clusters by bond-order empirical potentials, Theor. Chem. Acc. 136 (2017) 1–13.

[32] C. Hornung, High-Resolution Experiments with the Multiple-Reflection Time-of-Flight Mass Spectrometer at the Fragment Separator FRS (Ph.D. thesis), Universität Gießen, 2018.

[33] X. Kong, Y. Huang, Y. Chen, Difference in formation of carbon cluster cations by laser ablation of graphene and graphene oxide, J. Mass Spectrom. 47 (4) (2012) 523–528.

[34] P. Fischer, S. König, L. Schweikhard, Concatenated multi-reflection time-of-flight spectra for wide-band mass spectrometry, Int. J. Mass Spectrom. 463 (2021) 116546.

[35] P. Fischer, L. Schweikhard, Disentangling polycationic fullerenes produced from glassy carbon with multireflection time-of-flight mass spectrometry, Phys. Rev. Res. 4 (4) (2022) 043187.

[36] P. Fischer, P.F. Giesel, L. Schweikhard, Photodissociation of small group-14 atomic clusters in a multi-reflection time-of-flight mass spectrometer, Eur. Phys. J. D 77 (2) (2023) 27.

[37] L. Varga, H. Wilsenach, O. Hall, T. Dickel, M. Reiter, D. Amanbayev, T. Davinson, D. Morrissey, I. Pohjalainen, N. Tortorelli, J. Yu, J. Zhao, S. Ayet, S. Beck, J. Bergmann, Z. Ge, H. Geissel, L. Heitz, C. Hornung, N. Kalantar-Nayestanaki, E. Khan, G. Kripko-Koncz, I. Mardor, M. Narang, W. Plass, C. Scheidenberger, M. Simonov, S. Singh, A. State, C. Theisen, M. Vandebrouck, P. Woods, Novel device to study double-alpha decay at the FRS Ion Catcher, Nucl. Instrum. Methods Phys. Res. A 1063 (2024) 169252.