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Removal of molecular contamination in low-energy RIBs by the isolation-dissociation-isolation method

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ABSTRACT

Experiments with low-energy rare ion beams often suffer from a large amount of molecular contaminant ions. We present the simple isolation-dissociation-isolation method to suppress this kind of contamination. The method can be applied to almost all types of low-energy beamlines. In a first step, a coarse isolation of the mass-to-charge ratio of interest is performed, e.g. by a dipole magnet. In a second step, the ions are dissociated. The last step is again a coarse isolation of the mass-to-charge ratio around the ion of interest. The method was tested at the FRS Ion Catcher at GSI with a radioactive ion source installed inside the cryogenic stopping cell as well as with relativistic ions delivered by the synchrotron SIS-18 and stopped in the cryogenic stopping cell. The isolation and dissociation, here collision-induced dissociation, have been implemented in a gas-filled RFQ beamline. A reduction of molecular contamination by more than 4 orders of magnitude was achieved.

1. Introduction

A common problem faced when using low-energy beams of exotic nuclei is molecular contamination. Its origin can vary, e.g. hot surfaces of ion sources [1] or ionization of the residual gas within gas filled parts of the system [2–4]. This molecular contamination can render high precision measurements impossible. Thus, reduction or complete removal of this contamination is important for experiments at low-energy rare ion beam (RIB) facilities.

With collision-induced dissociation (CID) [5] a tool for breaking up molecules is readily available at most low-energy beamlines [6]. CID can be done, depending on the layout of the facility, by capturing the ions after an electrostatic beamline with increased kinetic energy (tens of eV instead of few eV) in an RF-quadrupole (RFQ) cooler/buncher or, as presented here, in a gas-filled RFQ beamline. During the CID process the molecules are excited by collisions with neutral gas atoms. Due to this excitation the molecules can break up. Thereby removing contaminant ions that had before the break-up the same mass-to-charge ratio as the ion of interest (IOI). But, molecular ions with a mass-to-charge ratio larger than that of the IOI might break up into fragments with a mass-to-charge ratio of the IOI, so the molecular contamination of the IOI is only changed, but not removed. This problem is solved by applying an isolation step before the dissociation. An isolation step after the dissociation remove the fragmented molecules from the beam. The isolation steps can be performed for DC beams by a dipole magnet [3] or RF mass filter [7] and for pulsed beams with a Bradbury-Nielsen gate [8]. This method of consecutive steps of isolation, dissociation and again isolation is the so-called IDI method (Fig. 1) and is described in the following. This method was successfully tested in the RFQ beamline at the FRS Ion Catcher (FRS-IC), GSI.

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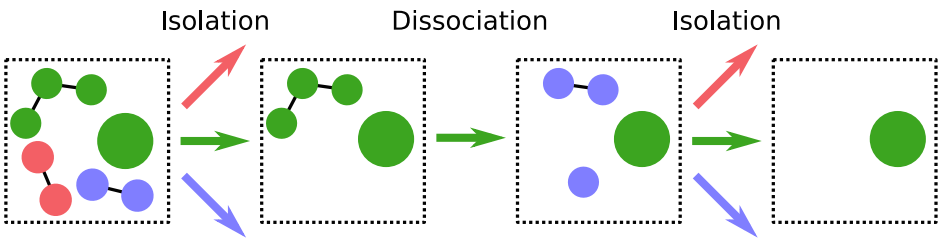

Fig. 1. Principle of removing molecular contamination by the IDI method. Colours represent the mass-to-charge ratio, where green is the mass-to-charge ratio of the IOI, blue is lower and red higher mass-to-charge ratios. During the first isolation step, mass-to-charge ratios higher and lower than the mass-to-charge ratio of interest are removed. Due to the dissociation the remaining molecules are fragmented. These fragments are removed during the second isolation step.

2. Experimental setup

The FRS-IC is located at the final focal plane of the Fragment Separator (FRS) [9]. The main parts of the FRS-IC are a cryogenic stopping cell (CSC) [10], a gas-filled RFQ beamline and a multiple-reflection time-of-flight mass spectrometer (MR-TOF-MS) [11].

At the FRS, exotic nuclei are produced in-flight by fragmentation and fission reactions. In the CSC the selected ions are stopped and thermalized in the gas. The stopping gas is helium at pressures of 50–70 mbar and a temperature of about 90 K. The stopped ions are transported by a DC field to the extraction region. Here the RF carpet provides a repelling force to prevent that the ions will hit the surface and guide them to the exit nozzle. The ions are extracted by the gas flow through the nozzle into the RFQ beamline, and transported to the MR-TOF-MS, where precision mass measurements are performed. For the first isolation step the extraction RFQ is operated as an RF mass filter [12,7], with a mass resolving power of about 10 to 20. The second isolation step is performed with the RFQ at the entrance to the MR-TOF-MS. This RFQ is operated at increased RF amplitudes, thereby low mass ions are not transported, the so-called low mass cut-off. A detailed description of the RFQ beamline and the FRS Ion Catcher in general can be found in [13].

3. Experiment and results

Tiny amounts (on the level of parts-per-billion) of impurities remain in the helium stopping gas despite the different measures that are taken to ensure highest cleanliness, e.g. cryogenic operation of the CSC [14] and the use of getter purifiers (SAES Pure Gas, MicroTorr) in the helium supply line. Due to the high ionization in the CSC, about 10 million He^+/e^- pairs are generated for each IOI entering the CSC, the impurities are ionized and result in molecular contamination in the low energy ion beam. The reduction of this molecular contamination is an important step to enable accurate measurements of rare isotopes [15].

To test the IDI concept at the FRS Ion Catcher, systematic studies with a ^{228}Th radioactive ion source as well as with relativistic ions were performed. The contamination in the mass-to-charge range of the singly-charged ions from the ^{228}Th source is shown in Fig. 2 top panel. These ions are generated from contamination on the surface of the ^{228}Th source and are not observed in experiments with beams from the FRS. To fragment these molecules CID was used by applying a voltage step of 60 volts in the RFQ beamline after the extraction RFQ. In the middle panel of Fig. 2 the result of this CID is shown. One can see that the mass lines at ~ 230 u/e disappear, but new mass lines appear. These are fragments of molecules that are broken apart due to CID. It can be seen that it is not sufficient to apply only CID; CID can even increase the amount of contamination at the mass-to-charge ratio of interest. By adding the isolation steps before and after the CID a contaminant-free spectrum was produced (Fig. 2 bottom panel). In this measurement the first isolation step was performed by operating the extraction RFQ with U/V = 0.12 (m/Δm ~ 10), where U is the DC and V the AC amplitude of the quadrupolar field on the extraction RFQ.

The experiments with relativistic ions were carried out with a ^{124}Xe primary beam extracted from the SIS at 600 MeV/u. These ions were slowed down and stopped in the CSC. Despite the excellent cleanliness

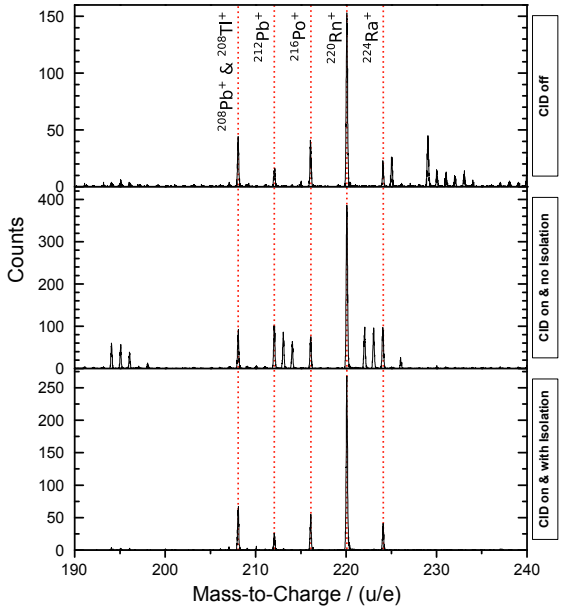

Fig. 2. Low-resolution mass-to-charge spectra of a ^{228}Th radioactive ion source mounted in the CSC. Ions generated by α-decay are marked in red. Top panel: No IDI is applied. Contaminants around mass-to-charge ratio 195 u/e and 230 u/e can be seen. Middle panel: After applying CID these molecules break up, but fragments from molecules with the higher mass-to-charge ratios now contaminate the mass-to-charge range of interest (212–224 u/e). Bottom panel: IDI is applied. The mass-to-charge spectrum is free of molecular contamination. Note, that in this case a larger mass-to-charge range has been selected in the isolation stages to transport all singly charge atomic ions from the source.

of the CSC, the identification of the stopped ^{124}Xe was possible only after applying the IDI method. Without it the mass line of $^{124}Xe^+$ was covered under $^{80}Kr^{12}C^{16}O_2^+$. The mass-to-charge difference is less than 300 keV/(c^2e), corresponding to a mass resolving power of more than 400,000. To fully separate (baseline separation) these masses, a mass resolving power of more than one million would be necessary. With IDI $^{80}Kr^{12}C^{16}O_2^+$ and all others molecular ions were removed. For $^{84}Kr^{12}C^{16}O_2^+$ the suppression is more than 4 orders of magnitude. The efficiency for the IOI was measured to be unchanged when applying the IDI method. Note that due to the special conditions inside the CSC (high cleanliness), "unusual" molecules can be formed, e.g. molecules containing noble elements (Fig. 3 top panel).

The only feasible way to remove these contaminants is the IDI method. In the spectrum (Fig. 3 bottom panel), where IDI was used, only $^{124}Xe^+$ ions are seen and can now be clearly identified. The most intense peak disappeared completely, corresponding to a suppression of the contaminants of more than a factor 10^4.

4. Conclusion

The concept of IDI for removing molecular contaminants was implemented within the RFQ beamline of the FRS-IC and tested during several measurements. A reduction of molecular contamination by

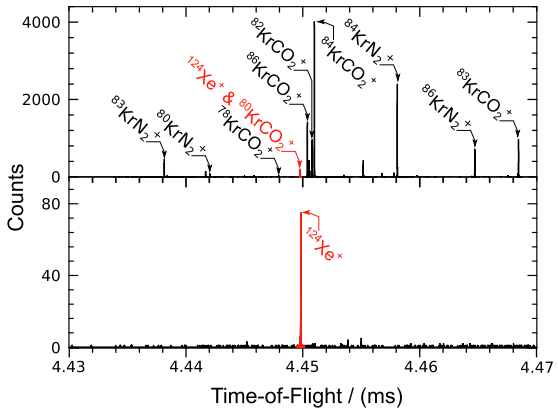

Fig. 3. High-resolution time-of-flight spectrum ($m/\Delta m \sim 150{,}000$) measured with the MR-TOF-MS of ions extracted from the CSC, while ^{124}Xe primary beam is stopped in the CSC. Ions from different mass numbers appear in the spectrum; they do different numbers of turns in the MR-TOF-MS. Top panel: MR-TOF-MS spectrum without using the IDI method. The ^{124}Xe peak (in red) is covered under the $^{80}Kr^{12}C^{16}O_2^+$ peak. Bottom panel: Spectrum with using the IDI method. The measurement time here is four times longer. Contaminants are suppressed by a factor of 10^4.

more than four orders of magnitude was achieved. In this work the MR-TOF-MS was very useful in the development of the IDI method. The method is universal and can be adapted to other facilities.

To further increase the suppression of more strongly-bound molecules, a second CID step is planned in future measurements. This is readily available due to the recent upgrade to the RFQ beamline of the FRS-IC [16].

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