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Single-particle detection of products from atomic and molecular reactions in a cryogenic ion storage ring

C. Krantz[a,1], O. Novotný[a,*], A. Becker[a], S. George[a], M. Grieser[a], R. von Hahn[a], C. Meyer[a], S. Schippers[b], K. Spruck[a,c], S. Vogel[a], A. Wolf[a]

[a] *Max-Planck-Institut für Kernphysik, Saupfercheckweg 1, 69117 Heidelberg, Germany*
[b] *I. Physikalisches Institut, Abt. Atom- und Molekülphysik, Justus-Liebig-Universität Gießen, Heinrich-Buff-Ring 16, 35390, Gießen, Germany*
[c] *Institut für Atom- und Molekülphysik, Justus-Liebig-Universität Gießen, Leihgesterner Weg 217, 35392 Gießen, Germany*

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ABSTRACT

We have used a single-particle detector system, based on secondary electron emission, for counting low-energetic (~keV/u) massive products originating from atomic and molecular ion reactions in the electrostatic Cryogenic Storage Ring (CSR). The detector is movable within the cryogenic vacuum chamber of CSR, and was used to measure production rates of a variety of charged and neutral daughter particles. In operation at a temperature of ~6 K, the detector is characterised by a high dynamic range, combining a low dark event rate with good high-rate particle counting capability. On-line measurement of the pulse height distributions proved to be an important monitor of the detector response at low temperature. Statistical pulse-height analysis allows to infer the particle detection efficiency of the detector, which has been found to be close to unity also in cryogenic operation at 6 K.

1. Introduction

Single-particle counting detectors are important instruments in many atomic and molecular physics experiments on fast-propagating ion beams [1,2]. In such experiments, an ion beam is guided through a target medium which can consist, e.g., of photons, electrons, neutral atoms, or molecules. Reactions of the projectile ions with the target particles typically lead to products of different charge-to-mass ratio. This results in the formation of daughter beams of different ion-optical rigidity compared to the parent, which can be separated from the latter by electric or magnetic analysing fields. At known intensity of the parent beam and thickness of the target, detection of the daughter particles reveals the rate coefficients of the processes involved in their production. Due to the typically low ion numbers and reaction cross-sections, the product detection needs to be done on the single-particle level.

Heavy-ion storage rings enhance such target experiments by their ability to store the projectiles for extended periods of time. Due to energetic processes in the ion source, unknown, highly-excited quantum states are often populated in atomic or molecular ions directly after production. In many cases storage of the ions enables them to reach a well-understood state population by spontaneous decay before undergoing the actual experiment. The extended storage time also allows phase-space manipulation of the ion beam, such as electron or stochastic cooling, or initial-state preparation techniques as required for laser- or collision-driven pump-probe experiments [3,4].

For years, medium-energy magnetic ion synchrotrons have been used very successfully for these kinds of experiments—a remarkable development considering that the technology of those machines was originally aimed at nuclear physics applications [5–7]. Based on that success, a new class of heavy-ion storage rings has emerged, with designs that are optimised for experiments on atomic and molecular physics. They use purely electrostatic ion optics, matching the output energy of relatively simple electrostatic injectors that can be flexibly equipped with state-of-the art molecular ion sources [8–10]. The most advanced set-ups use cryogenic cooling machines to reduce the temperature of their beam guiding vacuum vessels down to values near that of liquid helium [11–15]. On the one hand, this results in a vastly improved residual gas pressure compared to conventional ultra-high vacuum (UHV) set-ups, with correspondingly longer ion storage times [16,17]. On the other hand, storage in such a cold environment allows infra-red-active molecular ions to de-excite to their lowest rovibrational levels prior to starting experiments—a significant improvement over room-temperature ion-storage facilities [18,19].

* Corresponding author.
E-mail addresses: claude.krantz@med.uni-heidelberg.de (C. Krantz), oldrich.novotny@mpi-hd.mpg.de (O. Novotný).
[1] Present address: Marburg Ion-Beam Therapy Centre, 35043 Marburg, Germany.

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The advantages of these cryogenic ion storage rings come with technological challenges with respect to the particle detector equipment. A restriction regarding possible detection principles arises from the low energy of the product particles. Limited by available high-voltage technology, typical kinetic energies in electrostatic storage devices are of order a few keV/u or below. This rules out detection mechanisms where the counting volume of the detector is covered by significant layers of passive material—as is the case for surface-barrier semi-conductor counters [20] and, to lesser extent, for scintillators [21]. Open micro-calorimetric detectors are a promising option for product detection at cryogenic storage rings, which is presently under investigation [22–24]. Their fabrication and operation are however extremely difficult and expensive, such that their use may be limited to selected experiments in the foreseeable future.

Suitable detectors for cryogenic storage rings, which can be widely deployed at acceptable manufacturing and operating costs, are therefore based on surface secondary-electron emission with subsequent multiplication [13,15]. This detection technique has proven itself also at particle energies below 1 keV/u [25], but the low-temperature environment does come with new challenges. Besides engineering problems related to thermal expansion and embrittlement of materials, the efficiency of charge multiplication stages commonly used in low-energy ion detection is known to suffer in cold operation. Due to their semi-conductor-like properties, the electric resistance of micro-channel plates (MCPs) and single-channel electron multipliers (CEMs) rises strongly upon cooling into the cryogenic regime. The high resistance can lead to decreased gain or even complete charge depletion, especially at elevated particle hit rates. Depending on the application, MCPs have been used near ~10 K with varying degrees of success [26–28]. Even less is known about the low-temperature behaviour of CEMs [29].

In a recent publication, we have presented the design of a movable single-particle counting detector for the Cryogenic Storage Ring (CSR) of the Max Planck Institute for Nuclear Physics (MPIK) in Heidelberg, Germany [30]. Here, we report on the first operation of this device under real-life experimental conditions at the CSR.

This paper is structured as follows: In Section 2 we briefly describe the instrument. In Section 3 we present the most important findings from the first operation of the detector system with the storage ring CSR at its lowest temperature of ~6 K. In Section 4 we quantify and discuss the results from that series of experiments, with emphasis on the single-particle detection efficiency of the set-up. Section 5 closes with a summary and outlook onto future developments.

2. Overview of the experimental set-up

The CSR is a fully electrostatic storage ring designed for positive or negative ions of kinetic energies up to 300 keV per unit of charge [15]. The beam guiding vacuum vessel as well as the ion optics contained therein can be cooled to temperatures of ~6 K by a closed-loop liquid-helium refrigerator. For thermal insulation, the beam line is enclosed in an additional isolation vacuum vessel and protected by several layers of black-body-radiation shields.

With an orbit circumference of 35 m, the storage ring (cf. Fig. 1) consists of four identical ion-optical sectors which enclose four field-free drift sections. While one of the latter is occupied by the beam diagnostic instrumentation of the storage ring [31], the other three are free for installation of experimental equipment. The counting detector (lower panel of Fig. 1) is located downstream from an experimental section, within one of the ion-optics sectors of CSR. The technology of the detector system has been described extensively in a dedicated publication [30], hence we limit ourselves to a brief overview here.

Equipped with a 20-mm-wide entrance window for heavy particles, the detector is movable transversely to the beam direction in the plane of the storage ring. It is installed 1.0 m downstream of a short (6°) electrostatic bending dipole of the storage ring. Product particles generated from the stored ions are deflected at a characteristic angle in the dipole element. By placement at a suitable horizontal position, the detector can intercept products with a charge-to-mass ratio that differs from that of the stored parent beam by more than 100% in both directions. Specifically, it can detect neutral products on axis of the ion beam in the experiment as well as, e.g., ionisation products up to the double charge of a stored atomic cation beam [30].

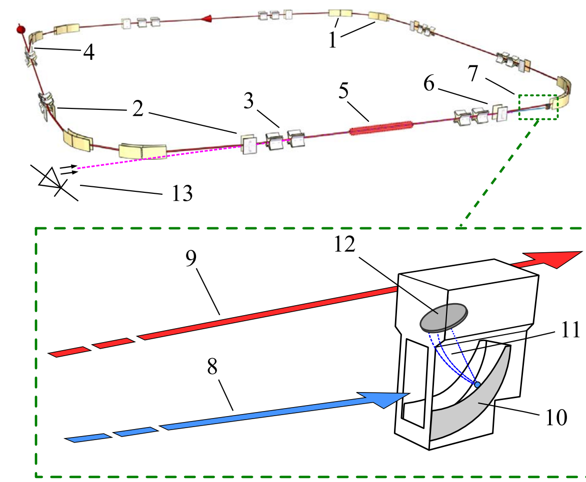

Fig. 1. Schematic view of the experimental set-up, consisting of the storage ring CSR [15] (top) and the COMPACT detector [30] (bottom). The lattice of CSR is a four-fold symmetric, approximately quadratic, and purely electrostatic beam-line, consisting of a total of eight 39° deflectors (1), eight 6° deflectors (2), and eight focussing quadrupole doublets (3). One of the 6° deflectors is fast-switchable to allow ion beam injection (4). In many of the here-reported measurements a laser beam overlapped the stored ions in an experimental target section (5). The 6° deflector (6) directly following the target acts as charge-to-mass analyser preceding the movable COMPACT detector (7). The latter can be positioned to intercept the product particles (8) from atomic reactions in the target while allowing the parent ion beam (9) to circulate unhindered in the CSR. In the detector, the product particles hit a secondary-electron emitting cathode (10). These electrons (11) are accelerated towards a micro-channel plate stack (12) where they are multiplied to form the detector current pulse. For off-beam testing, the detector can be irradiated by an ultra-violet (UV) light emitting diode (LED, 13) installed in the opposite sector of CSR. For details see text and refs. [15,30].

Eventually, the detector is designed to intercept product particles originating from ion-electron interactions in the future electron cooler of CSR—like electron recombination or electron impact ionisation [32,33]. In contrast to the detector set-up, the cooler was not yet operational during the 2015 experiments. Instead, an ion-photon interaction beam line was installed in the experimental CSR section preceding the detector [15]. It allowed to overlap the stored ions at grazing angle with laser beams of various wavelengths that were coupled into CSR using a system of broadband view-ports and mirrors in the cryogenic vacuum chamber. This in-ring laser target was used in experiments on photo-induced electron detachment of stored anions. In addition, without using the laser beams, experiments on auto-detachment and auto-fragmentation of excited molecular and cluster ions were performed using the same set-up. At higher CSR operating temperatures, products of electron transfer from the residual gas to stored cations were observed. For testing purposes, the detector can be irradiated by photons from an ultra-violet (UV, 245(5) nm) light emitting diode (LED) [30]. The UV-LED is located in a room temperature annex of the CSR sector opposite of the detector. The beam of photons from the LED is practically uncollimated and enters the CSR vacuum chamber via a set of UV-grade sapphire view-ports.

The detector employs a variant of the 'Daly' ion detection principle, where incident massive particles impinge onto a secondary-electron emitting cathode made of aluminium [25,30,34]. The secondary electrons released in each hit are accelerated by 1.2 kV towards a small chevron micro-channel plate stack (cf. Fig. 1). The latter acts as secondary-electron multiplier, while being protected from direct hits by