The Cryogenic Storage Ring and its application to molecular ion recombination physics

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Abstract. The Cryogenic Storage Ring (CSR), presently under construction at the Max-Planck-Institut for Nuclear Physics, will allow the storage of large ionic molecules under optimum experimental conditions. The electrostatic beam optics and the presence of a low-energy electron cooler will allow highly-precise recombination experiments with molecular ions of 160 atomic mass units per charge state. The all-cryogenic design of the storage ring will provide unprecedented vacuum conditions and assure long storage times even for very heavy ion beams. Suppression of the black body radiation background of the beam pipe in combination with electron cooling will give access to internal ion temperatures of 10 K and allow state-selective experiments on infrared-active species, impossible in present-day room-temperature storage rings. We give an overview of the CSR project, point out the scientific opportunities arising from its unique design, and give an outlook on possible first molecular recombination experiments after commissioning of the storage ring.

1. Introduction
Since the 1990s, electron cooler heavy-ion storage rings, besides their applications in other fields of molecular, atomic and nuclear physics, have contributed some of the most important experimental data on the dissociative recombination (DR) of molecular ions with electrons. Using relatively simple systematics, the merged-beams technique provides an energy resolution in the order of 1 meV in slow electron-ion collision reactions [1]. Storage ring experiments are thus a particularly reliable source of low-temperature DR rate coefficients, as needed e.g. for the understanding of reaction kinematics in the chemistry of cold interstellar media, star forming regions, or circumstellar disks [2]. Moreover, the well-defined initial-state conditions that can be achieved by radiative and electron cooling of molecular ions in the storage ring allow precision studies of the DR process itself. Due to the high velocity of the merged electron and ion beams in the laboratory frame, DR product detectors cover an effective 4π solid angle. Setups with neutral fragment imaging and timing capabilities allow complete reconstruction of the DR breakup momentum distributions [3, 4, 5]. In studies of low-energy neutral-product DR, these systems thus fulfil a role similar to that of COLTRIMS in the case of high-velocity charged-product reactions.

The value of the experimental input that magnetic heavy ion storage rings [6], like the Test Storage Ring (TSR) in Heidelberg or CRYRING in Stockholm, have provided to the field of dissociative recombination physics [7] is undisputed. Designed in the 1980s, these machines
rely on the principle of magnetic deflection and focusing of the stored ions. This storage technology was originally developed to fulfil the demands of nuclear and particle physics, i.e. handling of species with high charge-to-mass ratios. As research in the field of molecular ions progresses towards more and more complex but still only singly (or weakly) charged systems, the design of classic storage rings becomes a limitation with regard to the nature of the species that can be successfully studied. In addition, the rotational transition energies of molecules are so low that rotations can be populated by the black body radiation from the room-temperature vacuum chamber walls of existing storage rings, which complicates the experimental initial-state conditions. The Cryogenic Storage Ring (CSR) presently under construction [8, 9] at the Max-Planck-Institute for Nuclear Physics (MPIK) will address these limitations and provide the experimental platform for the next generation of precision merged-beam DR measurements.

2. Limitations of the present electron cooler storage ring TSR

We will outline the limitations imposed on DR experiments by magnetic storage rings using as an example the Test Storage Ring TSR of the MPIK in Heidelberg, Germany.

2.1. Limits of the ion beam optics

Magnetic storage rings are momentum selective. The storage condition for particles of charge-to-mass ratio \( q/m \) is fulfilled if the ion momentum \( p \) divided by the charge state \( q \) matches the magnetic rigidity \( \rho B \) of the ring:

\[
\frac{p}{q} = \frac{\gamma mv}{q} = \rho B ,
\]

(1)

where \( \gamma \) is the Lorentz factor. The maximum rigidity \( \rho B_{\text{max}} \) is defined by the maximum induction \( B_{\text{max}} \) of the dipole magnets and the curvature radius \( \rho \) of the closed orbit in the storage ring. For the TSR, which was designed for experiments on highly charged atomic ions (HCI), \( \rho B_{\text{max}} \approx 1.4 \text{Tm} \). This value is adapted to the range of HCI beam energies that can be produced at MPIK: with typical charge-to-mass ratios of 0.33 \( \text{u}^{-1} \) to 0.5 \( \text{u}^{-1} \), HCI can be stored in the TSR at velocities \( v \sim 0.1 \text{c} \) which conveniently match the typical output velocity of the accelerator facilities at MPIK. Storage times for HCI in the TSR can be very long, as ion loss cross sections due to reactions with residual gas molecules become small at elevated collision velocities.

For molecular ions, the (positive) charge state is normally limited to unity, as stripping of more than one valence electron from the parent molecule is normally not possible without disrupting the chemical bonds. Even for relatively small and simple molecules, the charge-to-mass ratios of the corresponding ions become very small compared to HCI. E.g. for \( \text{D}_2\text{O}^+ \), \( \text{DCND}^+ \) and \( \text{CF}^+ \), three of the most heavy systems that have been studied at the TSR, \( q/m \) reduces to 0.045 \( \text{u}^{-1} \), 0.033 \( \text{u}^{-1} \), and 0.032 \( \text{u}^{-1} \), respectively. As can be seen from eq. (1), the maximum storage velocity at given maximum magnetic rigidity behaves like

\[
v_{\text{max}} \sim m^{-1} .
\]

(2)

Given this decrease of the beam velocity with rising ion mass, it is not surprising that storage times of heavy molecular species are short compared to HCI, typically of a few seconds only, as the residual gas induced ion loss rate rises at lower velocities. With respect to DR experiments, there is consequently an effective maximum ion mass. It is reached at the point where the ion storage time \( \tau_{\text{ion}} \) becomes shorter than the internal and phase-space cooling times \( \tau_{\text{cool}} \). Beyond this point, control of the initial-state conditions of the experiment is no longer assured, strongly affecting the precision of possible DR measurements.
2.2. Limitations of the room-temperature environment
At a given ion velocity, the beam lifetime in a storage ring is defined by the mean free path length of the ion in the residual gas of the beam pipe. The TSR features a vacuum pressure of a few $10^{-11}$ mbar all along its 55-m-long vacuum chamber. This value is close to the optimum pressure that can be achieved in a room-temperature setup, as the residual gas is in this case dominated by H₂, the main degassing component of the stainless steel chamber walls.

A more fundamental limitation of the 300-K environment is related to molecular physics itself. While vibrational excitations are too high-energetic to be excited by far-infrared black body radiation, so that molecular ions can be safely assumed to relax to their vibrational ground state within the storage time in the TSR, this is not true for rotational excitations. Although rotational single-state experiments have been possible in the TSR in the special case of molecules with zero electric dipole moment using a state-selective ion source [10], the rotational states of stored ions generally have to be expected to assume a population that is in thermal equilibrium with the background radiation of 300 K. Due to this initial-state multiplicity, theoretical predictions of DR rate coefficients have to be convolved with a relatively broad range of rotational states, which fundamentally limits the quality of experimental data as theory benchmarks.

3. The Cryogenic Storage Ring project
The Cryogenic Storage Ring CSR is designed to provide the platform for the next generation of precision experiments with molecular ions. The CSR is an all-cryogenic electrostatic electron cooler storage ring [8, 9] and will feature experimental facilities for studies of ion-neutral, ion-photion and ion-electron interactions.

3.1. Beamline
Unlike in the case of magnetic deflection, the electrostatic beam optics is energy-selective. At a given charge state, $q$, the maximum kinetic energy, $E$, is defined by the bending radius, $\rho$, and the maximum electric field, $E_{\text{max}}$, that can be applied within the deflector elements:

$$\frac{E}{q} \leq \rho E_{\text{max}}.$$  \hspace{1cm} (3)

For the CSR, $\rho E_{\text{max}} = 300$ kV, hence the ring can store particles at maximum kinetic energies of 300 keV per charge state independent of their mass. In contrast to the Lorentz force, the electric deflecting force does not depend on the ion momentum. As a consequence, the dependence of the maximum storage velocity on the particle mass is weaker in comparison to (2):

$$v_{\text{max}} \sim m^{-1/2}.$$  \hspace{1cm} (4)

Thus, although in an electrostatic ring the velocity of light ions is limited compared to a magnetic ring by the typical value of $\rho E_{\text{max}}$, very heavy species can be stored at higher velocity. While several electrostatic storage rings have already been commissioned [11, 12] or are currently under construction [13], the CSR will be by far the largest device of this type, with a closed-orbit circumference of approximately 35 m and, to-date, the highest value of $\rho E_{\text{max}}$.

In order to achieve long storage times even for very heavy species, the vacuum system of the CSR is designed to provide very low residual gas pressures. Using a combination of ion-getter pumps, non-evaporative getter (NEG) coatings and UHV-bakeable cryopumps, the residual gas pressure will be reduced to $\sim 10^{-11}$ mbar during room-temperature operation. Further reduction of the residual gas density is achieved by cooling the entire vacuum chamber and ion optics to below 10 K using a superfluid helium cryocooler [8]. Therefore, the beam line has to be enclosed in a cryostat consisting of two layers of thermal radiation shields, several layers of superinsulation...
and a large isolation vacuum chamber. In cold operation, the dominating residual gas component \( \text{H}_2 \) can no longer be pumped by NEG coatings. Instead, it will be removed by cryocondensation on dedicated copper surfaces in the beam line which are cooled to below 2 K by direct contact with the superfluid helium circuit [14]. This reduces the pressure in the ring to approximately \( 10^{-13} \text{ mbar} \) (room temperature equivalent pressure yielding identical residual gas density), thus improving ion storage times by more than 2 orders of magnitude compared to warm operation. The cryogenic and vacuum concepts of the CSR have been tested in a prototype setup and have met their design goals, as reported recently [15].

A 300 kV injector platform is presently under construction at MPIK. Several types of ion sources dedicated to CSR experiments are planned, including, but not limited to, an EBIT for HCI production and specialised sources for production of cold molecular ion beams. The latter can be supersonic expansion jet [16] sources, electrospray ion sources for generation of large molecular ions [17], or buffer-gas cooled radiofrequency traps which allow relaxation of initially hot ion bunches prior to injection into the CSR [18]. The 300 kV high-voltage platform is sufficiently spacious for several sources to be installed simultaneously, which ensures flexibility in the planning of CSR experiments.

### 3.2 Experimental facilities

The CSR has a quadratic layout with four straight beamline sections enclosed by four identical corner sections. The latter contain all of the beam bending and focussing optics. Furthermore, the ion beam injection will be performed in a corner section by fast switching of one of the bending elements. Thus, four \( \sim 2\text{-m-long} \) linear sections are available for experimental instrumentation.

A first straight section is dedicated to ion beam diagnostics, featuring a capacitive pickup electrode for highly-sensitive current measurements. A second section houses a transverse gas-jet target backed by a reaction microscope for studies of fast ion-neutral interactions. The linear chamber following the injection corner will be available for future experimental equipment, but will also include a collinear merging region with a neutral beam. The latter will be fed through the injection beamline at variable velocity and allow experiments on slow ion-neutral collisions.
The fourth experimental section contains the electron cooler and thus the key ingredient to recombination experiments. Electron cooling of slow, massive ions is a challenging task. While pioneering work with slow merged electron beams has been done at the KEK electrostatic storage ring [19], CSR is the first machine of this kind to aim at strong phase-space compression by electron cooling, using long ion storage times and specialised sources for cold, slow electrons. For a wide range of molecular species, this combination of a cooled ion beam and a very slow merged electron beam make the CSR the only next-generation storage ring suited for experiments on low-energy DR.

3.3. Electron cooler

The electron cooler is designed to operate at electron energies $E_e$ from 1 eV to 1 keV. Considering the velocity-matching condition $E_e = E_i m_e/m_i$ (with $e$ and $i$ denoting the energy and mass of electron and ion, respectively), the energy range $E_e > 160$ eV is needed only for the generation of non-zero collision energies in merged-beams experiments. For cooling of singly charged, massive molecular ions, electron energies of only a few eV or less are necessary. The electrons are overlapped with the stored ion beam using a novel magnetic beam-merging scheme, which minimises disturbance of the ion trajectory [20].

Well-controlled manipulation of very slow electron beams is difficult, as effects such as the beam space-charge or workfunction differences between the cathode and the beam pipe materials, which are only minor corrections in existing coolers, become dominant contributions to the final electron energy. In addition, the available electron density at low energy is severely limited by the electron gun pervance, as the extraction voltage at the cathode cannot be arbitrarily higher than the final acceleration potential. In the absence of high electron densities, the electron temperature has to be as low as possible in order to assure an acceptable cooling force [21]. Hence, the CSR electron cooler will employ a cryogenic GaAs(Cs,O) photocathode as an electron source, similar to the existing high-resolution electron target of the TSR, which has already demonstrated its ability to cool slow ion beams within a few seconds of storage time [22].

Recent experiments performed at the existing photocathode electron beam setup of the TSR have established a procedure that allows reliable production of very slow electron beams of well-defined energy and density, taking space-charge and contact potential effects into account [23]. The measurements have shown that stable beams with a narrow velocity-spread can be obtained at mean kinetic electron energies down to 1 eV or lower. Given the CSR’s ion energy limit of 300 keV, the mass range of singly-charged ion species for which velocity matching can be obtained in the electron cooler extends to at least 160 u.

3.4. Detectors

Similarly to the other experimental facilities, the electron cooler will be backed by particle detectors for rigidity-changed or neutralised products in the bending section located after it in the CSR. The low ion beam energy in combination with the cryogenic extremely high vacuum environment severely limits the spectrum of available detector technologies that can be employed.

Rigidity-changed products, such as those arising from dissociative excitation or ion-pair formation events, will be recorded using counting detectors based on charge multiplication by secondary electron emission from a converter electrode. The secondaries are then detected using an MCP backed by a collector anode. Secondary electron emitters have the advantage of not presenting any dead layer which the impinging products have to traverse for successful detection, so that also low-energetic particles can be counted. Detectors based on this principle [24] have already been successfully used at the TSR for several years.

Neutral DR fragment imaging will be performed using an MCP backed by delay-line readout anodes, which provide multi-hit time and position sensitivity and thus allow measurement of kinetic energy releases in breakup events, in addition to the pure recombination rate counting...
capability. Successful operation of such a detector in a cryogenic environment has already been reported in [25] and is currently being tested in the CSR prototype (described in [15]).

The explosion cone aperture for detectable DR fragments is limited to $\pm 1^\circ$ by the geometry of the CSR bending elements. For symmetric breakups of the parent molecule into neutral fragments of approximately equal mass, this limitation is insignificant, as only unrealistically large kinetic energy releases of several tens of eV could cause the recombination products to miss the imaging detector. For strongly asymmetric breakups, where the mass of one or several fragments is much smaller than that of the parent molecule, products can miss the detector if the energy release of the recombination reaction exceeds a critical value. The maximum allowed kinetic energy release is in this case proportional to the mass of the light fragment and inversely proportional to the mass of the parent molecule. If, e.g., a single hydrogen atom is emitted from a parent of mass 100 u, stored in the CSR at 300 keV, the geometrical efficiency will be limited already for kinetic energy releases exceeding $\sim 1$ eV. However, for those events where both particles do hit the MCP, the large velocity difference between the fragments will in this case allow inclusion of time-of-flight measurements in the analysis of the kinetic energy release. Thanks to the much slower ion beam velocity of the CSR, this will be possible with much higher precision compared to present fast-beam setups.

While an MCP-based fragment detector lacks the product mass sensitivity of the present setup at TSR [4], fragmentation branching ratios can be obtained from statistical analysis of the fragment velocity distributions if the emittance of the parent ion beam is small, i.e. if the centre-of-mass of the molecular breakup is sufficiently well-defined by electron cooling [5]. Furthermore, the cryogenic environment of the ring offers realistic opportunities for future fragment imaging setups involving low-temperature micro-calorimeters [26], which can offer even higher fragment mass resolution than present surface-barrier semiconductor detectors.

4. Opportunities for molecular ion recombination experiments at the CSR
CSR is currently the only next-generation electrostatic storage ring for which also DR experiments are planned. Its high mass acceptance combined with electron cooling and the cryogenic environment opens the way to a range of experiments not feasible in present setups.

4.1. DR in a cold environment
With its cold vacuum chamber, the CSR will be the first electron cooler storage ring that truly simulates the environmental conditions found in the interstellar medium (ISM). It will thus allow the study of DR reactions relevant to the chemistry of the ISM without interference by an unrealistic 300-K black-body background radiation.

Aided by superelastic electron collision (SEC) cooling [27], by the quasi radiation-free storage volume, and by the long storage times in the extremely high vacuum of the CSR, initially hot molecules produced in the ion sources will be allowed to relax down to their rovibrational ground-state. Experiments seeking to benchmark theory will greatly profit from this possibility to prepare and maintain pure internal state populations. Possible candidates for first state-selective DR experiments at the CSR are HD$^+$ and HeH$^+$, ions which are easy to produce and store at high current intensity and which are known to efficiently relax radiatively. Theory of the simple molecule HD$^+$ is in fairly good agreement with present room-temperature convolved DR rate coefficients and could thus benefit most from single-state recombination data. For HeH$^+$, updated theoretical DR rate coefficients have been published recently, but can, up to now, only be compared to rotational-state convolved experimental datasets [28].

4.2. DR of massive molecular ions
The CSR will extend the mass range of molecular ions accessible for precision DR studies. It will be able to store heavy, slow ion species long enough to render electron cooling possible.
Internal relaxation of these ions will be achieved by combination of the cold ion sources on the injector platform (Sec. 3.1), which pre-cool all internal molecular modes including those of low or vanishing infrared activity, and the low-temperature environment in the storage ring, which prevents the re-excitation of modes with stronger infrared activity. Candidates for experiments taking advantage of the high mass range of the CSR include e.g. the carbon fluorine ions CF$_n^+$, which play an important role in industrial plasmas and of which only the lightest specimen CF$^+$ could be studied at a high energy resolution in the TSR up to now. Another promising field is the DR of large hydrocarbon or small biomolecule ions. Finally, even electron collision studies of cluster ions will be an option at the CSR, which will allow, e.g., to study the dependence of the recombination process on the internal cluster temperature.

5. Present status and outlook

We have introduced the CSR project as the future platform for next-generation precision DR experiments. The CSR and its ion source array are presently being built at the MPIK (see fig. 1) with the first corner section to be completely assembled in early 2011. The remaining parts of the ring lattice as well as the electron cooler and the detector systems will be constructed during the next two years, such that first stored beams are expected at the beginning of 2013.

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