Electron Cooling of Molecular Rotations in the Low-Energy Electrostatic Storage Ring CSR

Andreas Wolf
Cryogenic Storage Ring (CSR Heidelberg) electron cooling team

Max Planck Institute for Nuclear Physics, Heidelberg, Germany

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Outline

- Energy damping and electron collisions

- The CSR laboratory

- Rotational cooling of CH^+ ions
  CSR measurement and quantitative analysis

- Conclusions

Energy damping and electron collisions

Energy damping in ion beams

… or ions in a plasma environment

External motion

Ion charge

"Natural" non-conservative forces on storage-ring orbit
very small (… synchrotron damping …)

acceleration

$$P = \frac{2}{3}\,\frac{q^2}{4\pi\epsilon_0}\,\frac{a^2}{c^3}$$

Larmor formula

Coulomb-type friction → electron cooling
energy carried away by electrons

Ionic charge (monopole) → Long-range Coulomb force

Internal motion

Atomic or molecular dipole moment

Natural damping by dipole oscillations (spontaneous emission)
- fast for near optical frequency (electronic [nanoseconds], vibrational [milliseconds])
- slow for rotation ← minutes … hours

Quantum levels

E_e

$$\omega = \frac{E_e - E_g}{\hbar}$$

E_g

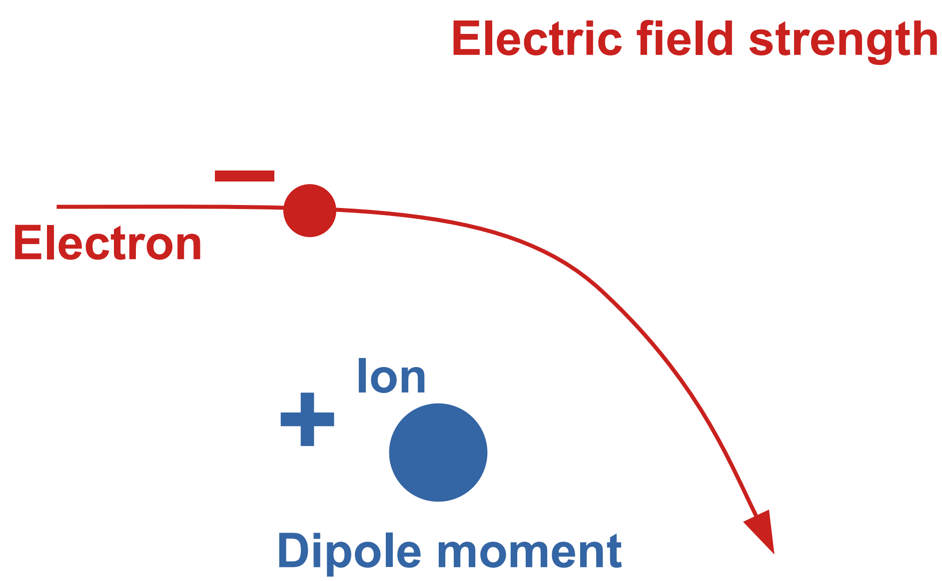
$$a \propto \omega^2$$

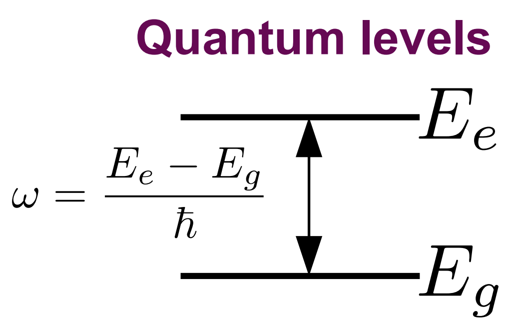
$$P \propto \omega^4$$

Electron collisions → excitation or *damping* of internal motion

Oscillating multipole moments → Short-range forces

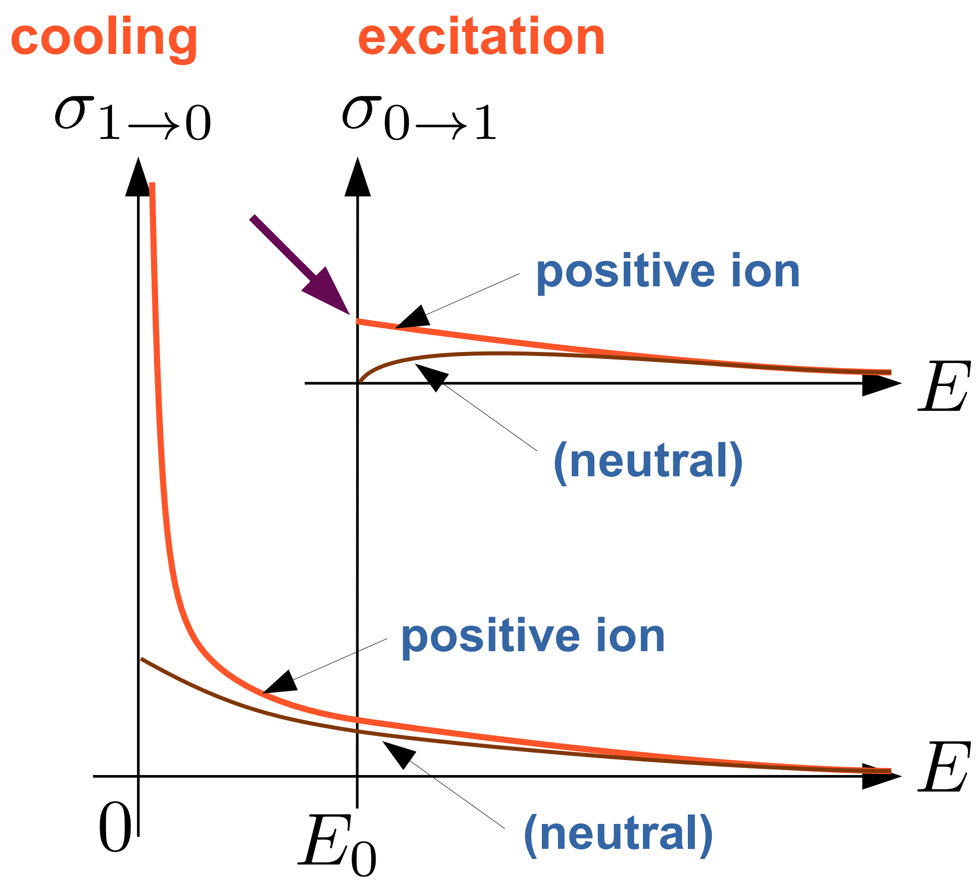
Charged-particle collisions and internal states

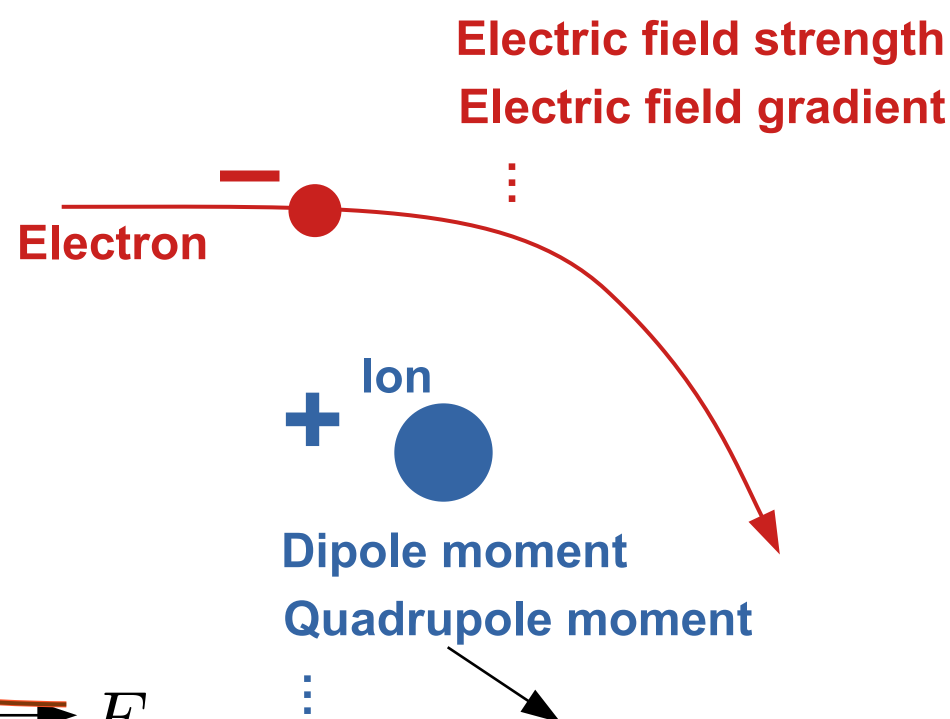




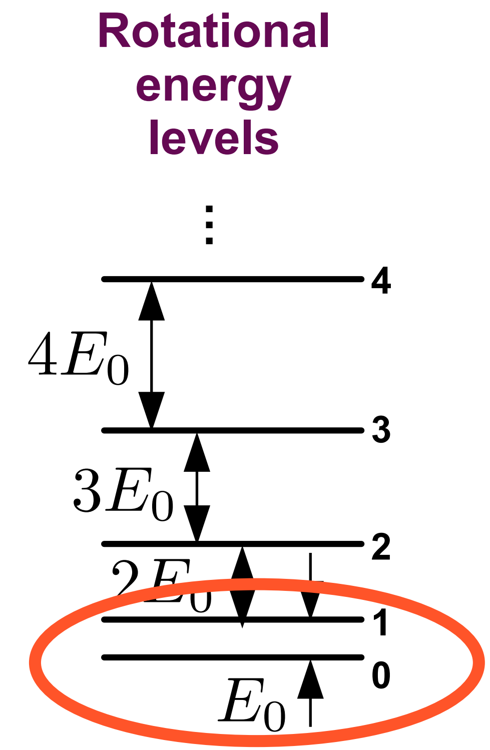
Charged-particle collisions and internal states

Cross sections





$$\sigma_{0\to 2} \approx \sigma_{0\to 1}/10$$



$E_0 \approx 3.5\ \mathrm{meV}$
(case of CH^+)

Rough approximation

$$\sigma_{0\to 1}(E_0) \approx 10^{-16}\,\mathrm{cm}^2 \cdot \frac{\mu_0^2}{E_0}$$

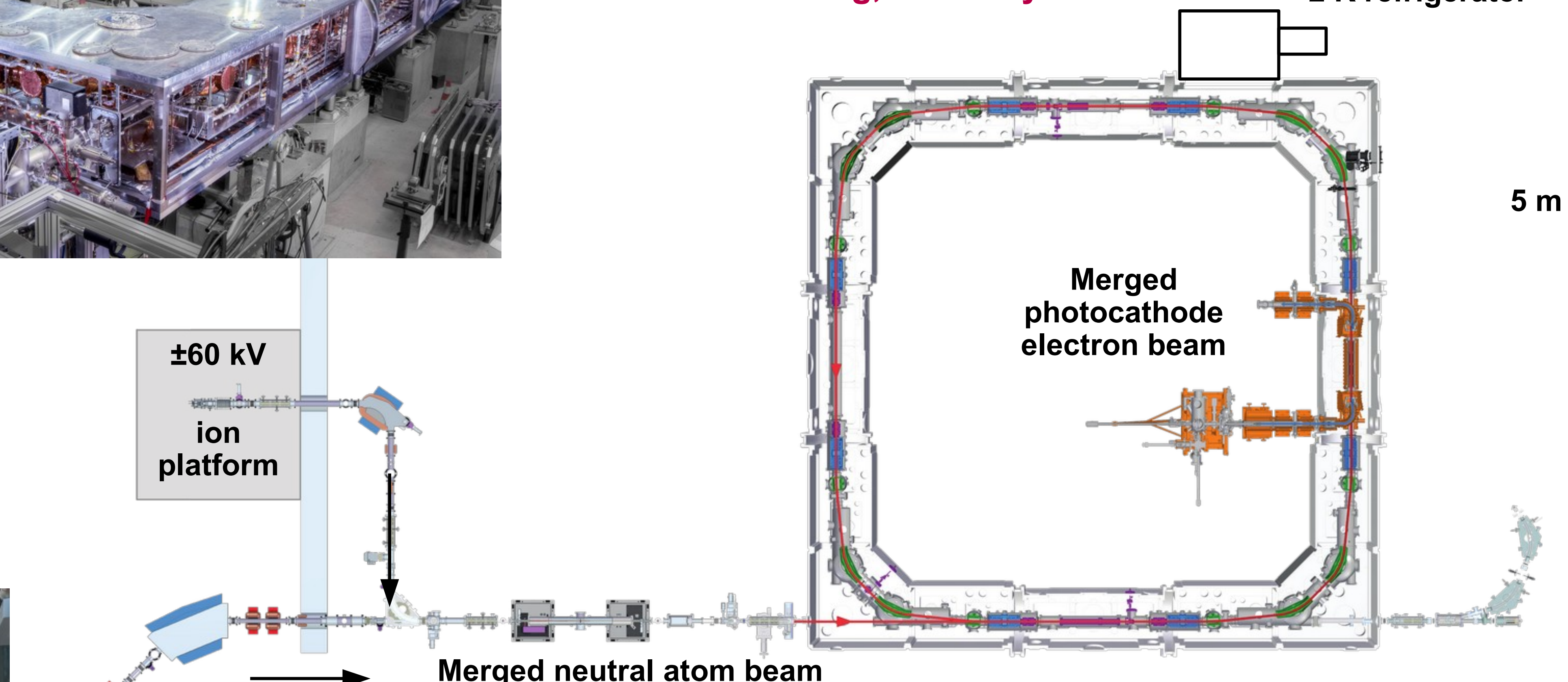
μ_0 in ea_0 (a_0 = Bohr radius)

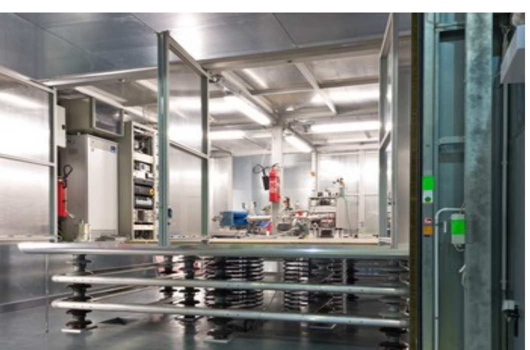
E_0 in units of $27.2\ \mathrm{eV}$

The CSR laboratory

CSR laboratory

Max Planck Institute for Nuclear Physics
Heidelberg, Germany





35 m circumference
cryogenic electrostatic
ion storage ring (300 keV·*q*)

R. von Hahn et al.,
Rev. Sci. Instrum. 87, 063115 (2016)

CSR – machine view

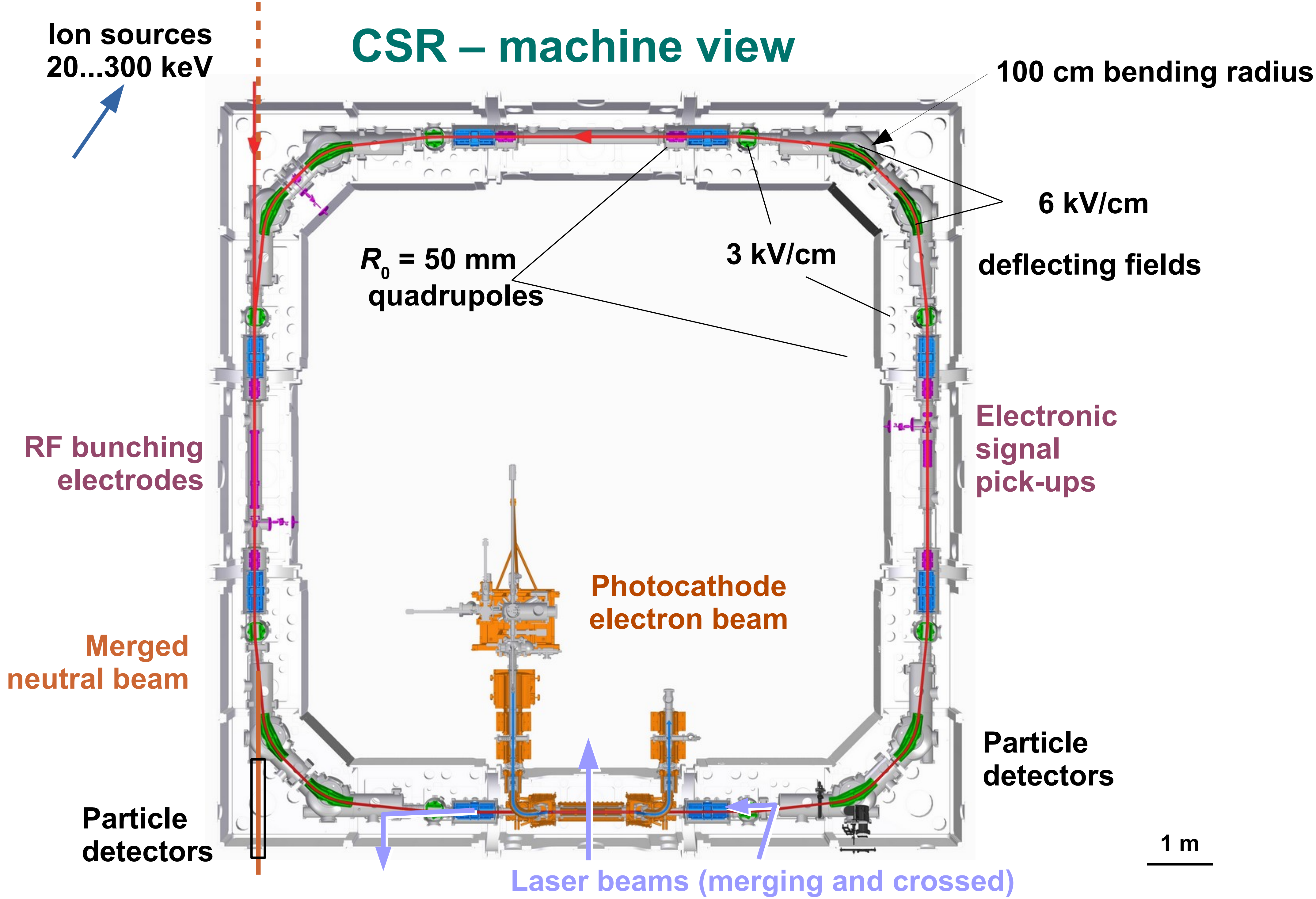
Examples:

Mass number *A*

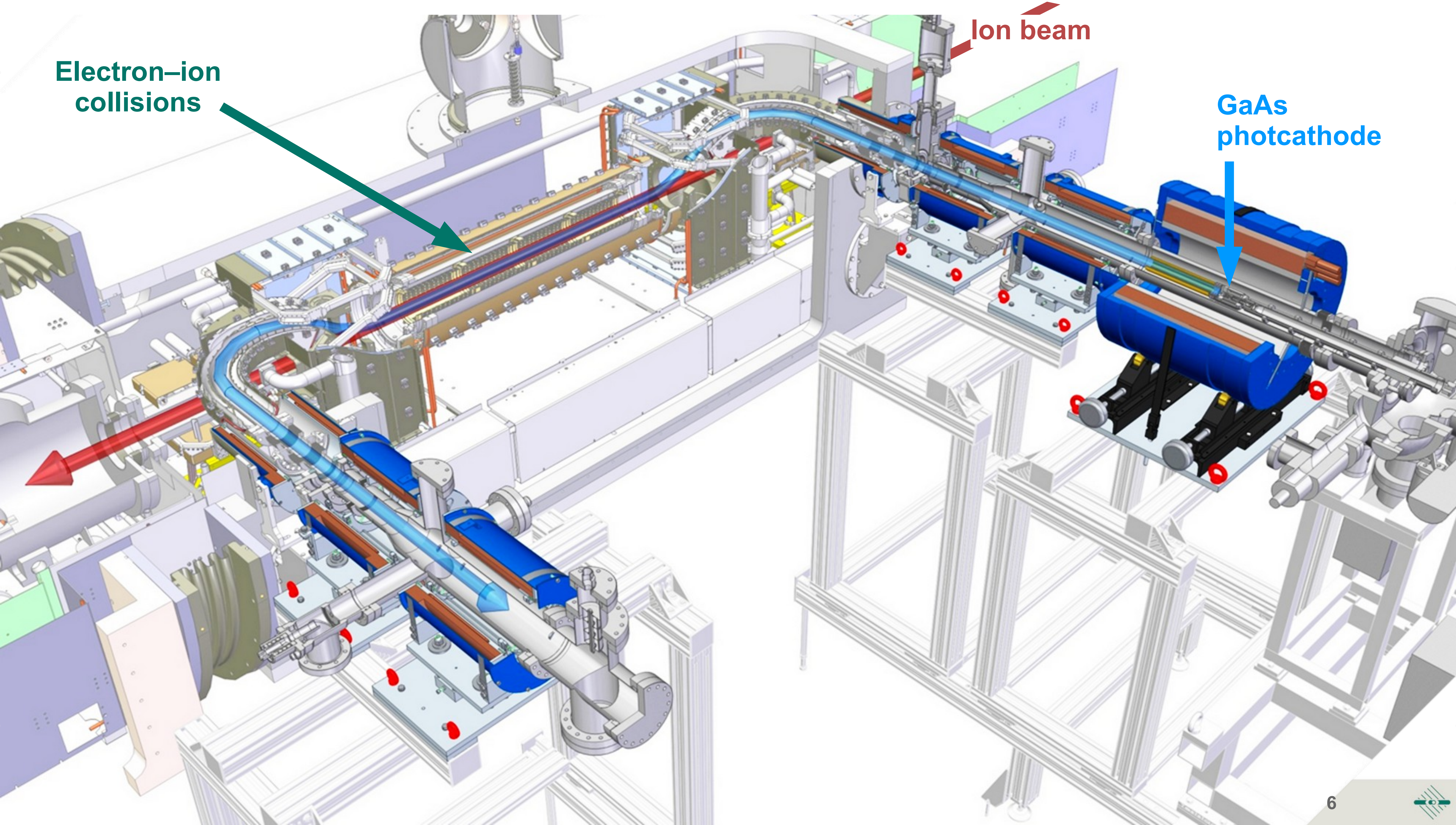
18 O^+
40 Ar^+

5 HeH^+
13 CH^+
17 OH^+
34 $^{16}O^{18}O^+$
64 TiO^+

17 OH^-
24 C_2^-
26 CCH^-
108 Al_4^-
216 Ag_2^-

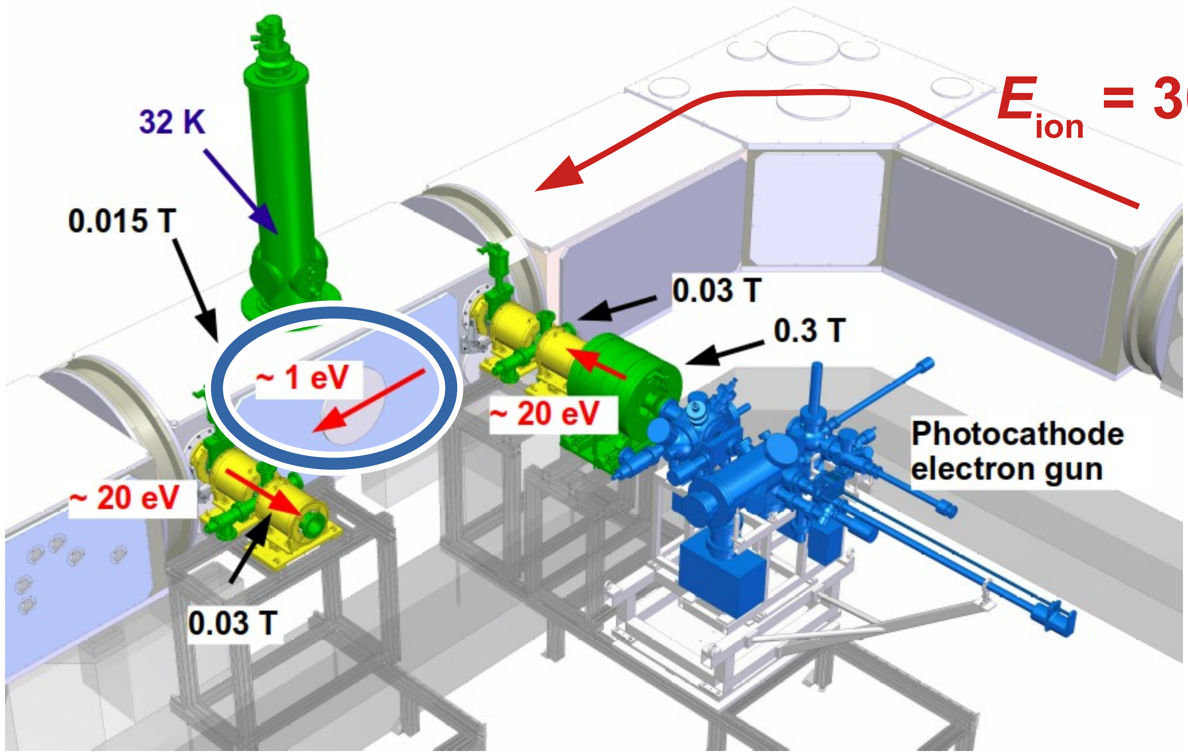


CSR photocathode electron cooler



Velocity-matched merged beams at low energy

C. Krantz, S. Vogel
S. Lohmann
P. Wilhelm
D. Paul
O. Novotný



Last week (end Oct 2021): cooling force measurements for a 4.8 eV electron beam

Mass number *A*

18 O^+
40 Ar^+

$$E_e = \frac{E_{ion}}{A\,u}$$

5 HeH^+
13 CH^+ ← 12.6 eV
17 OH^+
34 $^{16}O^{18}O^+$ ← 4.8 eV
64 TiO^+ ← 2.6 eV

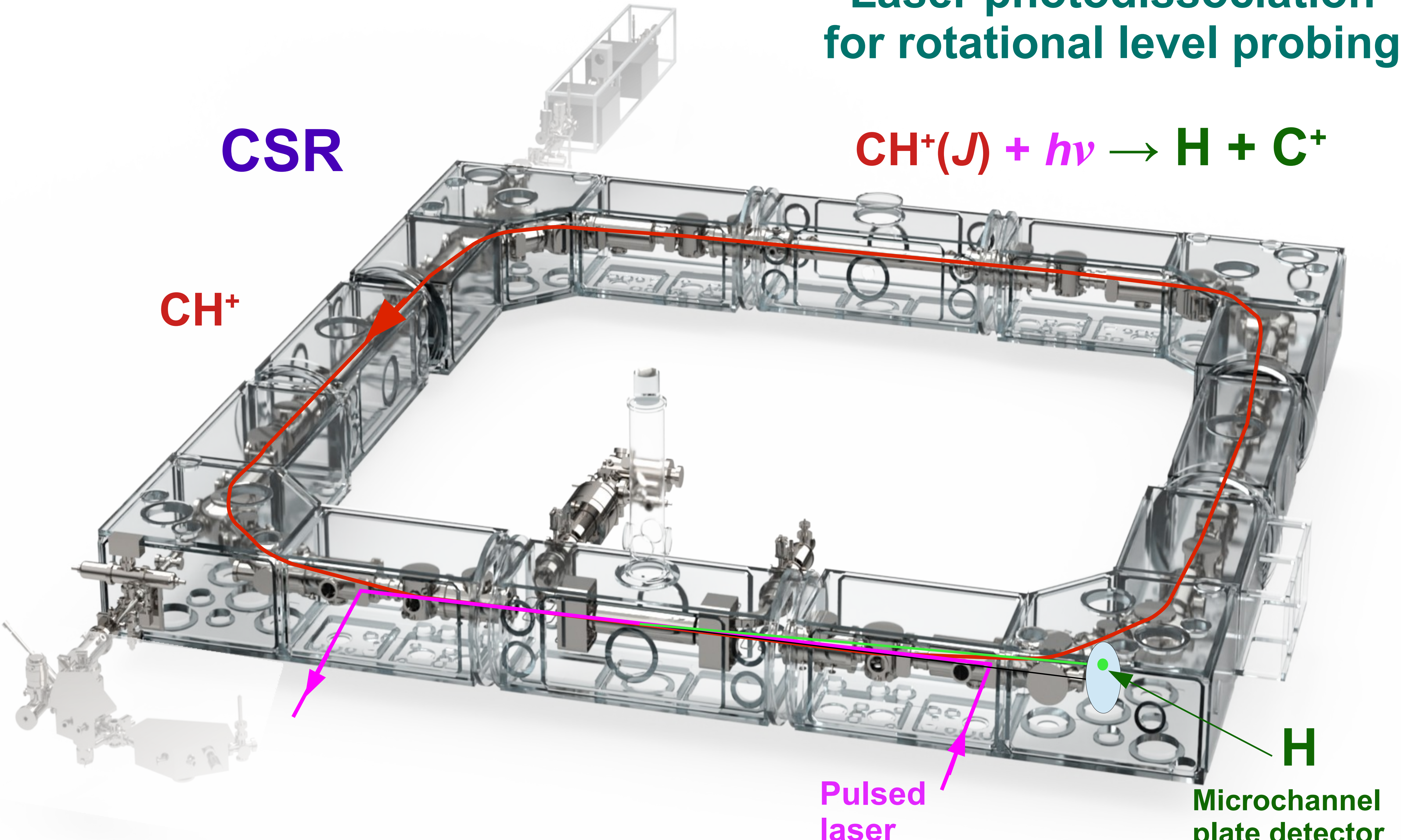
17 OH^-
24 C_2^-
26 CCH^-
108 Al_4^-
216 Ag_2^-

1 eV photocathode electron beam deceleration and transport:
A. Shornikov et al., PRSTAB 17, 042802 (2014)

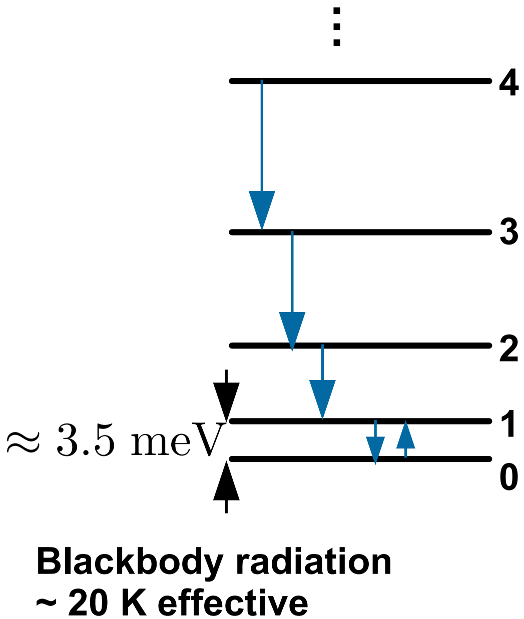
Rotational cooling of CH^+ ions

Radiative cooling only

Laser photodissociation for rotational level probing



Laser photodissociation for rotational level probing

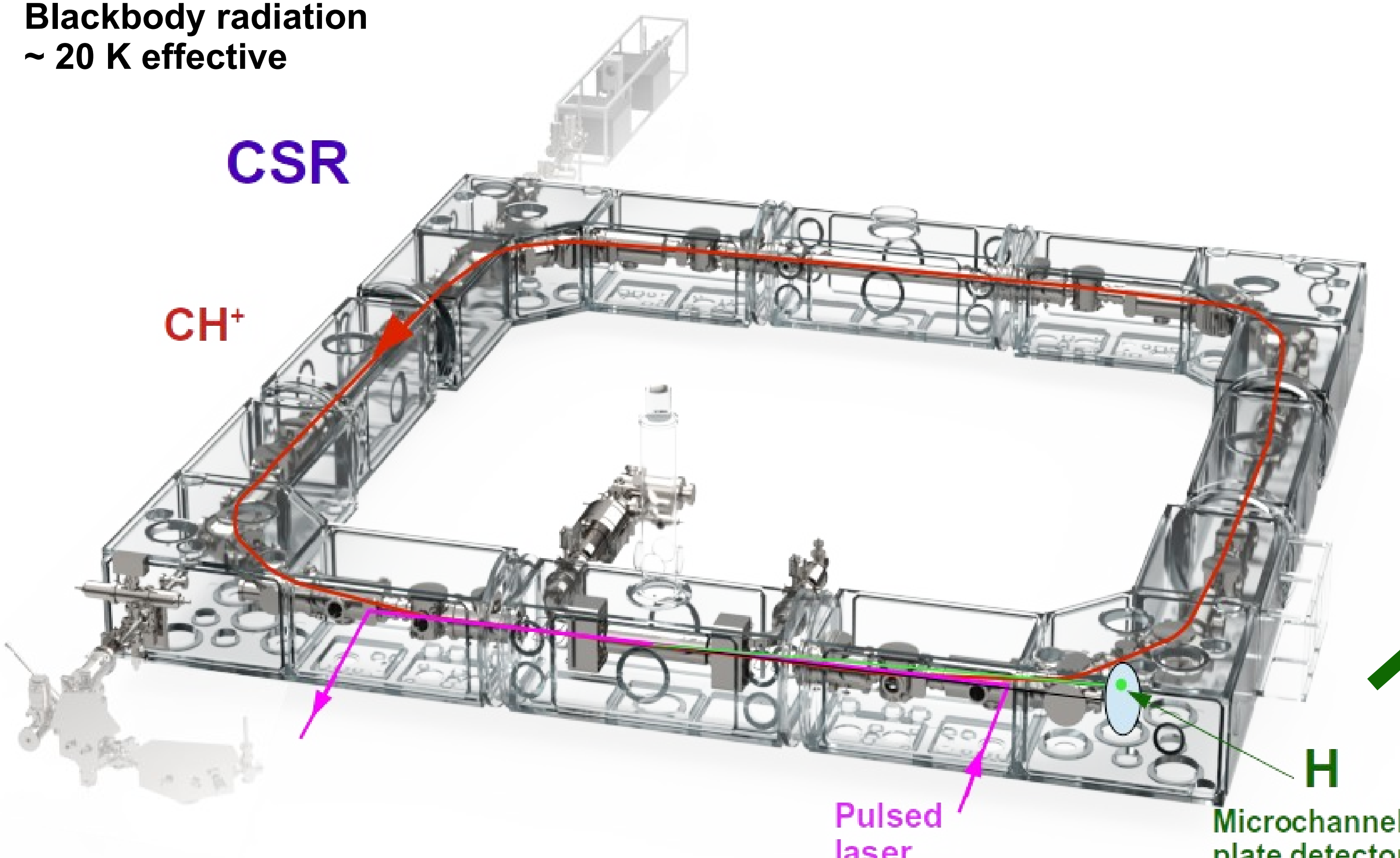


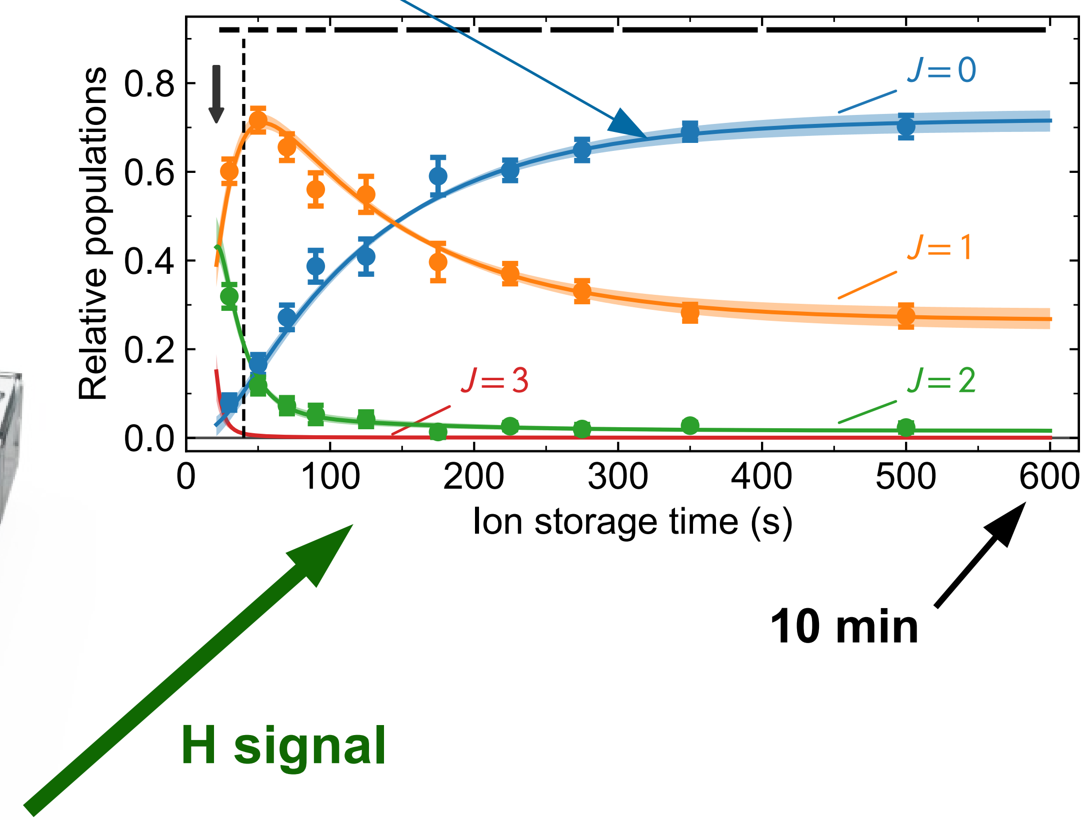
Probing wavelenghts for $J = 0, 1, 2$

Rotational cooling cascade by infrared emission

Modeling by rate equations

$CH^+(J) + h\nu \rightarrow H + C^+$

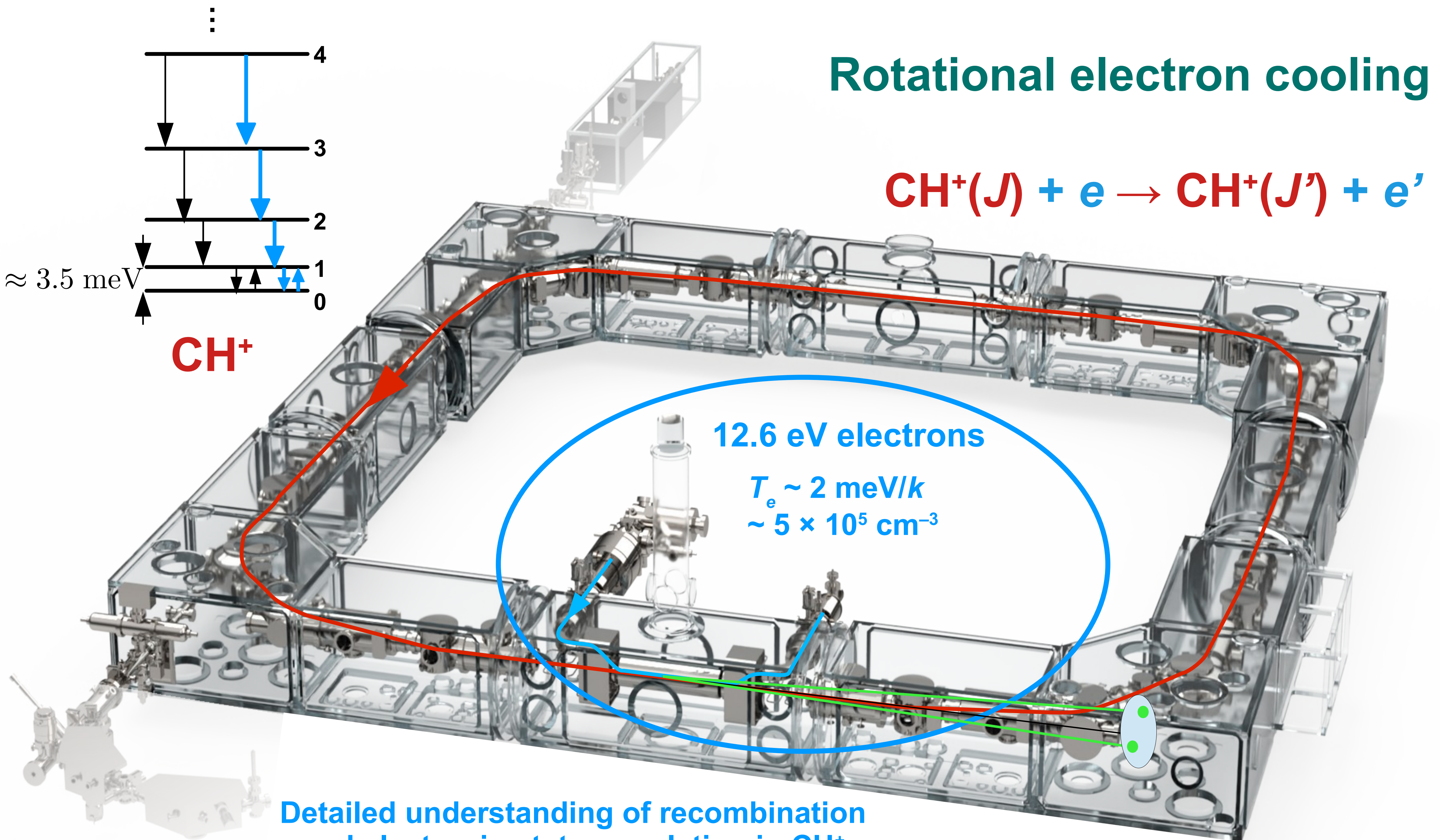




Rotational cooling of CH^+ ions

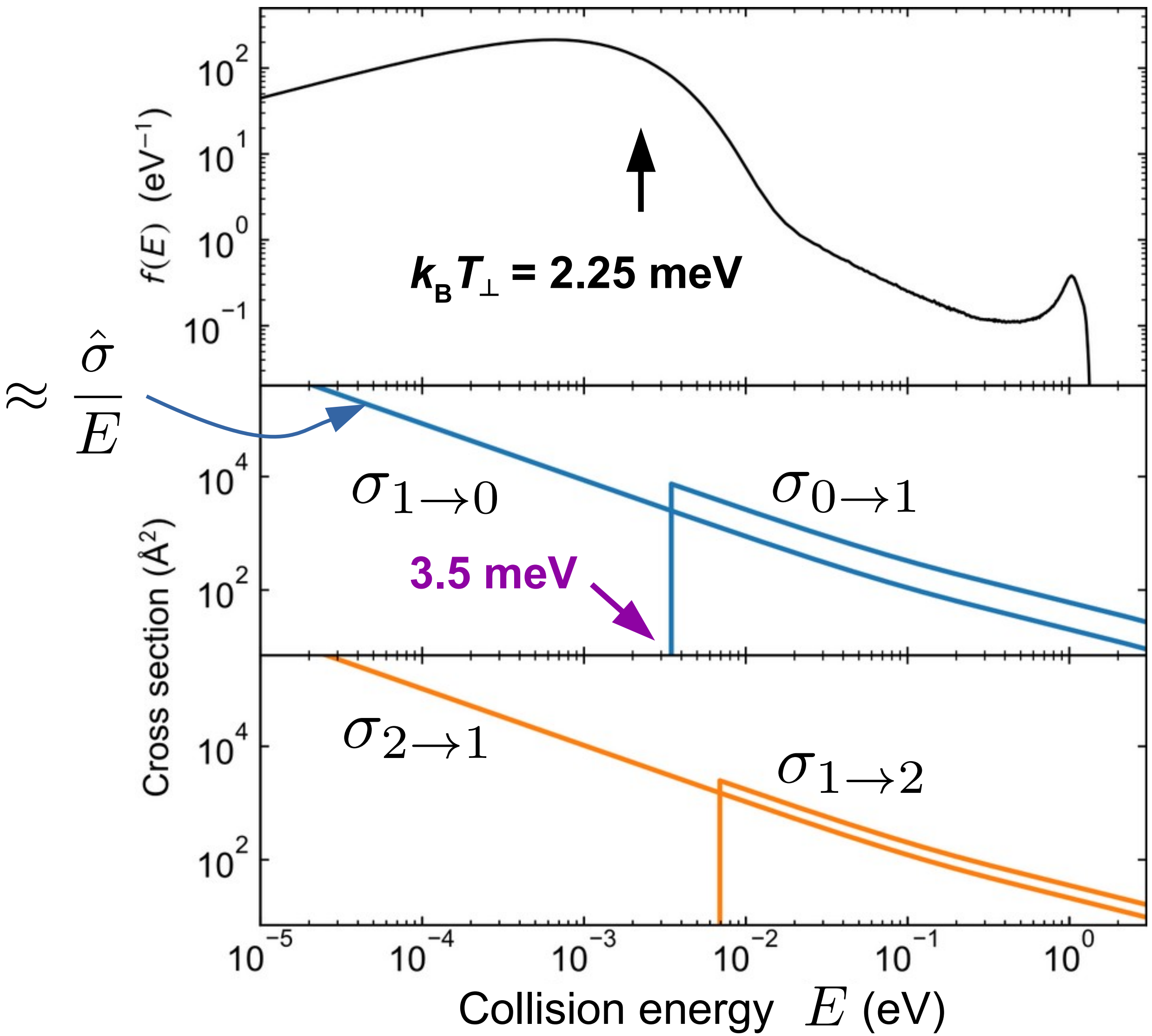
Rotational electron cooling measurement

Rotational electron cooling



Detailed understanding of recombination and electronic-state population in CH^+ from recombination measurements …

Electron interaction cross sections



Co-moving electron energy distribution

$$k_B T_\perp = 2.25 \text{ meV}$$

$$k_B T_\parallel = 0.6 \text{ meV}$$ **(effective)**

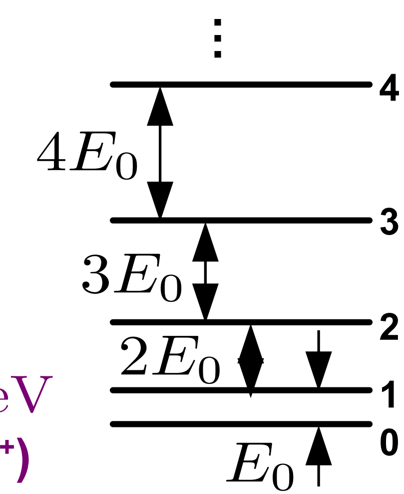
$$n_e = 7 \times 10^5 \text{ cm}^{-3}$$ **80 cm merging**

$$n_e = 4 \times 10^4 \text{ cm}^{-3}$$ **ring-averaged**

Cooling and excitation cross sections

Calculated in dipole–Born approximation (Coulomb scattering waves)

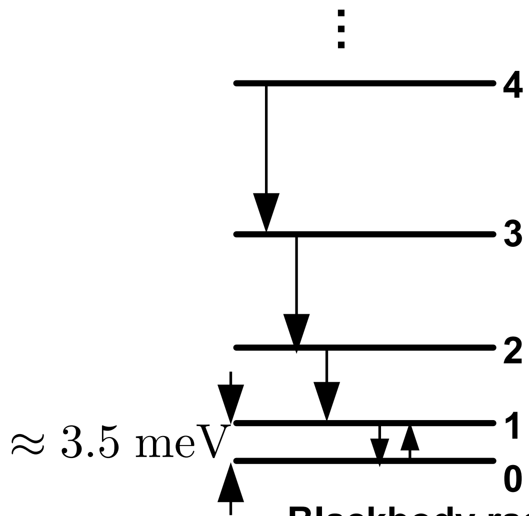
Rotational energy levels



$E_0 \approx 3.5$ meV

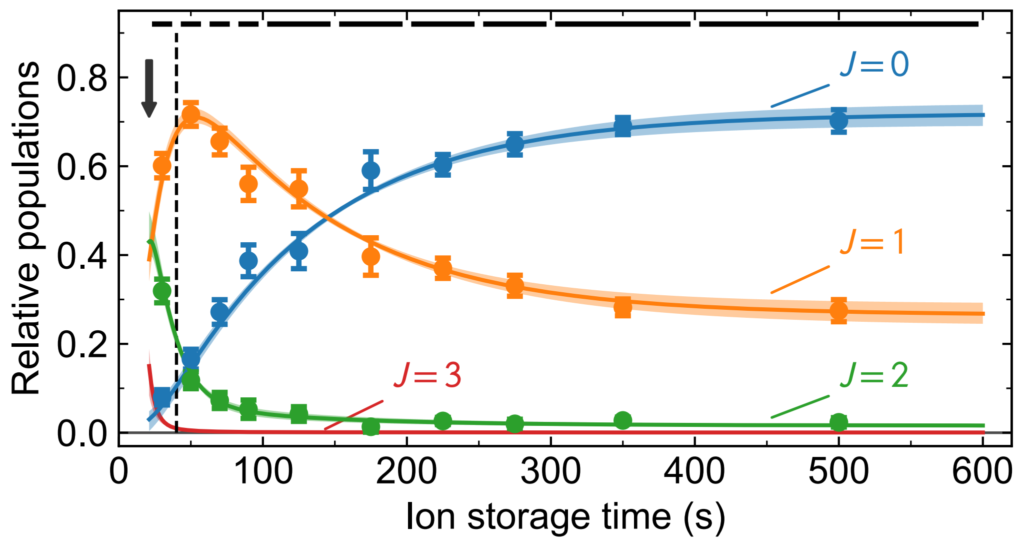
(case of CH$^+$)

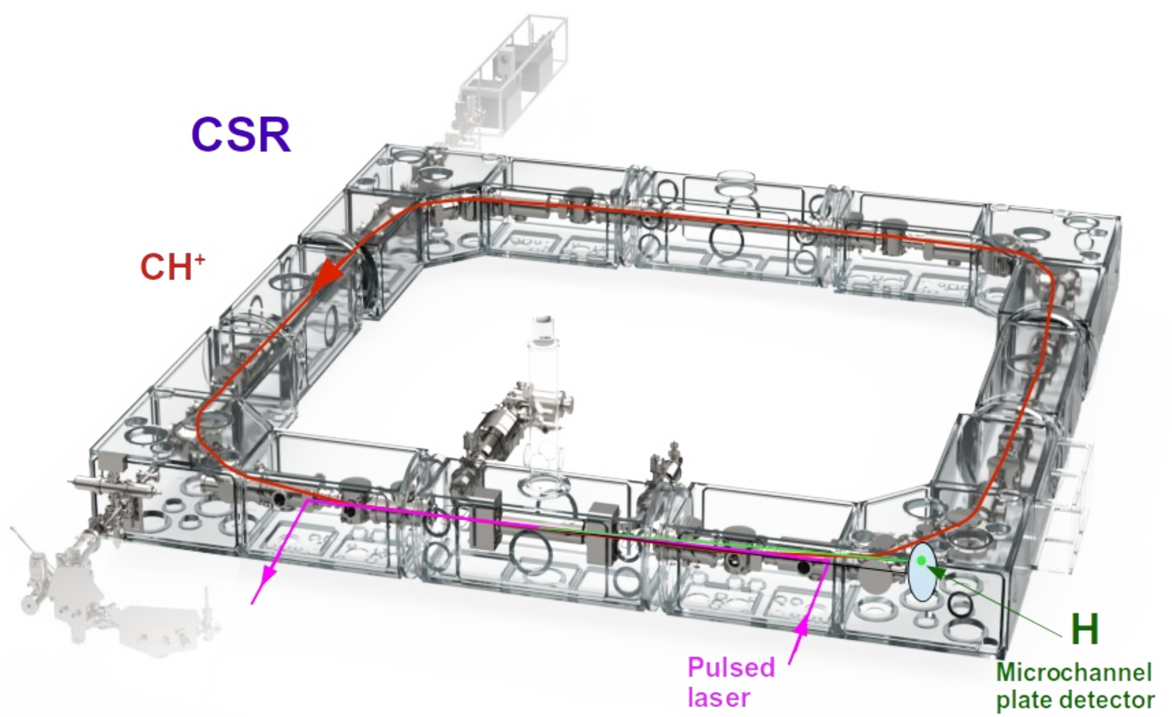
Rotational electron cooling measurement



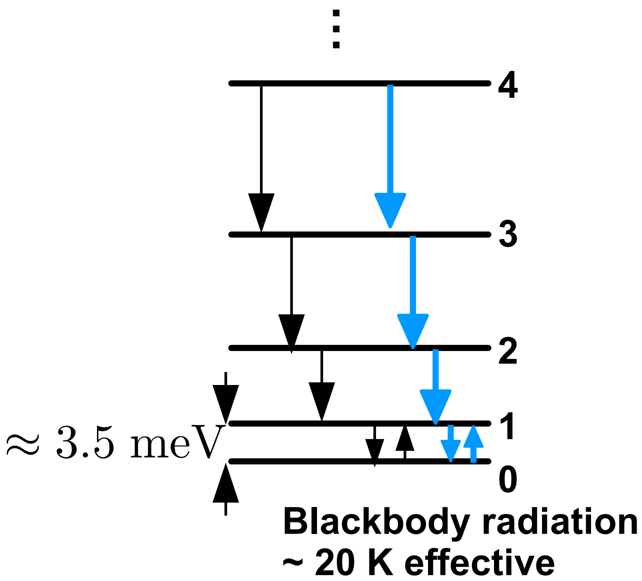
Blackbody radiation
~ 20 K effective

Probing results for radiative cooling only





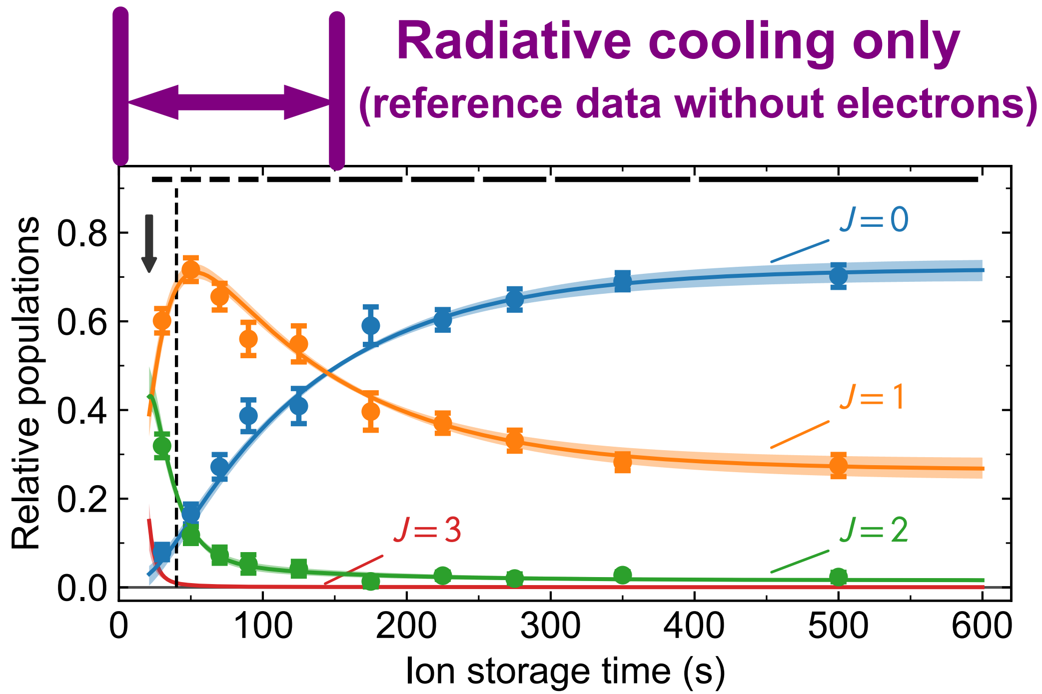
Rotational electron cooling measurement

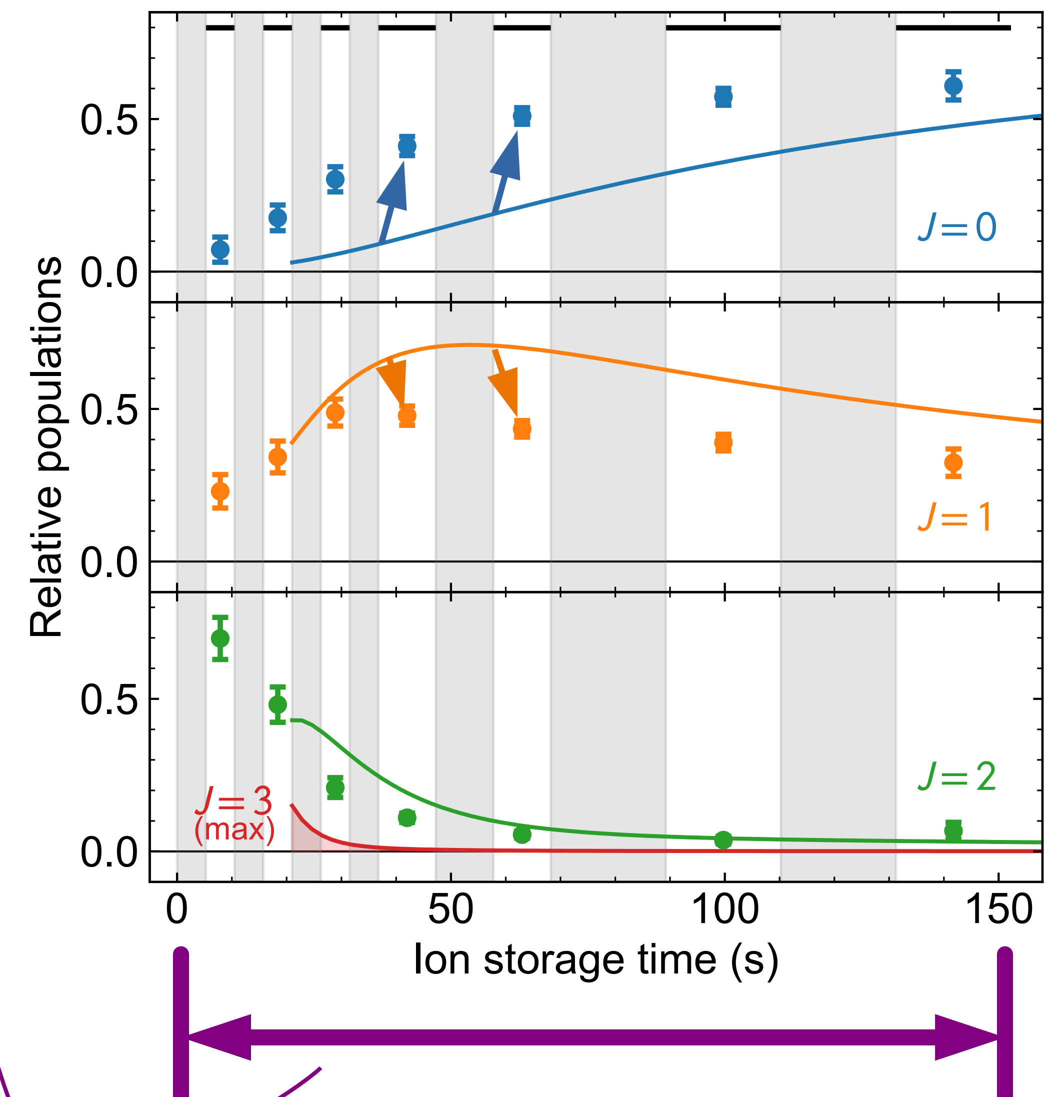


Electrons at E_{rel} = 0
(~50% duty cycle)

Electron-induced rotational cooling

Radiative cooling only
(reference data without electrons)



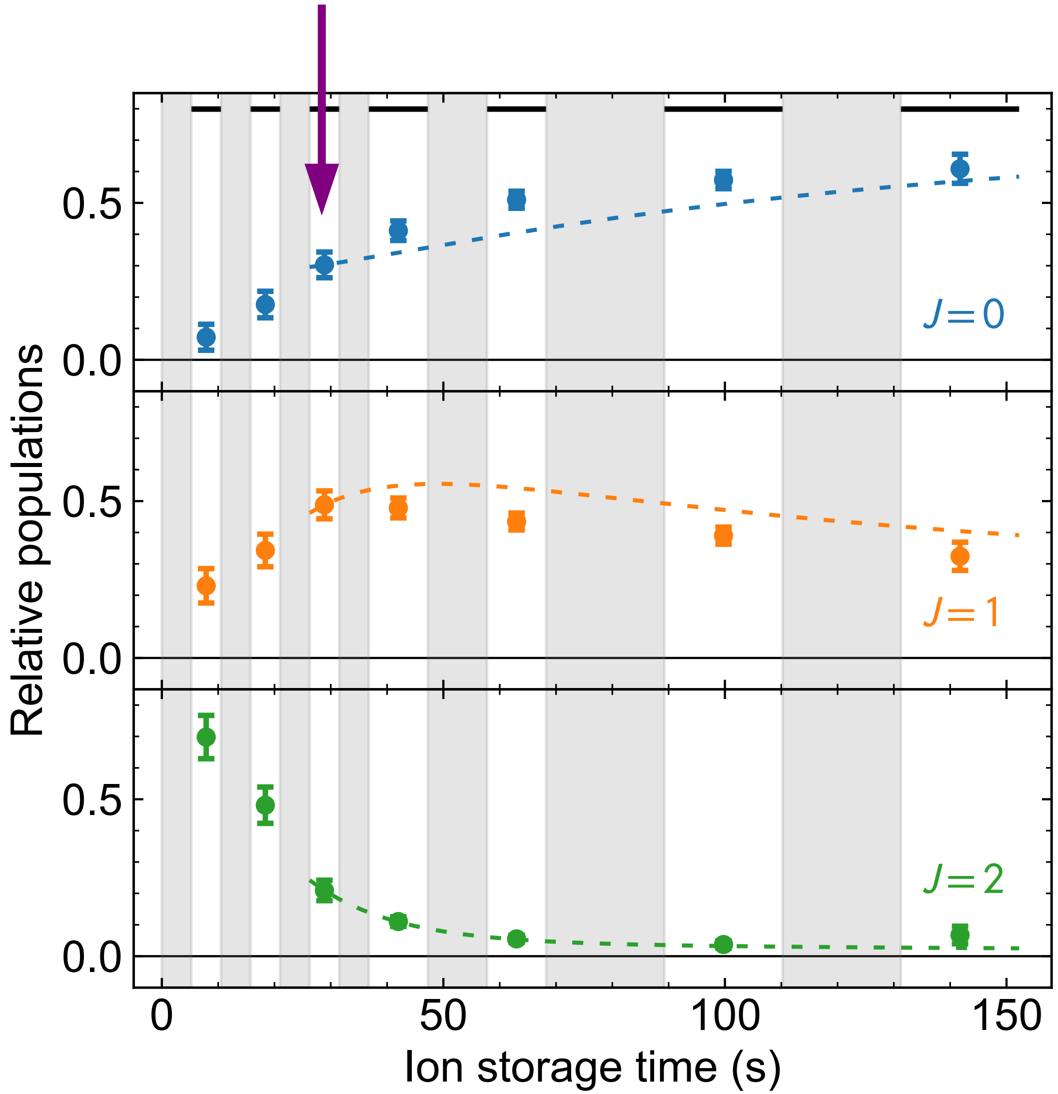


> 50% in *J* = 0: ~150 s without electrons
~ 50 s with electrons

Rotational cooling of CH^+ ions

Quantitative analysis

Modeling with theoretical collision cross sections

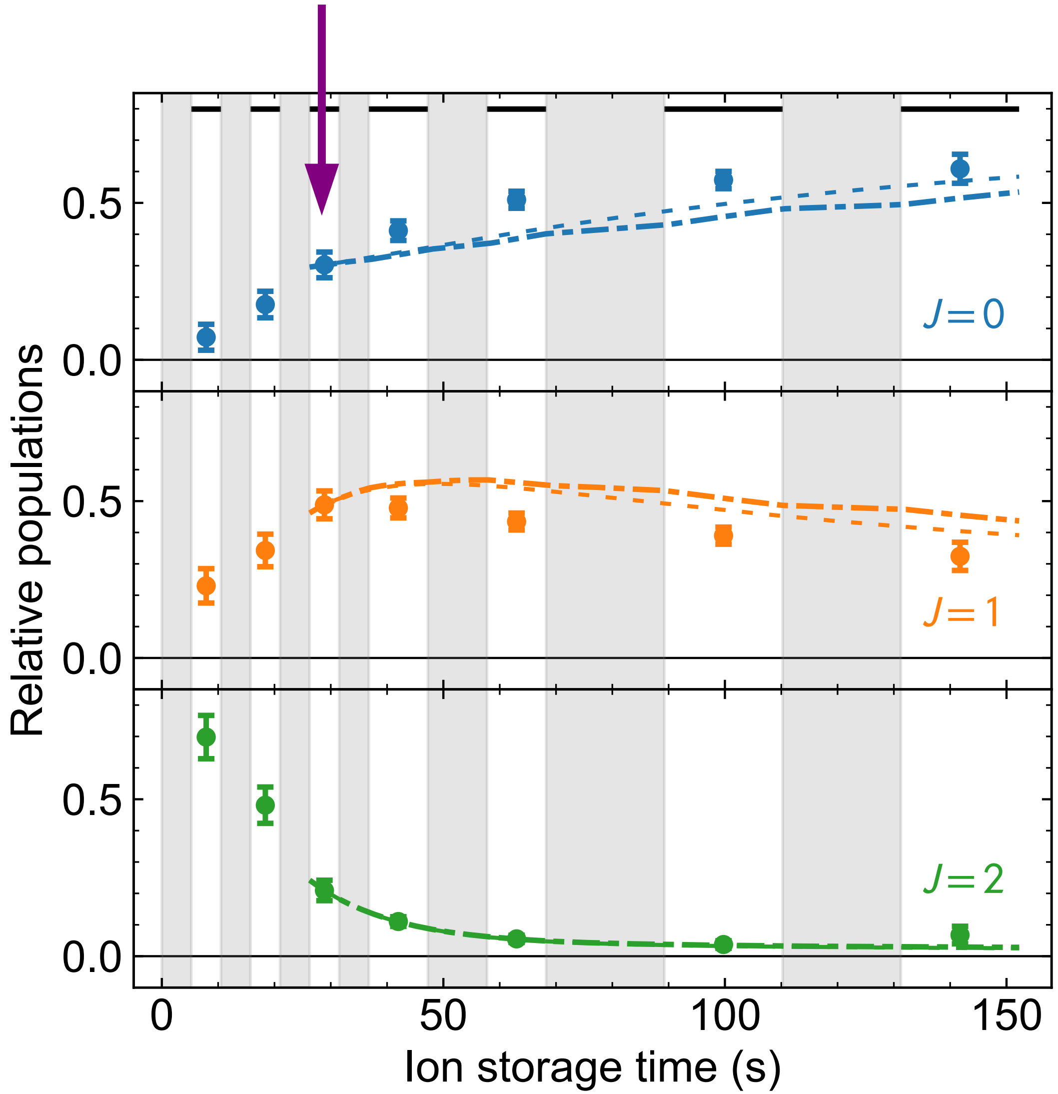


Start at third probing window

Rate equations for *J*-level populations:

Radiative cooling only

Modeling with theoretical collision cross sections



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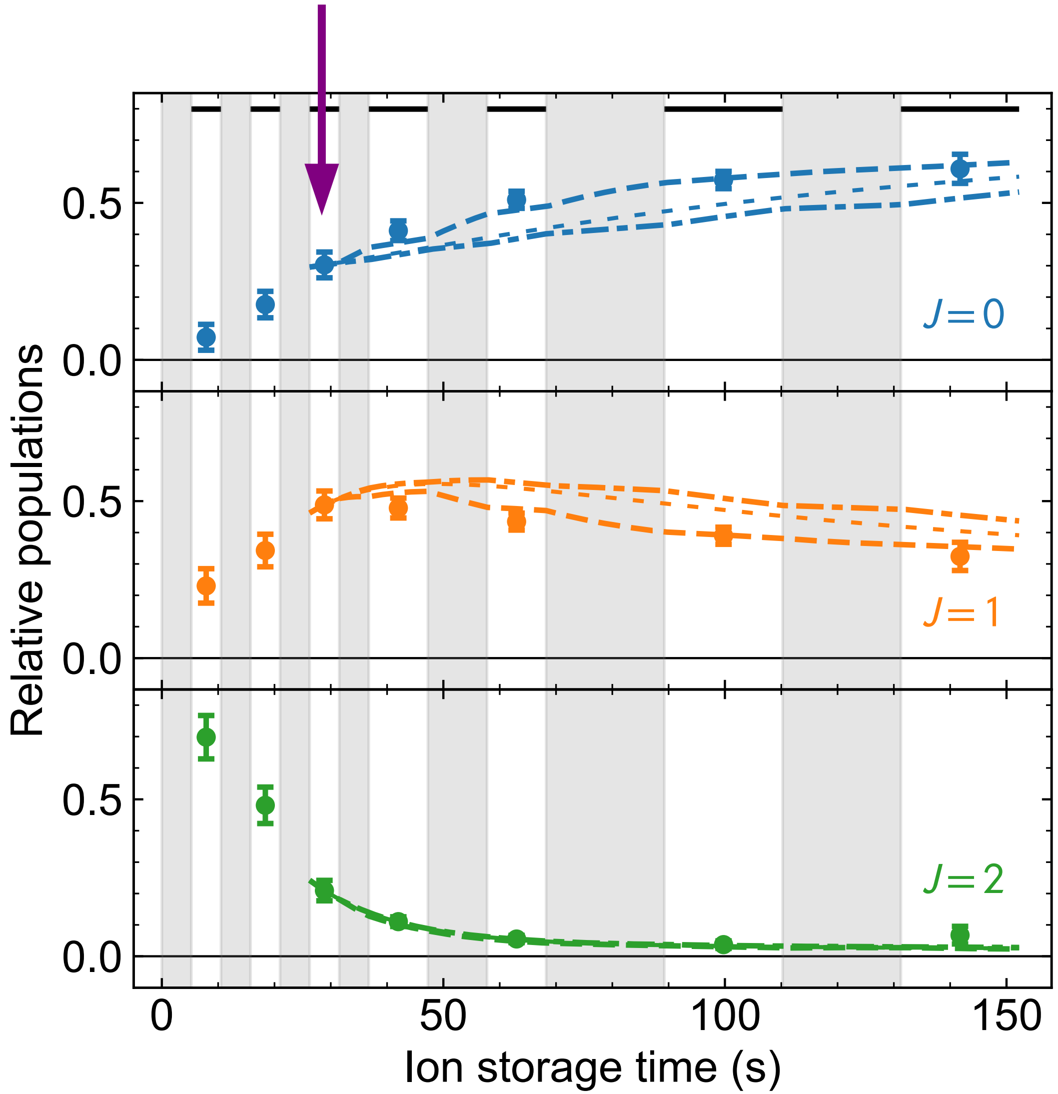
Rate equations for *J*-level populations:

Radiative cooling only

\+ *J*-dependent loss by recombination of CH^+

Modeling with theoretical collision cross sections



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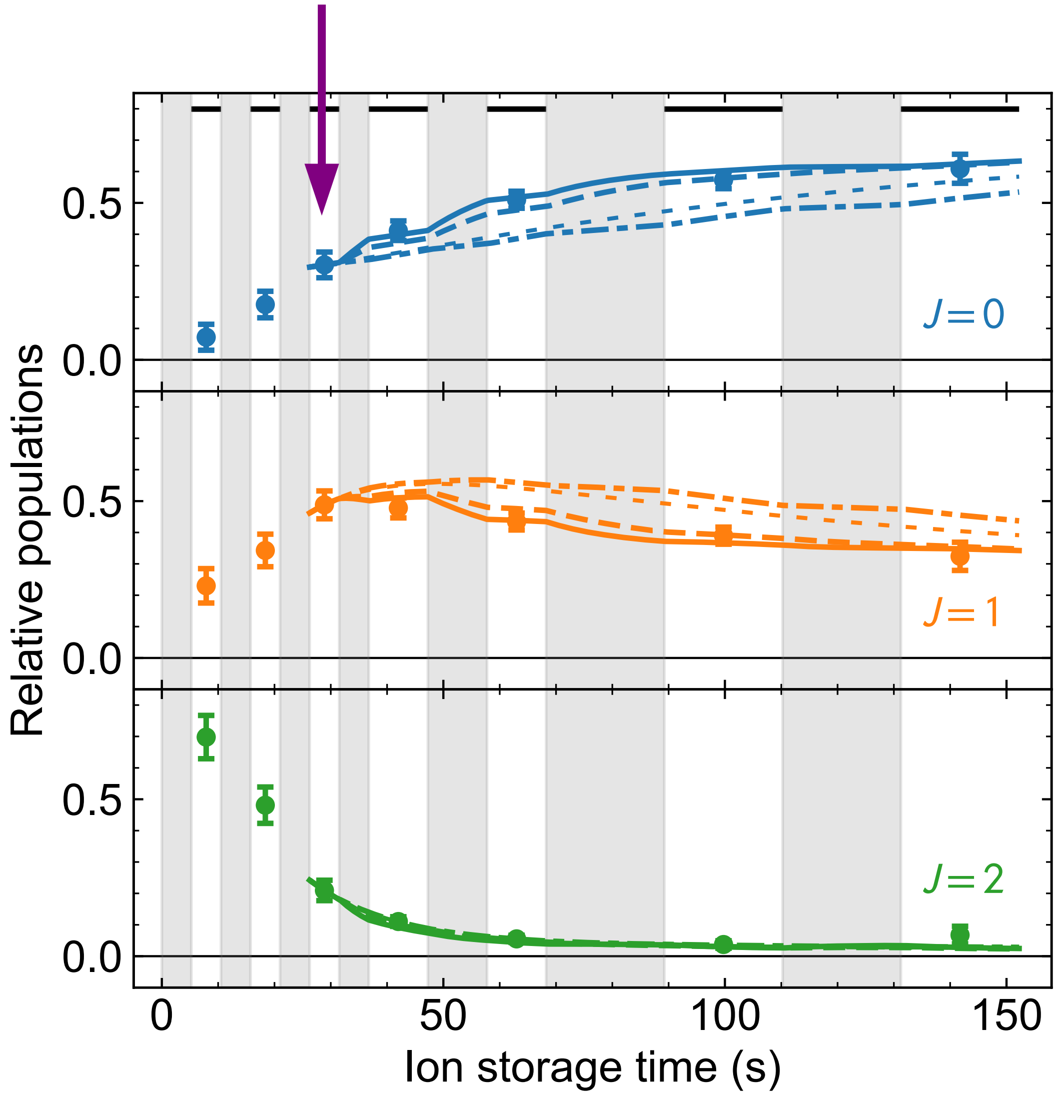
Rate equations for *J*-level populations:

Radiative cooling only

+ *J*-dependent loss by recombination of CH^+

+ Inelastic collisions in Coulomb–Born approximation

Modeling with theoretical collision cross sections



Start at third probing window

Rate equations for *J*-level populations:

Radiative cooling only

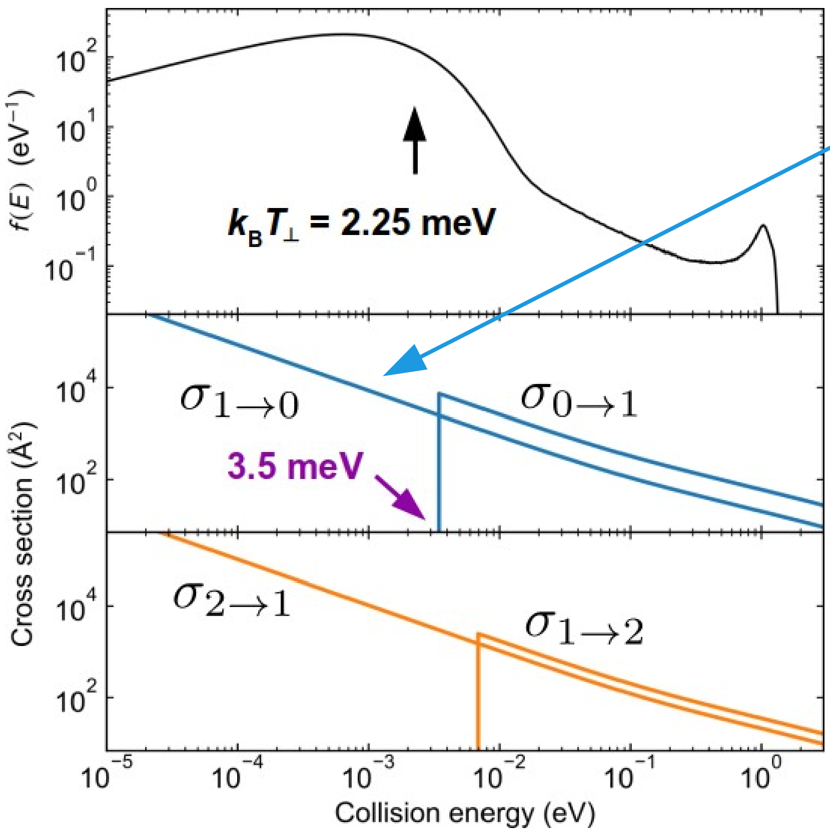
+ *J*-dependent loss by recombination of CH^+

+ Inelastic collisions in Coulomb–Born approximation

Inelastic collisions from recent *R*-matrix molecular calculations

J. R. Hamilton, A. Faure, and J. Tennyson, Mon. Not. R. Astron. Soc. 455, 3281 (2016).

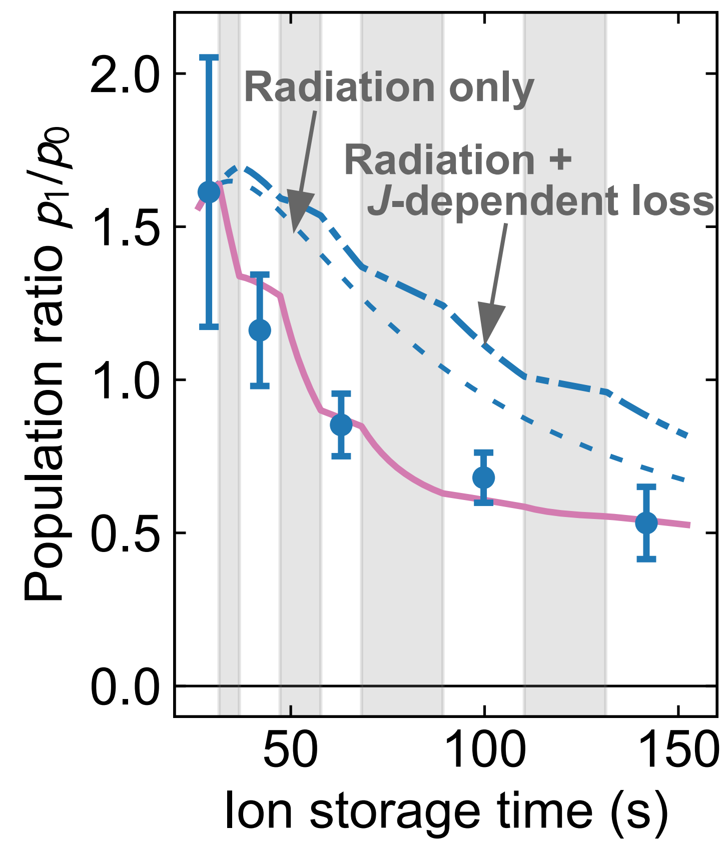
Experimental $J = 1 \leftrightarrow 0$ inelastic cross section



$$\sigma_{1\to0} = \frac{\hat{\sigma}}{E}$$

$\sigma_{0\to1}$ follows from the principle of detailed balance

Fitting the measured population ratio p_1/p_0



$\hat{\sigma}$ = 1.23 × 10^{-15} cm^2 eV

Uncertainties in 10^{-15} cm^2 eV:

fit statistics	±0.26
systematics	±0.46

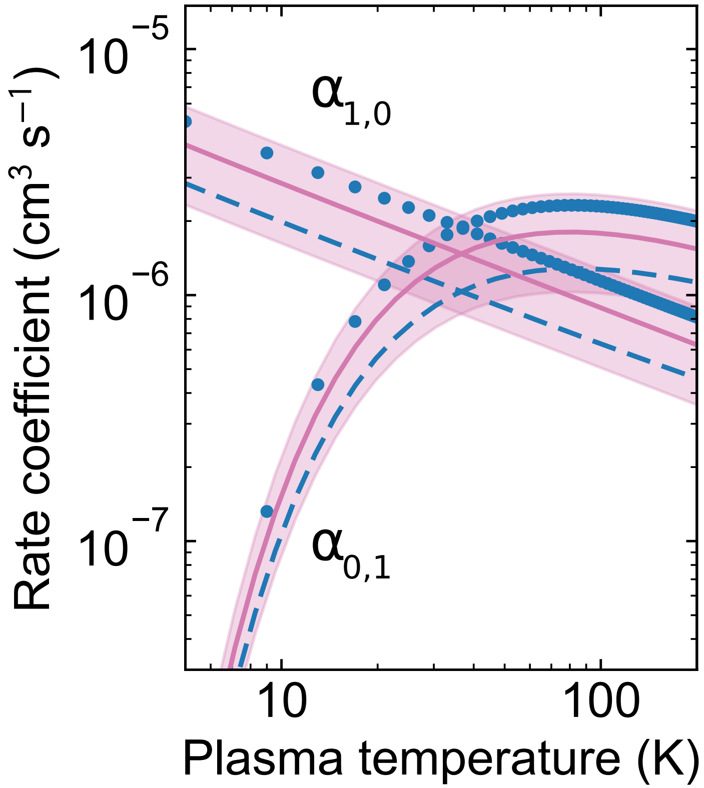
(~43% relative uncertainty)

Rate coefficients in cold plasma for *J*-changing electron collisions
(astrophysics etc.)

$$\alpha_{1,0}(T) = \langle\ v\sigma_{1\to 0}(v)\ \rangle_T$$

$$\alpha_{0,1}(T) = \langle\ v\sigma_{0\to 1}(v)\ \rangle_T$$

Thermal rate coefficients with the fitted 0↔1 cross sections



Hamilton et al. (2016)

***R*-matrix theory**

Rotational cooling measurement at CSR

Coulomb–Born

(~43% relative uncertainty)

Conclusions

Last week (end Oct 2021):
cooling force measurements
for a 4.5 eV electron beam

Cryogenic photocathode electron cooler in CSR
with electron energies $\lesssim$ 10 eV

Electron cooling of molecular rotations
substantially speeds up rotational relaxation in a stored ion beam

Rotational electron cooling rates measured by laser probing
on diatomic hydride CH^+

Damping of oscillating dipoles in electron collisions:
theory quantitatively confirmed

Outlook

Stronger electron cooling expected for smaller rotational constants (heavier molecules)

Electron cooling expected to remain significant for molecules without dipole moment

In development: general, sensitive laser probing of rotational populations
based on spectrally resolved, multistep photodissociation

Authors of submitted paper on [Rotational electron cooling measurement at CSR](#)

Á. Kálosi, M. Grieser, R. v. Hahn, U. Hechtfischer, C. Krantz, H. Kreckel, D. Müll, D. Paul, D. W. Savin, P. Wilhelm, A. Wolf, O. Novotný

CSR electron collisions beamtime team

Stored and Cooled Ions Division (K. Blaum)

MAX-PLANCK-INSTITUT FÜR KERNPHYSIK

Oldřich Novotný

Daniel Paul

Columbia Univ.
New York

Ábel Kálosi

COLUMBIA UNIVERSITY IN THE CITY OF NEW YORK

Manfred Grieser

Damian Müll

Leonard Isberner

Justus-Liebig University Gießen

Lisa Gamer

Selina Geisser

Claude Krantz

GSI Darmstadt

Patrick Wilhelm

MAX-PLANCK-GESELLSCHAFT

Merged electron beam experiments at the CSR

Collaborations

Holger Kreckel

MAX-PLANCK-INSTITUT FÜR KERNPHYSIK

Daniel Savin

COLUMBIA UNIVERSITY IN THE CITY OF NEW YORK

Daniel Zajfman
Oded Heber

Weizmann Institute of Science

Stefan Schippers

JUSTUS-LIEBIG-UNIVERSITÄT GIESSEN

Xavier Urbain

UCLouvain

Andreas Fleischmann
Christian Enns

UNIVERSITÄT HEIDELBERG ZUKUNFT SEIT 1386

Kirchhoff Institute of Physics

Alexander Terekhov

Rzhanov Institute of Semiconductor Physics
Acad. Sci. Novosibirsk

MAX-PLANCK-GESELLSCHAFT