Storage Ring Experiments on Electron – Molecular Ion Interactions

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Molecular processes in interstellar environment: Challenges for laboratory studies

Storage-ring studies of dissociative recombination

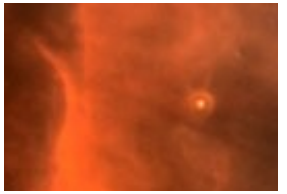
Recent results with improved experimental tools

Developments toward cryogenic ion storage ring experiments

Molecules in space

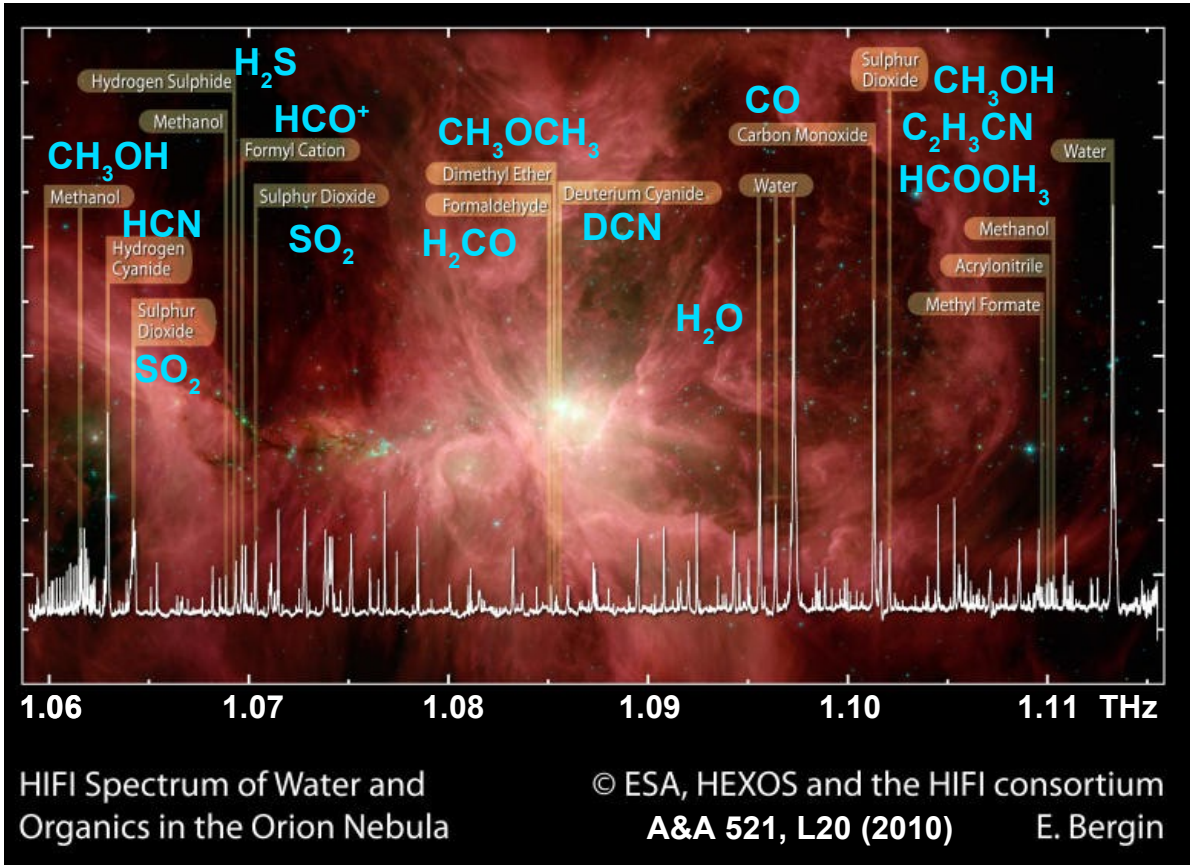
Cooling of gas clouds

Interstellar medium
Molecular clouds near galactic centres
Star-forming regions
Protoplanetary discs



Orion nebula
Hubble

Chemical complexity



Isotope (deuterium) fractionation

Spectroscopic diagnostic (temperature, radiation, dynamics)

Interstellar environment – challenges for the laboratory

Low density and temperature

Down to 1 cm^{-1}, 10 K

High reaction rates
Barrier-less reactions

Ions and radicals as drivers of chemistry

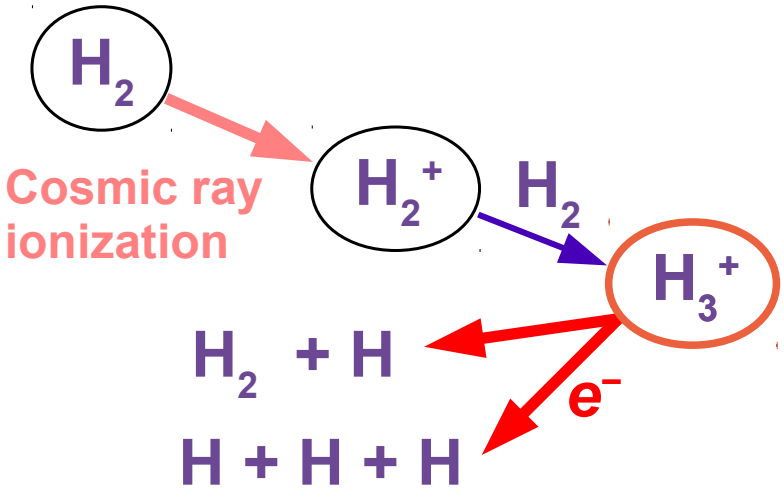
Unstable species
Special production mechanisms

Wide range of molecular species

Choice of representative model systems
Benchmarking of theories with high predictive power

Knowledge of quantum levels, innermolecular interactions,
needed for many individual cases

Chemical evolution driven by ionic collisions



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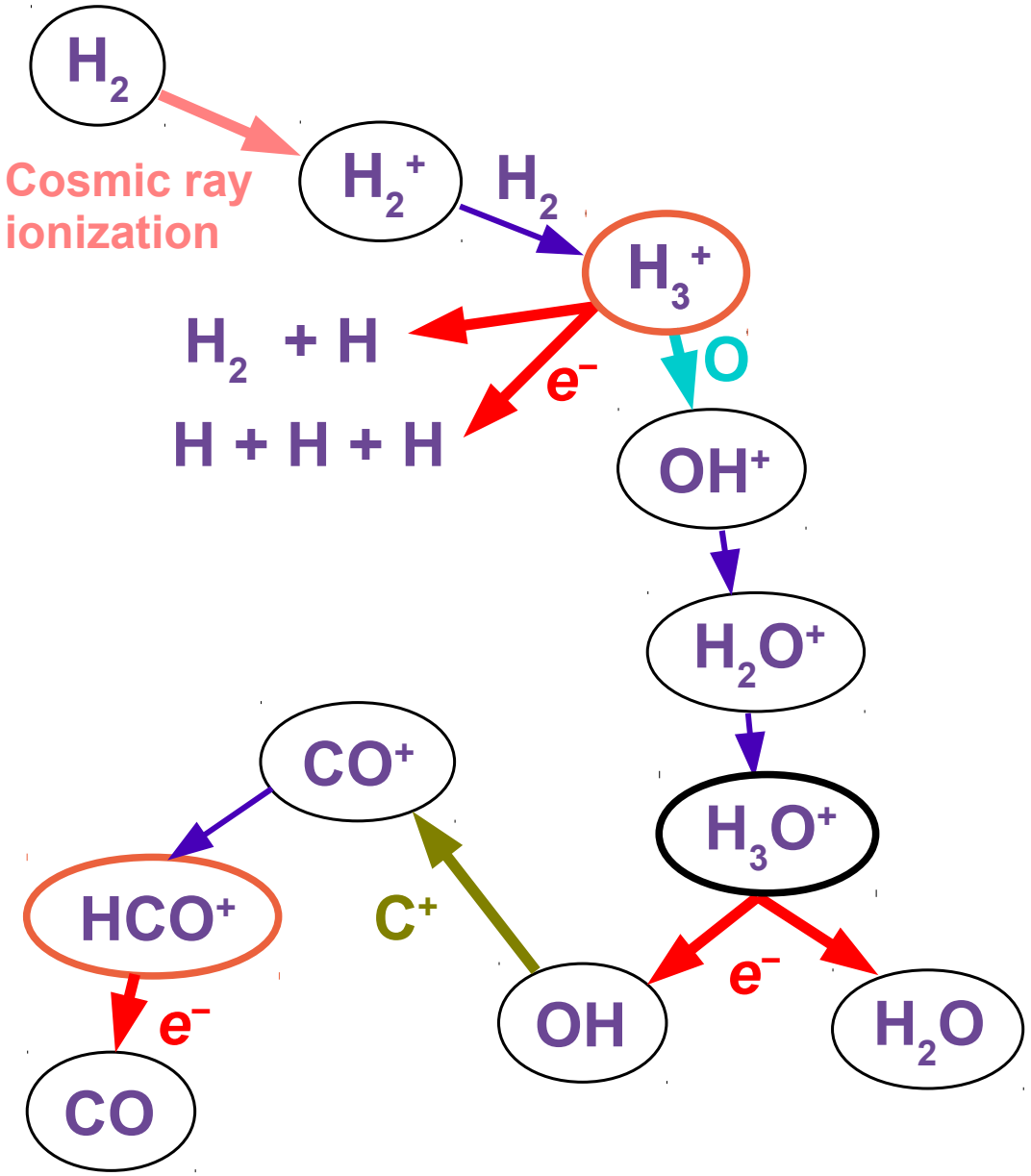

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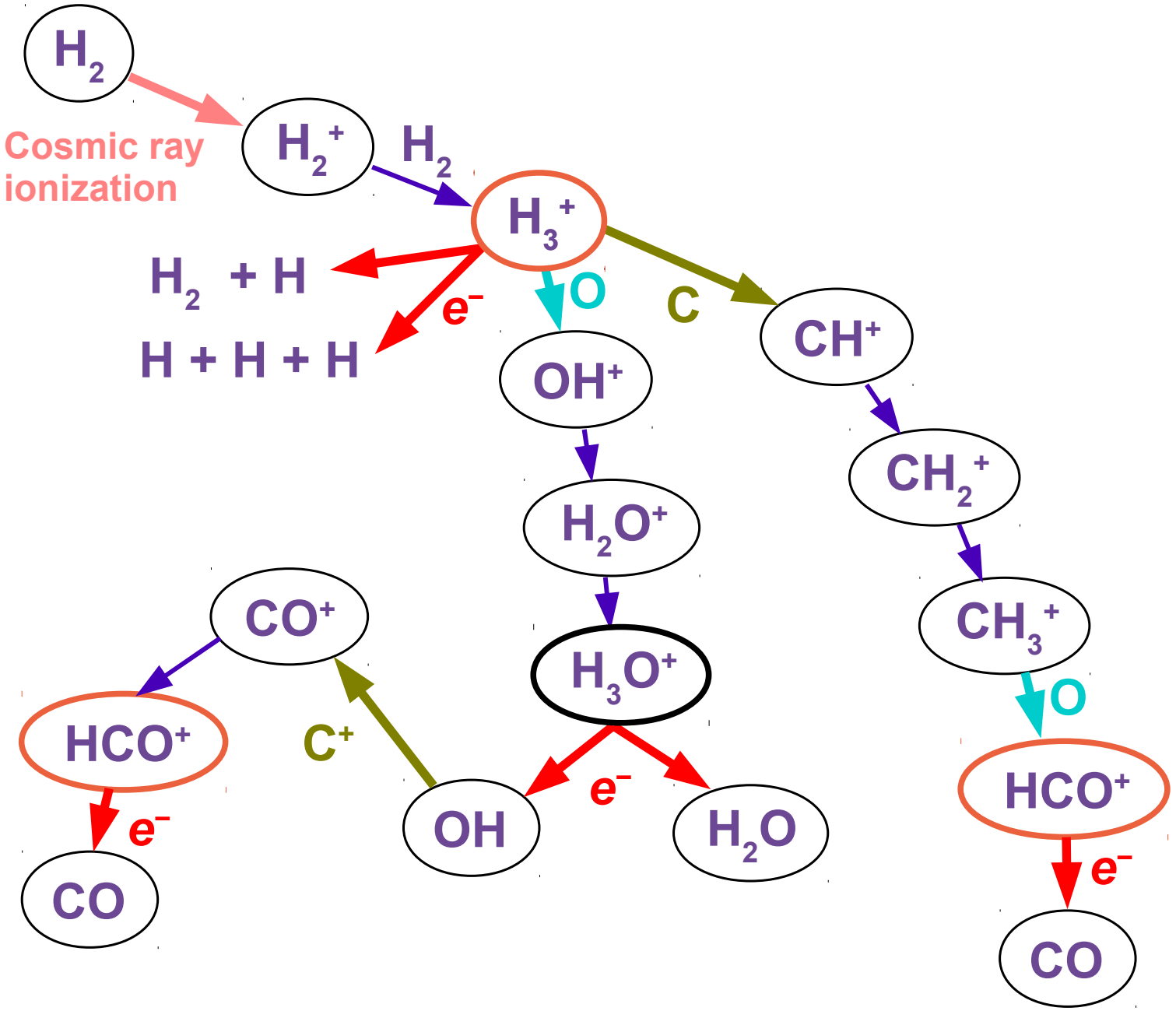

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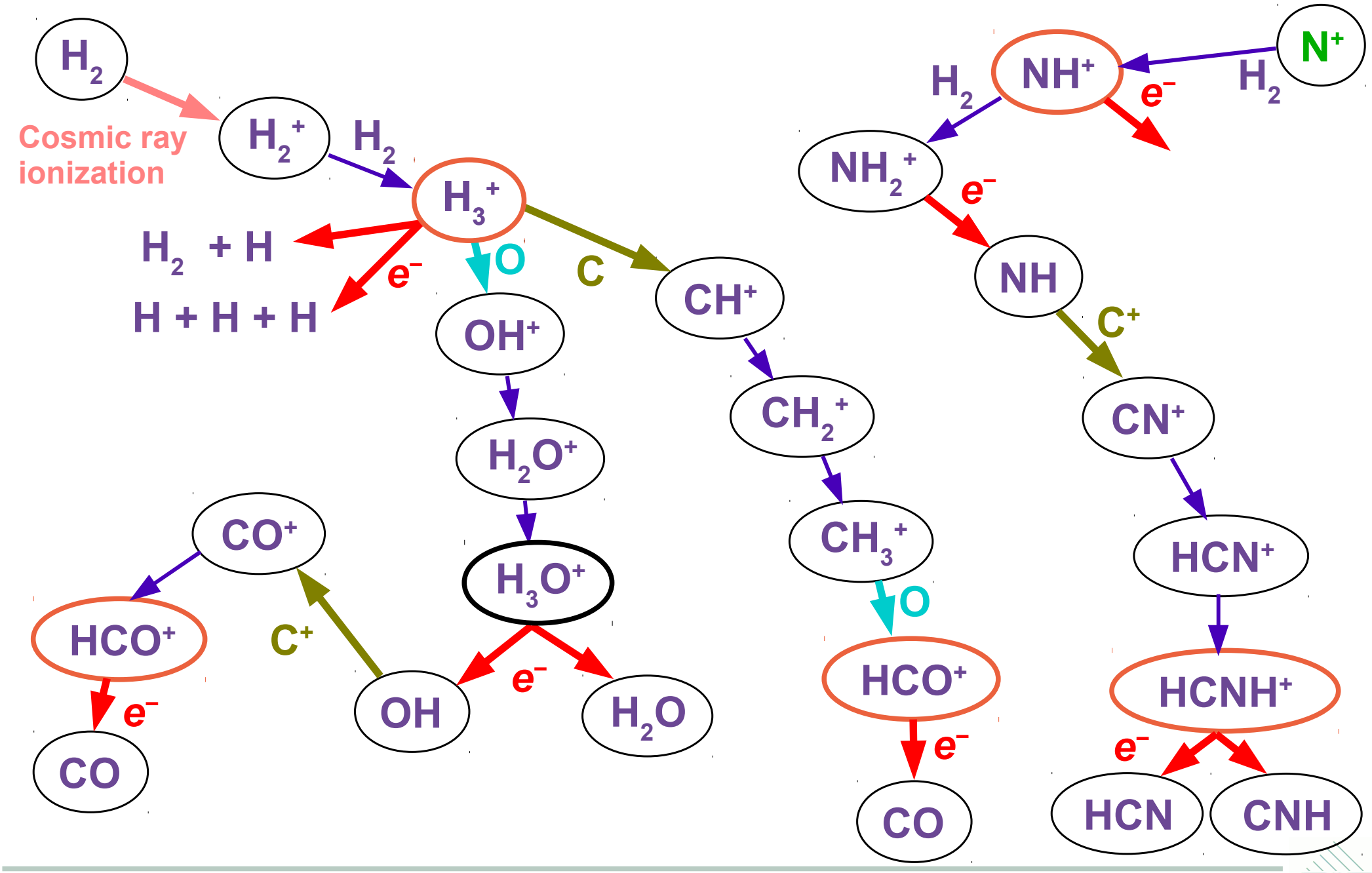

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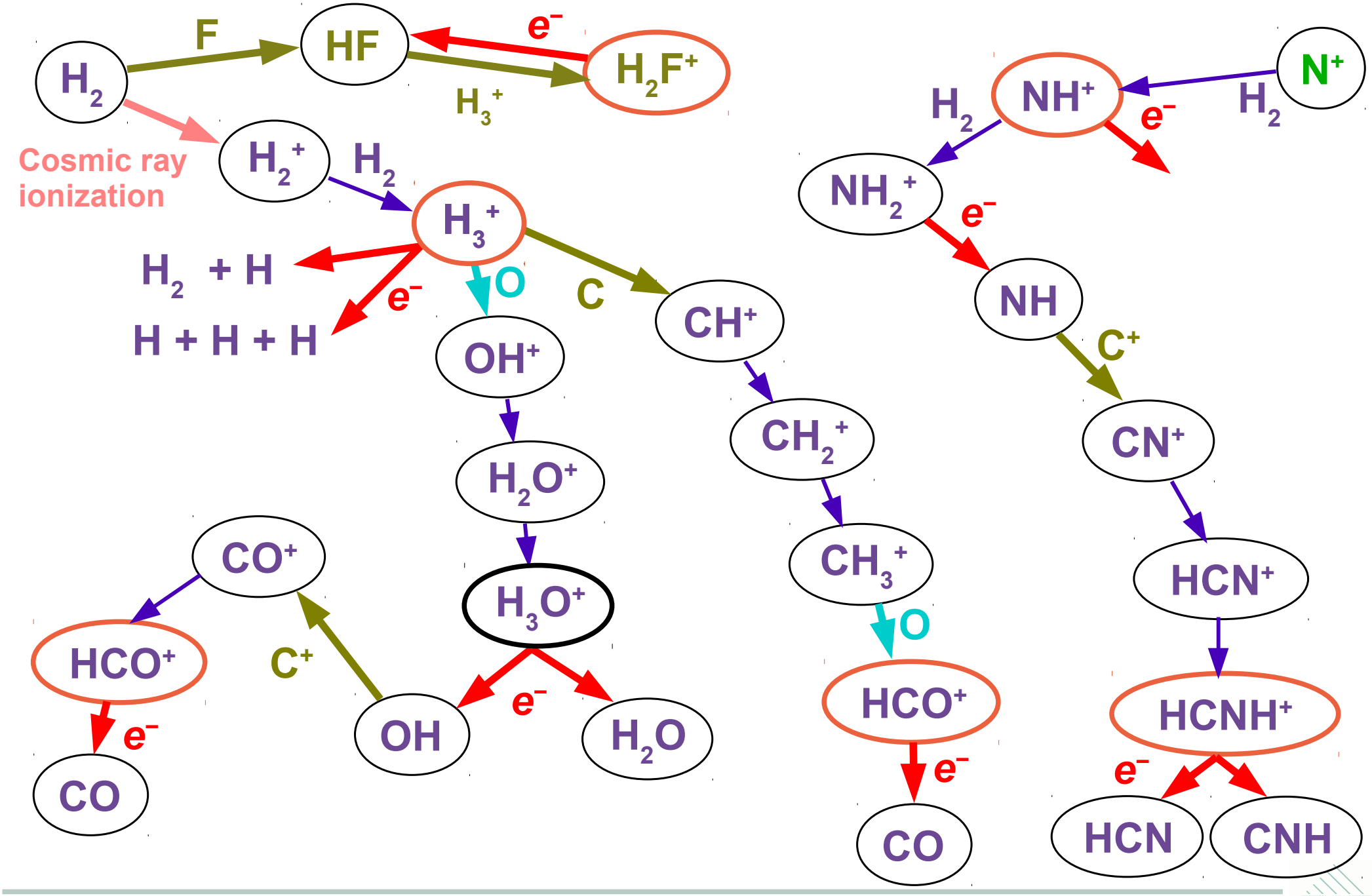
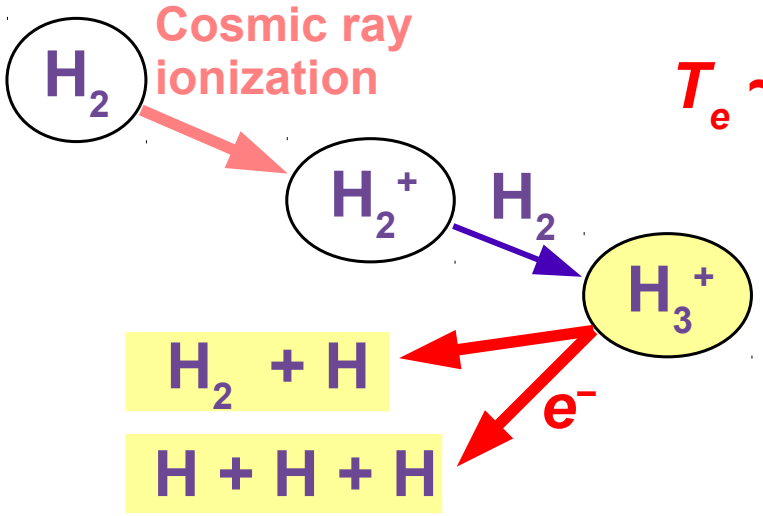


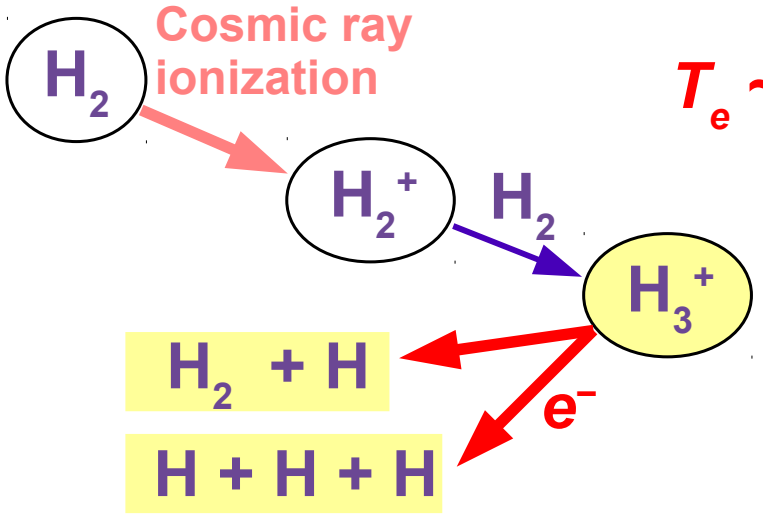
Dissociative recombination: flea and elephant



$T_e \sim 10$ K

$$H_3^+ + e^- \rightarrow H_3^{**} \rightarrow H_2 + H$$
$$\rightarrow H + H + H$$

Dissociative recombination: flea and elephant



$T_e \sim 10$ K

$$H_3^+ + e^- \rightarrow H_3^{**} \rightarrow H_2 + H$$
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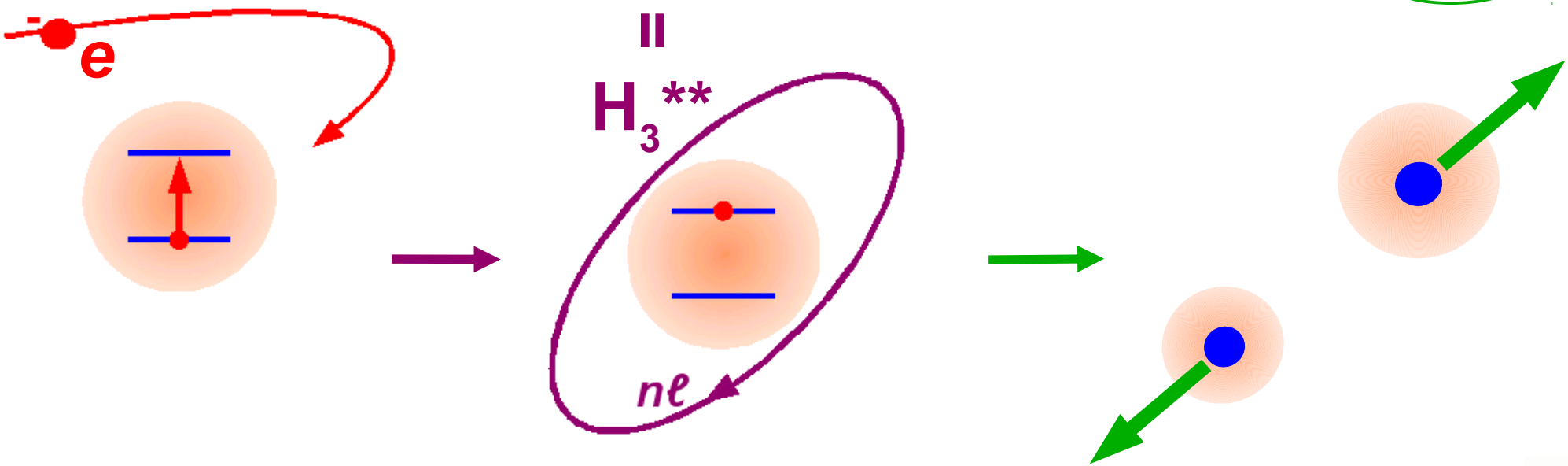
Ro-vibrationally
excited
Rydberg resonance

$H_3^+(v,J) + e$ $\quad$ $H_3^+(v',J') + n\ell$ $\quad$ $H + H_2 + E_{kin}$



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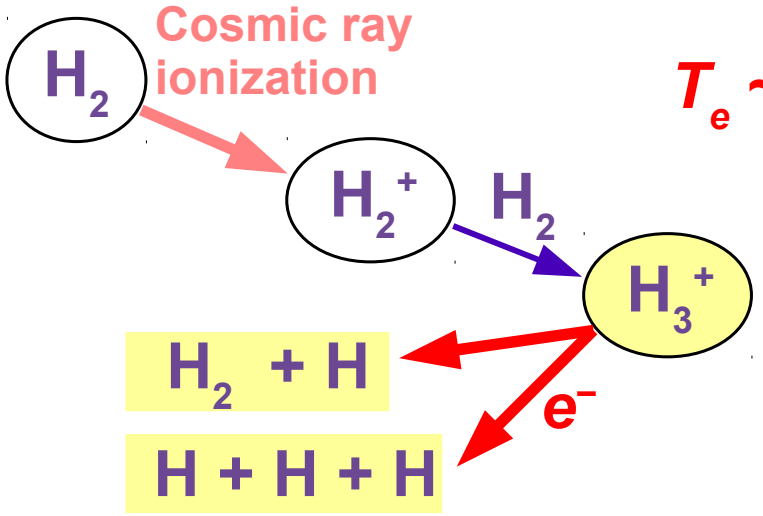

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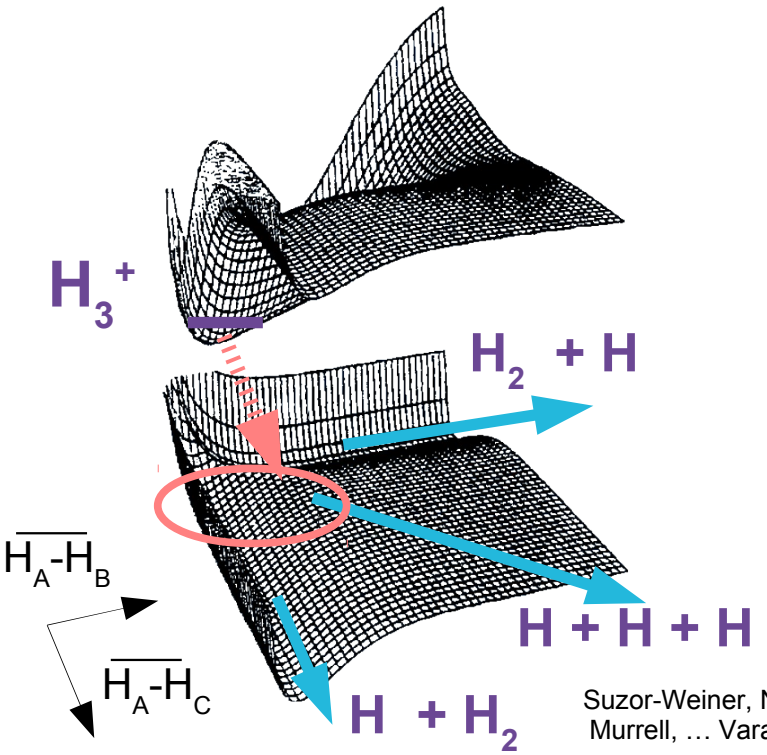
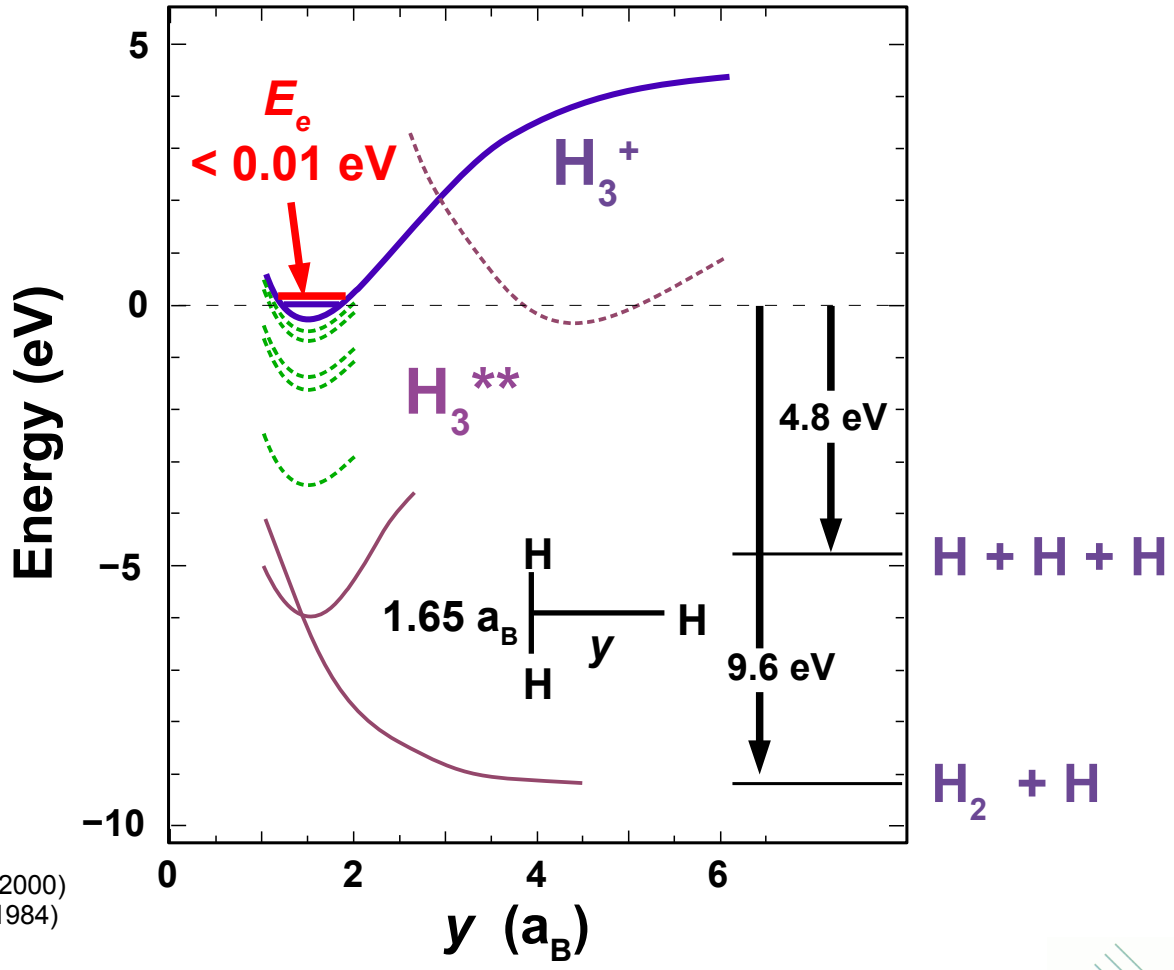
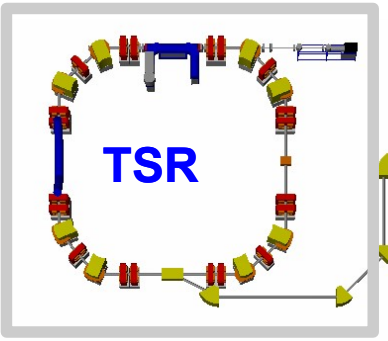


Suzor-Weiner, Nature (2000)
Murrell, … Varandas (1984)



Electron–ion merged beams
Ion storage ring TSR, Heidelberg



10 m

25 MV
cavity
linac

0.5 MV
RFQ
linac

12 MV
tandem
accelerator

Ion
sources

2.5 MeV
pelletron

Electron
cooler

$v_e = v_i$

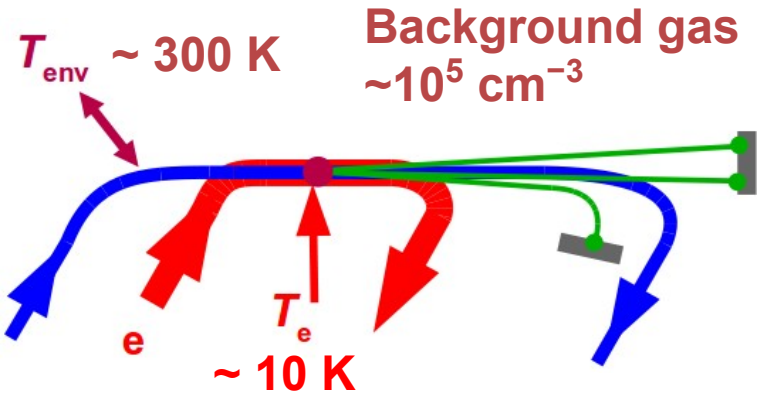
$kT_e \sim$
4 meV

Cooled
MeV ion
beam

10^{-11} mbar
300 K

Extracted ions
Coulomb explosion beamline

Electron–ion merged beams
Ion storage ring TSR, Heidelberg



~10 K interaction energy

Photocathode electron target/cooler

$v_e \neq v_i$

12 m

Neutral fragment detectors

kT_e ~ 1 meV

Electron cooler

$v_e = v_i$

kT_e ~ 4 meV

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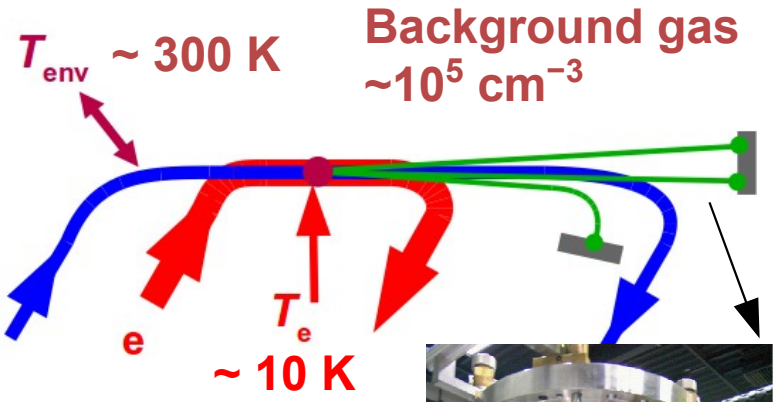

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EMU@TSR

H. Buhr et al. PRA 81, 062702 (2010)

Neutral fragment identification

Fragment kinetic energy:

$E_{kin} = ½ m_i v_{beam}^2$

→ fragment mass

Energy sensitive detector
- (Multistrip) surface-barrier detector
- Cryo calorimeter

Results from storage-ring merged beams

Product rates

Cross section (collision energy dependent)

Thermal rate coefficients

Product masses

Reaction branching ratios

Product kinetic energies

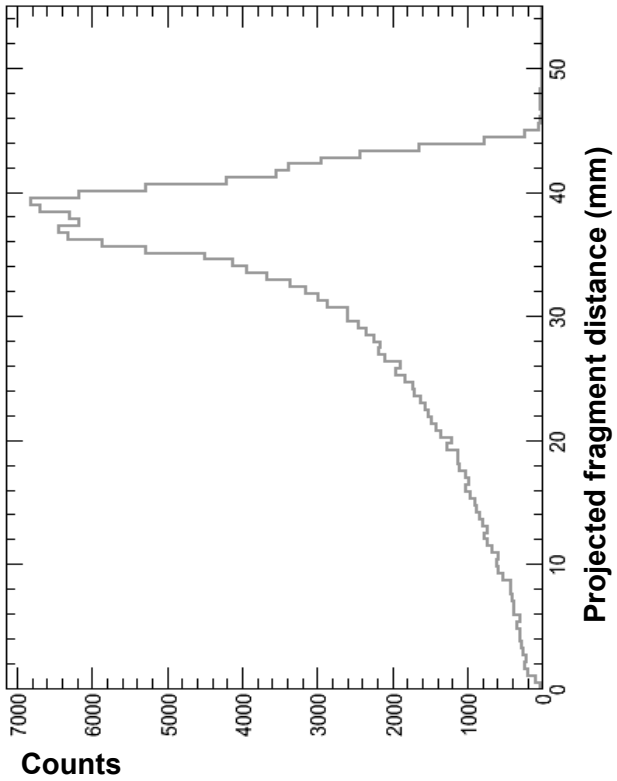
Internal excitation of molecular products

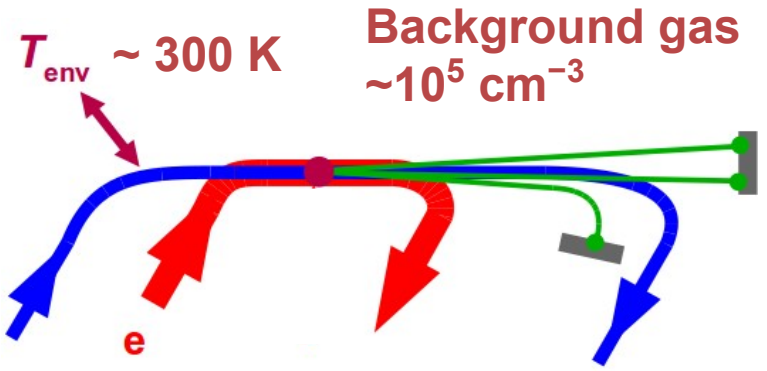
Internal excitation of incident molecular ion

Atomic final states and
measurement of dissociation energies
(diatomic reactions)

Fragment imaging and rate coefficient

$NH^+ + e \rightarrow N + H$





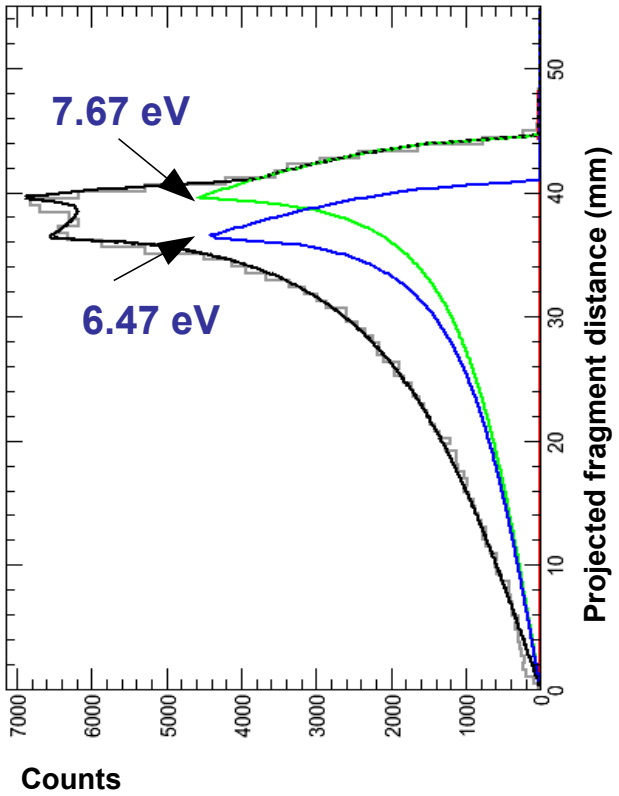
$E_{cm} = 0$

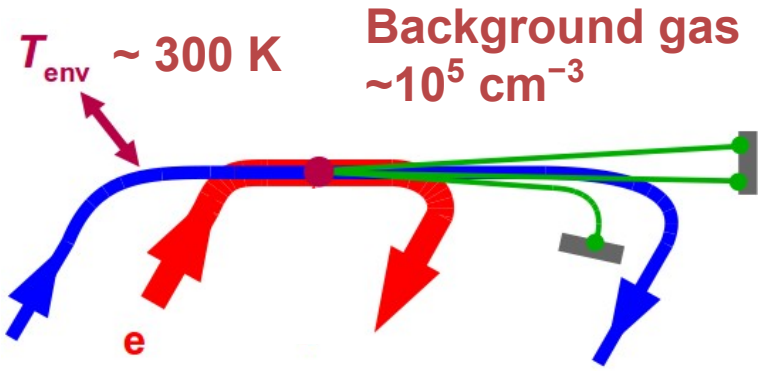
$NH^+(X^2\Pi) + e^- \rightarrow$

- $N(^4S^o) + H(^2S)$ + 10.05 eV
- $N(^2D^o) + H(^2S)$ + 7.67 eV
- $N(^2P^o) + H(^2S)$ + 6.47 eV

Fragment imaging and rate coefficient

$NH^+ + e \rightarrow N + H$





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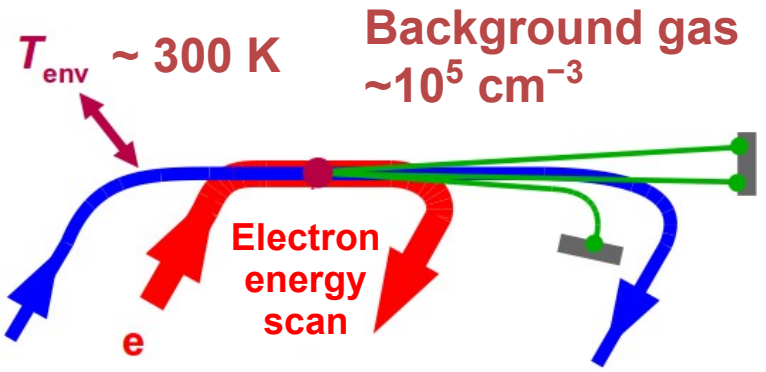

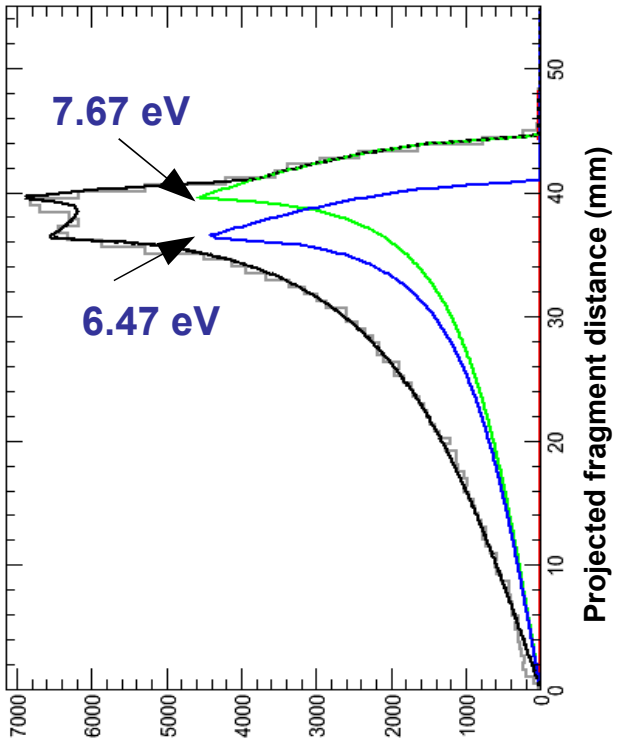

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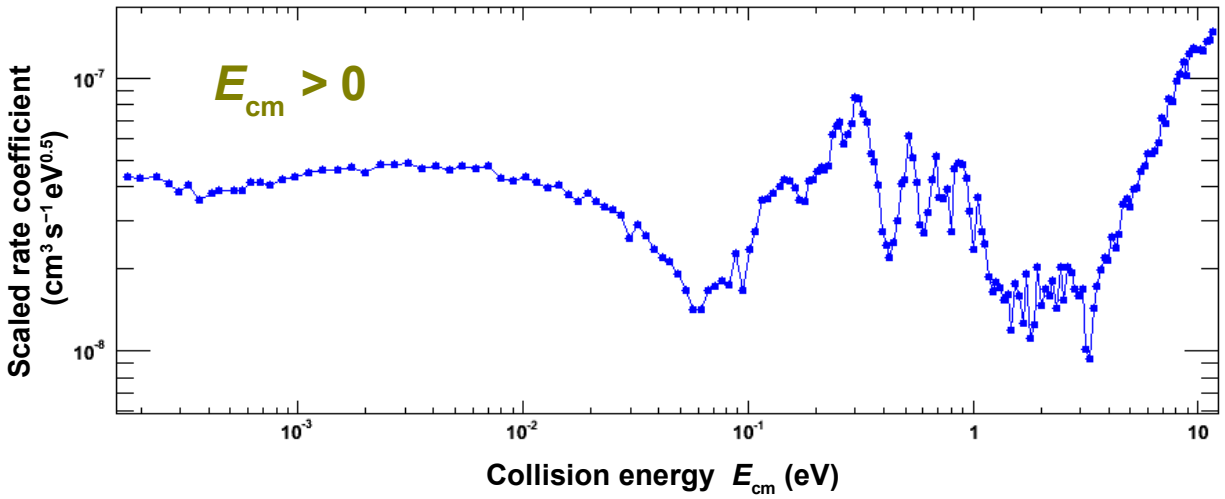
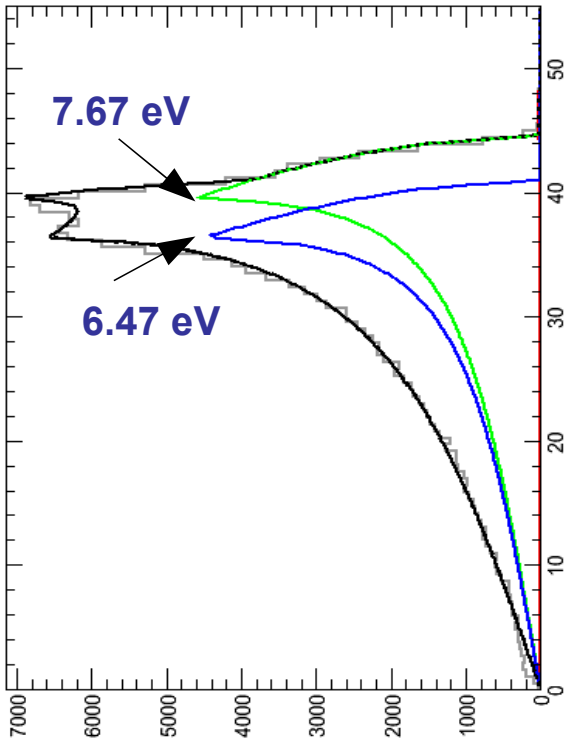


Fragment imaging and rate coefficient

$NH^+ + e \rightarrow N + H$

EMU detector:
High-count-rate fragment imaging at $E_{cm} > 0$

Molecular-ion dissociation energies probed on <0.1 eV level (sometimes to <1 meV)



Counts

$E_{cm} = 0$

$NH^+(X^2\Pi) + e^- \rightarrow$

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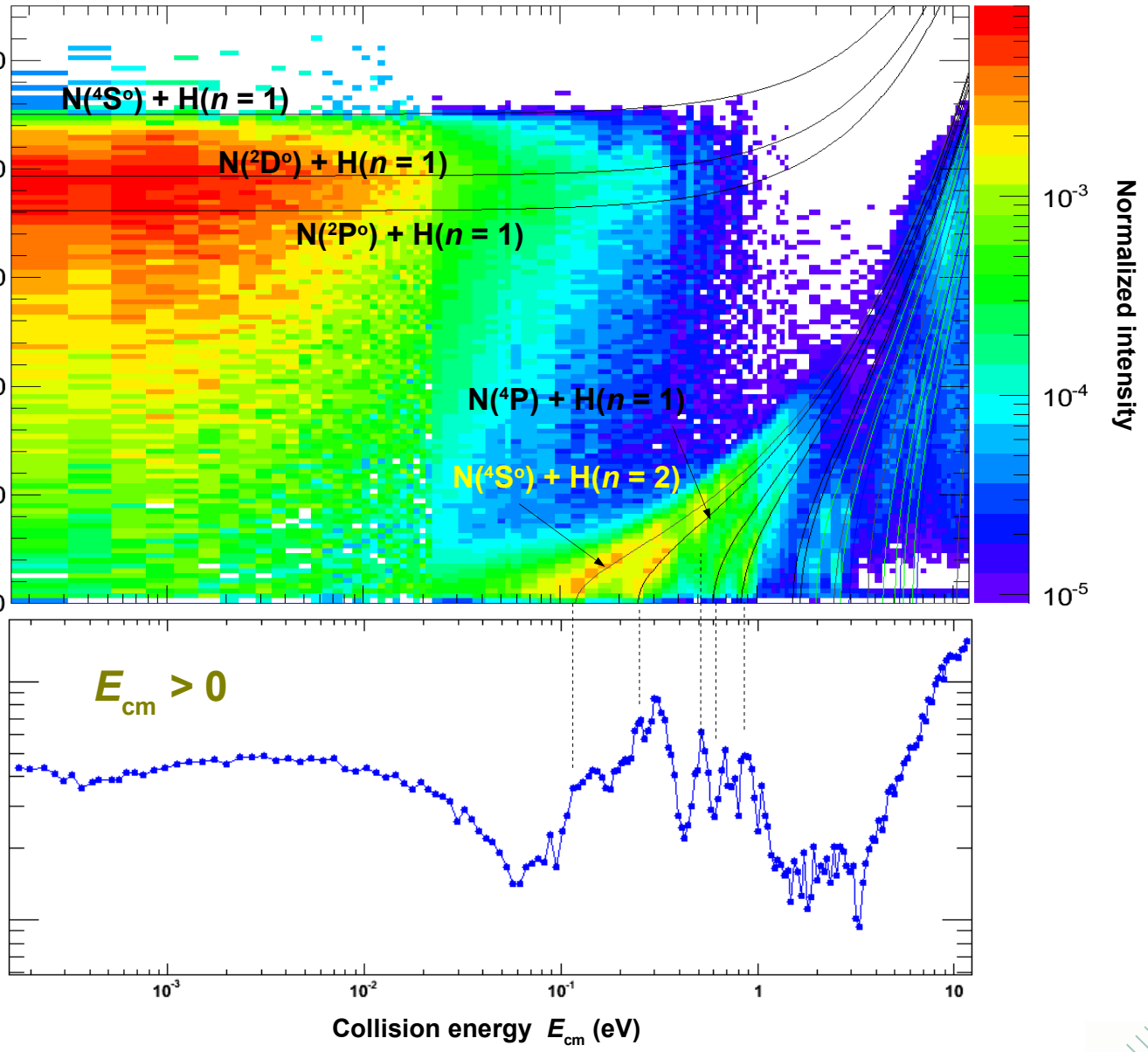
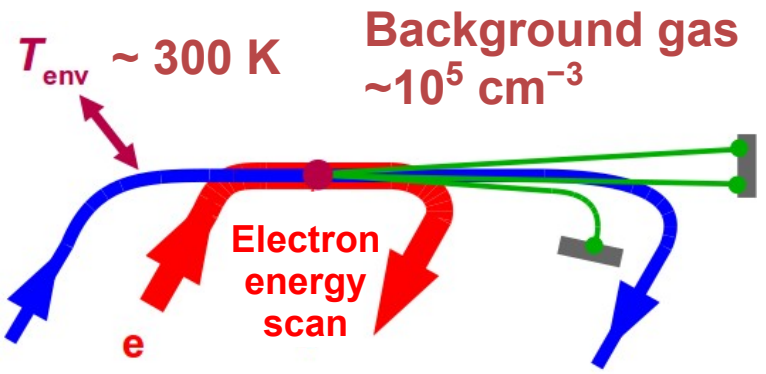
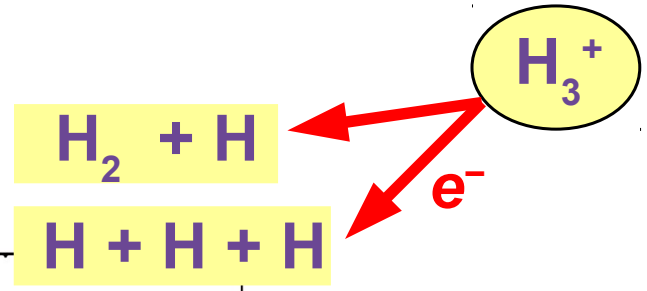
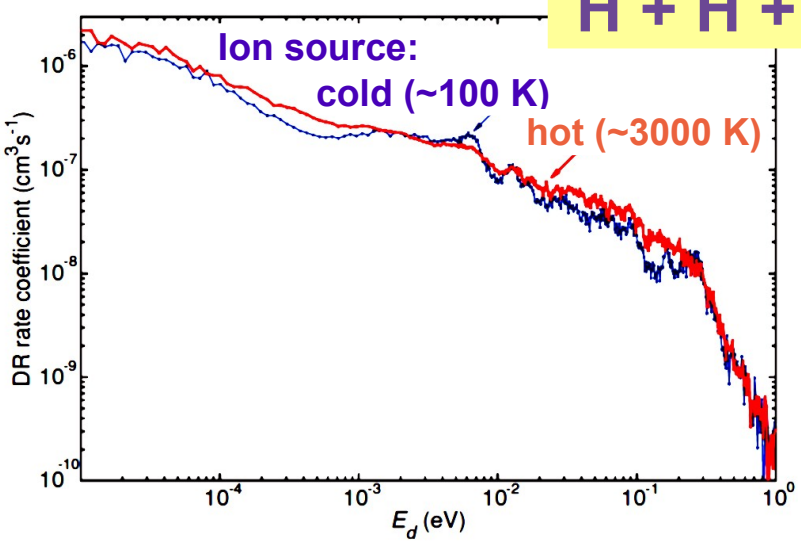


Probing the Rydberg resonances that drive recombination

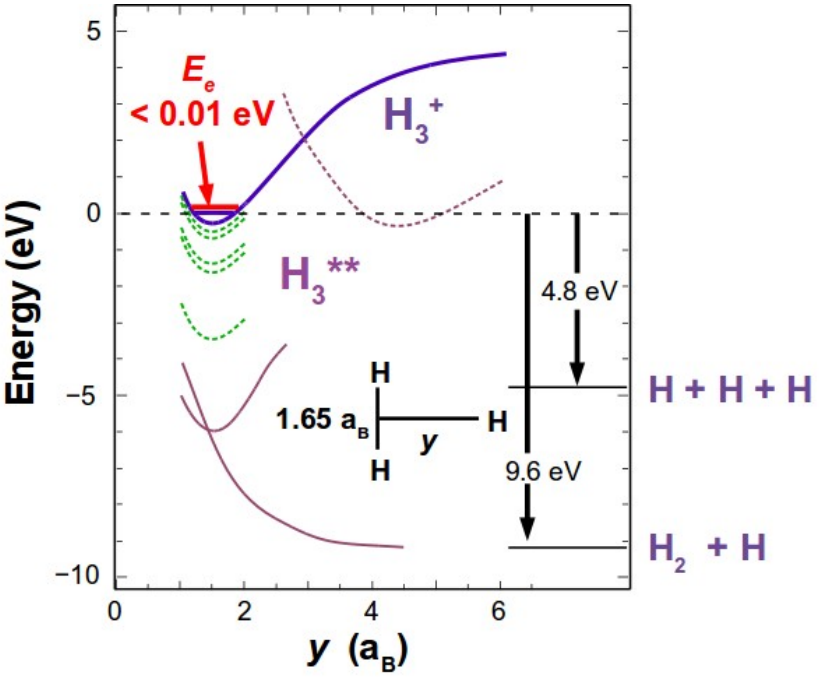


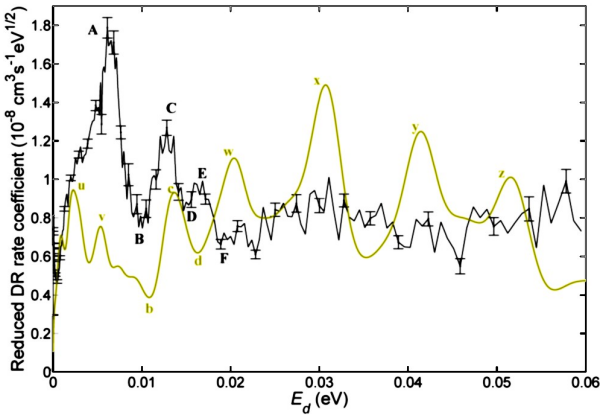
Electron capture by ro-vibrational excitation of the target ion

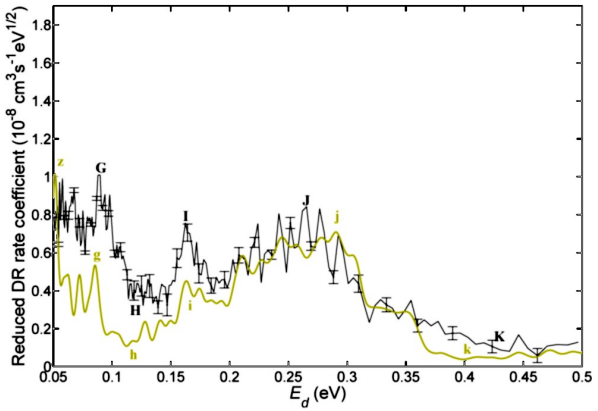




Petrignani et al.,
PRA 83, 032711 (2011)



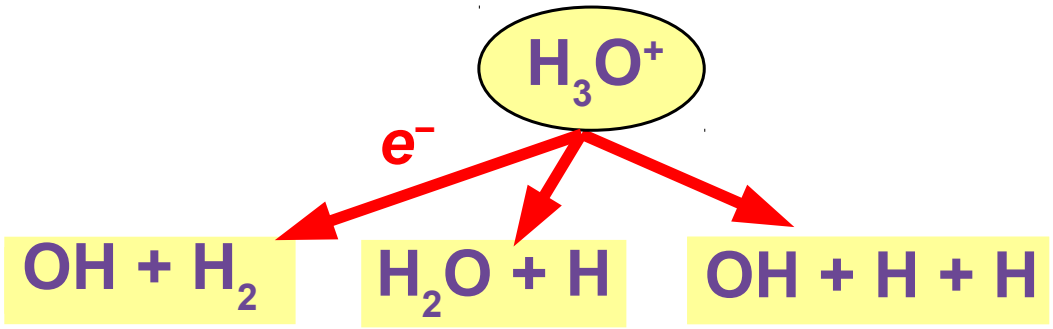


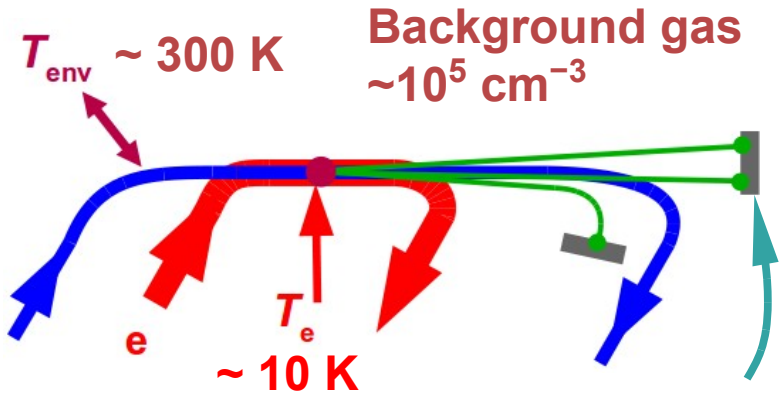


Theory of Rydberg recombination resonances

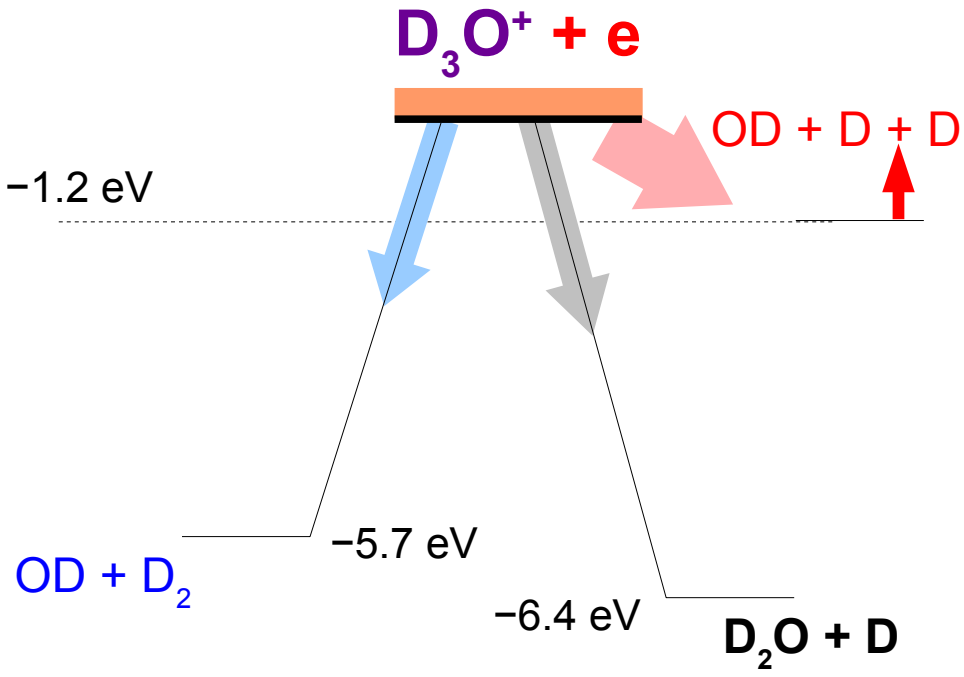
C. H. Greene, V. Kokoouline
(Boulder / Miami / Paris)

Probing the fragmentation into molecular products

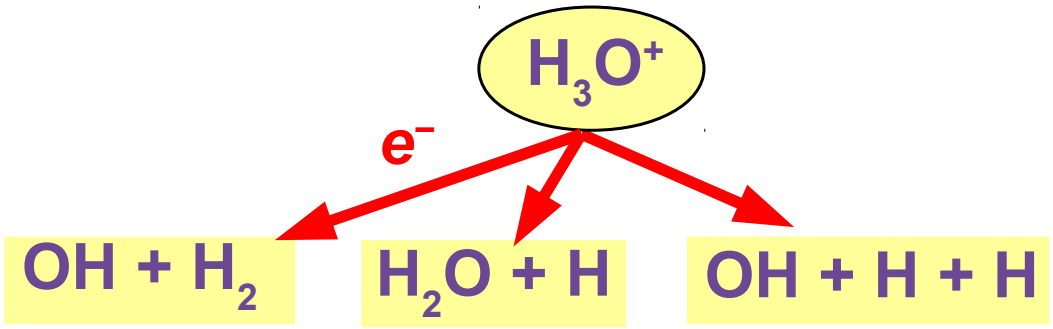


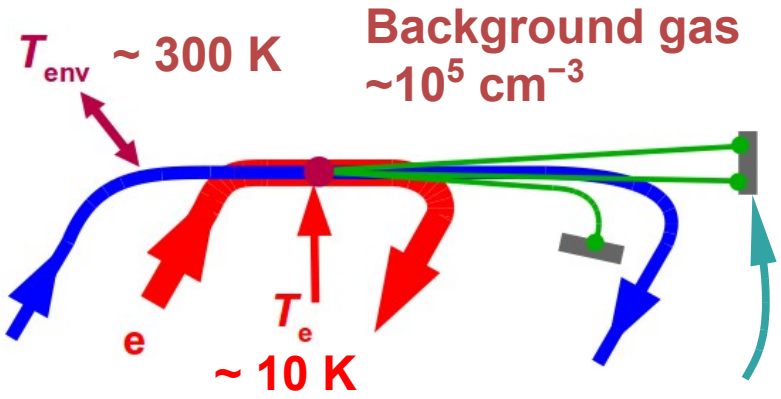


EMU detector:
Fragment mass identification and momentum spectroscopy



Probing the fragmentation into molecular products





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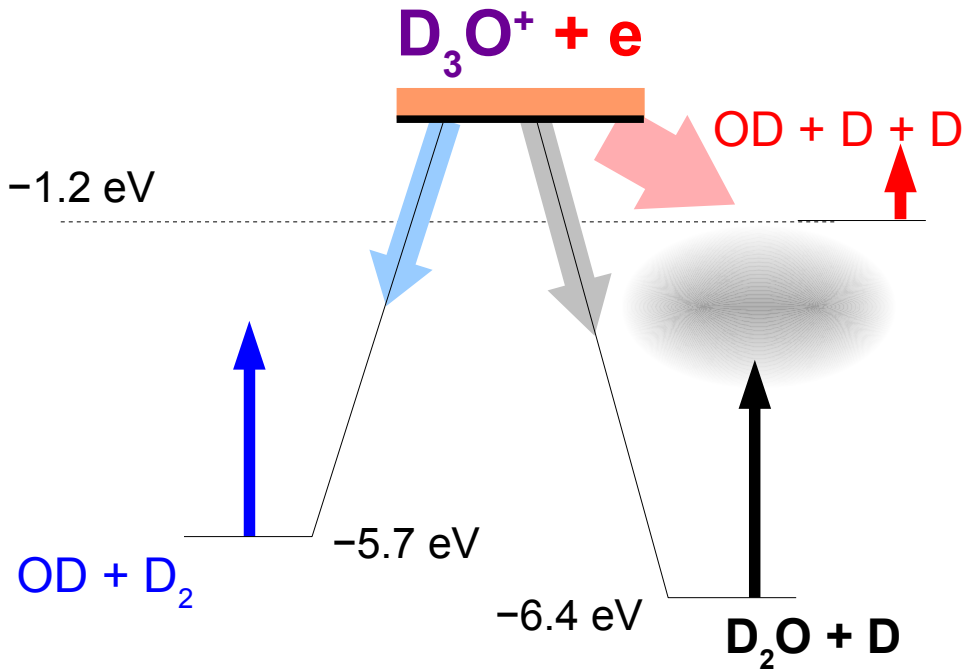
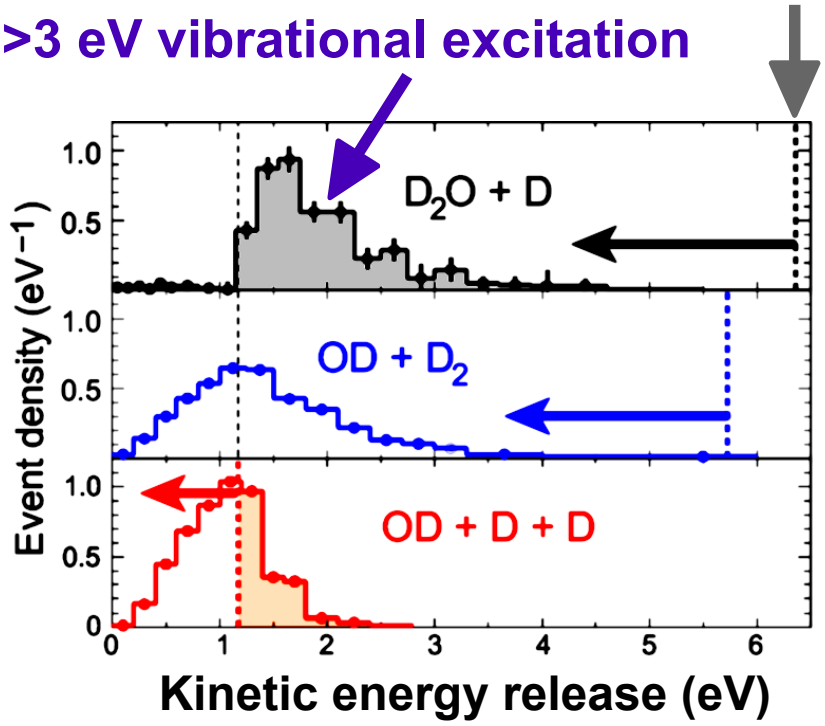


Ground state water product

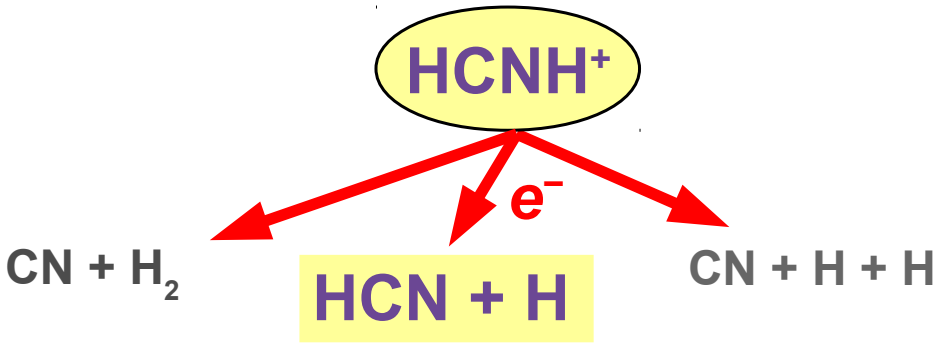
>3 eV vibrational excitation

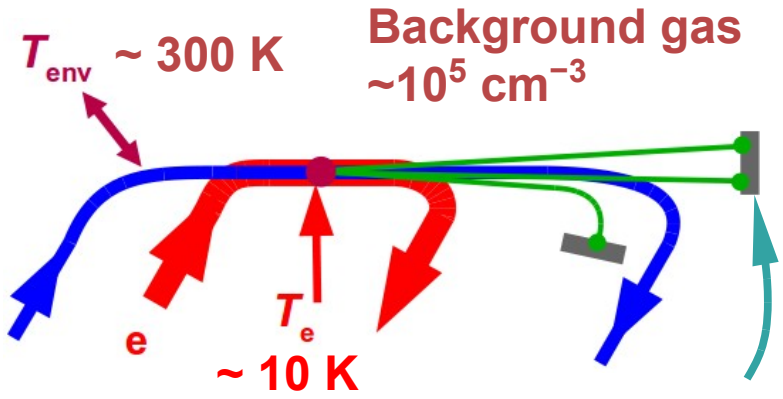


Water formed with extreme inner excitation

H. Buhr et al., Phys. Rev. Lett. 105, 103202 (2010)

Production of molecular isomers by dissociative recombination





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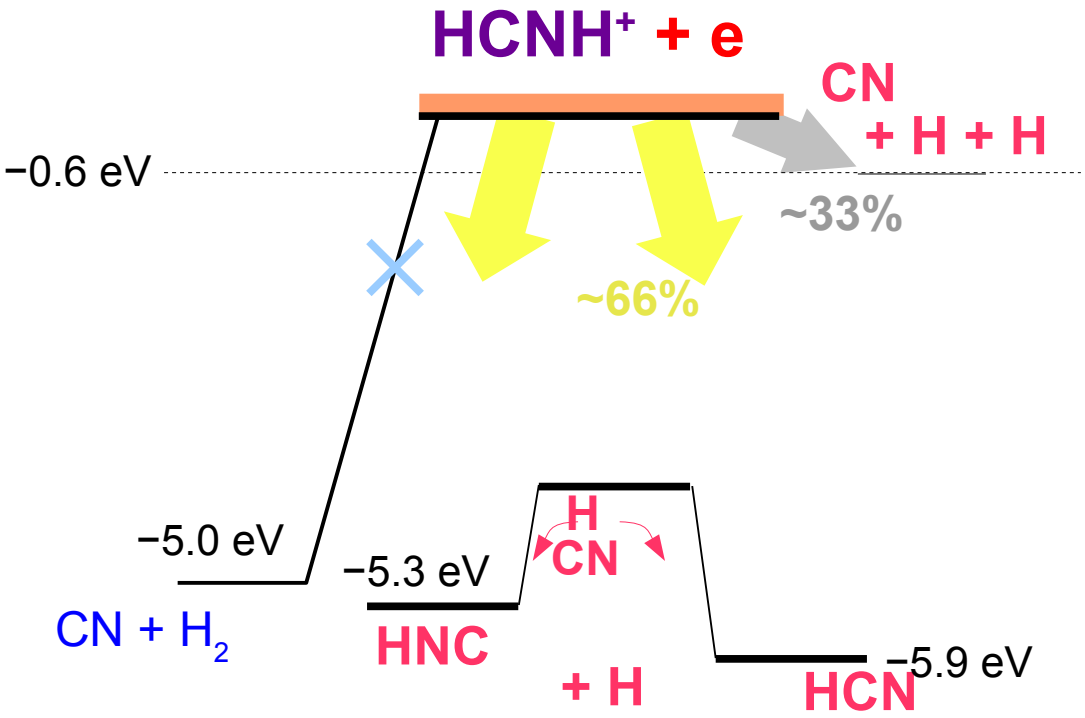

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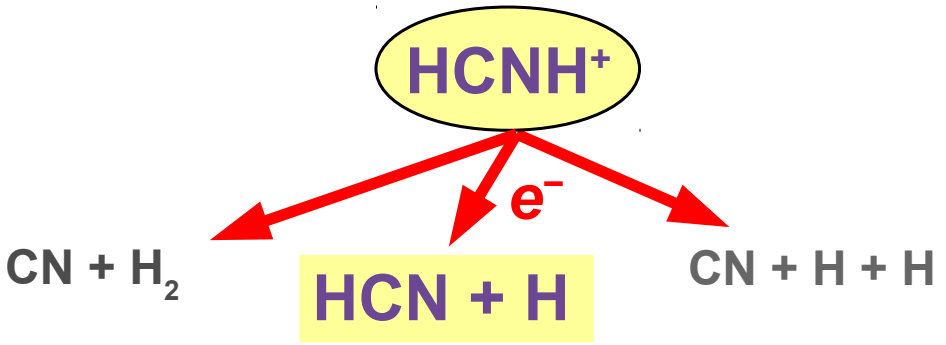

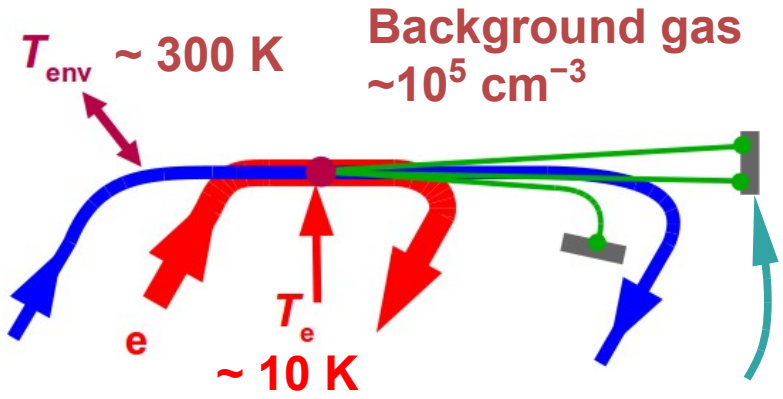

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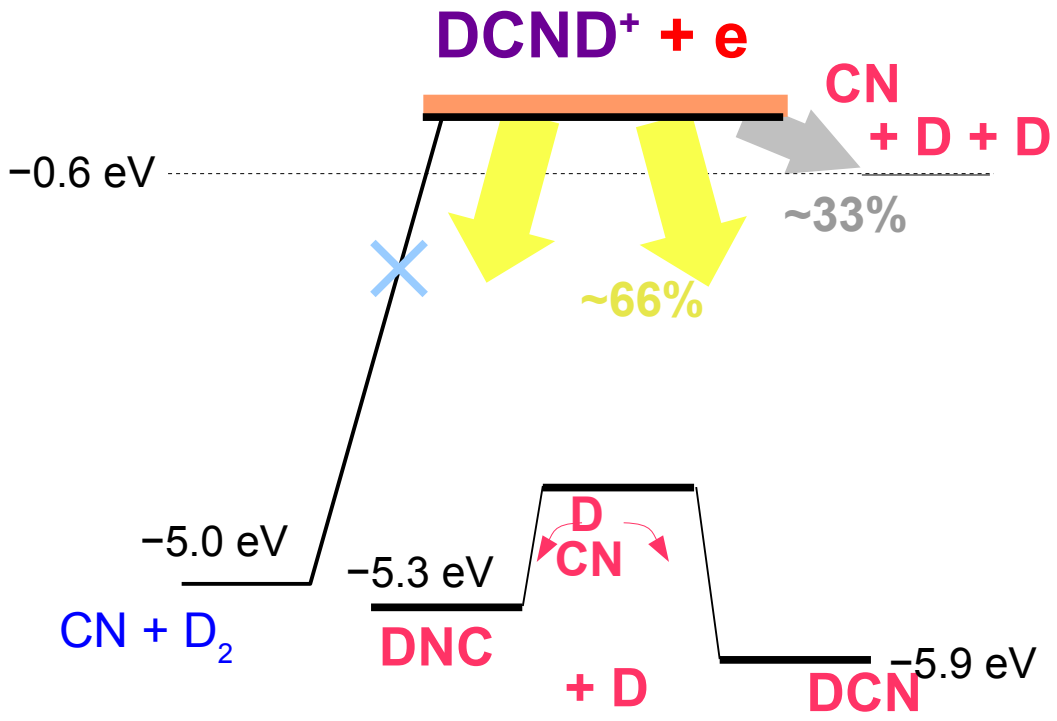

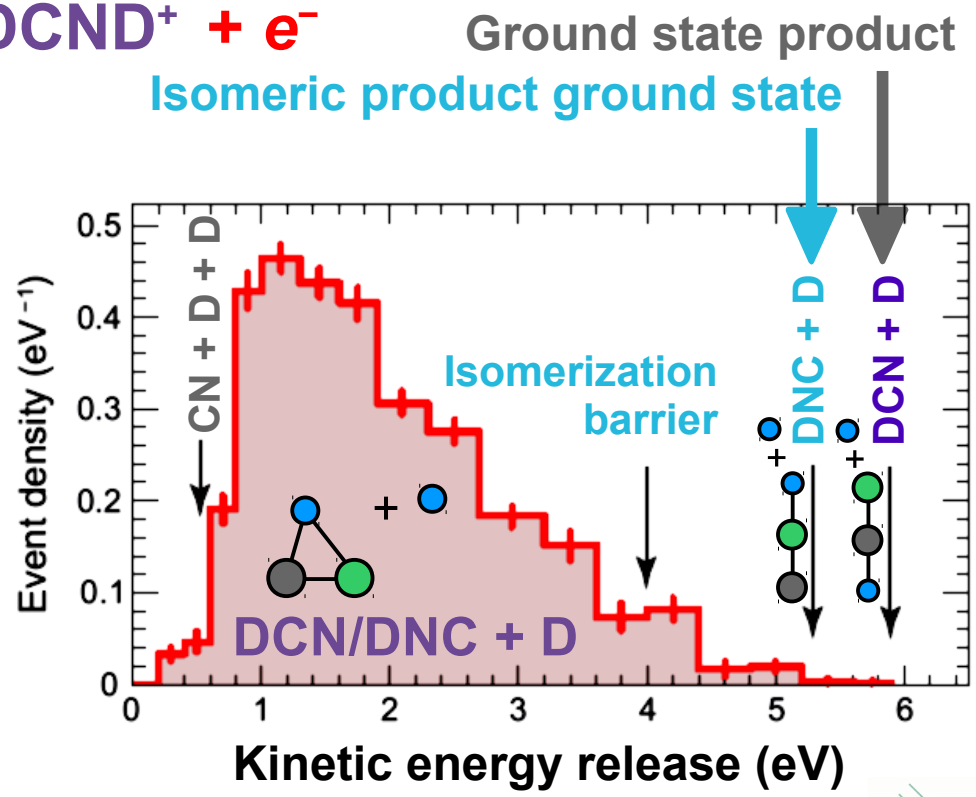

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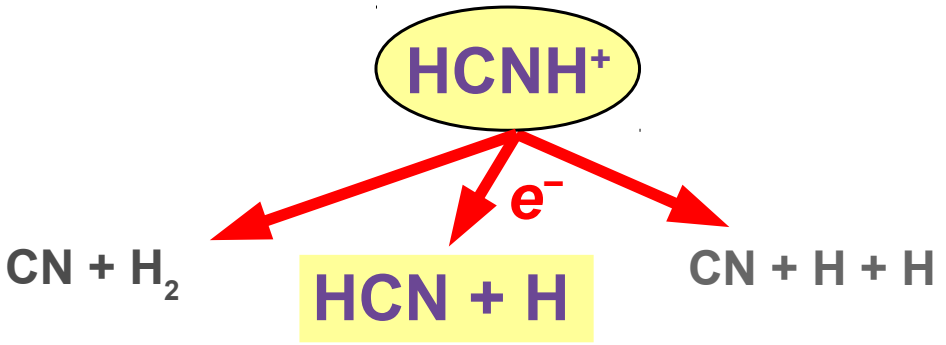

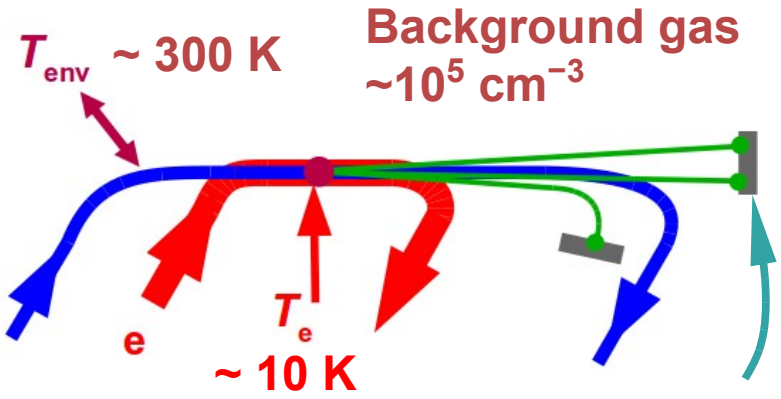

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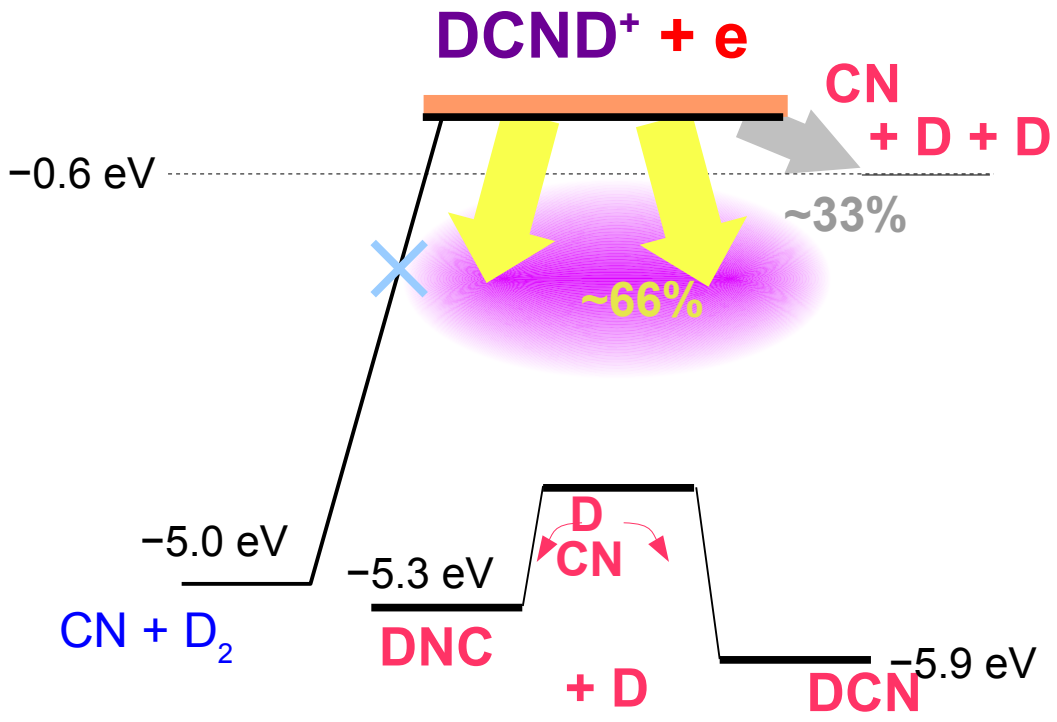
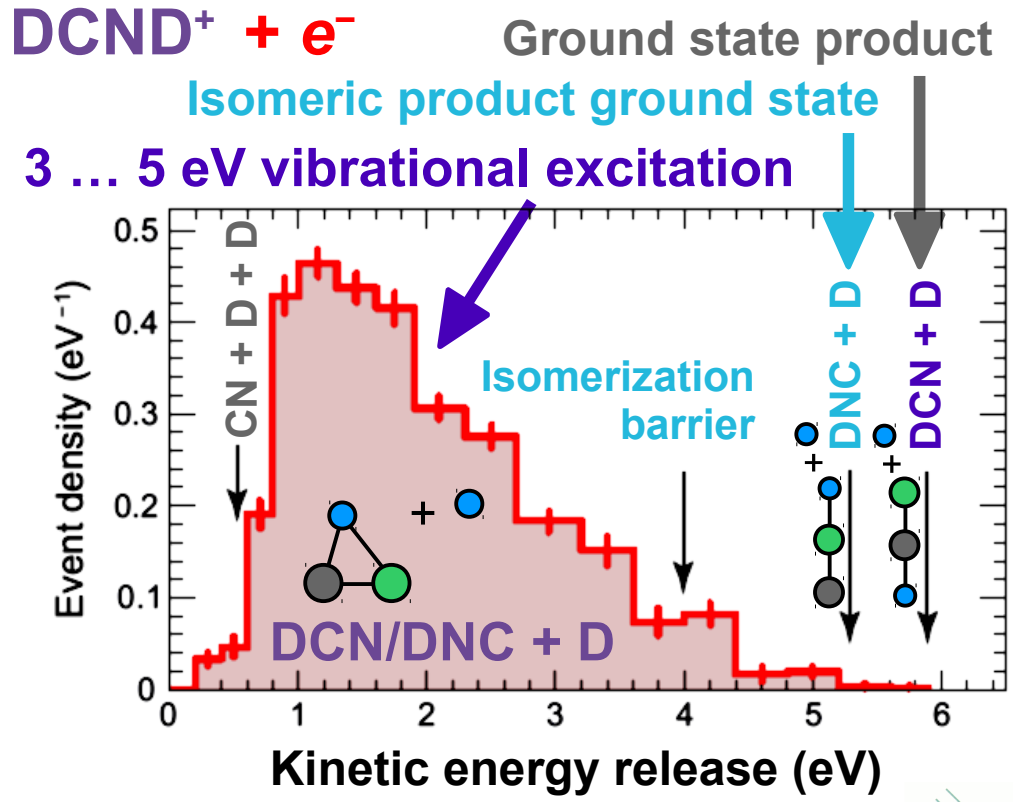
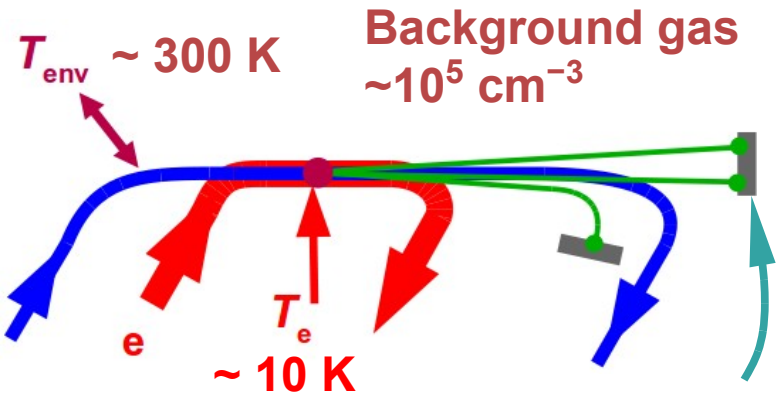
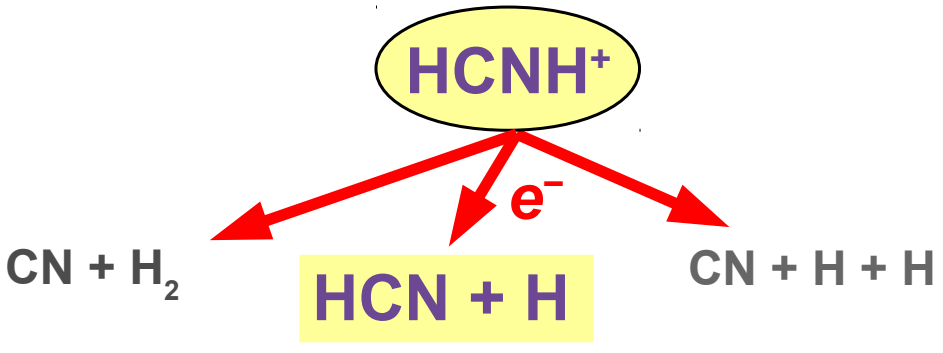


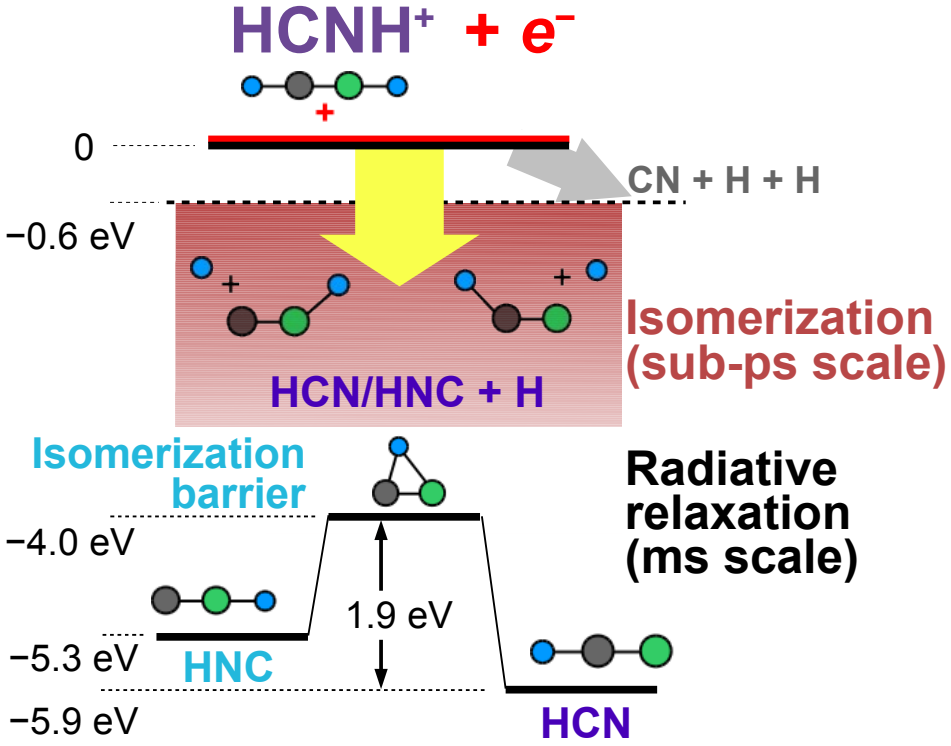


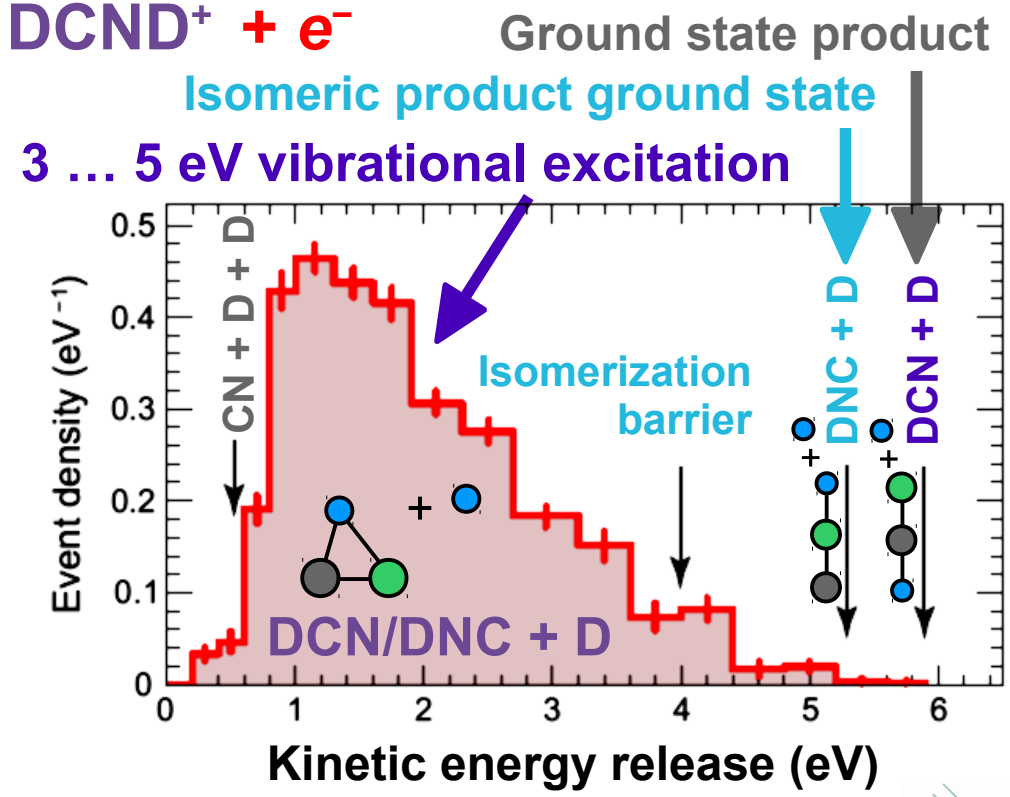
Production of molecular isomers by dissociative recombination



EMU detector:
Fragment mass identification
and momentum spectroscopy



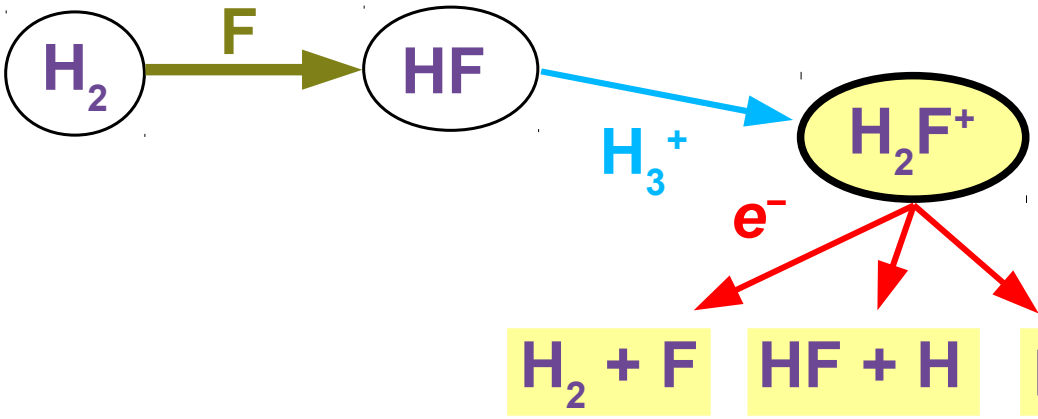


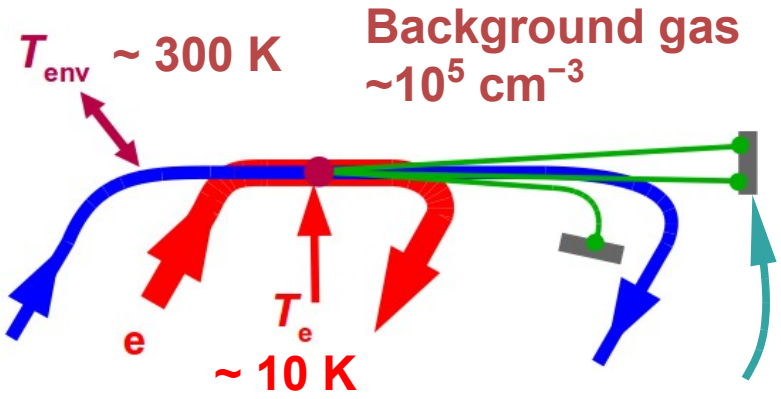


[HCN]/[HNC] of ~1:1 by electron recombination

M. Mendes et al., Astrophys. J. Lett. 746, L8 (2012)

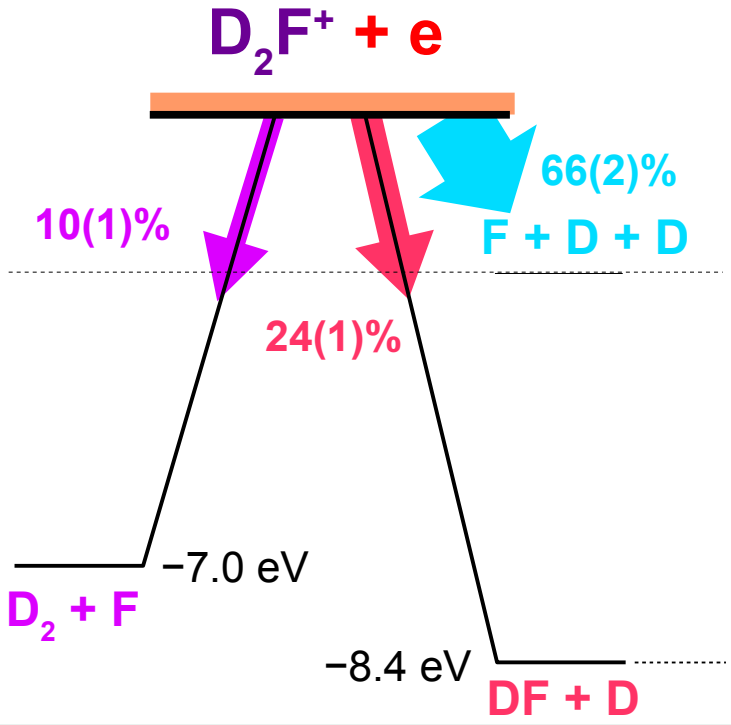
Halogen chemistry: destruction of HF





EMU detector:
Fragment mass identification
and momentum spectroscopy

Strong three-body (fully atomic) fragment channel



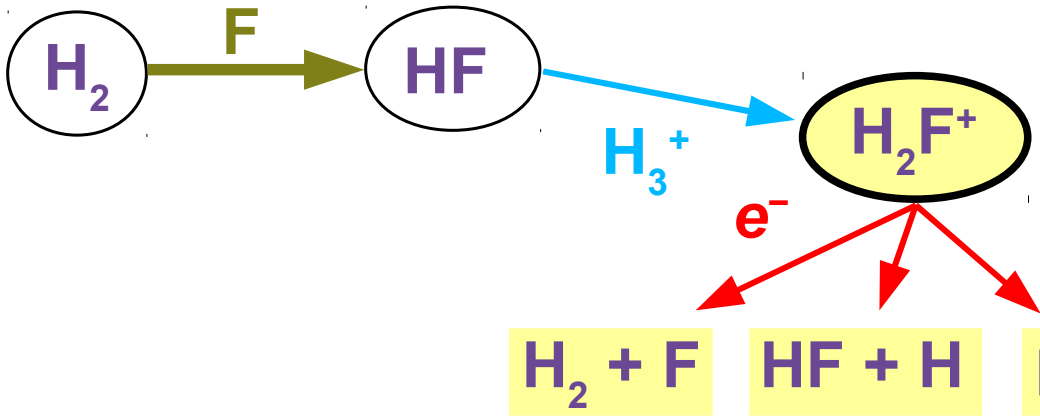
O. Novotný

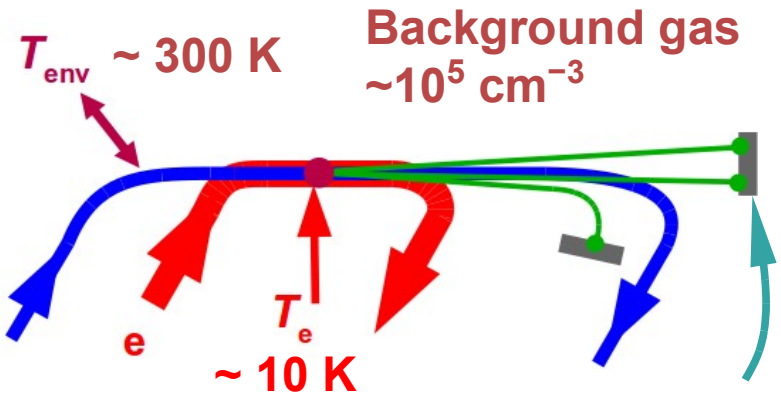
Columbia/
Heidelberg
collaboration

2010/11

(also with
W. Geppert
M. Hamberg,
Stockholm Univ.)

Halogen chemistry: destruction of HF





EMU detector:
Fragment mass identification and momentum spectroscopy

Strong three-body (fully atomic) fragment channel

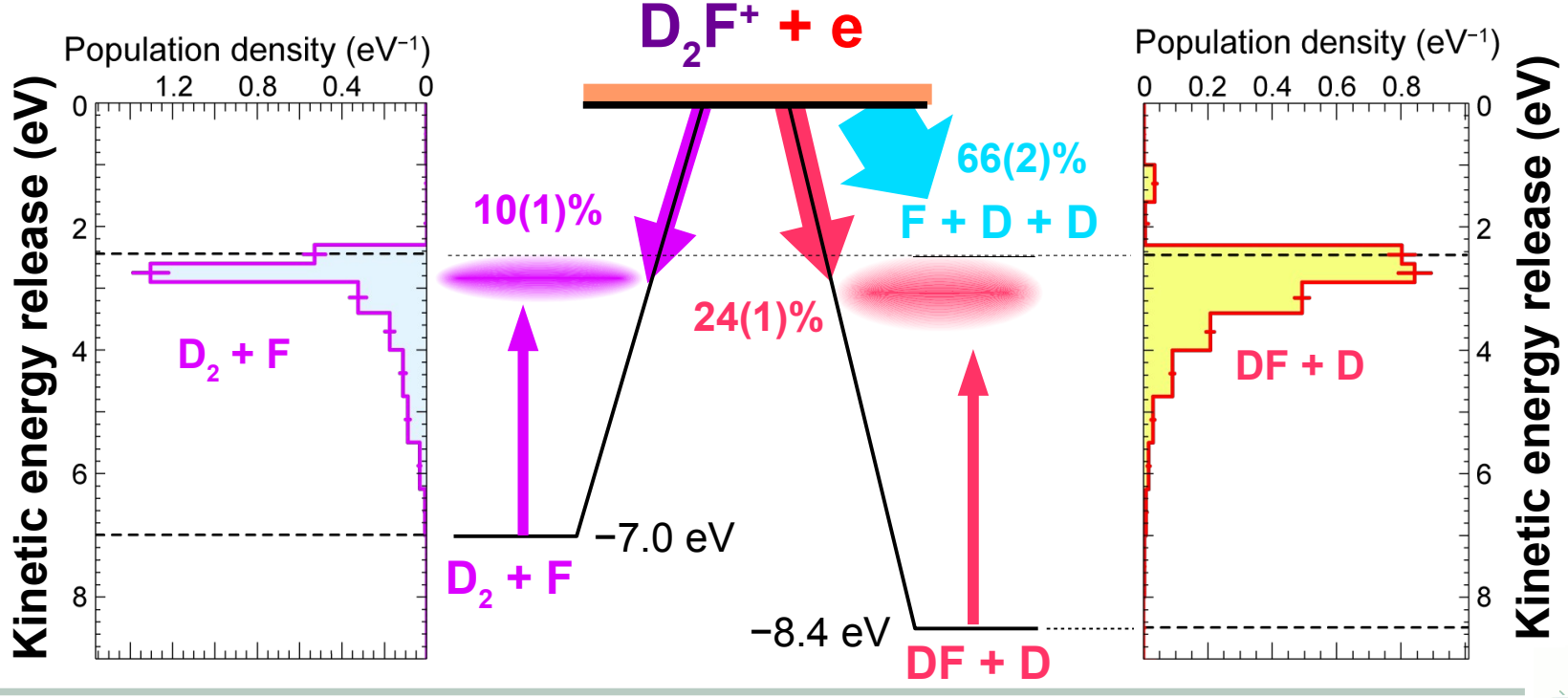
Extreme vibrational excitation in all molecular product channels

O. Novotný

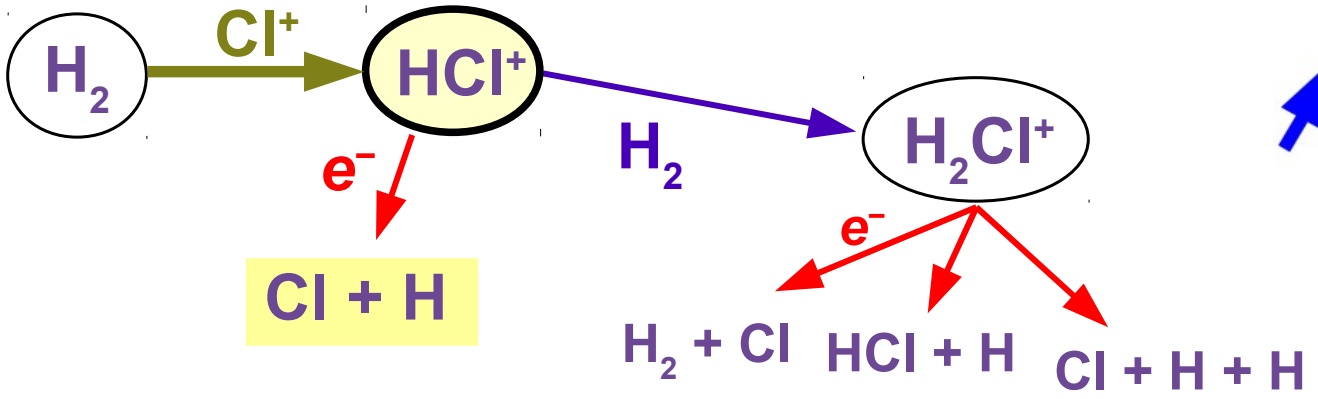
Columbia/
Heidelberg
collaboration

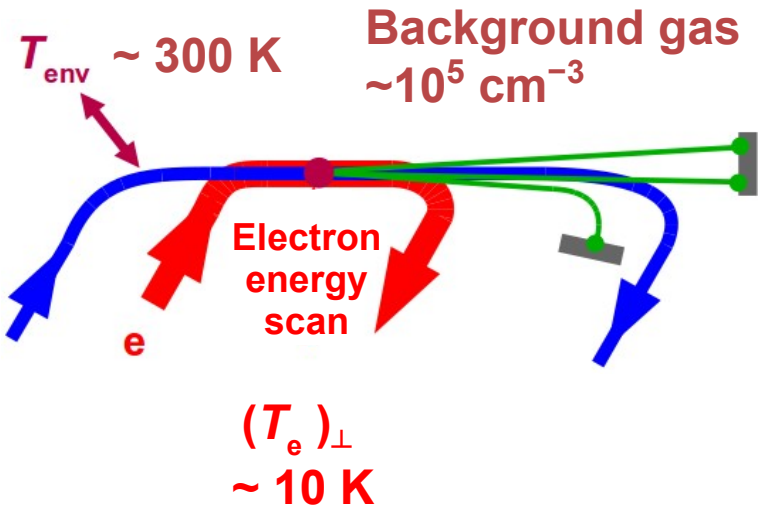
2010/11

(also with
W. Geppert
M. Hamberg,
Stockholm Univ.)



Halogen chemistry: HCl^+ DR rate



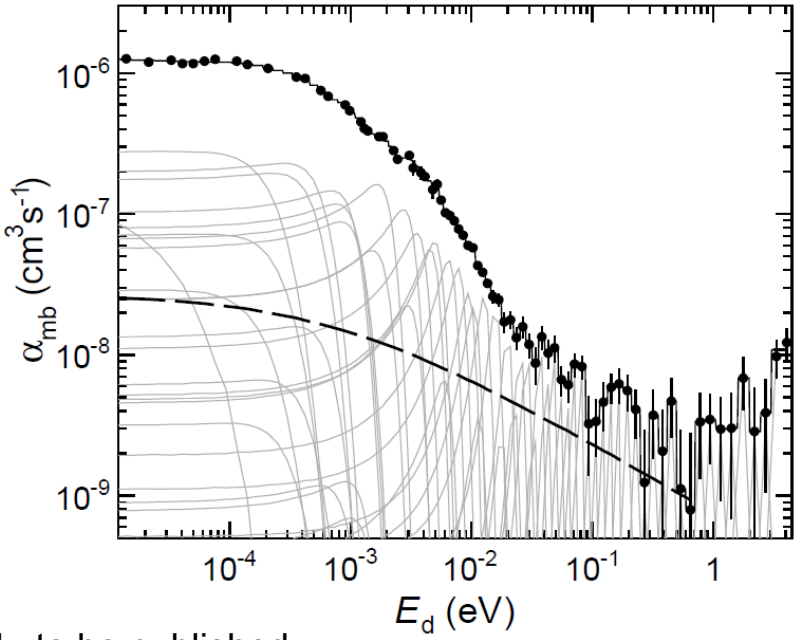


Fitting-deconvolution method for low-energy DR cross section

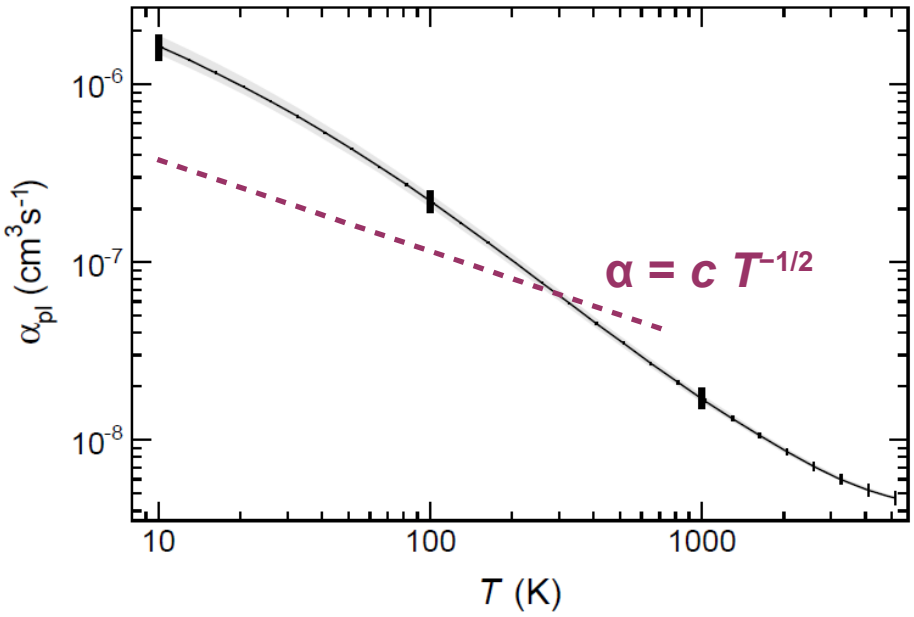
O. Novotný

Columbia/
Heidelberg
collaboration

2010/11



Plasma rate coefficient

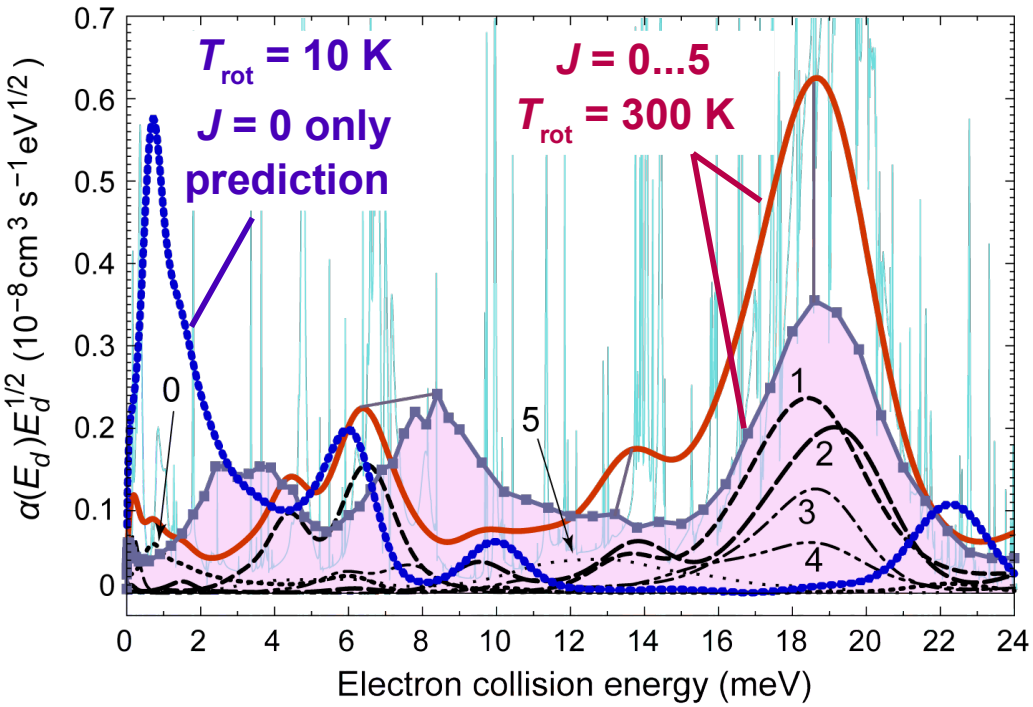


O. Novotný et al., to be published

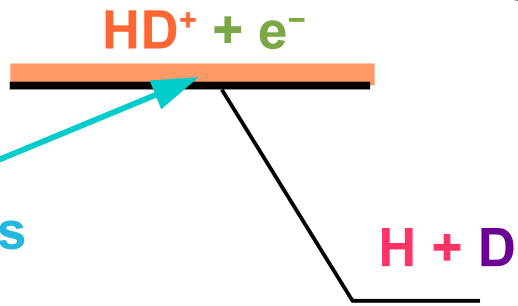
Toward cryogenic ion beam experiments

TSR result for recombination cross section

$HD^+ + e^- \rightarrow H + D$

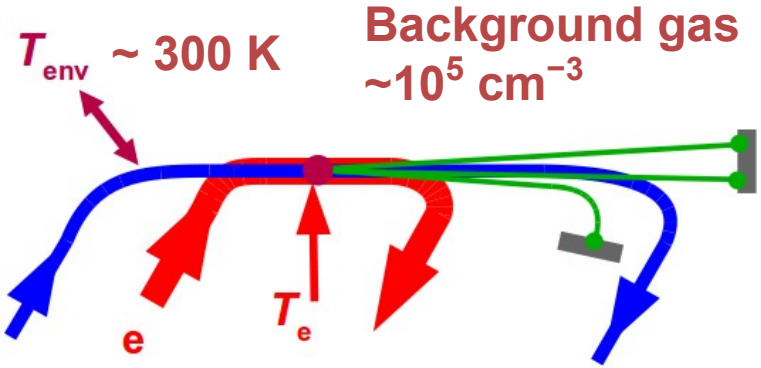


Waffeu-Tamo et al.,
Phys. Rev. A 84, 022710 (2011)

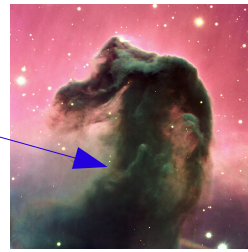


Rovibrational capture resonances

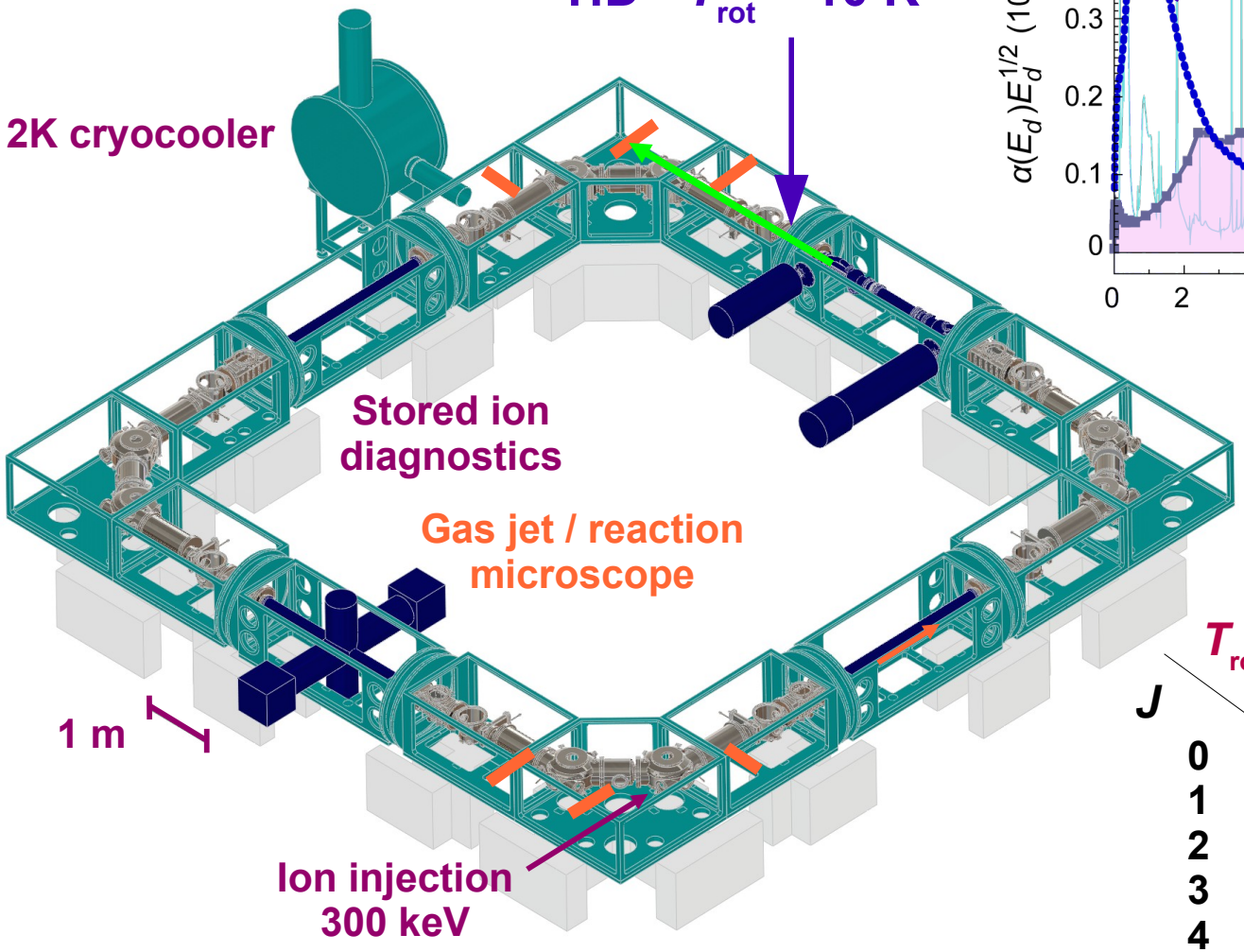
Measured recombination cross section dominated by $J \geq 1$

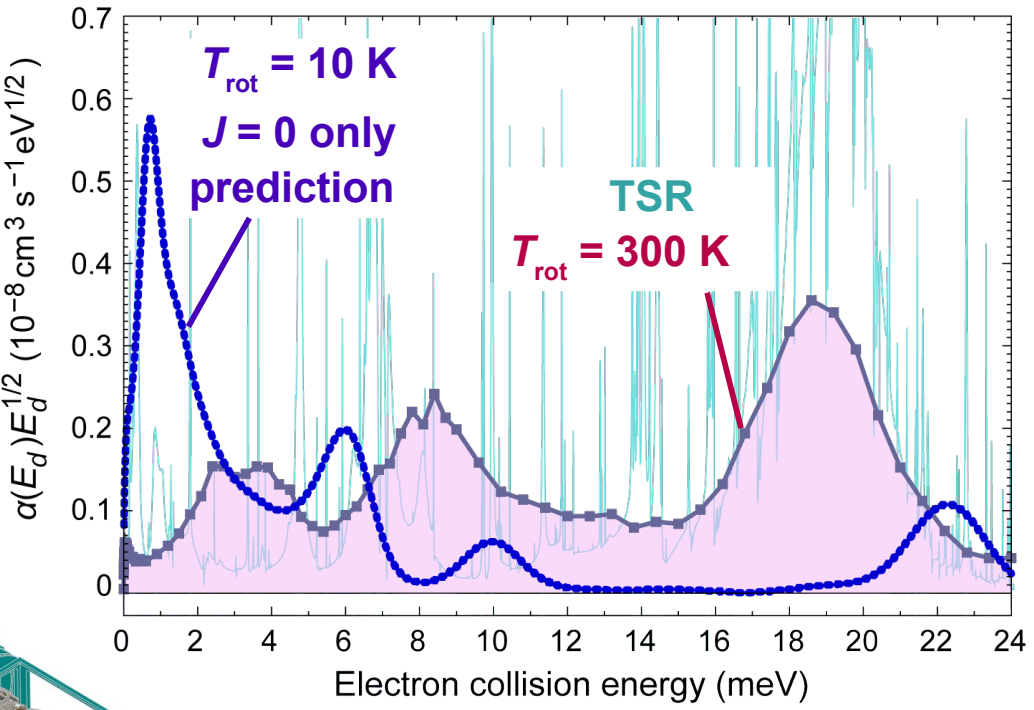


J	T_{rot} 300 K	10 K
0	0.104	0.995
1	0.251	0.005
2	0.271	0.0
3	0.199	0.0
4	0.108	0.0



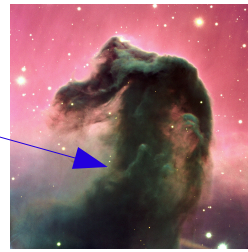
Toward cryogenic ion beam experiments





Rydberg capture resonances at low rotation

J	T_{rot} 300 K	10 K
0	0.104	0.995
1	0.251	0.005
2	0.271	0.0
3	0.199	0.0
4	0.108	0.0



Cryogenic electrostatic storage ring CSR

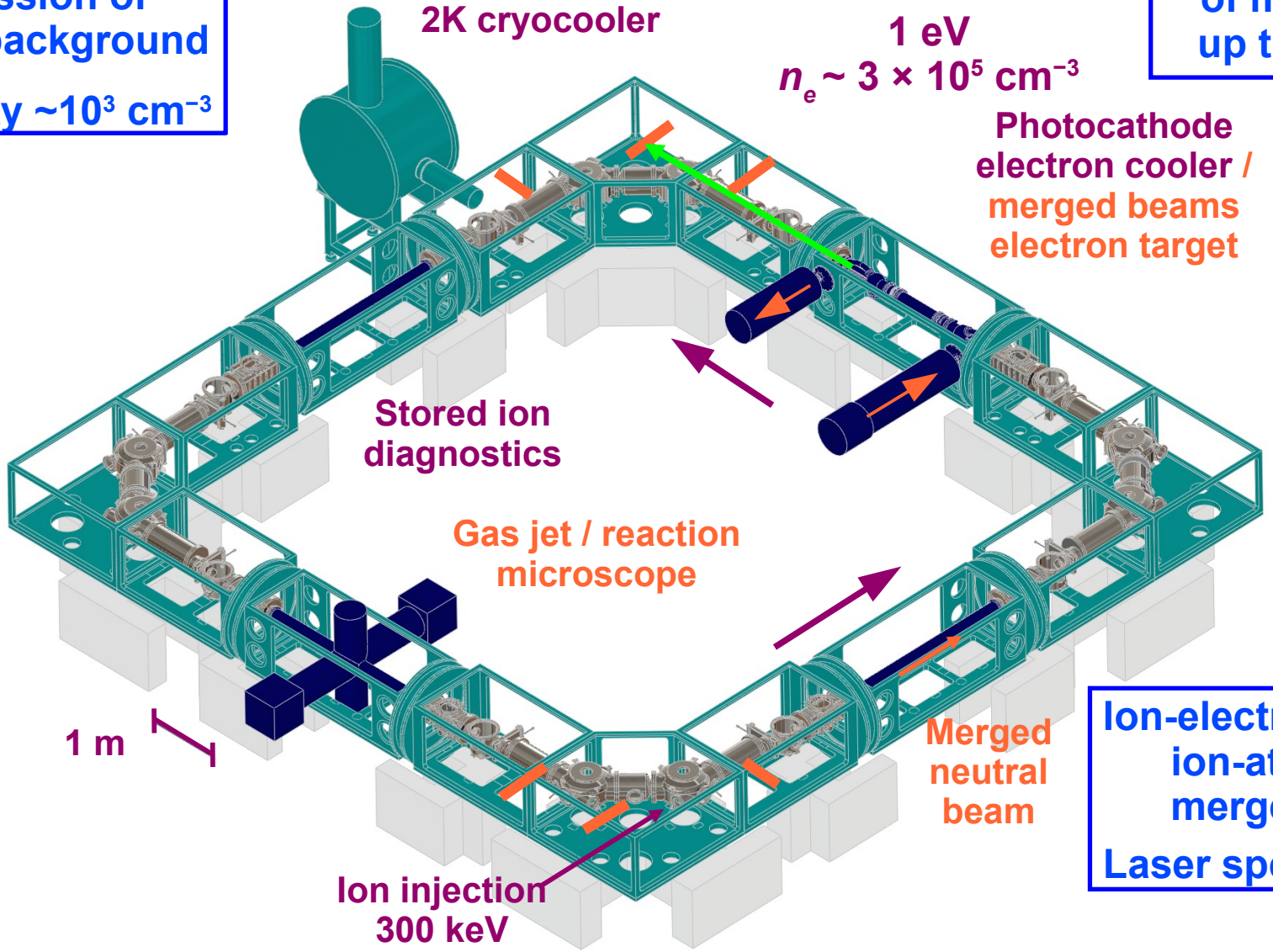
A 300 keV ion storage ring with electrostatic beam optics for molecular ions and clusters (cations, anions),

heavy atomic beams, highly charged ions

2 K cryopumping and suppression of radiation background

Gas density ~10^3 cm^{-3}

Electron cooling of molecules up to A ~ 160

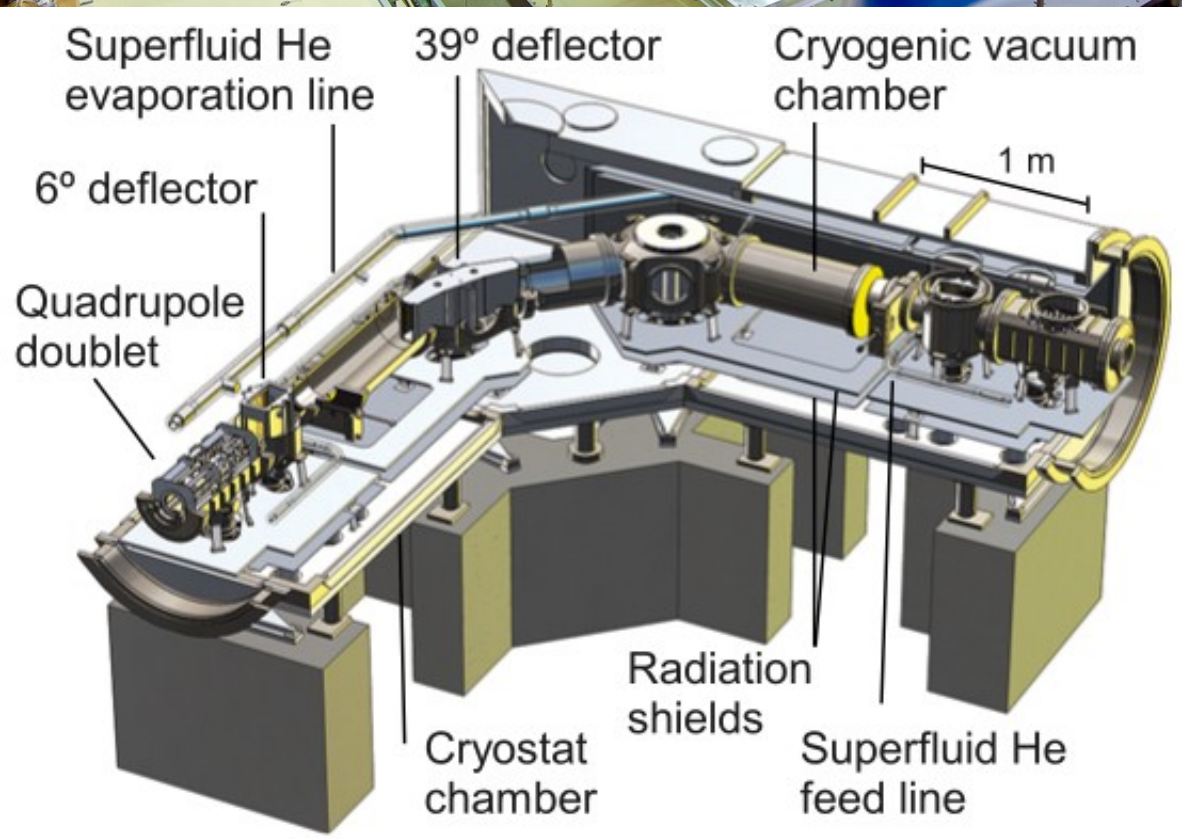


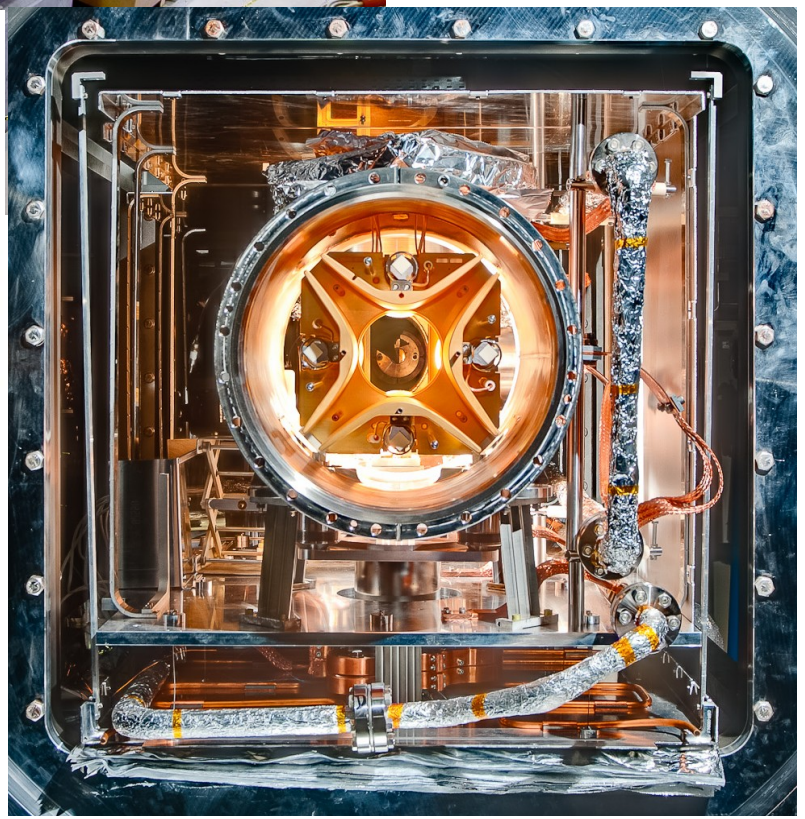
D. A. Orlov, C. Krantz, A. Shornikov et al.

Ion-electron and ion-atom merged beams

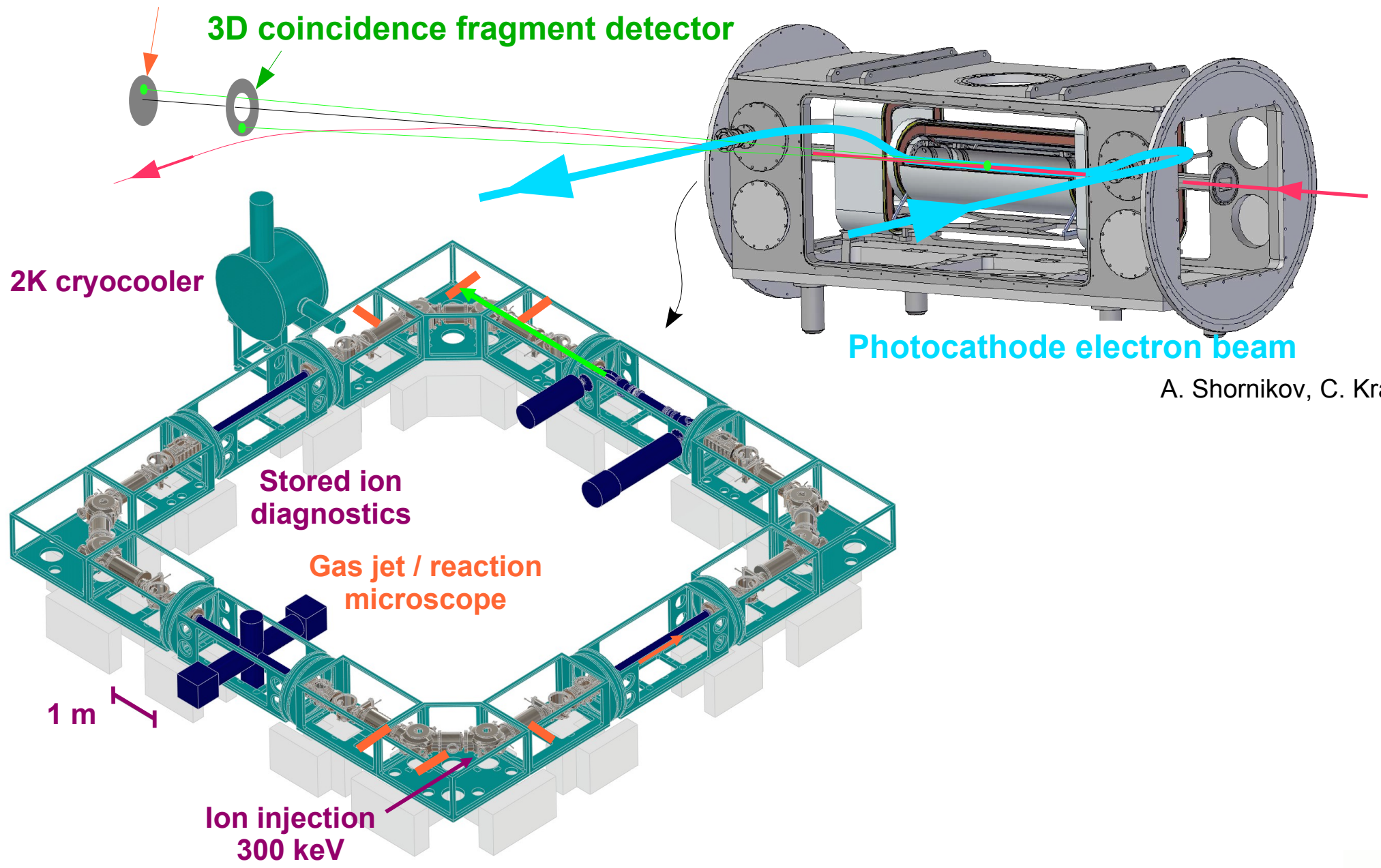
Laser spectroscopy

Cryogenic storage ring CSR



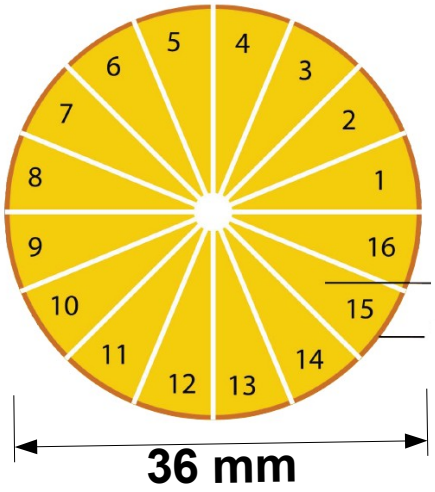


Cryogenic merged electron–ion beams



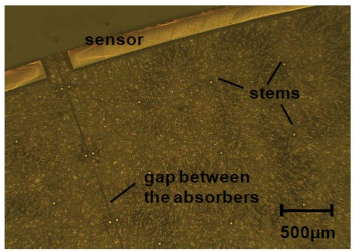
A. Shornikov, C. Krantz

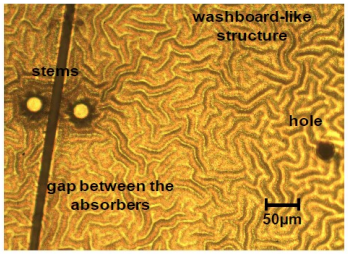
Fragment imaging microcalorimeter

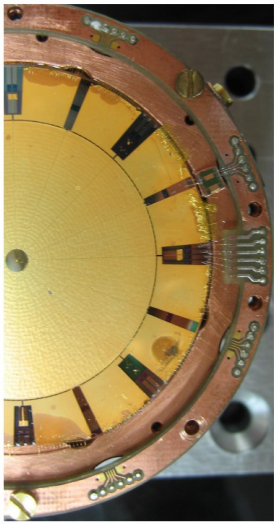


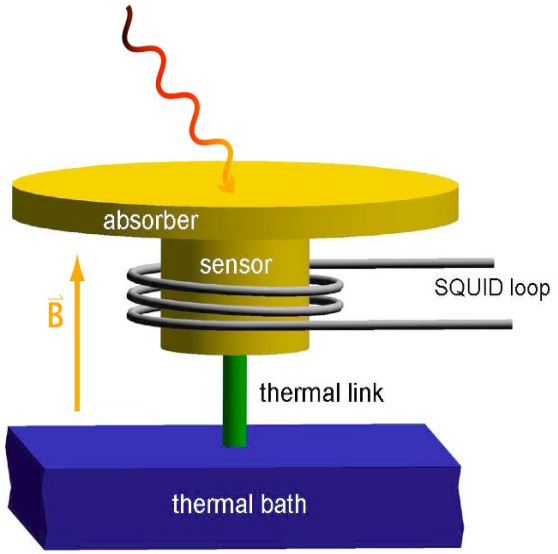
Absorber

Thermal sensor







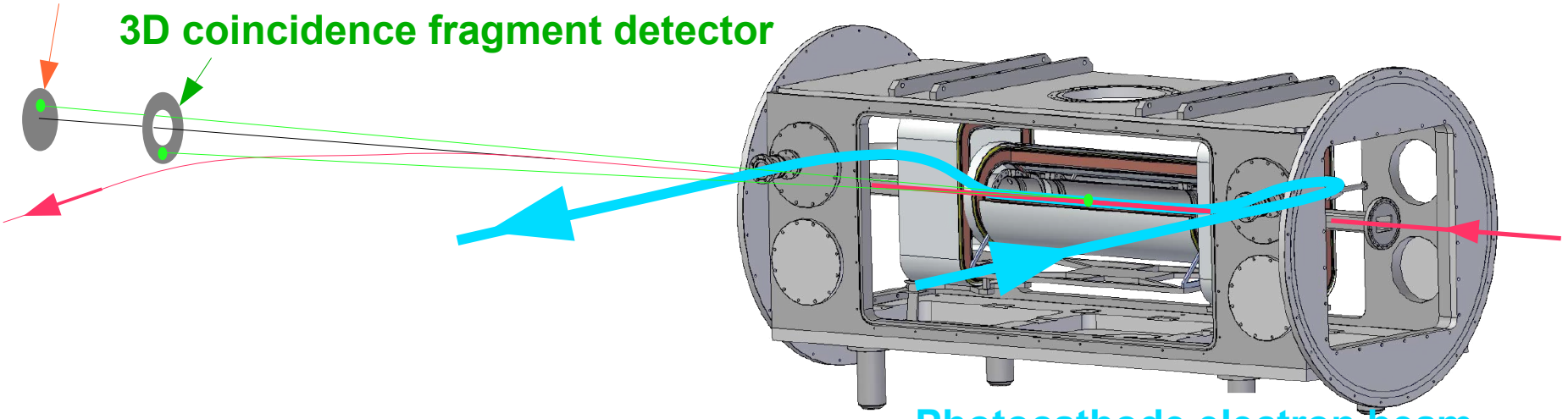


A. Fleischmann, C. Enss et al.

In development at
Kirchhoff Institute of Physics
University of Heidelberg

Segmented microcalorimeter detector

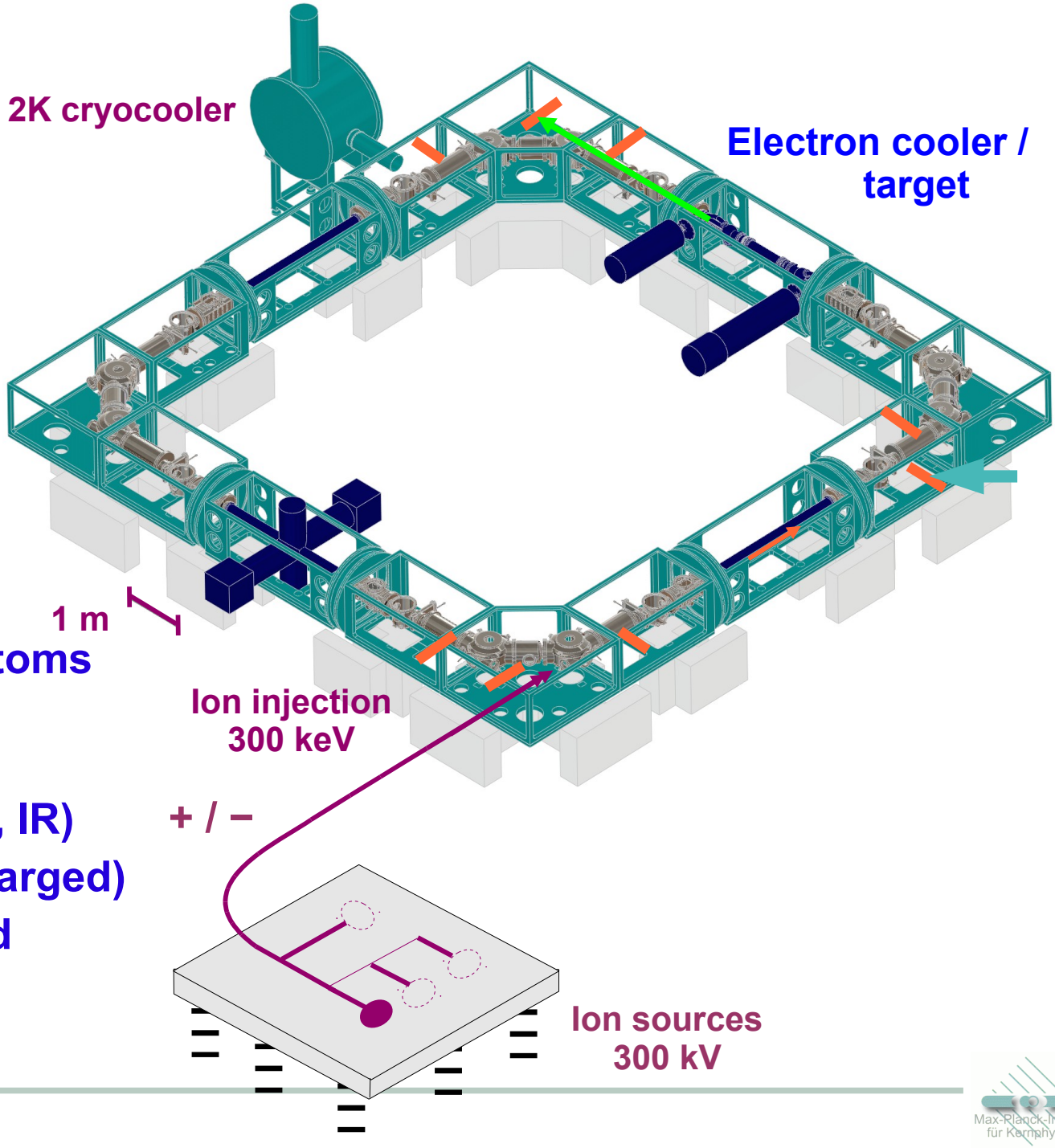
3D coincidence fragment detector



Photocathode electron beam

A. Shornikov, C. Krantz

Cryogenic ion beam experiments at CSR



- Stored ion beams at 10 K internal temperature
- Organic molecules, heavy atoms (300 keV for all masses)
- Rotationally resolved ion spectroscopy (laser, IR)
- Negative ions (also multi-charged)
- Cluster systems, H_2O-loaded

Cryogenic ion beam experiments at CSR

Holger Kreckel group @ MPIK

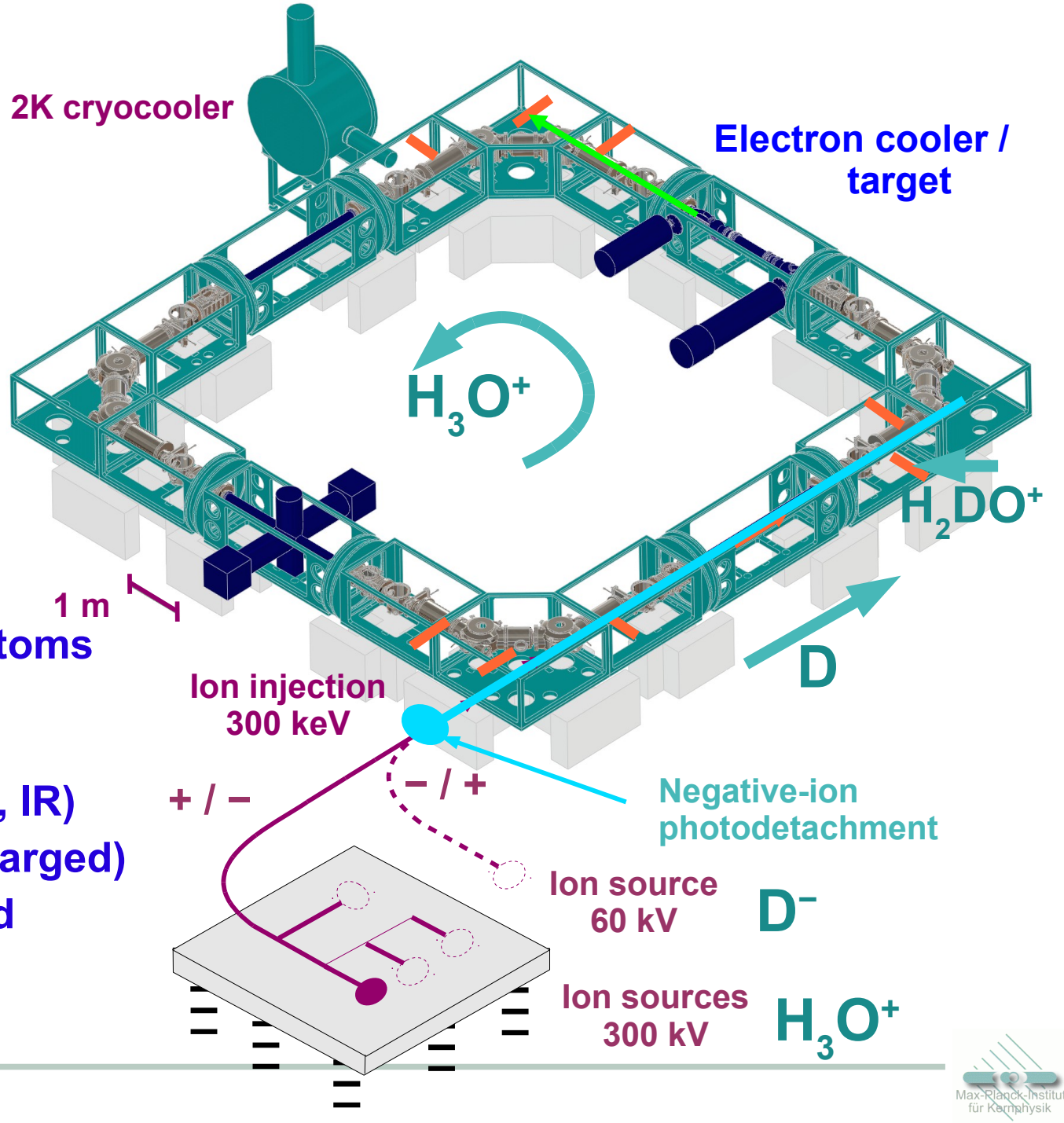
Ion–atom merged beams

Example:

$D + H_3O^+ \rightarrow H + H_2DO^+$

→ Deuterium enrichment by ion chemistry

- Stored ion beams at 10 K internal temperature
- Organic molecules, heavy atoms (300 keV for all masses)
- Rotationally resolved ion spectroscopy (laser, IR)
- Negative ions (also multi-charged)
- Cluster systems, H_2O-loaded



Summary

Storage-ring merged beams technique for molecular fragmentation
- **Cross sections, branching ratios, product excitation**

Experimental advances:
Collision energy resolution down to ~10 K (photocathodes)
Mass sensitive fragment imaging detectors

Recent results for dissociative recombination:
- **Collision energy dependence of final-channel partial rates**
- **Deconvolution of low-energy rate coefficients**
- **Internal excitation of molecular fragments**

New developments using cryogenic ion storage:
- **Internal state control**
- **Merged beams experiments in electrostatic rings**
- **Goal to implement cryogenic microcalorimeters**

The collaboration

Max-Planck-Insitut für Kernphysik, Heidelberg

Division on Stored and Cooled Ions (K. Blaum)

Atomic and molecular quantum dynamics

O. Novotný(*), C. Krantz, M. Froese, M. Lange

PhD: **A. Shornikov, S. Menk, C. Domesle, K. Spruck, A. Becker, P. Herwig, Bian Yang**

Recently completed post-docs: **H. Buhr, A. Petrignani**

Recently completed PhD: **J. Stützel, M. Berg, M. Mendes, D. Bing, F. Laux**

Cooled and stored ions instrumentation

M. Grieser, R. von Hahn, R. Repnow

PhD: **F. Fellenberger, F. Berg**

Heidelberg collaborations

Kirchhoff Inst., Univ. Heidelberg

A. Fleischmann
C. Enss

Microcalorimeter ion detector

Max-Planck-Institut für Astronomie

External collaborations

Weizmann Institute of Science Rehovot, Israel

D. Zajfman, O. Heber, D. Schwalm

DFG ISM-SPP

Columbia Univ., NYC

D. Savin, O. Novotný(*)

Univ. Giessen, Germany

S. Schippers, A. Müller

Univ. of Illinois, Urbana

B. McCall, H. Kreckel

Univ. Stockholm

W. D. Geppert

Univ. Louvain-La-Neuve, Belgium

X. Urbain

MAX-PLANCK-INSTITUT FÜR KERNPHYSIK