Effects of Molecular Rotation in Low-Energy Electron Collisions of H_3^+

Andreas Wolf

Max-Planck Institute for Nuclear Physics, Heidelberg

Physics, Chemistry and Astronomy of H_3^+, London, 16-19 January 2006

- Low-energy H_3^+ collision processes
- Cold collisions in stored ion beams: instrumentation
  - Cryogenic injector ion trap
  - Photocathode electron beam
- Energy-resolved low-energy recombination (DR) of cold H_3^+
- Spin-symmetry effect on DR: exploratory study
- Long-time heating and cooling of H_3^+ probed by low-energy DR
- Summary and outlook

Ion Storage and Molecular Quantum Dynamics

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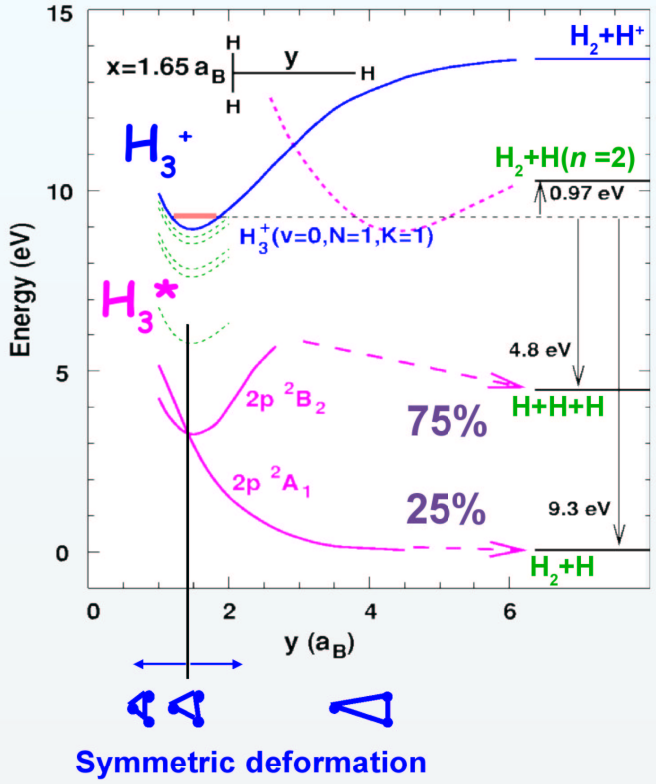
German-Israeli Foundation for Fundamental Research (GIF) 2002–05

European Research Network "Electron transfer reactions" 2000–04

Low-energy collisions of H_3^+



$H_3^+ + e$ **(00) *JK***

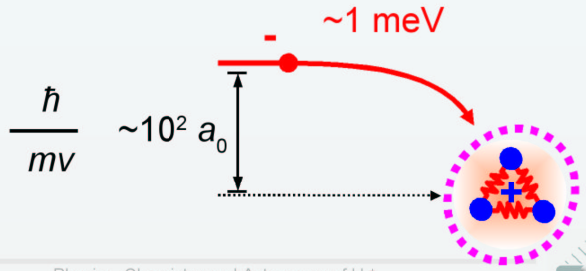
Autoionizing Rydberg resonance

H_3^* **$(v_1' v_2')$ *J'K'***

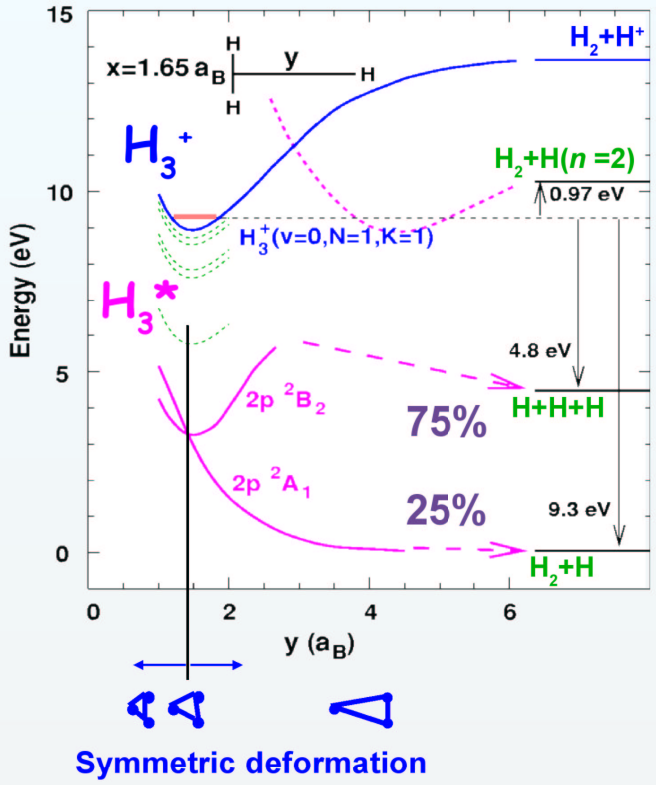
$H + H + H$

$H_2 + H$

Dissociative recombination



Low-energy collisions of H_3^+



$H_3^+ + e$ **(00) *JK***

Autoionizing Rydberg resonance

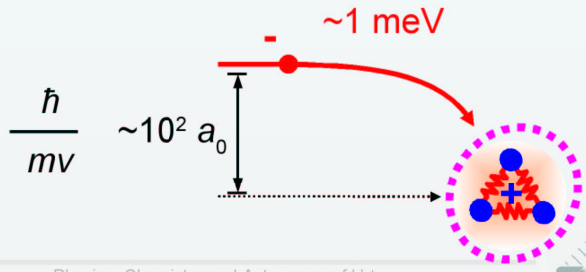
H_3^* **$(v_1' v_2')$ *J'K'***

$H + H + H$ $(H_3^+)' + e'$

$H_2 + H$ **$(v_1'' v_2'')$ *J''K''***

Dissociative recombination

Super-elastic collisions
Electron impact excitation



Effects of Molecular Rotation in Low-Energy Electron Collisions of H_3^+ Physics, Chemistry and Astronomy of H_3^+ London, 16-19 January 2006 Andreas Wolf

Storage ring DR measurements

Milli-eV electron collisions

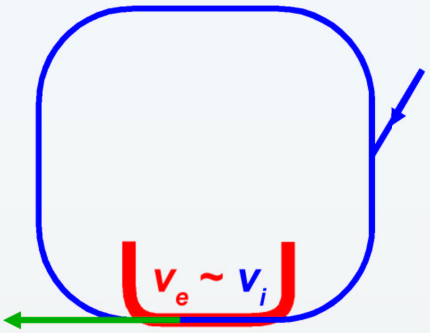
Merged beams, ~5 K electrons

Control of molecular excitation

Ion storage

Neutral fragments with eV energies

Fast (MeV) beam
→ efficient detection



Ion storage ring (~ MeV energy)

Merged electron beam (~keV energy)

Electron cooling: $v_i \stackrel{!}{=} v_e$

Collision measurements: $v_e \stackrel{!}{\neq} v_i$

⇒ collision energy
~1 meV up to keV

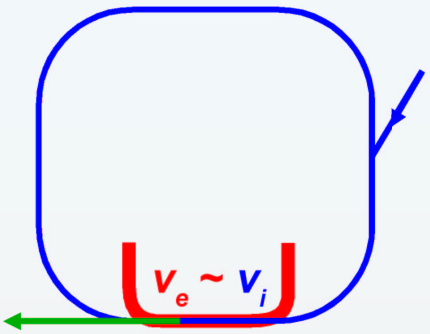
Storage ring DR measurements

Storage time up to 80 s

Radiative relaxation
Black-body equilibrium
Cooling by milli-eV electron collisions

Cold ion sources

Cooled hollow cathode
Expanding jet discharge
10 K buffer-gas cooling injector trap



Ion storage ring (~ MeV energy)

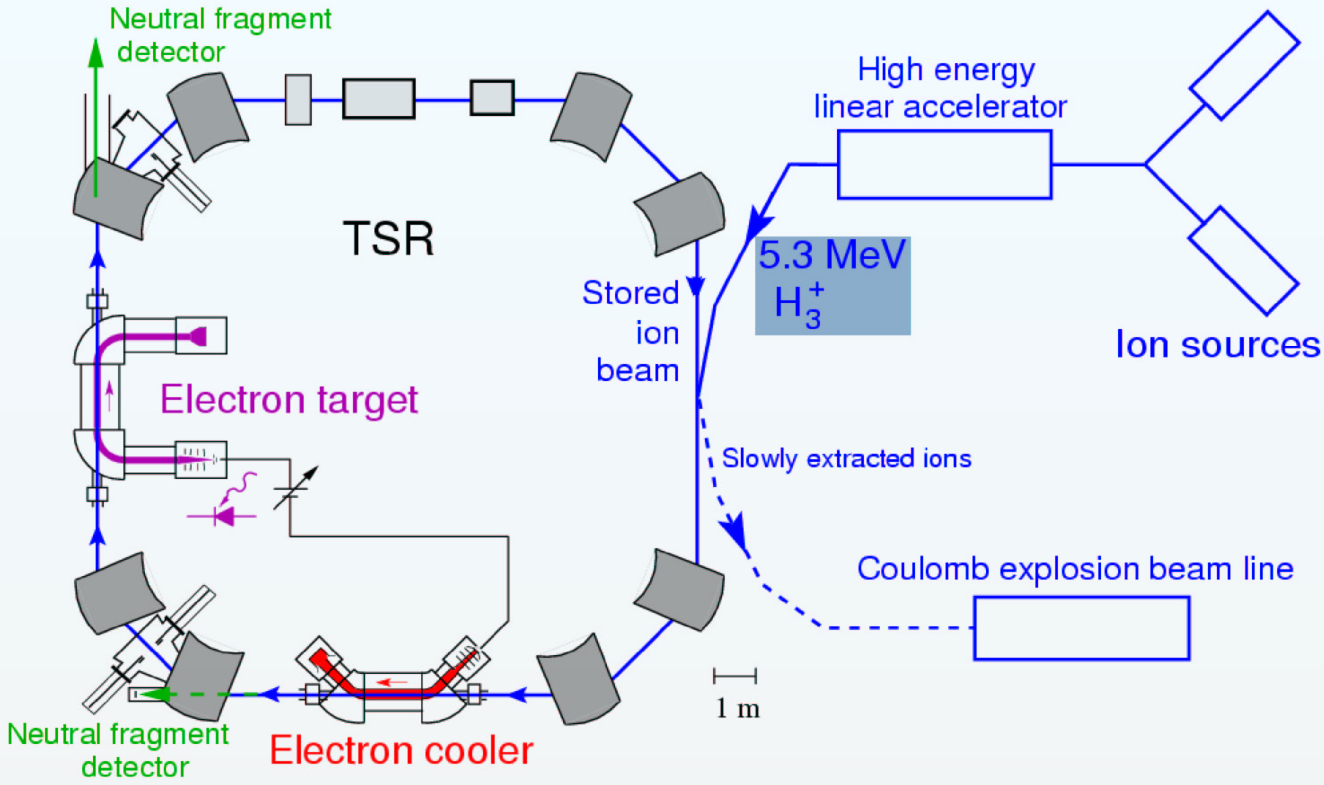
Merged electron beam (~keV energy)

Electron cooling: $v_i \stackrel{!}{=} v_e$

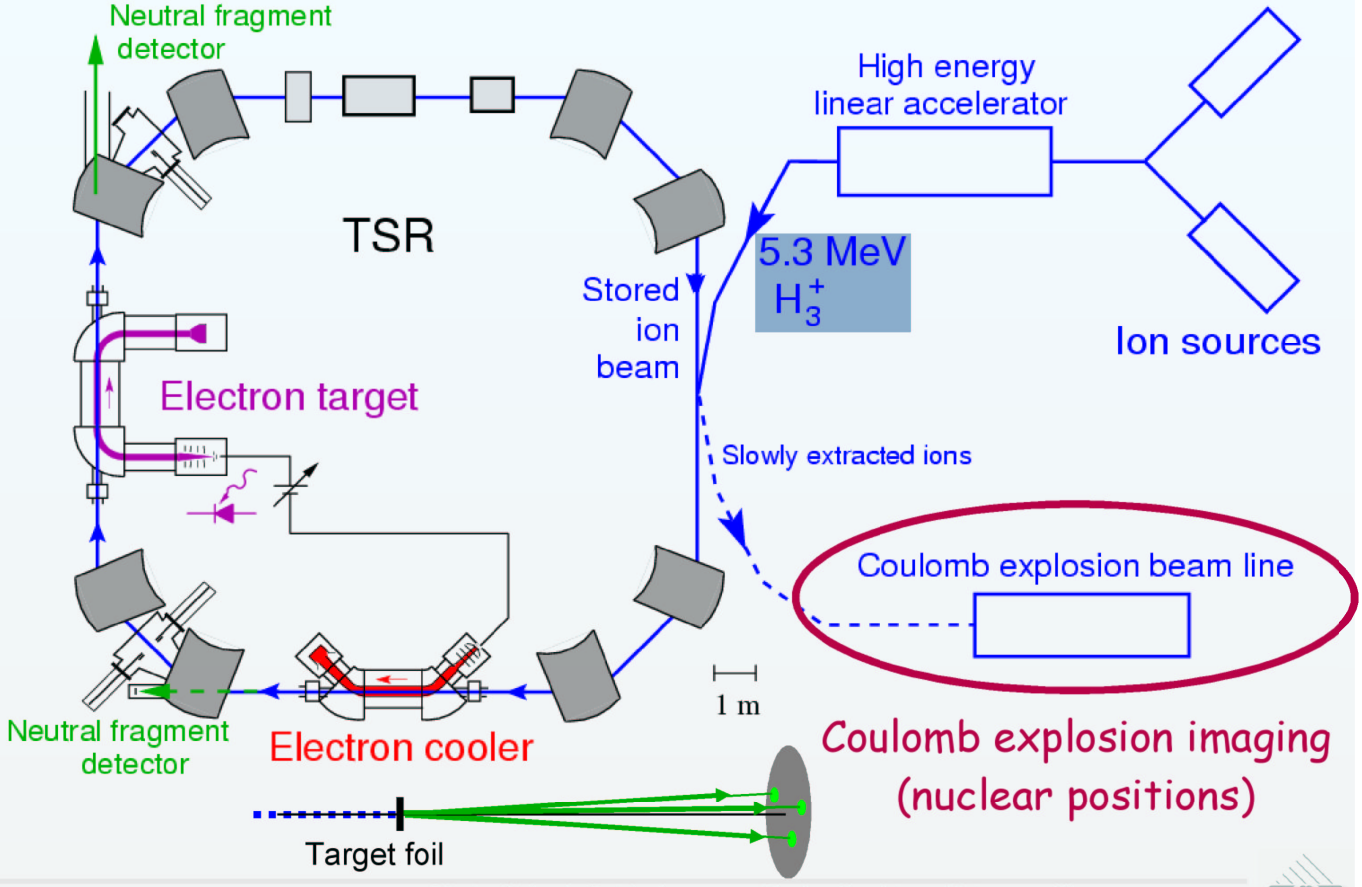
Collision measurements: $v_e \stackrel{!}{\neq} v_i$

⇒ collision energy
~1 meV up to keV

Fast stored H_3^+ ion beams

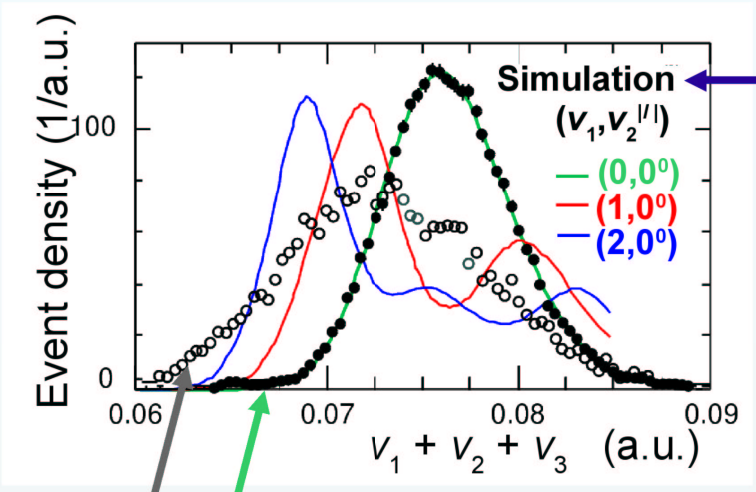


Fast stored H_3^+ ion beams



Fast stored H_3^+ ion beams

Velocity sum after Coulomb explosion of H_3^+ nuclei

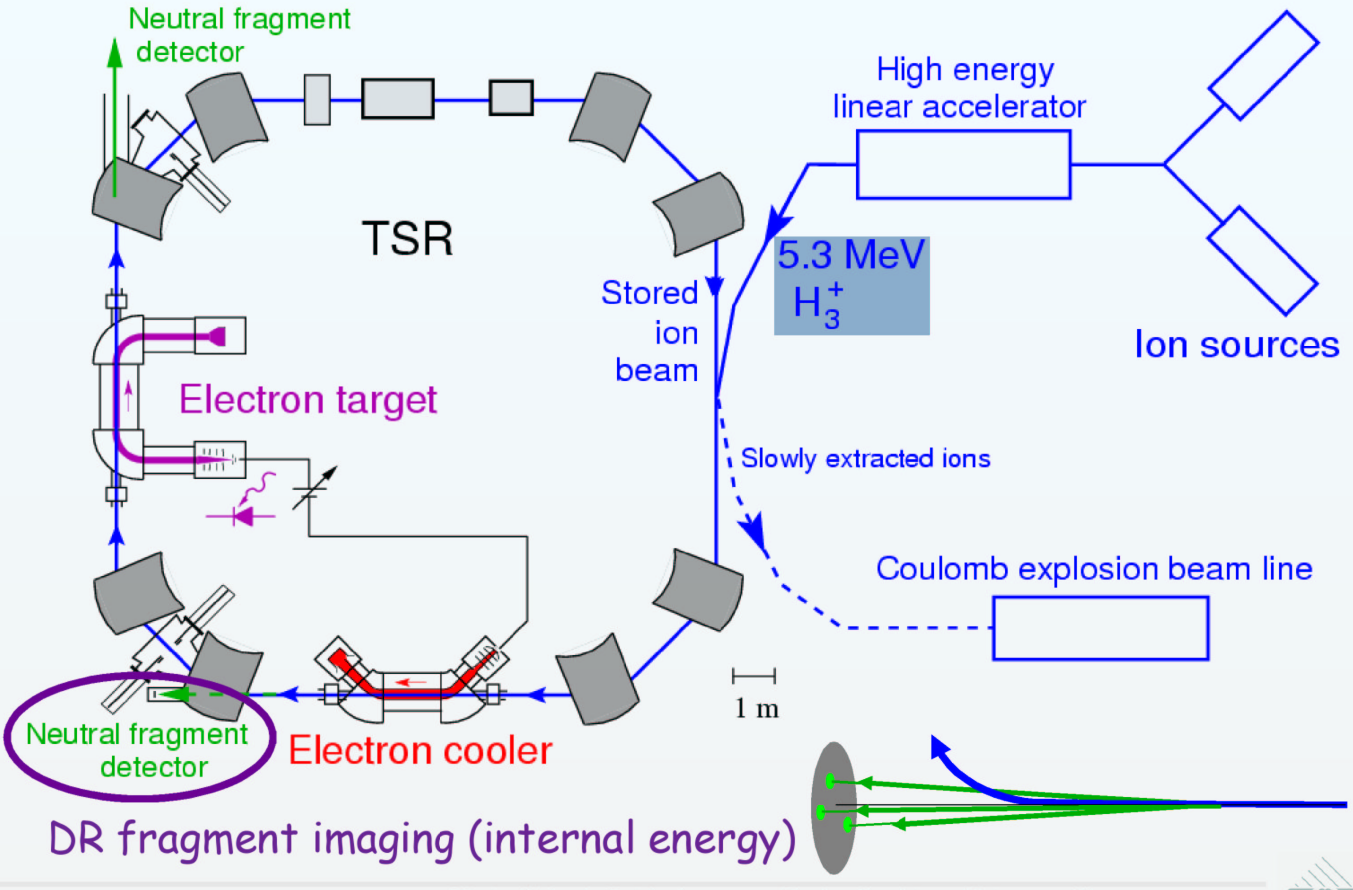


Distribution of initial nuclear coordinates from vibrational wave function

H. Kreckel
Wednesday 11:15

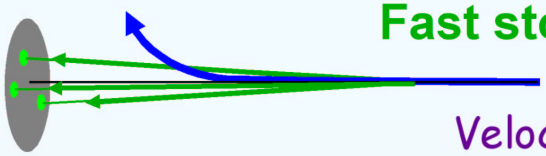
Complete relaxation of bending (and breathing) vibrations after ~ 2 seconds

Fast stored H_3^+ ion beams

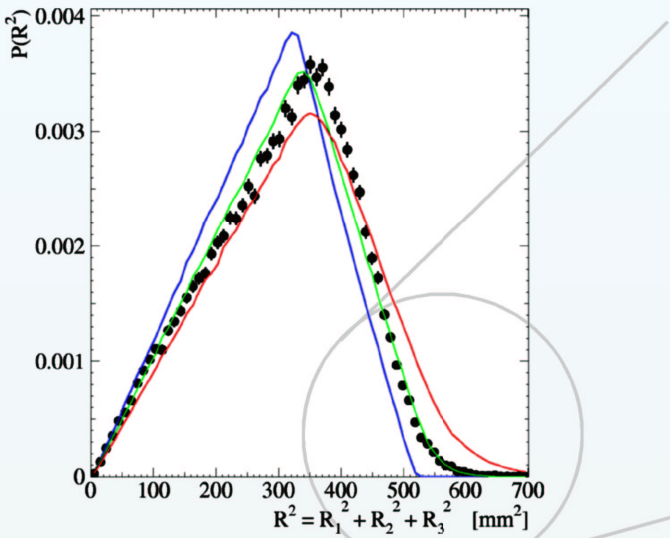


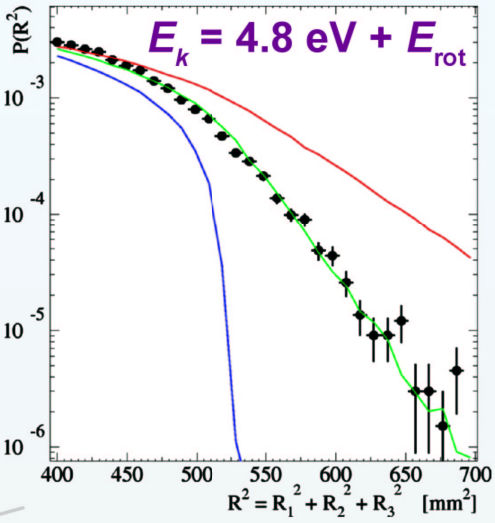
DR fragment imaging (internal energy)

Fast stored H_3^+ ion beams



Velocity sum after 3-body dissociation in DR



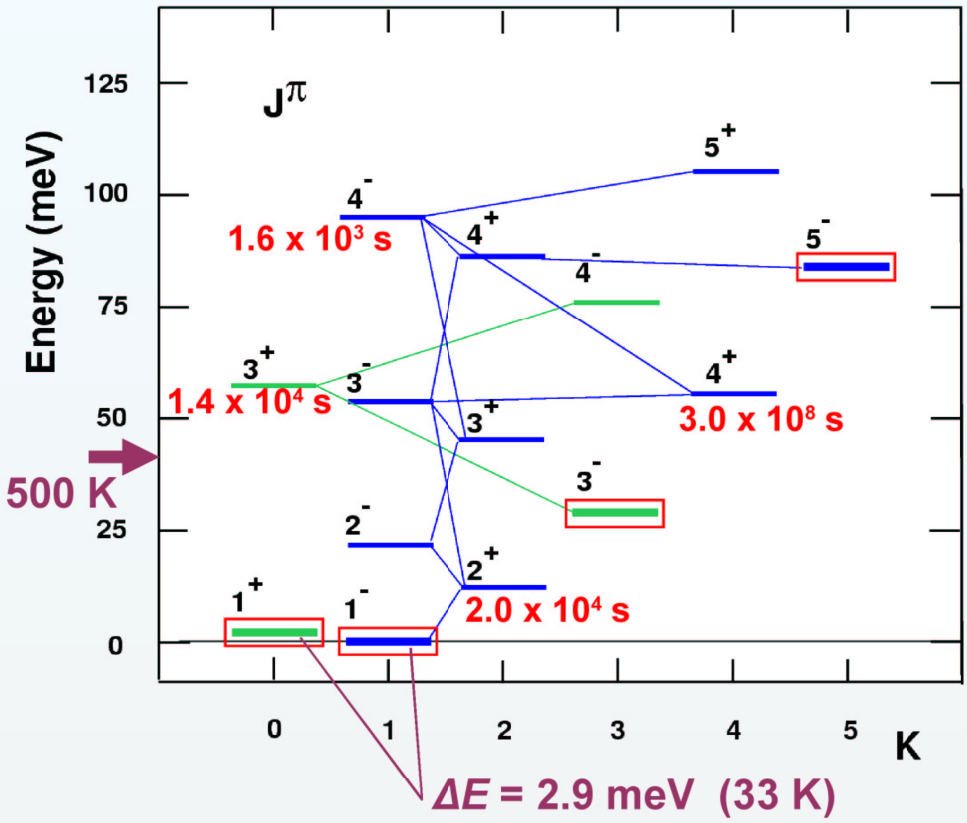


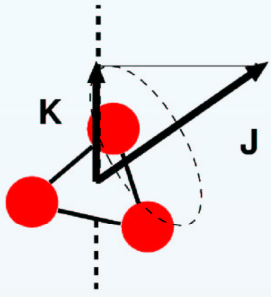
D. Zajfman
Tuesday 09:15

- Experiment 3-10 sec
- simulation (no rot.)
- simulation (T=230 meV)
- simulation (T=500 meV)

Strong quasi-stable rotational excitation (~ 2500 K)
J up to ~14

Long lived rotational levels in H_3^+



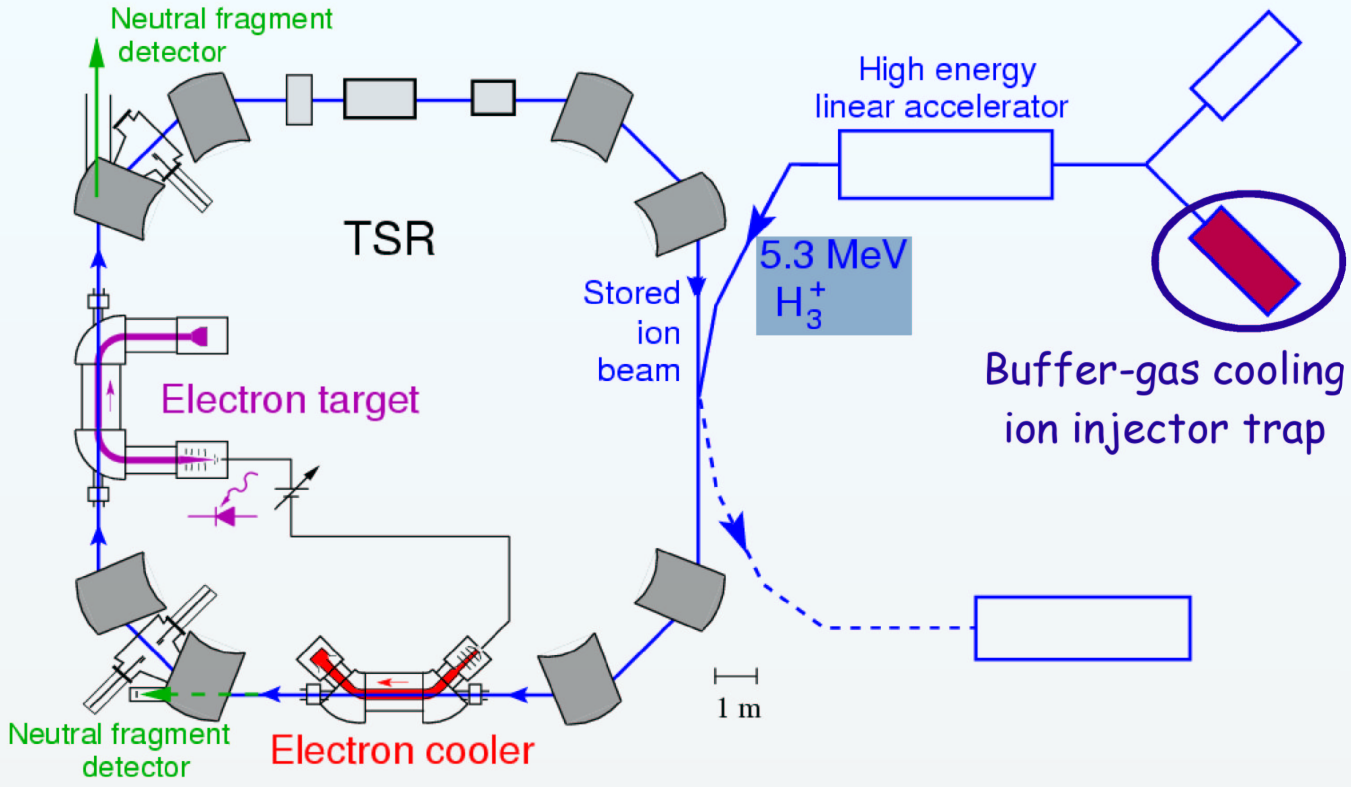


„Forbidden" E1 transitions

Radiatively stable

Ortho-H_3^+ (I = 3/2)
Para-H_3^+ (I = 1/2)

TSR cryogenic injector trap



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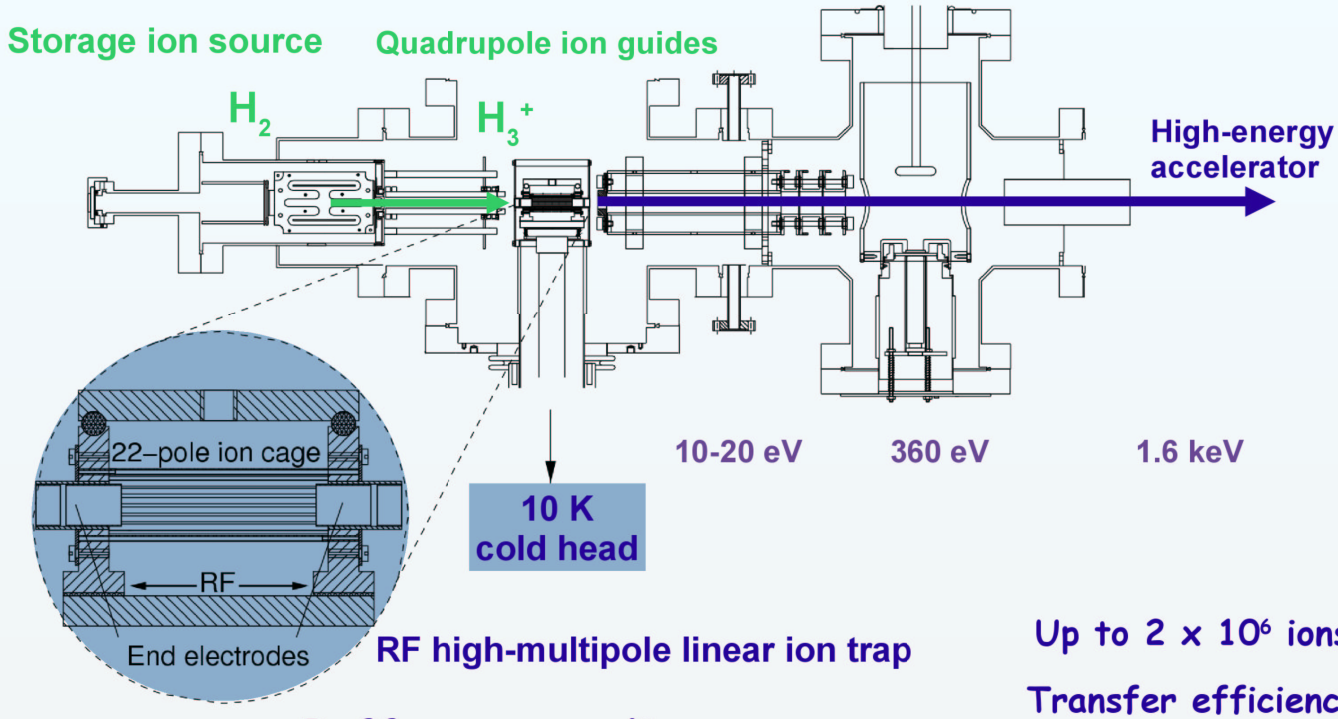


RF high-multipole linear ion trap

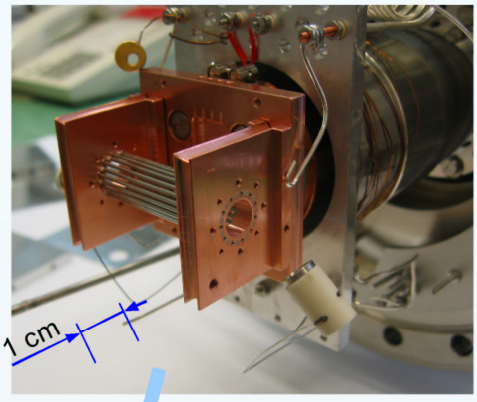
Up to 2 x 10^6 ions

Transfer efficiency 40-50%

Buffer gas cooling

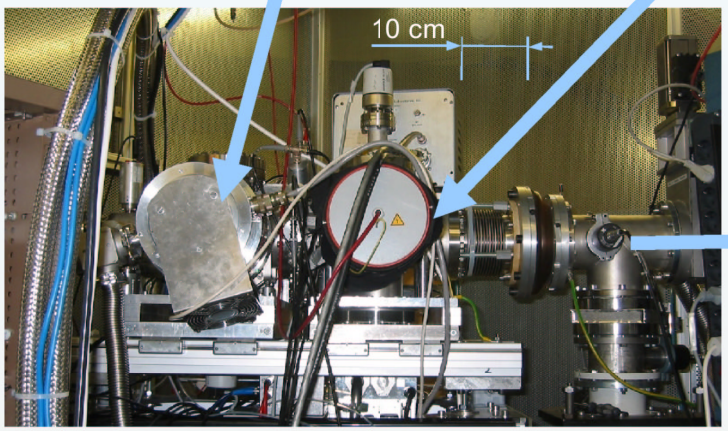
~ 10^{-2} mbar He, 1-100 ms

TSR cryogenic injector trap



Ion trap on
10 K cold head

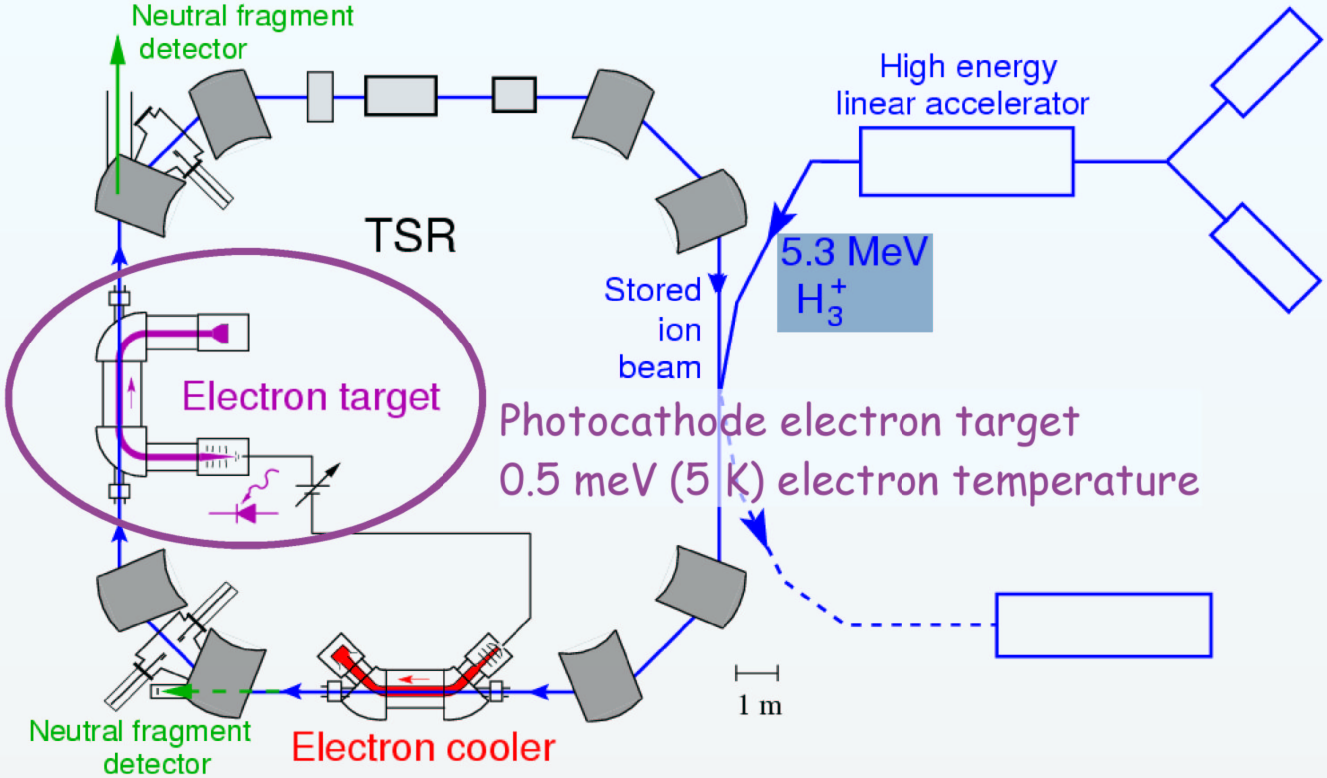
~ 5 eV quadrupole
ion guide and
mass filter



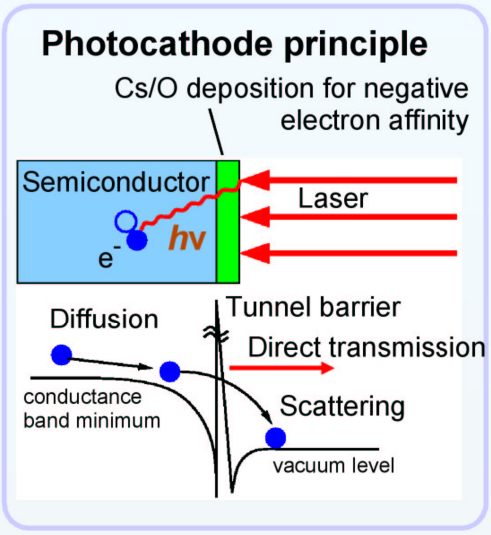
12 keV H_3^+ ions
to linear accelerator
and TSR

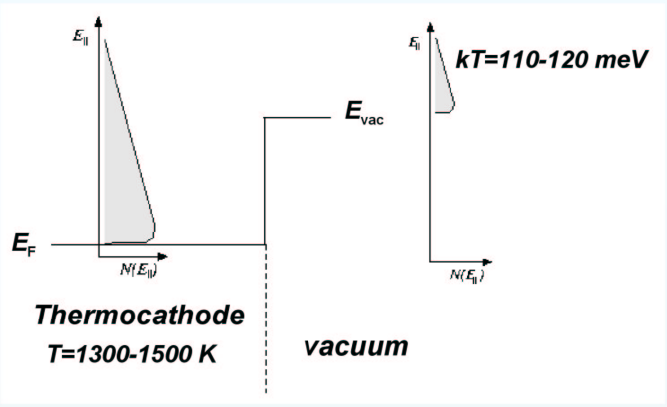
Oct. 2004: **H. Kreckel**
M. Motsch
J. Mikosch

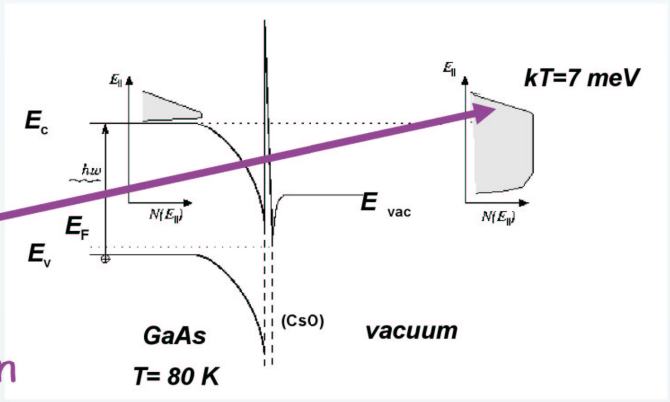
TSR photocathode electron source



TSR photocathode electron source

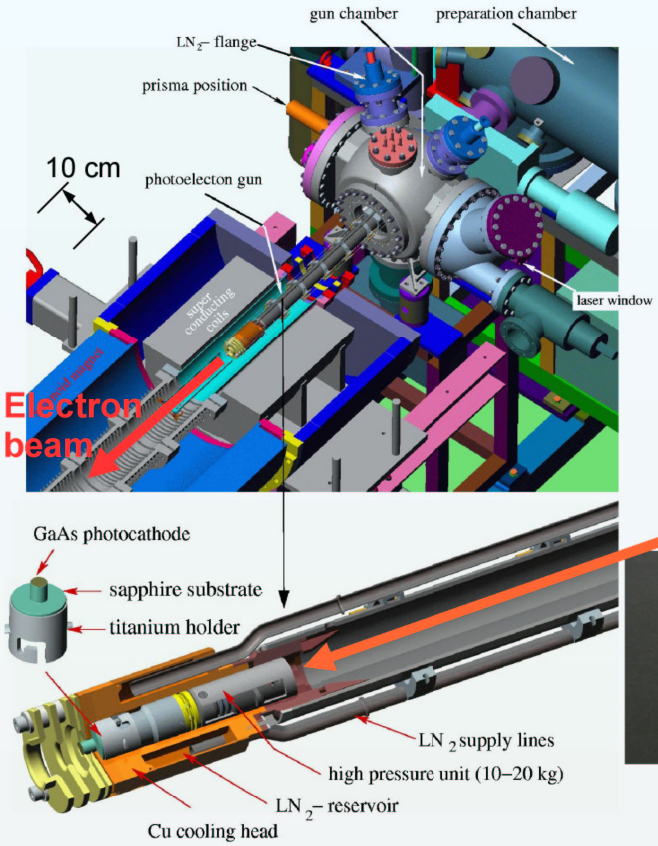






Cold electron source
0.5 meV (5 K)
after magnetic expansion

TSR photocathode electron source



Photocathode gun

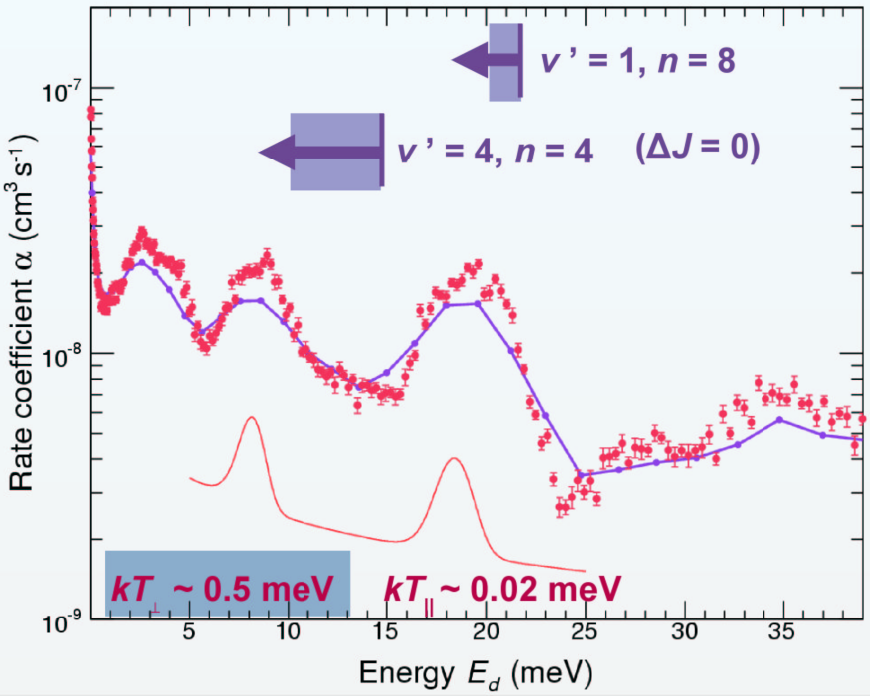
Laser illumination up to 1 W
Temperature rise 15-20 K/W at 90 K

U. Weigel et al., NIM A 536 (2005) 323

TSR photocathode electron source

Energy resolution test: DR of HD^+

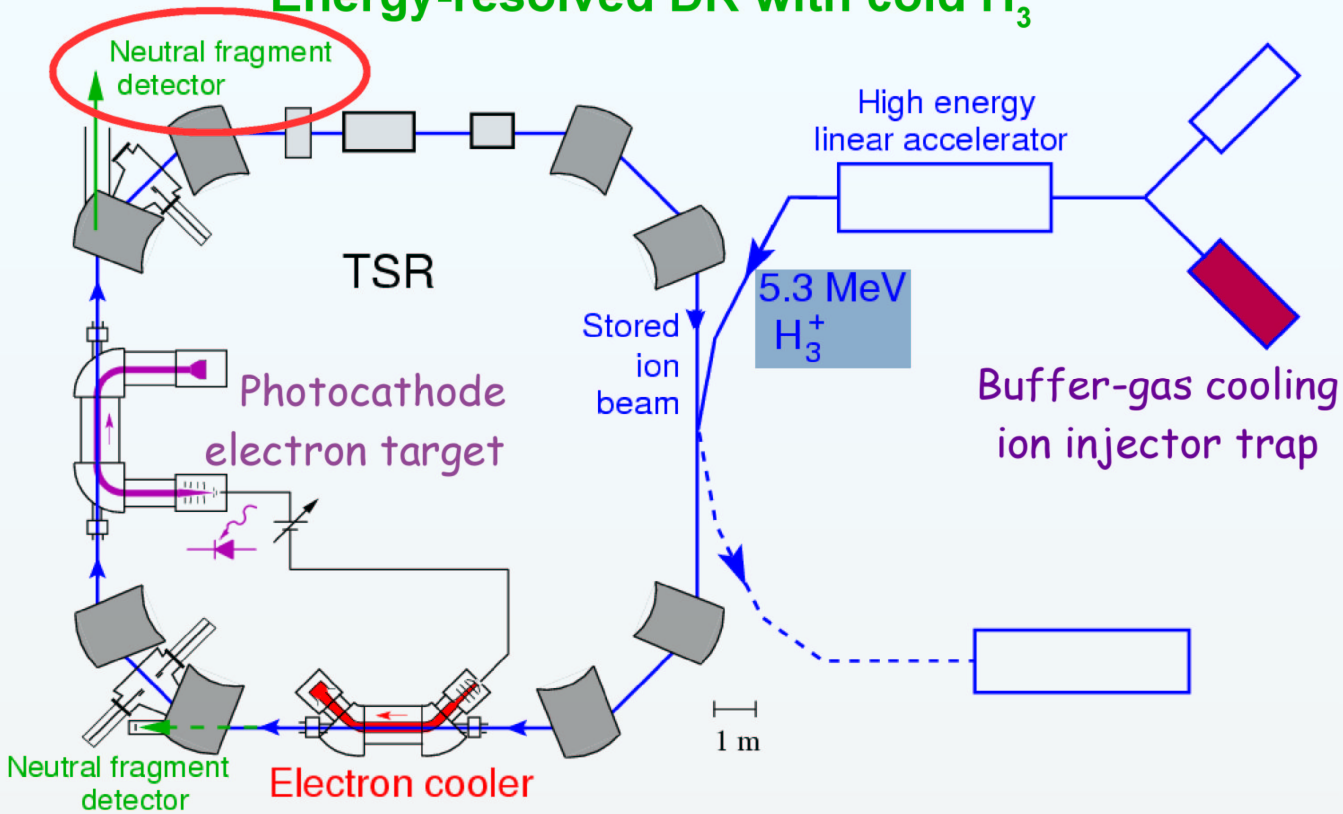
$$HD^+(1s\sigma, v = 0, J) + e \rightarrow HD^{**}(1s\sigma\, n\ell\lambda,\ v'J') \rightarrow H + D$$



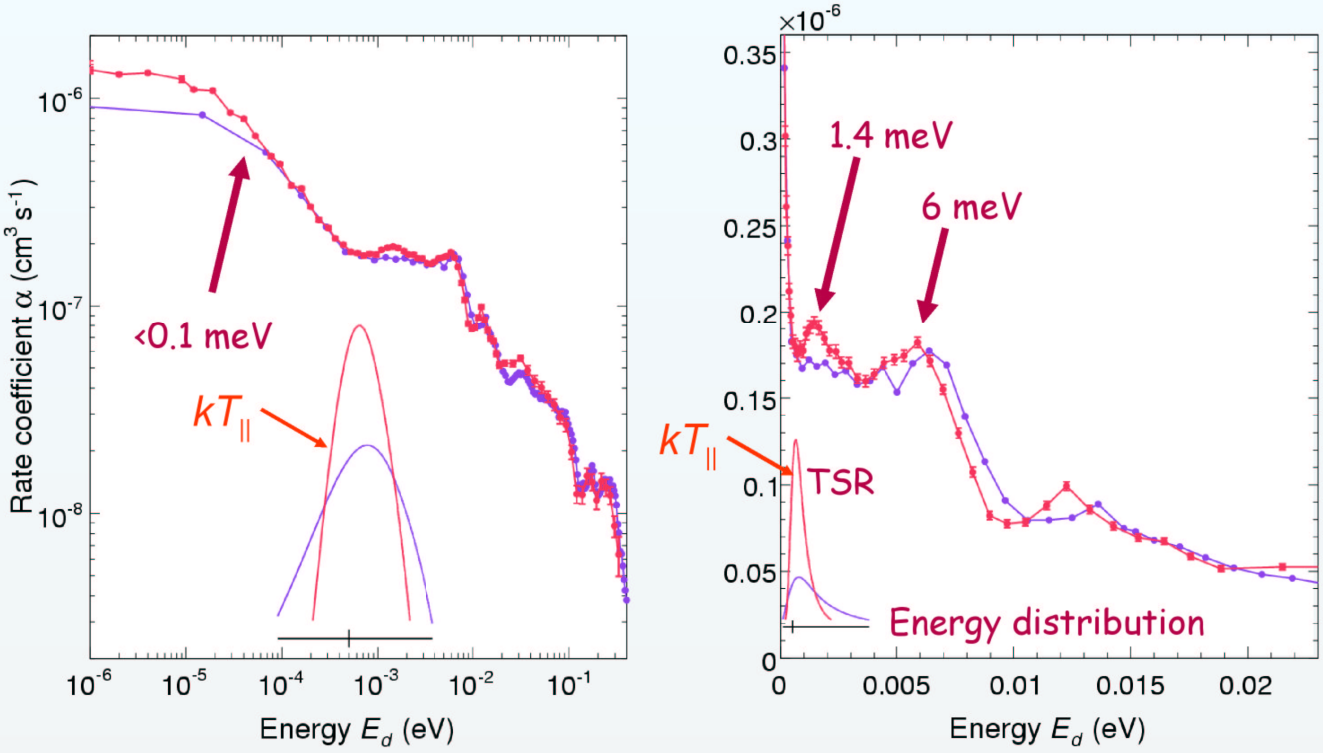
CRYRING (2001)
$kT_\perp \sim 2$ meV

TSR
E-target and photocathode (2004)

Energy-resolved DR with cold H_3^+



Energy-resolved DR with cold H_3^+



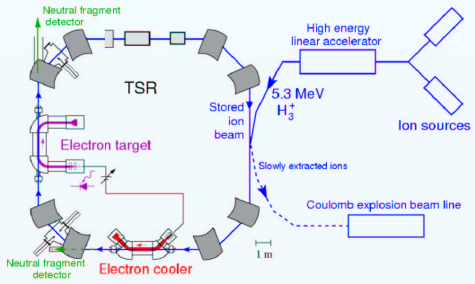
Absolute scale from 10 eV peak of CRYRING data

TSR: $kT_\perp \sim (0.5 \pm 0.15)$ meV

CRYRING: $kT_\perp = (2 \pm 0.5)$ meV

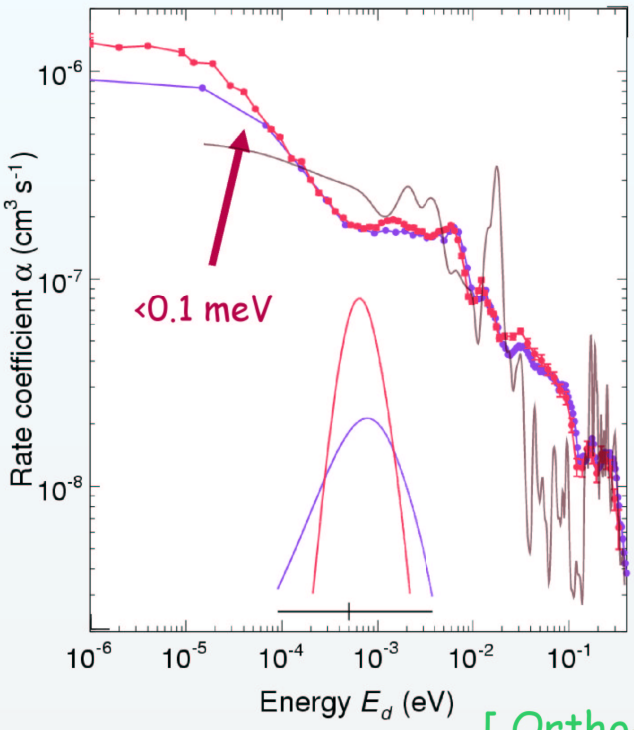
Energy-resolved DR with cold H_3^+

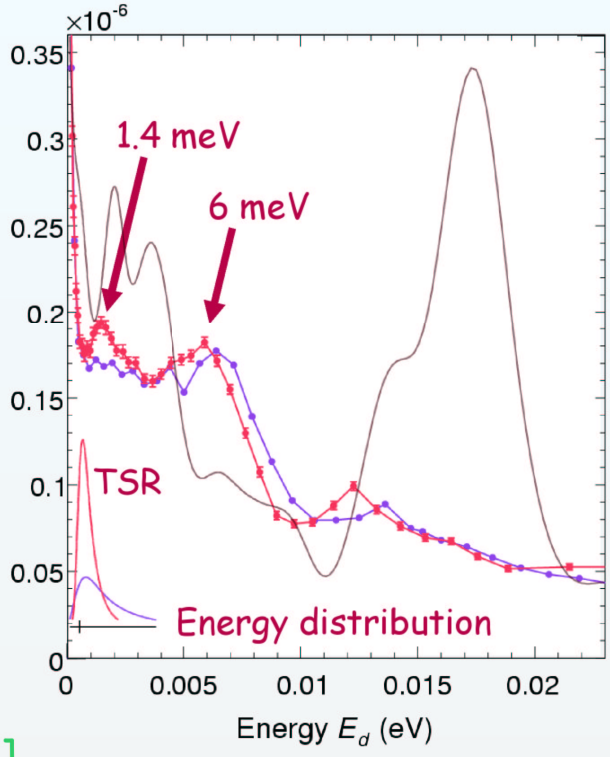
Experimental parameters



	CRYRING	TSR
Cold H_3^+ source	Expanding jet discharge	RF buffer gas cooling trap
Acceleration	12.1 MeV in the ring, ~1 s	5.3 MeV before injection, 1 μs
Electron density	6 x 10^6 cm^{-3}	4 x 10^5 cm^{-3}
Magnetic field (interaction region, nanosecond time dependence)	0.03 T	0.075 T

Energy-resolved DR with cold H_3^+



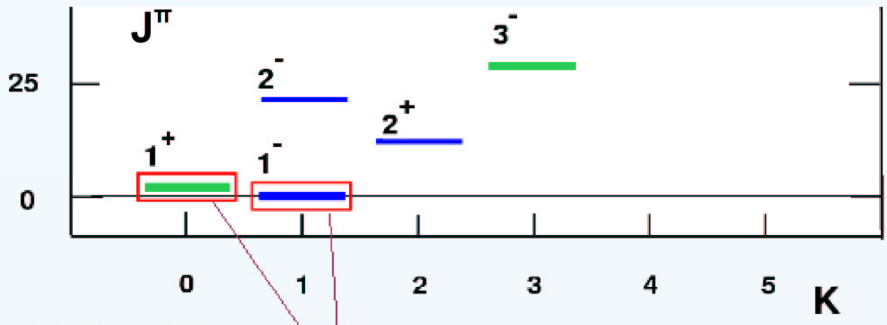


Theory: Kokooline & Greene (2003)

$$\frac{[\ \text{Ortho-}H_3^+\ ]}{[\ \text{Para-}H_3^+\ ]} = 1$$

Kreckel et al. (2005)

Spin symmetry variations: exploratory study



Ortho-H_3^+ (I = 3/2)

Para-H_3^+ (I = 1/2)

ΔE = 2.9 meV (33 K)

CRYRING and TSR high-resolution measurements:

$$\frac{[\ \text{Ortho-}H_3^+\ ]}{[\ \text{Para-}H_3^+\ ]} \sim (0.7 \ldots 1)$$

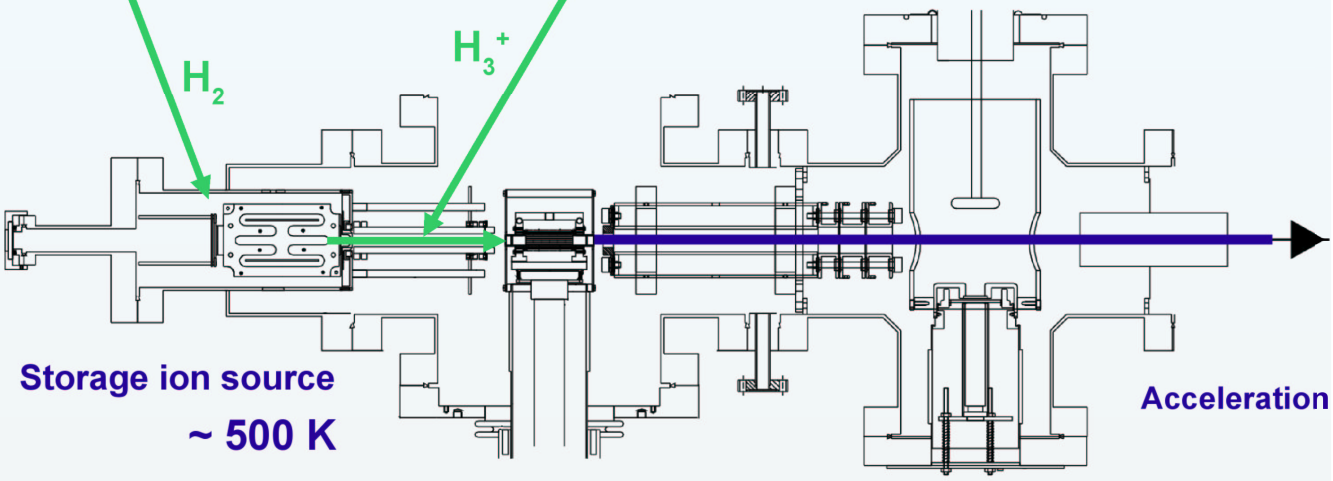
TSR: vary ortho/para ratio alone (single setup)

Spin symmetry variations: exploratory study

Para-H_2
Normal H_2

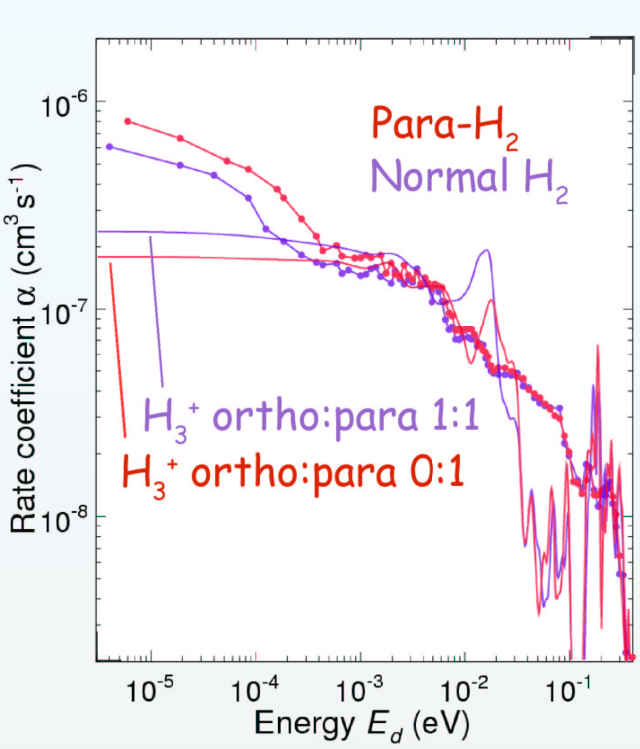
$$\frac{[\text{Ortho-}H_3^+]}{[\text{Para-}H_3^+]} = \begin{cases} \sim 0.1 \ldots 0.8 \\ 1 \end{cases}$$

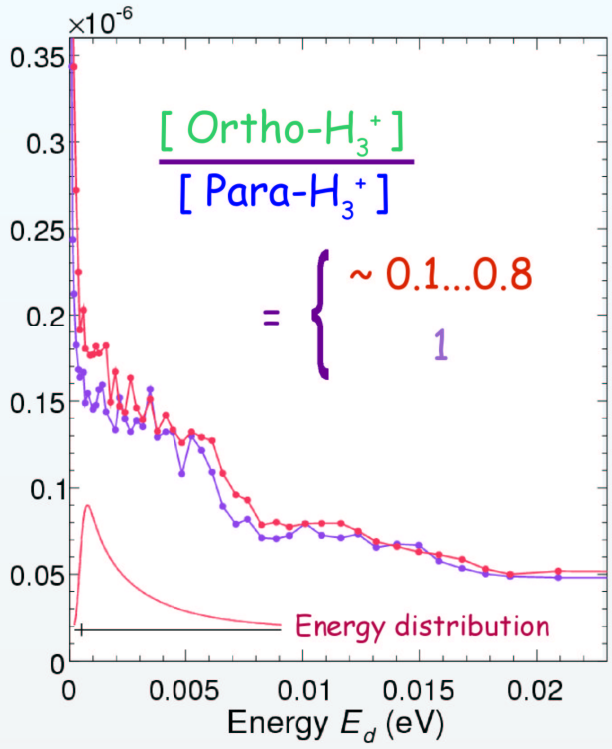
Cordonnier et al. (2000)



H_3^+ nuclear spin symmetry determined by ion source

Spin symmetry variations: exploratory study



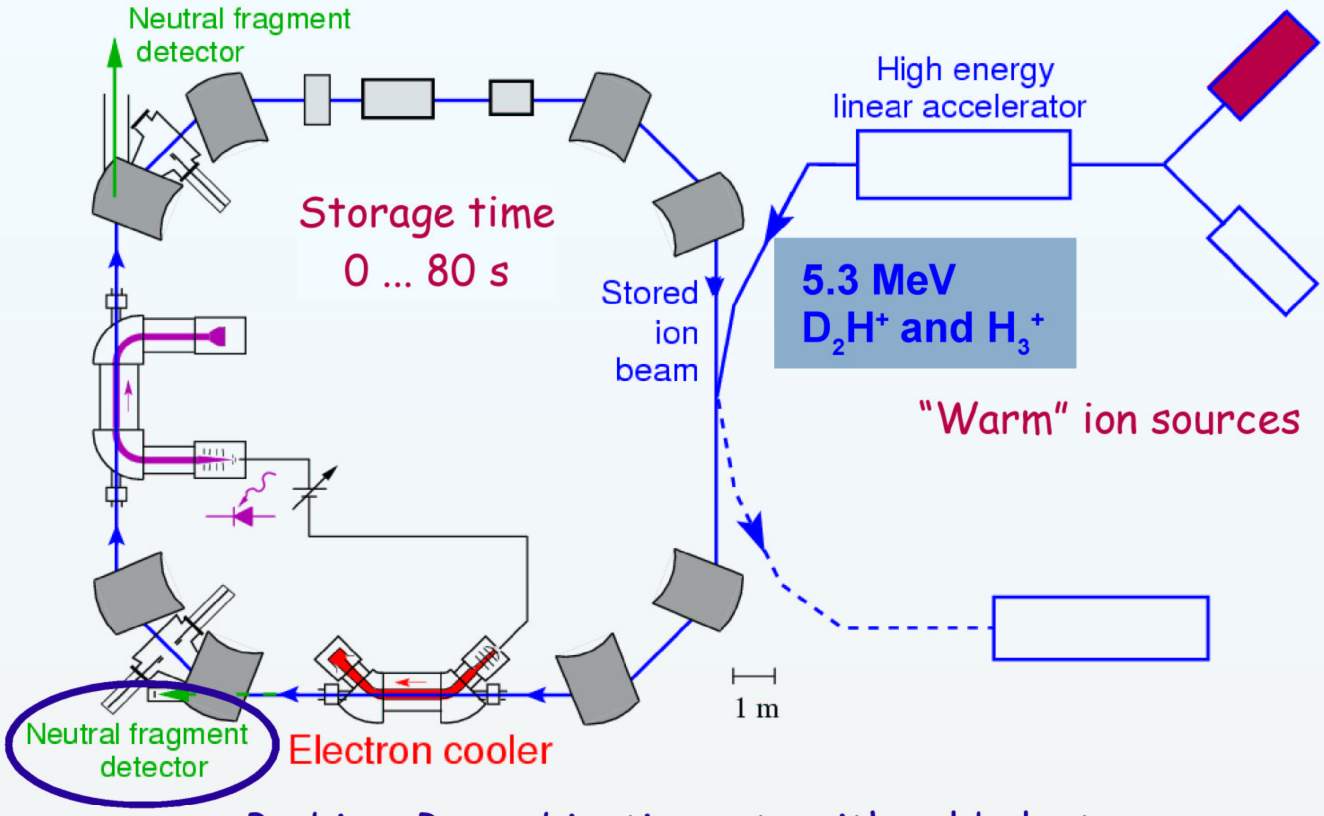


Theory: Kokooline & Greene (2003)

TSR: $kT_\perp \sim 4$ meV

Kreckel et al. (2005)

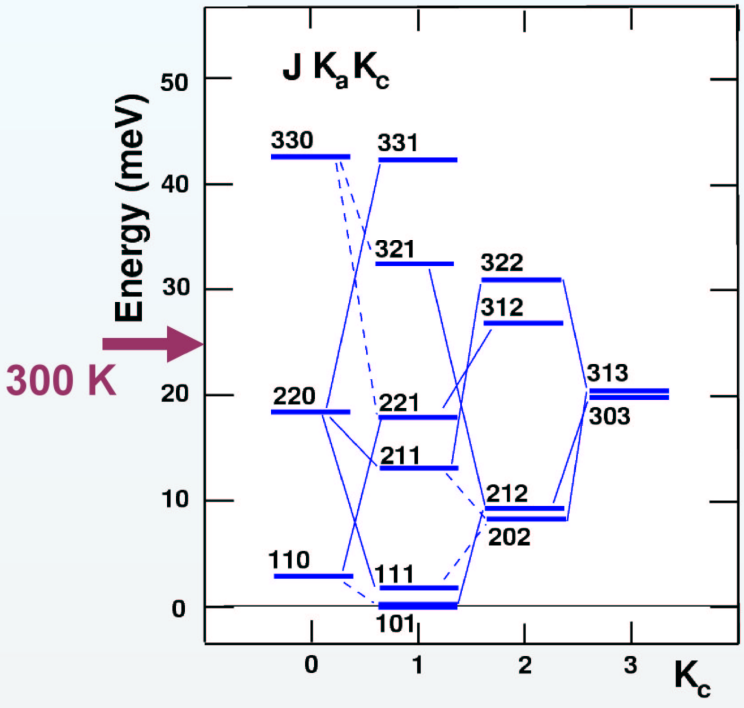
Probing of low-energy rate coefficient



Probing: Recombination rate with cold electrons

Probing of low-energy rate coefficient

D_2H^+ rotational levels

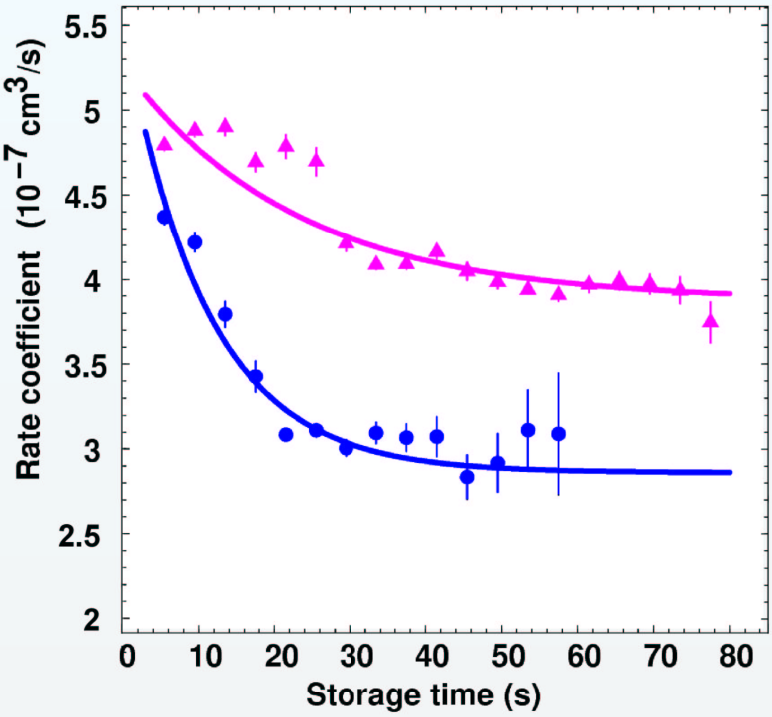


Radiative lifetimes
~ 10...50 s

Miller, Tennyson & Sutcliffe 1989

Probing of low-energy rate coefficient

D_2H^+ interaction with low-temperature (140 K) electrons



Time-averaged interaction strength

0.2 R_0

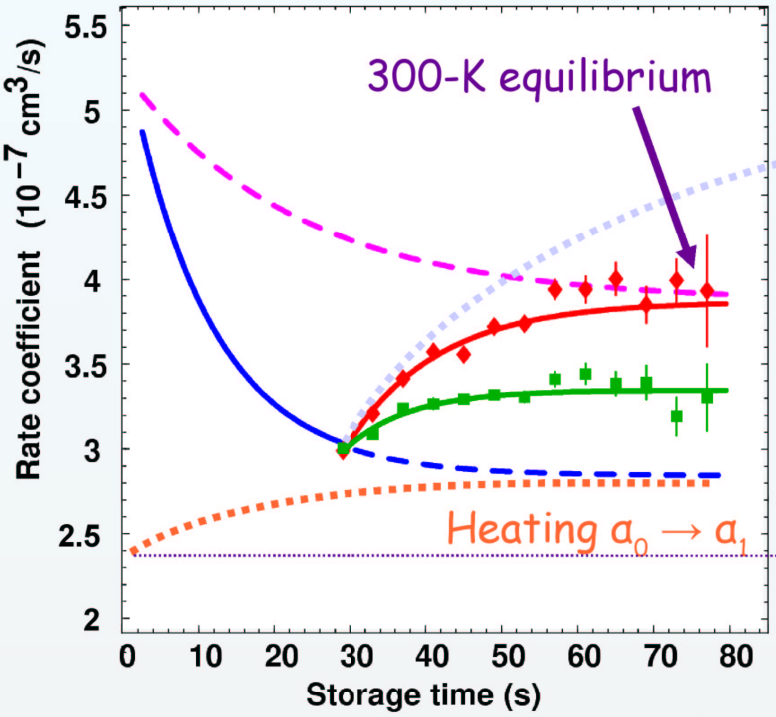
R_0

20%

Lammich et al. (2003)

Probing of low-energy rate coefficient

D_2H^+ interaction with low-temperature (140 K) electrons



$$\alpha_1 = C \frac{R_{heat}}{R_{cool}}$$

$\alpha_0 + 5\alpha_1$

0.2 R_0

0.5 R_0 $\alpha_0 + 2\alpha_1$

R_0 $\alpha_0 + \alpha_1$

α_0

20%

$\alpha_1 / \alpha_0 = 0.2$

Residual gas excitation

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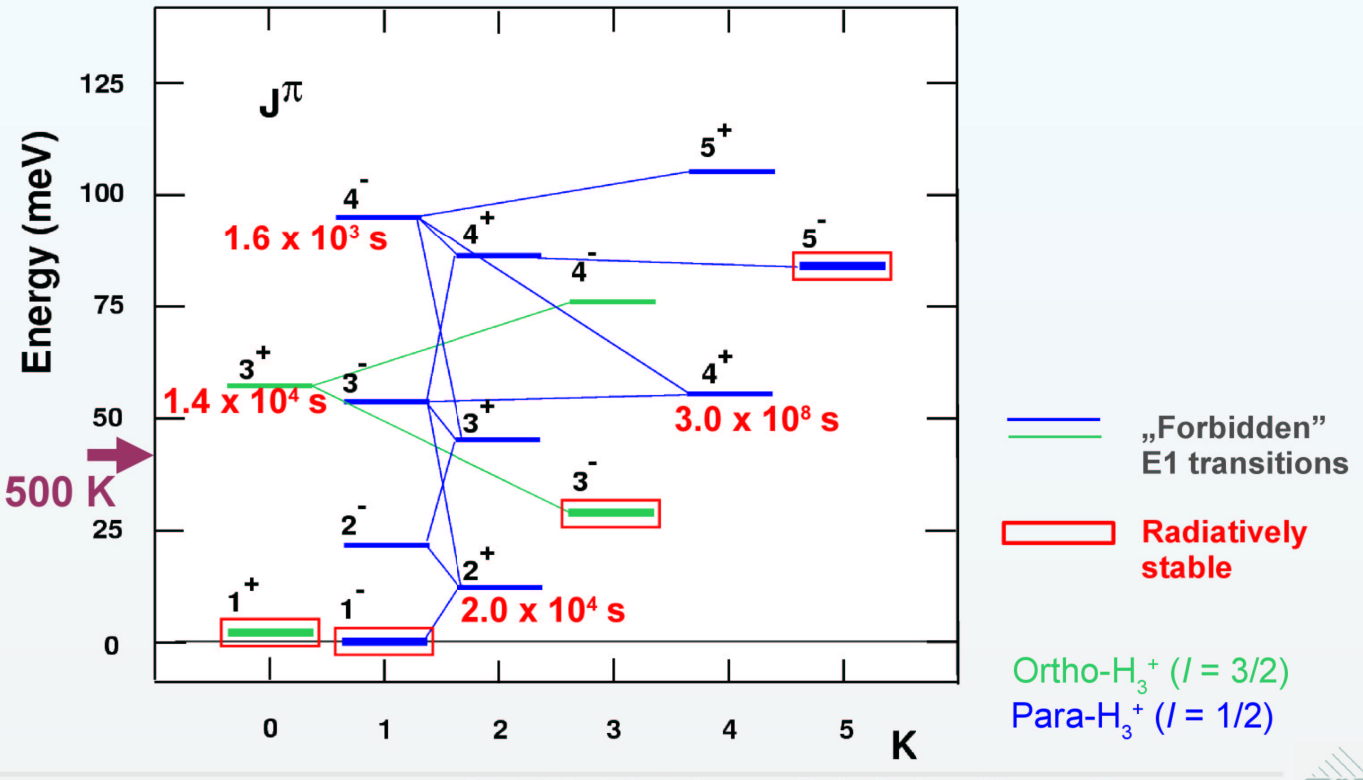
Physics, Chemistry and Astronomy of H_3^+ London, 16-19 January 2006
Andreas Wolf

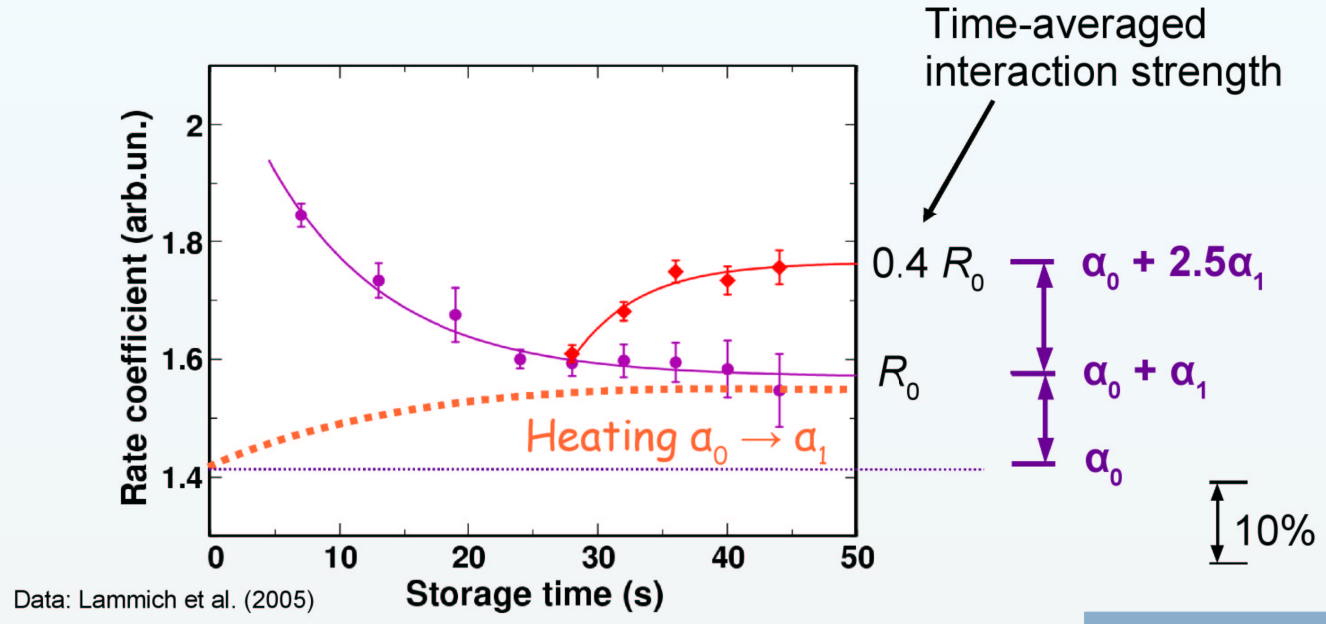
Probing of low-energy rate coefficient

H_3^+ rotational levels



Probing of low-energy rate coefficient

H_3^+ interaction with low-temperature electrons

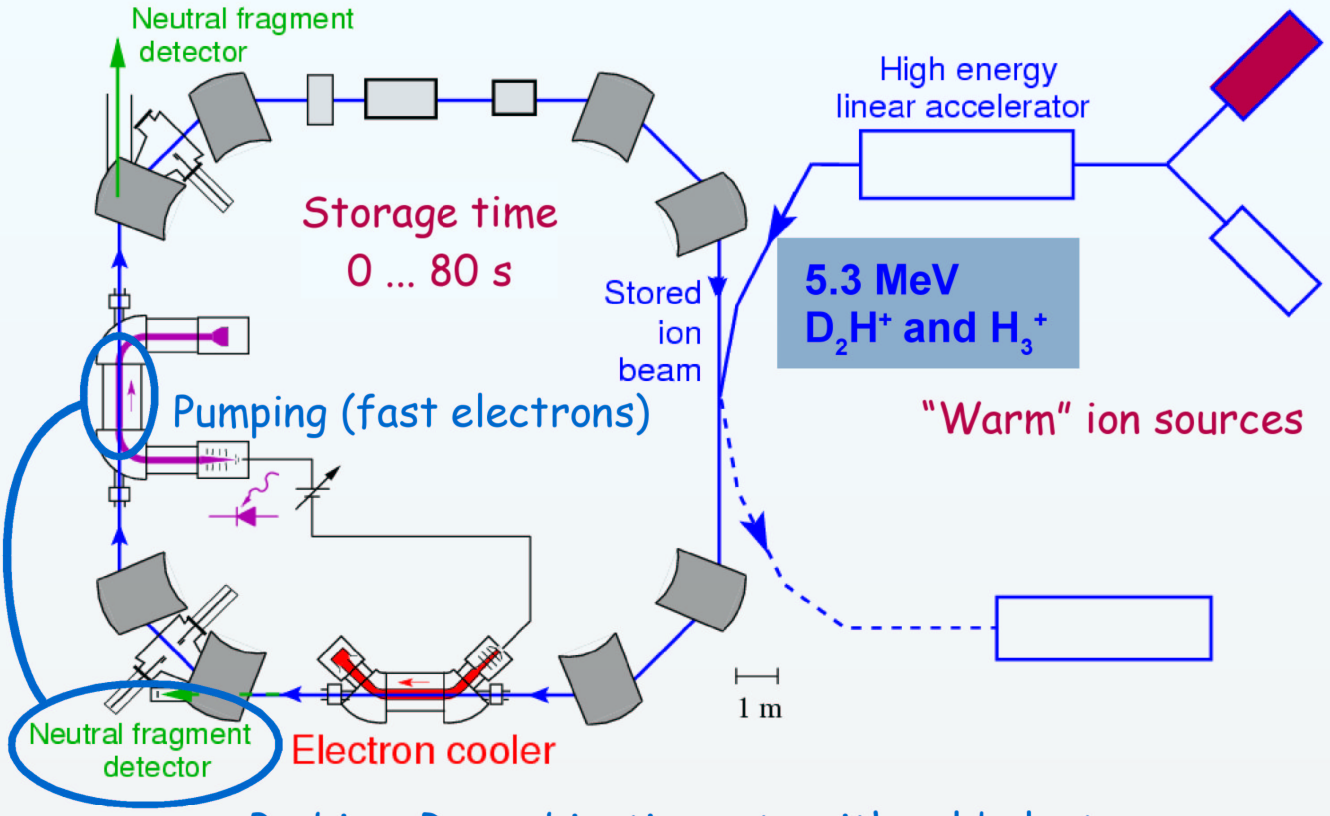


Data: Lammich et al. (2005)

~ 10% rotational excitation by residual gas

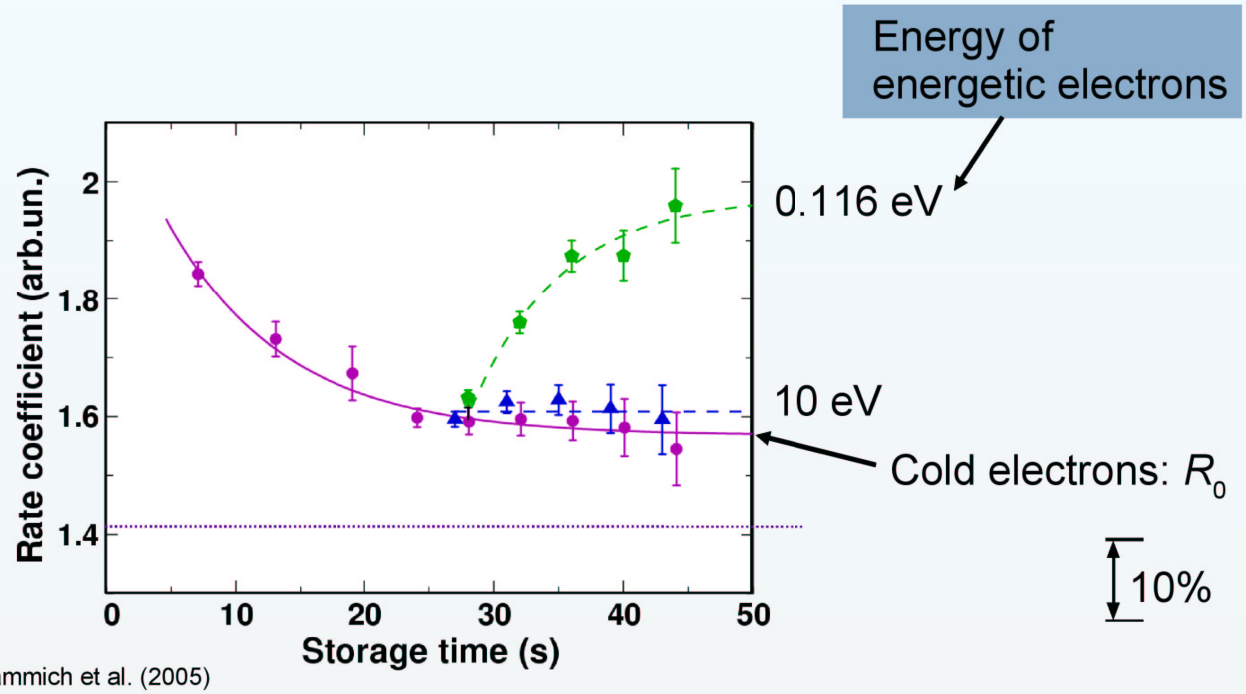
$\alpha_1 / \alpha_0 = 0.08$

Pump–probe measurement of electron impact excitation



Probing: Recombination rate with cold electrons

Pump–probe measurement of electron impact excitation

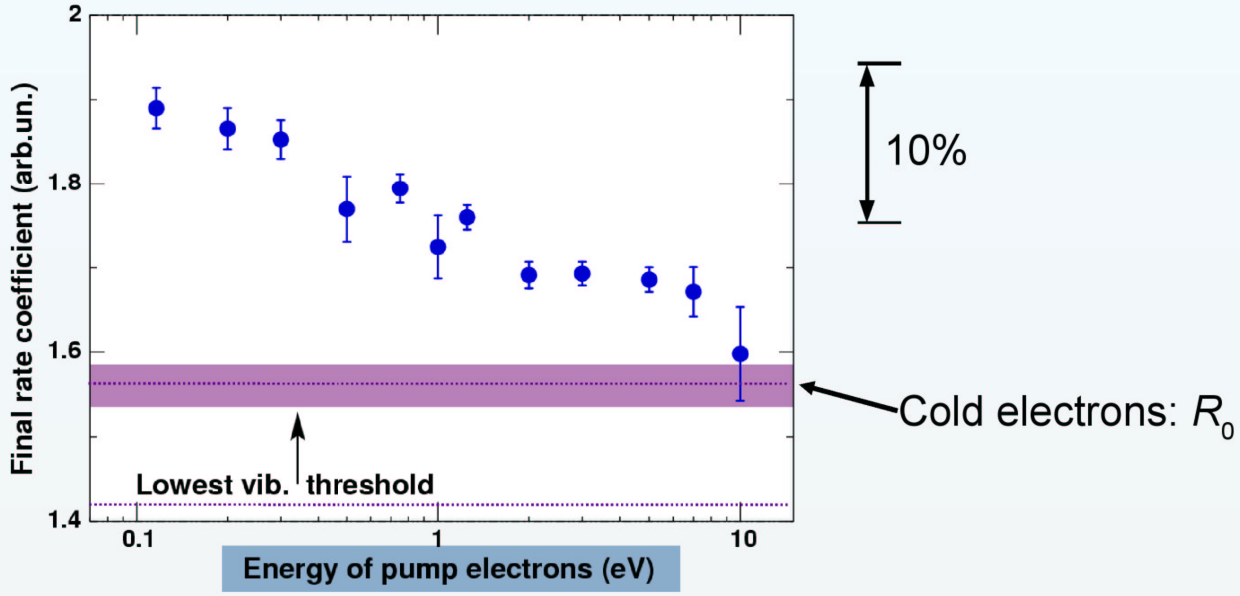


Data: Lammich et al. (2005)

~ 2% rotational excitation by 10-eV electrons (normalisation measurement)

Pump–probe measurement of electron impact excitation

Rotational impact excitation curve by energetic electrons



Summary and outlook

Excitation in stored, fast H_3^+ ion beams

Probing methods

- Vibrations: Coulomb explosion imaging
- High rotations: Molecular fragment imaging
- Low rotations: Long-time (~60 s) variation of low-temperature rate coefficient

H_3^+ rotational cooling by cold electron interaction (140 K)

Differential depletion by DR, or superelastic collisions

H_3^+ rotational excitation by residual gas: ~10%

by 10 eV electrons: ~2%

Electron impact rotational excitation rate coefficient ~ $E^{-1/2}$ at 0.1...10 eV

Summary and outlook

Merged beams experiments with cold electrons

Control of internal excitations and electron collision energies (few-Kelvin level)
- Cryogenic photocathode electron beam
- Cryogenic RF trap coupled to accelerator/storage ring

Low-energy rate coefficient
- High-resolution low-energy structure of DR rate coefficient
- Variation of low-energy DR rate coefficient with nuclear spin symmetry

Rotational resonance structure
- Low energy resonances at <0.1 meV, 1.4 meV, 6 meV
- Much less resonant structure than predicted at >10 meV

Summary and outlook

Outlook

Spectroscopic determination of H_3^+ ortho:para ratio in ion injector trap

Sensitive in-situ diagnostic for rotational level populations in stored H_3^+ beam
- Ortho:para H_3^+ contributions to DR
- Electron impact excitation and cooling

High-resolution low-energy DR measurement for D_3^+

Rotational relaxation by radiation, cryogenic temperatures
- Low-energy cryogenic storage ring with cold electron beam (under construction)
- High-resolution low-energy resonances for isotopologues H_2D^+, D_2H^+ , cold