Ion - neutral reactions

Dieter Gerlich

Introduction
Instruments for ion chemistry

Typical applications
Selected systems: old or up to date?
Negative ions
Spectroscopy

Carbon reactions
Formation of $C_3H_n^+$

Deuteration of H_3^+
Non statistical behavior

Reactions with H atoms
Beam-trap combination

Outlook
Tucson, sub-K cooling, nano-particles

Ion chemistry: instruments

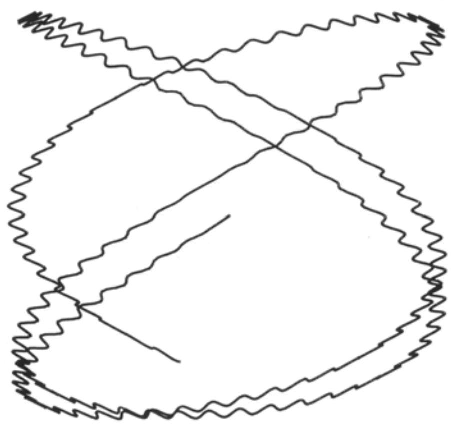
Environment
Astrochemistry
....

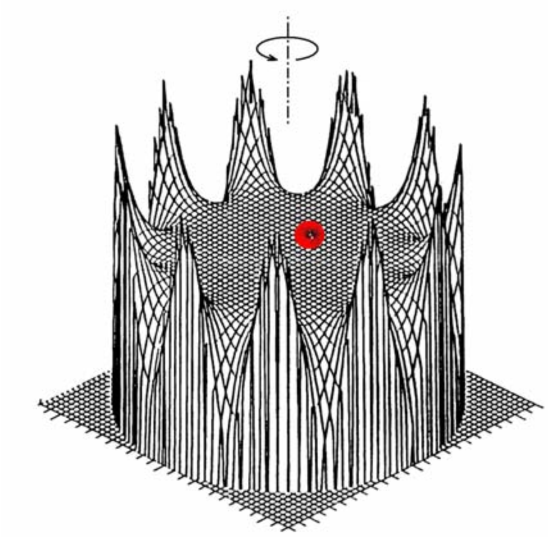
Reaction dynamics
Structure & function
Elemental analysis
....

Ion and neutral beams
Mass spectrometry
SIFT*, DRIFT
Traps (ICR, Paul)
Multi-electrode rf traps

....

INHOMOGENEOUS RF FIELDS: A VERSATILE TOOL FOR THE STUDY OF PROCESSES WITH SLOW IONS**





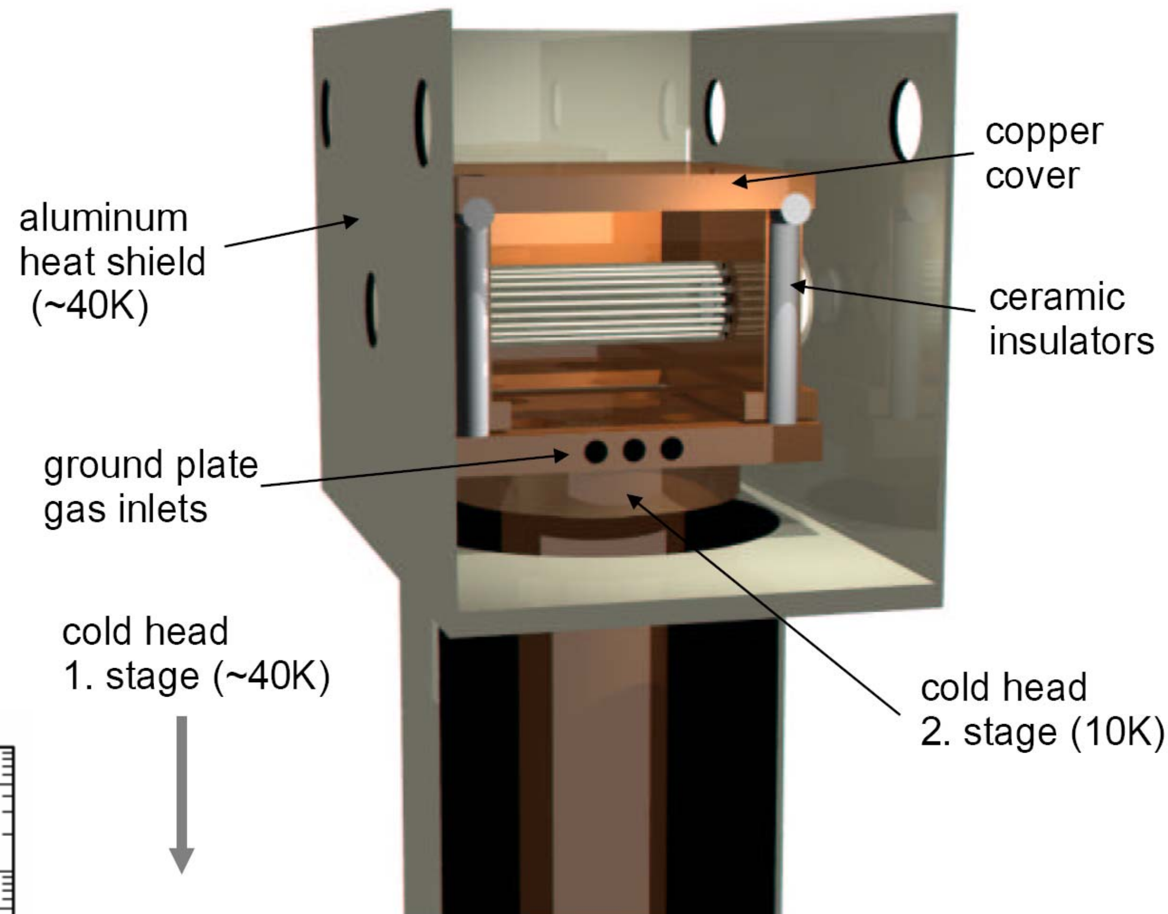
*Canterbury SIFT (also known as 'the Southern Hemisphere SIFT")
S. Petrie· D. K. Bohme, *Mass Spectrometric Approaches to Interstellar Chemistry*, Top Curr Chem (2003) 225: 37–75
**D. Gerlich, Adv. in Chem. Phys. 82 (1992) 1

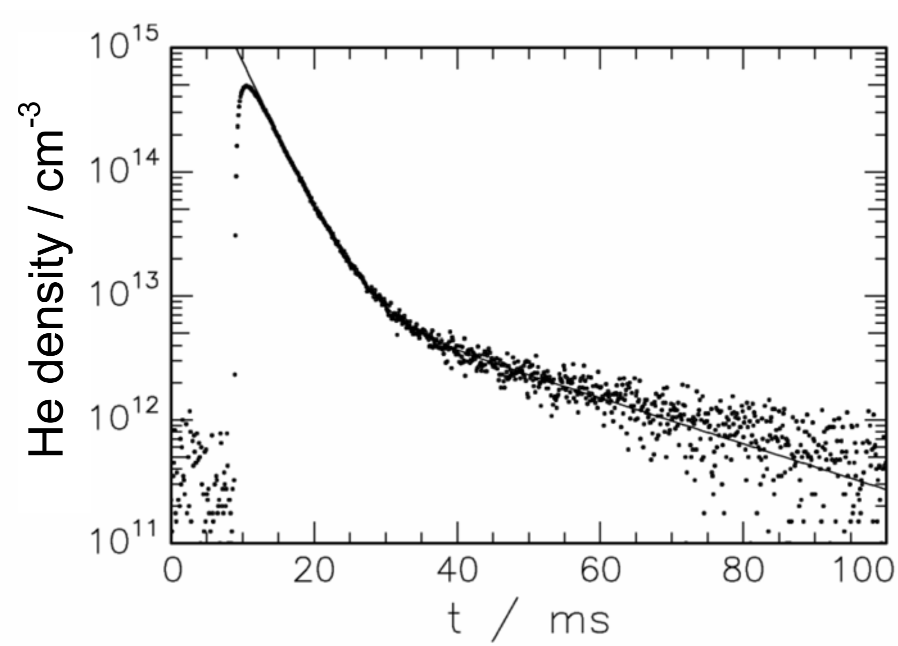
Buffer gas cooling in an rf trap

Dynamic traps such as
Penning, storage rings, cone trap
do not work

Paul trap does not work
η = const

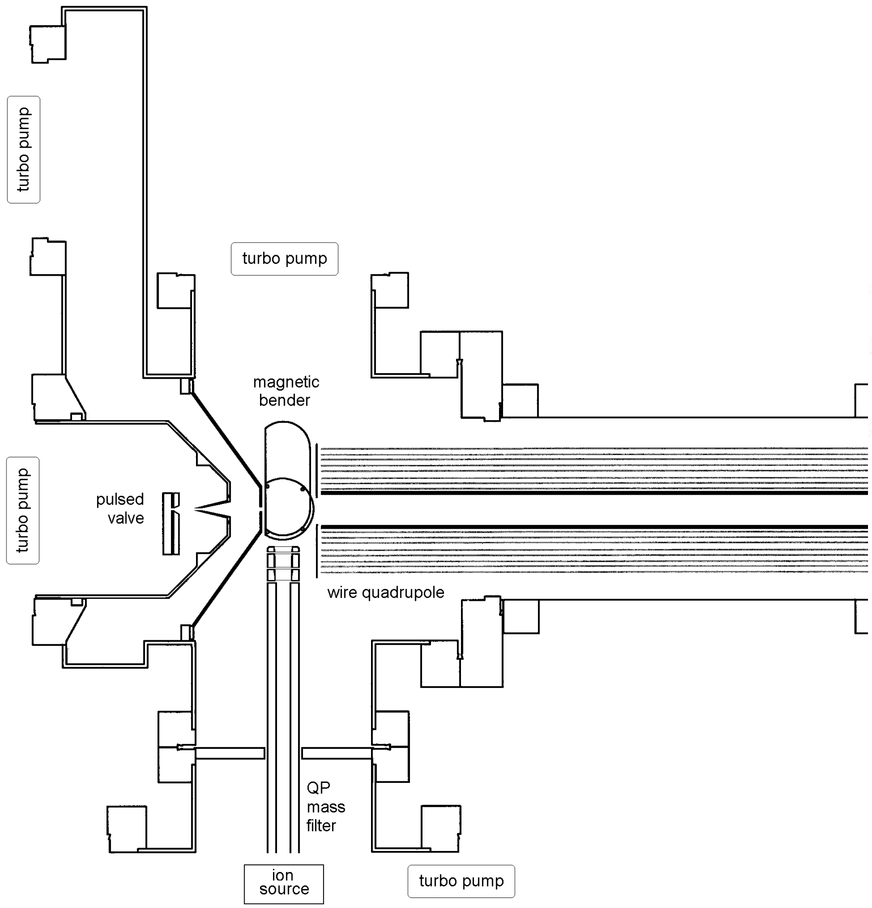
Only way to cool efficiently
internal degrees of freedom
are
rf multielectrode traps

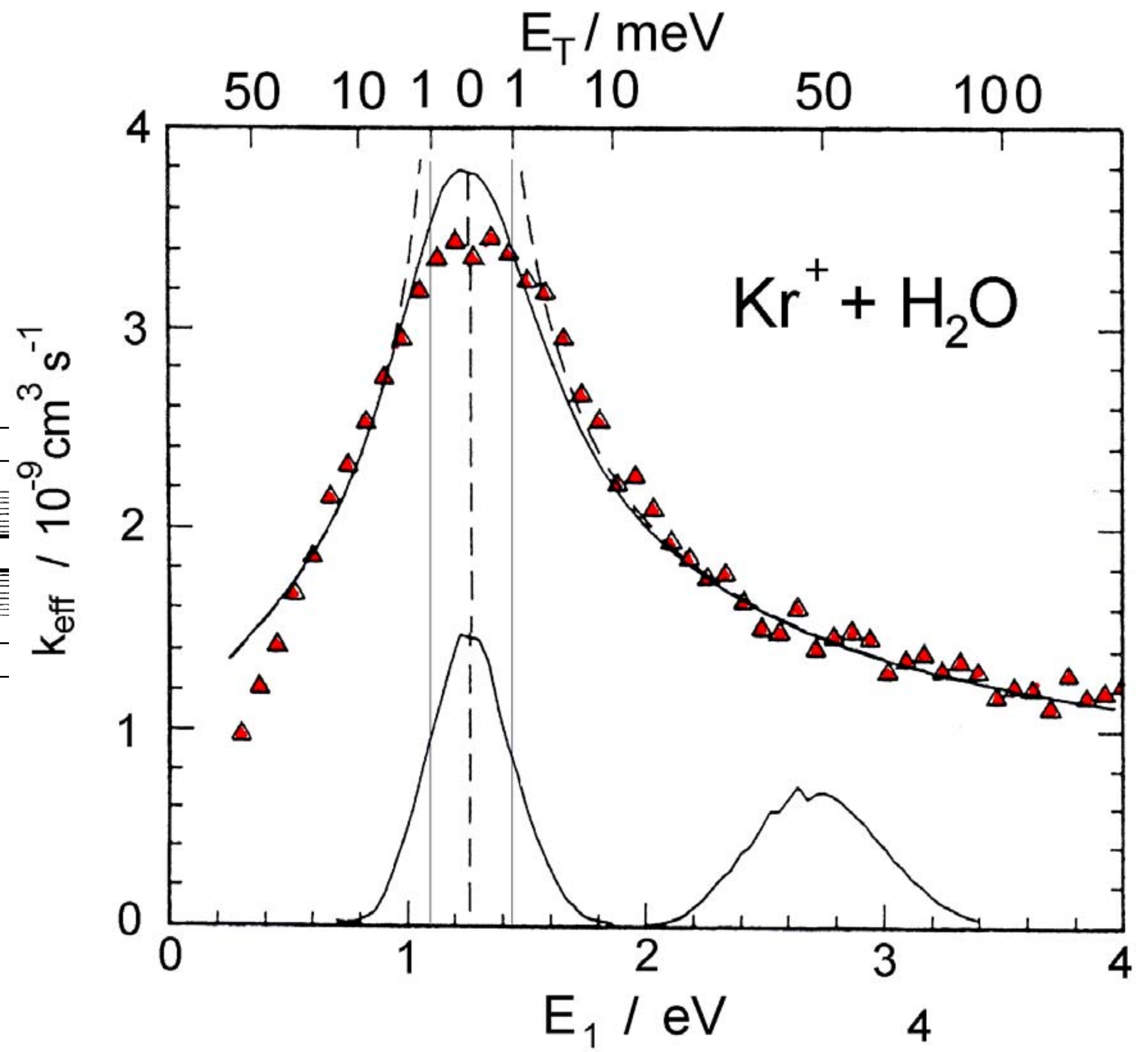




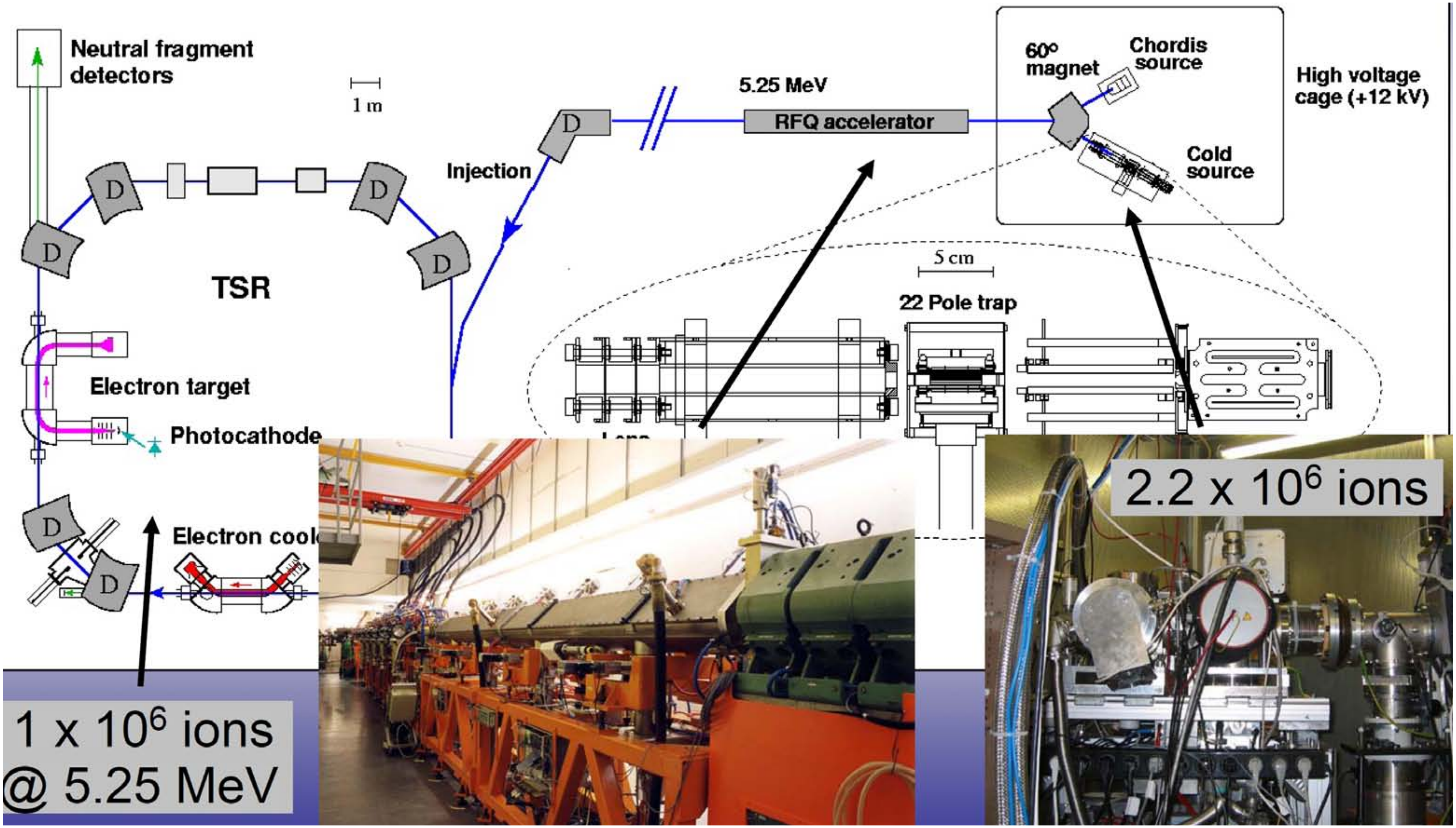
sub K:
cold pulsed effusive beam

Merged beams





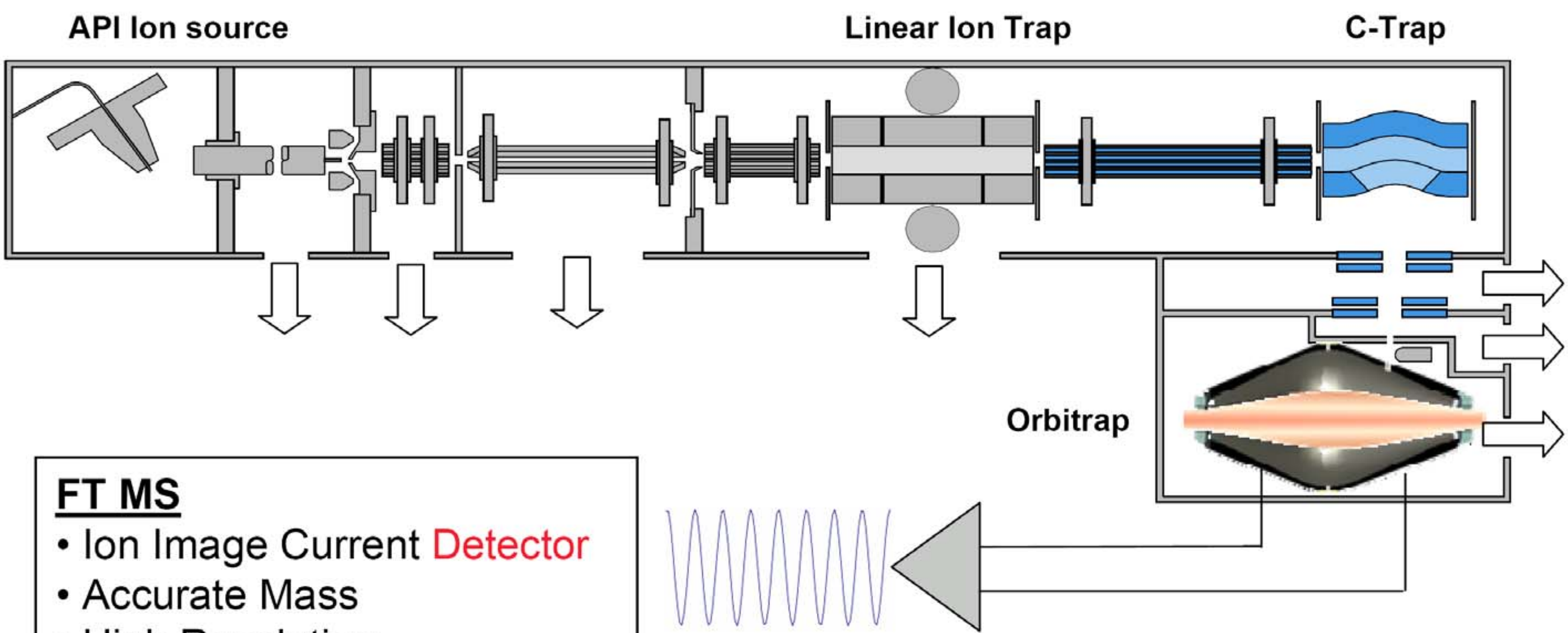
22PT + TSR Heidelberg



LTQ Orbitrap

Linear Ion Trap MS
- MS, MS/MS and MS^n Analysis
- **AGC Control**
- Secondary Electron Multiplier Detector

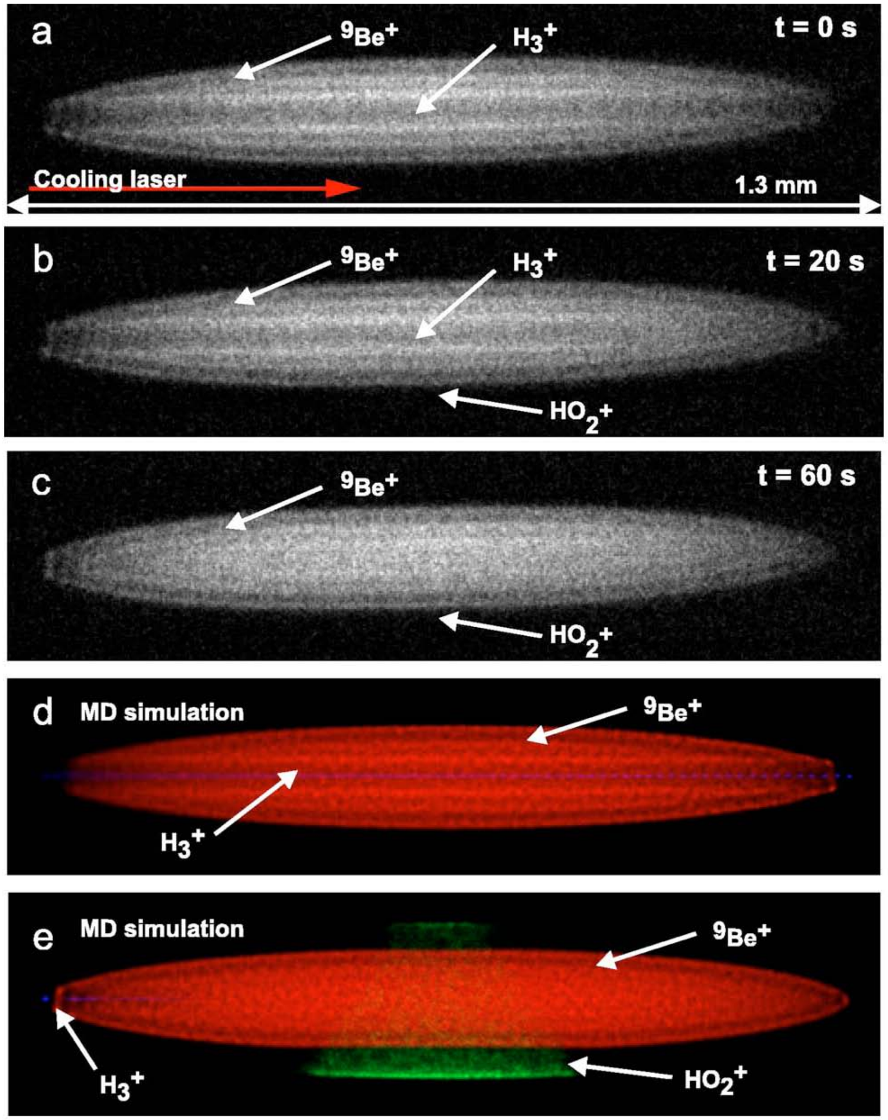
Two Detectors
Two Data Signals
One Instrument



FT MS
- Ion Image Current Detector
- Accurate Mass
- High Resolution

S. Horning Thermo 2005

Coulomb clusters, sympathetic cooling



Ultracold $^9Be^+$ ions

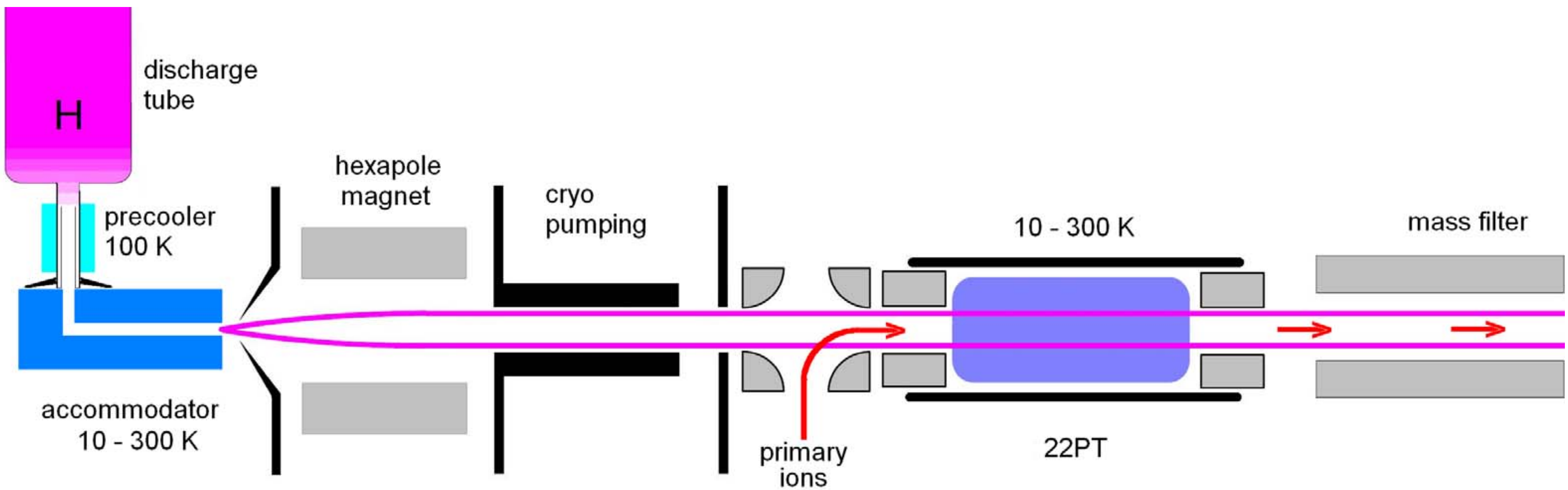
$H_3^+ + O_2$

"Ultracold laser-cooled and sympathetically cooled ions in traps open up the possibility for high-precision measurements on well-localized systems"

Useful for astrophysics and -chemistry?

B. Roth *et al. Ion-neutral chemical reactions between ultracold localized ions and neutral molecules with single-particle resolution Phys. Rev.* **A 73** (2006) 42712-1

Focusing H atoms: E_H < 1meV



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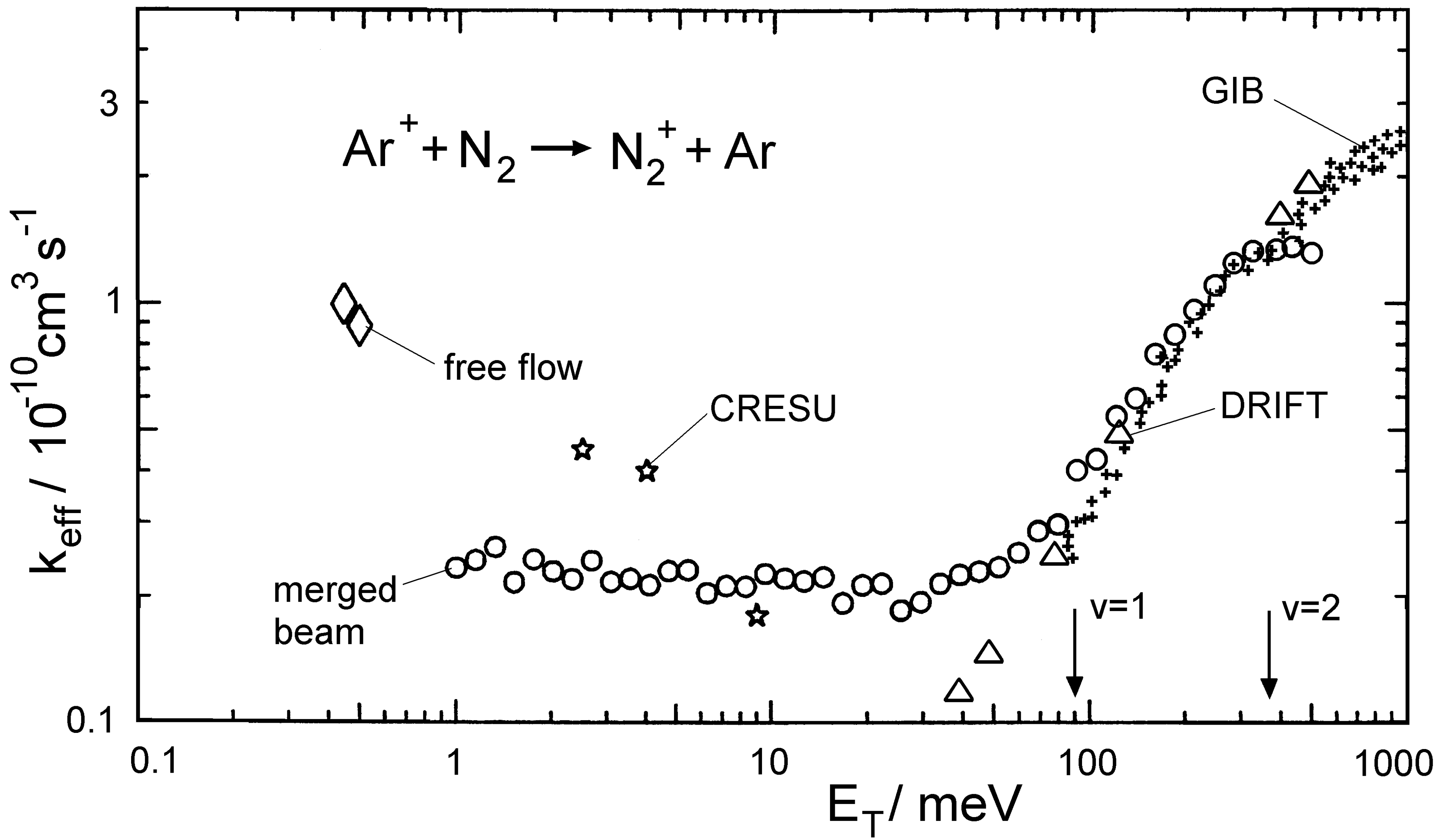
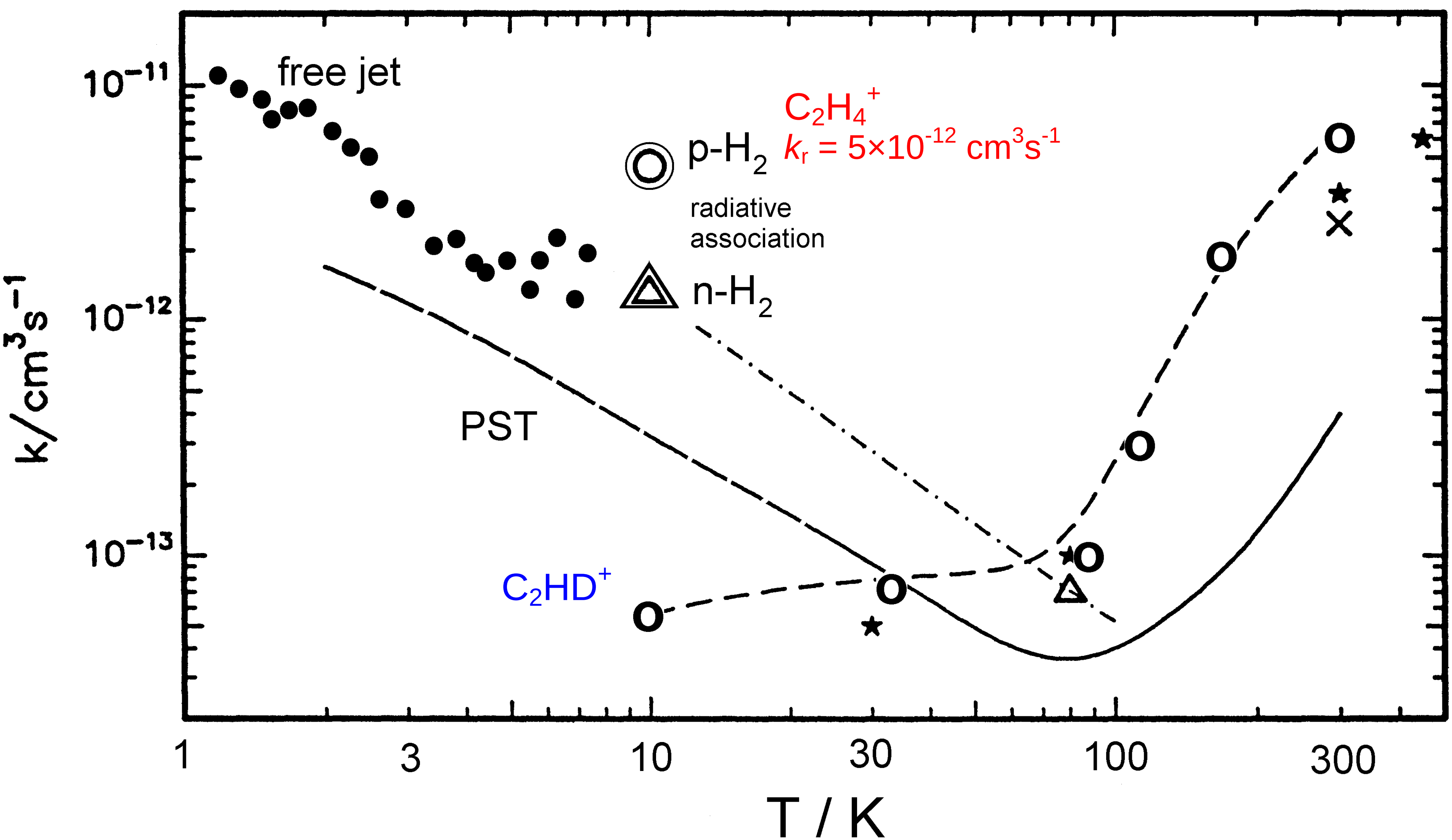
Test reaction $Ar^+ + N_2$

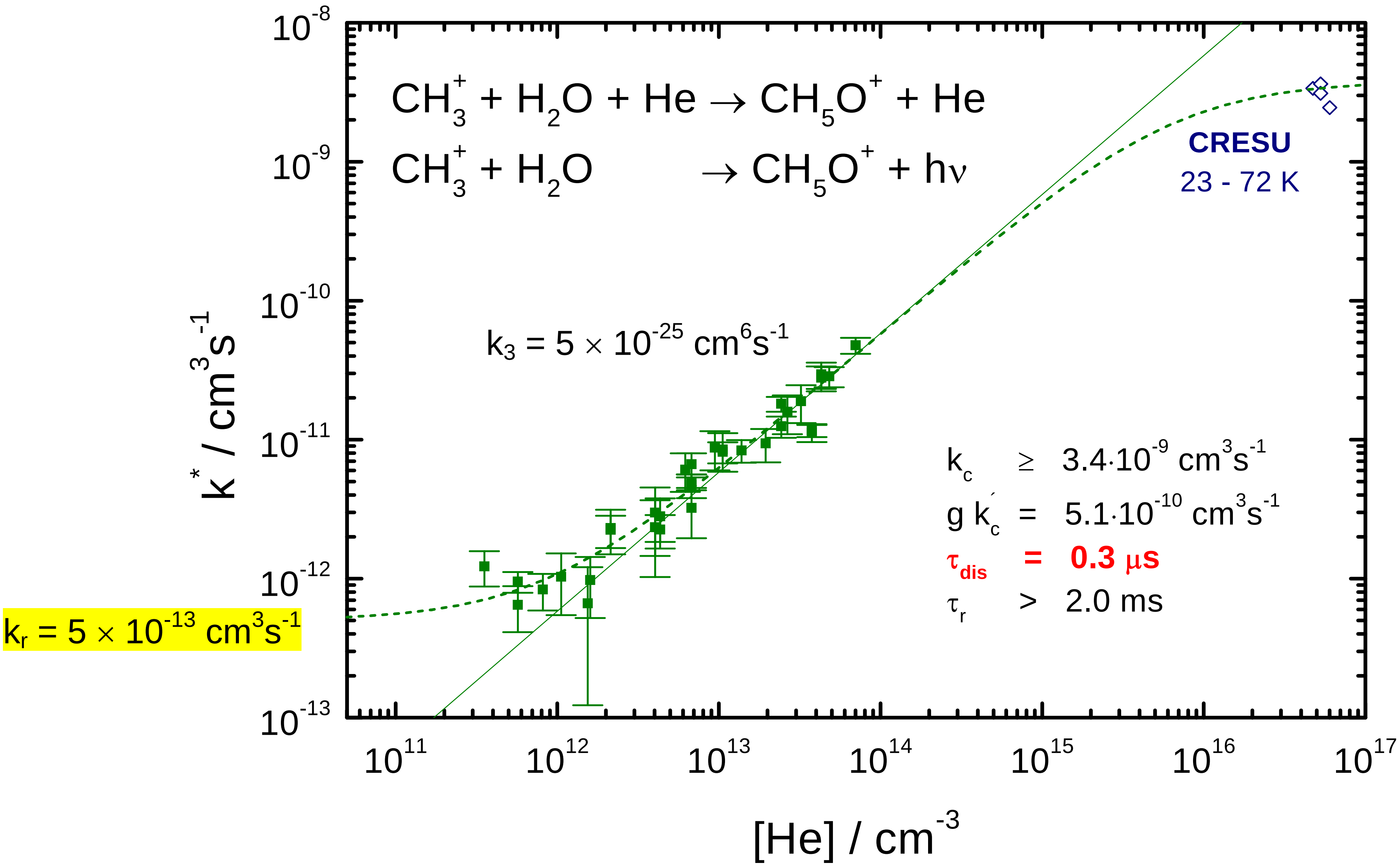


$C_2H_2^+ + H_2$



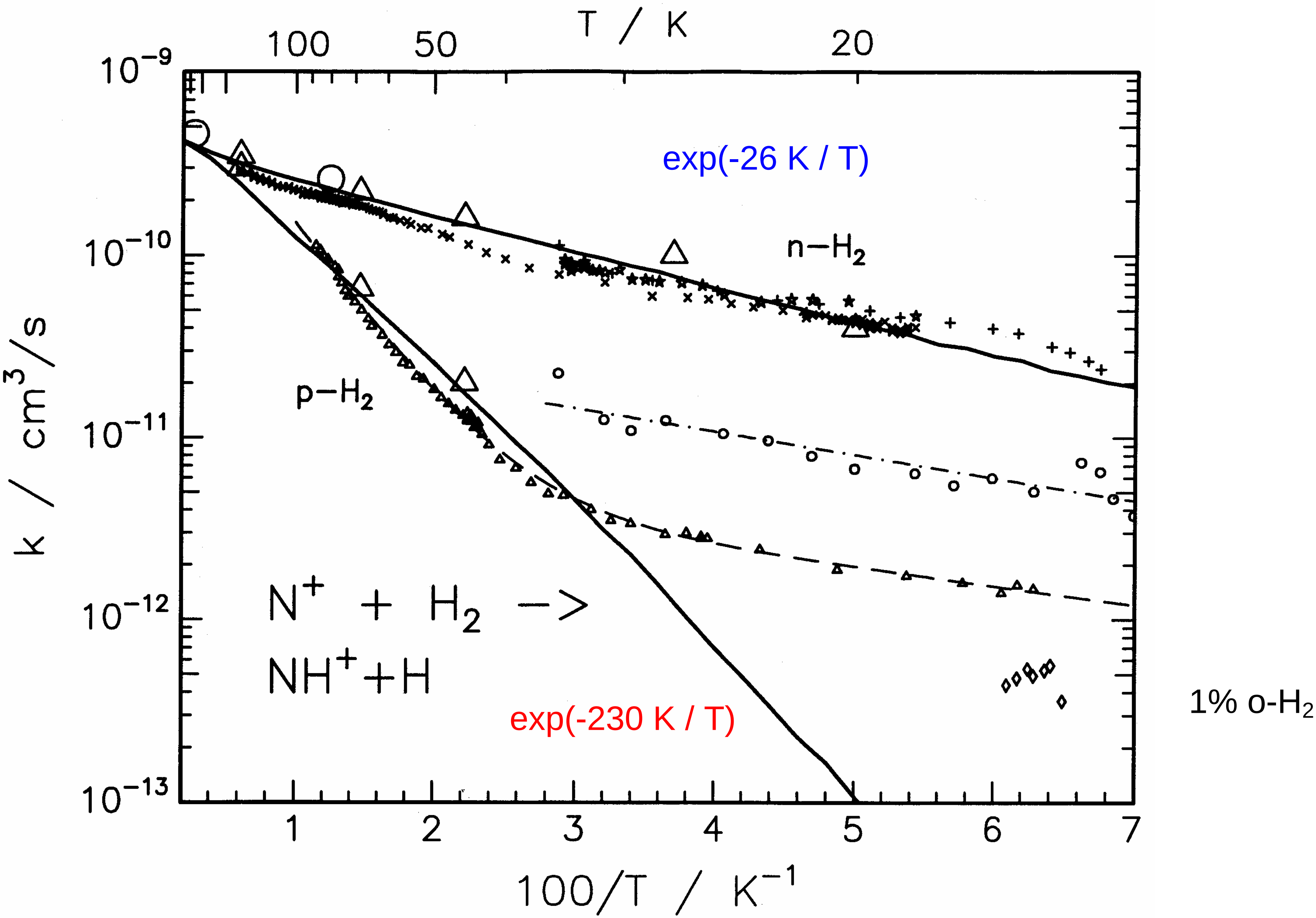
D. Gerlich, *in: "Molecules and Grains in Space"*
I. Nenner (ed.) AIP Press, New York (1994) 489

Formation of Methanol in space?



A. Luca, D. Voulot, D. Gerlich 2002

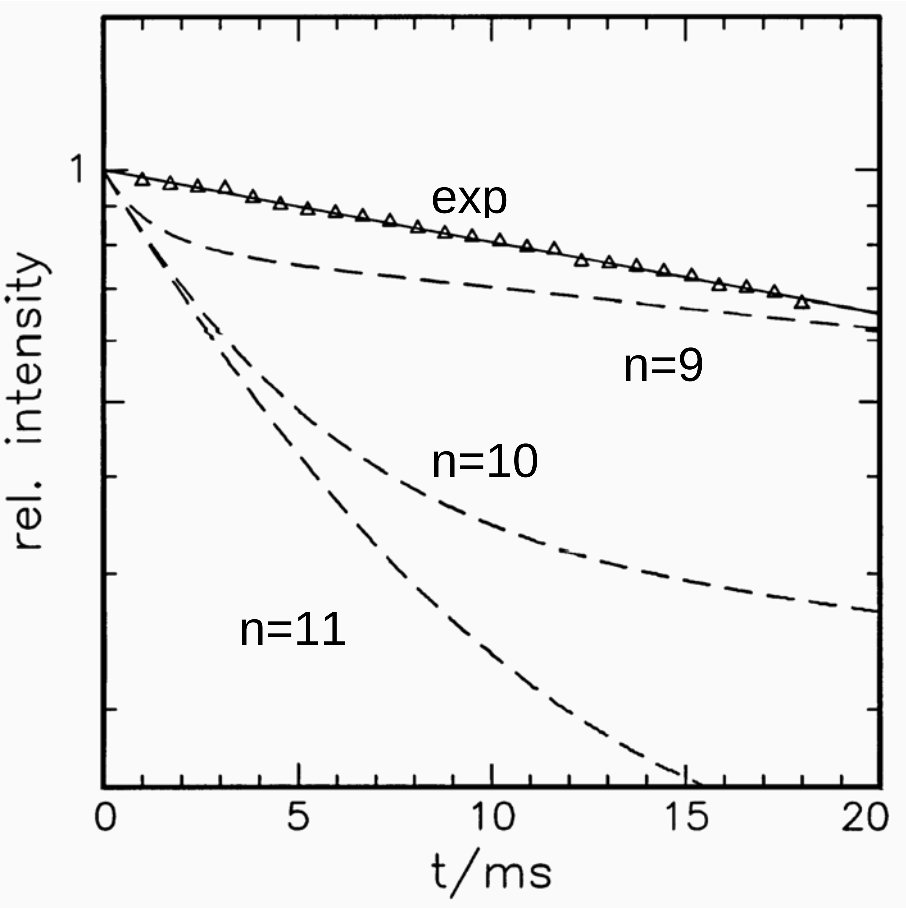
N^+ + p/n-H_2: temperature dependence

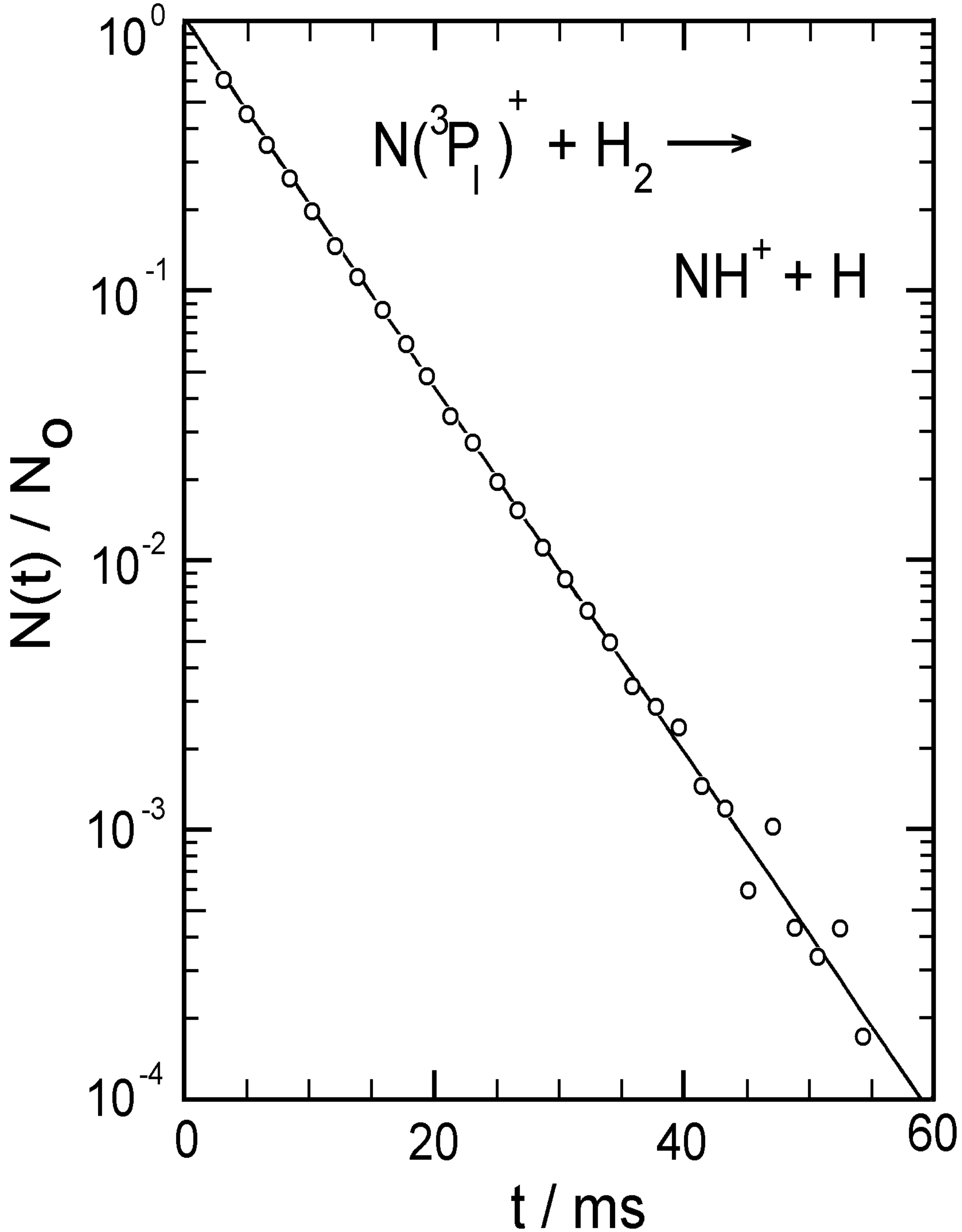


D. Gerlich "Experimental Investigation of Ion-Molecule Reactions Relevant to Interstellar Chemistry"
J. Chem. Soc. Farraday Trans., 89 (13), 2199-2208, (1993)

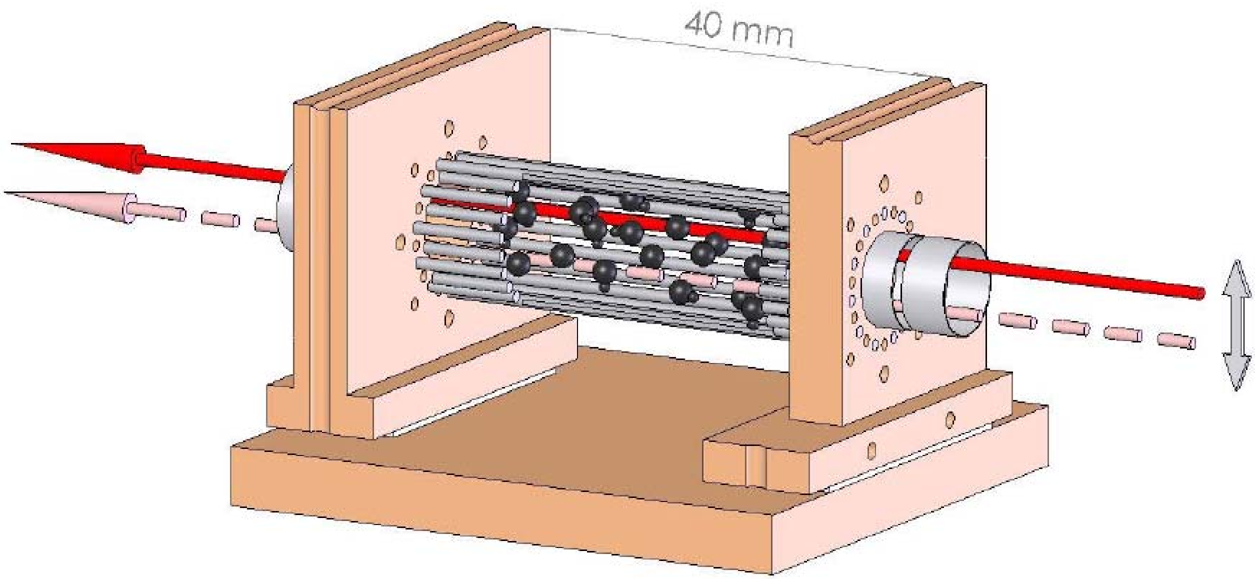
$N(^3P_J)^+ + H_2$ relaxation vs. reaction

Phase space theory	$k(^3P_0) = 1.7\times10^{-11}$ cm^3/s
	$k(^3P_1) = 2.6\times10^{-10}$ cm^3/s
	$k(^3P_2) = 4.6\times10^{-10}$ cm^3/s
Relaxation rate coefficients	$k(^3P_l\rightarrow{}^3P_{l-1}) = 10^{-n}$ cm^3/s n=9, 10 or 11
Exp.: [n-H_2] = 5.6×10^{11} cm^{-3}	$k_{EXP} = 3.83\times10^{-11}$ cm^3/s





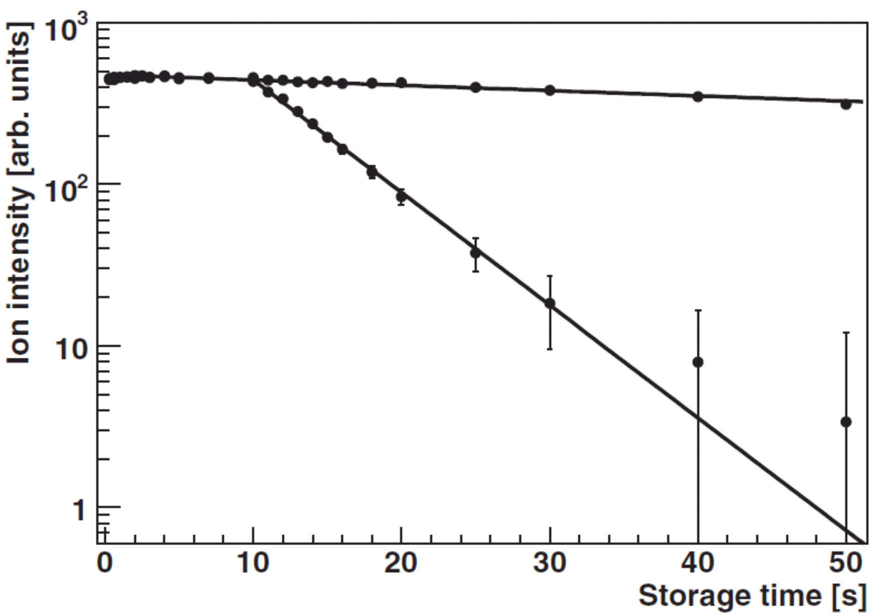
Photodetachment: OH^-

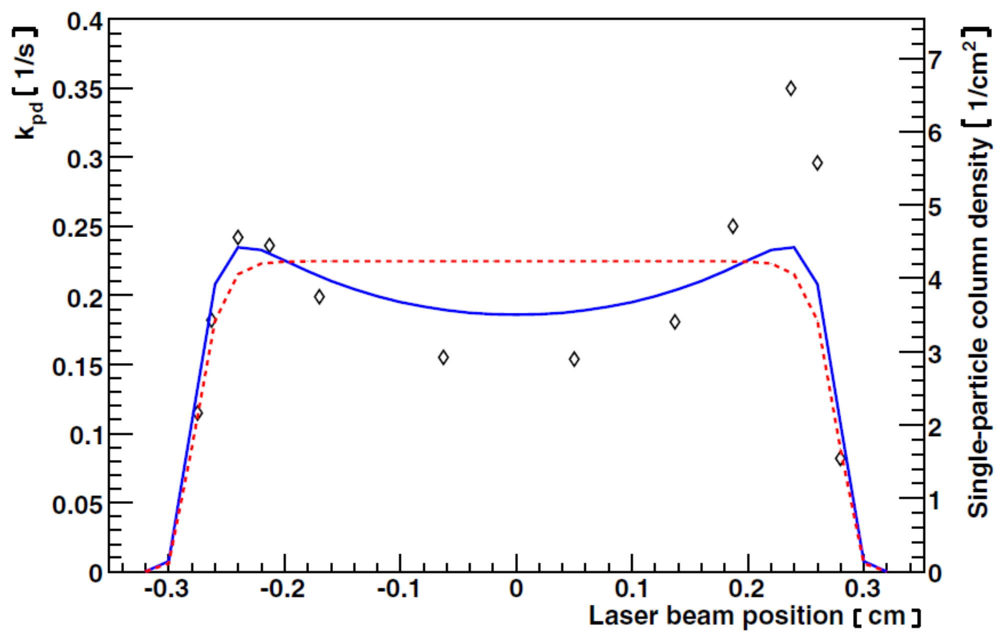


T = 170 K

$\sigma = 5.6 \times 10^{-18}\ cm^2$

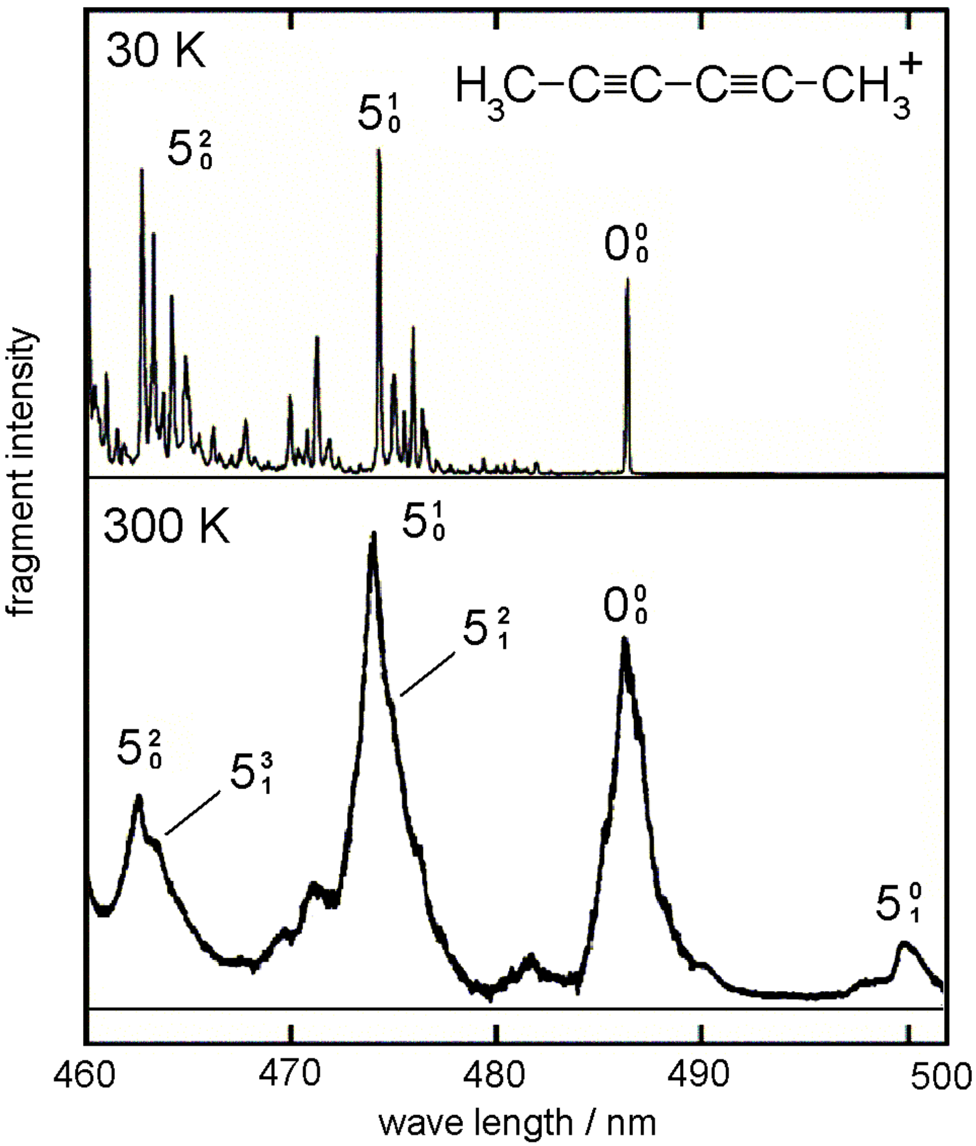
$\sigma \sim (2J+1) \times 10^{-18}\ cm^2$



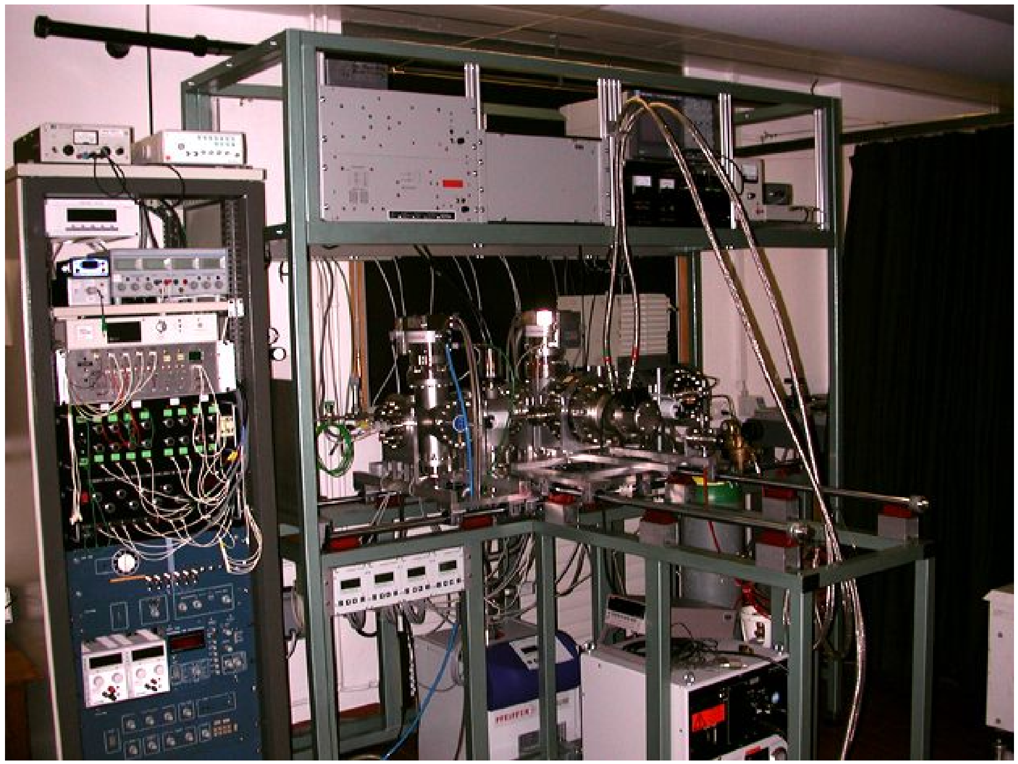


S. Trippel *et al.* (2006) *Photodetachment of cold OH− in a multipole ion trap* *Phys. Rev. Lett.* **97:** 193003-1.

Electronic spectra: the Basel 22PT

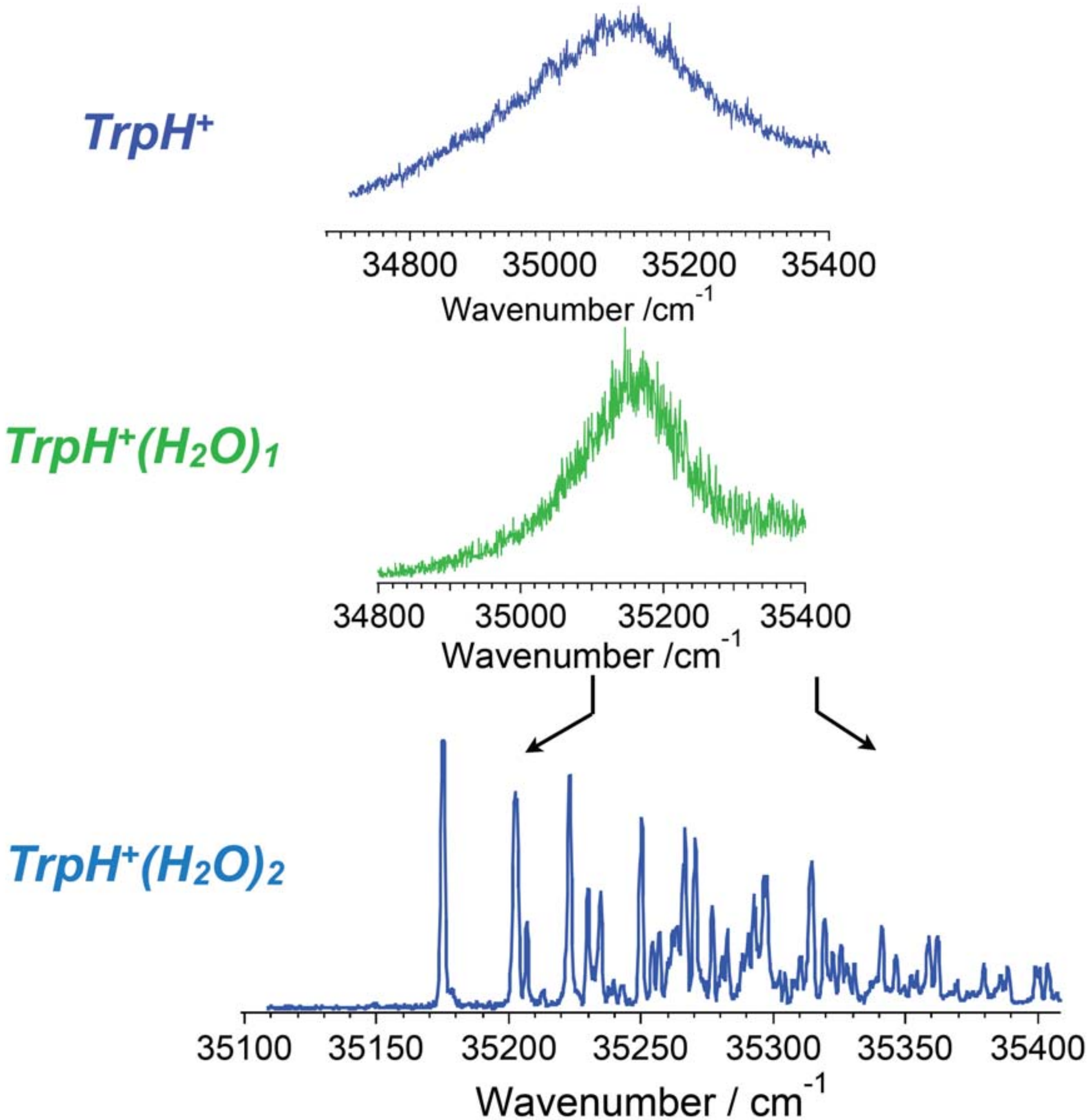


one photon dissociation spectrum
30 K
300 K



A. Dhzonson, J.P. Maier *Electronic absorption spectra of cold organic cations: 2,4-Hexadiyne. Int. J. Mass. Spec.* **255** (2006)139

Microsolvation of cold, protonated tryptophan



O. Boyarkin, S. Mercier, A. Kamariotis, and T. Rizzo
J. Am. Chem. Soc.; **128** (2006) 2816

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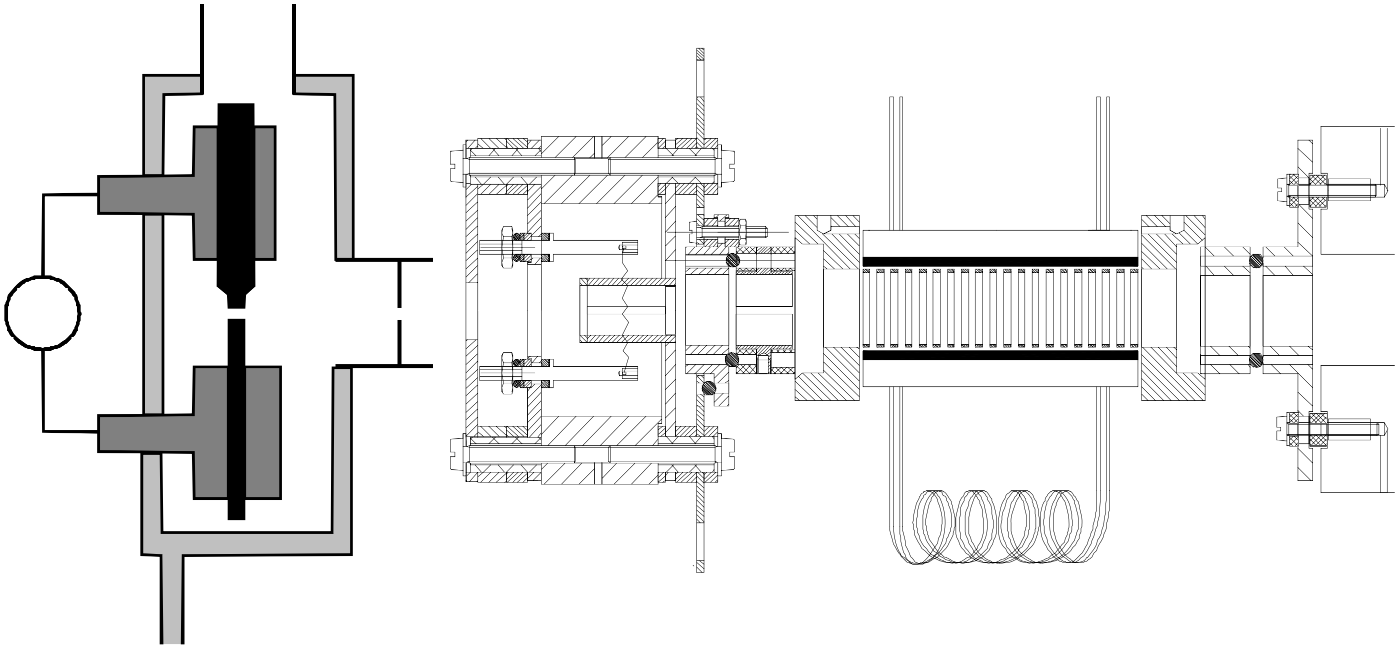
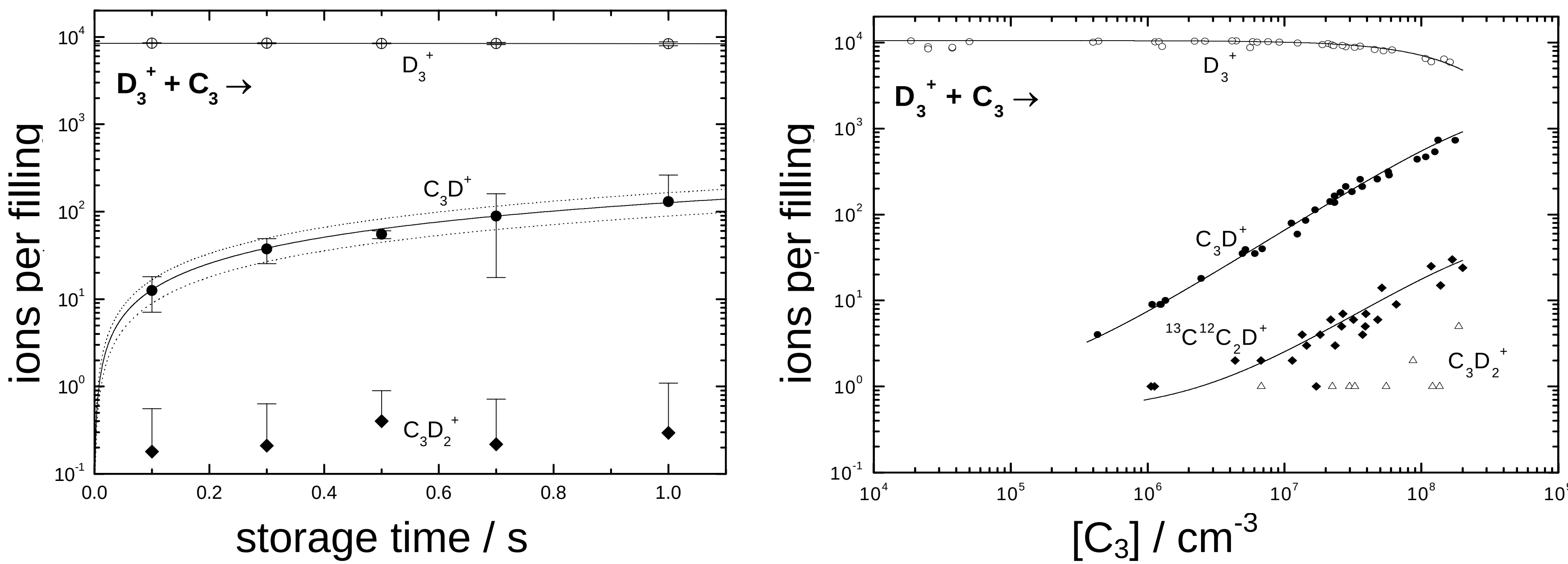
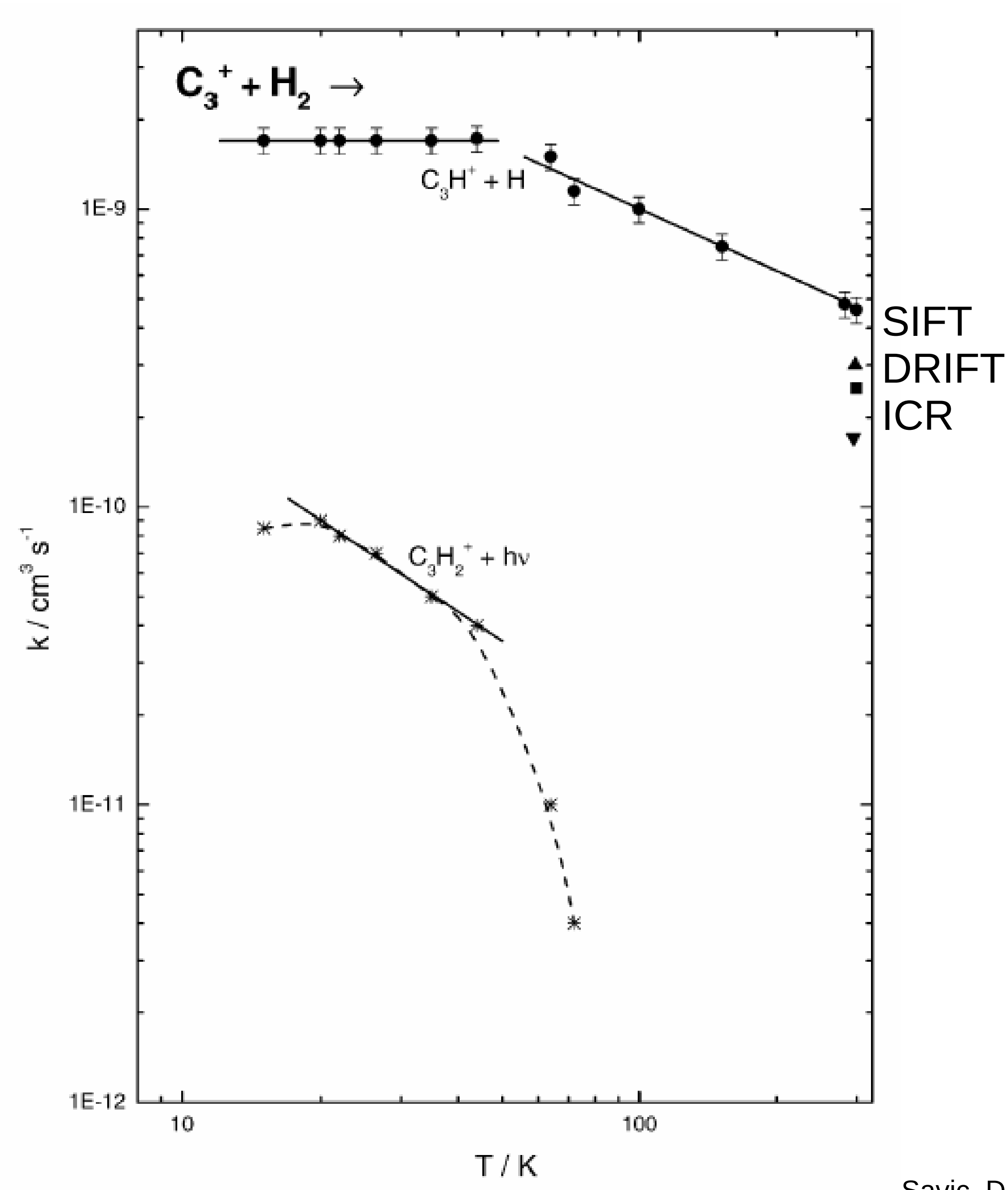
carbon beam - ionizer - ring electrode trap





I. Savic, I. Cermak, D. Gerlich, Int. J. Mass Spectrom., **240** (2005) 139

$C_3^+ + H_2$



197 kJ mol^{-1} exothermic!

why temperature dependent?

radiative association 5 % !

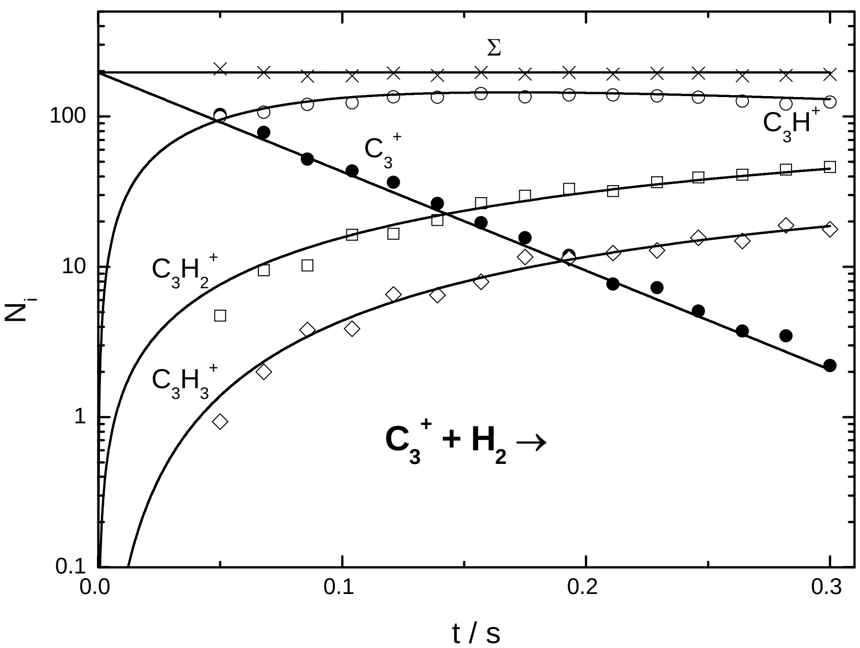
bending modes?

ground state structure?

linear below 50 K?

Savic, D. Gerlich, Phys. Chem. Chem. Phys. 7, 1026-1035 (2005)

$C_3H_n^+ + H_2$, HD and D_2 @ 15 K



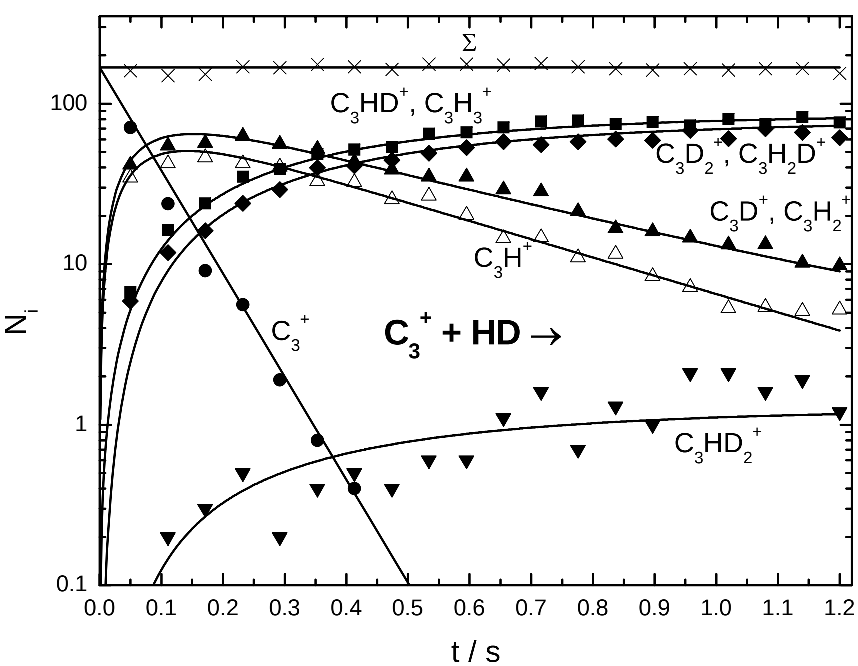


Table 1 Measured reaction rate coefficients[a] for the indicated reactions

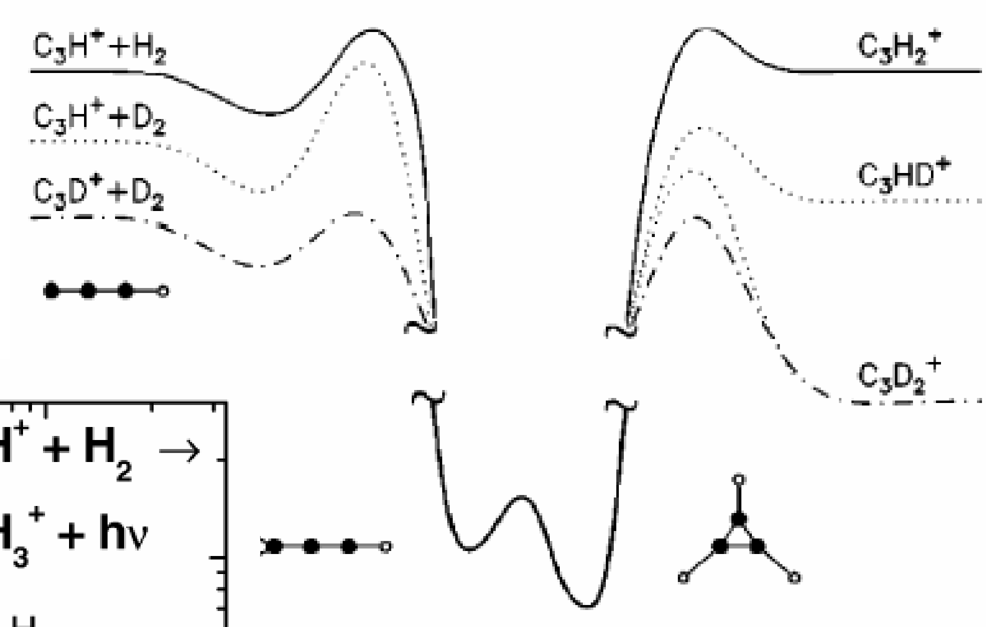
Reaction				$k/\text{cm}^3\ \text{s}^{-1}$ or $\alpha/\text{cm}^3\ \text{s}^{-1}$
C_3^+	+ n-H_2	→ C_3H^+	+ H	1.7(−9)
				4.7(−10)
				4.6(−10)
		→ $C_3H_2^+$	+ $h\nu$	5.7(−12)
				8.5(−11)
		→ C_3H^+	+ H	2.0(−9)
	+ p-H_2			2.0(−9)
				1.8(−10)
				2.5(−10)
				3.0(−10)
	+ HD	→ C_3D^+	+ H	9.3(−10)
		→ C_3H^+	+ D	7.6(−10)
		→ C_3HD^+	+ $h\nu$	5.9(−11)
	+ n-D_2	→ C_3D^+	+ D	1.3(−9)
				1.3(−10)
				1.8(−10)
				1.5(−10)
C_3H^+	+ n-H_2	→ $C_3H_2^+$	+ H	1.5(−10)
				1.4(−11)
				1.1(−10)
				1.5(−11)
				5.0(−11)
	+ p-H_2			4.0(−11)
	+ n-H_2	→ $C_3H_3^+$	+ $h\nu$	2.3(−11)
				6.0(−11)
				2.0(−10)
	+ p-H_2			2.2(−10)
				3.4(−12)
	+ HD	→ C_3D^+	+ H_2	5.6(−11)
		→ C_3HD^+	+ H	4.6(−10)
		→ $C_3H_2^+$	+ D	3.0(−12)
		→ $C_3H_2D^+$	+ $h\nu$	3.2(−11)
	+ D_2	→ C_3D^+	+ HD	3.0(−13)
		→ C_3HD^+	+ D	1.0(−11)
		→ $C_3D_2^+$	+ H	2.7(−11)
		→ $C_3HD_2^+$	+ $h\nu$	4.0(−12)
C_3D^+	+ HD	→ C_3HD^+	+ D	1.0(−10)
		→ $C_3D_2^+$	+ H	8.3(−11)
		→ $C_3HD_2^+$	+ $h\nu$	8.0(−12)
	+ D_2	→ $C_3D_2^+$	+ D	1.7(−10)
		→ $C_3D_3^+$	+ $h\nu$	1.3(−10)

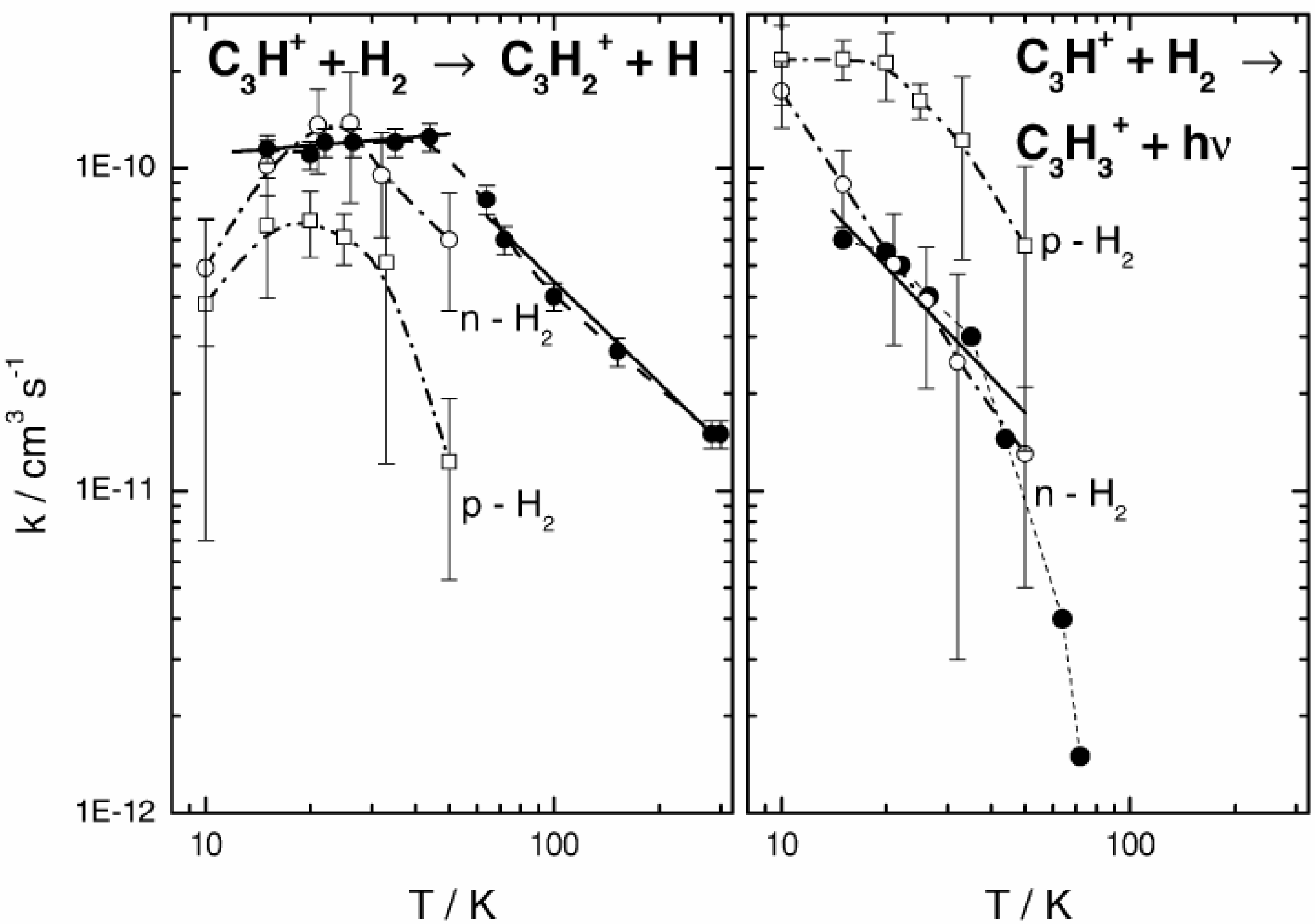
I. Savic, D. Gerlich, Phys. Chem. Chem. Phys. 7, 1026-1035 (2005)

$C_3H^+ + H_2$: competing channels

C_3H^+ + HD →

C_3HD^+: $k = 4.6 \times 10^{-10}$ cm^{3}s^{-1}
$C_3H_2^+$: $k = 3.0 \times 10^{-12}$ cm^{3}s^{-1}





strong isotope effects
competition
lifetimes 50 µs

barriers
zero point energies
exchange symmetry
nuclear spin

I. Savic, D. Gerlich, Phys. Chem. Chem. Phys. 7, 1026-1035 (2005)

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Complete depletion in prestellar cores

Reaction
$H + H \rightarrow H_2(p)$
$H + H \rightarrow H_2(o)$
$H + D \rightarrow HD$
$H + crp \rightarrow H^+ + e^-$
$He + crp \rightarrow He^+ + e^-$
$H_2(p) + crp \rightarrow H^+ + H + e^-$
$H_2(o) + crp \rightarrow H^+ + H + e^-$
$H_2(p) + crp \rightarrow H + H$
$H_2(o) + crp \rightarrow H + H$
$H_2(p) + crp \rightarrow H_2^+(p) + e^-$
$H_2(o) + crp \rightarrow H_2^+(o) + e^-$
$H^+ + H_2(o) \rightarrow H^+ + H_2(p)$
$H_3^+(p) + H_2(o) \rightarrow H_3^+(o) + H_2(p)$
$H_3^+(o) + H_2(o) \rightarrow H_3^+(p) + H_2(p)$
$H^+ + H_2(p) \rightarrow H^+ + H_2(o)$
$H_3^+(p) + H_2(p) \rightarrow H_3^+(o) + H_2(o)$
$H_3^+(o) + H_2(p) \rightarrow H_3^+(p) + H_2(o)$
$H_2^+(o) + H_2(o) \rightarrow H_3^+(p) + H$
$H_2^+(o) + H_2(o) \rightarrow H_3^+(o) + H$
$H_2^+(p) + H_2(o) \rightarrow H_3^+(p) + H$
$H_2^+(p) + H_2(o) \rightarrow H_3^+(o) + H$
$H_2^+(o) + H_2(p) \rightarrow H_3^+(p) + H$
$H_2^+(o) + H_2(p) \rightarrow H_3^+(o) + H$
$H_2^+(p) + H_2(p) \rightarrow H_3^+(p) + H$
$He^+ + H_2(p) \rightarrow H^+ + H + He$
$He^+ + H_2(o) \rightarrow H^+ + H + He$
$H^+ + e^- \rightarrow H + photon$
$H_2^+(p) + e^- \rightarrow H + H$
$H_2^+(o) + e^- \rightarrow H + H$
$He^+ + e^- \rightarrow He + photon$
$H_3^+(p) + e^- \rightarrow H + H + H$
$H_3^+(o) + e^- \rightarrow H + H + H$
$H_3^+(p) + e^- \rightarrow H_2(p) + H$
$H_3^+(p) + e^- \rightarrow H_2(o) + H$
$H_3^+(o) + e^- \rightarrow H_2(o) + H$

Reaction
$HD^+ + H_2(p) \rightarrow H_2D^+(p) + H$
$HD^+ + H_2(p) \rightarrow H_2D^+(o) + H$
$HD^+ + H_2(o) \rightarrow H_2D^+(p) + H$
$HD^+ + H_2(o) \rightarrow H_2D^+(o) + H$
$HD^+ + H_2(p) \rightarrow H_3^+(p) + D$
$HD^+ + H_2(o) \rightarrow H_3^+(o) + D$
$HD^+ + H_2(o) \rightarrow H_3^+(p) + D$
$H_2^+(p) + HD \rightarrow H_2D^+(p) + H$
$H_2^+(p) + HD \rightarrow H_2D^+(o) + H$
$H_2^+(o) + HD \rightarrow H_2D^+(p) + H$
$H_2^+(o) + HD \rightarrow H_2D^+(o) + H$
$H_2^+(p) + HD \rightarrow H_3^+(p) + D$
$H_2^+(o) + HD \rightarrow H_3^+(p) + D$
$H_2^+(o) + HD \rightarrow H_3^+(o) + D$
$H_3^+(p) + D \rightarrow H_2D^+(p) + H$
$H_3^+(p) + D \rightarrow H_2D^+(o) + H$
$H_3^+(o) + D \rightarrow H_2D^+(o) + H$
$H_2D^+(p) + H \rightarrow H_3^+(p) + D$
$H_2D^+(o) + H \rightarrow H_3^+(o) + D$
$H_2D^+(o) + H \rightarrow H_3^+(p) + D$
$H_3^+(p) + HD \rightarrow H_2D^+(p) + H_2(p)$
$H_3^+(p) + HD \rightarrow H_2D^+(p) + H_2(o)$
$H_3^+(p) + HD \rightarrow H_2D^+(o) + H_2(p)$
$H_3^+(p) + HD \rightarrow H_2D^+(o) + H_2(o)$
$H_3^+(o) + HD \rightarrow H_2D^+(p) + H_2(o)$
$H_3^+(o) + HD \rightarrow H_2D^+(o) + H_2(o)$
$H_2D^+(p) + H_2(o) \rightarrow H_2D^+(o) + H_2(p)$
$H_2D^+(o) + H_2(p) \rightarrow H_2D^+(p) + H_2(o)$
$H_2D^+(o) + H_2(o) \rightarrow H_2D^+(p) + H_2(p)$
$H_2D^+(p) + H_2(p) \rightarrow H_2D^+(o) + H_2(o)$
$H_2D^+(p) + H_2(p) \rightarrow H_3^+(p) + HD$
$H_2D^+(p) + H_2(o) \rightarrow H_3^+(p) + HD$
$H_2D^+(p) + H_2(o) \rightarrow H_3^+(o) + HD$
$H_2D^+(o) + H_2(p) \rightarrow H_3^+(p) + HD$
$H_2D^+(o) + H_2(o) \rightarrow H_3^+(p) + HD$
$H_2D^+(o) + H_2(o) \rightarrow H_3^+(o) + HD$

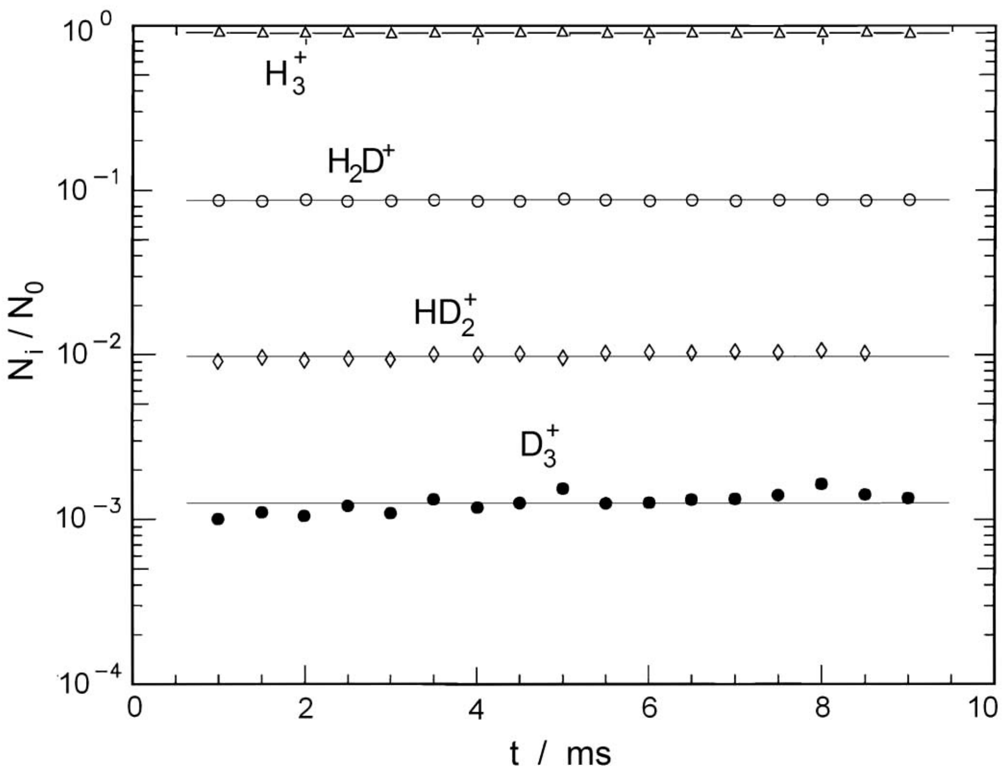
Reaction
$D_3^+ + H_2(o) \rightarrow H_2D^+(o) + D_2$
$D_3^+ + H_2(p) \rightarrow HD_2^+ + HD$
$D_3^+ + H_2(o) \rightarrow HD_2^+ + HD$
$D_3^+ + e^- \rightarrow D + D + D$
$D_3^+ + e^- \rightarrow D_2 + D$
$D_2^+ + D_2 \rightarrow D_3^+ + D$
$D_3^+ + HD \rightarrow HD_2^+ + D_2$
$g^0 + secpho \rightarrow g^+ + e^-$
$g^- + secpho \rightarrow g^0 + e^-$
$g^0 + e^- \rightarrow g^- + photon$
$g^- + H^+ \rightarrow g^0 + H$
$g^- + H_3^+(p) \rightarrow g^0 + H_2(p) + H$
$g^- + H_3^+(p) \rightarrow g^0 + H_2(o) + H$
$g^- + H_3^+(o) \rightarrow g^0 + H_2(o) + H$
$g^- + H_3^+(p) \rightarrow g^0 + 3H$
$g^- + H_3^+(o) \rightarrow g^0 + 3H$

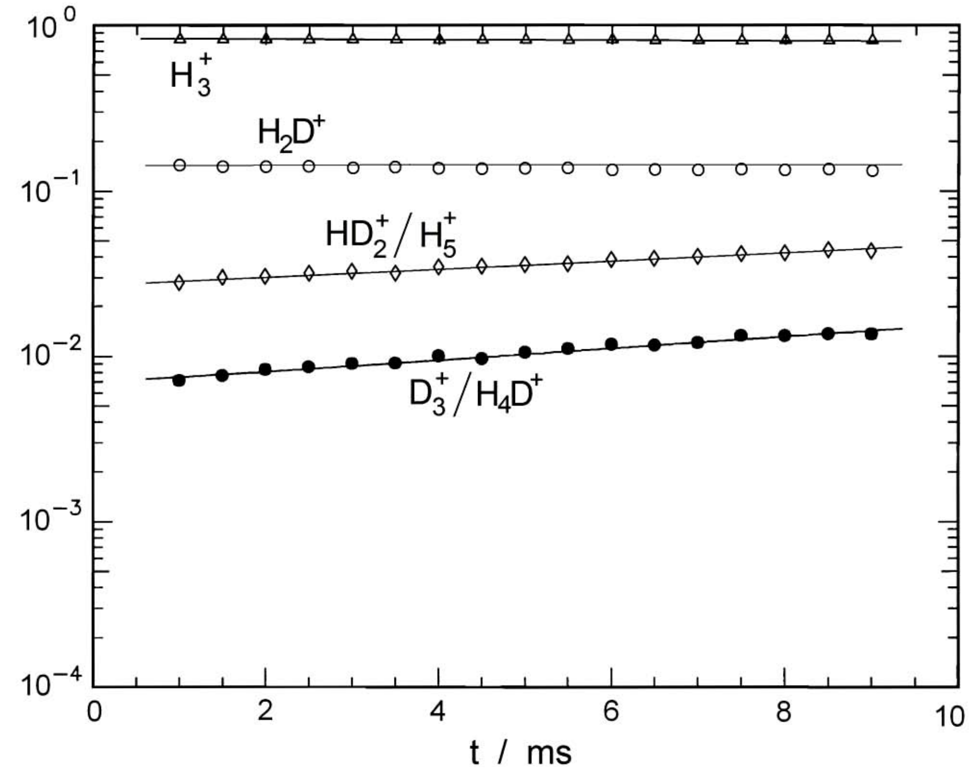
more than 250 reactions

ionization
neutralization
ortho para transitions
deuteration
excitation

C. M. Walmsley, D. R. Flower, G. Pineau des Forets
Astron. & Astroph., 418, 1035-1043, (2004)

H_3^+ + HD: equilibrium of of H_3^+, H_2D^+, HD_2^+, and D_3^+

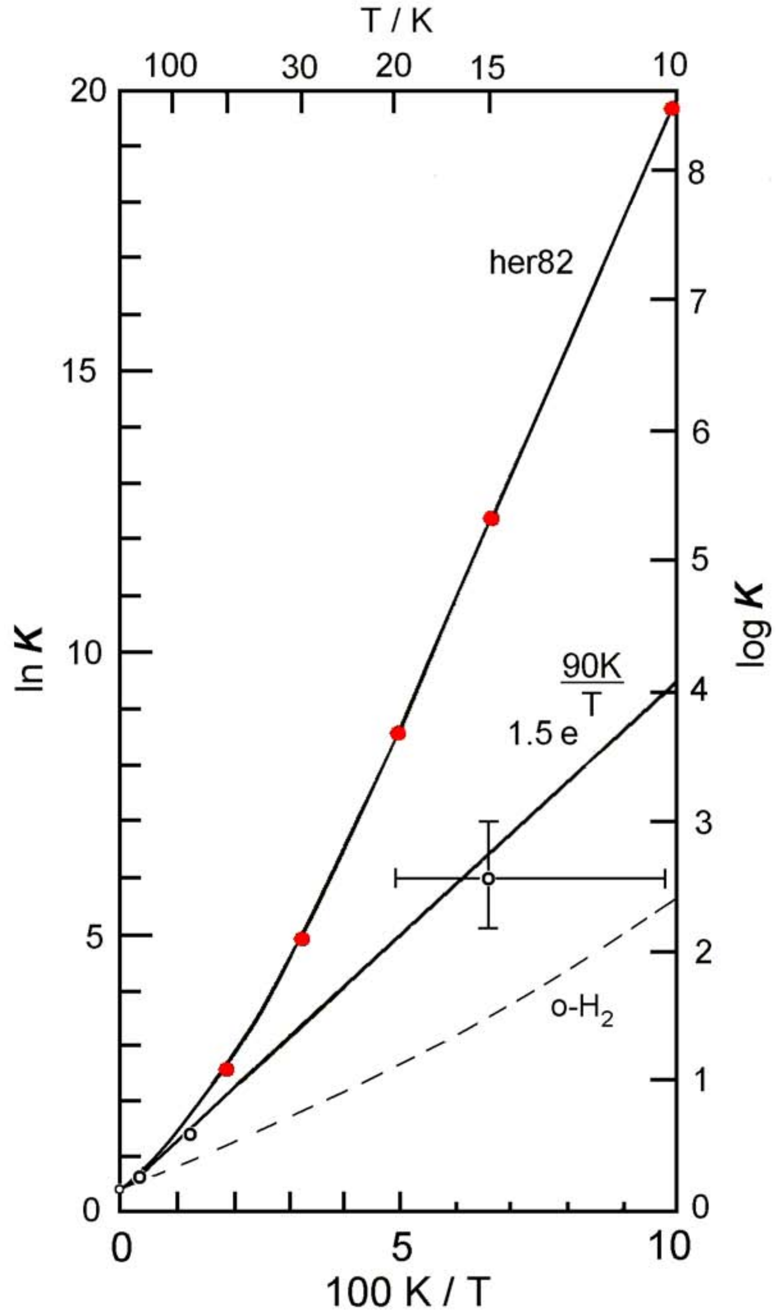




HD $= 9.3 \times 10^{11}$ cm^{-3}
n-H_2 $= 1.4 \times 10^{13}$ cm^{-3}

HD $= 5.7 \times 10^{13}$ cm^{-3}
n-H_2 $= 1.9 \times 10^{14}$ cm^{-3}
$k_3 = 5 \times 10^{-29}$ cm^{6}s^{-1}

$H_3^+ + HD \leftrightarrow H_2D^+ + H_2$
thermodynamic equilibrium?



$$\Delta G = \Delta H - T\Delta S$$

G Gibbs free energy
H enthalpy
S entropy

$$K(T) = \exp\left(-\Delta G / RT\right)$$

equilibrium coefficient

$$K(T) = (\mu'/\mu)^{3/2} \times$$

$$\times \frac{q(H_2D^+)\,q(H_2)}{q(H_3^+)\,q(HD)}$$

$$\times \exp\left(\Delta E_0 / kT\right)$$

$$q(T) \quad \sum_i g_i \exp\left(-\varepsilon_i / kT\right)$$

partition function

$\Delta E_0 =$ 231,8 K

ε_i relative to the lowest state

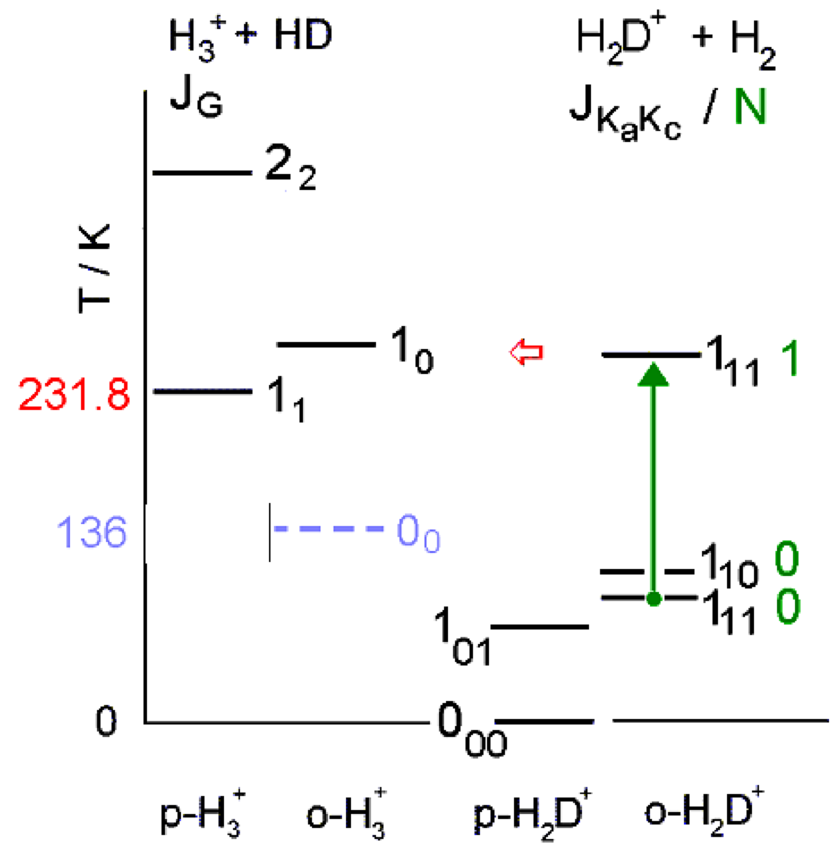
Deuteration $H_3^+ + HD \leftrightarrow H_2D^+ + H_2$: equilibrium constant *K*?

$K \sim \exp(231.8\ K / T)$

T (K)	Adams and Smith	Herbst	Ramanlal
80	4.5 (±1.3)	5.9	6.82
200	2.4 (±0.7)	2.6	1.52
295	2.0 (±0.6)	2.1[a]	1.07[a]

[a] The theoretical value is actually at 300 K.

	T = 10 K
Ramanlal & Tennyson	2.6(+12)
Gerlich et al. (2002)	n-H_2: *K* = 7.4 p-H_2: *K* = 390
T_{22PT} 10 K, (2005) $T_{p\text{-}H2\ Gen}$ 12.5K	>500

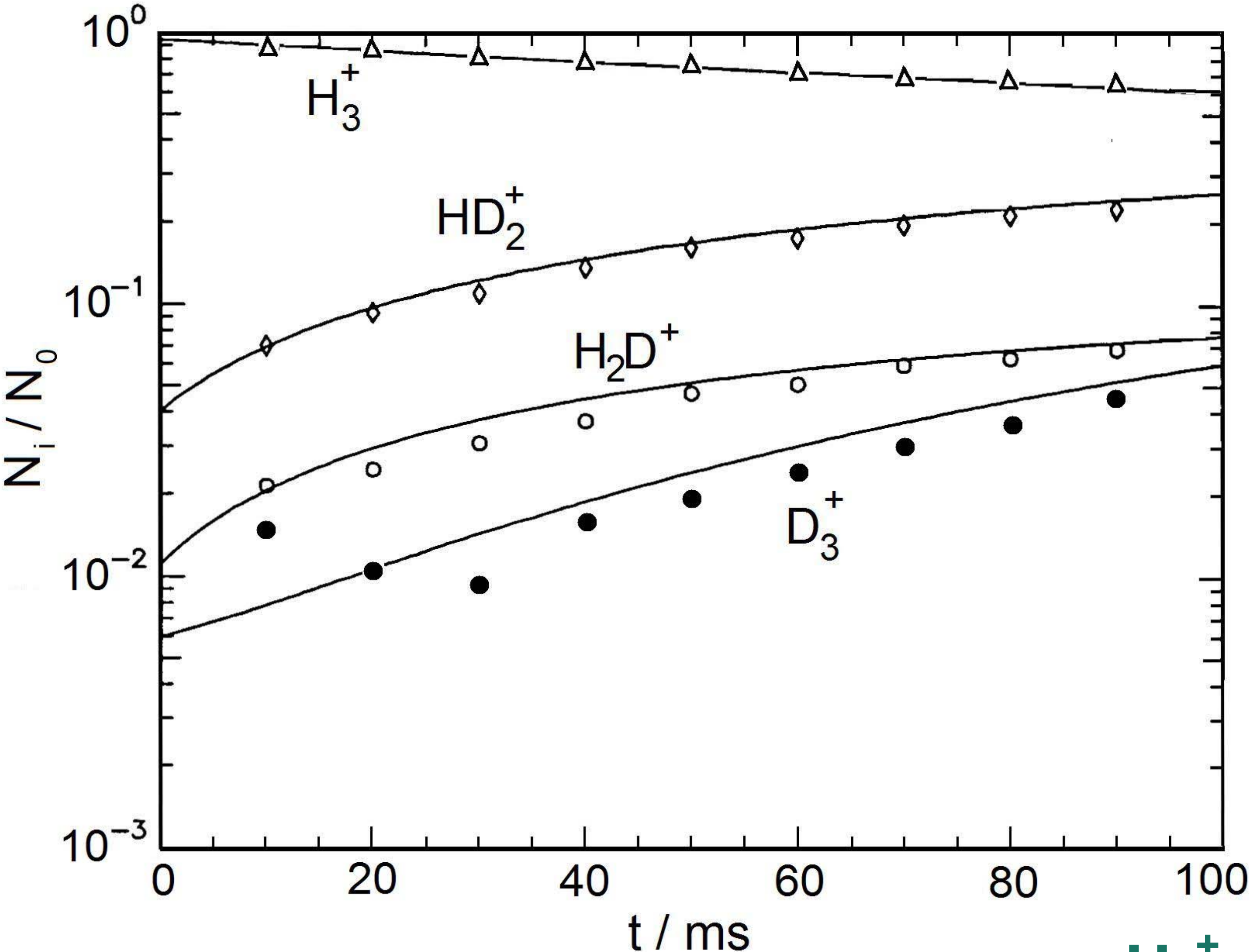


Ramanlal & Tennyson wrote in 2004:
trap experiment disagrees with calculations by
12 orders of magnitude

role of o-H_2 (N=1)
state specific k_i(T)
method overtone LIR

D. Gerlich et al. Phil. Trans. R. Soc. Lond. A 364, 3007–3034 (2006)

$H_3^+ + D_2$



nominal temperature

10 K

D_2 number density

2.8×10^9 cm^{-3}

$k(HD_2^+)$ =

11×10^{-10} cm^3 s^{-1}

$k(H_2D^+)$ =

3×10^{-10} cm^3 s^{-1}

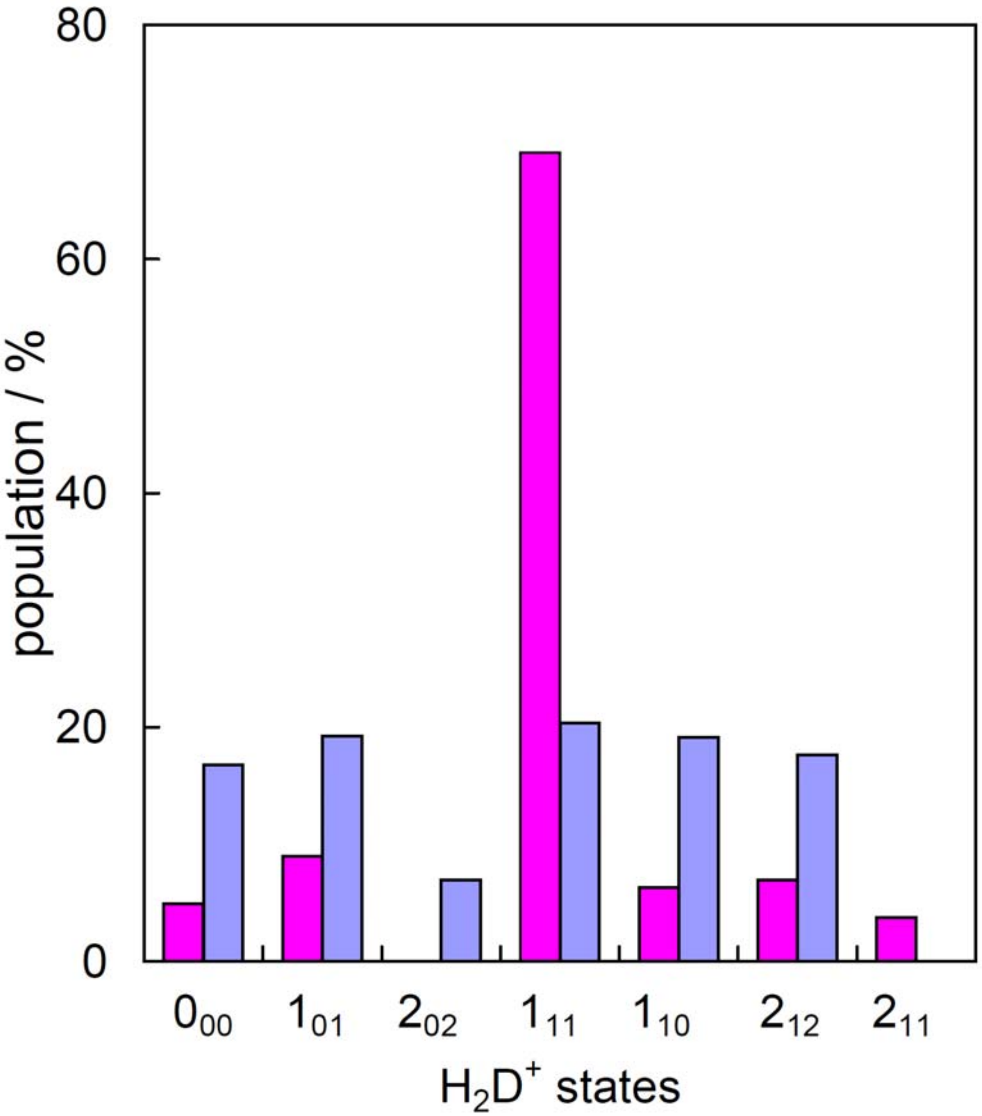
preference for
single proton jump

$H_3^+ + HD \rightarrow H_2D^+ + H_2$

3×10^{-10} cm^3 s^{-1}

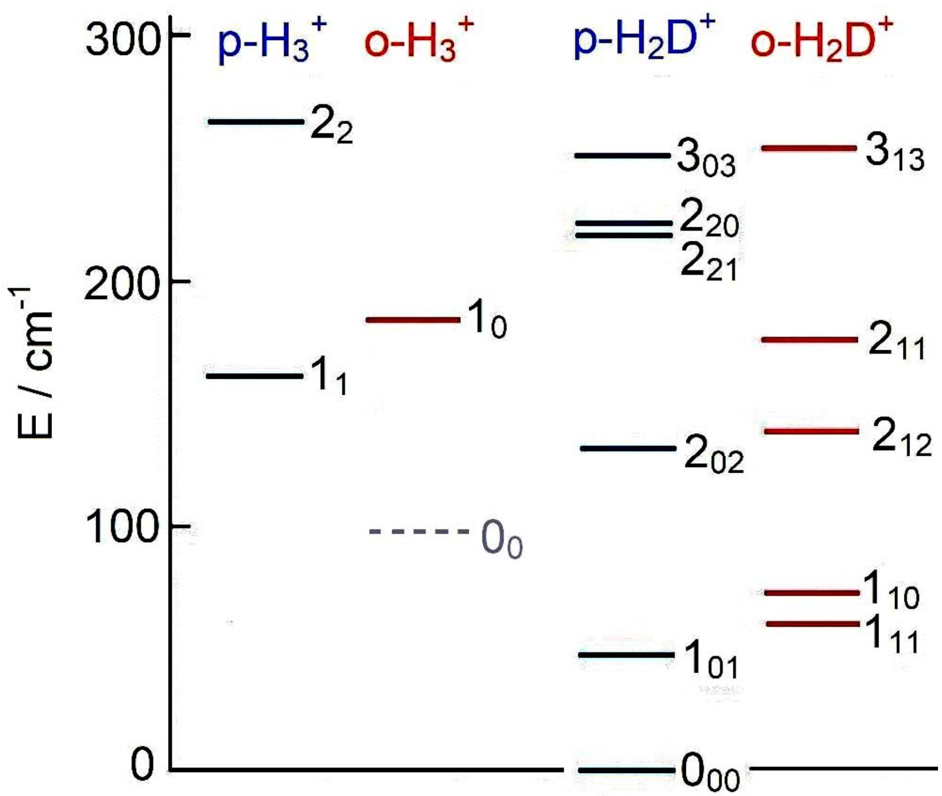
D. Gerlich *et al.* (2006) *Dynamical constraints and nuclear spin caused restrictions in HmDn+ collision systems and deuterated variants Phil. Trans. R. Soc. Lond. A* **364:** 3007

Nuclear spin: propensity rules



o-H_3^+ + HD → H_2D^+

p-H_3^+ + HD → H_2D^+



D. Gerlich *et al.* Phil. Trans. R. Soc. Lond. **A 364** (2006) 3007

Ion - neutral reactions

Dieter Gerlich

Introduction

Instruments for ion chemistry

Typical applications

Selected systems: old or up to date?
Negative ions
Spectroscopy

Carbon reactions

Formation of $C_3H_n^+$

Deuteration of H_3^+

Non statistical behavior

Reactions with H atoms

Beam-trap combination

Outlook

Tucson, sub-K cooling, nano-particles

$CH_4^+ + H_2 \leftrightarrow CH_5^+ + H$

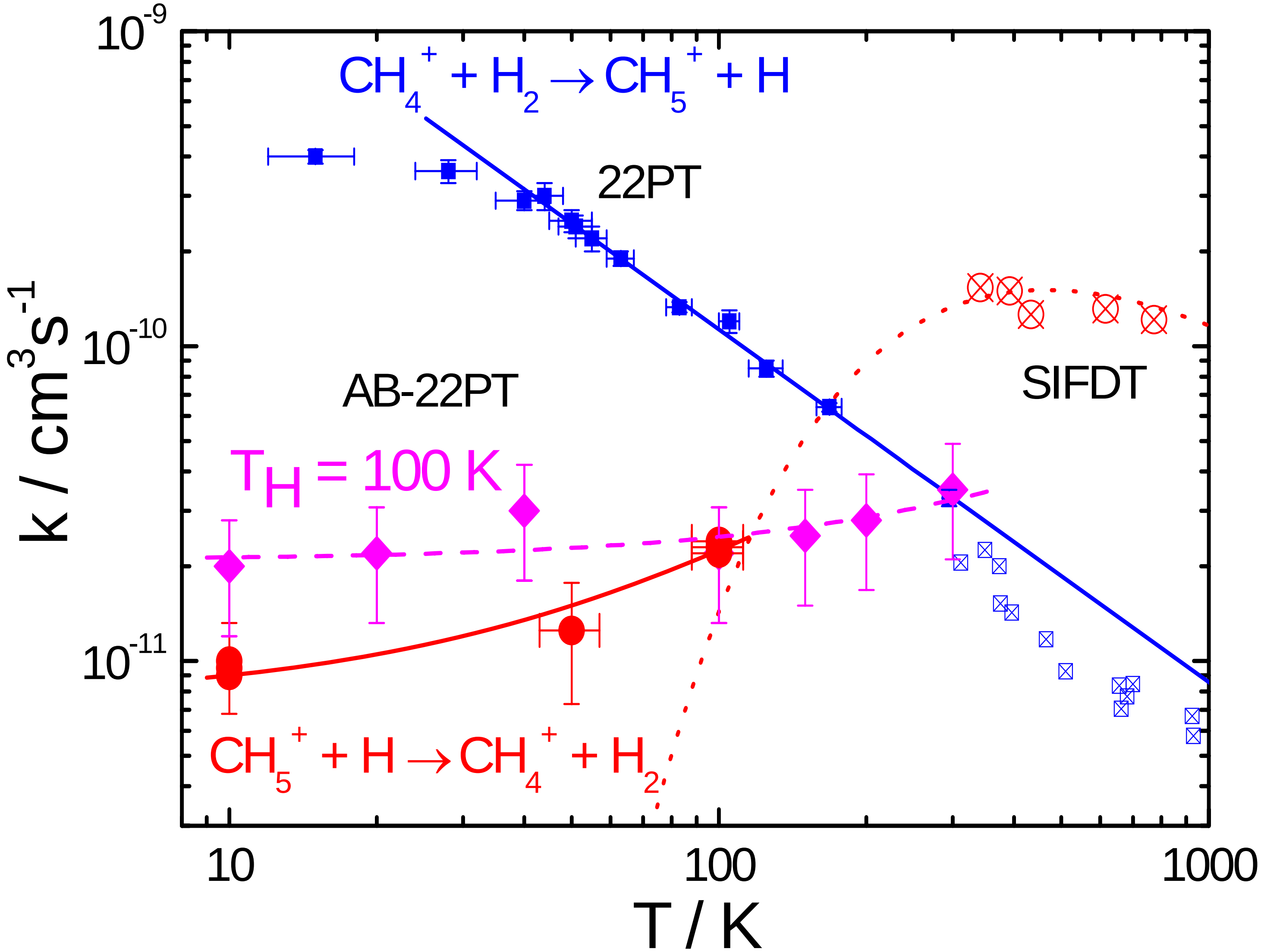
exothermic
but endoentropic?

free energy
$\Delta G = \Delta H - T\,\Delta S$

$\Delta G < 0$
for $T > 150$ K

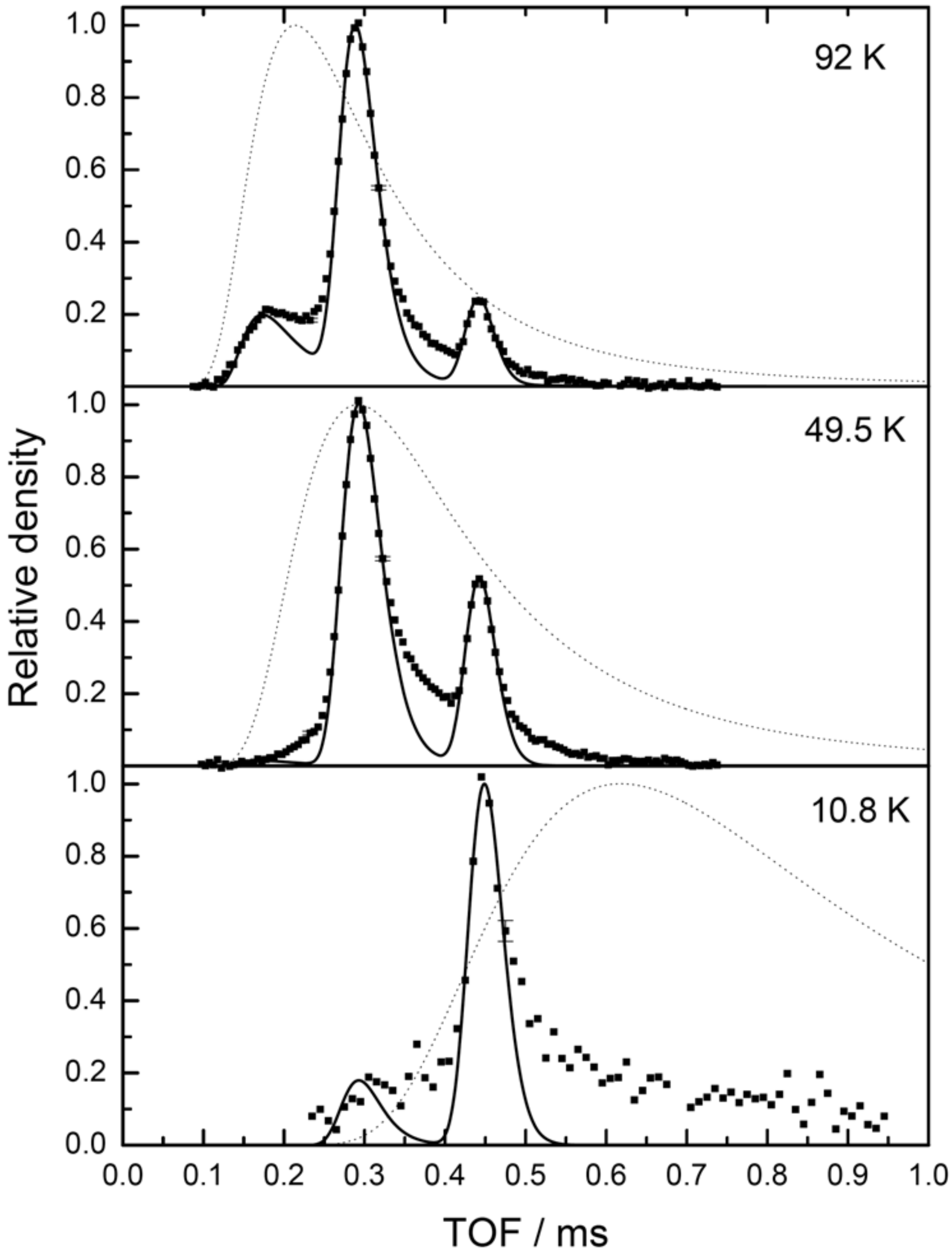
E_{kin}(H) or T_c?
direct collision?

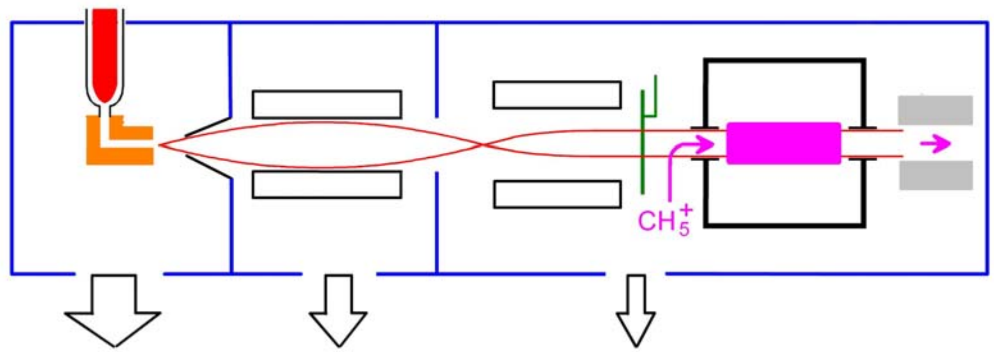
proton affinity
of methane



blue: O. Asvany, I. Savic, S. Schlemmer, and D. Gerlich, Chem. Phys. **298** (2004) 97
blue T>300K: Federer et al, Molecular Astrophysics (1985)
red: G. Borodi, A. Luca, D. Gerlich, in preparation

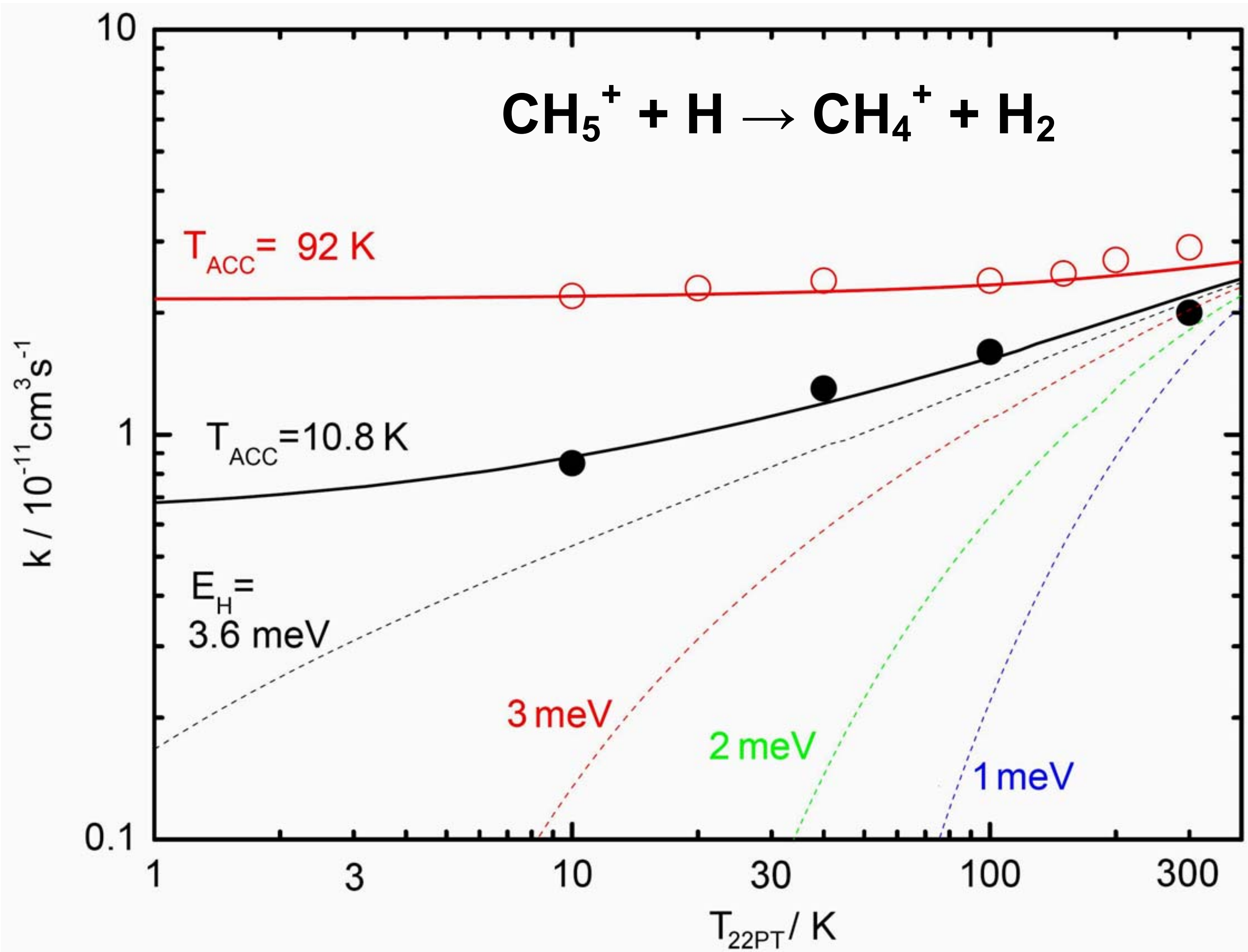
Focusing H atoms





Specific rate coefficients $k(T_{22PT}, E_t)$

$$k = k_0 \left((E_t + \alpha\, kT_{22PT} - E_0)/E_t \right)^{1/2}$$



E_0 =
(3.5 ± 0.1) meV

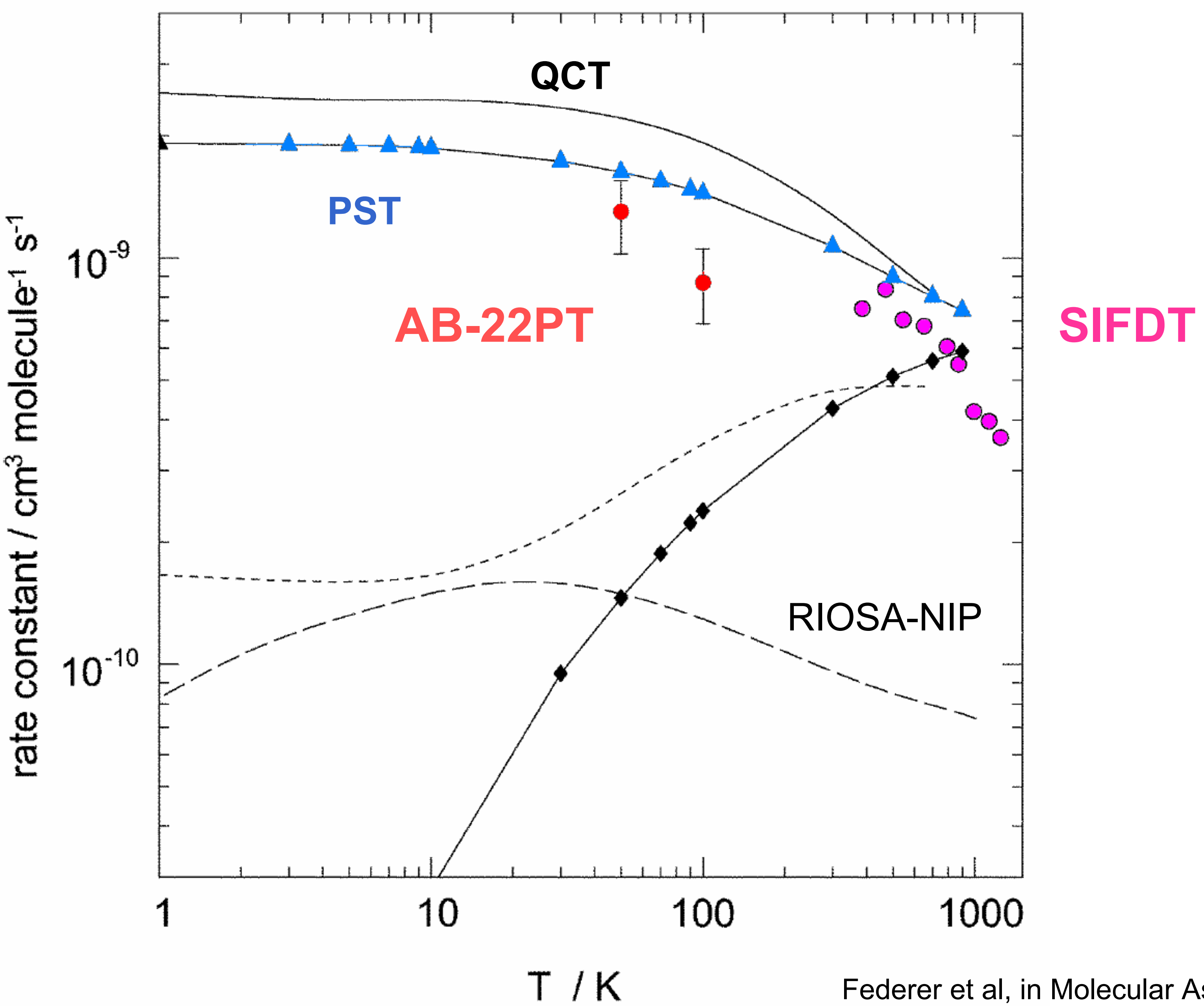
α =
0.1 ± 0.1

k_0 =
(3.2± 0.2) ×10^{-11} cm^3s^{-1}

$$k_{eff}(E_H, T_{22PT}) = \int f^*(g; E_H, T_{22PT})\, g\, \sigma(g;\, T_{22PT})\, dg$$

G. Borodi, A. Luca, C. Mogo, D. Gerlich
Collisions of cold trapped CH_5^+ ions with slow hydrogen atoms, (2008)

$CH^+ + H \rightarrow C^+ + H_2 + 0.398$ eV



Federer et al, in Molecular Astrophysics 1985
Chesnavitch et al, AJ 1984
Stoecklin and Halwick, PCCP 2005
Halvick et.al. PCCP 2007

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FG Laboratory Astrophysics 2000 - 2006

DFG research unit 388

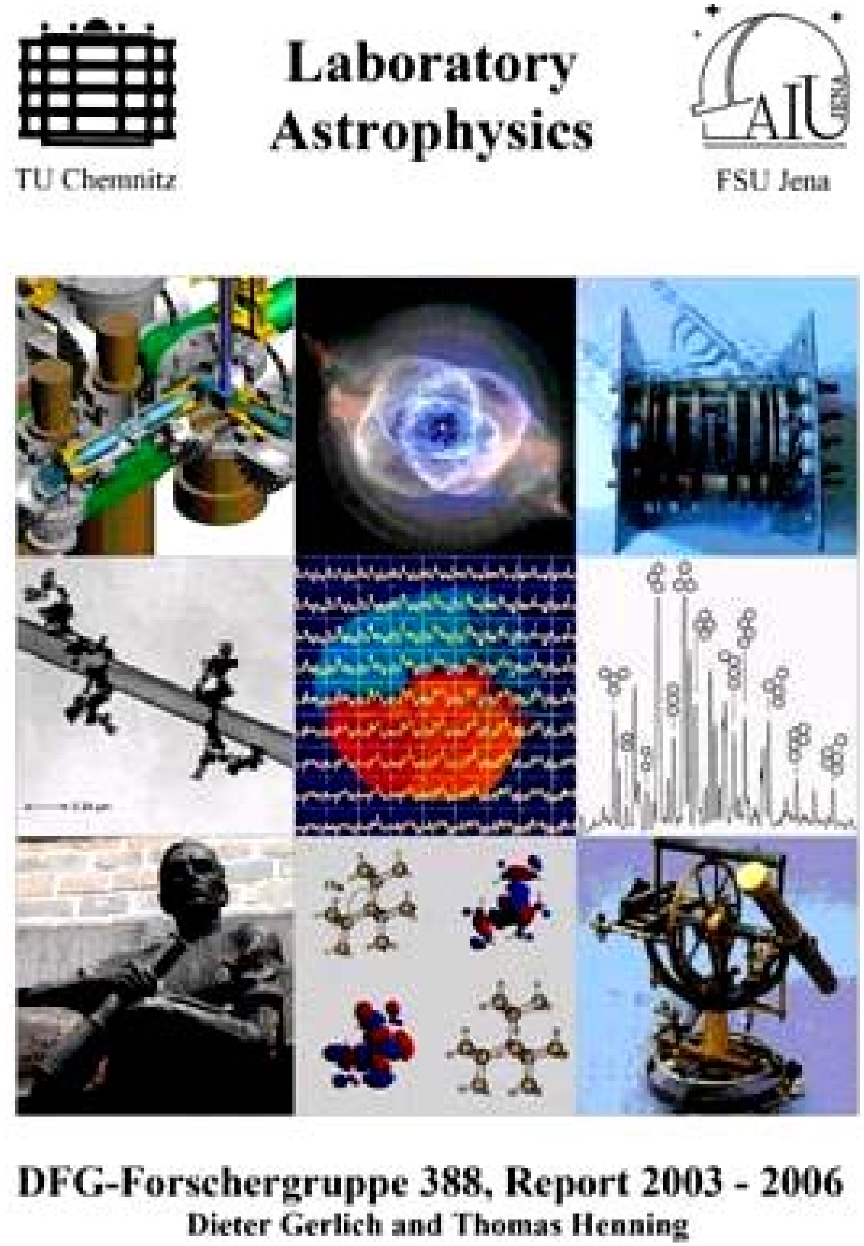
Deutsche
Forschungsgemeinschaft
DFG

Structure, dynamics and properties of molecules and grains in space

Sprecher: Prof. Dieter Gerlich, TU Chemnitz

From 2000 to 2006 the Deutsche Forschungsgemeinschaft has supported the Forschergruppe ***FGLA***.

Final report (Jan. 2007) see:
http://www.tu-chemnitz.de/FGLA



TECHNISCHE UNIVERSITÄT CHEMNITZ
FAKULTÄT FÜR NATURWISSENSCHAFTEN

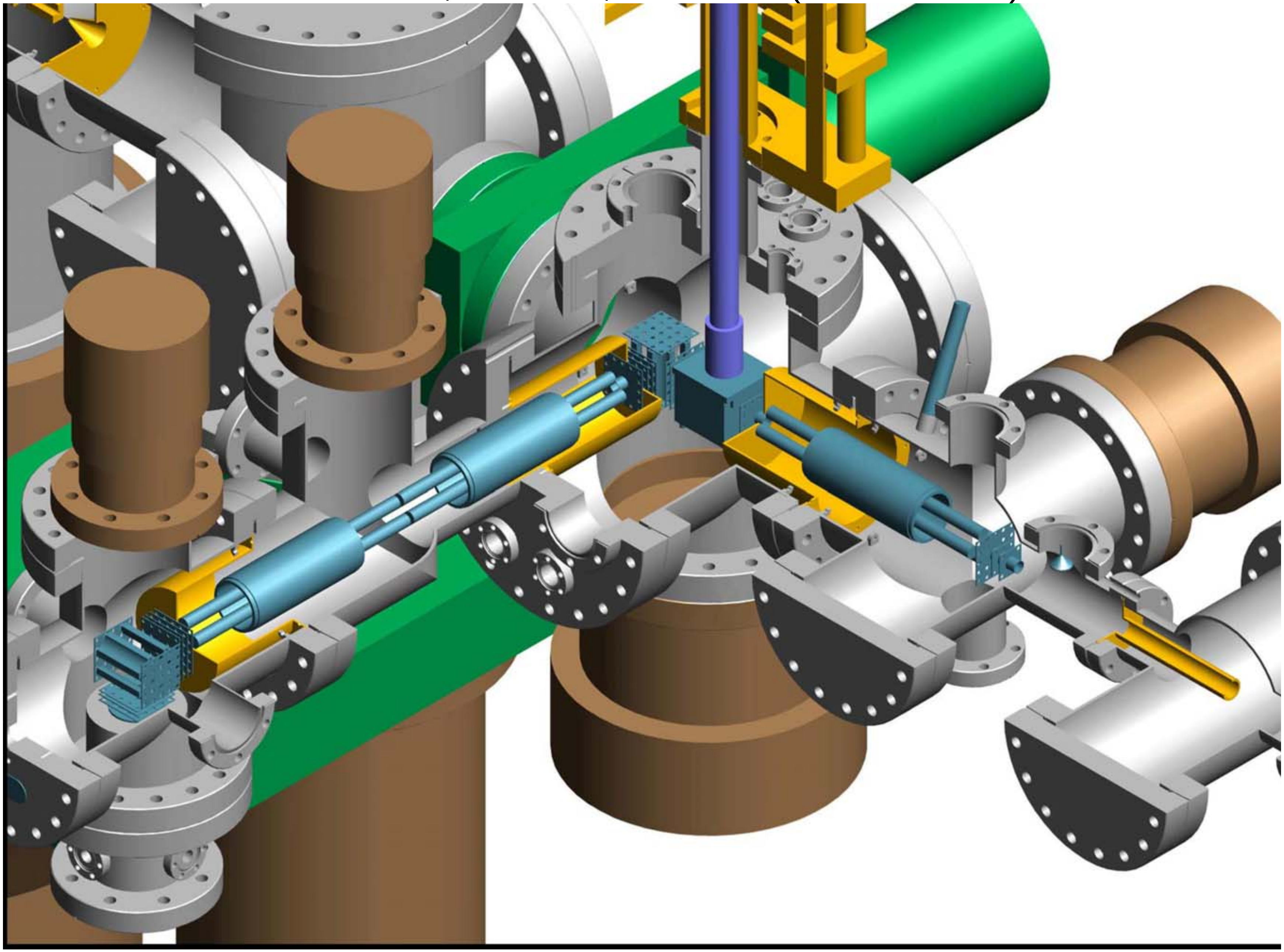
Gasentladungs- und Ionenphysik

DFG FG *Laboratory Astrophysics*

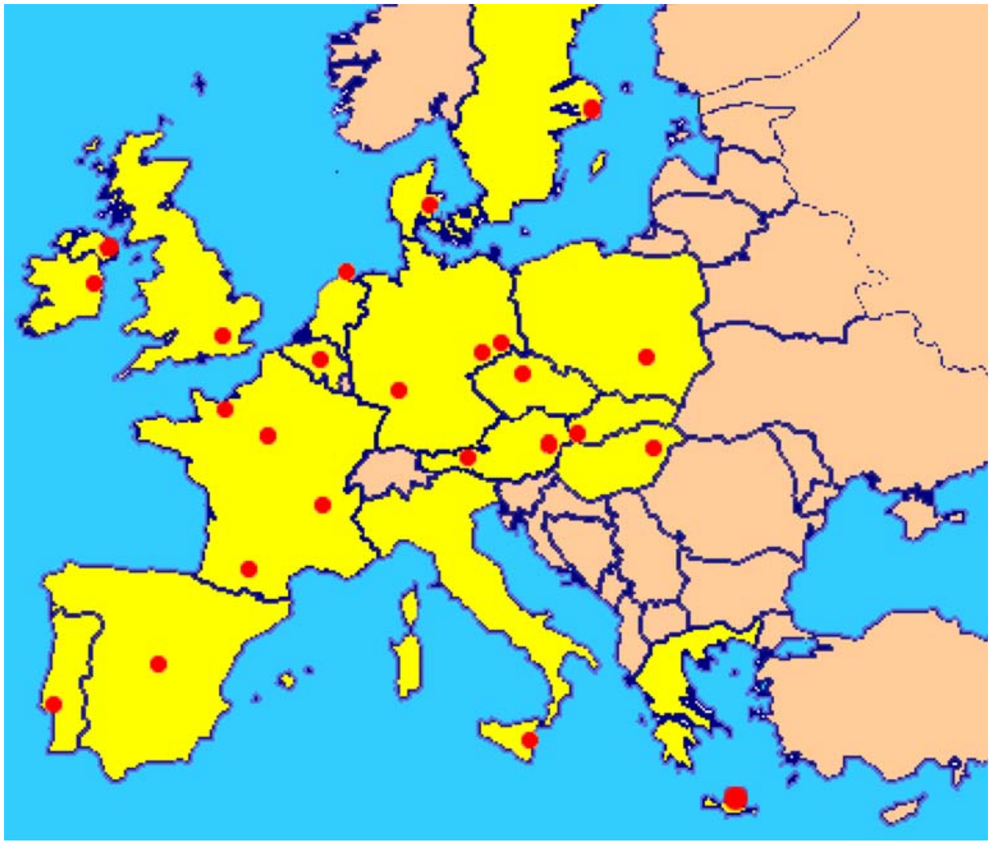
AB-22PT + H-beam	**A. Luca,** G. Borodi, C. Mogo
Black body radiation	**S. Decker**
RET + C_n-beam	I. Savic, S. Decker, I. Cermak
4K-22PT	J. Glosik, R. Plasil, F. Windisch
22PT-spectroscopy	J. Maier, Basel
Cold $TrpH^+$, $TyrH^+$	T. Rizzo, O. Boyarkin
Beam-Trap astrochemistry	**M. Smith**
22PT-TSR $H_3^+(J,K) + e^-$	D. Zajfmann, A. Wolf, H. Krekel, TSR HD
TV-22PT	**S. Schlemmer,** O. Asvany **(Köln)**

New Temperature Variable Ion Trap for Chemical Research

G Tikhonov, M. Smith, D. Gerlich (NSF CRIF ID)



ITS LEIF: Ion Technology and Spectroscopy at Low Energy Ion beam Facilities

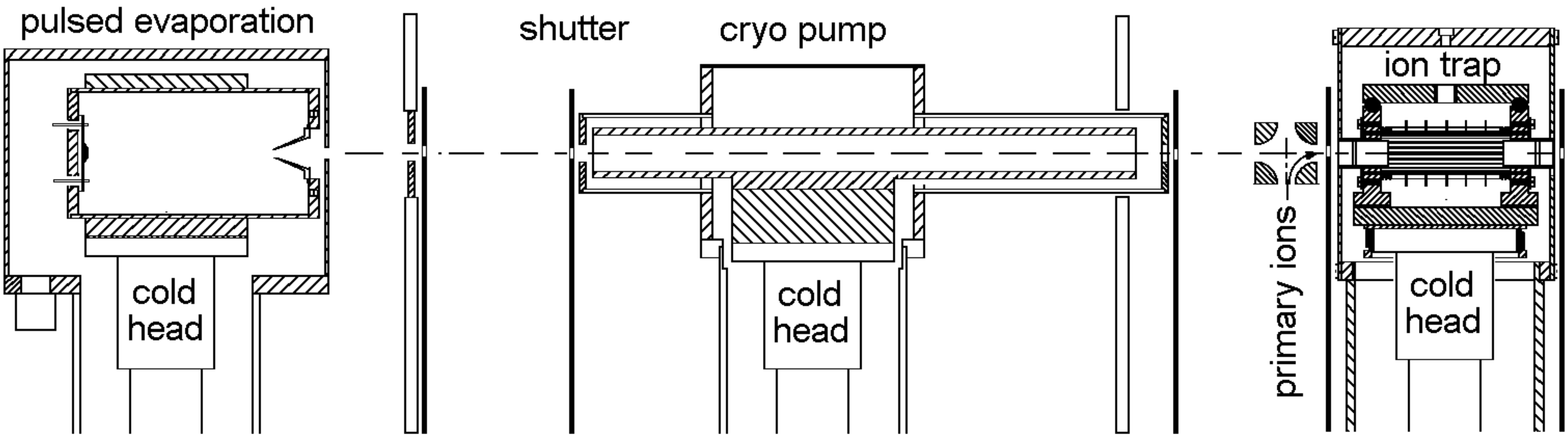


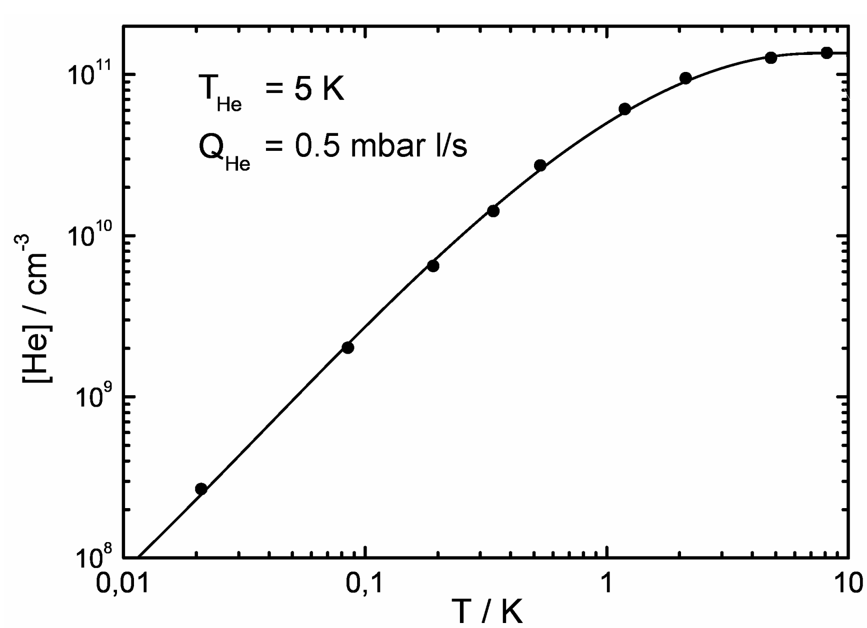
TU Chemnitz:

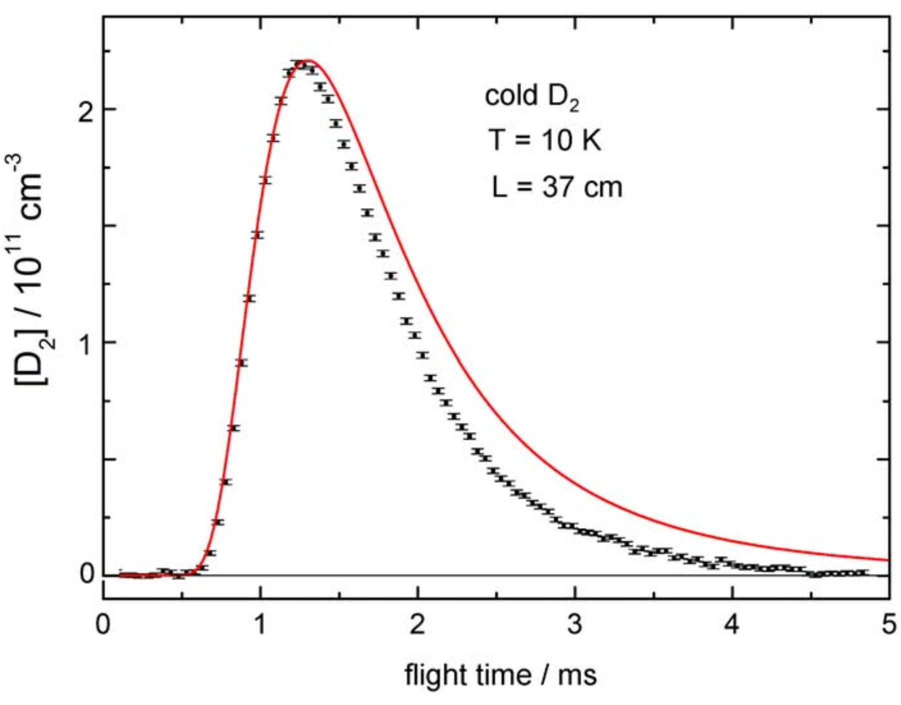
- production of brilliant beams of cold molecular ions, including biomolecules
- extending the unique sensitivity and versatility of rf multipole ion traps for in situ experiments (spectroscopy, photostability, chemical reactions, astrochemistry).
- characterizing the instrument as ion source
- Injecting externally created ions (e.g. from an ESI source)
- synthesizing ions in situ
- test object H3+ and deuterated variants
- new method for getting ultracold ions (sub-K domain, presently 5 K are reached)

closing the gap between "ultra cold molecules" (= laser cooled diatoms) and cold chemistry: s-wave reactions, state specific cross sections, role of nuclear spin, non rotating large molecules

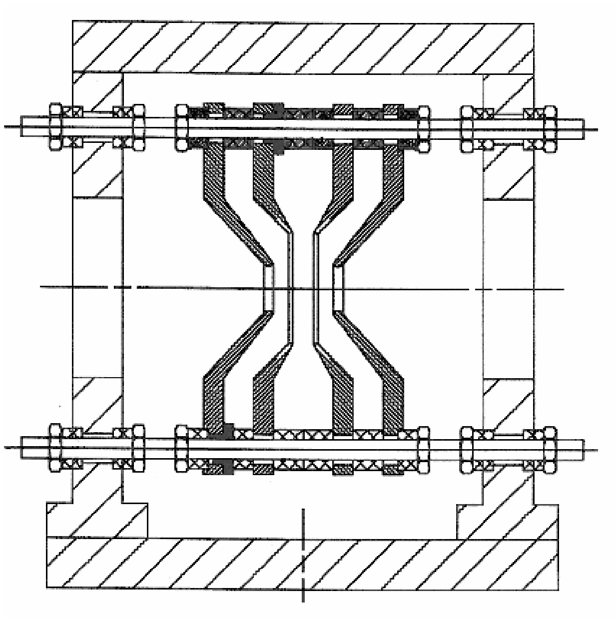
Sub-K cooling of stored ions

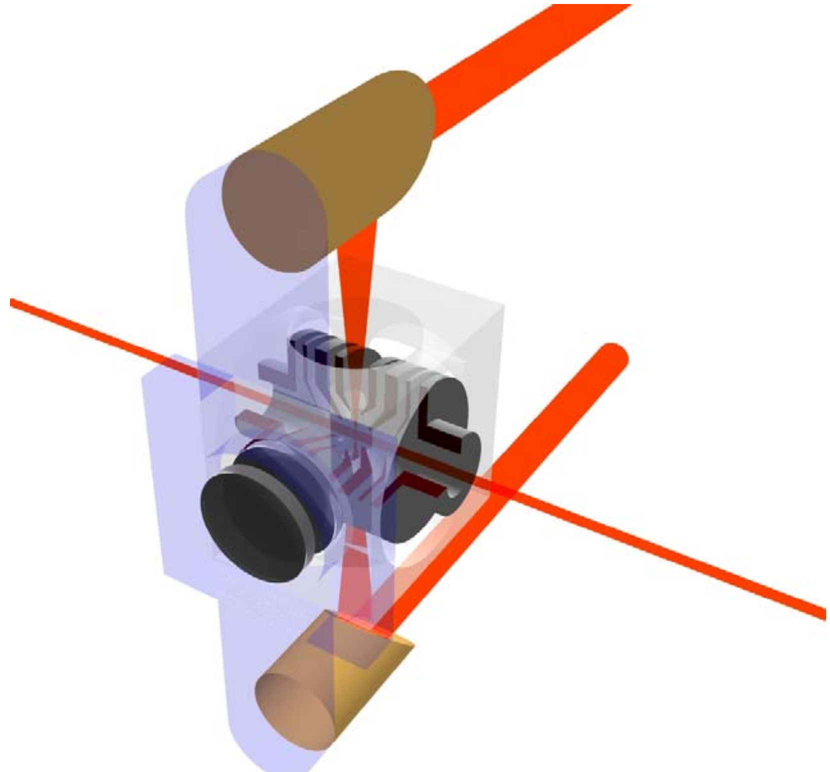






Heating stored C_{60}^{+} with a cw CO_2 laser





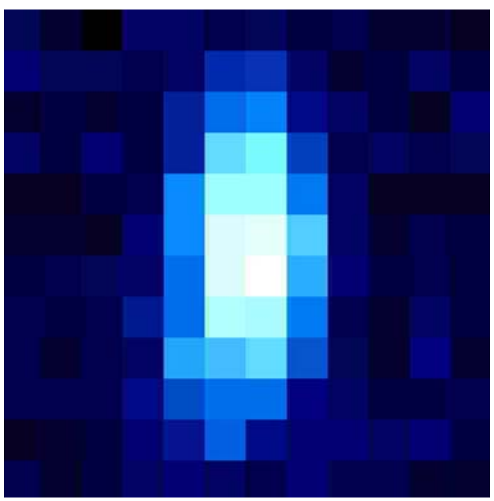


Image of 100 000 ions

100 × 150 µm (magnification ×6)
exposure 5 s
trapping time several min
T ~ 2300 ± 300
no decay
Laser power 20 W
Focus 50 µm
He buffer gas 1 $\times 10^{-4}$ mbar

Summary / perspectives of NPMS

- single particle (time average)
- non-destructive, absolute mass and charge determination
- long time trapping, isolation under UHV conditions
- high resolution of secular frequencies ($\Delta\nu/\nu < 10^{-6}$)

- experimental characterization of the trap, new trap design
  accuracy, precision, linearity, anharmonicity of potential
- small particles (1- 5 nm)
- optical detection (spectroscopy, light pressure,...)
- chemistry, agglomerates, magnetic properties
- temperature range: 5 K - 3000 K

Black body radiation of
carbonaceous material