Universitas Carolina
Charles University in Prague

Polyatomic ions in traps: from molecules via clusters to nanoparticles

Dieter Gerlich

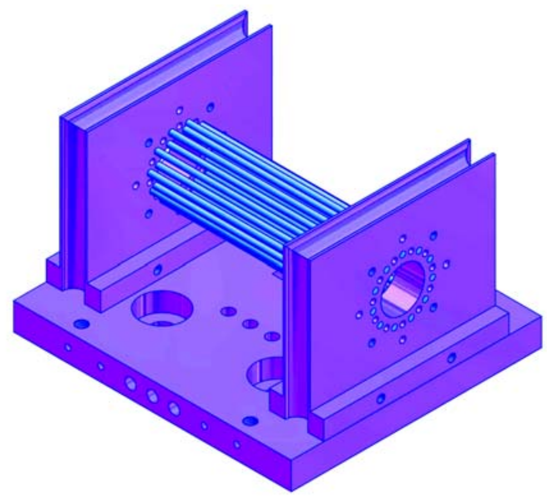
Introduction

Ions in rf fields

Basics, buffer gas cooling

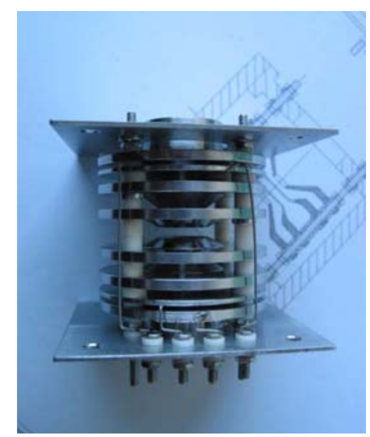
Typical tests

Spectroscopy
Association reactions, cluster
LIR: N_2^+ + Ar



Recent application

Reactions with H atoms
Deuteration, nuclear spin
State selective preparation



Nanoparticles

NPMS
HT - SRET, Decay of C_{60}^+

Summary and outlook

TECHNISCHE UNIVERSITÄT CHEMNITZ
FAKULTÄT FÜR NATURWISSENSCHAFTEN

Gasentladungs- und Ionenphysik

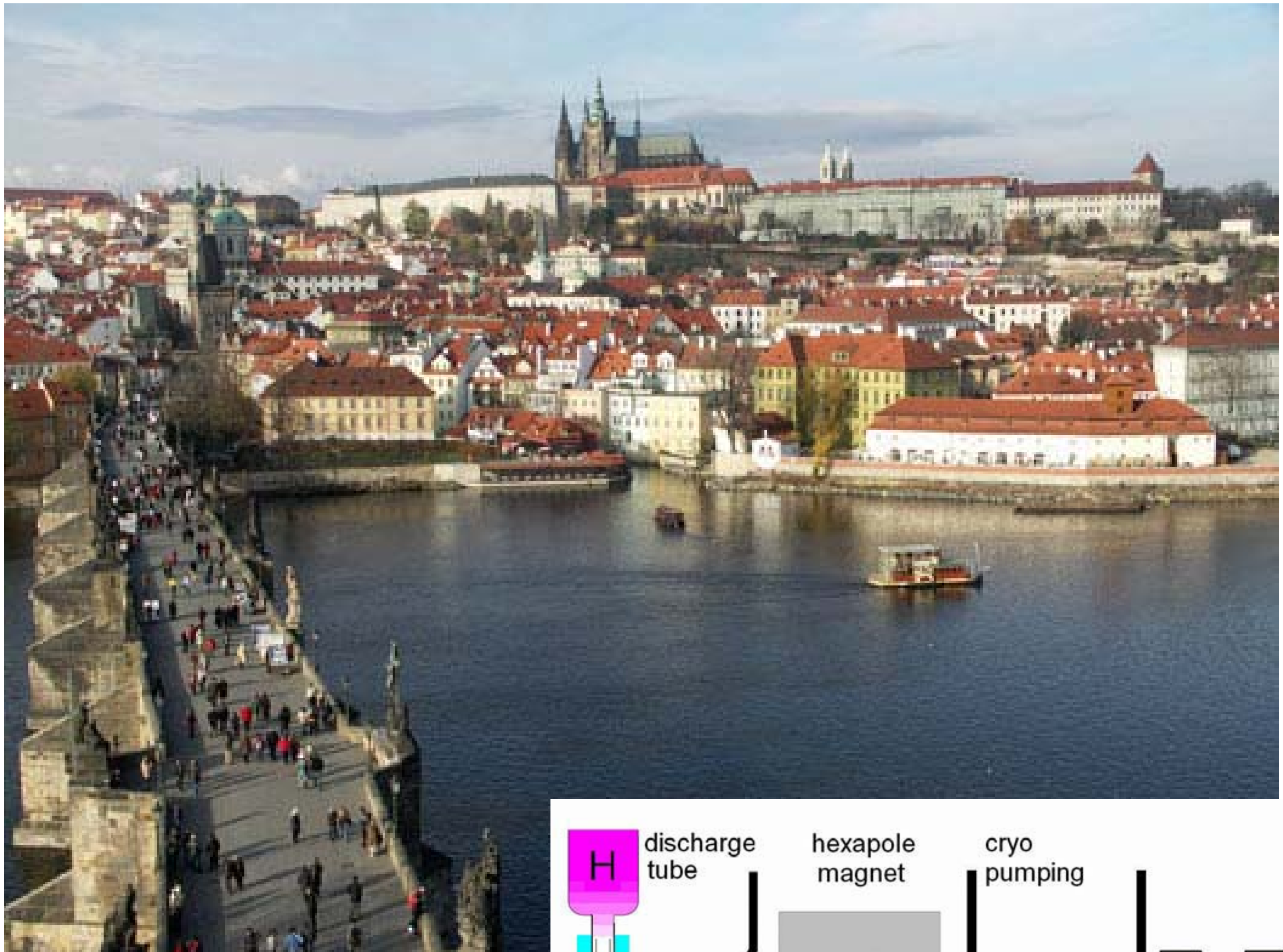
DFG FG *Laboratory Astrophysics*

1993 - 2009

TV-22PT	S. Schlemmer, O. Asvany (Köln)
AB-22PT + H-beam	A. Schlemmer, A. Luca, G. Borodi **J. Glosik, R. Plasil,** C. Mogo
RET + C_n-beam	I. Savic, S. Decker, I. Cermak
Black body radiation	S. Decker, M. Kämpf
Beam-Trap **Astrochemistry**	M. Smith — THE UNIVERSITY OF ARIZONA, TUCSON ARIZONA
22PT-spectroscopy	J. Maier, Basel
Cold $TrpH^+$, $TyrH^+$	T. Rizzo, O. Boyarkin

Transfer 22PT -> Prague

Charles University in Prague
Universitas Carolina

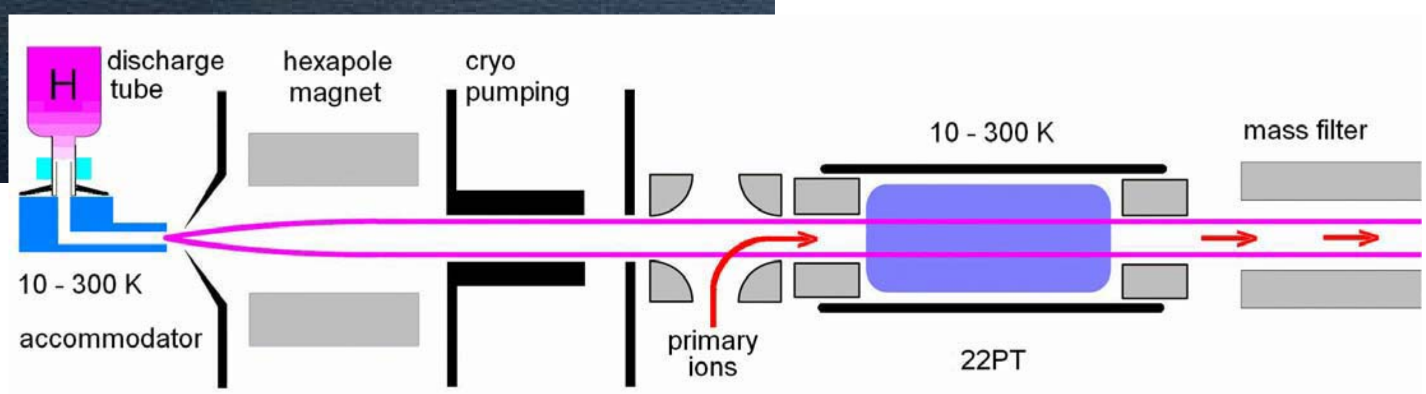


01.10.2009

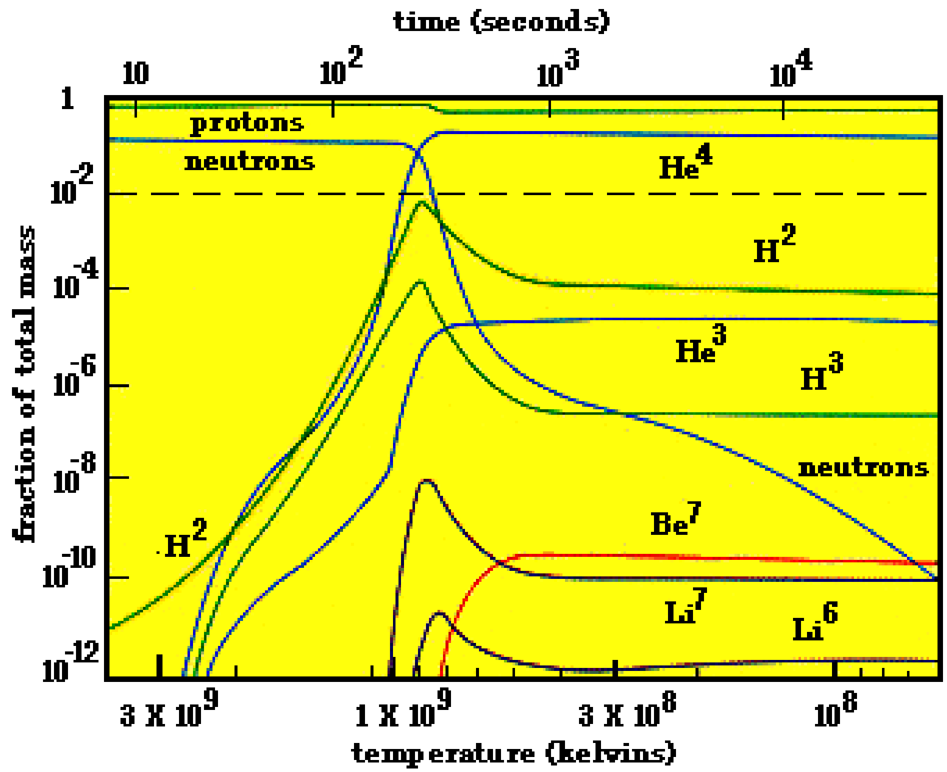
Reactions with H atoms

Neg ions H- + H

buffer gas cooling
ultracold ions: N_2^+



Big Bang Nucleosynthesis



< 1 s : n/p ~ 1

$p + e^{-} \longleftrightarrow n + \nu$

$n + e^{+} \longleftrightarrow p + \bar{\nu}$

> 1 s, $\tau_{1/2} \sim 615$ s

$n \longrightarrow p + e^{-} + \bar{\nu}$

100 s, n/p ~ 1/7

10^{9} K, kT ~ 0.1 MeV

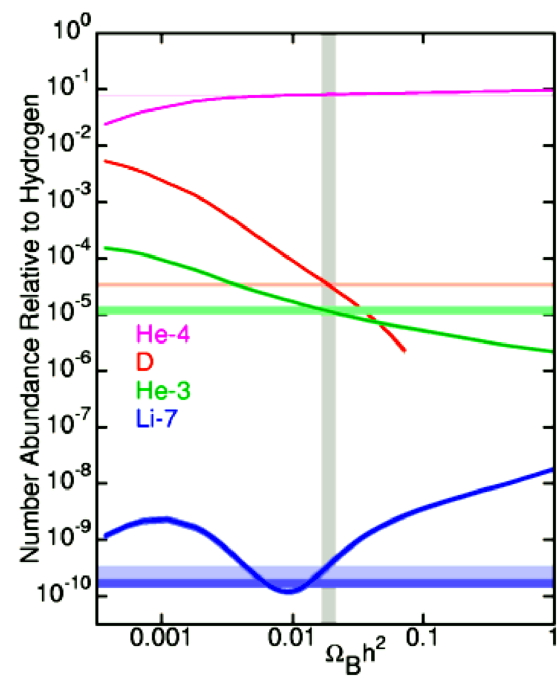
$p + n \longleftrightarrow d + \gamma + 2.2$ MeV

$d + n \longrightarrow H^{3} + \gamma$

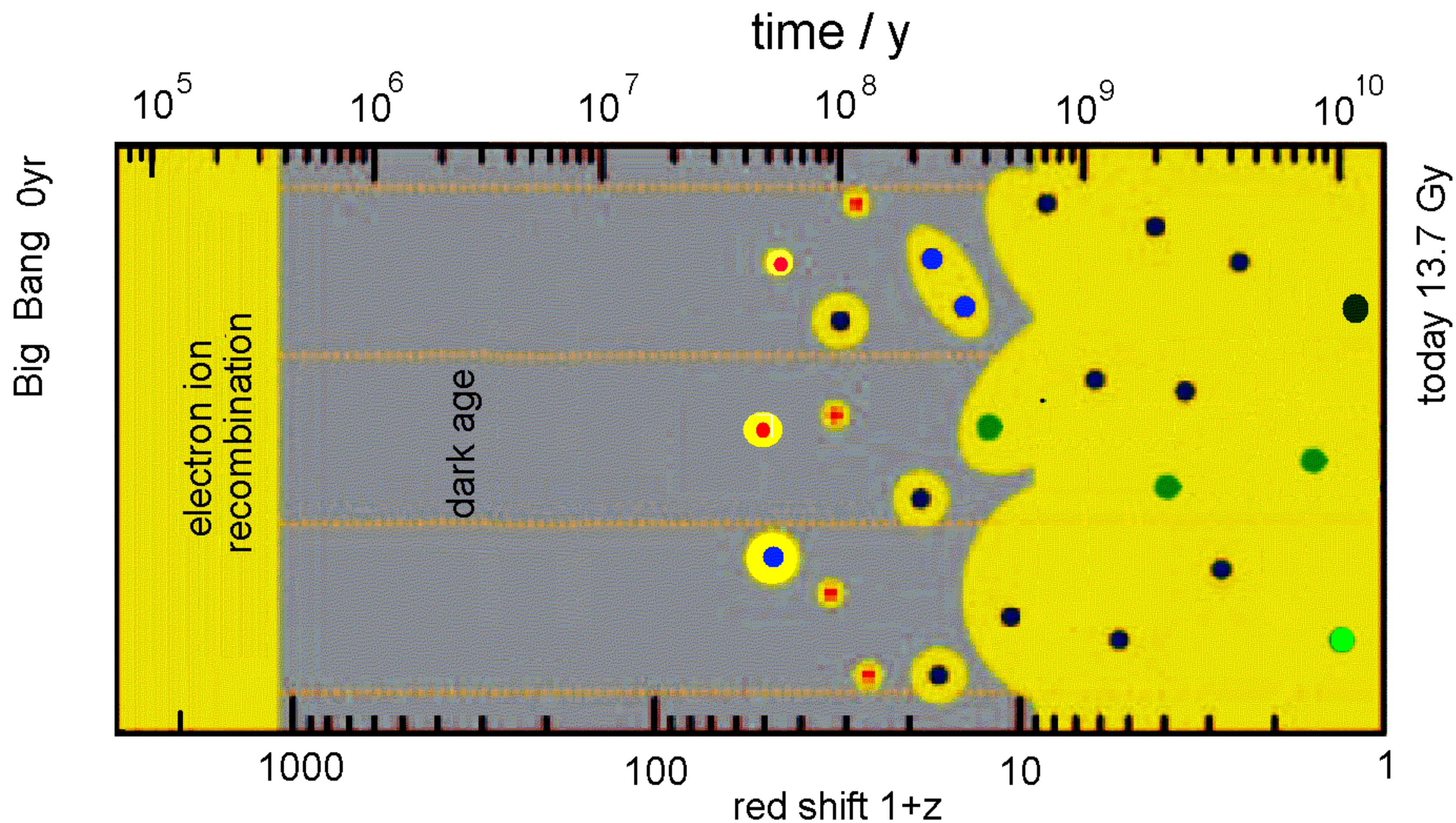
$H^{3} + p \longrightarrow He^{4} + \gamma$

$d + p \longrightarrow He^{3} + \gamma$

$He^{3} + n \longrightarrow He^{4} + \gamma$



Early universe chemistry: end of dark age

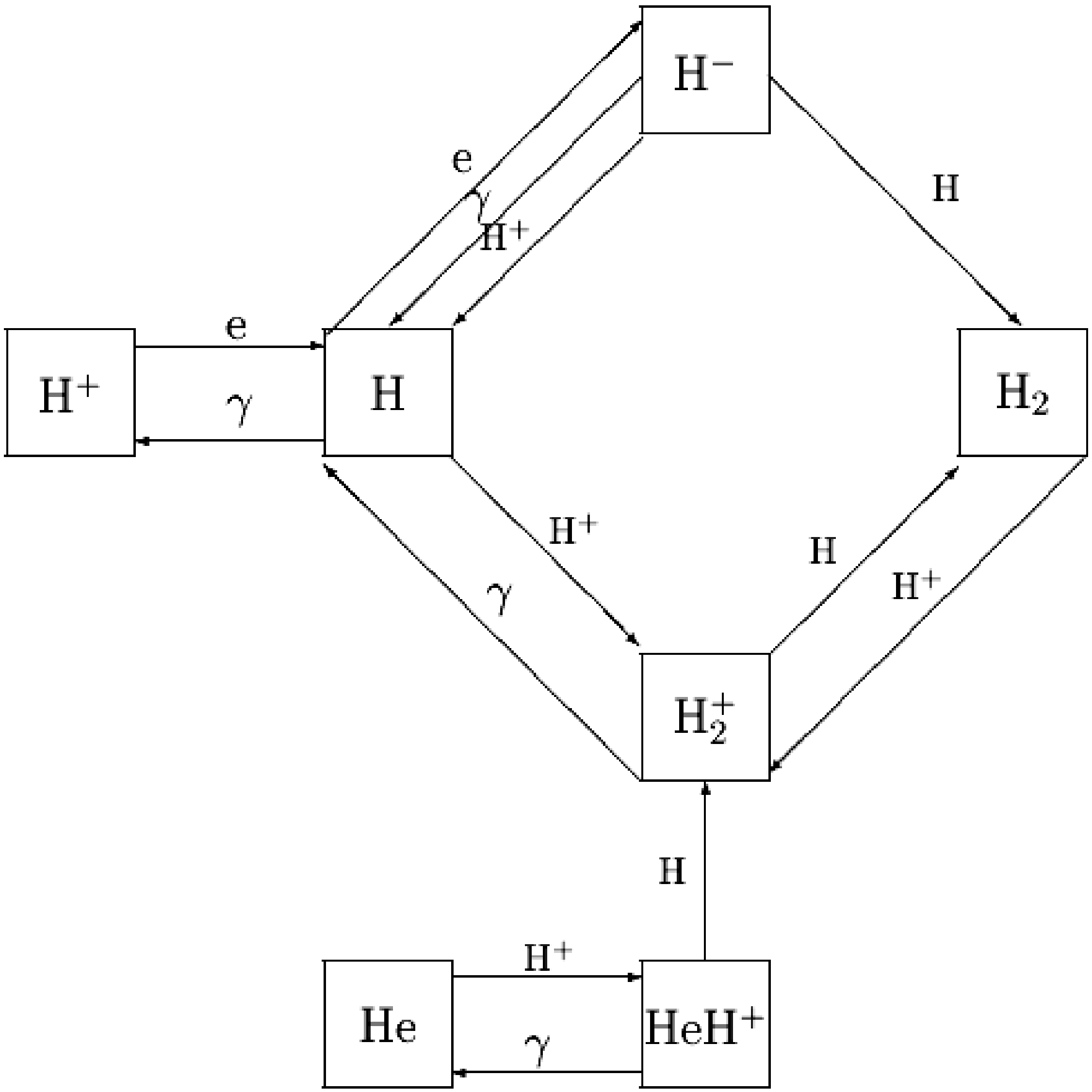


$$H^- + H \rightarrow H_2 + e^-$$

- important for cooling primordial clouds (8,000 - 200 K)
- uncertainties affect predictions for star formation

Volker Bromm, Physik Journal 7 (2008) Nr.4, 29

H_2 formation in the early Universe



radiative association
very slow
H + H $\rightarrow H_2 + h\nu$

early times
$H^+ + He \rightarrow HeH^+ + h\nu$
$HeH^+ + H \rightarrow H_2^+ \, He$
$H_2^+ + H \rightarrow H_2 + H^+$

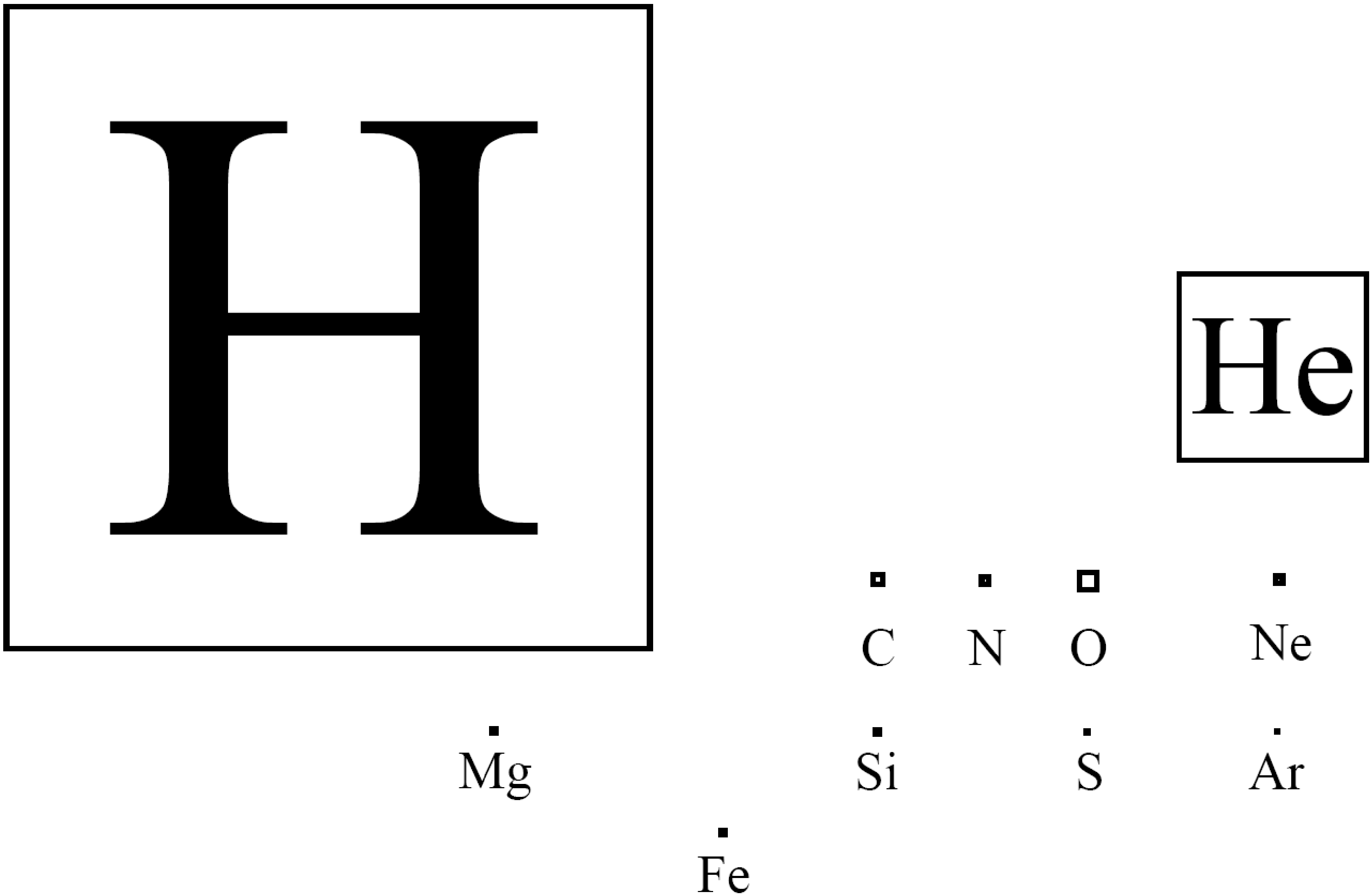
later times
$H^+ + H \rightarrow H_2^+ + h\nu$

$H^* + H \rightarrow H_2 + h\nu$
$\rightarrow H_2^+ + e^-$

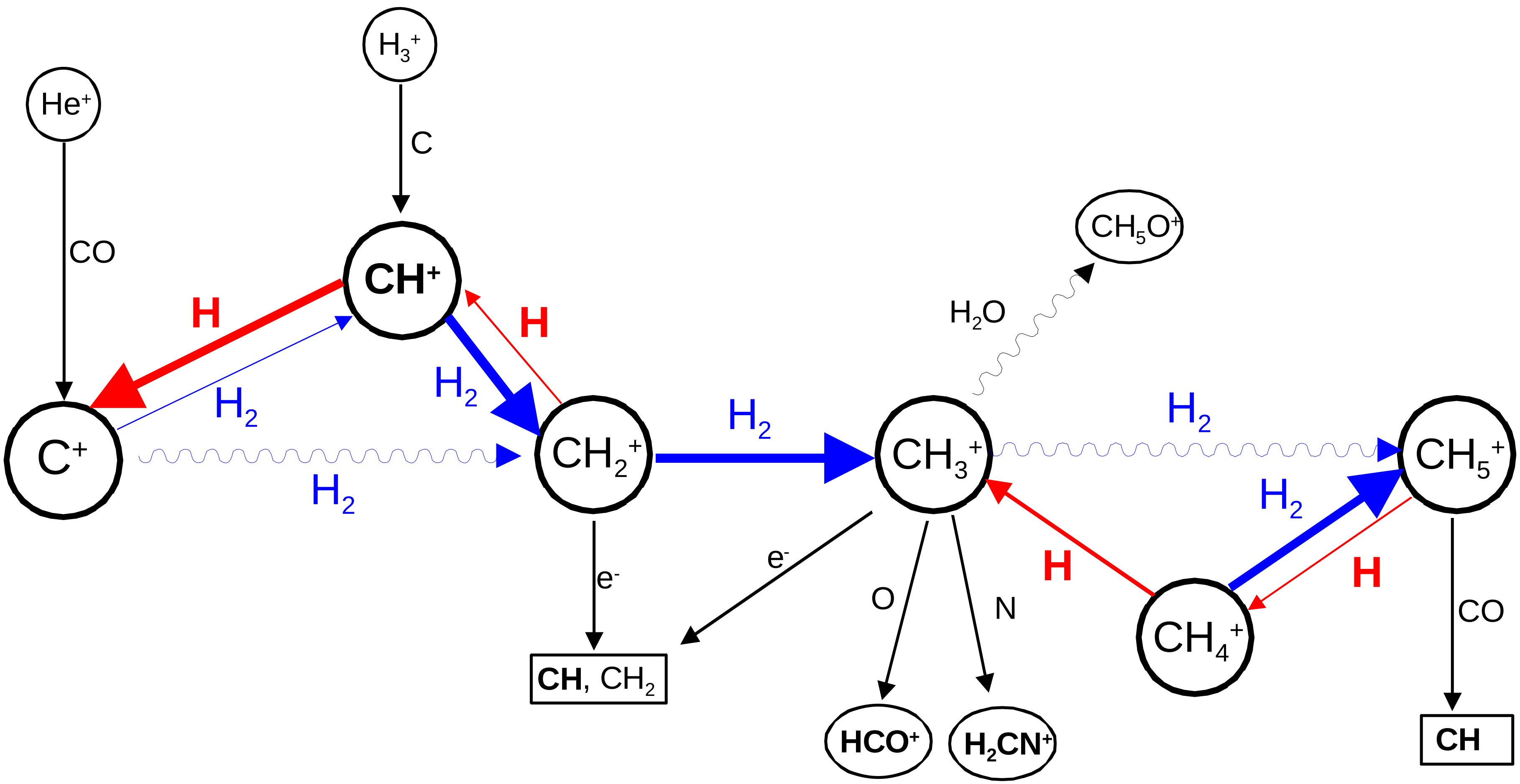
H^- channel
$H + e^- \rightarrow H^- + h\nu$
$H^- + H \rightarrow H_2 + e^-$

Lepp S., Stancil P.C., Dalgarno A., *J. Phys. B:* **35** (2002) R57

The periodic table of astronomers



Formation and destruction of CH_n^+ in space



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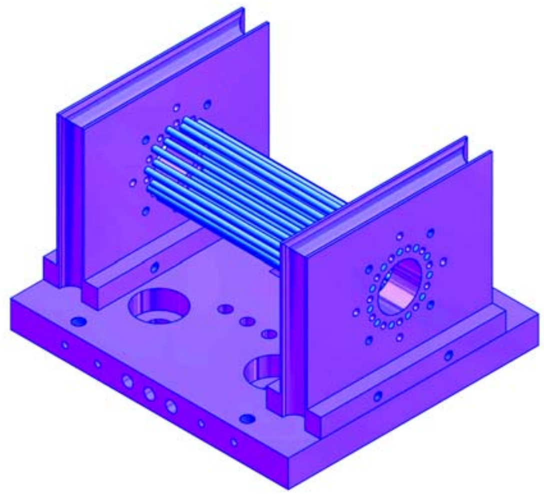
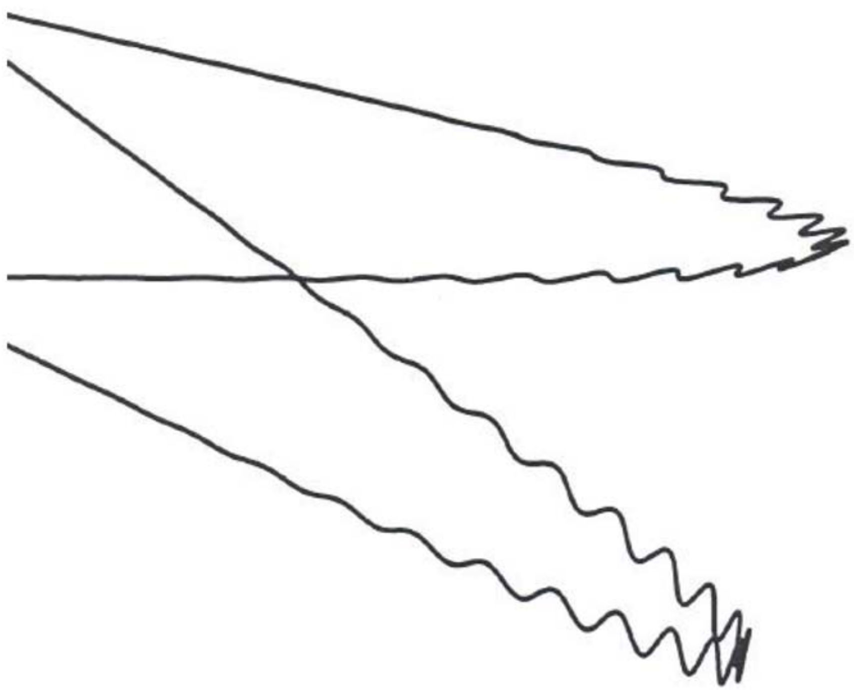


INHOMOGENEOUS RF FIELDS: A VERSATILE TOOL FOR THE STUDY OF PROCESSES WITH SLOW IONS

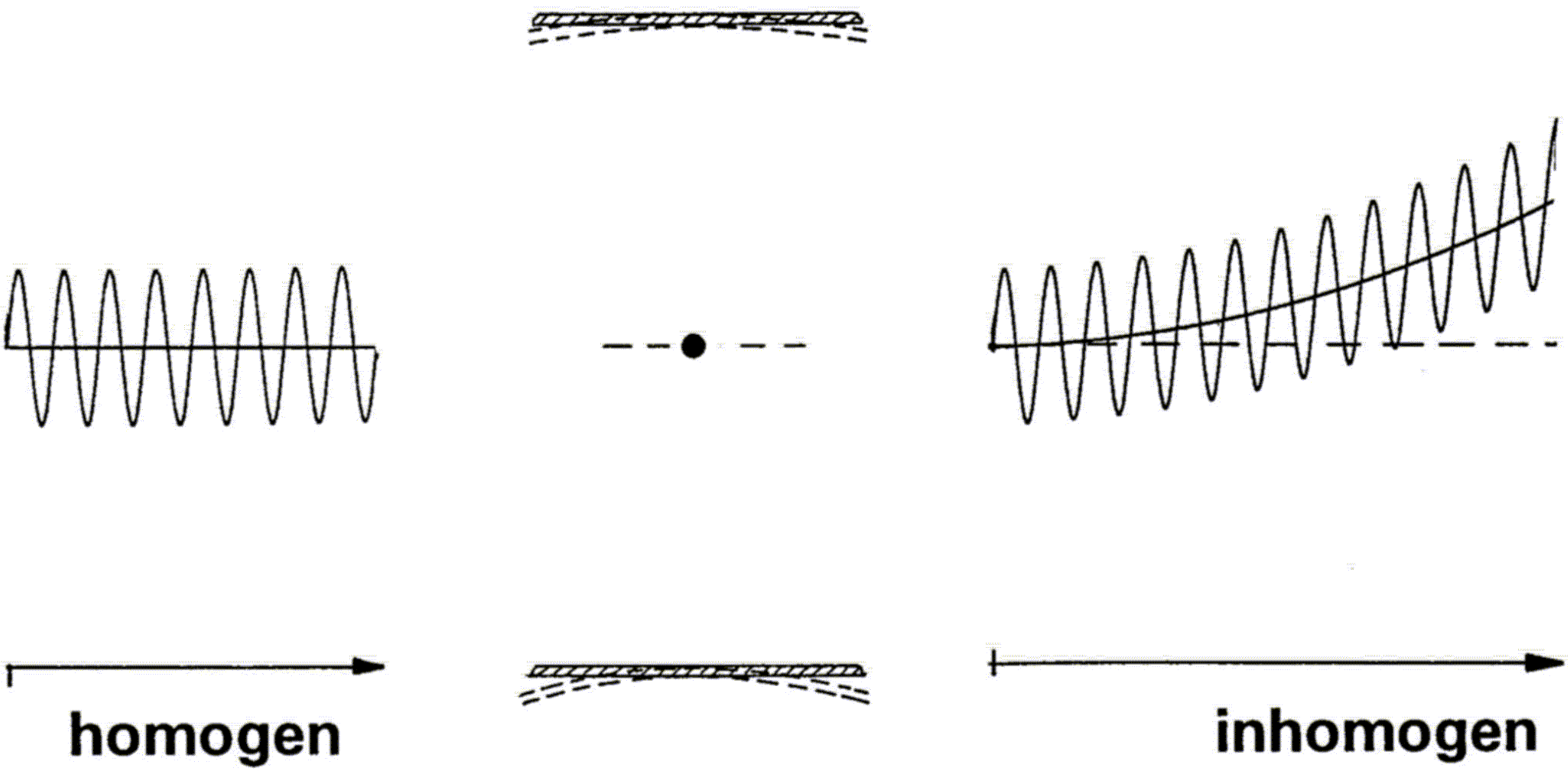
DIETER GERLICH



Advances in Chemical Physics, LXXXII, J. Wiley & Sons (1992)

Motion in a fast oscillating field

$$m\ddot{\mathbf{r}} = q\mathbf{E}_0(\mathbf{r})\cos(\Omega t + \delta) + q\mathbf{E}_s(\mathbf{r})$$



Kapitza 1951, Landau-Lifschitz Classical Mechanics 1962

Motion in a fast oscillating field

$$\mathbf{r}(t) = \mathbf{r}(0) - \mathbf{a}\cos(\Omega t)$$

$$\mathbf{a} = q\mathbf{E}_0/m\Omega^2$$

$$\mathbf{r}(t) = \mathbf{R}_0(t) + \mathbf{R}_1(t)$$

$$\mathbf{R}_1(t) = -\mathbf{a}(t)\cos\Omega t$$

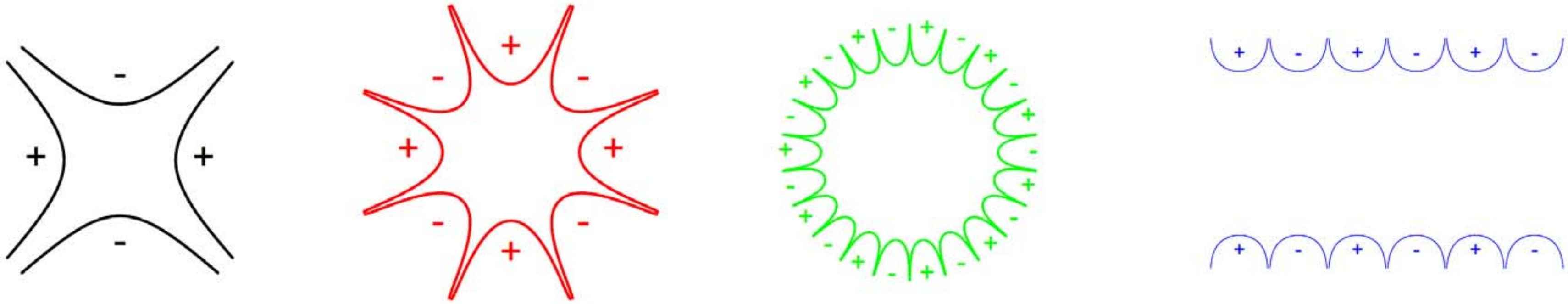
$$\mathbf{E}_0(\mathbf{R}_0 - \mathbf{a}\cos\Omega t) = \mathbf{E}_0(\mathbf{R}_0) - (\mathbf{a}\cdot\nabla)\mathbf{E}_0(\mathbf{R}_0)\cos\Omega t + \cdots$$

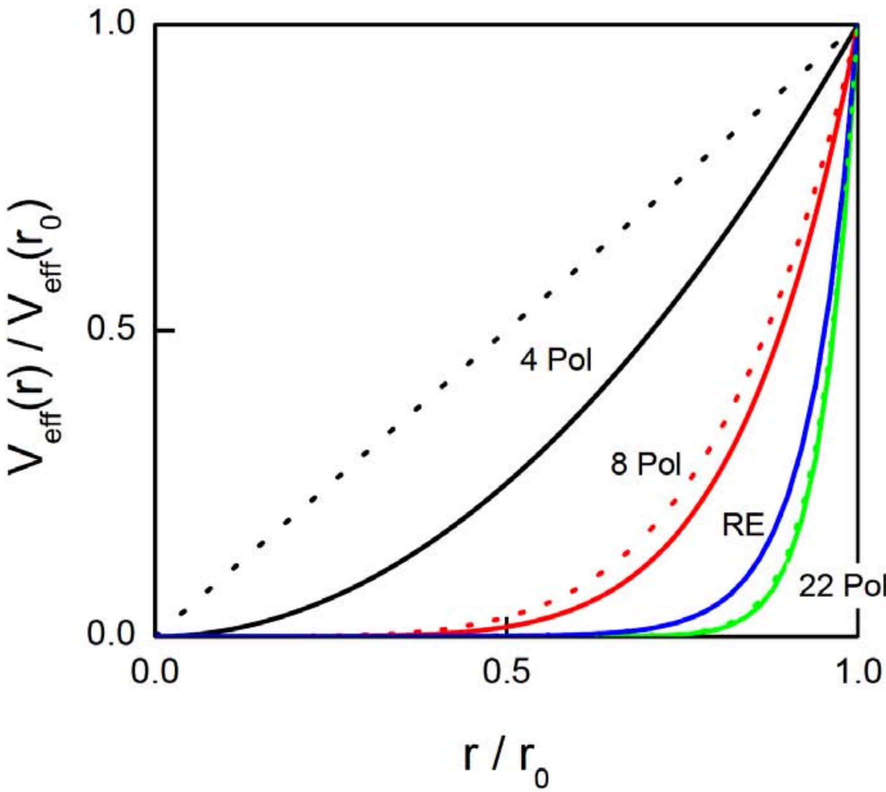
effective potential $V^*(\mathbf{R}_0) = q^2 E_0^2/4m\Omega^2$

adiabaticity parameter $\eta = 2q|\nabla E_0|/m\Omega^2$

Kapitza 1951, Landau-Lifschitz Classical Mechanics 1962

Ion trapping in rf fields





Effective Potential:

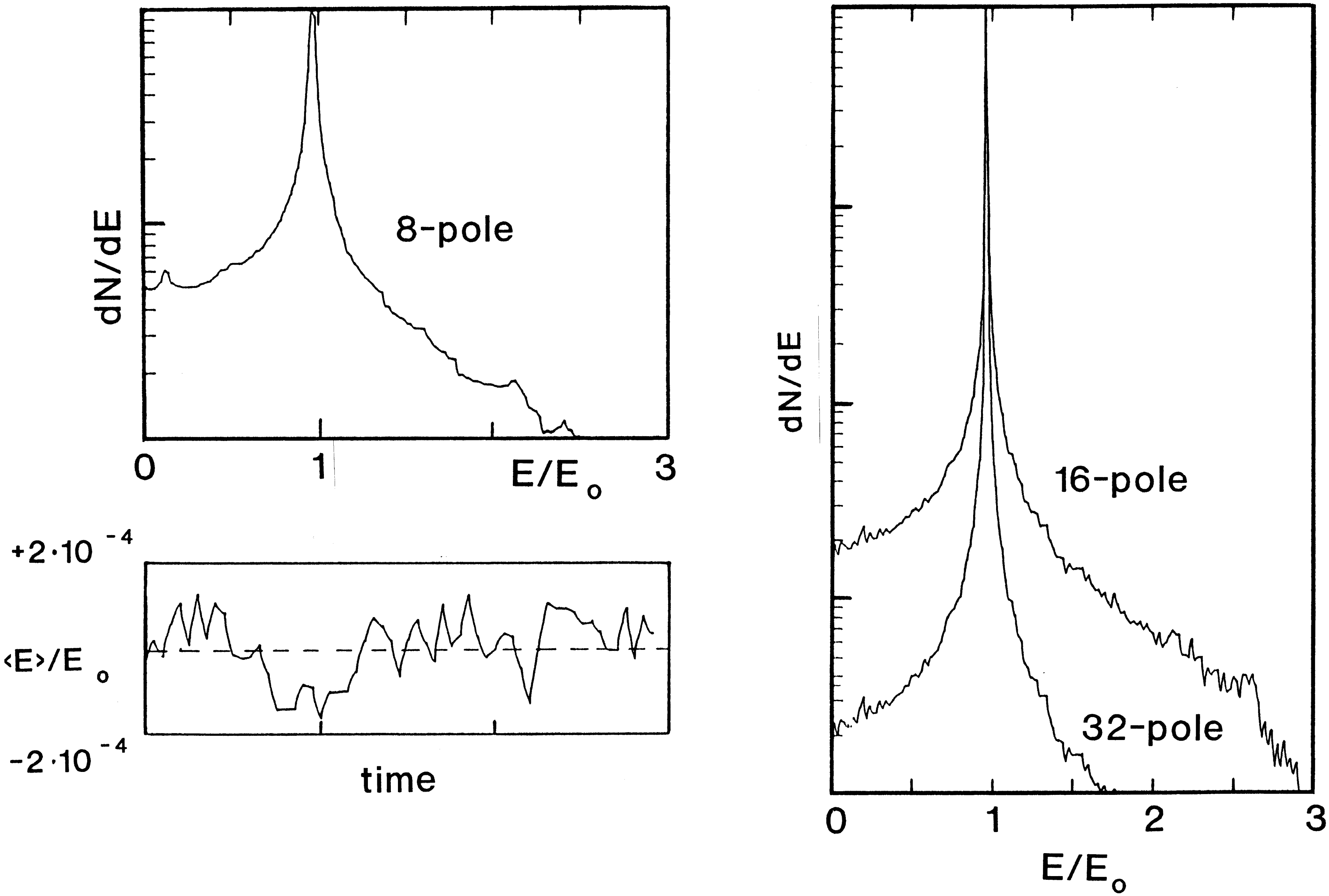
$$V_{eff} = \frac{q^2 E_0^2}{4m\Omega^2} \qquad \eta = \frac{2q\left|\nabla E_0\right|}{m\Omega^2}$$

2n-Pole:

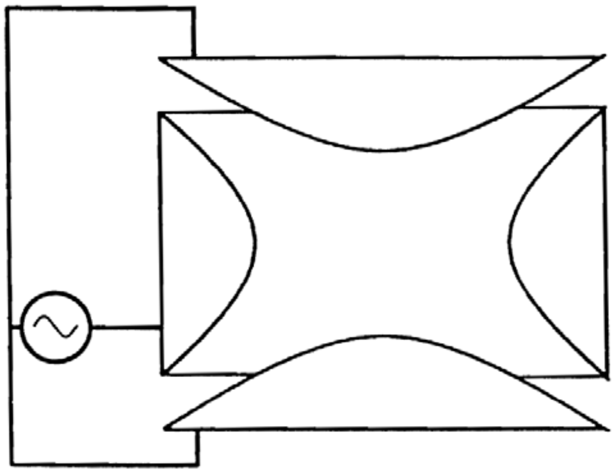
$$V_{eff} = \frac{n^2 q^2 V_0^2}{4m\Omega^2 r_0^2}\left(\frac{r}{r_0}\right)^{2n-2}$$

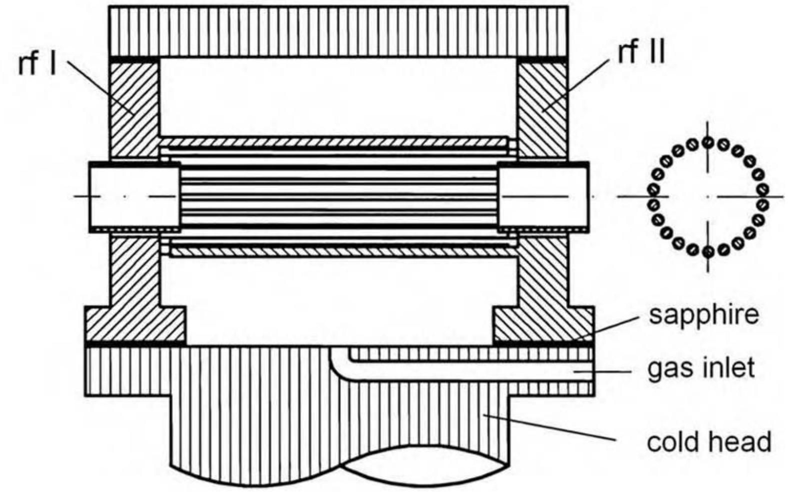
$$\eta = 2n(n-1)\frac{qV_0}{m\Omega^2 r_0^2}\left(\frac{r}{r_0}\right)^{n-2}$$

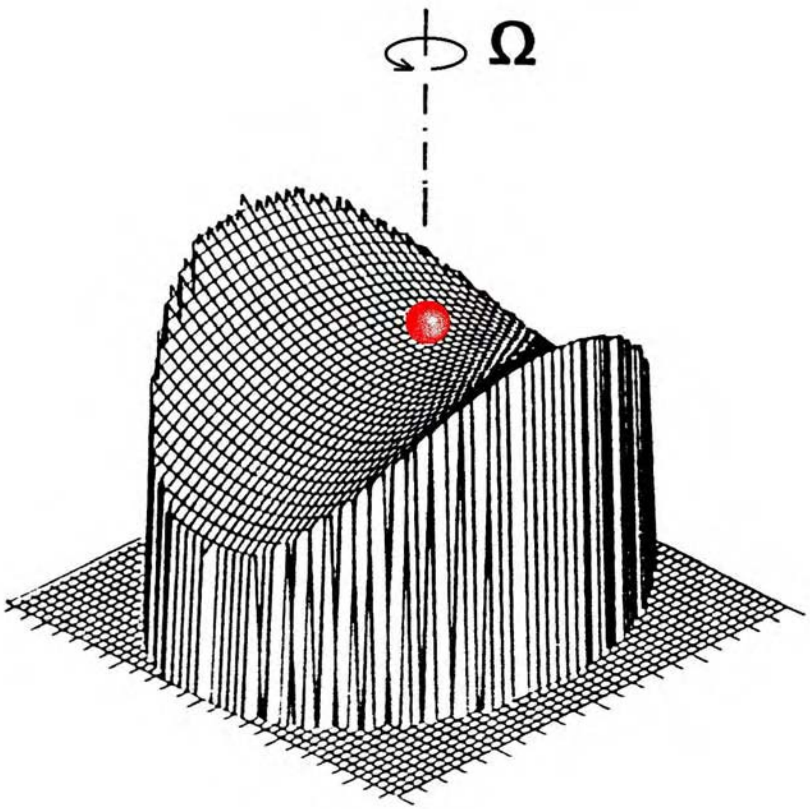
Kinetic energy distribution

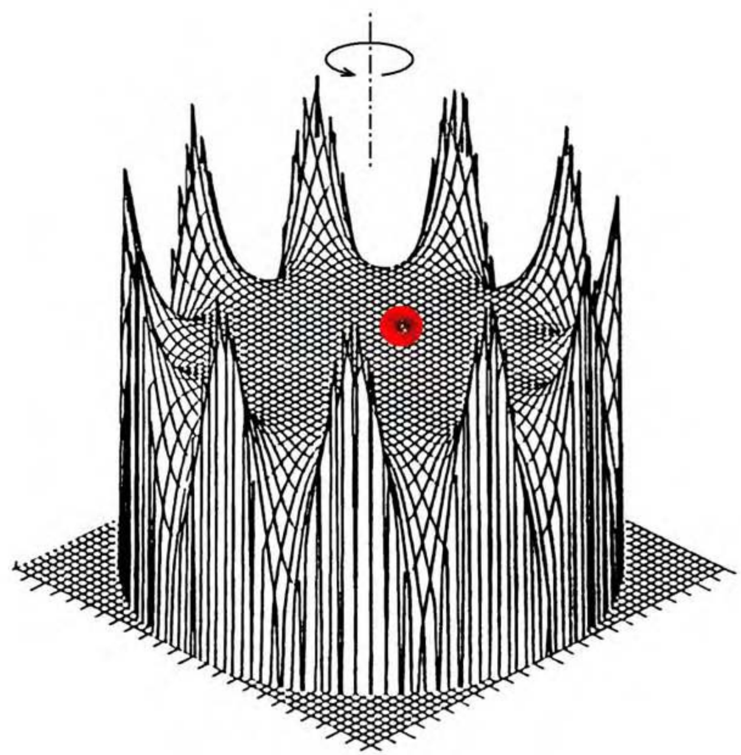


Confinement of charged particles in rf or AC fields

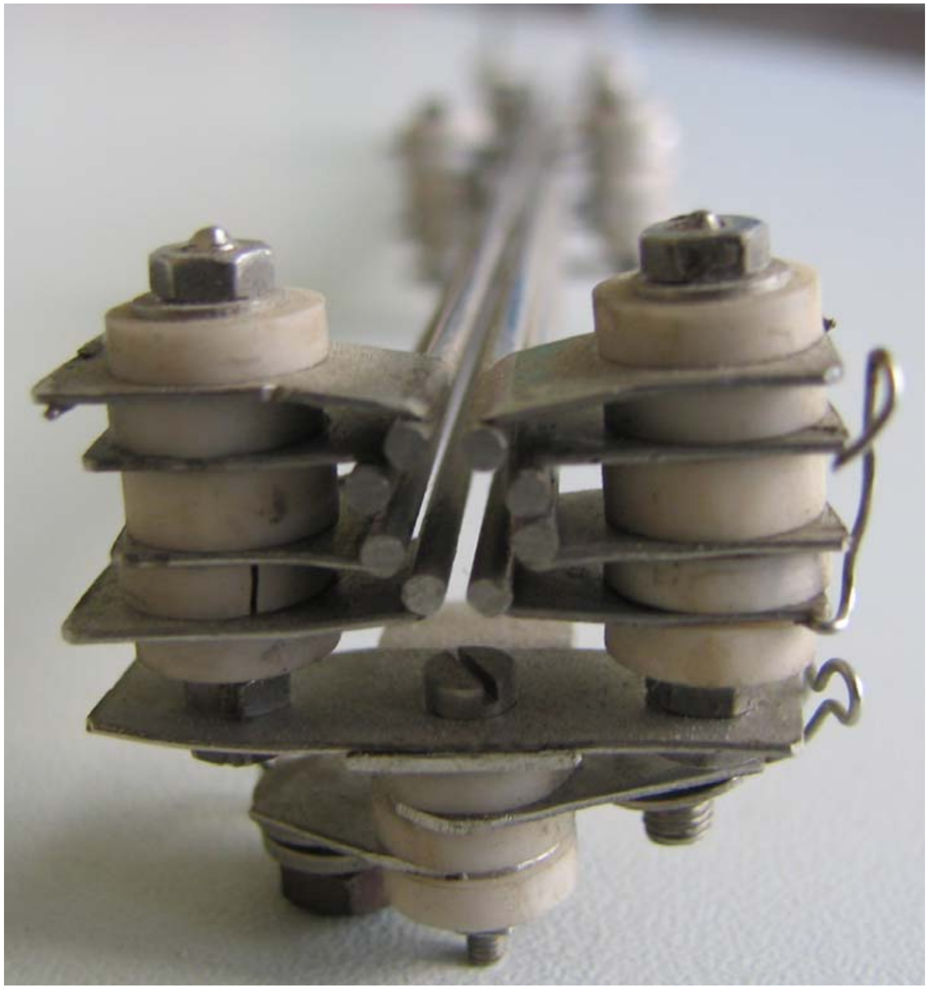




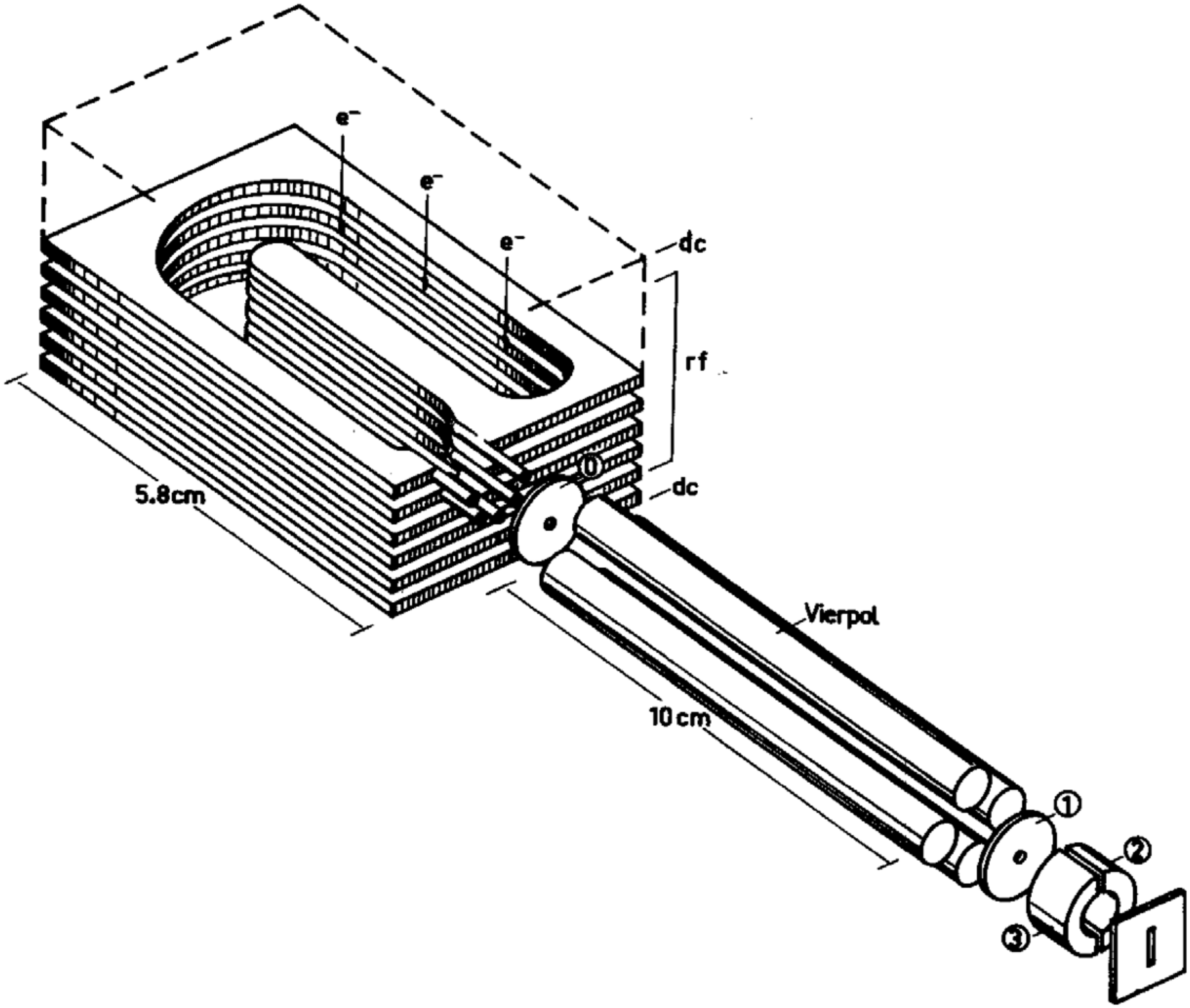




8-Pol (1969) / wire 4-Pol (1985)

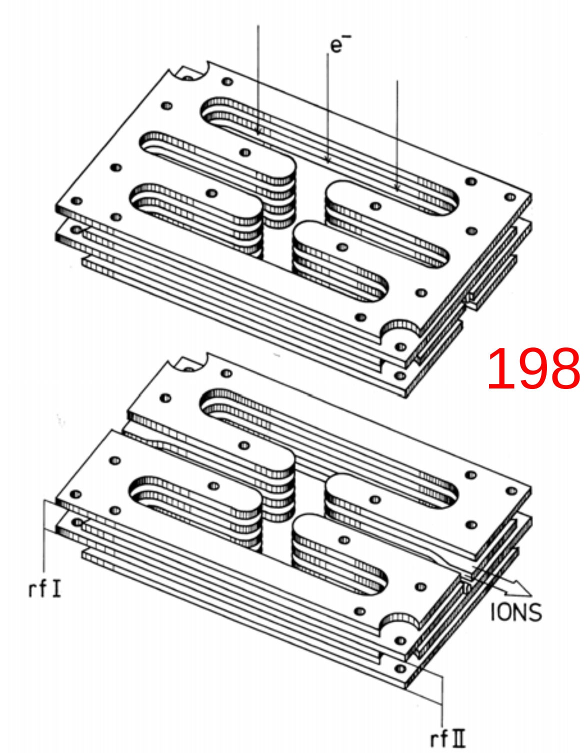


Storage ion source

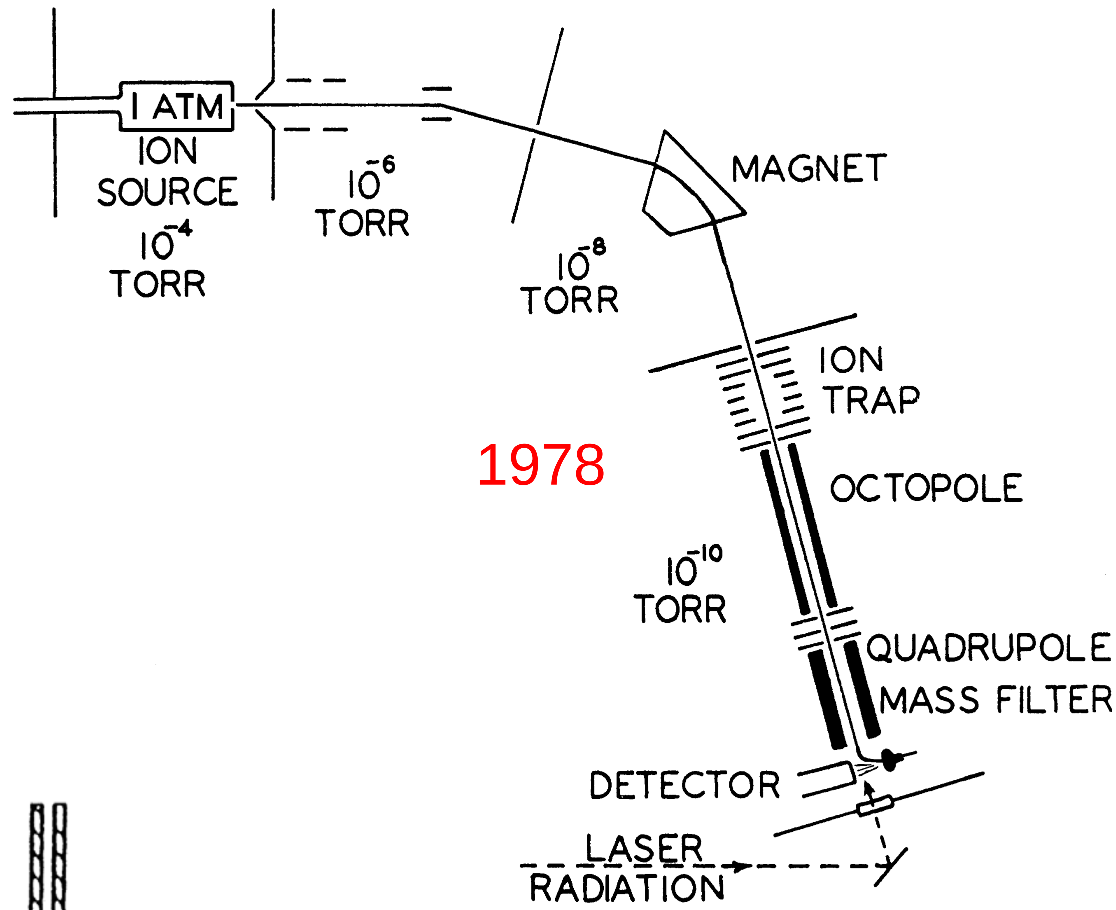


Gerlich 1977

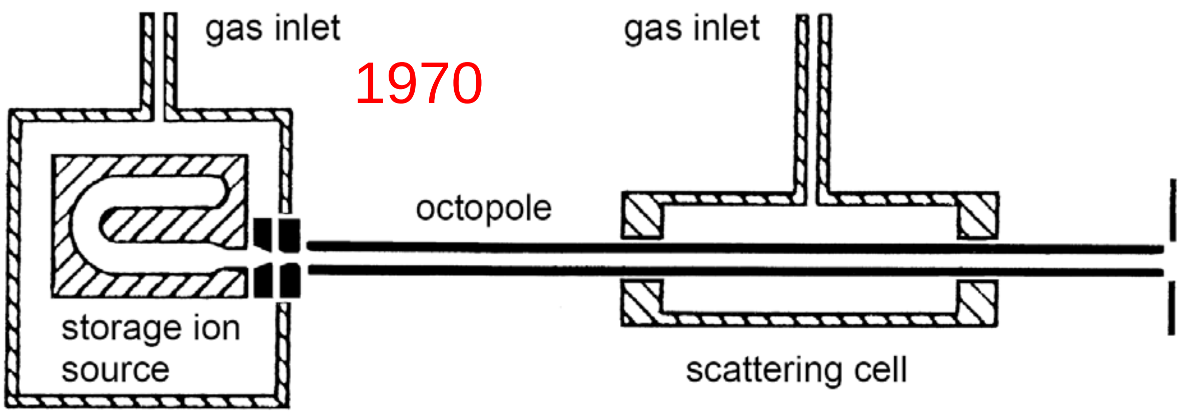
History: thermalizing ions



1985



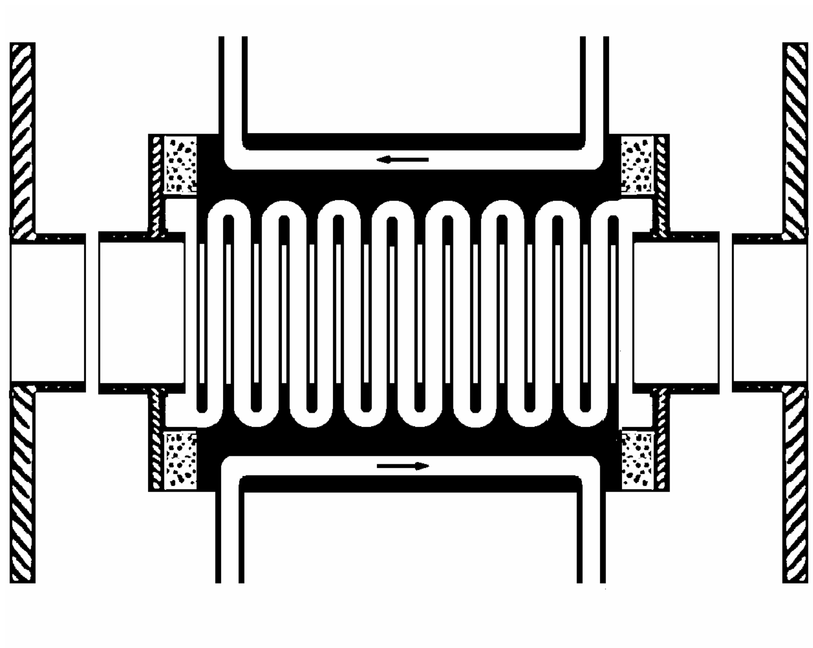
1978

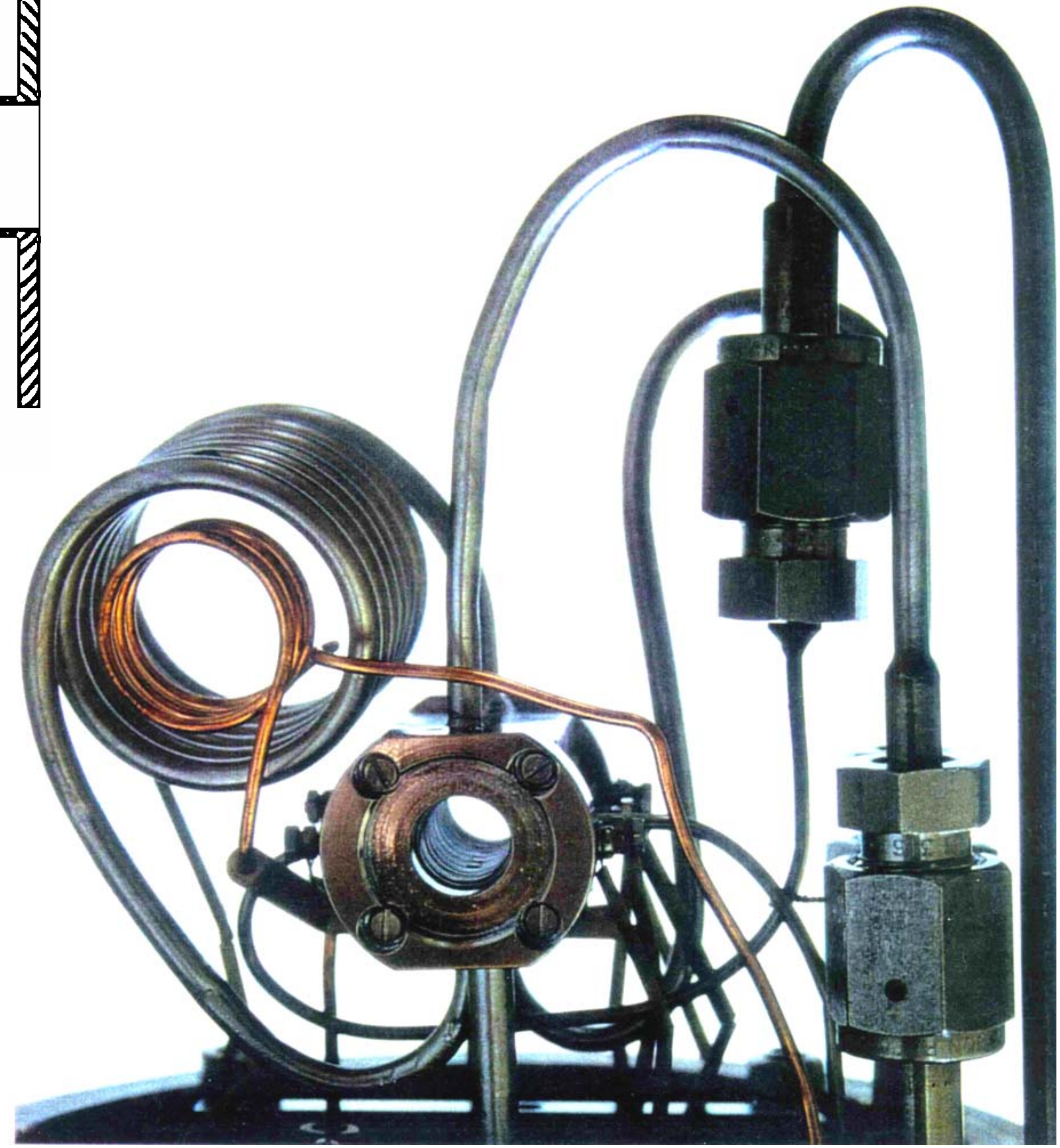


1970

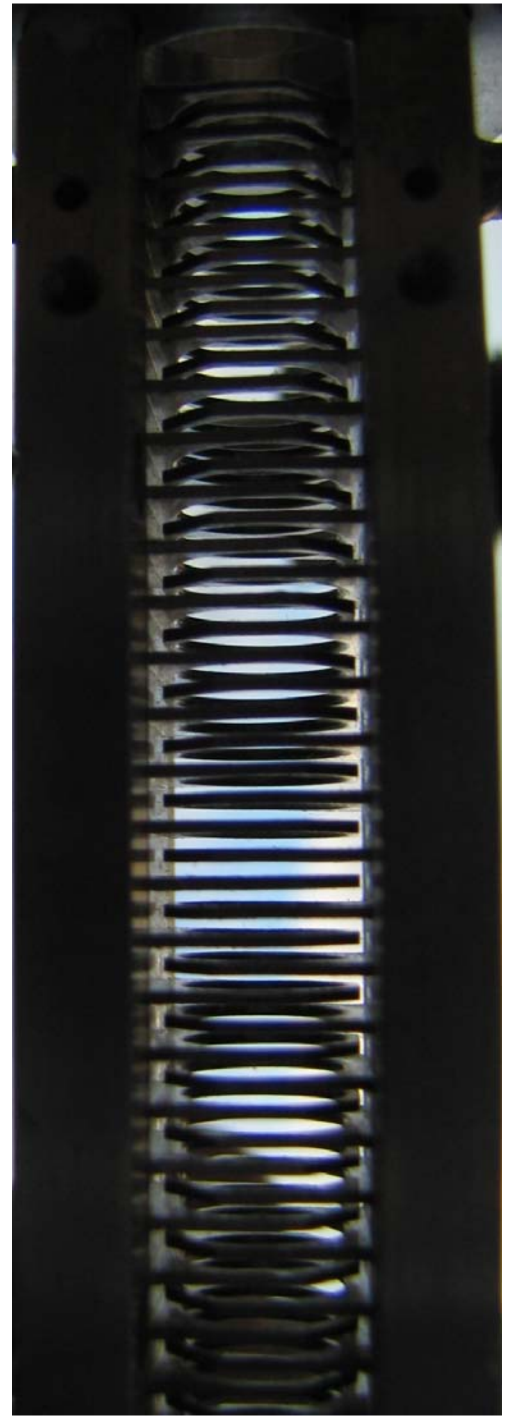
D. Gerlich, J. Anal. At. Spectrom., **19,** (2004) 581

Liquid N2 cooled trap (1988)

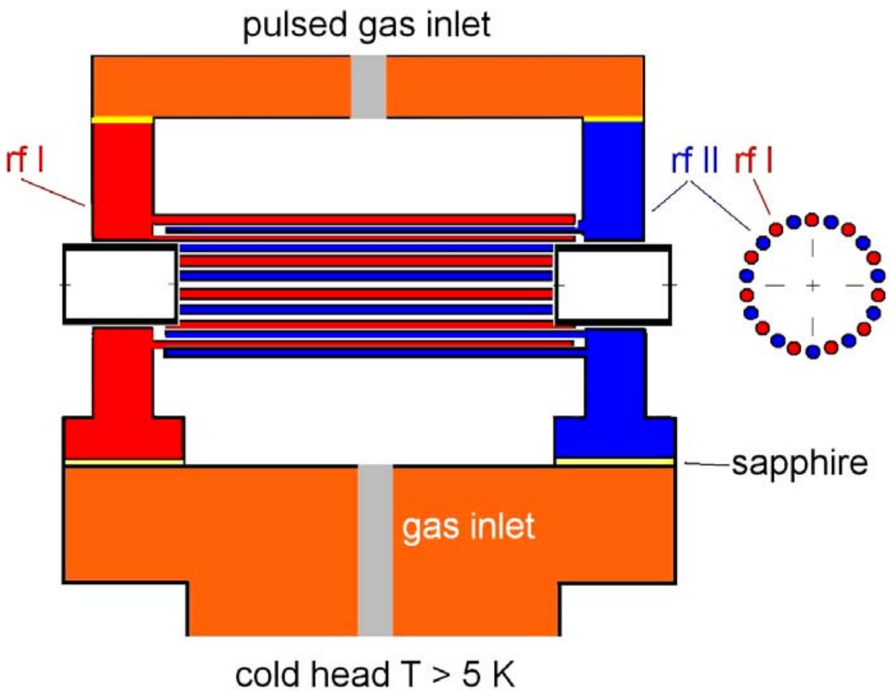




l-N_2 cooled RET (1990)

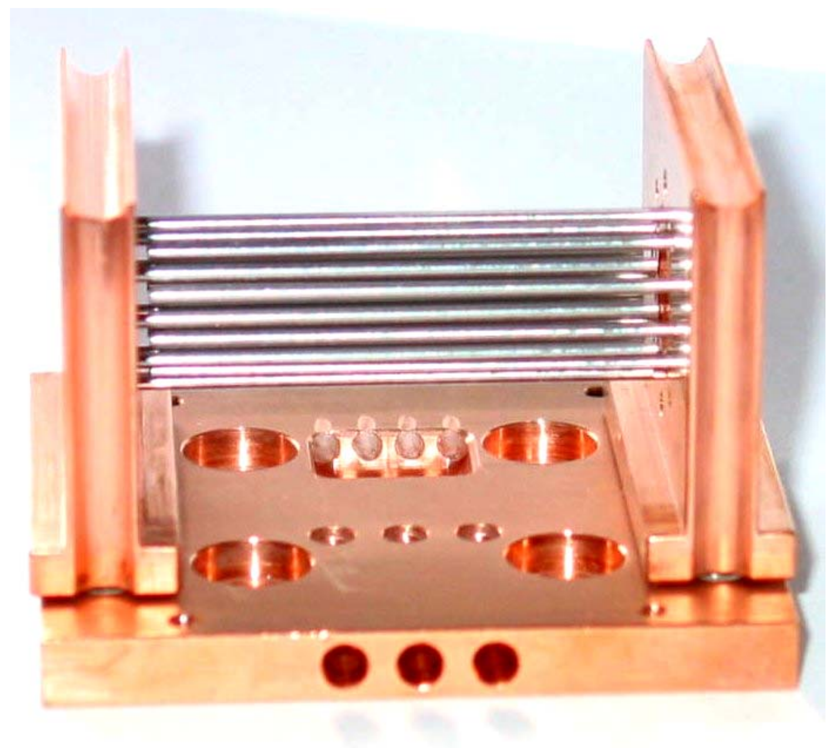


TV 22-pole trap



d = 1 mm
2 r_0 =10 mm

$r_0 = (n-1)\, d/2$
$2n = 22$



Effective potential V^*

$V^* = q^2\, E_0^{\,2} / \, 4m\Omega^2$

Adiabaticity parameter η

$\eta = 2\, q\, |\nabla E_0| \,/\, m\Omega^2$

parameters: q, m, E_0, Ω, scaling: $m\Omega^2$

$\eta \sim E_{max}^{(n-2)/(2n-2)} = E_{max}^{9/20}$

D. Gerlich Physica Scripta, **T59** (1995) 256

22: a powerful number

„The 22 is the most powerful of all numbers. It is often called the Master Builder. The 22 can turn the most ambitious of dreams into reality. It is potentially the most successful of all numbers".

Source:
http://www.decoz.com/Masternumbers.htm

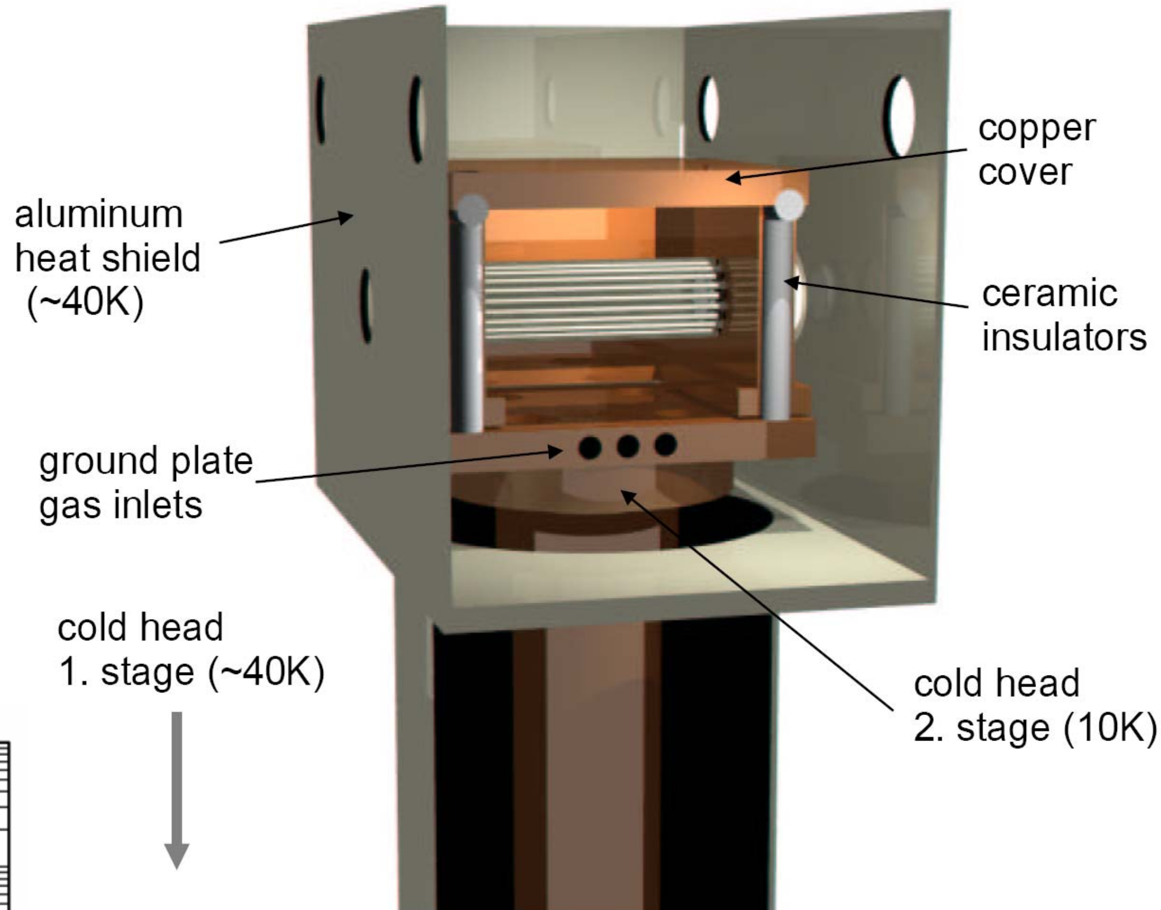
Master Numerologist Hans Decoz, author of Numerology; Key To Your Inner Self

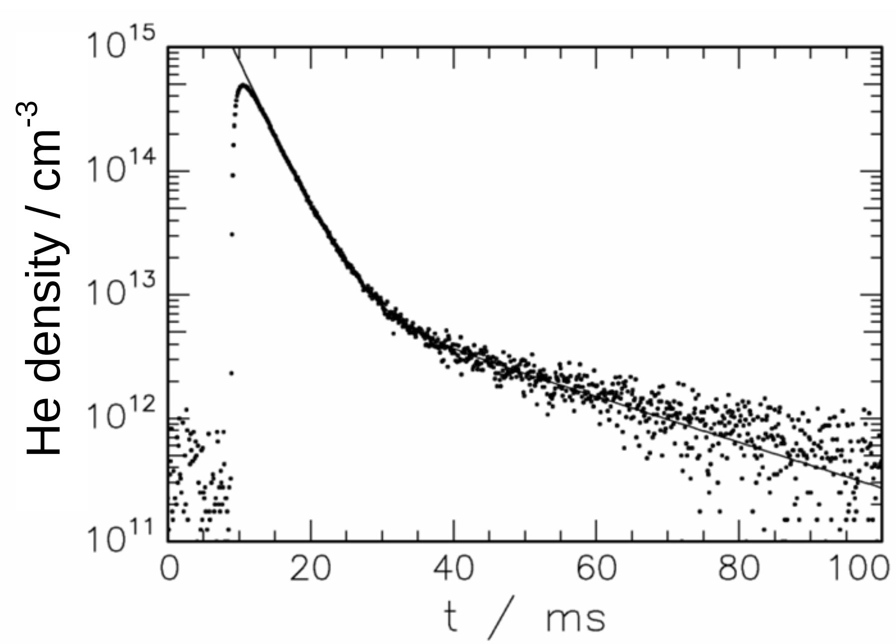
Buffer gas cooling in an rf trap

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

Paul trap does not work
η = const

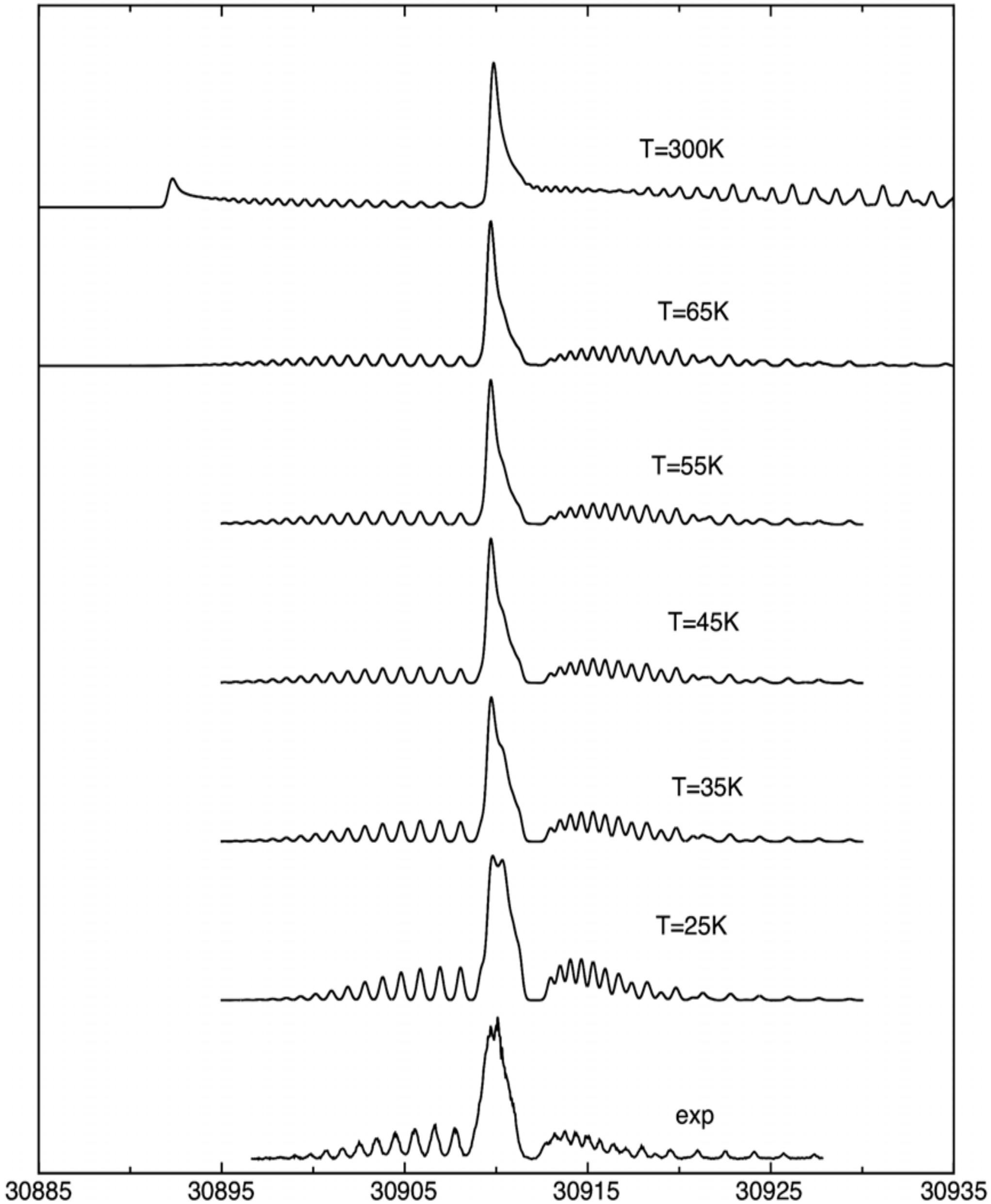
Only way to cool efficiently internal degrees of freedom are
rf multielectrode traps





sub K:
cold pulsed effusive beam

T: buffer gas, collision, rotation



N_2O^+ in cold He

m_{ion} = 44 u , m_{He} = 4 u

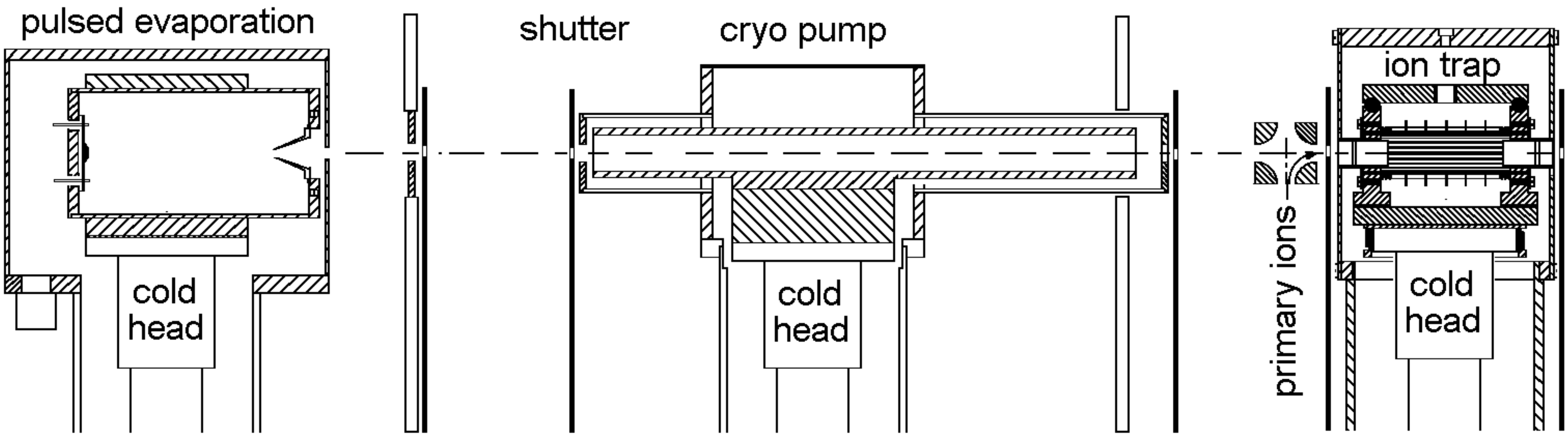
The rotational temperature of the ions is determined by the collision temperature T_{coll} which is given by:

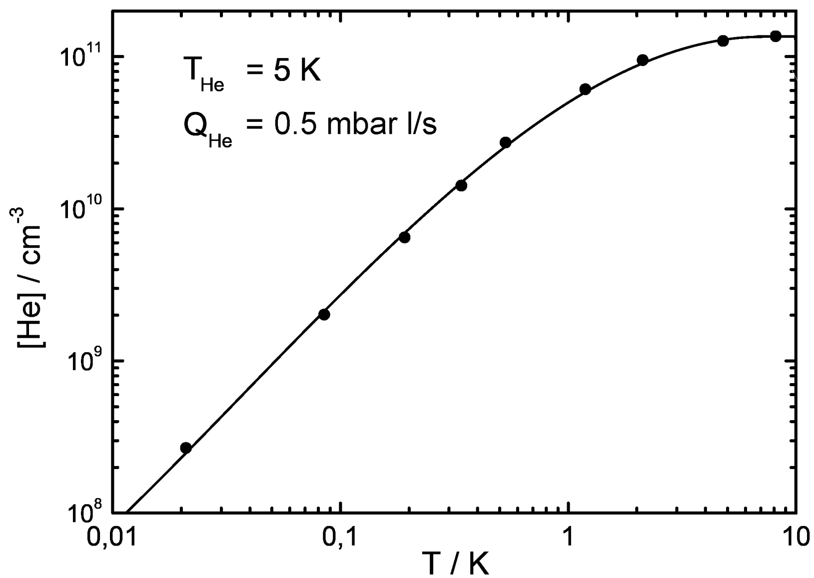
$$T_{coll} = (m_{ion}\ T_{He} + m_{He}\ T_{ion})/(m_{ion} + m_{He})$$

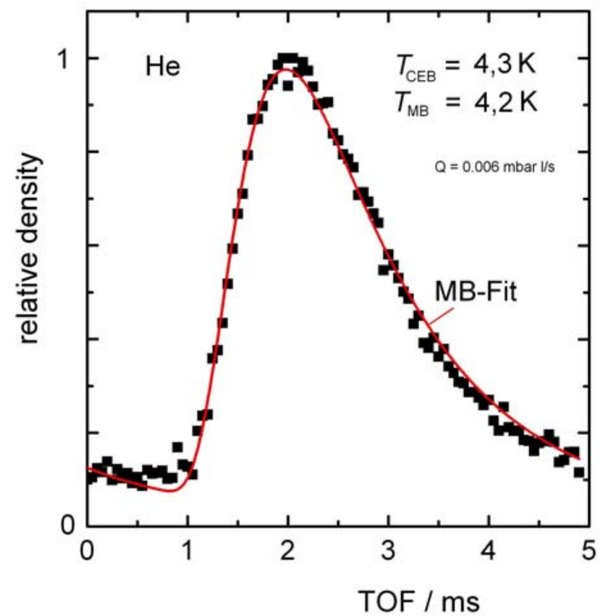
T_{He} = 5 K
$T_{rot} = T_{coll}$ = 25 K

T_{ion} = 245 K

Sub-K cooling of stored ions







D. Gerlich, G. Borodi, Faraday Discussion 142 (2009): *Cold and Ultracold Molecules*

Phase space compression in ion chemistry

Liouville theorem

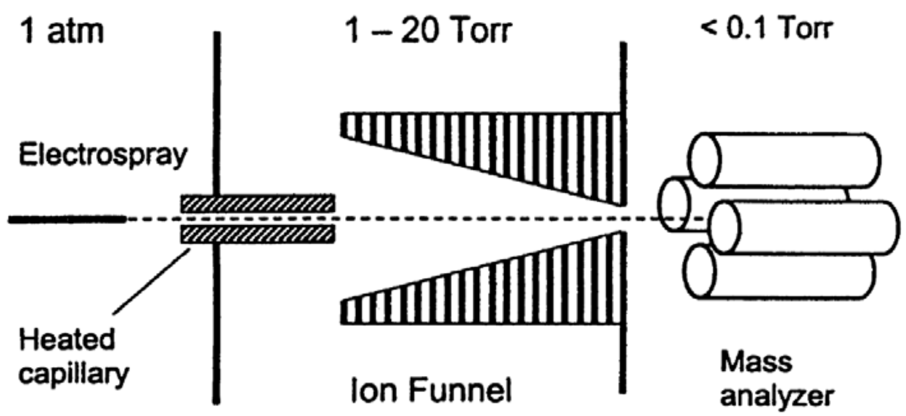
Patent US4963736 (1988)

Mass spectrometer and method and improved ion transmission

Patent suit 2002

Applied Biosystems and MDS won against Micromass to the tune of $47.5M.

Ion funnel PNNL, Richland 1998



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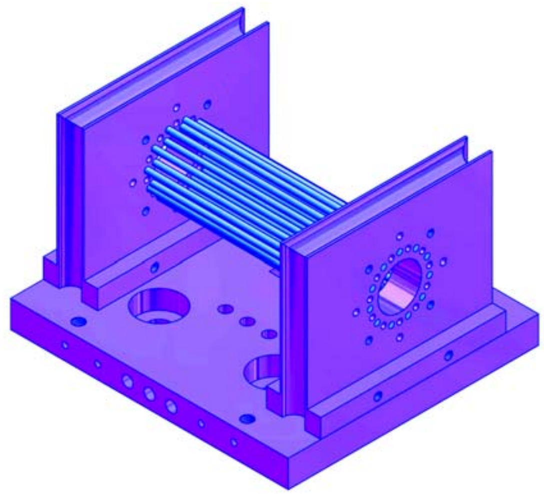
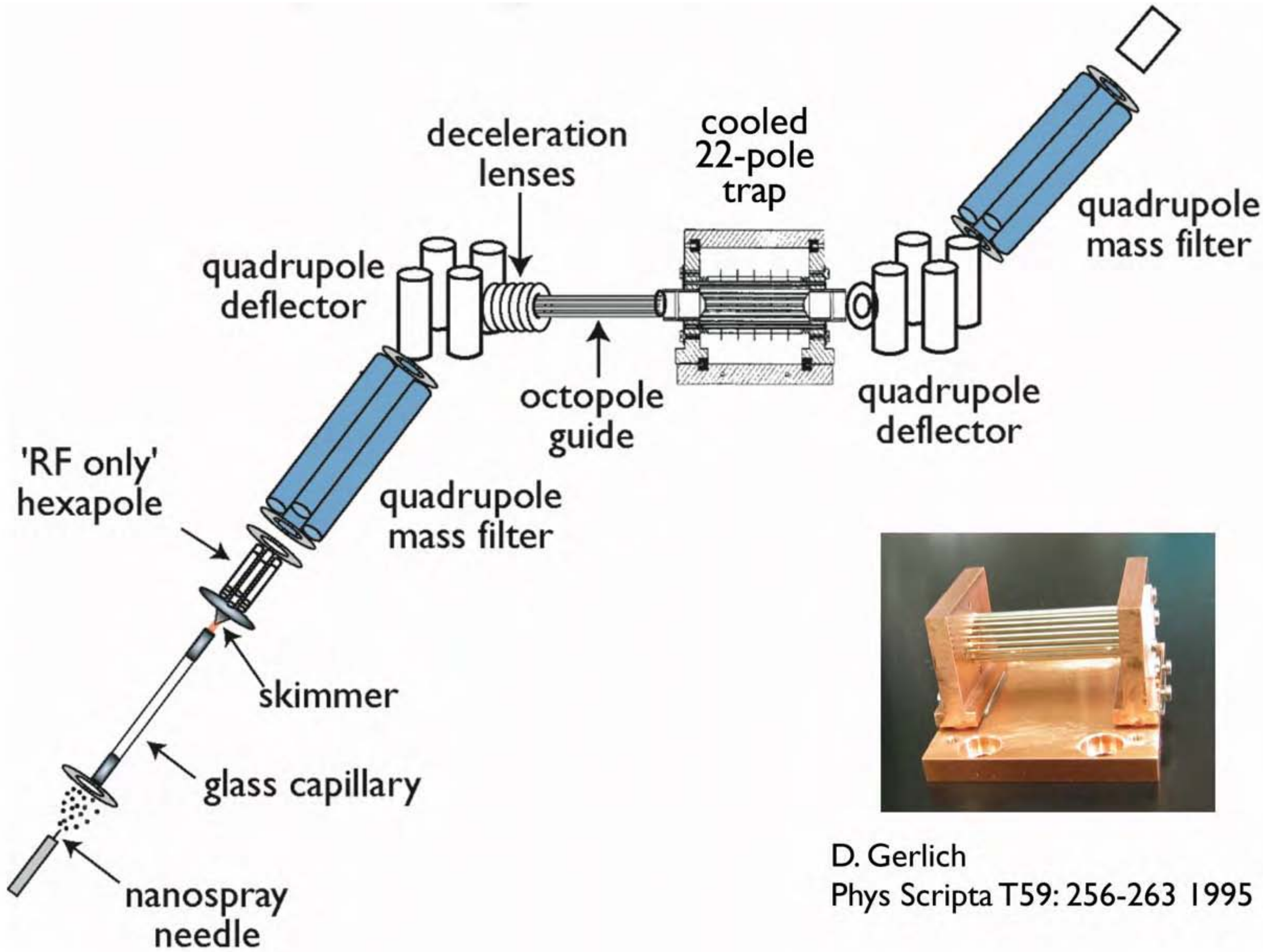
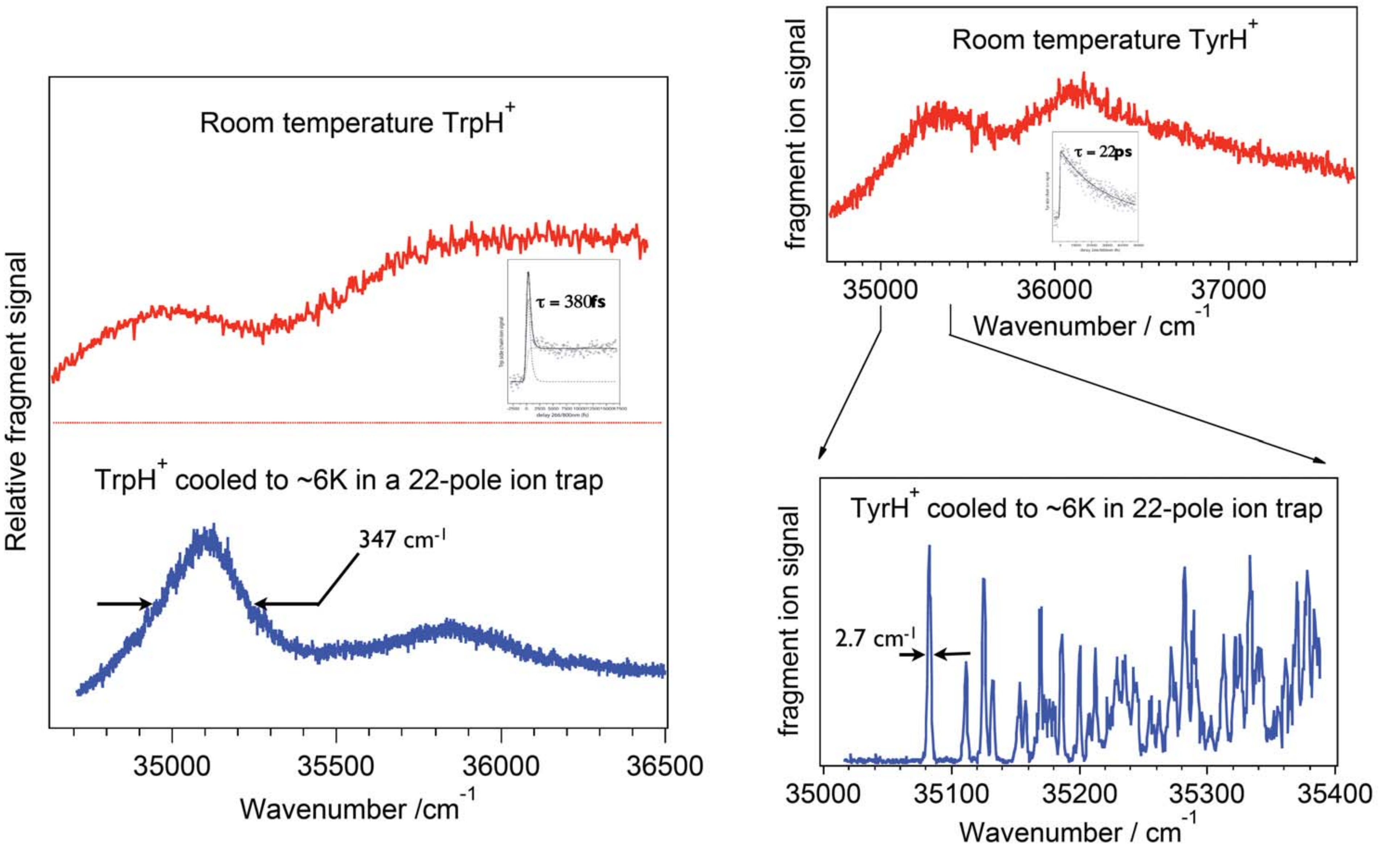


The Lausanne cooled ion spectrometer



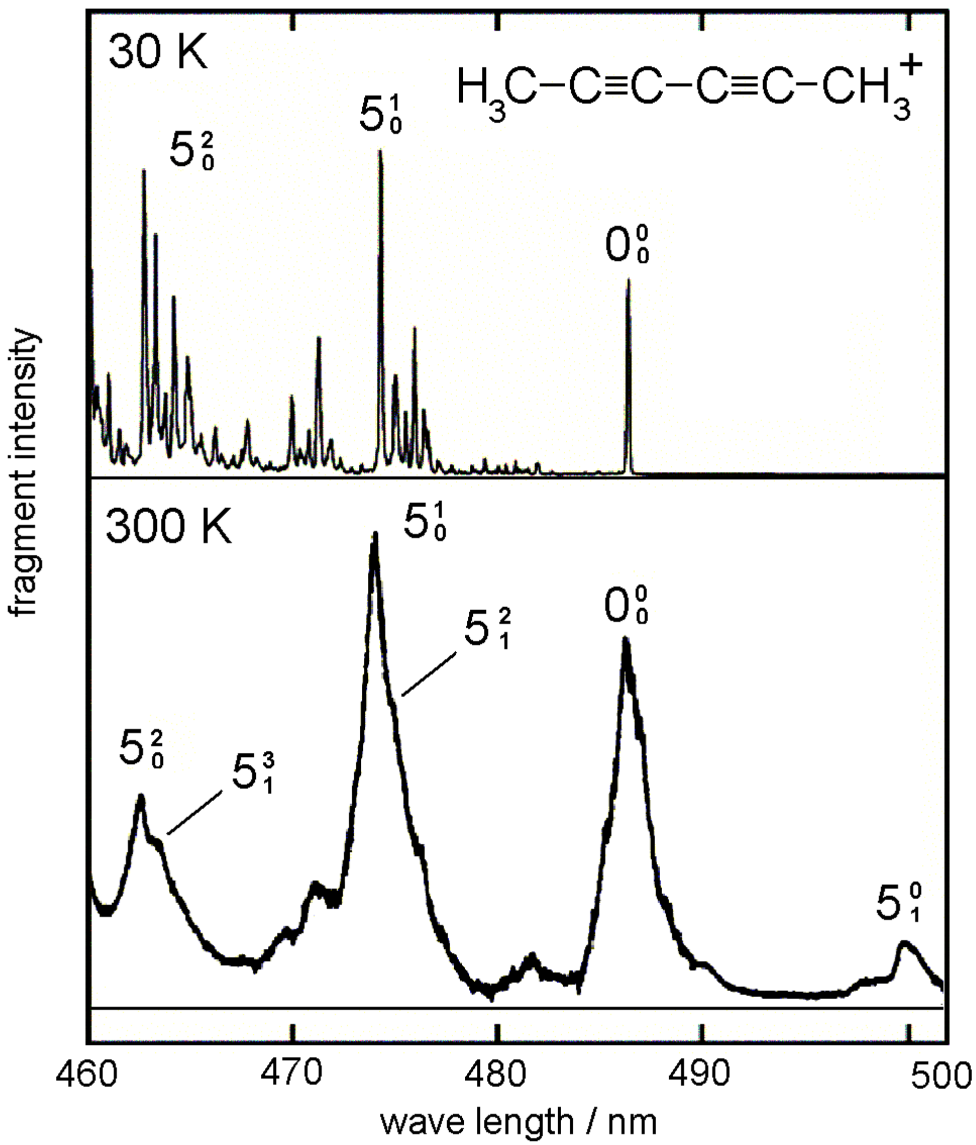
O. Boyarkin, S. Mercier, A. Kamariotis, and T. Rizzo
J. Am. Chem. Soc.; **128** (2006) 2816

Electronic spectra of cold, protonated amino acids

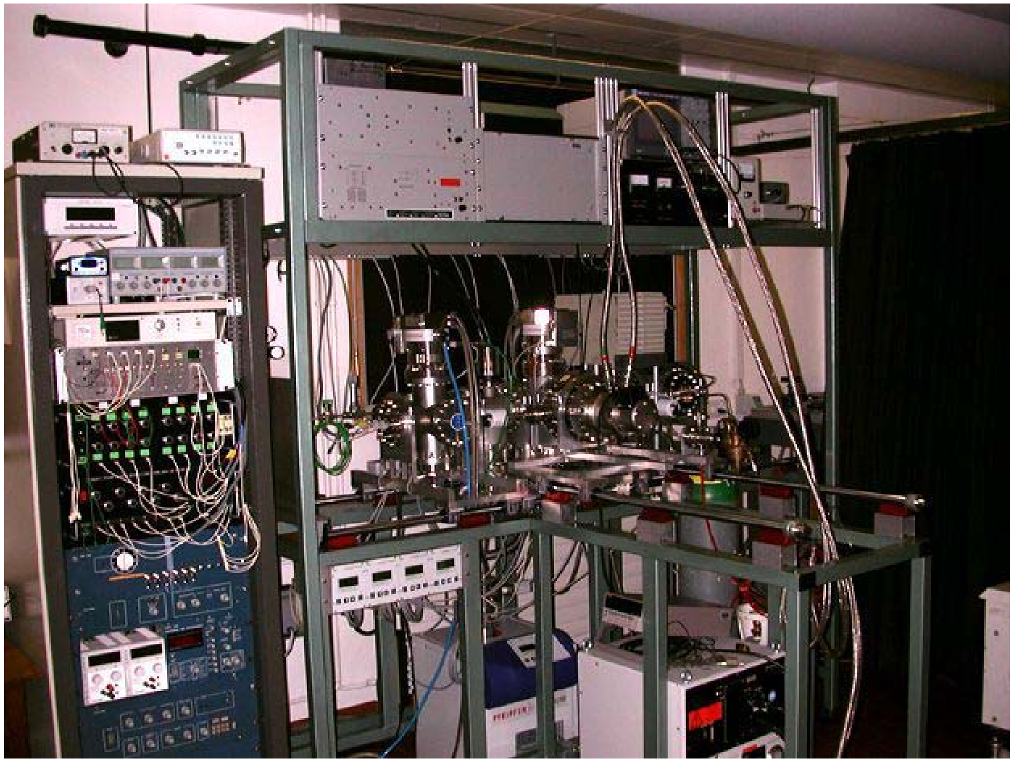


O. Boyarkin, S. Mercier, A. Kamariotis, and T. Rizzo
J. Am. Chem. Soc.; **128** (2006) 2816

Electronic spectra: the Basel 22PT



one photon dissociation spectrum
30 K
300 K



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

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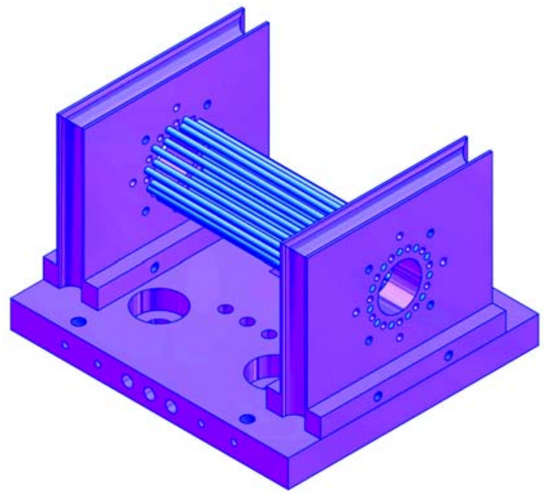
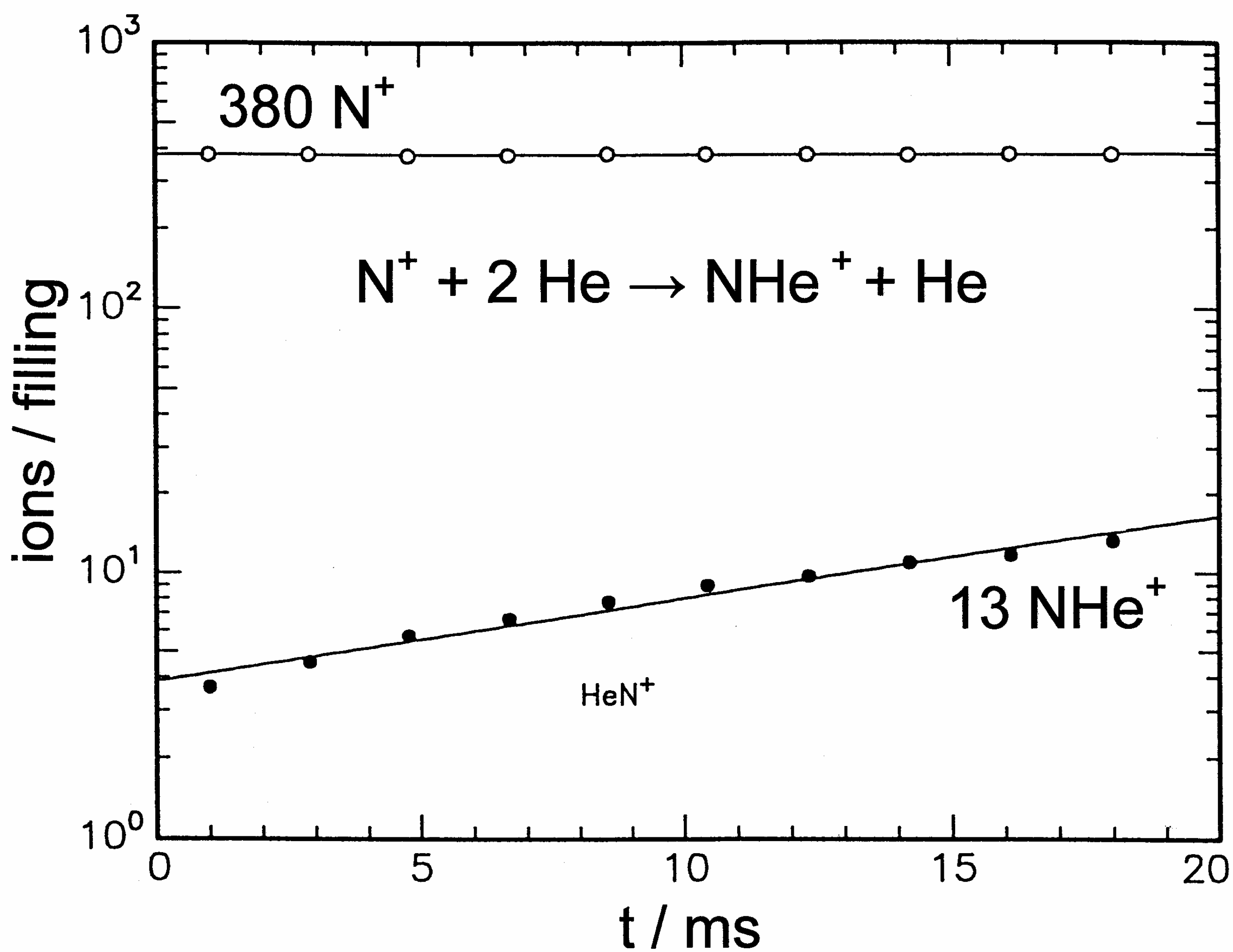


Ternary association: $N^+ + 2\,He$



T=(15±5)K

$[He] = 4.8\times10^{14}\ cm^{-3}$

collision rate $10^6\ s^{-1}$

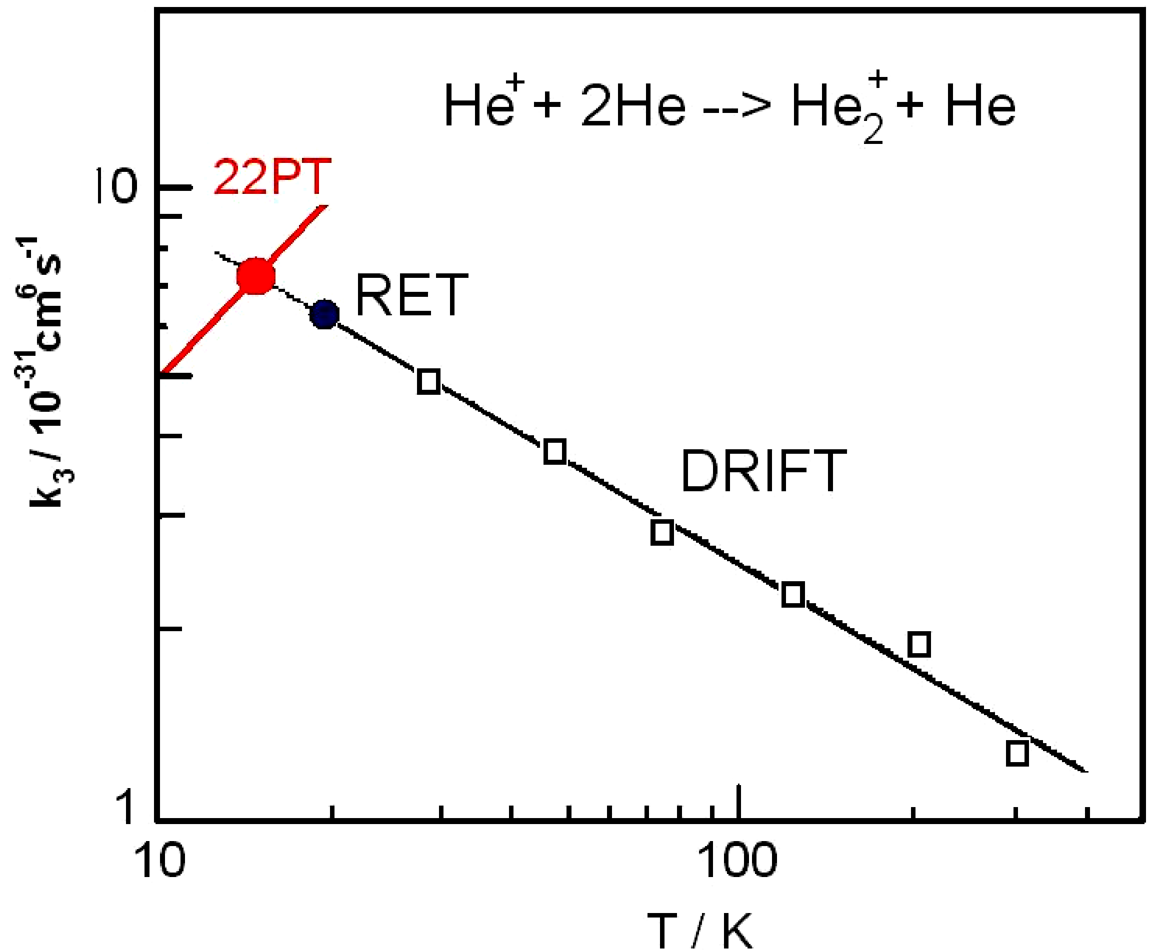
$k_{eff} = 8.7\times10^{-16}\ cm^3\ s^{-1}$

$k_3 = 4.6\times10^{-31}\ cm^6\ s^{-1}$

complex life time
1.6×10^{-11} s

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

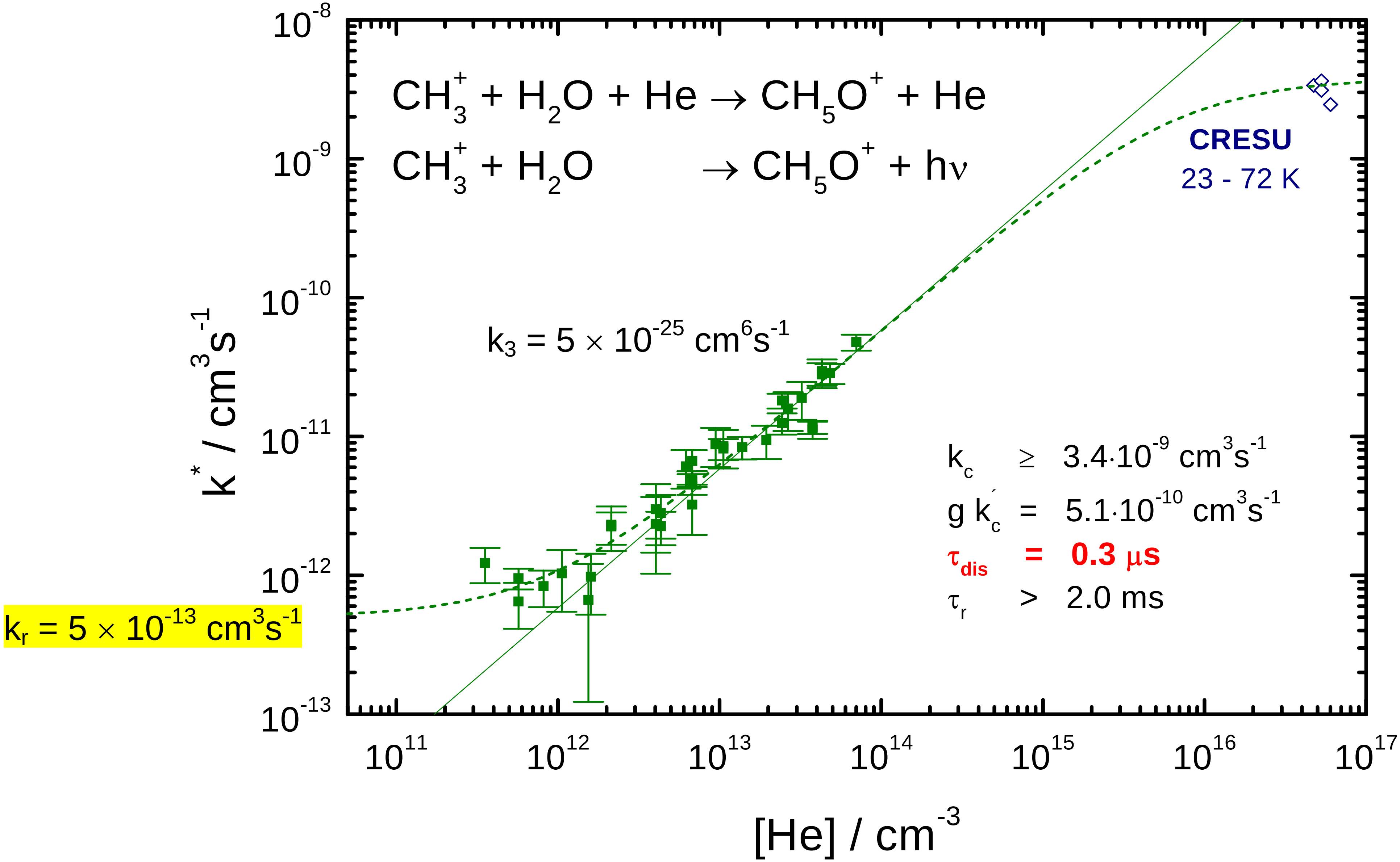
He^+ + 2 He: ternary association



$k_3 = 1.4 \times 10^{-31}\ (T/300\ K)^{0.6 \pm 0.1}\ cm^6 s^{-1}$

22PT: [pau94], DRIFT: Böhringer Arnold 1983

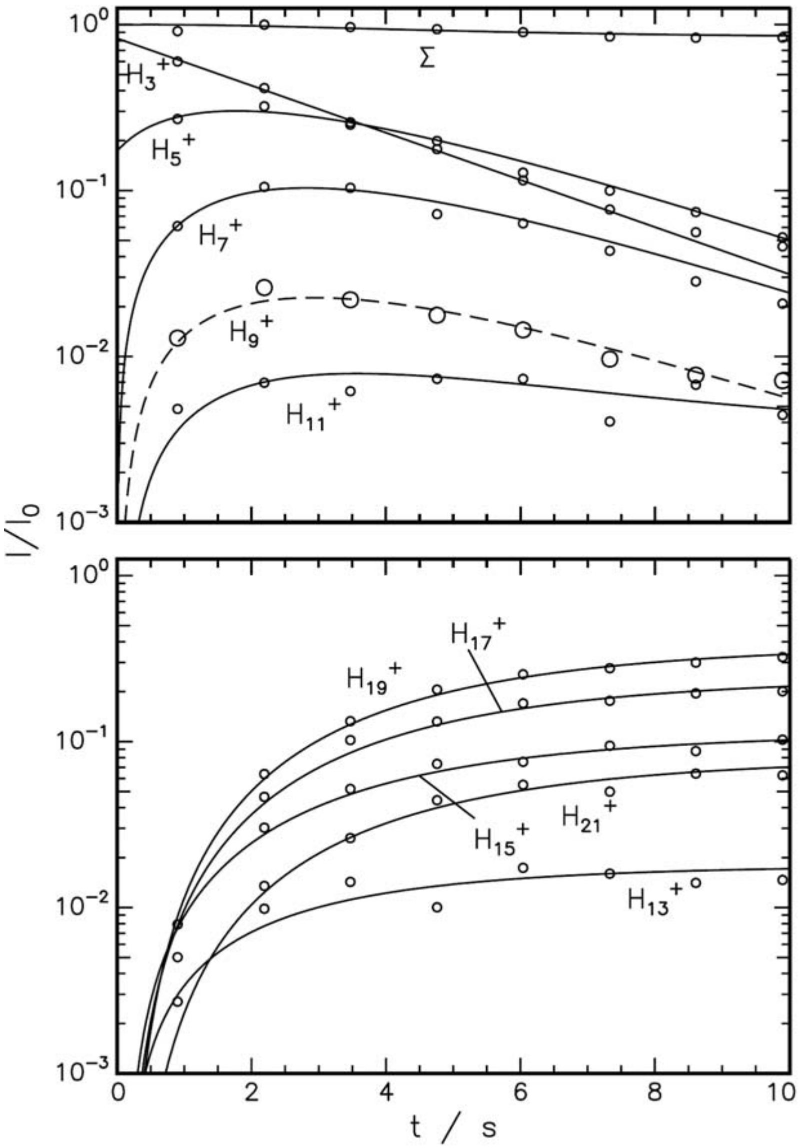
Formation of Methanol in space?

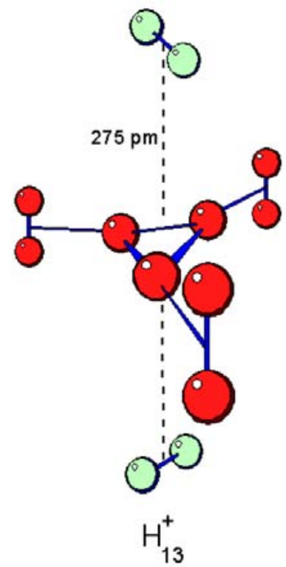


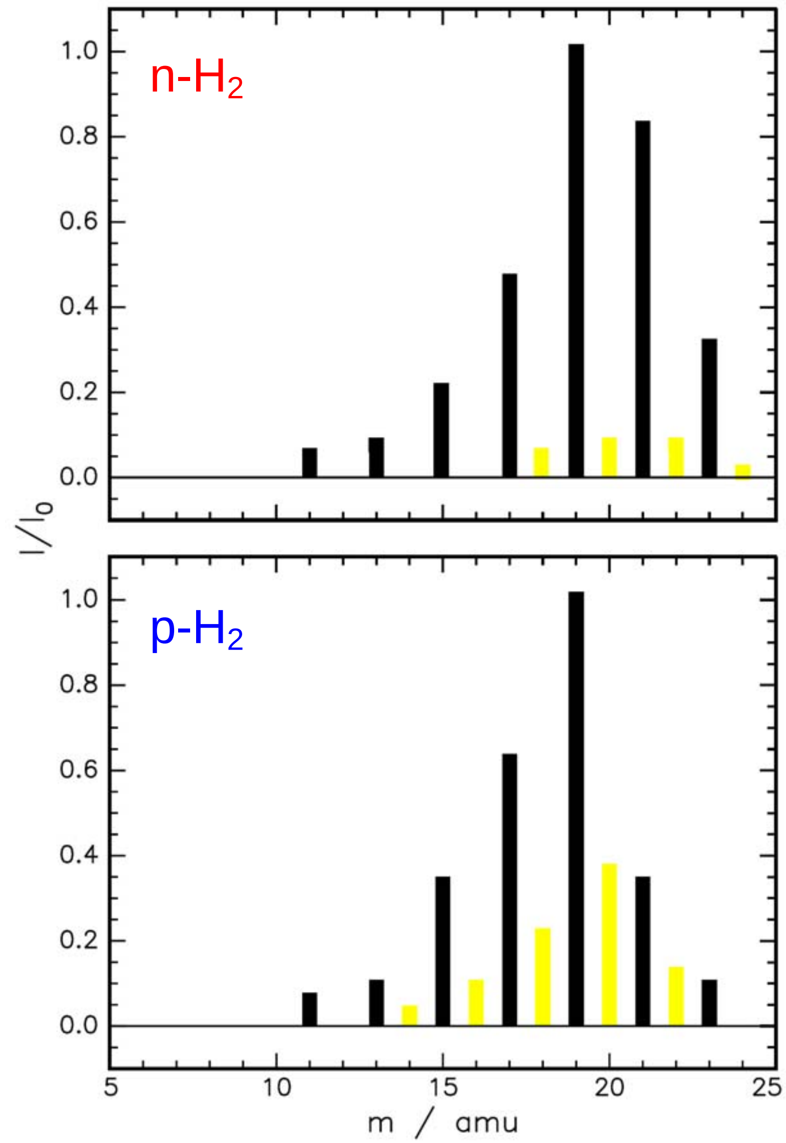
A. Luca, D. Voulot, D. Gerlich 2002

H_n^+ cluster growth stationary equilibrium: n = 19

T = 10 K, $[H_2] = 10^{14}$ cm^{-3}, storage time 10 s







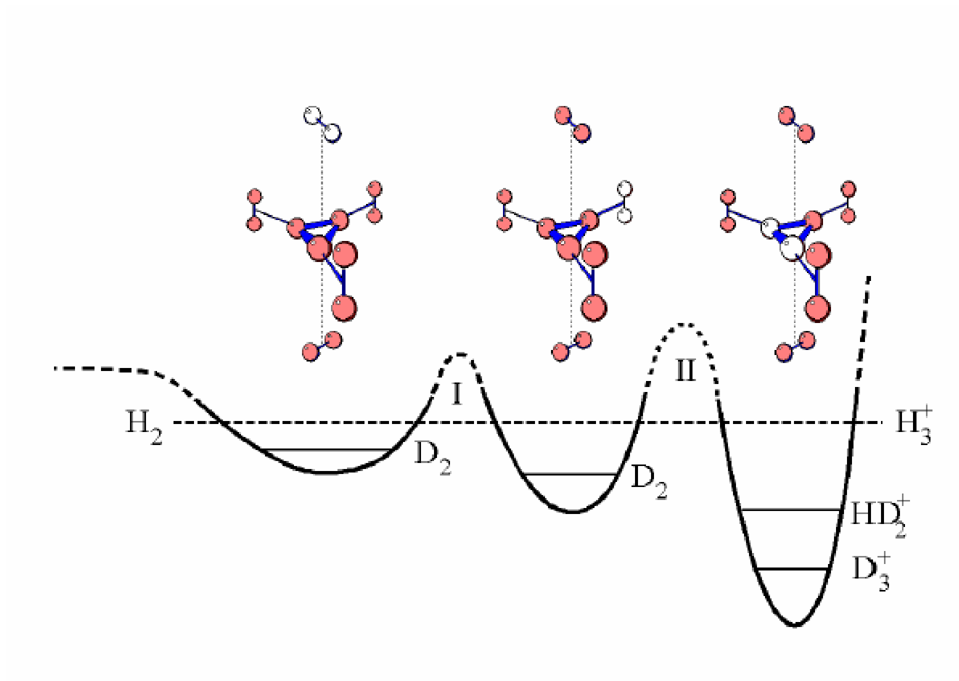
W. Paul, B. Lücke, S. Schlemmer, D. Gerlich, IJMS **149** (1995) 373

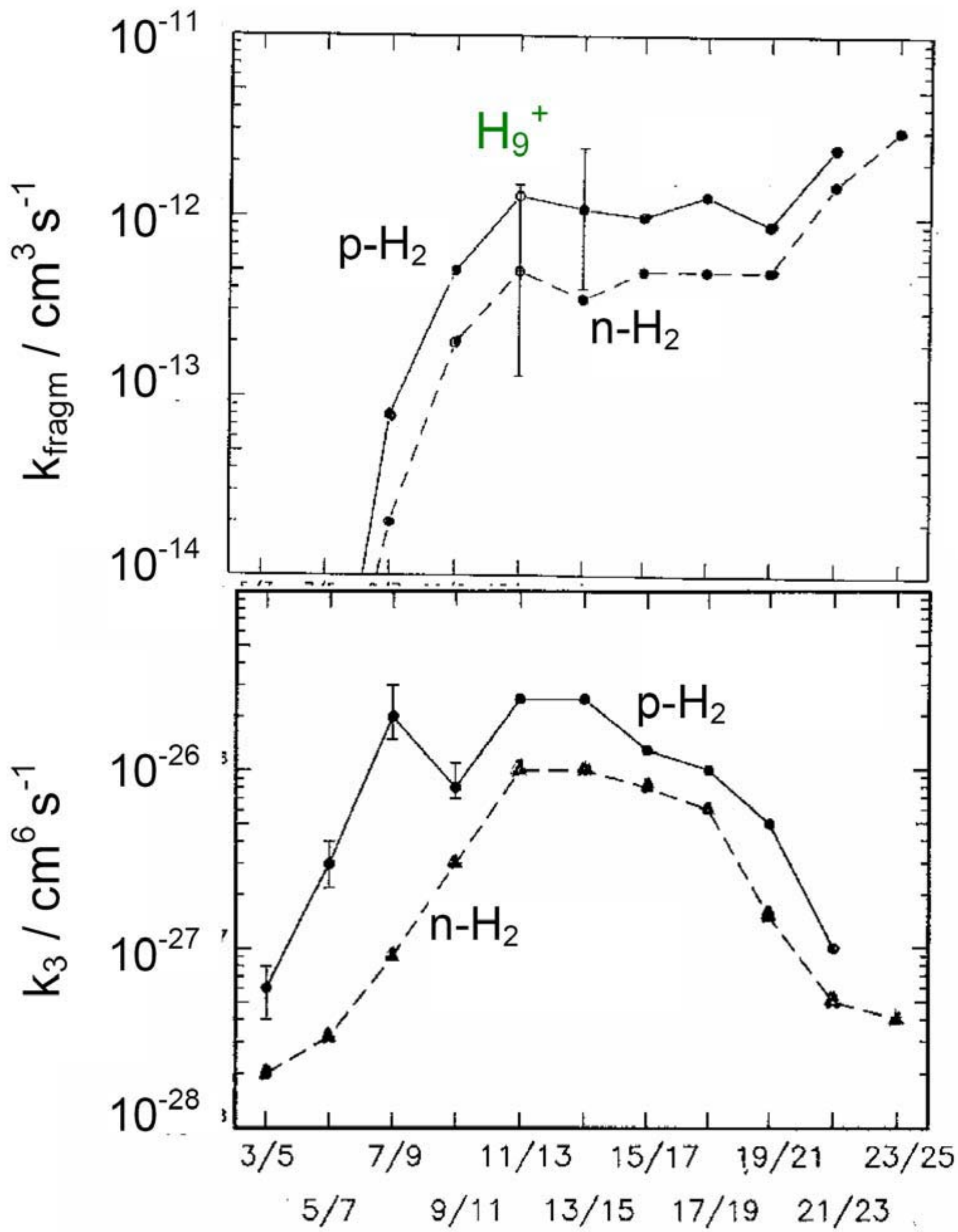
Growth and destruction of $((H_3^+)H_2)_n$ cluster

$p\text{-}H_2$ is more destructive!

large cluster grow better with $o\text{-}H_2$

deuteration:





Paul, Lücke, Schlemmer, and Gerlich, Int. J. Mass Spectrom. Ion Proc., **150** (1995) 373

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Polyatomic ions in traps: from molecules via clusters to nanoparticles

Dieter Gerlich

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Basics, buffer gas cooling

Typical tests

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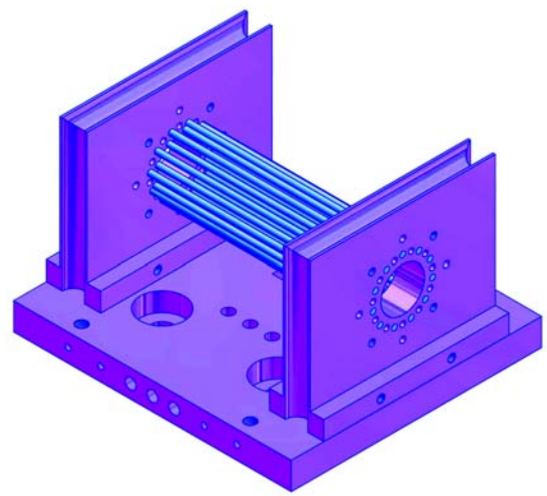
Association reactions, cluster
LIR: N_2^+ + Ar



Recent application

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State selective preparation

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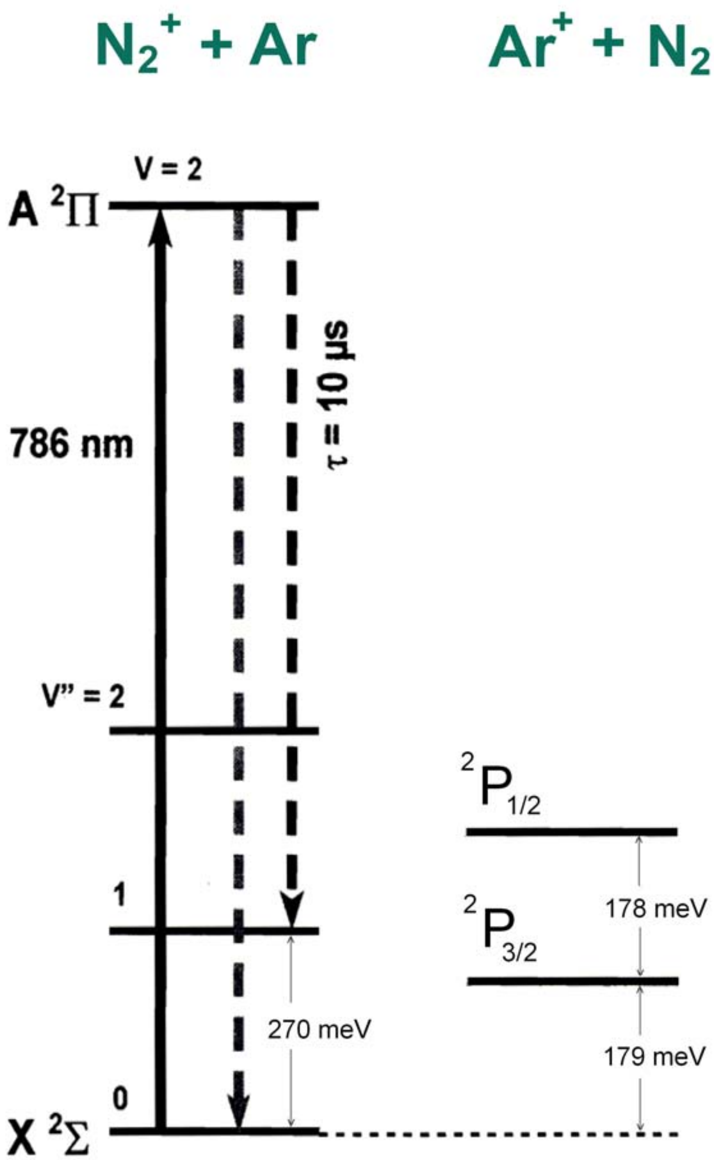
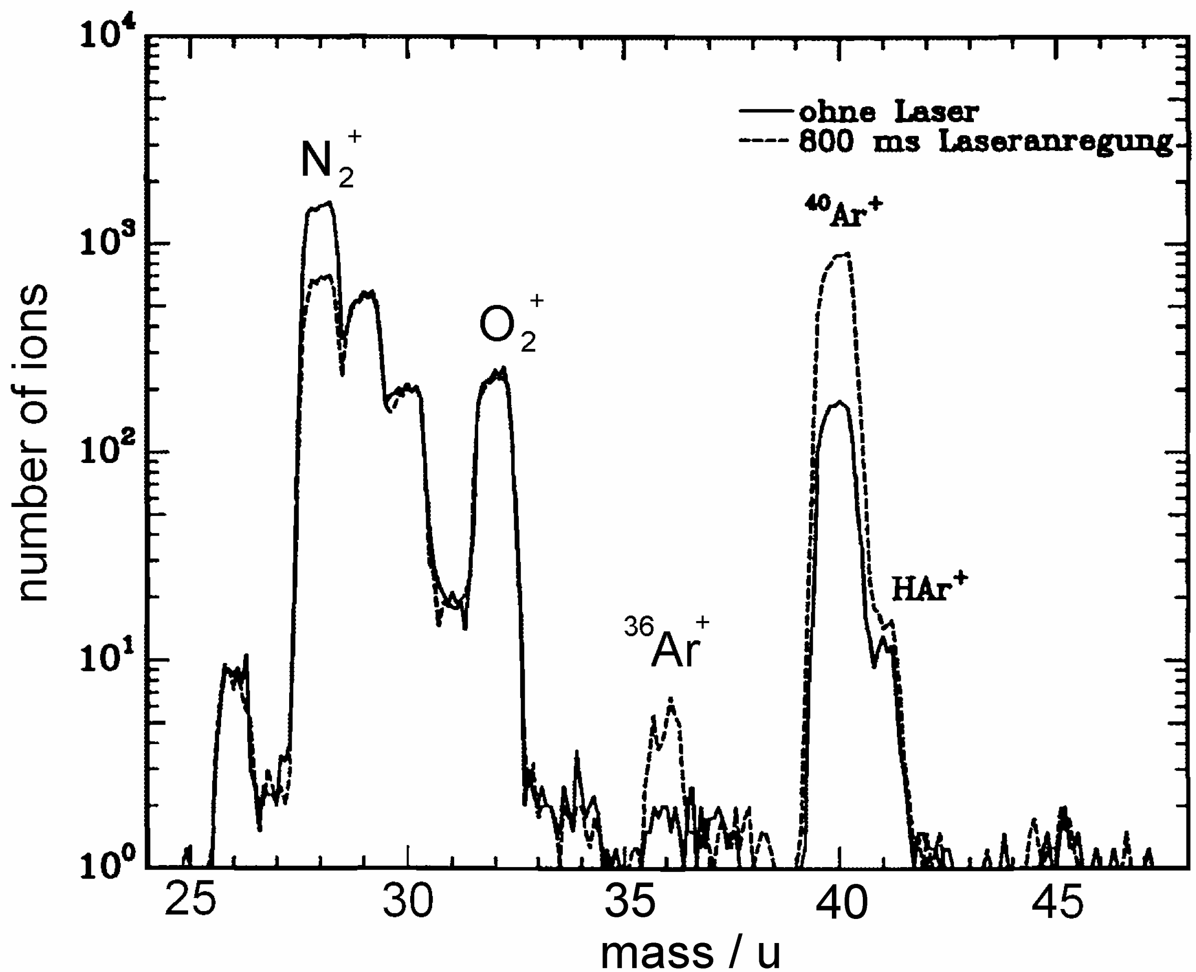
NPMS
HT - SRET, Decay of C_{60}^+

Summary and outlook

N_2^+ + Ar: LIR

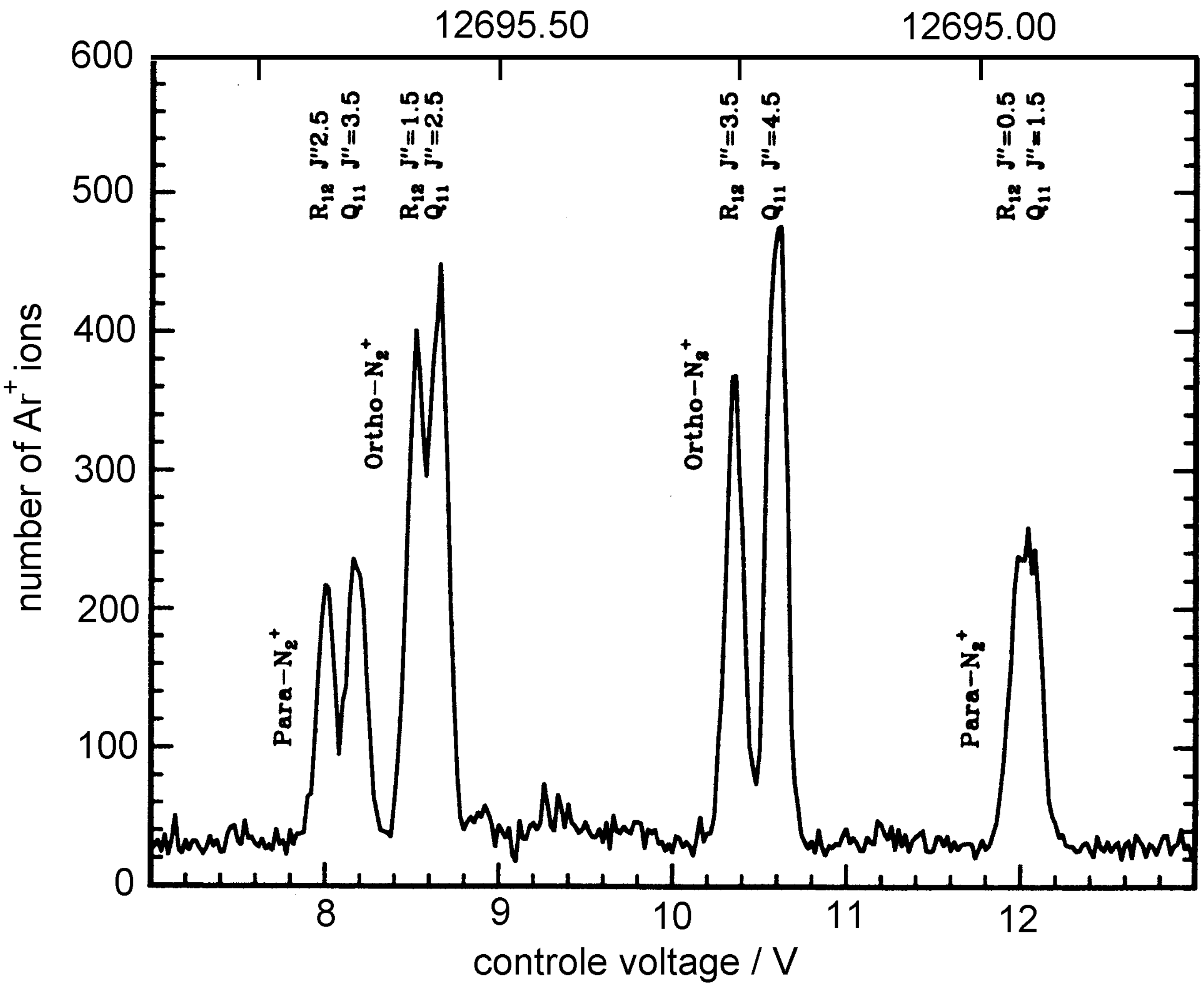


N_2^+ + Ar
Ar^+ + N_2
V = 2
A $^2\Pi$
786 nm
τ = 10 µs
V" = 2
1
0
X $^2\Sigma$
270 meV
$^2P_{1/2}$
$^2P_{3/2}$
178 meV
179 meV



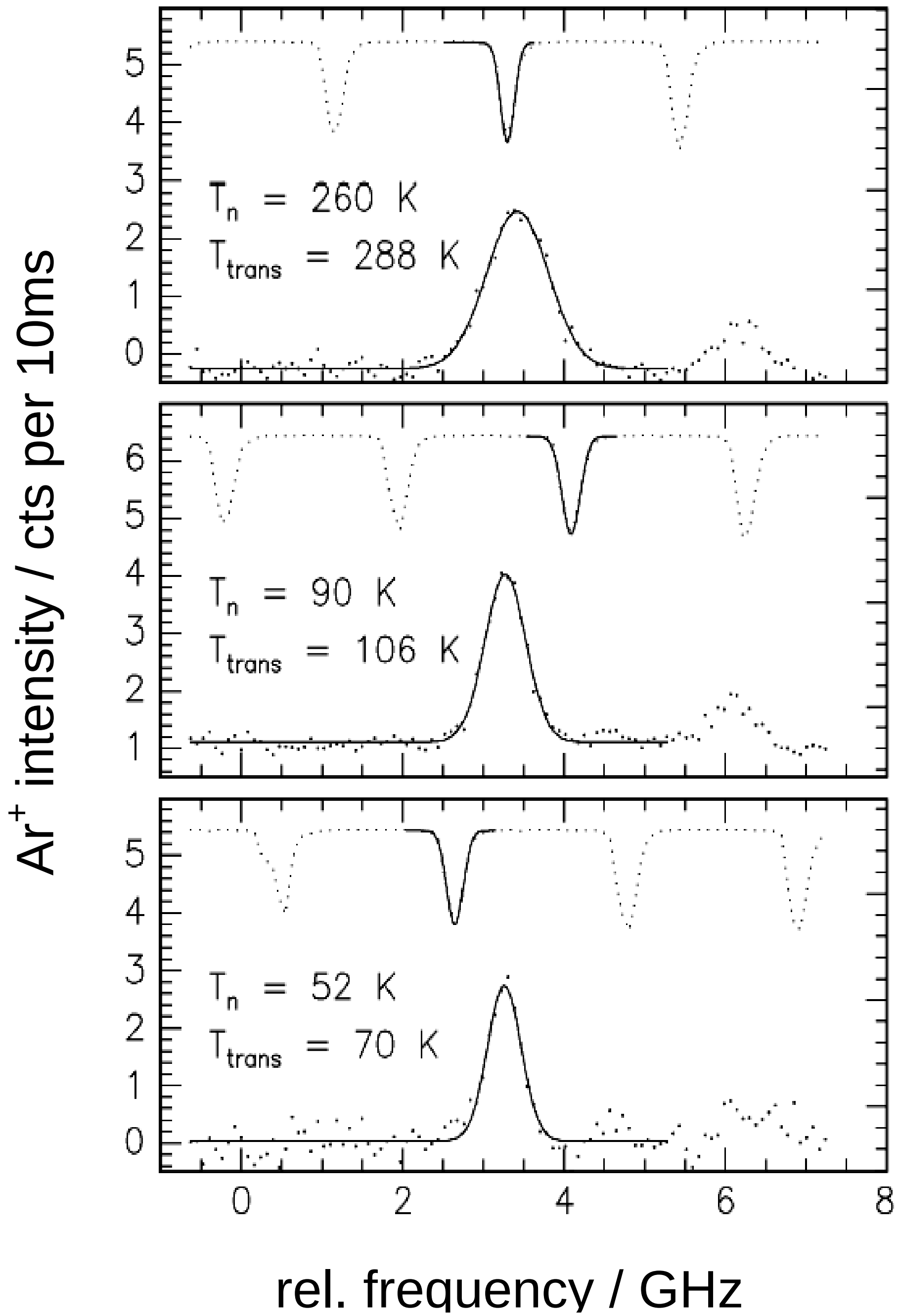
ohne Laser
800 ms Laseranregung
N_2^+
O_2^+
$^{40}Ar^+$
$^{36}Ar^+$
HAr^+
number of ions
mass / u
10^0
10^1
10^2
10^3
10^4
25
30
35
40
45

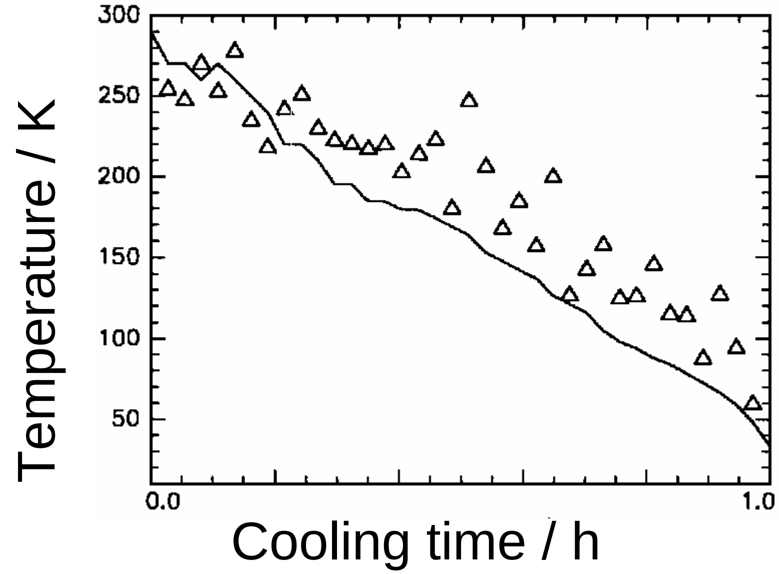
Low lying states N_2^+

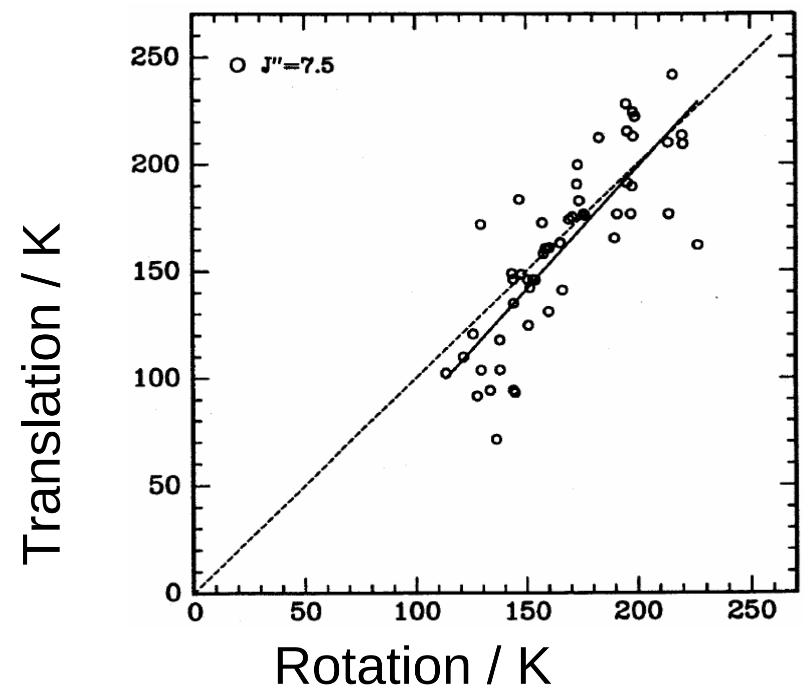


Linie		Messung
R_{12}	$J'' = 0.5$	12694.9641
Q_{11}	$J'' = 1.5$	12694.9460
R_{12}	$J'' = 1.5$	12695.5921
Q_{11}	$J'' = 2.5$	12695.5675
R_{12}	$J'' = 2.5$	12695.6875
Q_{11}	$J'' = 3.5$	12695.6548
R_{12}	$J'' = 3.5$	12695.2586
Q_{11}	$J'' = 4.5$	12695.2149

Temperature: LIR $N_2^+ + Ar \rightarrow Ar^+ + N_2$

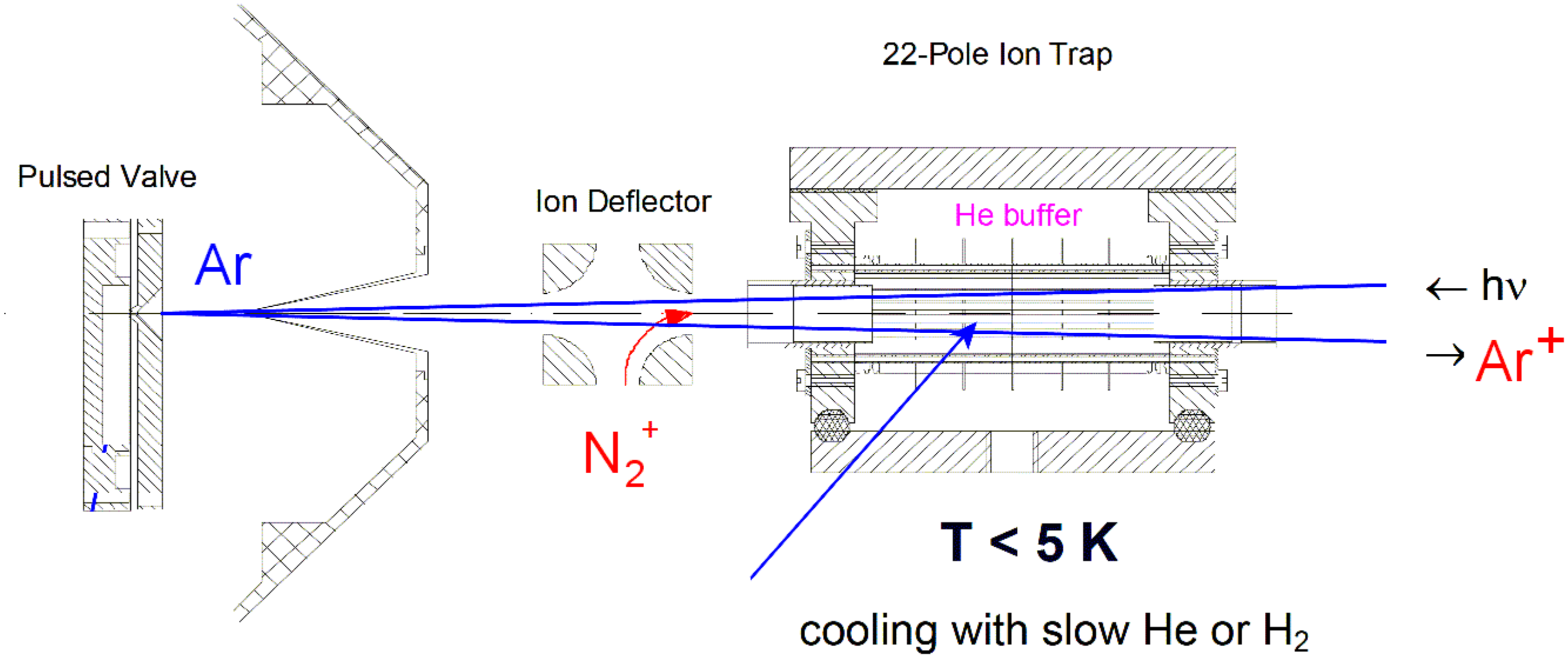


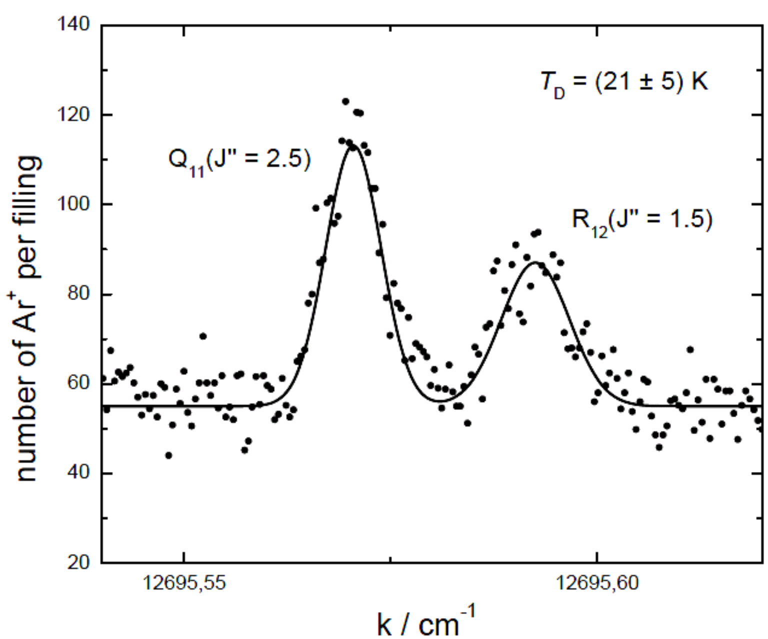




A. Sorgenfrei, PhD thesis, Freiburg, 1994

First LIR with an Ar beam



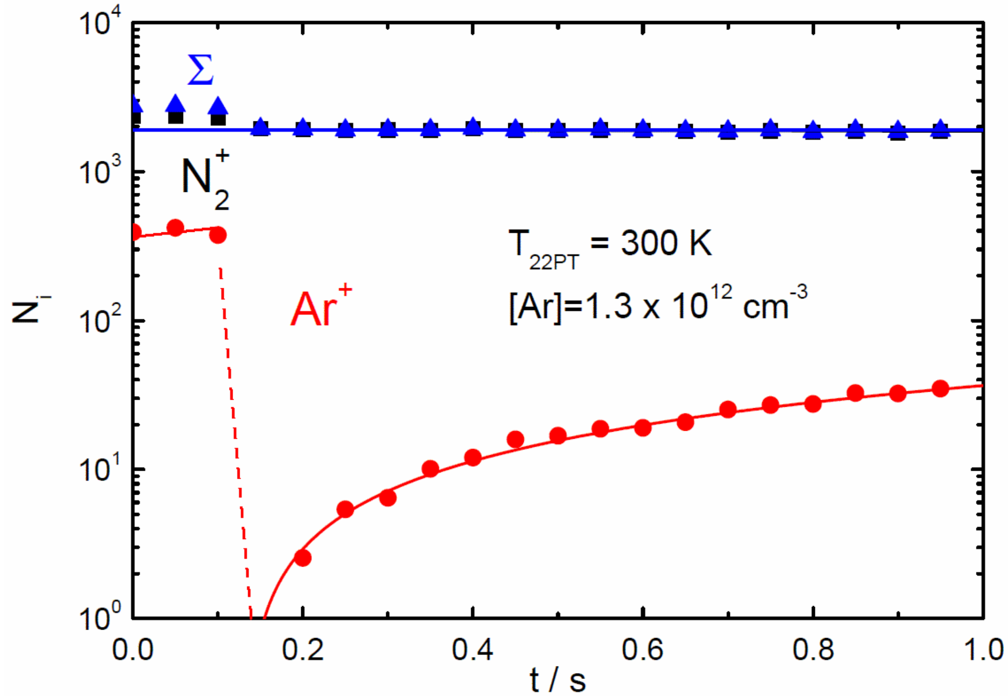


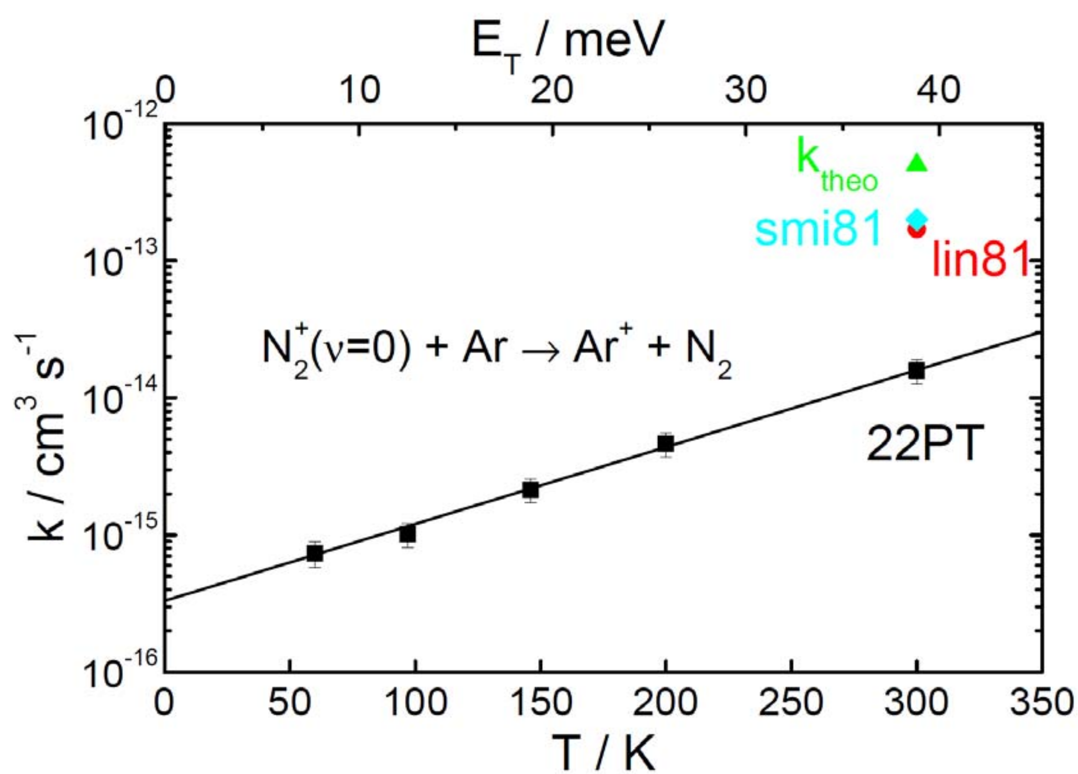
T_{22PT} 10 K

T_D (21 ± 5)K

Prag, 28.04.2010

$N_2^+ + Ar \rightarrow Ar^+ + N_2$





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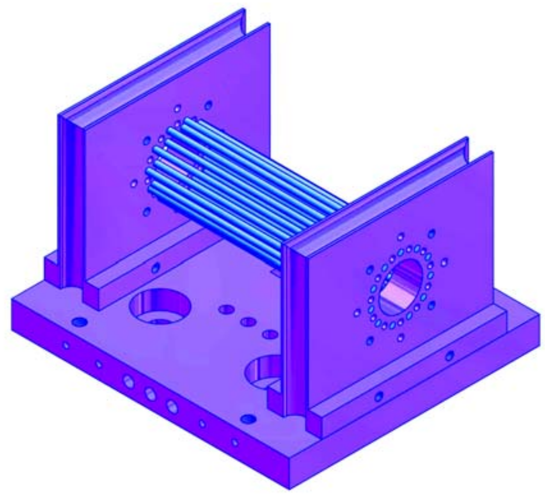

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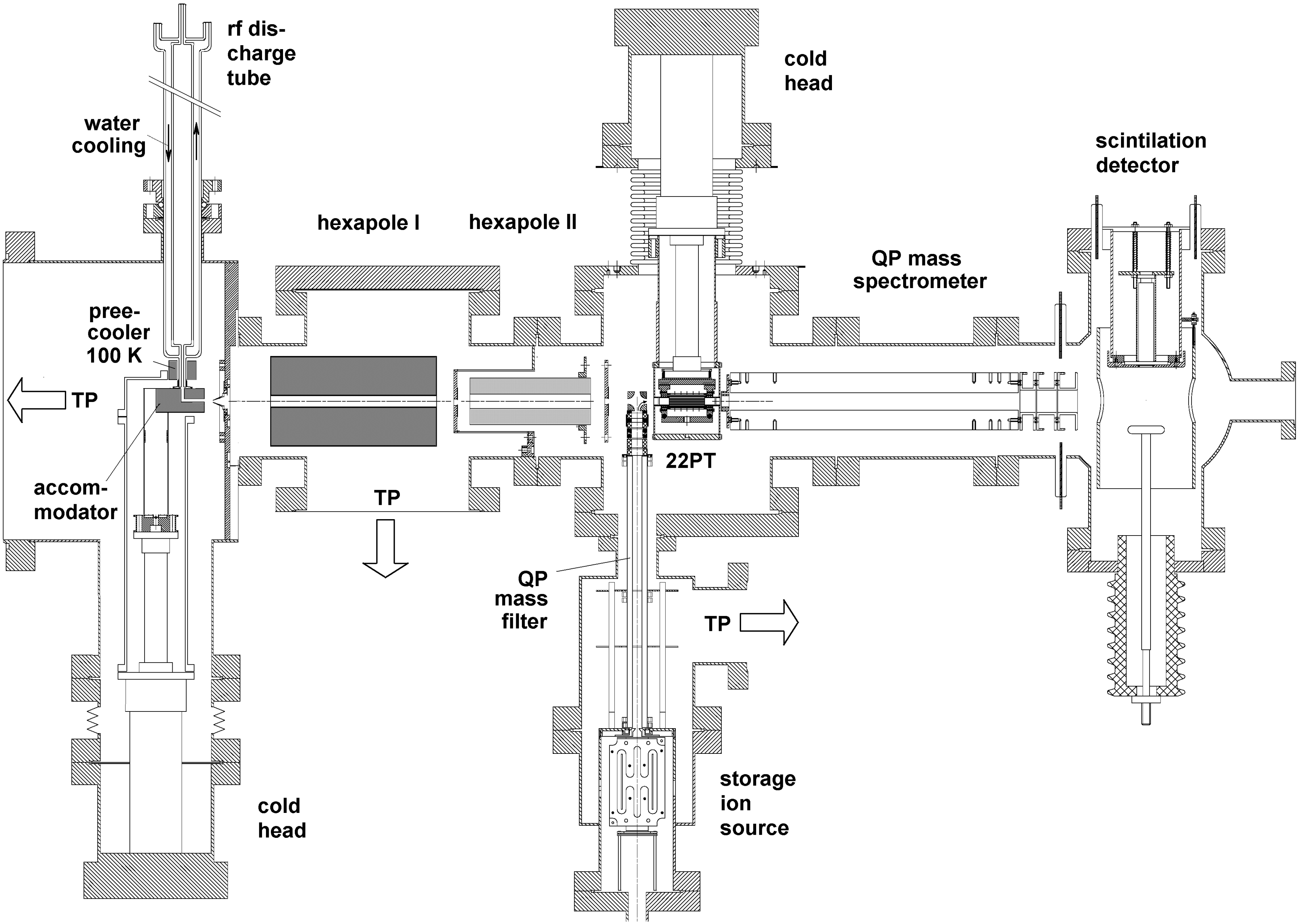
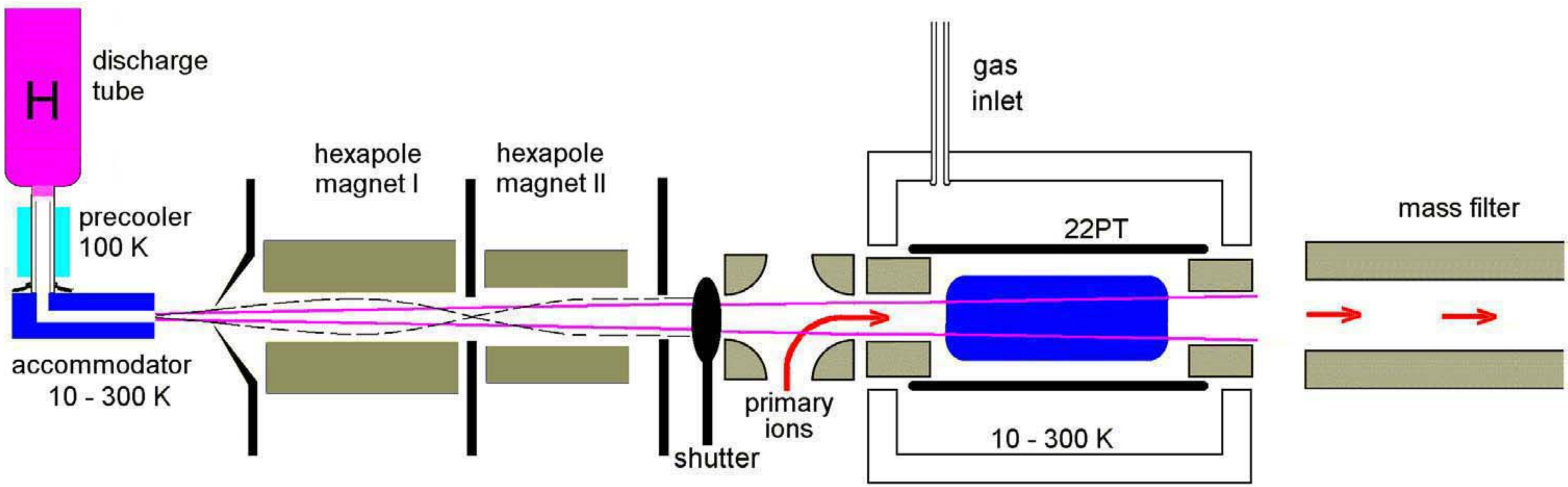
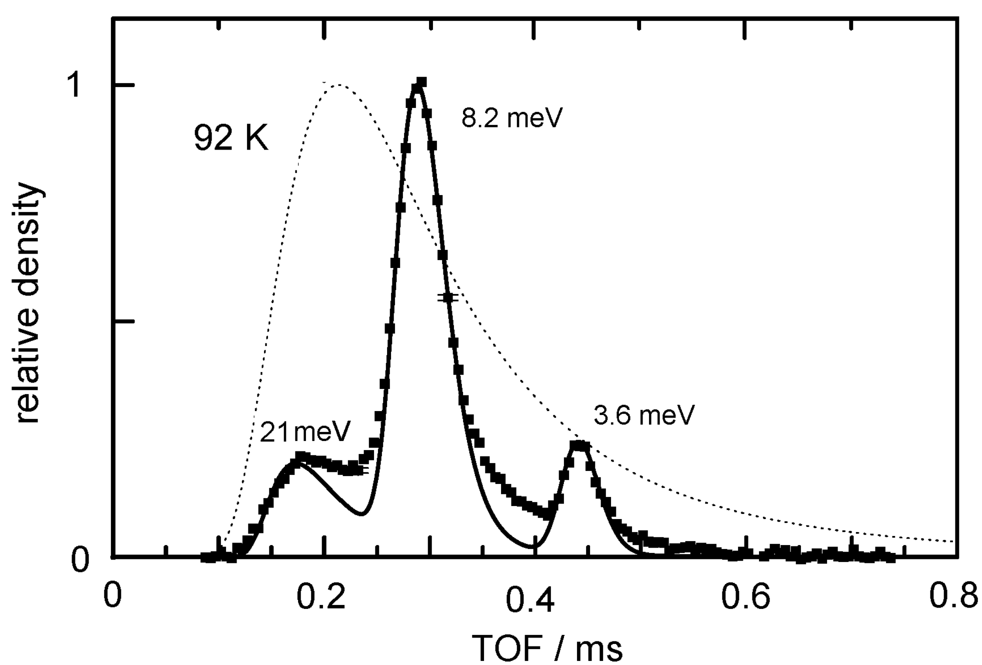
H-atom source + 22pole trap

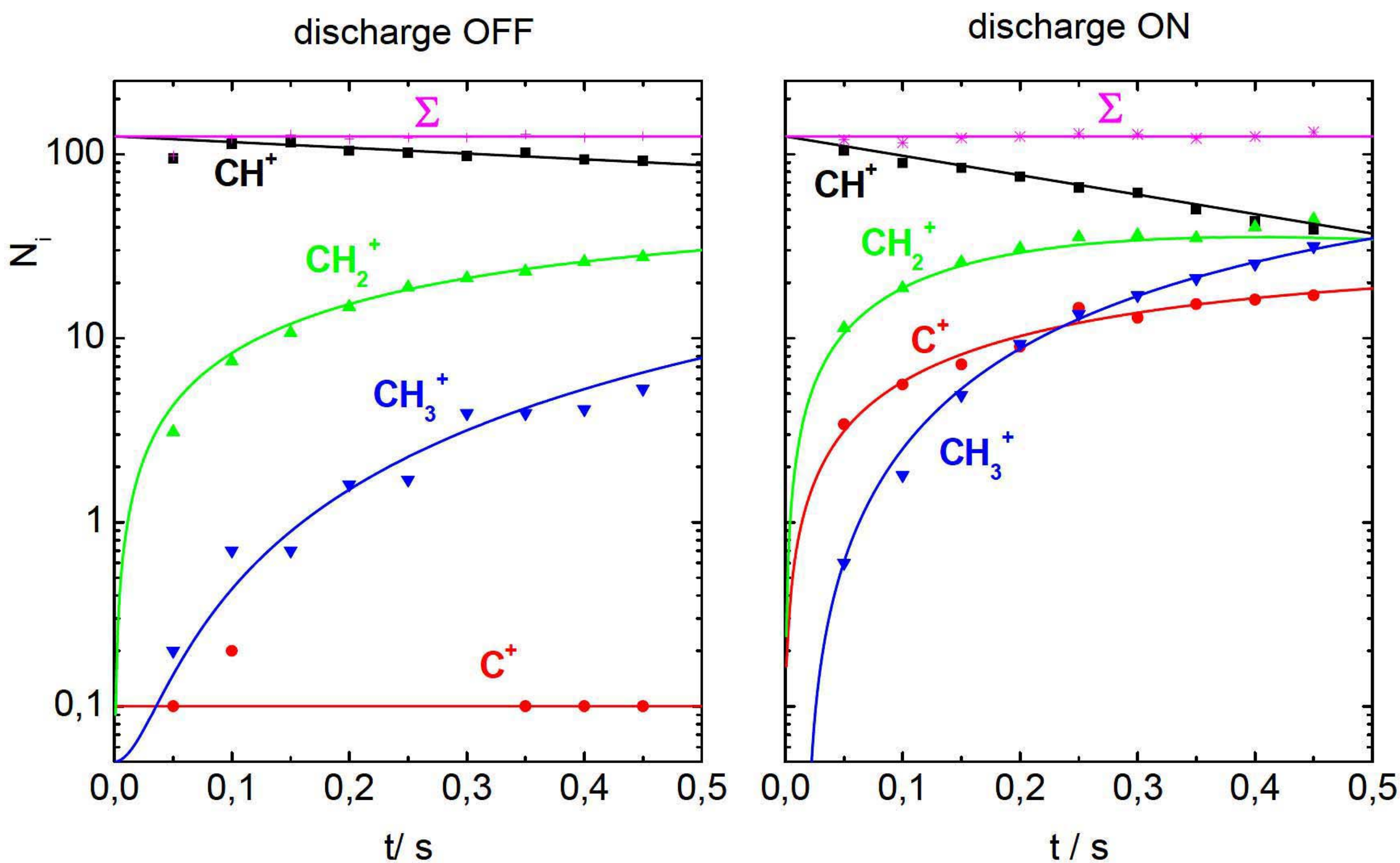


Focusing H atoms



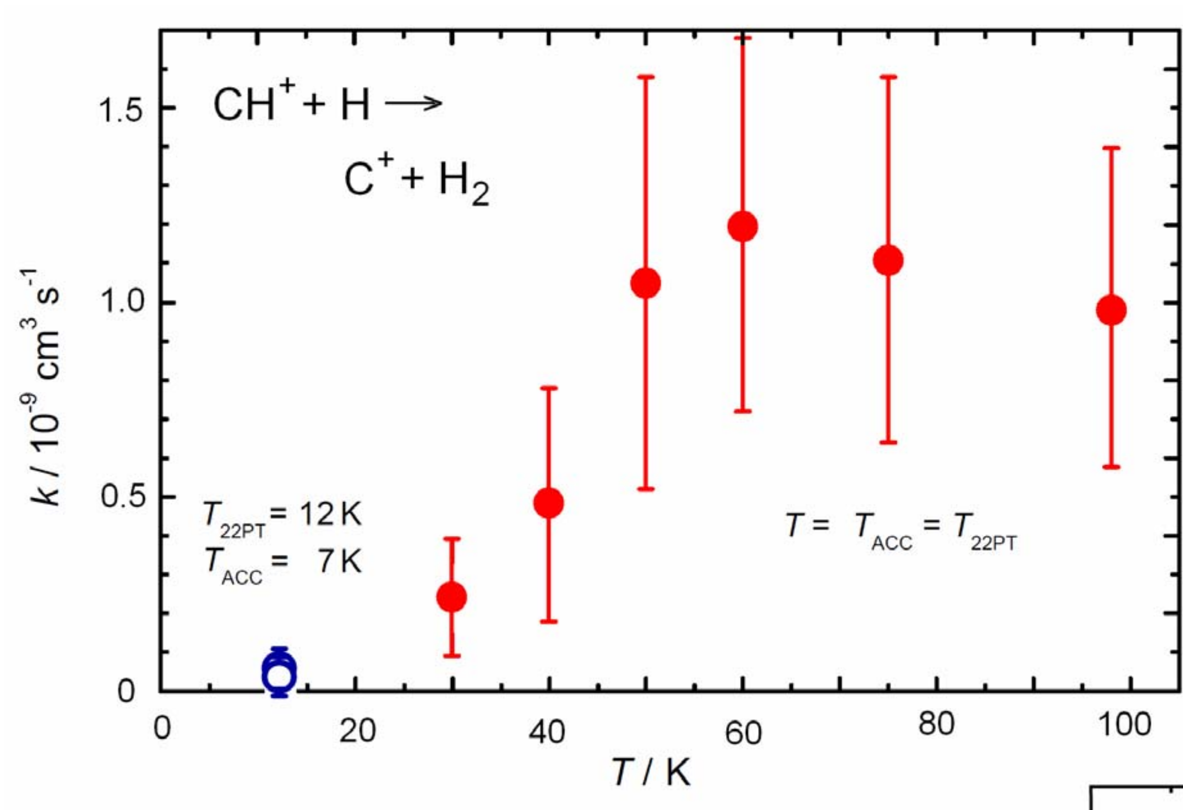


$CH^+ + H_2$ / $CH^+ + H$

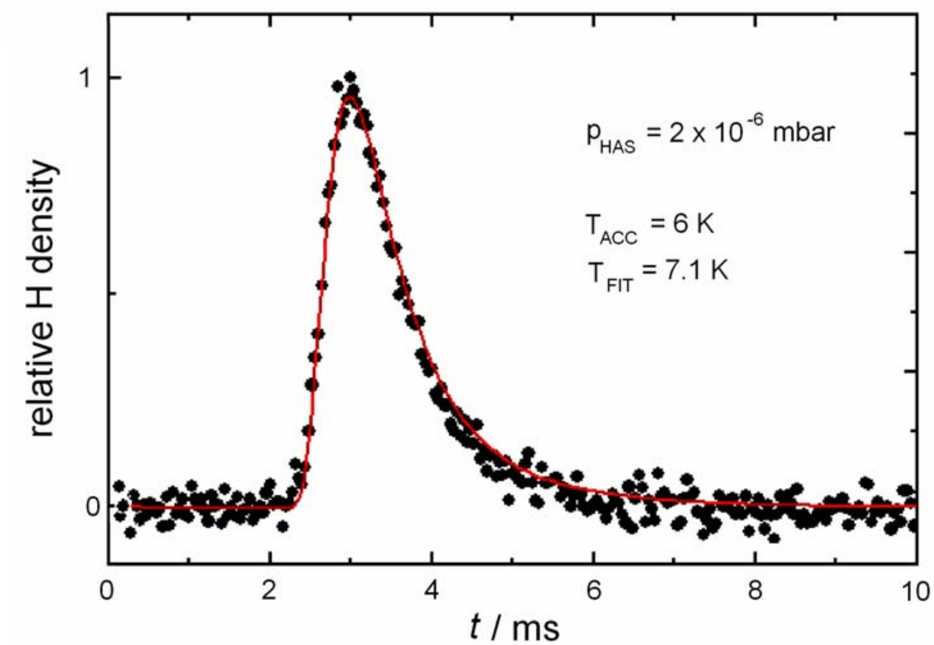


T_{22PT}=50 K
[He] = 1.5×10^{13} cm^{-3}
[H] = 4×10^{8} cm^{-3}
[H_2] = 1.7×10^{9} cm^{-3}

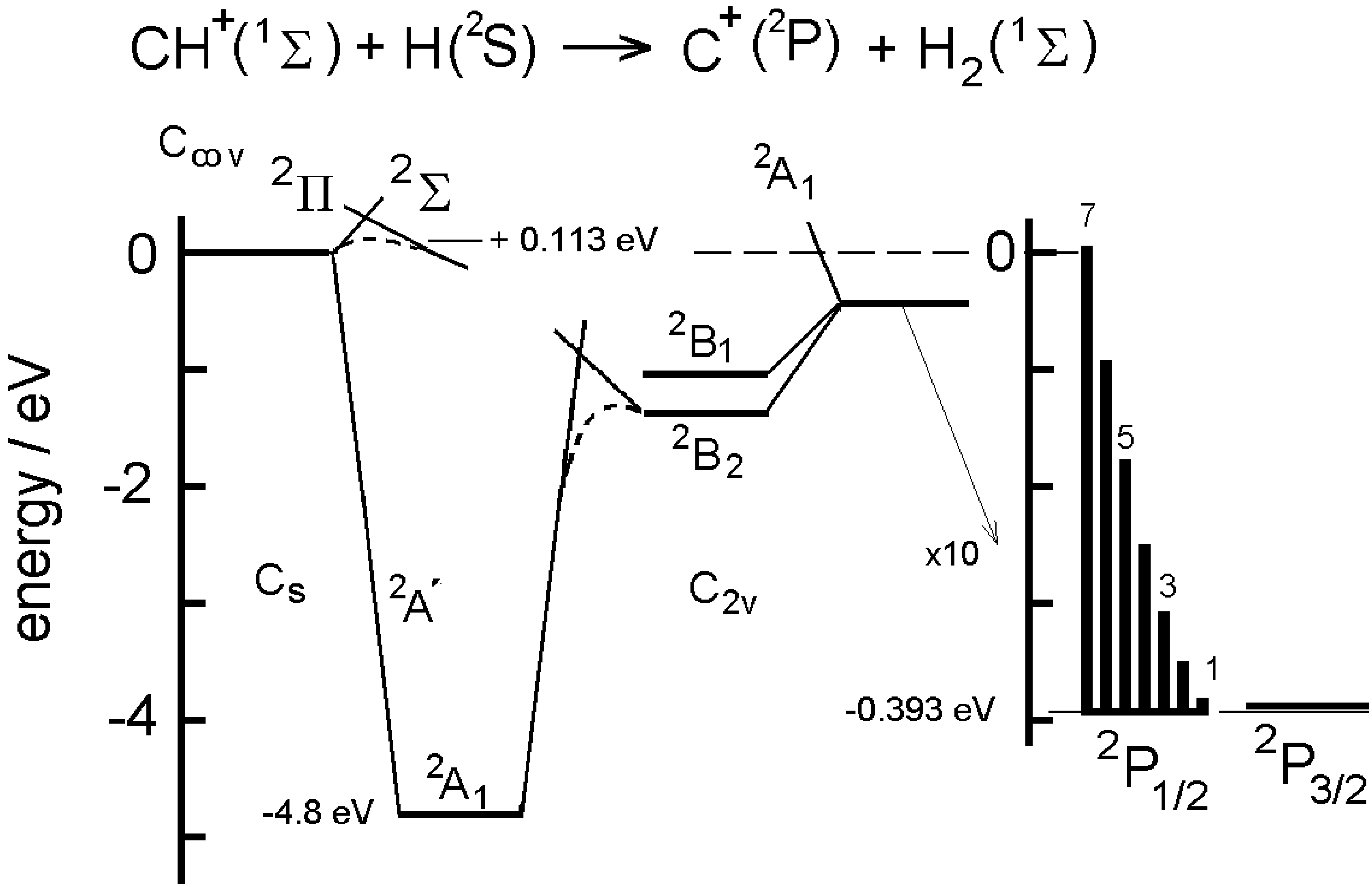
CH^+ + H: barrier!



k (7 K, CH+ (j=0) = $(5 \pm 3) \times 10^{-11}$ $cm^3 s^{-1}$



Where is the barrier?



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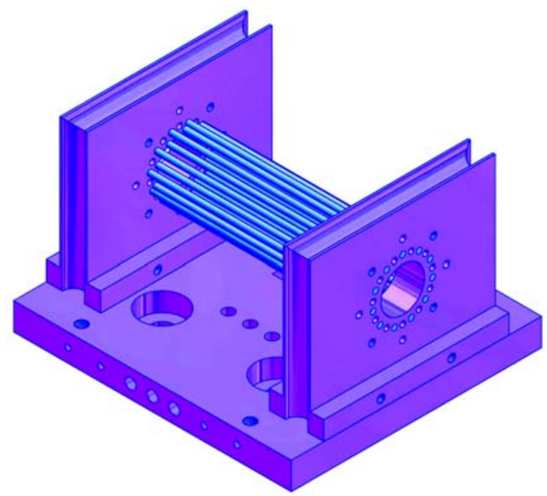


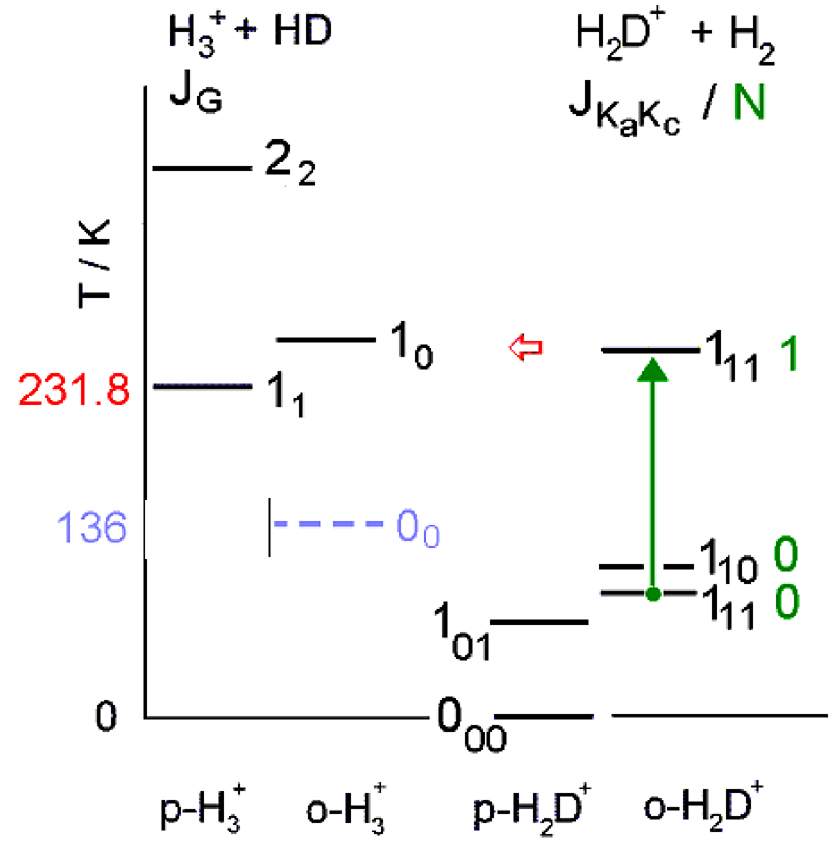
Deuteration $H_3^+ + HD \leftrightarrow H_2D^+ + H_2$: equilibrium constant *K*?

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

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**

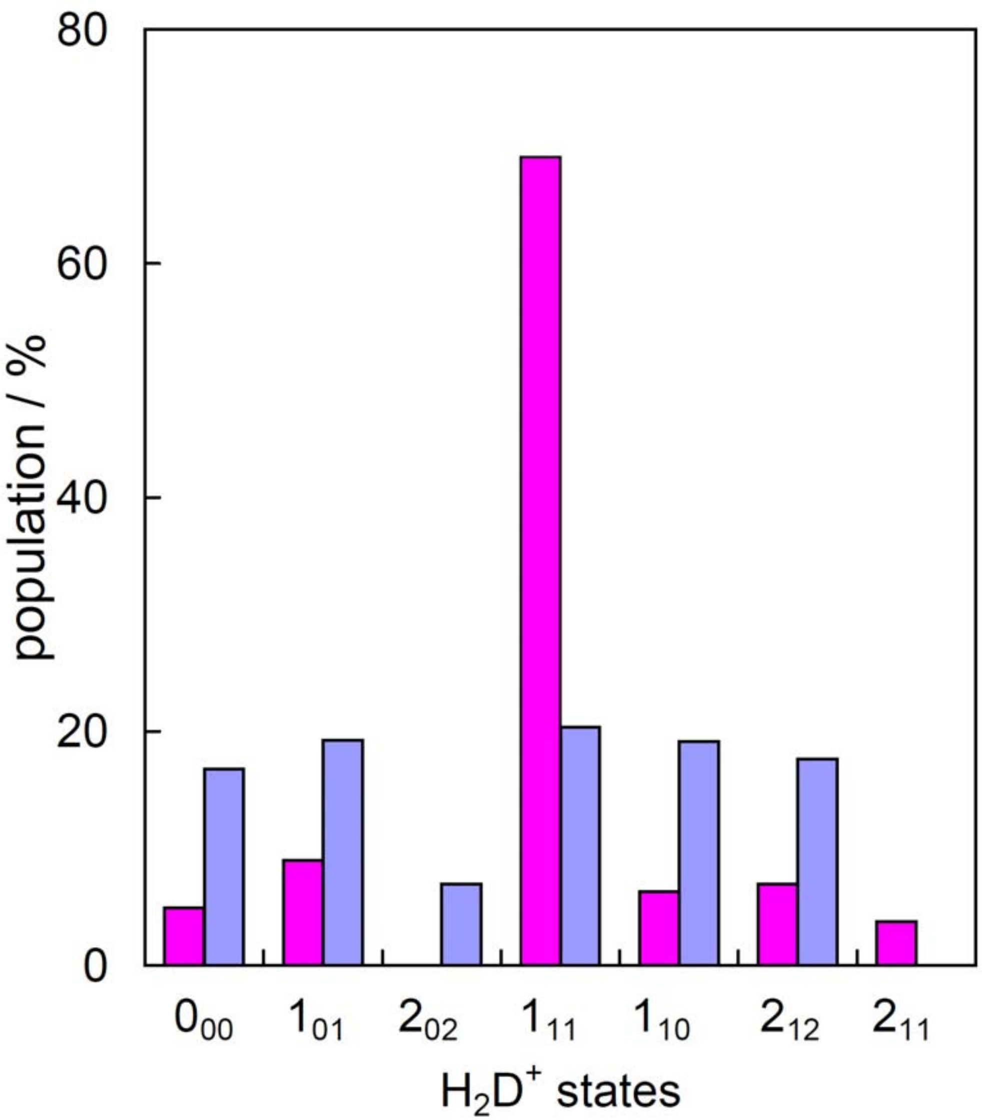


Ramanlal & Tennyson wrote in 2004:
trap experiment disagrees with calculations by
12 orders of magnitude

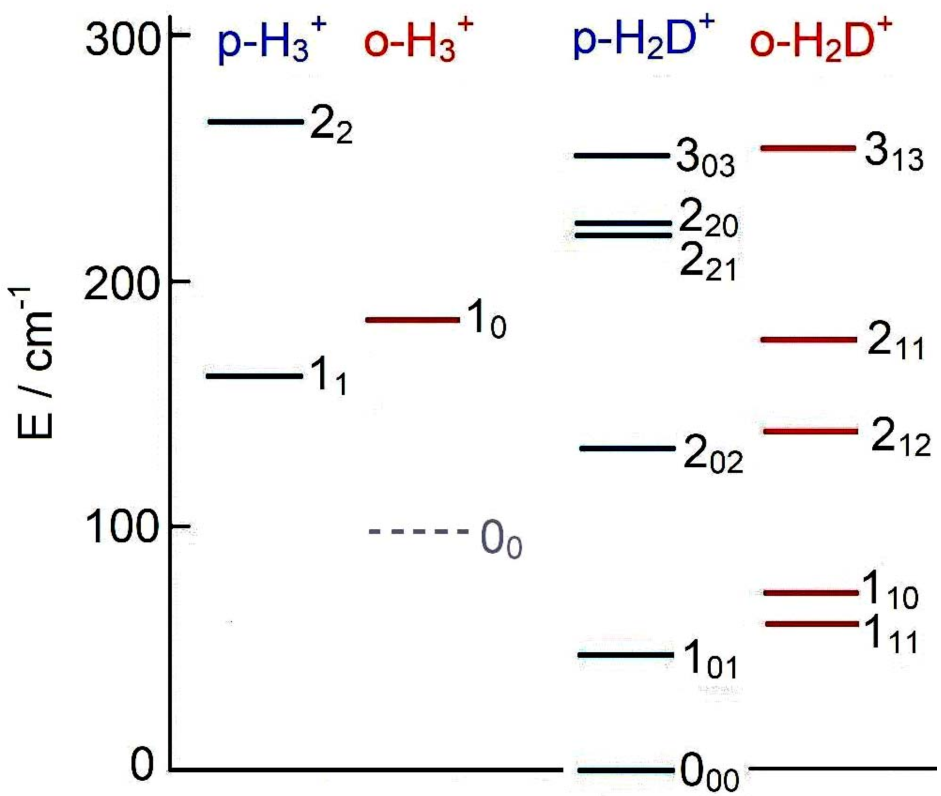
role of o-H_2 (N=1)
state specific k_i(T)
method overtone LIR

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

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

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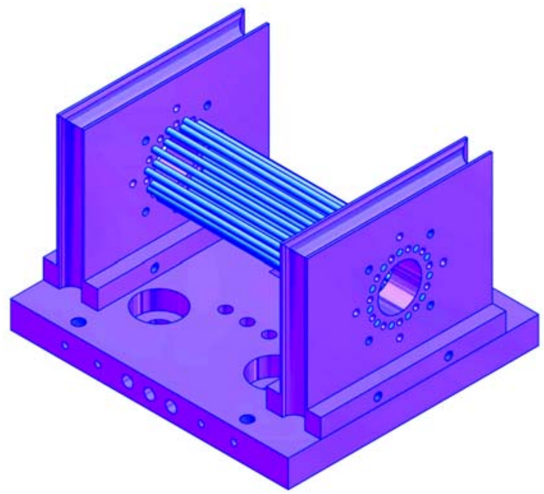
- LIR: N_2^+ + Ar



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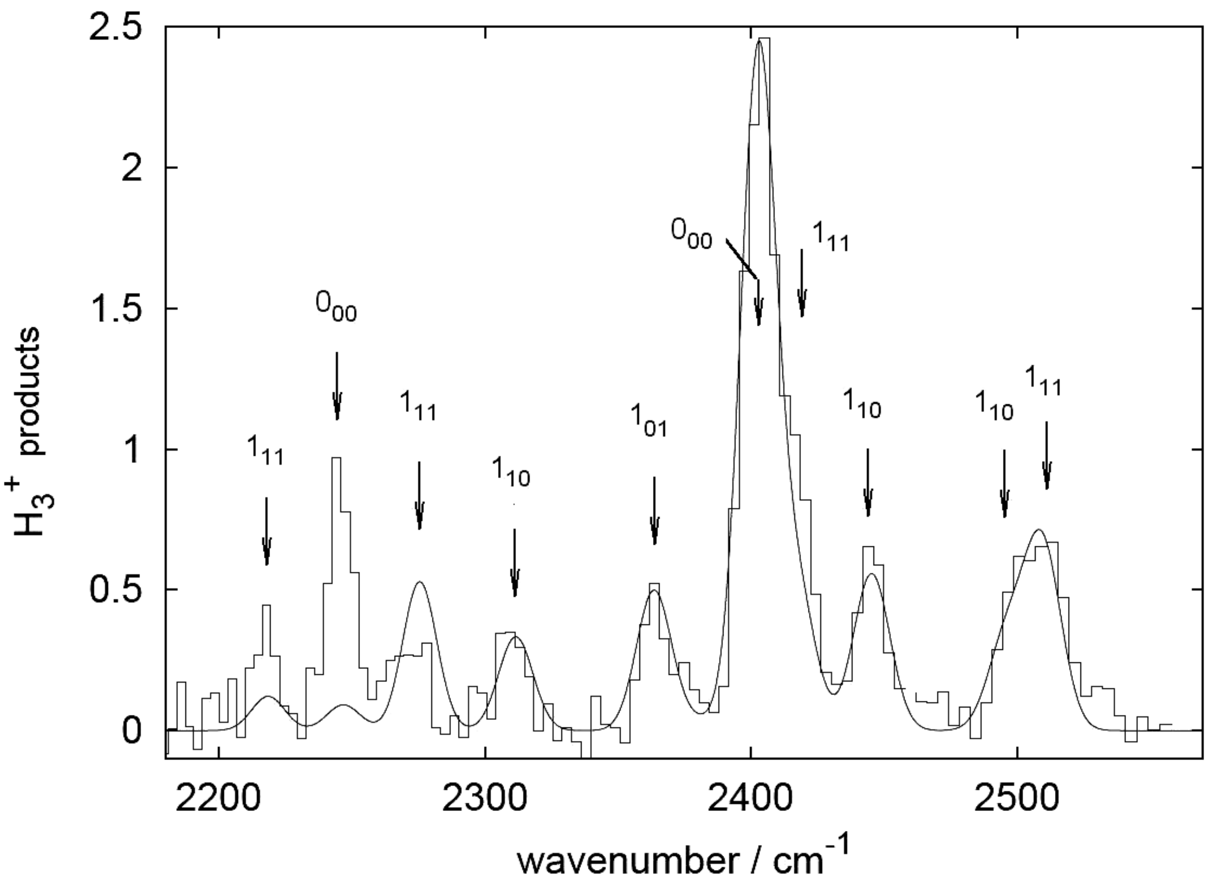
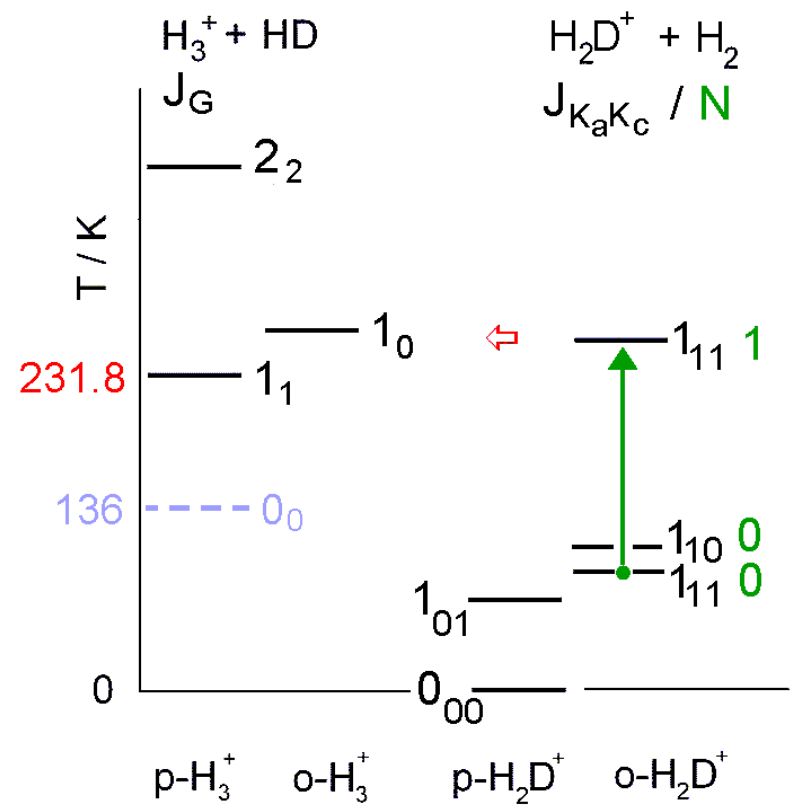
Summary and outlook

LIR spectrum of H_2D^+ (Δv_2 or Δv_3 = 1)

$H_2D^+ + H_2 + h\nu \rightarrow H_3^+$

$[H_2] = 7 \times 10^{10} cm^{-3}$ T = (25 ± 5) K



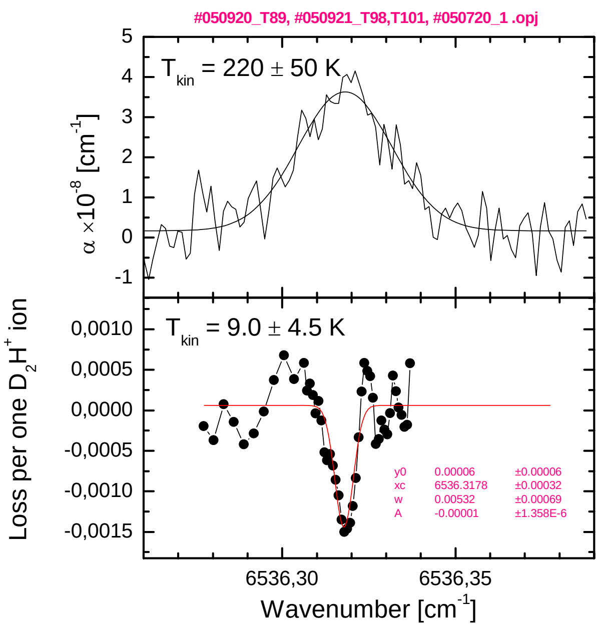


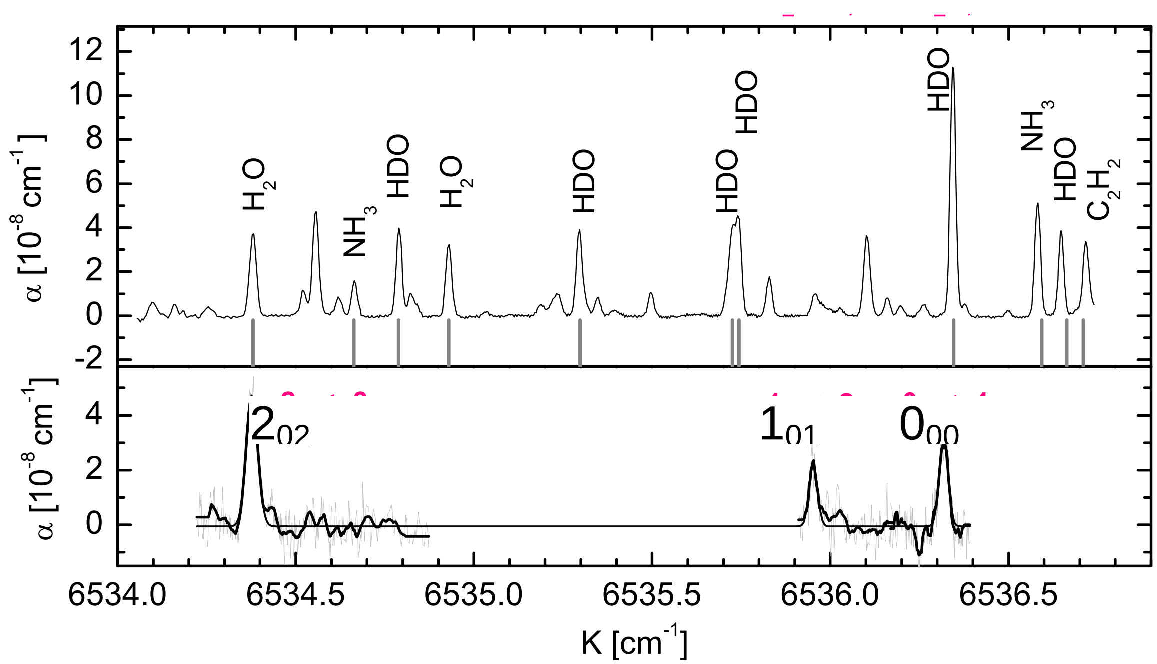
n- H_2 : o : p = 0.75 : 1

"p"- H_2 : o : p = 0.20 : 1

Schlemmer, Asvany, Hugo, Gerlich, Astrochemistry, IAU Symposium 231, 2005

Overtone detection of D_2H^+ (0_{00})





Doppler width

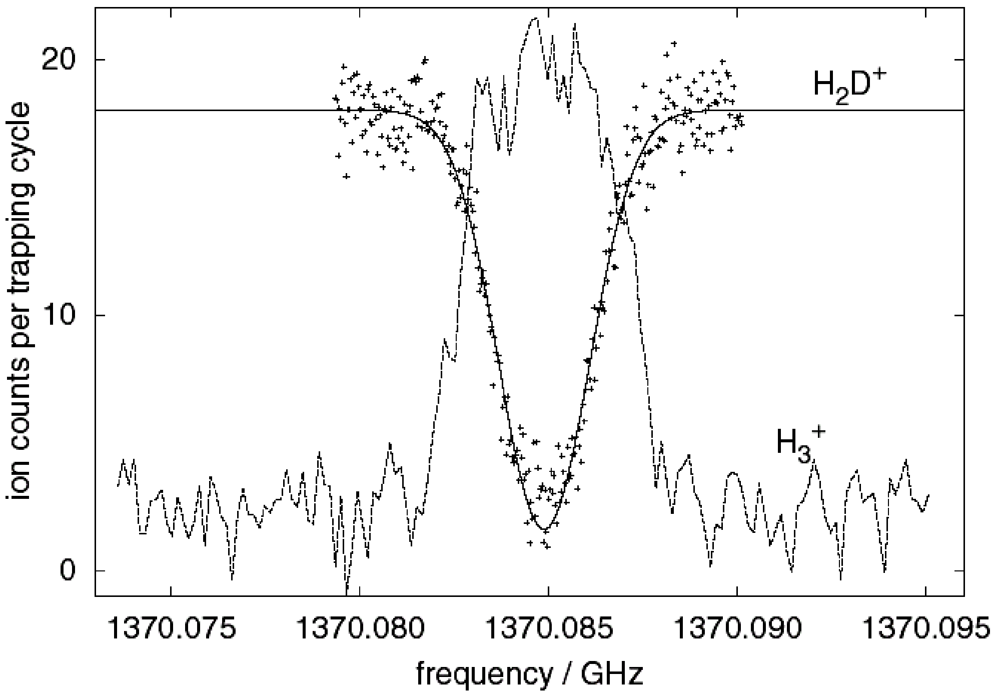
discharge 250 K
trap 9 K

Transition

calc.	6536.301
measured	6536.319

Hlavenka, Plasil, Bano, Korolov, Gerlich, Ramanlal, Tennyson, Glosik Int. J. Mass Spectrometry, **255** (2006) 170

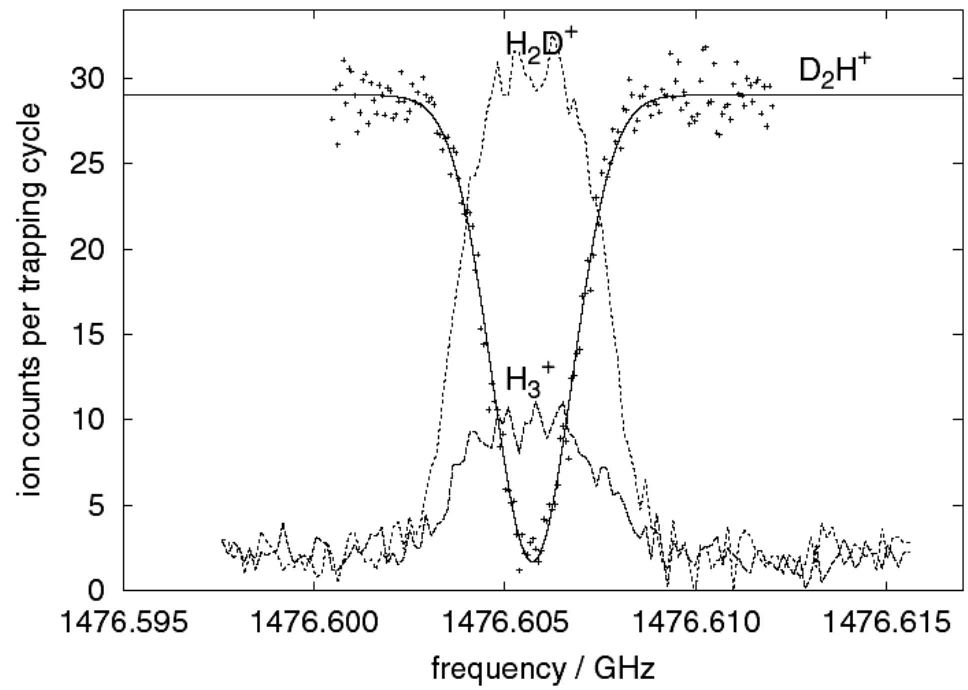
THz radiation induced D-H exchange



First LIR spectra
H_2D^+ 1370084.880(20) MHz
D_2H^+ 1476605.708(15) MHz

TABLE I. Frequencies for the H_2D^+ $1_{01} \leftarrow 0_{00}$ and D_2H^+ $1_{11} \leftarrow 0_{00}$ rotational lines in MHz. The numbers in parentheses give the experimental uncertainty of the last digit.

	H_2D^+ $1_{01} \leftarrow 0_{00}$	D_2H^+ $1_{11} \leftarrow 0_{00}$
this work	1370084.880(20)	1476605.708(15)
ab initio[a]	1369991.8	1476628.0
unpublished value[b]	1370146.0(3)	1476605.5(3)



O. Asvany *et al.* PRL **100** (2008) 233004

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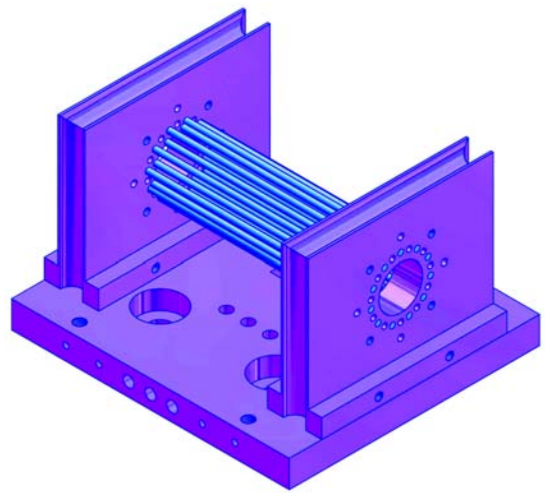

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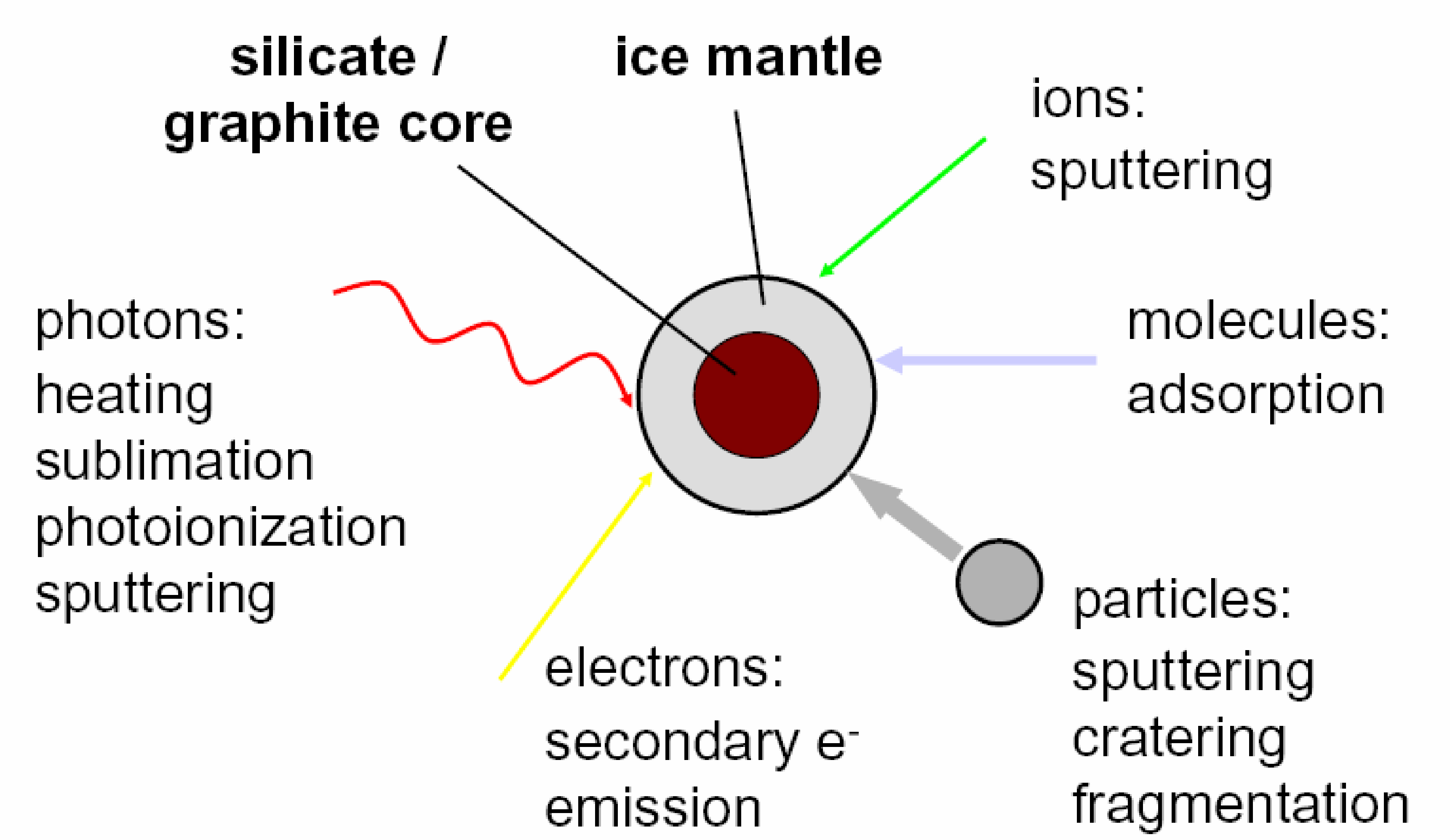
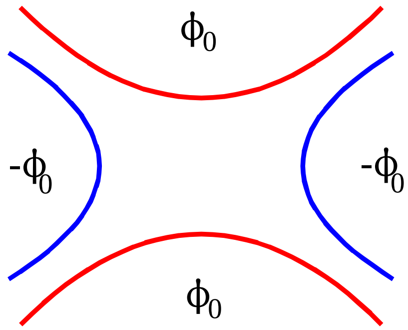
Evaporation and erosion of dust particles



Theory of Quadrupole Trap



Applied Field:

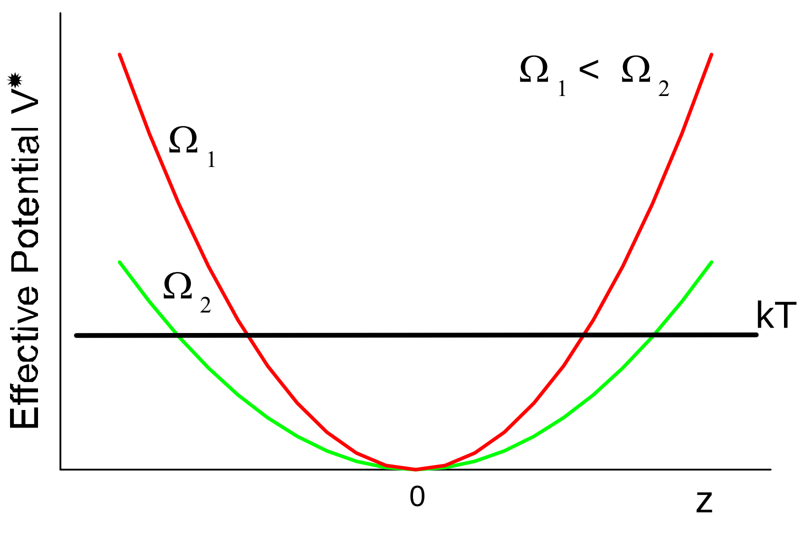
$\Phi_0 = U_0 - V_0 \cos(\Omega t)$

Secular Motion:

$$\omega_z = \frac{q_z}{\sqrt{2}} \frac{\Omega}{2}$$

Effective Potential:

$E_{pot} = 1/2\, M\omega^2 z^2$



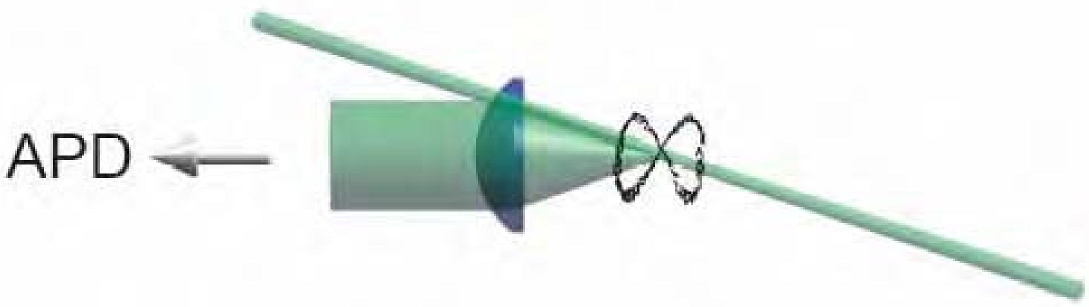
Stability Parameter:

$$q_z = \frac{4\,Q\,\Omega}{M\,z_0^2\,\Omega^2} < 0.3$$

Q/M-Determination:

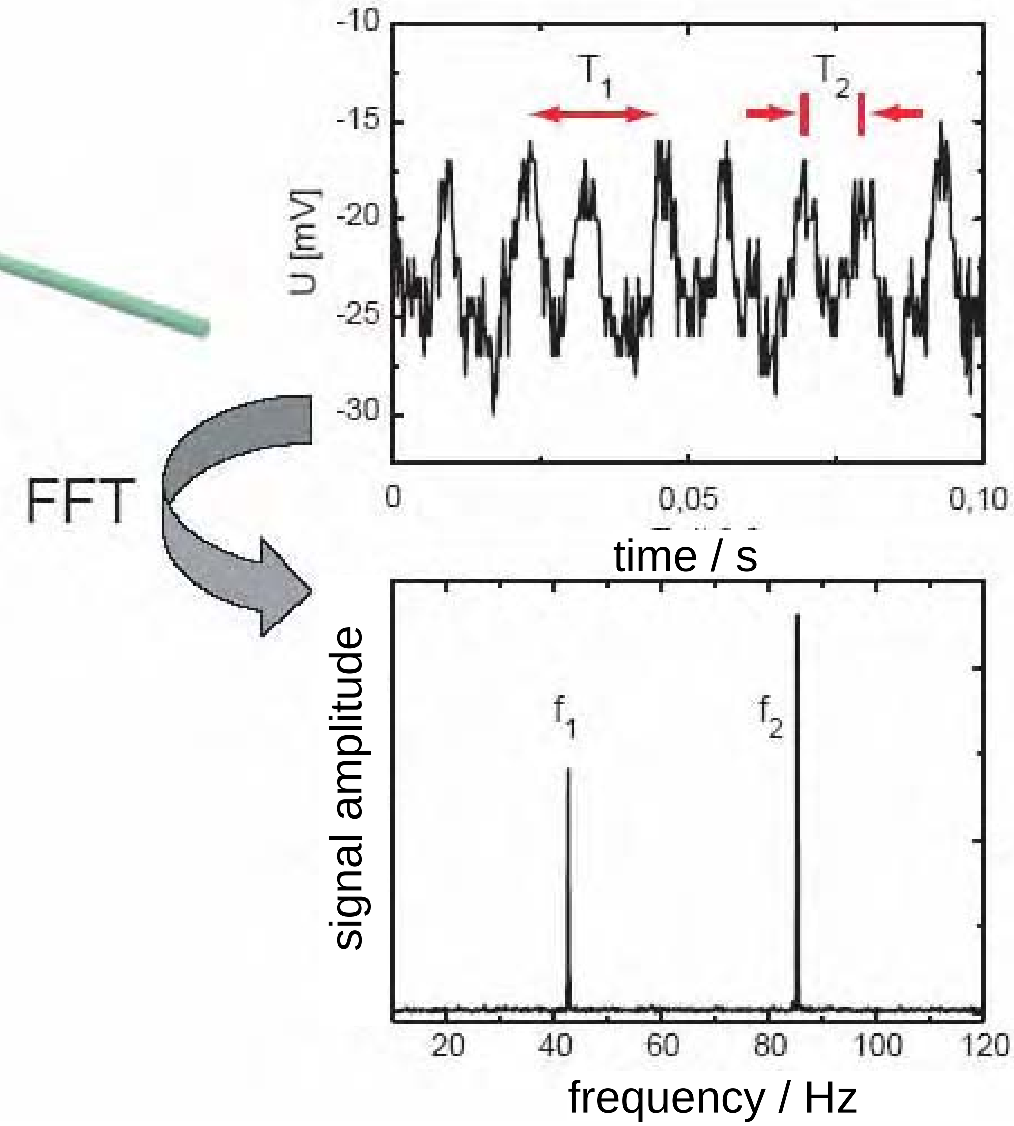
$$Q/M = \frac{z_0^2\,\omega_z\,\Omega}{\sqrt{2}\,V_0}$$

Determination of q/m

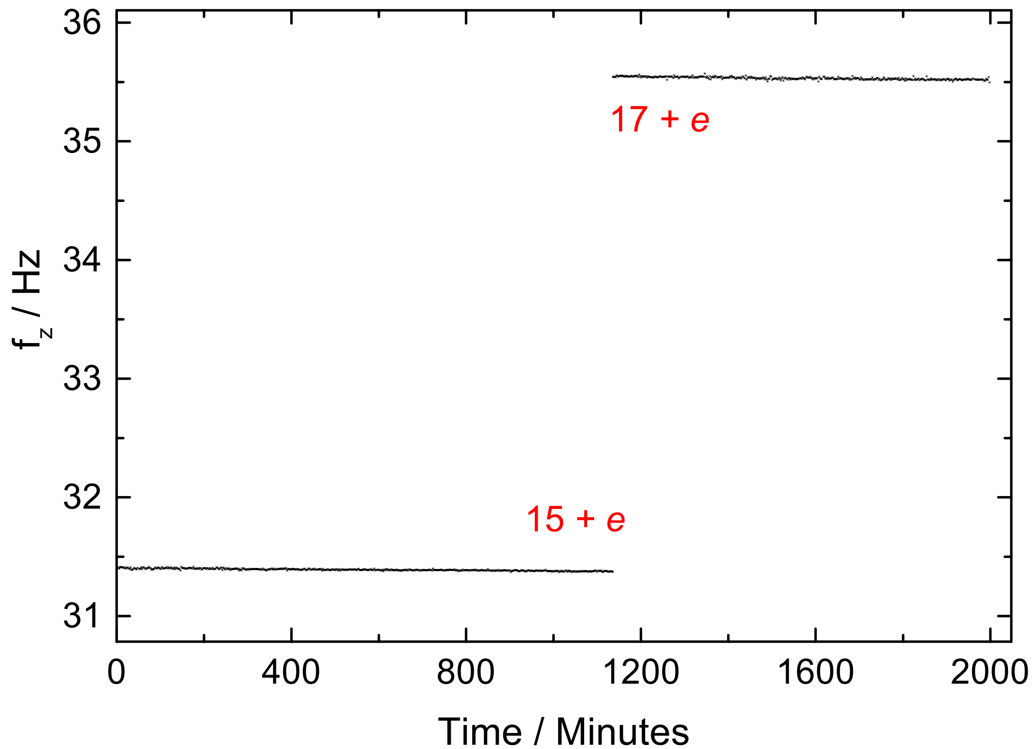


$\omega_r = 2\pi \cdot 42.73$ Hz
$\Omega = 2\pi \cdot 928$ Hz
$V_0 = 1365$ V

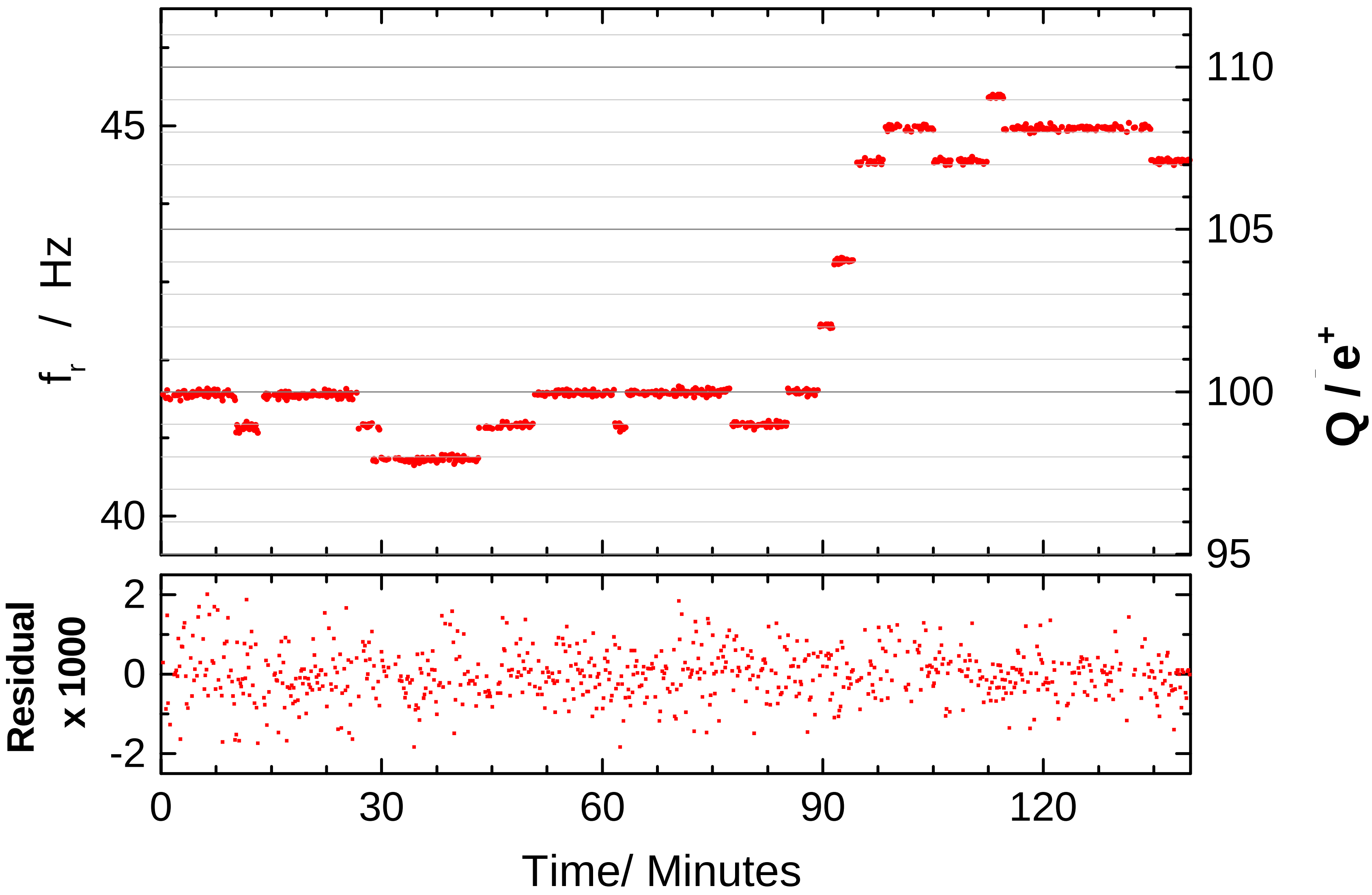
$Q/M = 98.13$ mC/kg
$d\omega/\omega = 6 \cdot 10^{-5}$



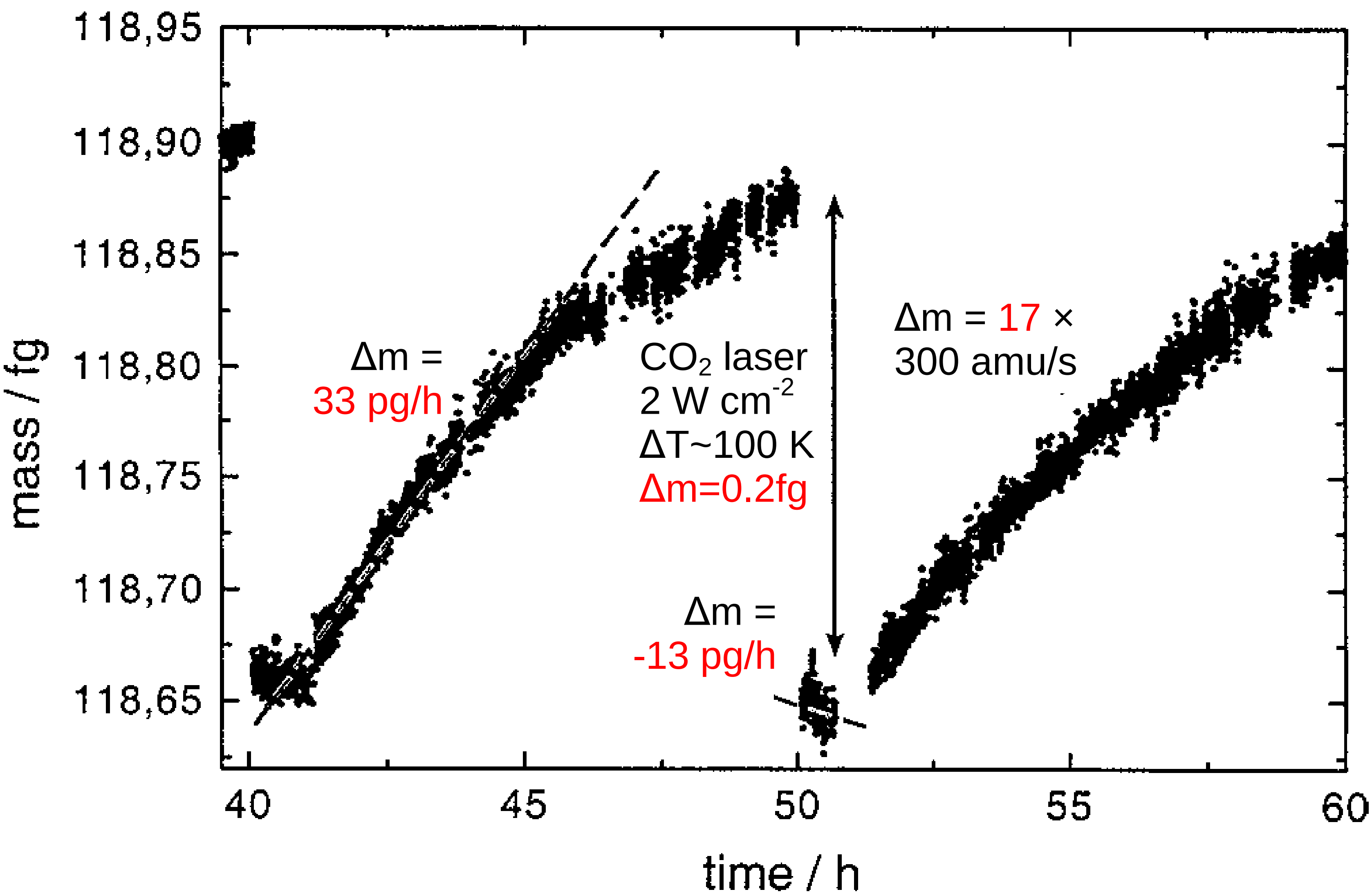
Cosmic rays induced change



Absolute charge state

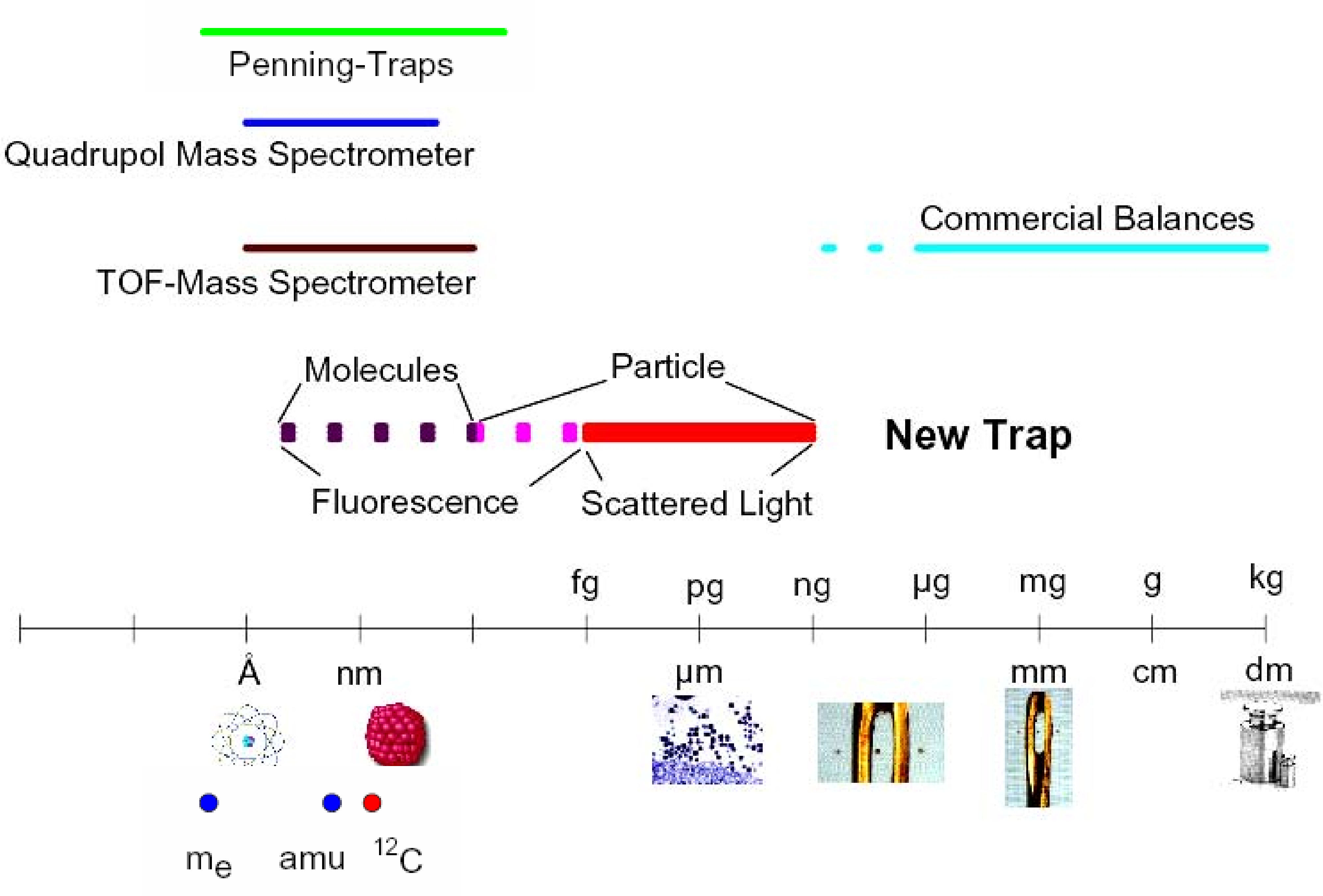


Gas ad- and desorption
500 nm diameter SiO_2 sphere



Molecular Ions and Nanoparticles in RF and AC Traps
D. Gerlich, Hyperfine Interactions 146 (2003) 293

mass spectrometers and scales



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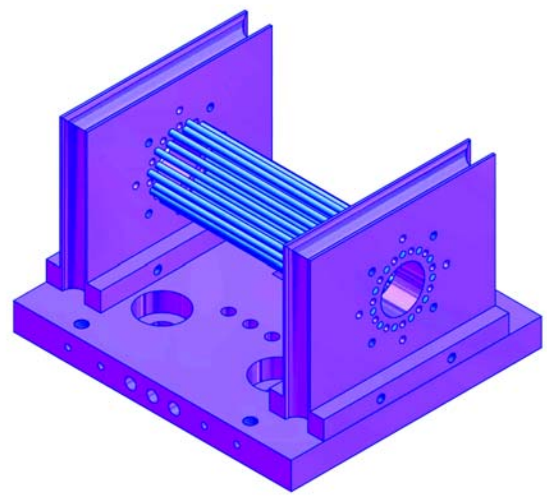

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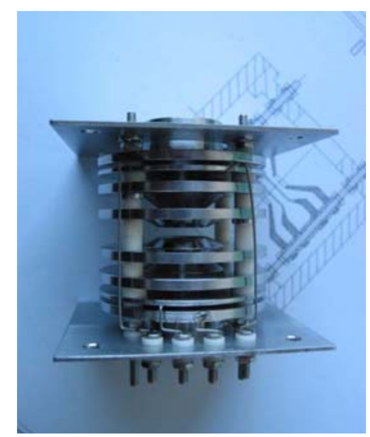

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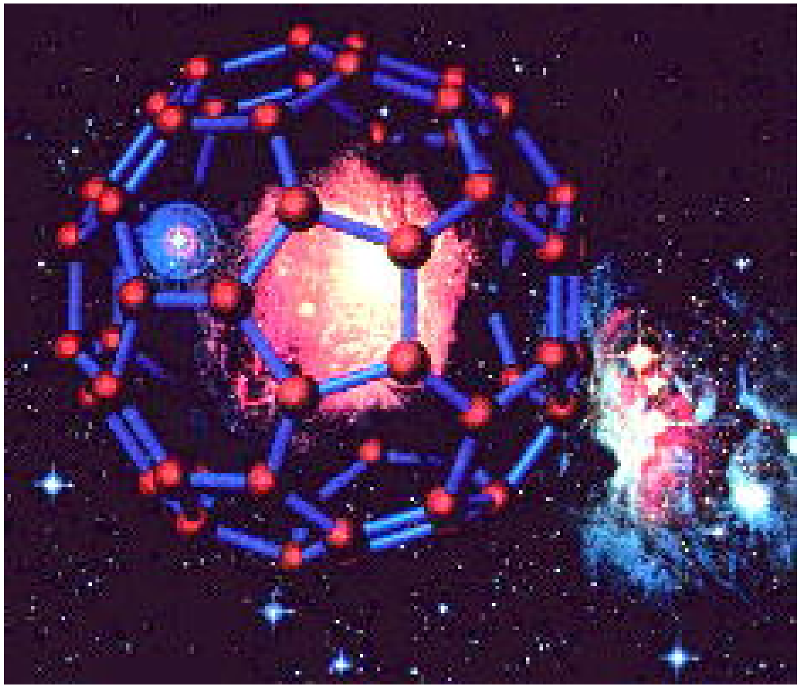
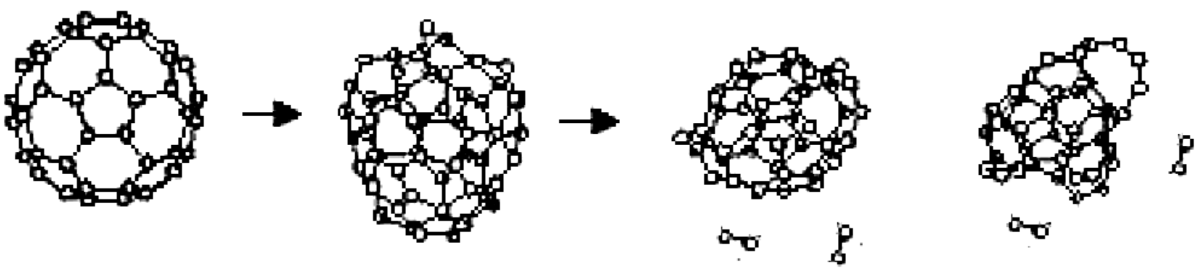
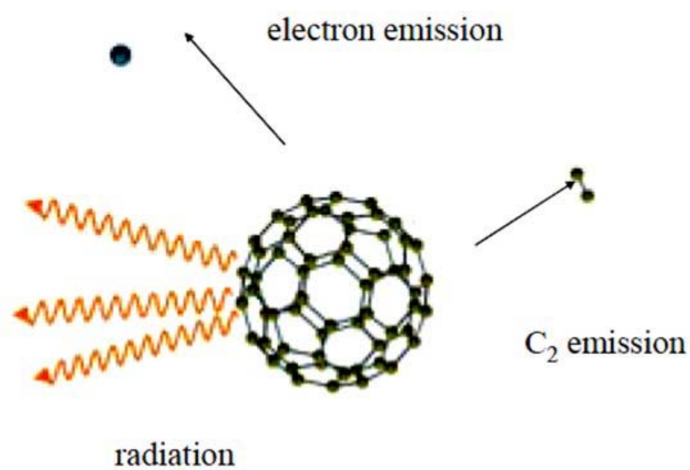
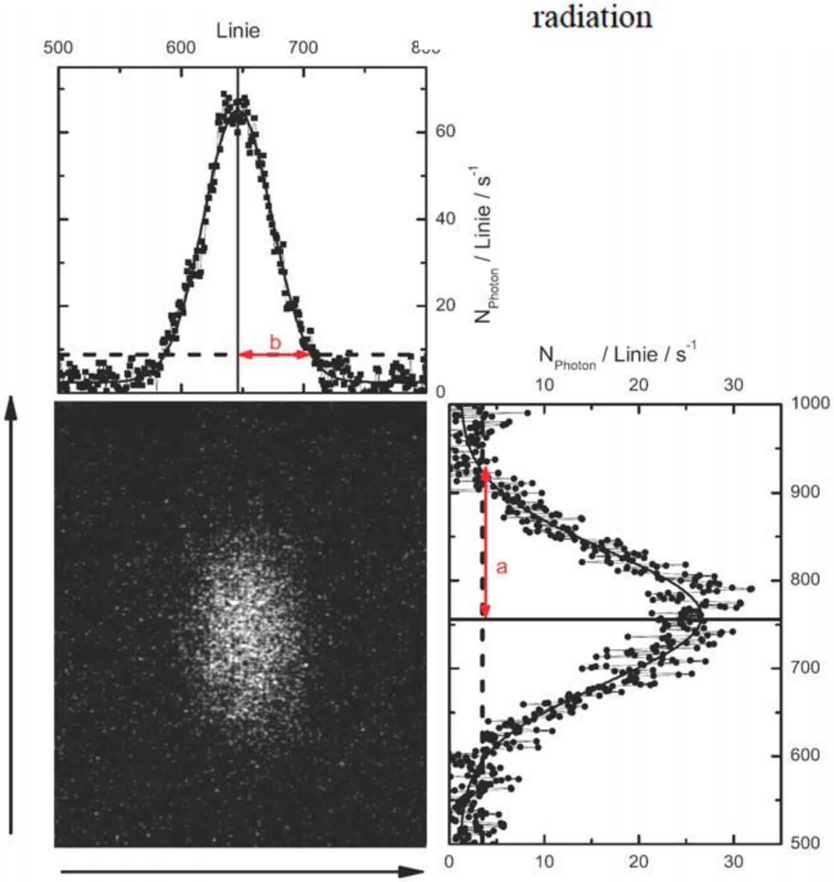
Hot carbon nanoparticles

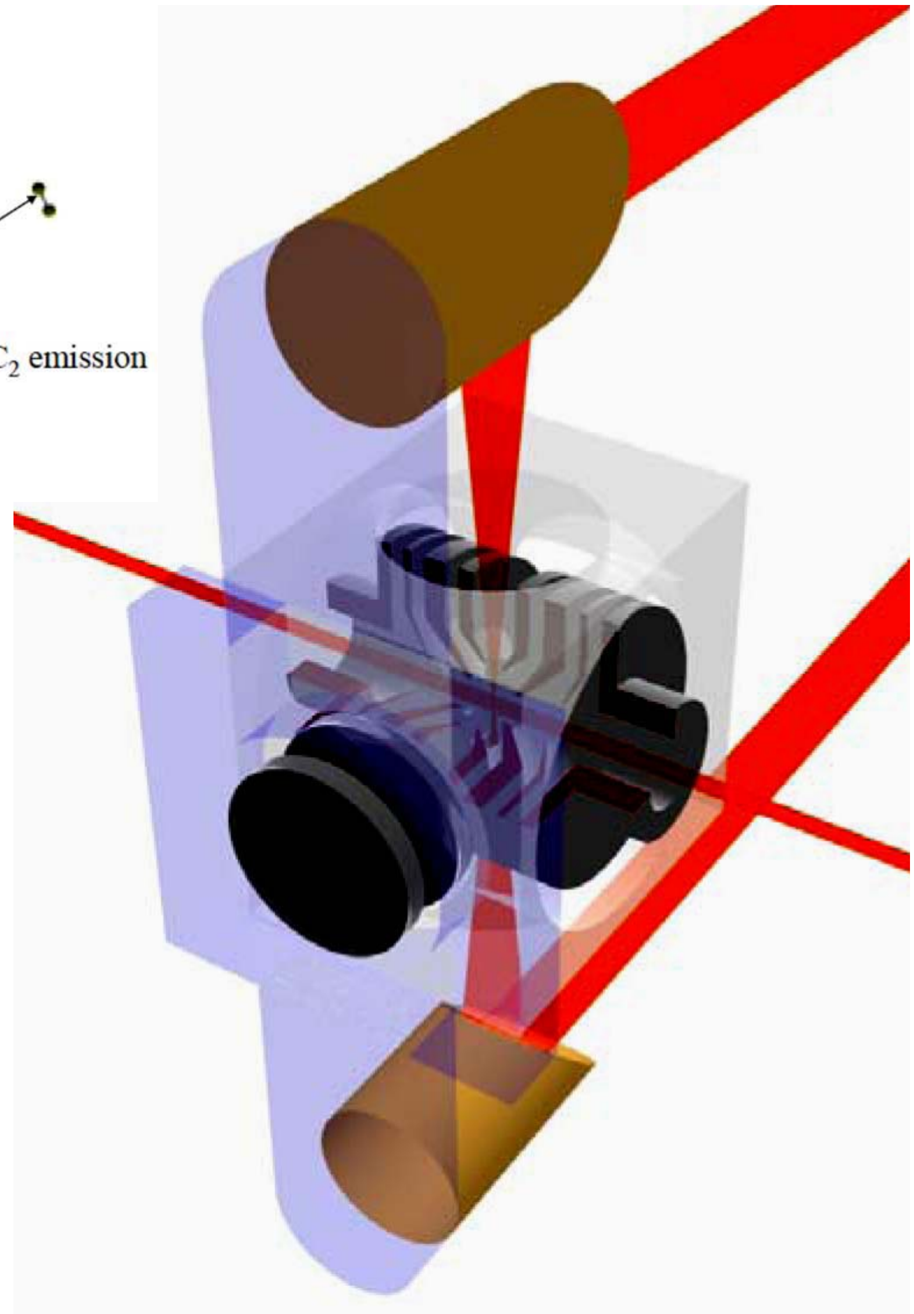




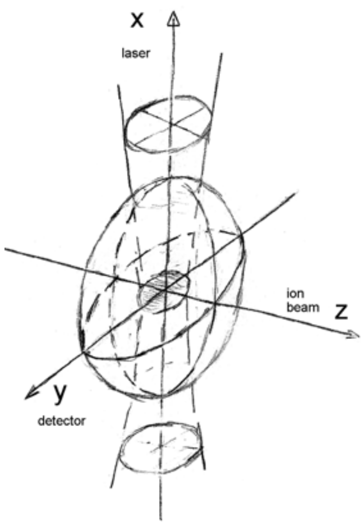
Ions at high temperatures

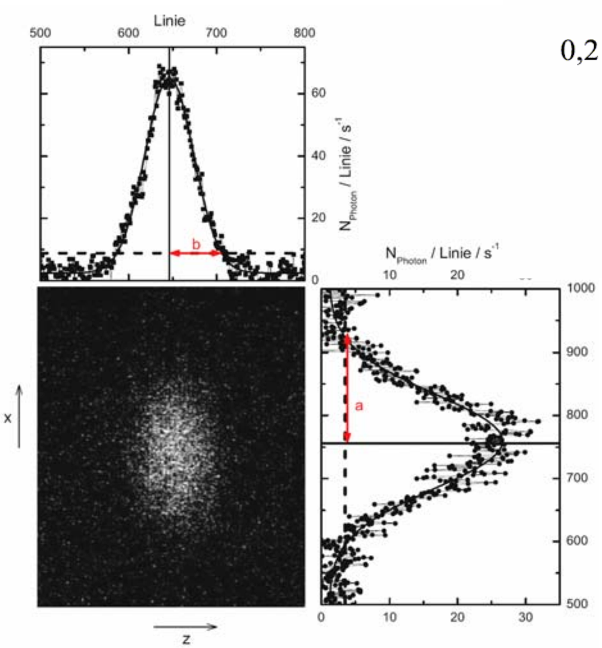


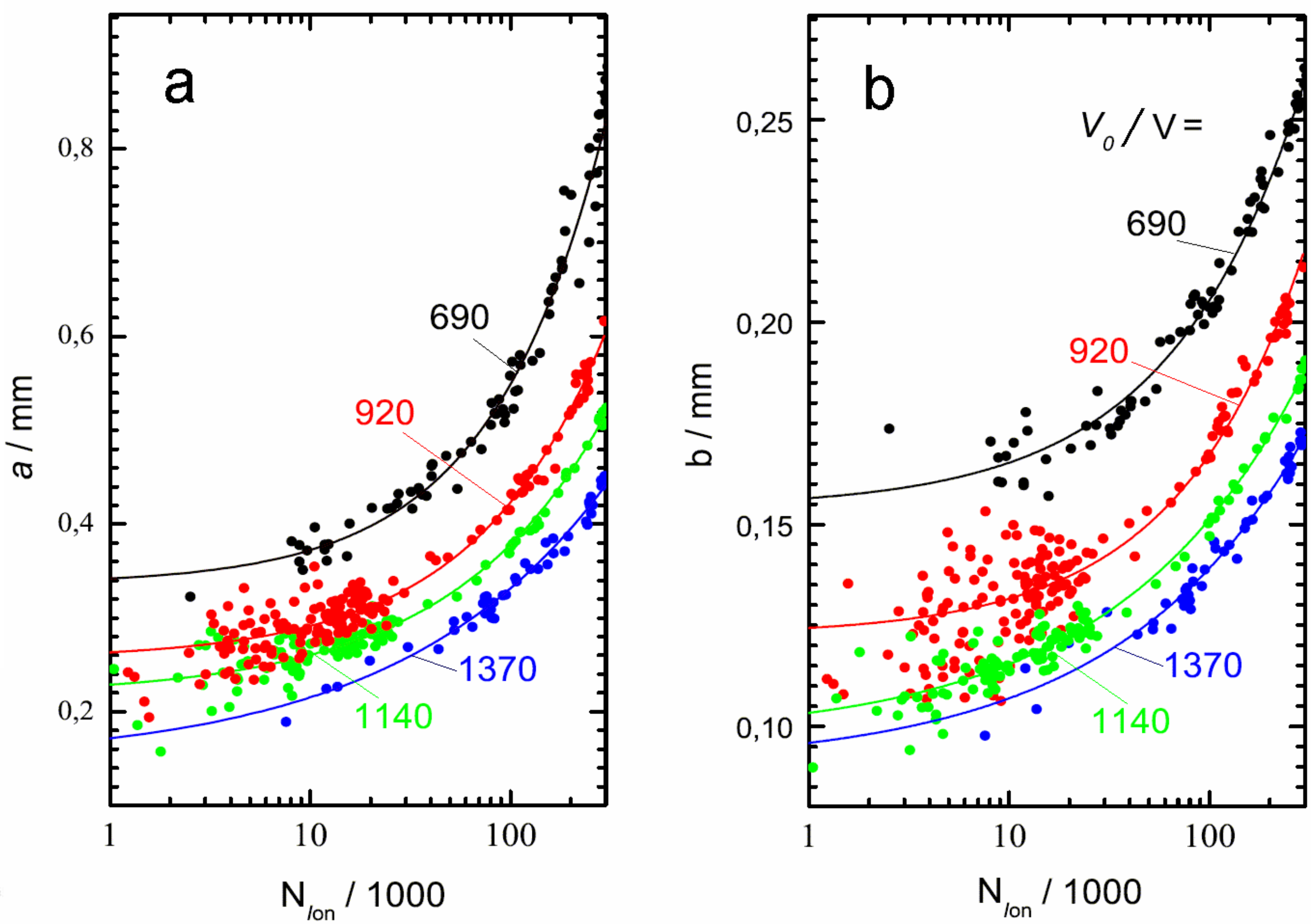




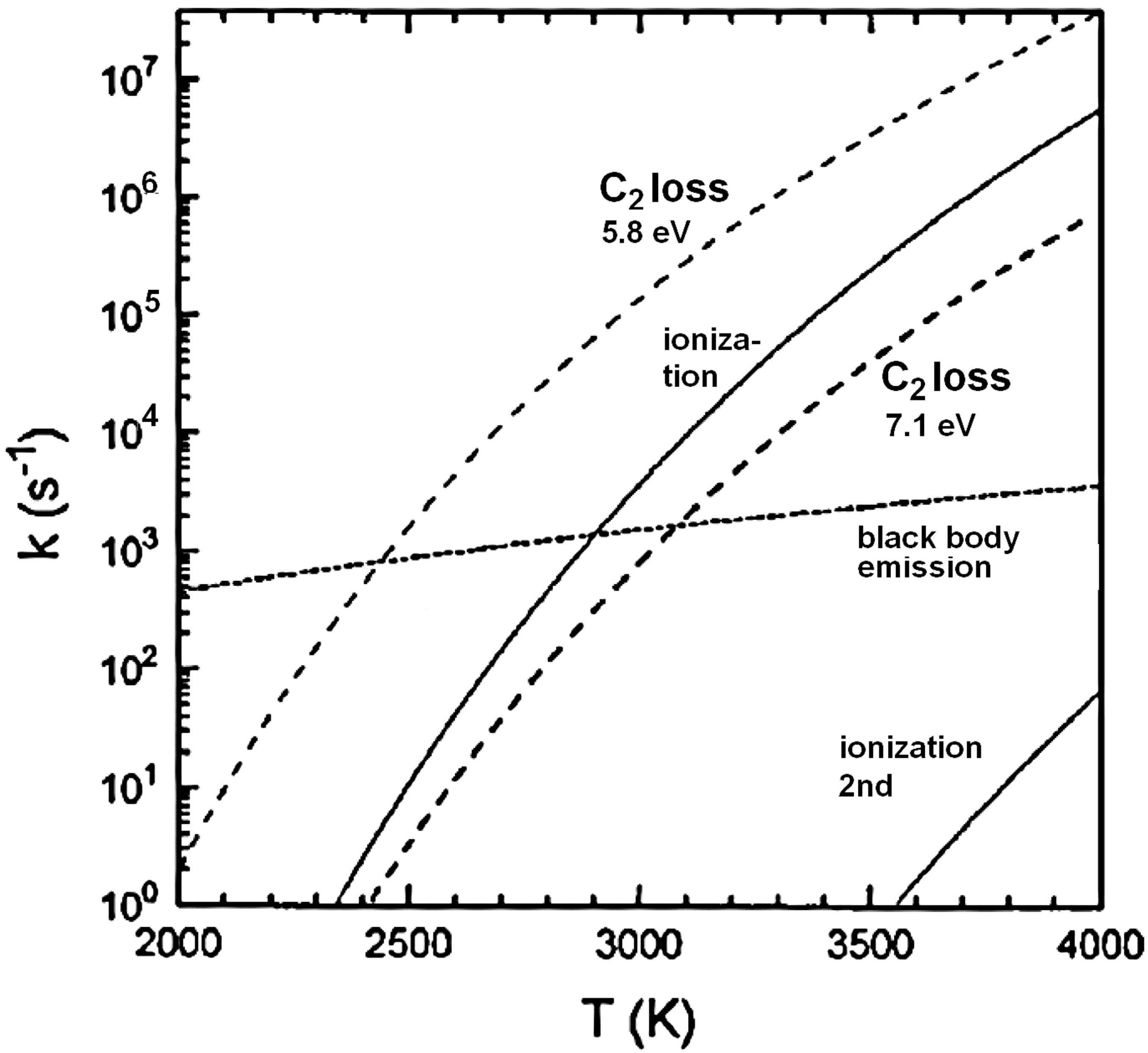
Space charge, amplitude dependence





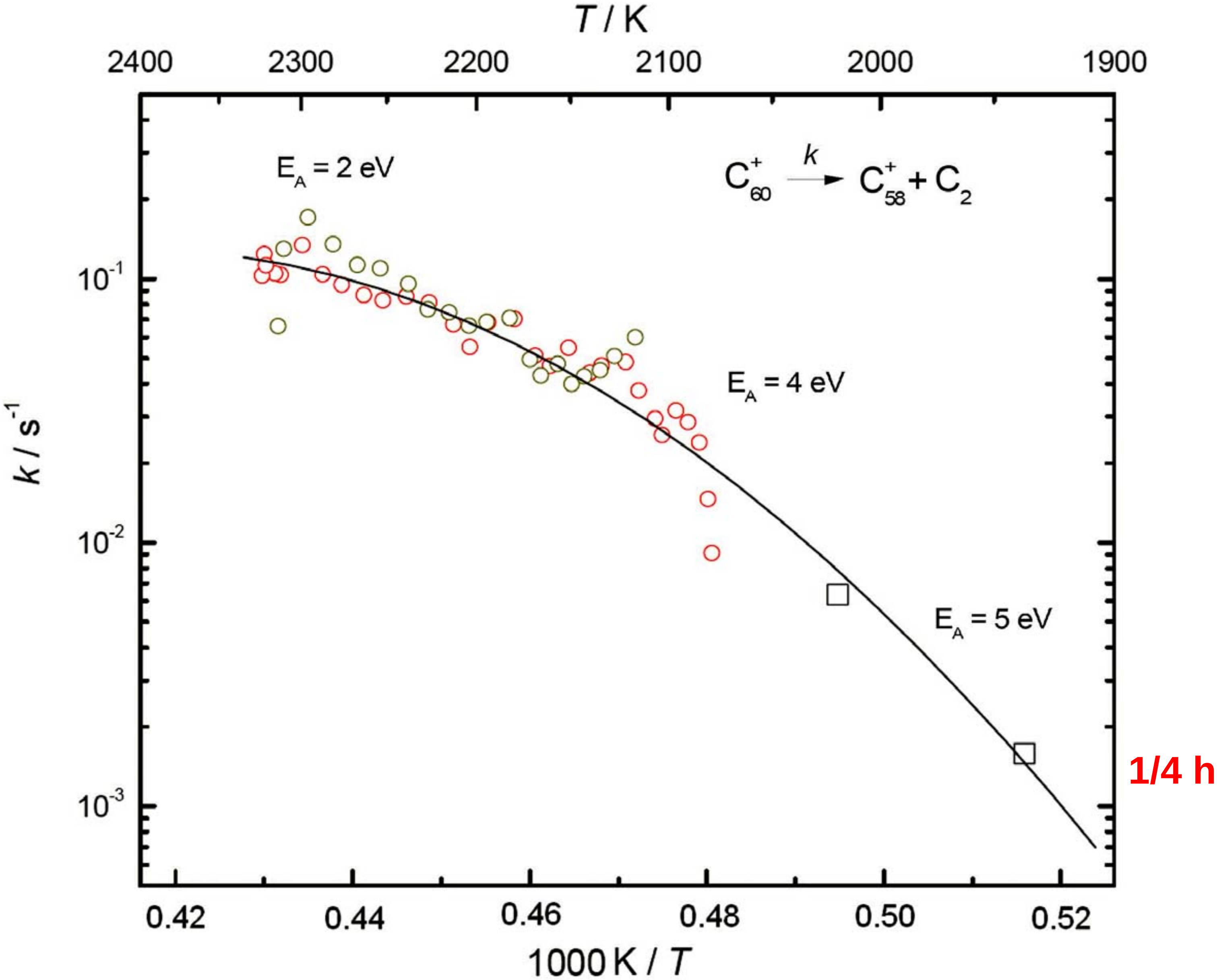


Cooling hot C_{60}

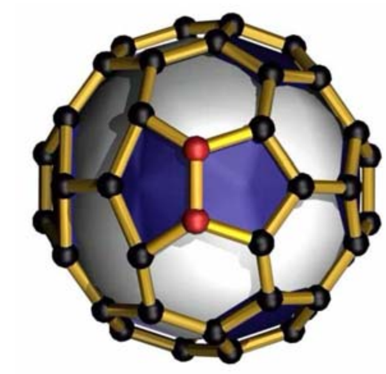


R. Mitzner, J. Chem. Phys. **103** (1995) 2445

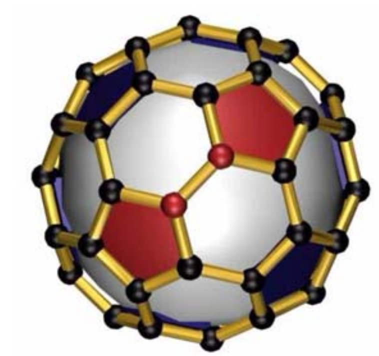
Thermal decay of C_{60}^+



C2 elimination



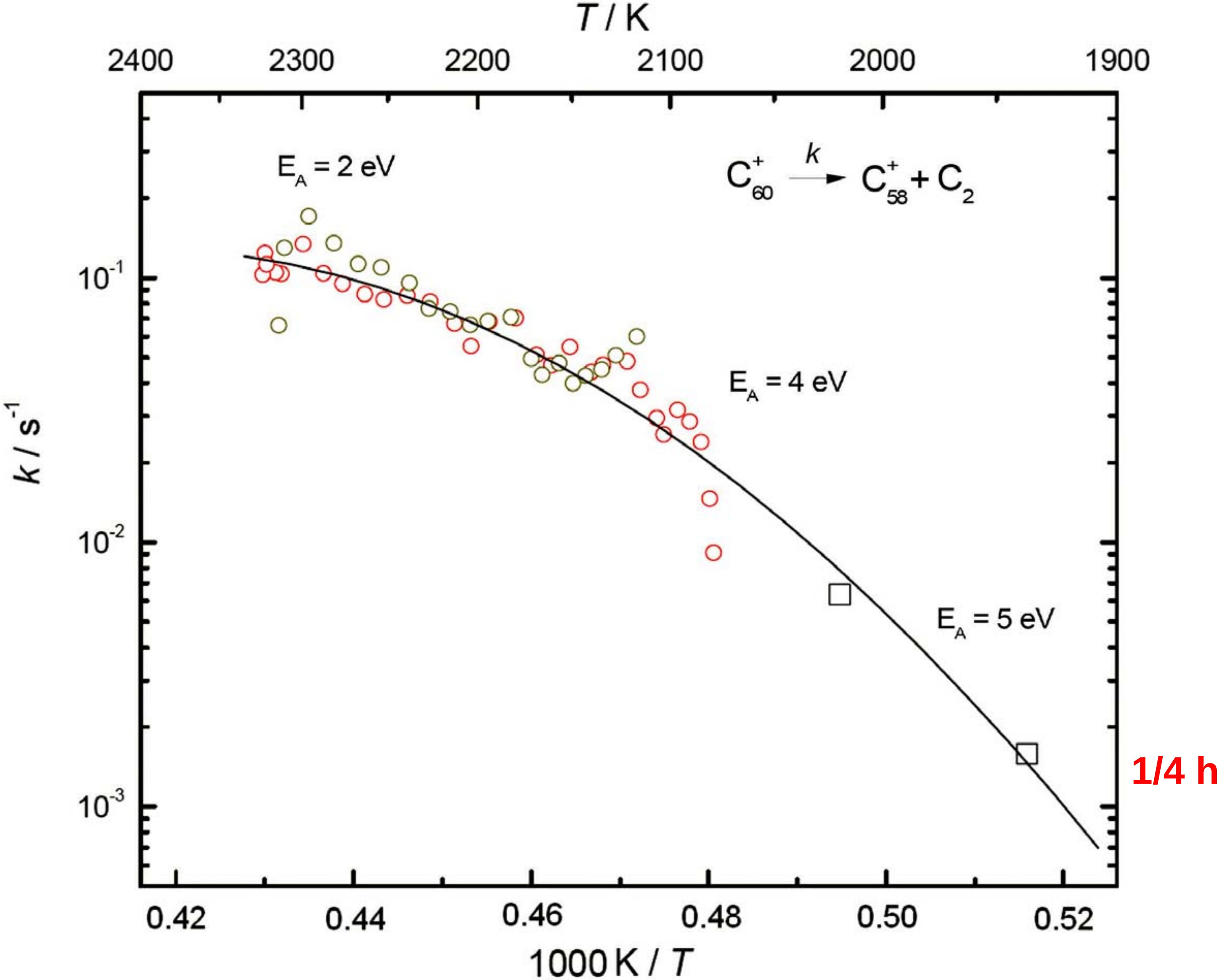
Isomerization



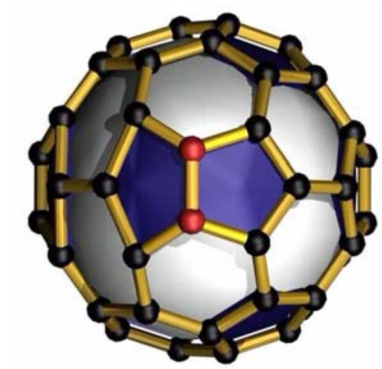
Arrhenius parameter

literature values: 3 - 12 eV

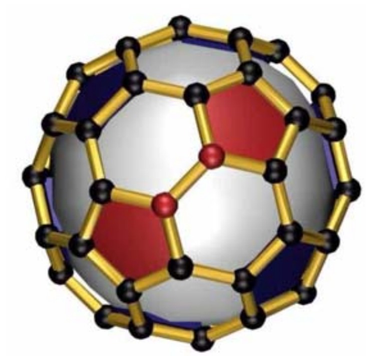
Thermal decay of C_{60}^+



C_2 elimination



Isomerization



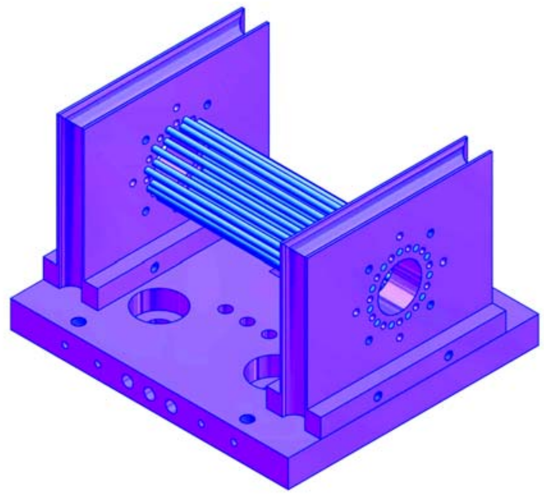
Arrhenius parameter

literature values: 3 - 12 eV

Universitas Carolina
Charles University in Prague

Polyatomic ions in traps: from molecules via clusters to nanoparticles

Dieter Gerlich



Introduction

Ions in rf fields
- Basics, buffer gas cooling

Typical tests
- Spectroscopy
- Association reactions, cluster
- LIR: N_2^+ + Ar

Recent application
- Reactions with H atoms
- Deuteration, nuclear spin
- State selective preparation

Nanoparticles
- NPMS
- HT - SRET, Decay of C_{60}^+

Summary and outlook

Cold ion chemistry in traps

Controlling all degrees of freedom

State to state reactions
Ultra slow relative velocities

Buffer gas cooling in RF traps

Cooling ions with cold effusive beams
Chopped, very slow beams of He
Cooling with slow H-atoms
Combination ion trap - H atom trap

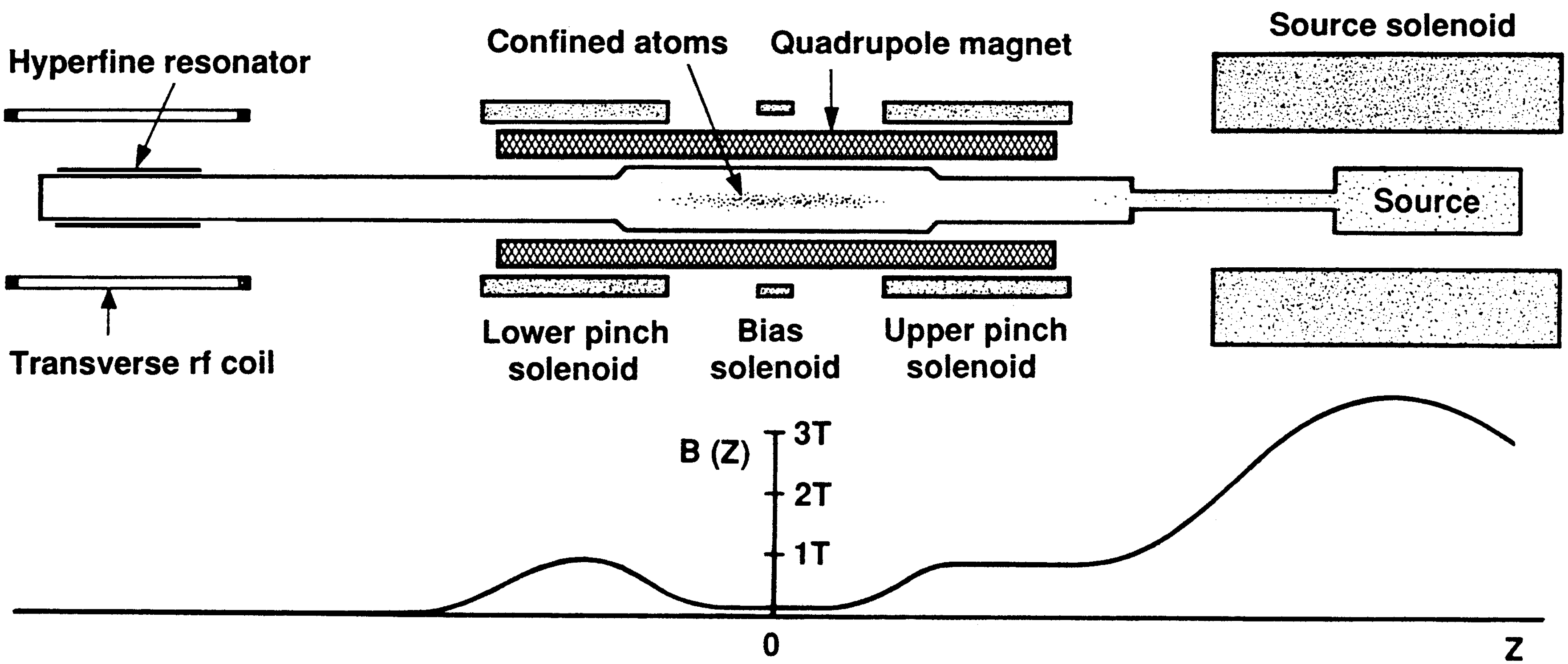
Problems with ion traps

Potential distortions on surfaces
High Q resonance circuit without parasitic oscillations
Superconducting electrodes
Magnetic fields

Coulomb crystals in rf ion traps

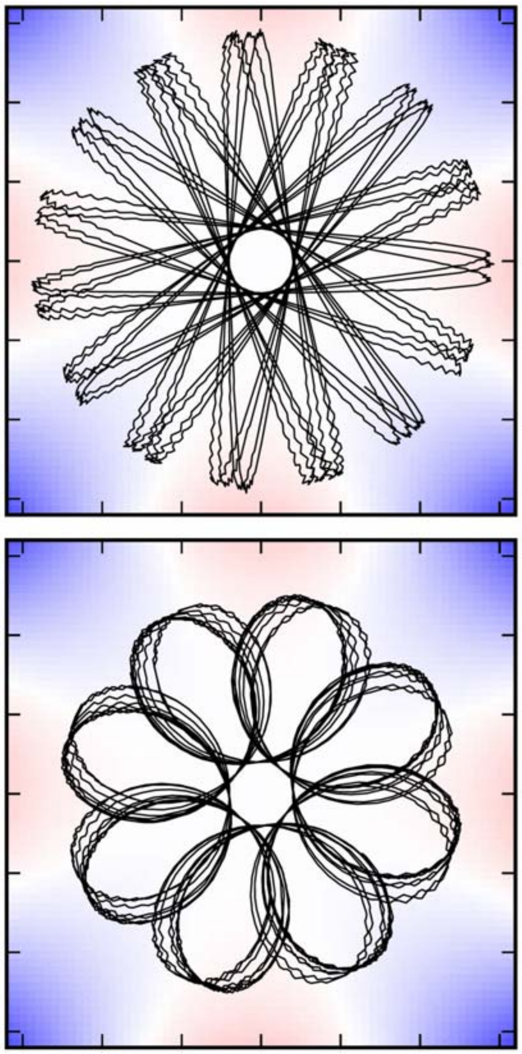
Laser cooling, sympathetic cooling

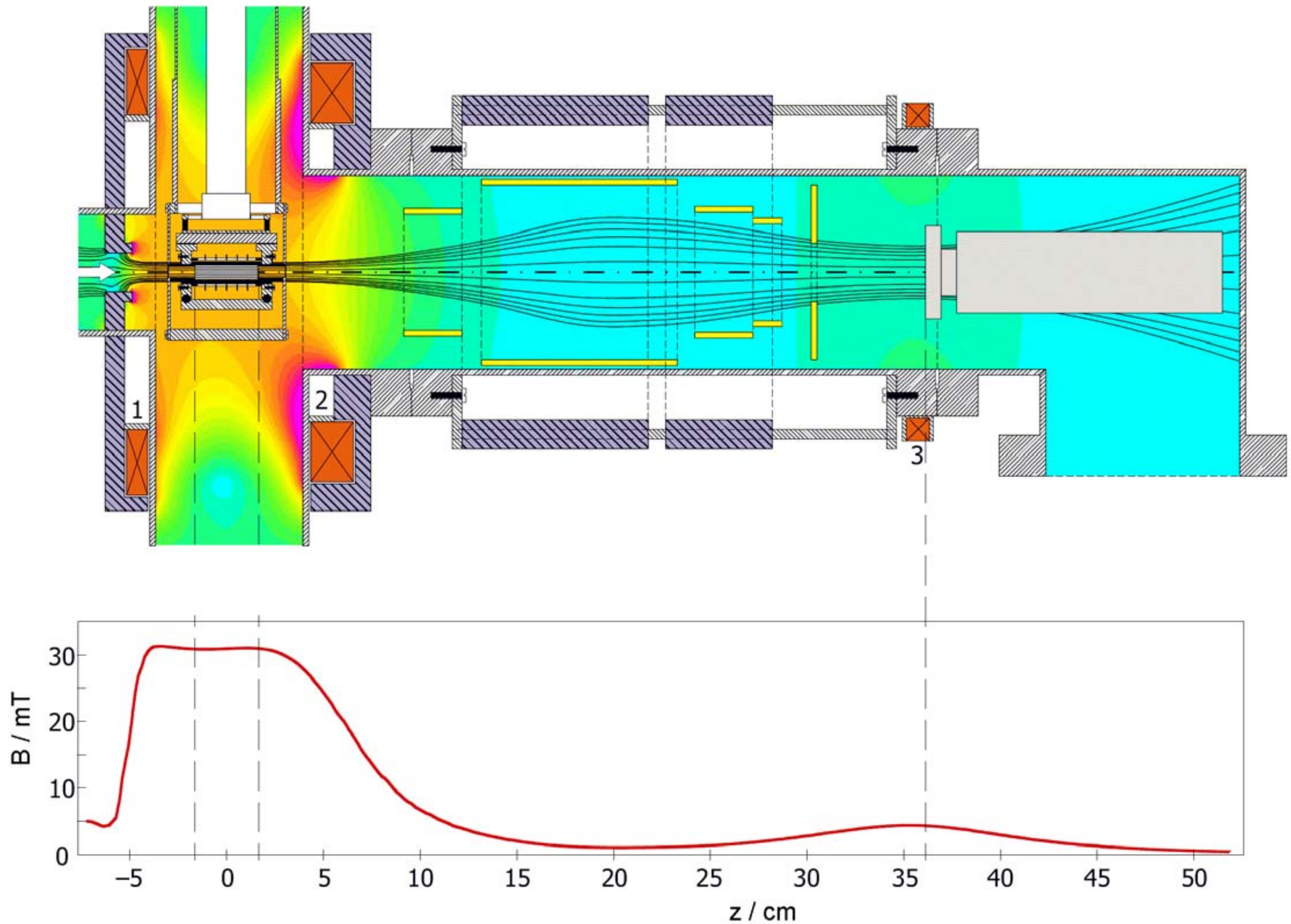
Trapping cold H atoms



Combination: rf ion trap and MAC e^- spectrometer

$H^- + H \rightarrow H_2 + e^-$ $k(T) / E_T(e^-)$





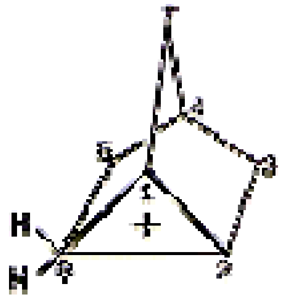
S. Roucka, P. Jusko, D. Gerlich, R. Plasil, J. Glosik, WDS'10 Proceedings of Contributed Papers Prague,Czech Republic, 2010

Nobel Prize in Chemistry 1994

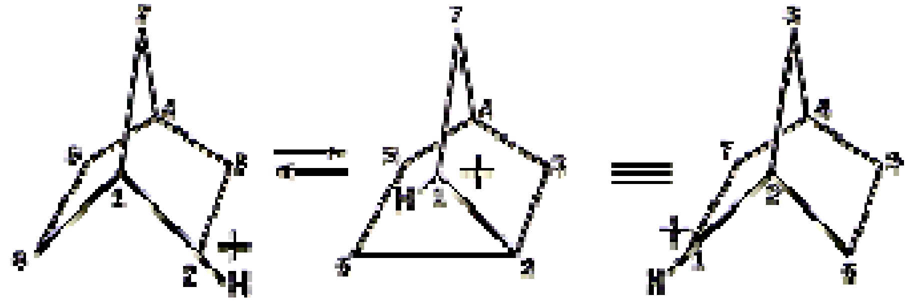
George A. Olah

"for his contribution to carbocation chemistry"

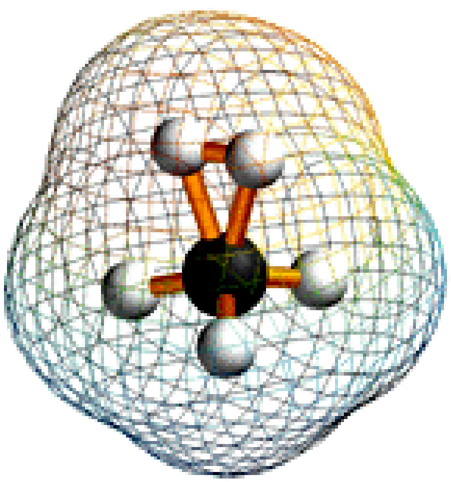
CH_5^+



penta-coordinated carbocation (carbonium ion)



equilibrating tri-coordinated carbocation (carbenium ion)



Carbocations

A cornerstone of the classical theory of structural chemistry since the time of Kekulé in the 1860s is that carbon can bind at most four other atoms (tetra-coordination).

Around 1950 S. Winstein in the USA found a short-lived carbocation that contained penta-coordinated carbon. He named the ion non-classical. Despite very great efforts by many leading physical organic chemists, the problem remained unsolved until Olah's method of preparing long-lived carbocations was applied.

http://nobelprize.org/nobel_prizes/chemistry/laureates/1994

NPMS: perspectives

- long time trapping, isolation under UHV conditions
- non-destructive, absolute mass and charge determination
- high resolution of secular frequencies ($\Delta\nu/\nu < 10^{-6}$)
- single particle: average over time (not ensemble)

- experimental characterization of the trap, new trap design
  accuracy, precision, properties of the 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