DESIREE

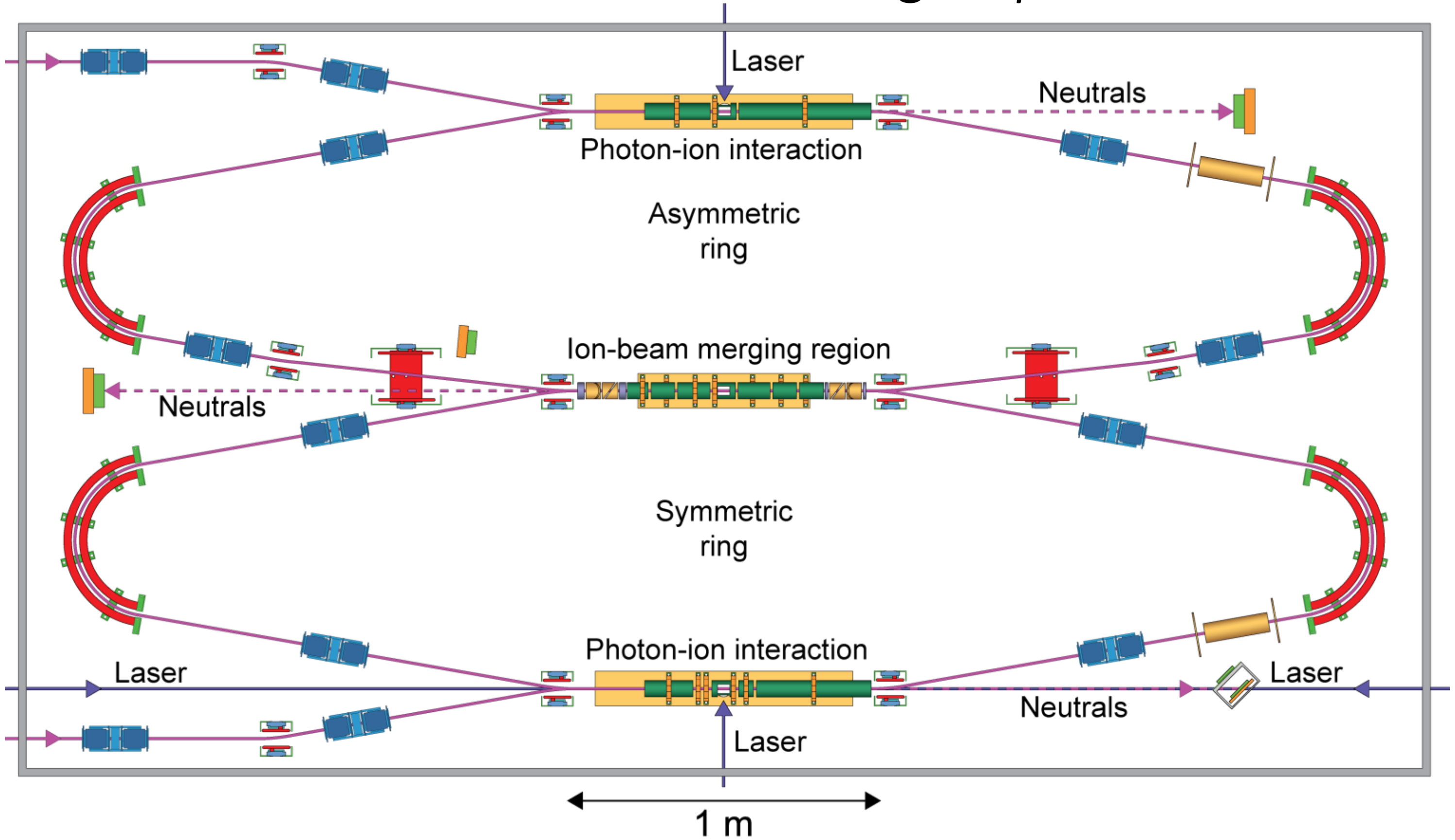
a cryogenic double-ring facility for merged ion-beam studies

Henning Schmidt, Stockholm University

DYMCOM
Institut Pascal, November 22, 2019

Stockholms universitet

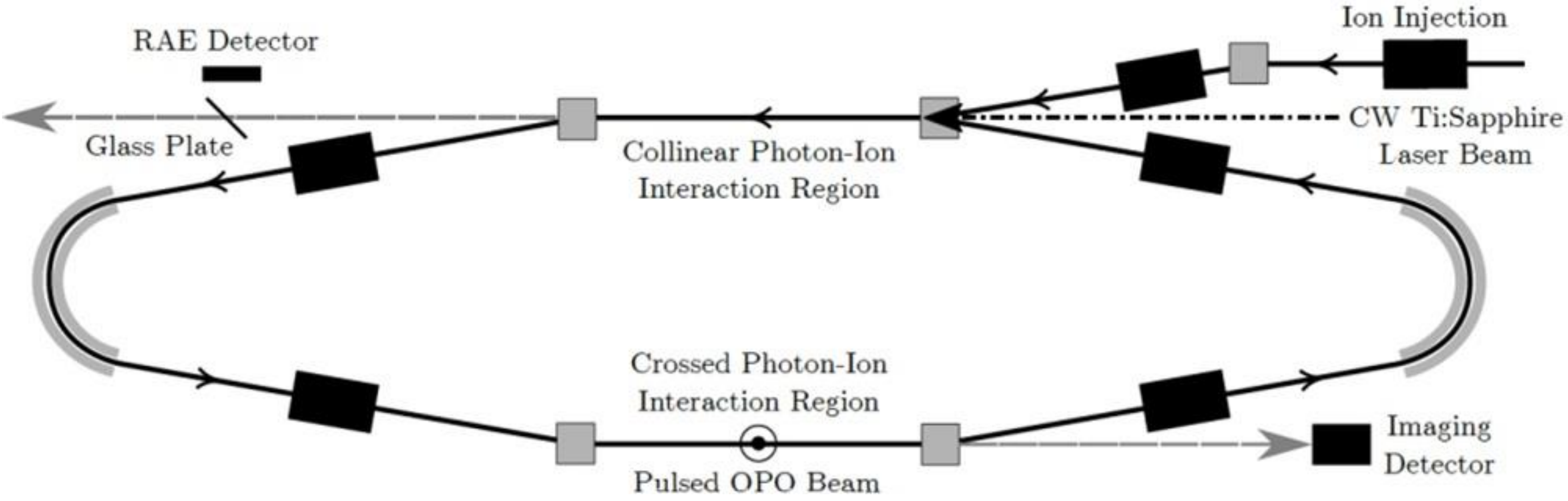
Double ElectroStatic Ion-Ring ExpEriment



Outline

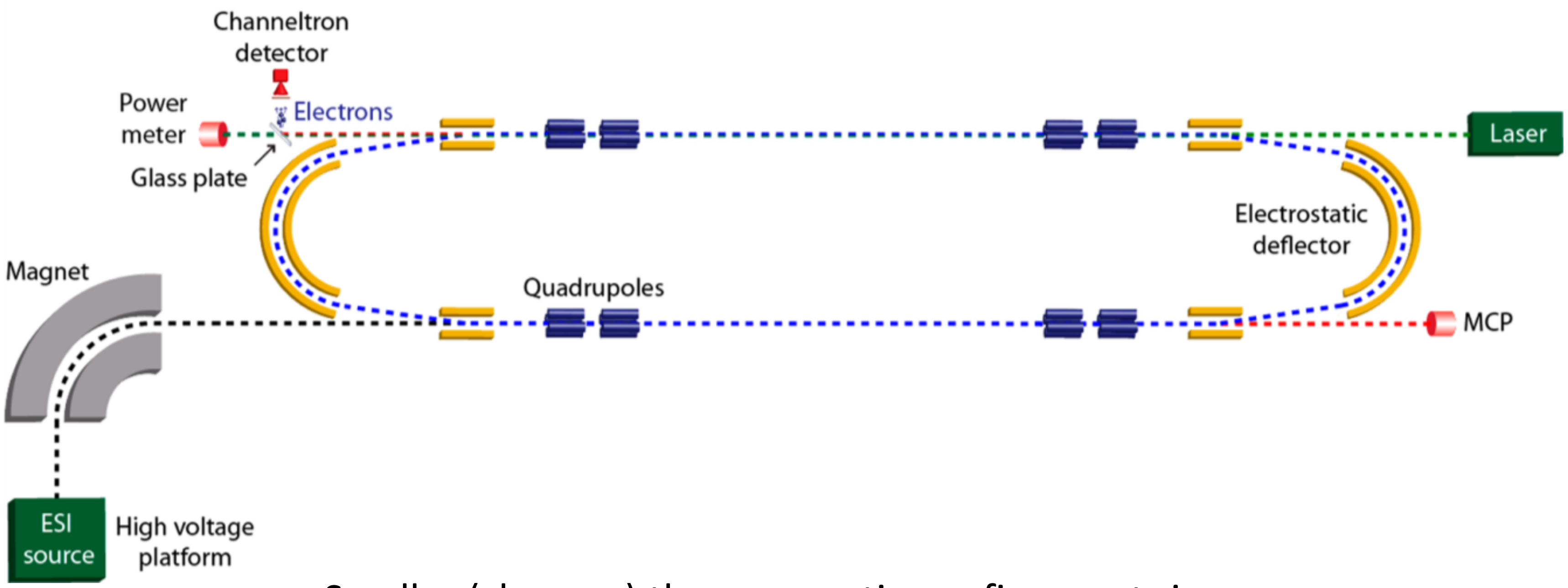
- A description of DESIREE
- Long storage time – cold ions
- Cold collisions??
- Hot stuff in a cold ring

Why use storage rings?



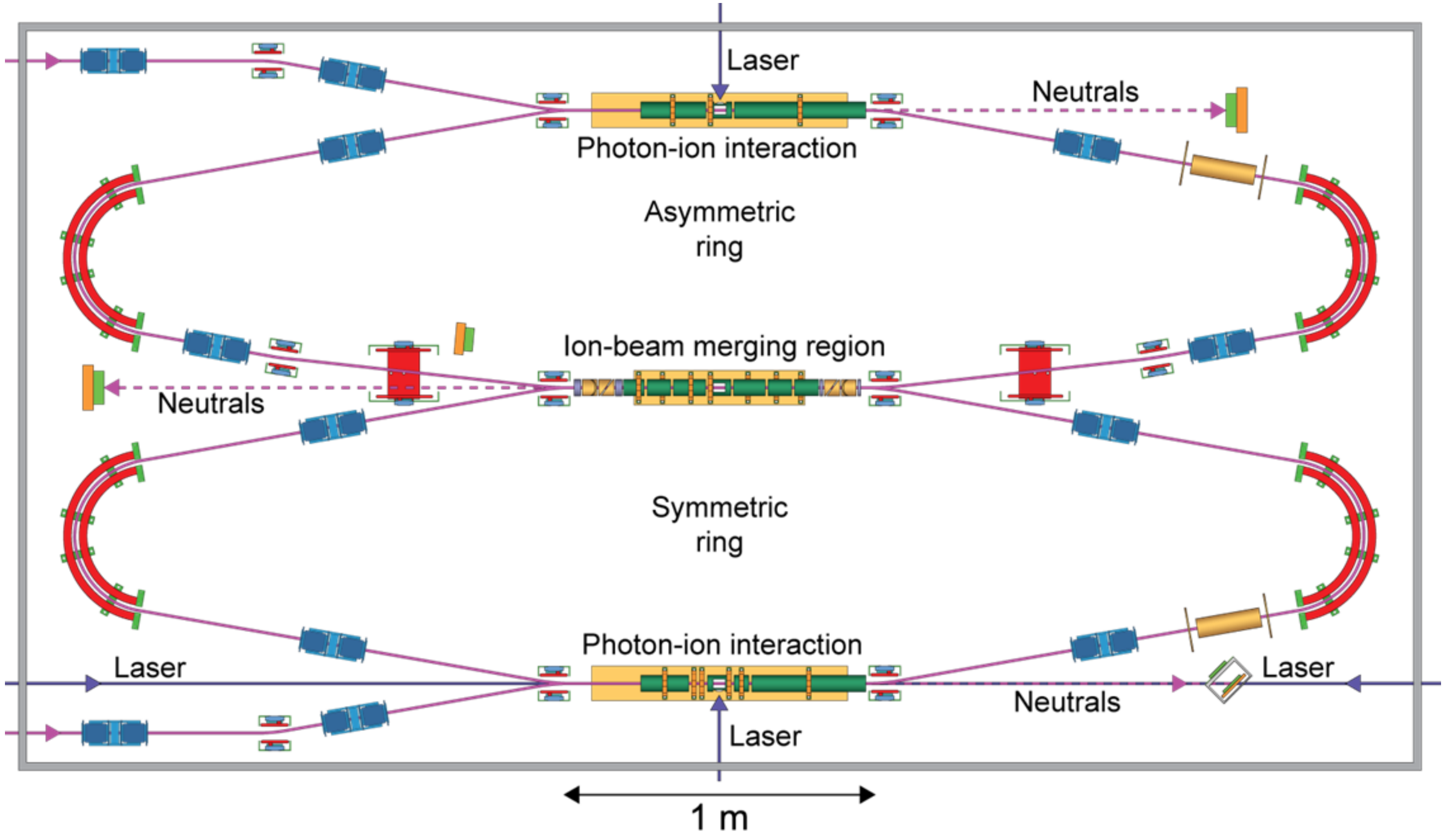
- Easy product detection (and laser access)
- Extended observation times

ELISA, Århus University S.P.Møller

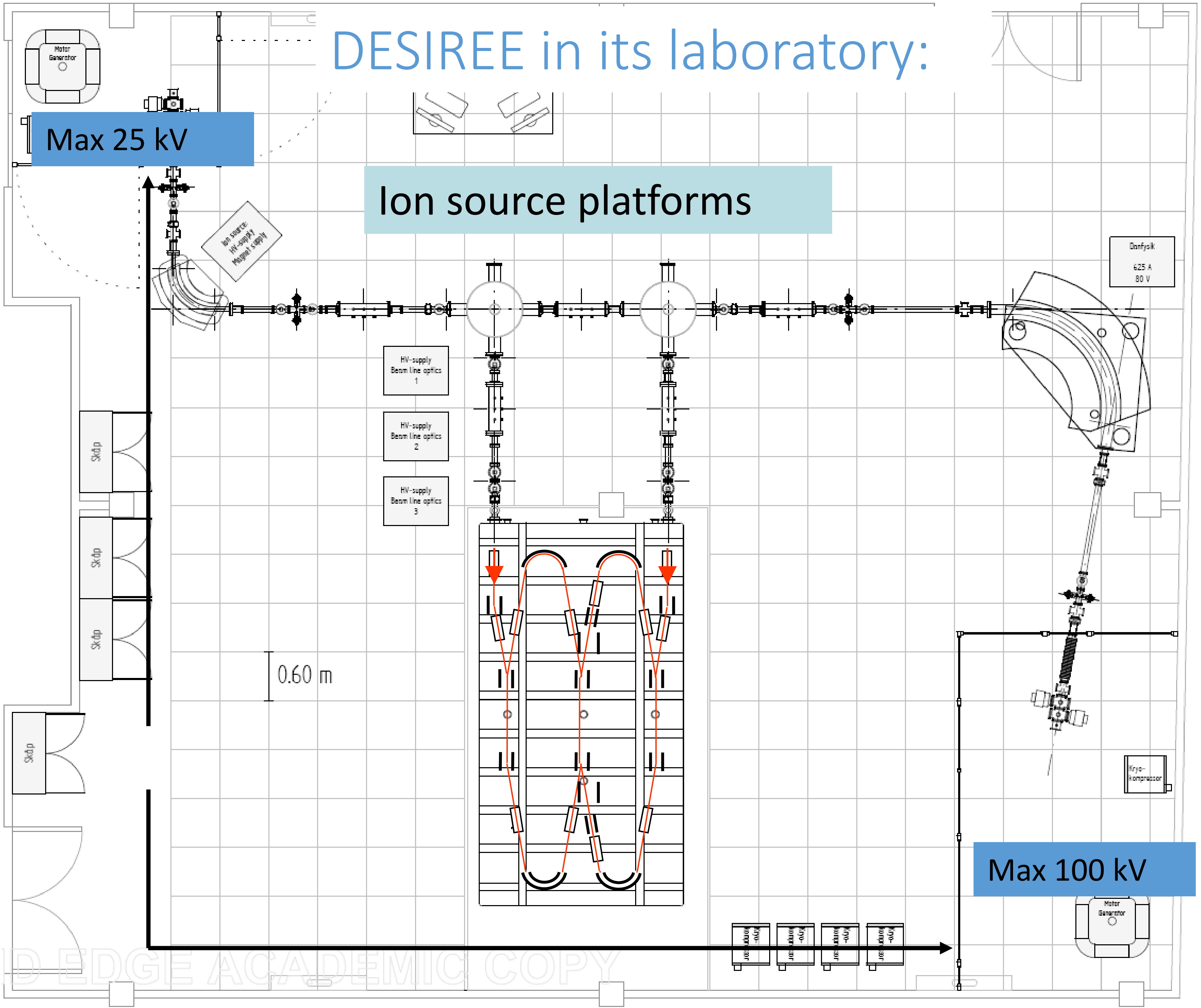


- Smaller (cheaper) than magnetic-confinement ring
- Mass-independent operation
- No magnetic-field mixing

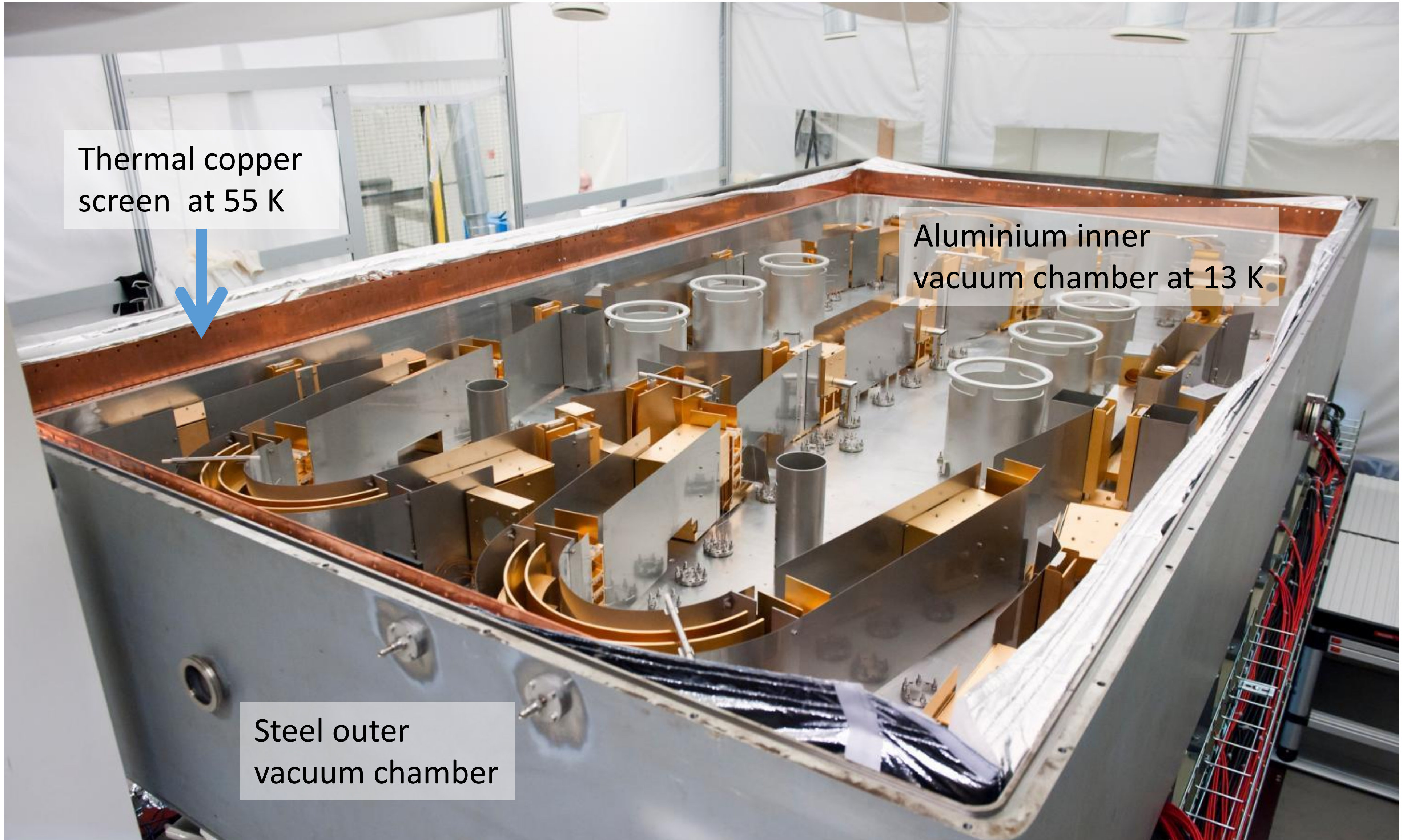
Electrostatic storage rings/traps


Laser
Neutrals
Photon-ion interaction
Asymmetric
ring
Ion-beam merging region
Neutrals
Symmetric
ring
Photon-ion interaction
Laser
Laser
Neutrals
Laser
1 m

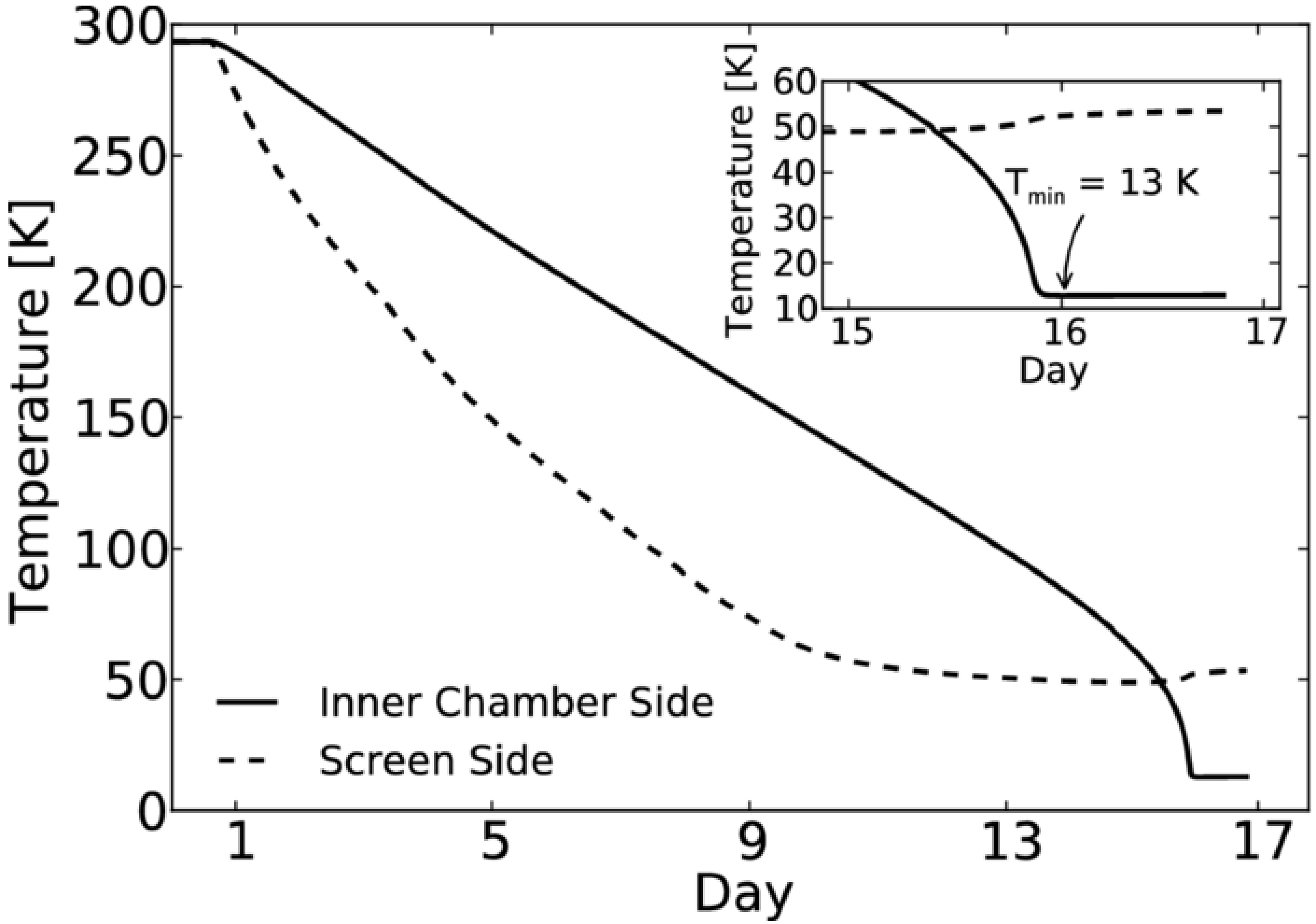
DESIREE in its laboratory:



A look inside DESIREE!



Cooling down DESIREE.



DESIREE is

- Electrostatic
- Cryogenic
- A double-ring system

Cheaper than magnetic ring
More compact
Good for (slow) heavy particles

Very good vacuum (a few residual gas molecules per mm^3)
Long (up to 1 hour) storage lifetimes
Low degree of excitation at thermal equilibrium.
Storage of fragile systems

Free of magnetic-mixing of fine- and hyper-fine structure levels.

Merged-beams studies of mutual neutralization and other two-ion processes at low and controlled CoM energies

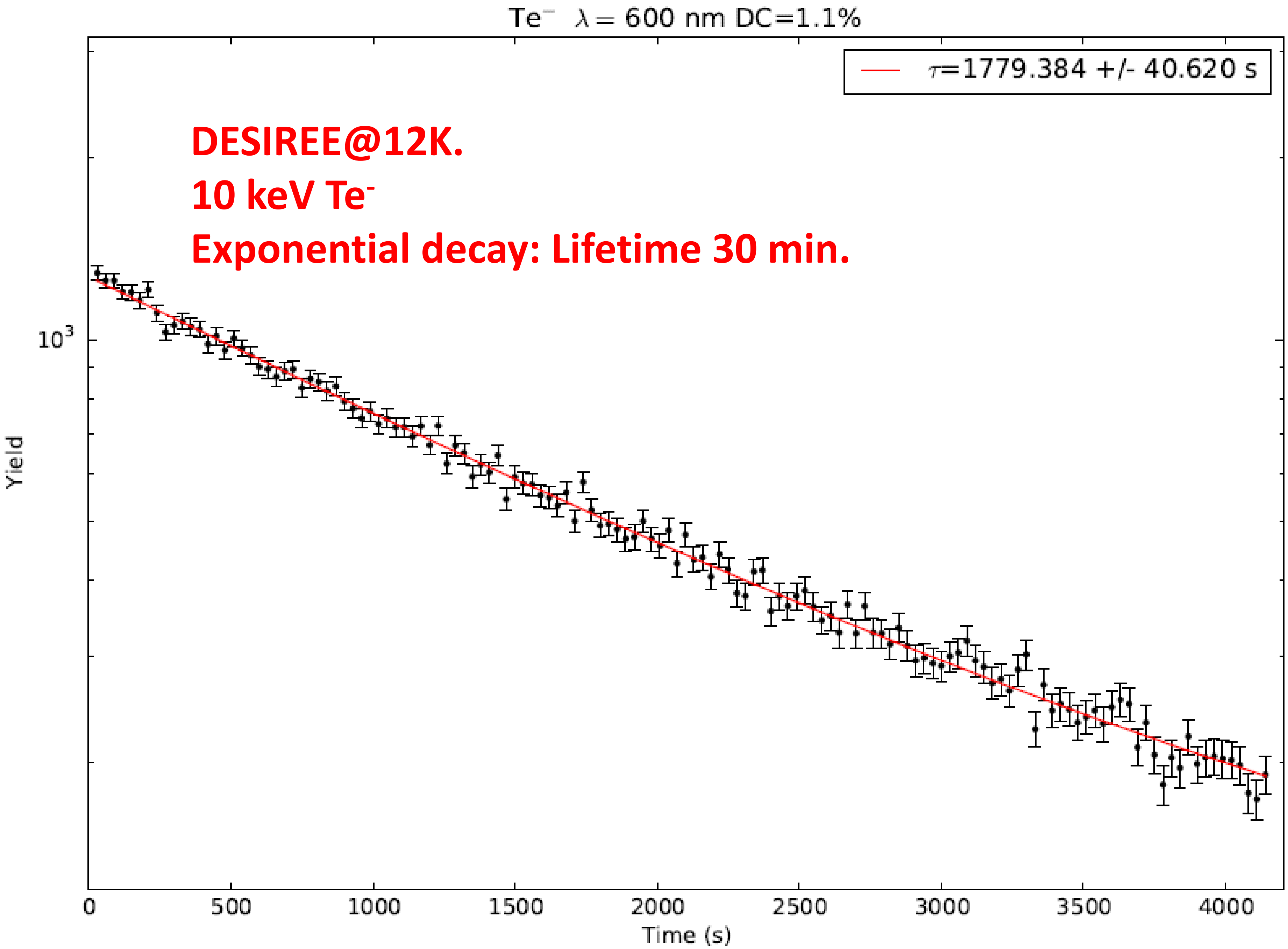
One major goal for DESIREE:

- Mutual neutralization with control of external and internal degrees of freedom.
  - External
    Merged co-propagating beams for CM energy control down to 10 meV.
  - Internal
    Long time storage
    Low thermal excitation
    Laser manipulation

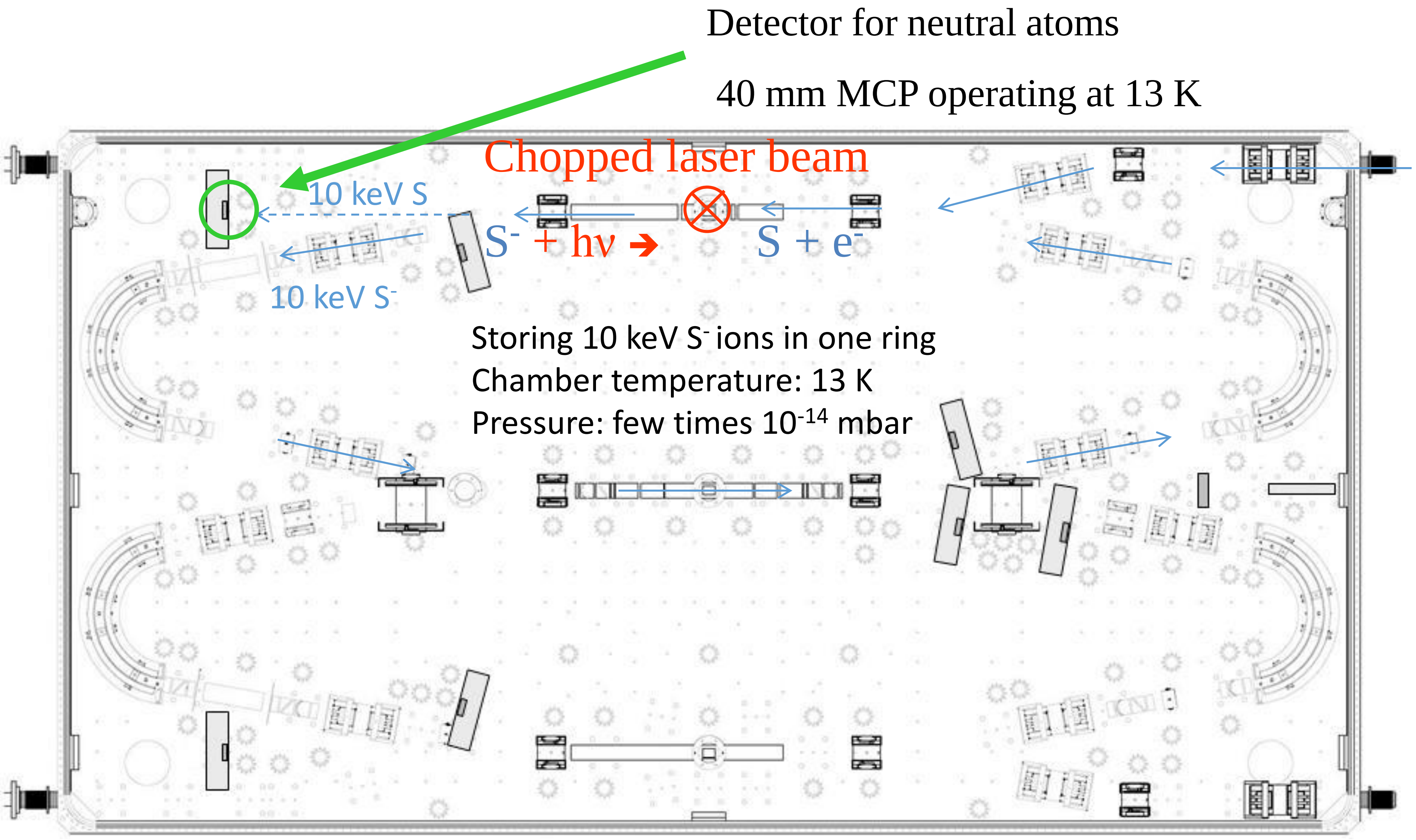
Cryogenic vacuum system!

Outline

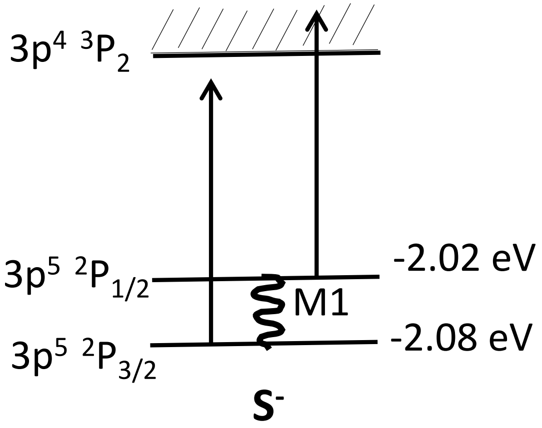
- A description of DESIREE
- Long storage time – cold ions
- Cold collisions??
- Hot stuff in a cold ring


Te⁻ λ = 600 nm DC=1.1%
τ=1779.384 +/- 40.620 s
DESIREE@12K.
10 keV Te-
Exponential decay: Lifetime 30 min.
10^3
Yield
0
500
1000
1500
2000
2500
3000
3500
4000
Time (s)

Laser-probing of the Metastable- (MS) and the Ground State (GS) populations in S^- as functions of time



Probing the decay of Metastable (MS) S^-



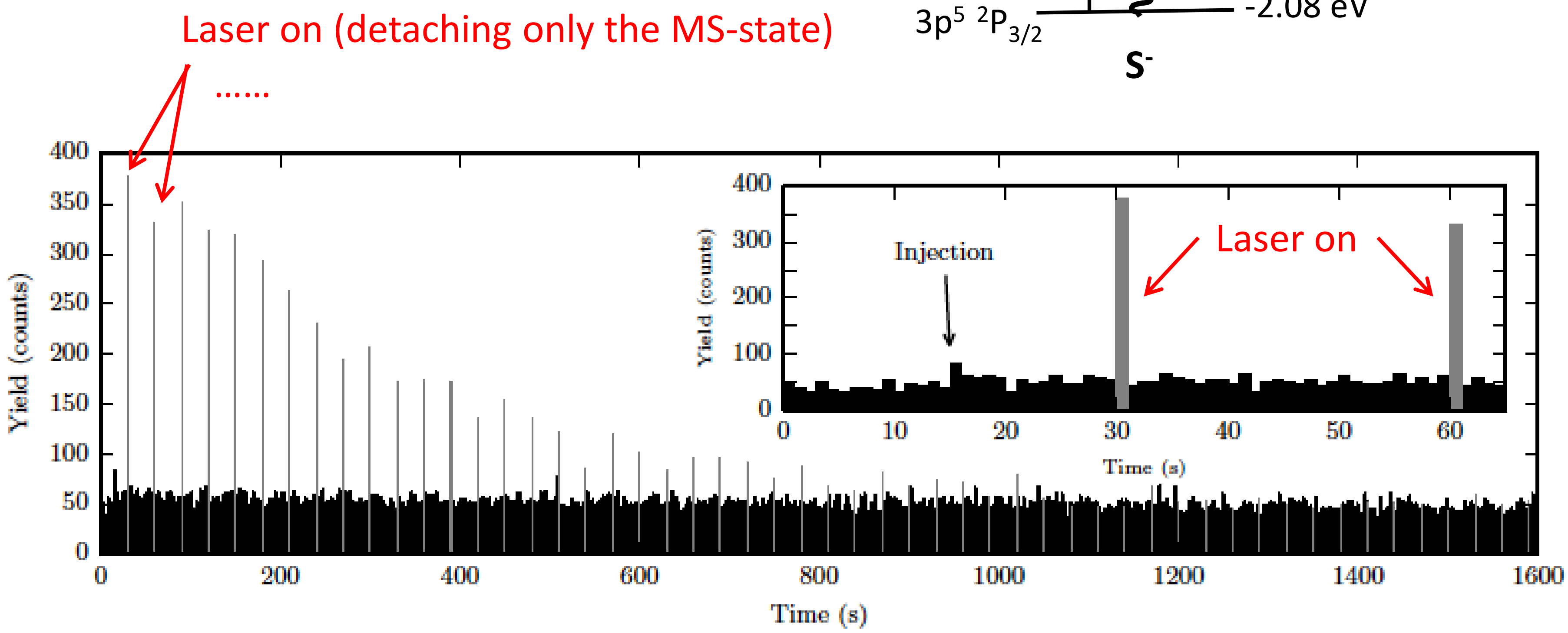
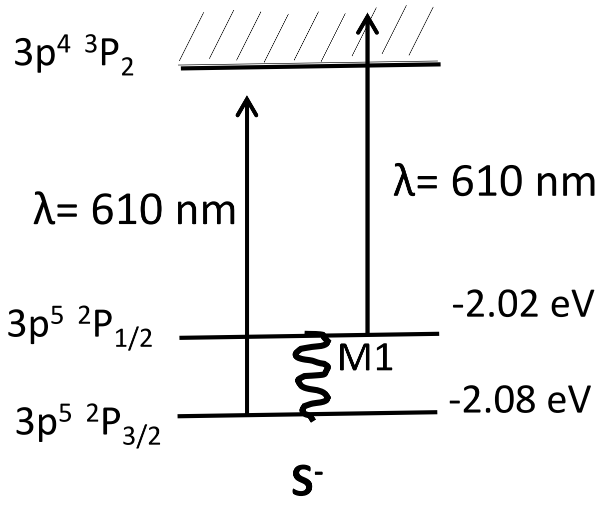
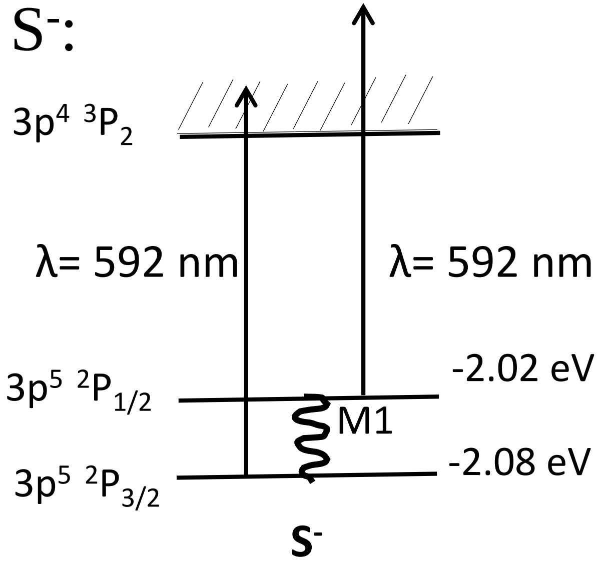


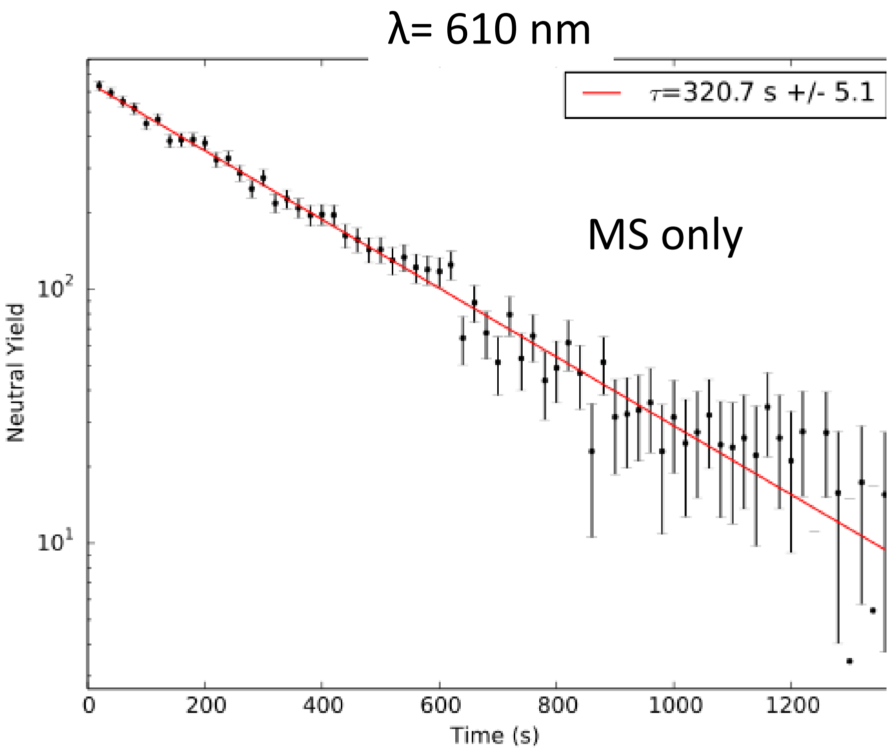
FIG. 3: The rate of neutrals recorded on the MCP as a function of time. The inset shows the first 65 seconds. Bins recorded when the laser shutter is open is grey. The increased count rate during injection time is also indicated in the figure.

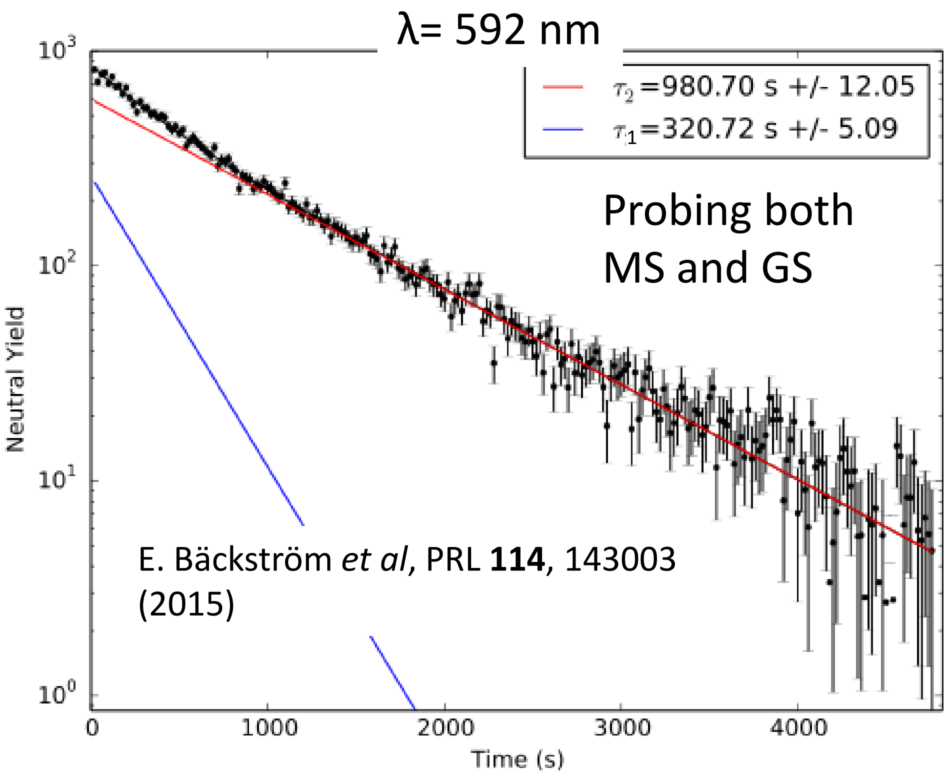
E. Bäckström *et al*, PRL **114**, 143003 (2015)

Laser probing of metastable population in S^-:









Metastable lifetimes in atomic negative ions

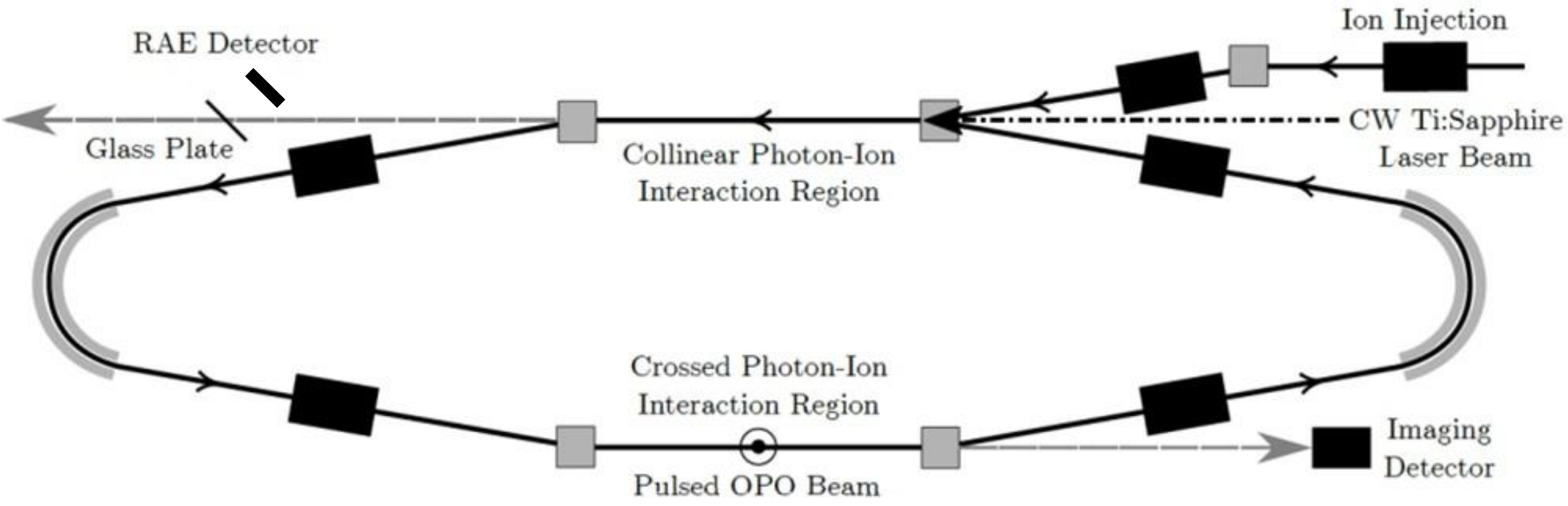
- sulfur, tellurium, selenium,
  - E. Bäckström *et al*, Phys. Rev. Lett. **114**, 143003 (2015)
- nickel
  - M. Kaminska *et al*, Phys. Rev. A **93**, 012512 (2016)
- platinum
  - KC. Chartkunchand *et al*, Phys. Rev. A **94**, 032501 (2016)
- iridium
  - M. K. Kristiansson *et al*,…

Single-ring experiments monitoring relaxation of stored ions

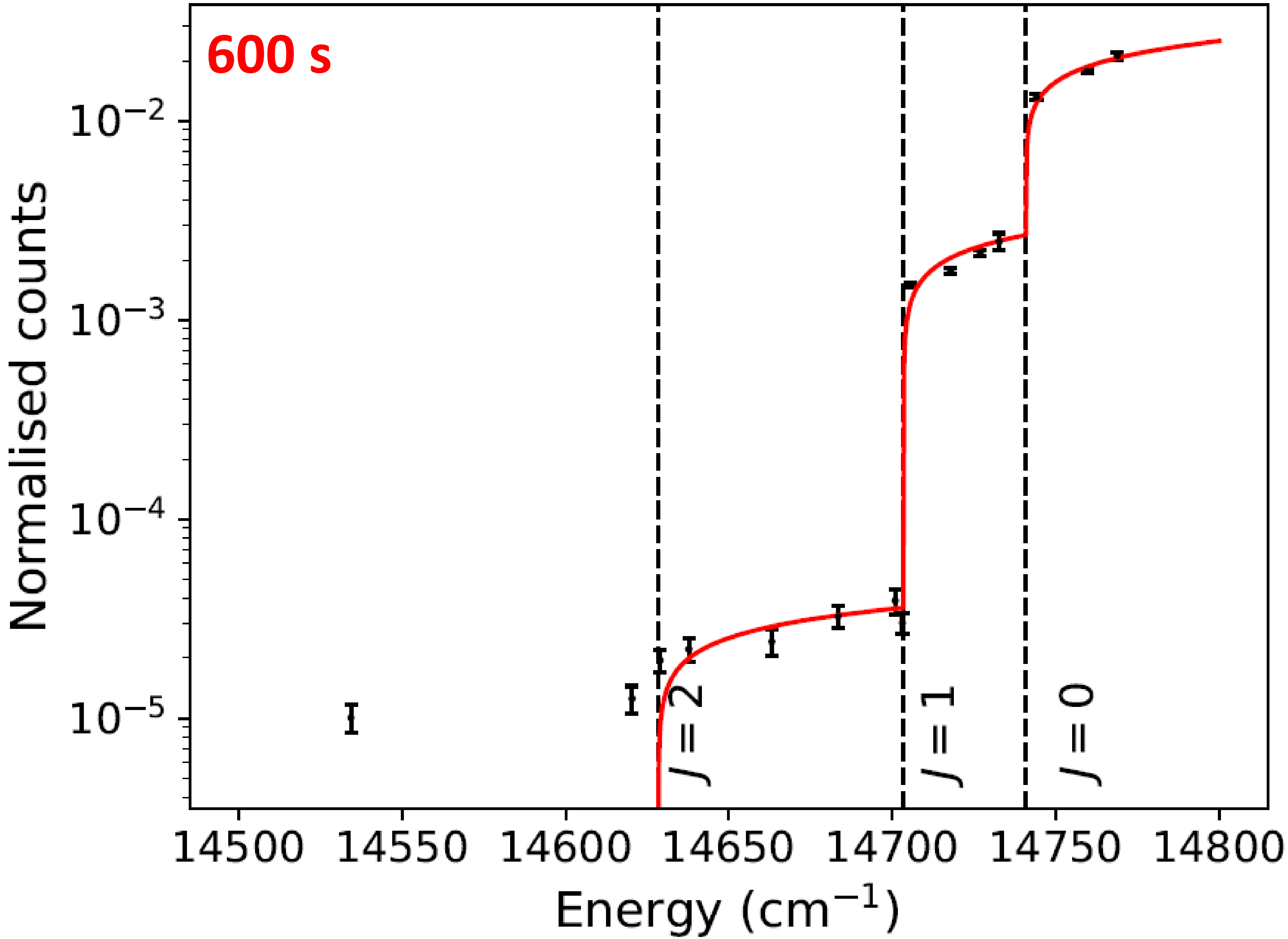
- Lifetimes of metastable states in atomic negative ions
- Rotational relaxation of cold molecular ions

OH^- in DESIREE

H. T. Schmidt *et al*, Phys. Rev. Lett. **119**, 073001 (2017)



H. T. Schmidt *et al*, Phys. Rev. Lett. **119**, 073001 (2017)



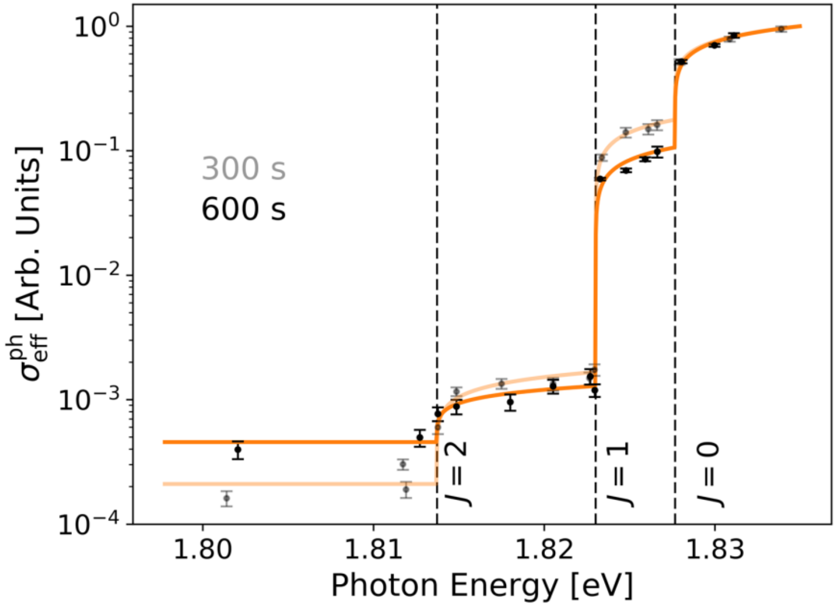
Threshold photodetachment thermometry analysis

- After 600 s of storage, T ~ 22 K provides good description
- DESIREE temperature 13.5 K
- Why?

–Longer storage times needed

–Background contribution from $^{17}O^{-}$

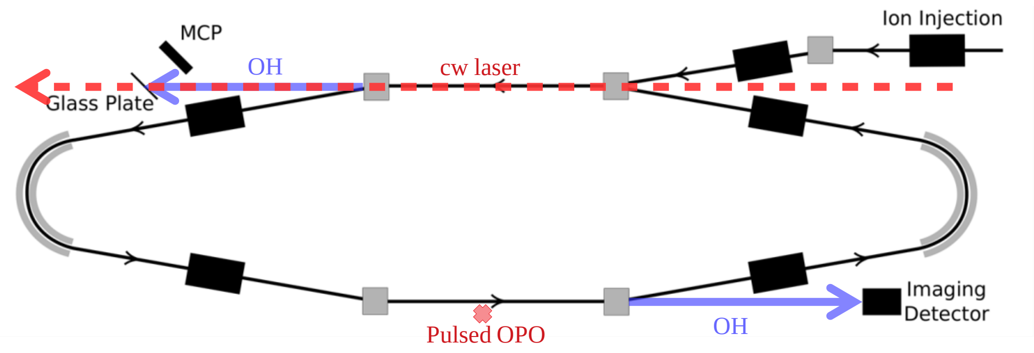
–Other effects?

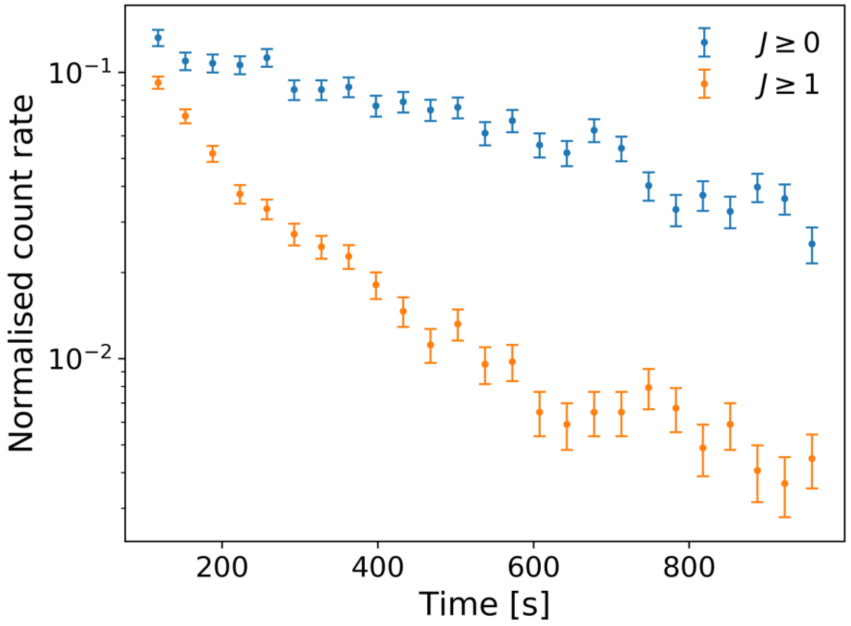


t [s]	$P(0)$ [%]	$P(1)$ [%]	$P(2)$ [%]	T [K]
50	5.5 ± 3.2	82.8 ± 2.9	11.7 ± 0.6	-
100	28.9 ± 2.7	68.8 ± 2.6	2.3 ± 0.1	-
150	44.4 ± 2.0	54.3 ± 2.0	1.27 ± 0.08	50.6 ± 3.0
300	67.0 ± 1.3	32.2 ± 1.3	0.85 ± 0.05	29.2 ± 0.9
600	79.2 ± 0.5	20.3 ± 0.5	0.51 ± 0.06	22.0 ± 0.3

Alternative experimental procedure

- Stored ions are probed using pulsed OPO
- Measurements at photon energies detaching from $J \geq 0$ and $J \geq 1$ respectively
- From measurements $P(1)$ can be determined as function of time
- Cw laser can be simultaneously applied to detach from $J \geq 1$





$P(1)$ as function of time

- Spontaneous decay
- Depletion

  $P(1) = 3.6 \pm 0.3$ %

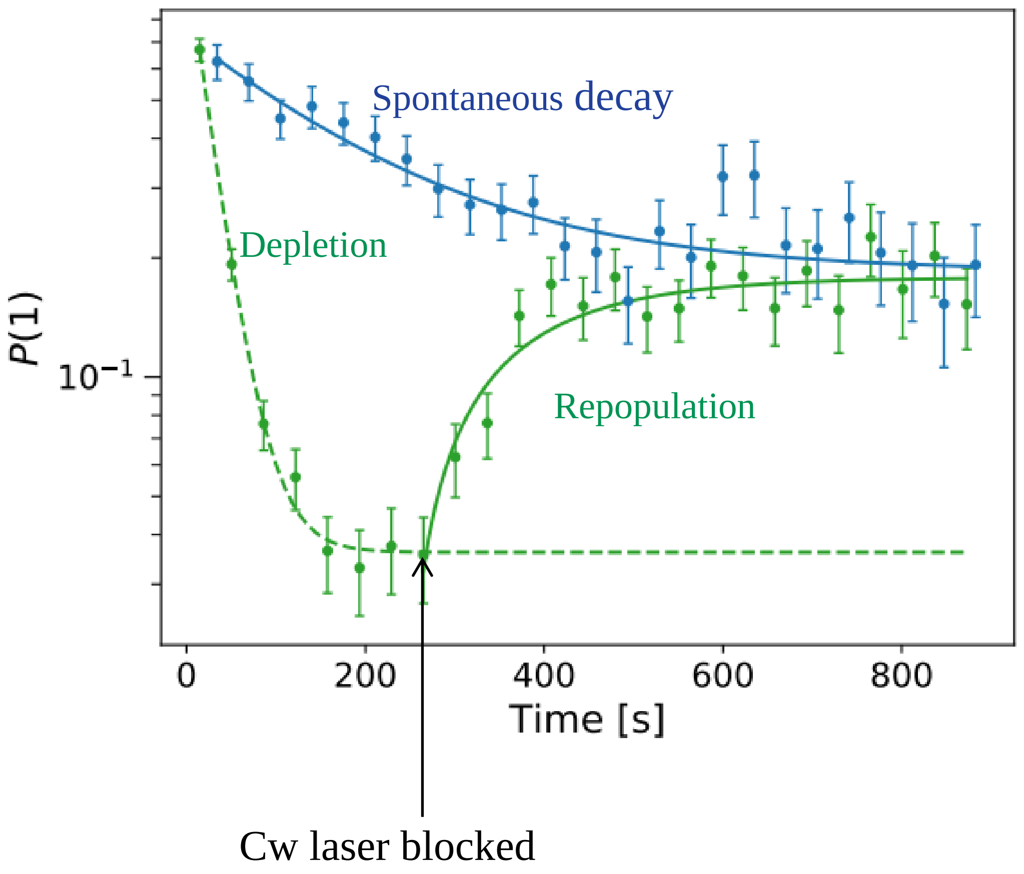
  $T = 12.3 \pm 0.2$ K
- Repopulation

  $\Gamma_{\mathrm{eff}}^{-1} = 134 \pm 25$ s

  $P(1) = 18.1 \pm 0.9$ %

  $T = 20.6 \pm 0.5$ K
- Intrinsic lifetime

  $1/A_{10} = 183 \pm 35$ s

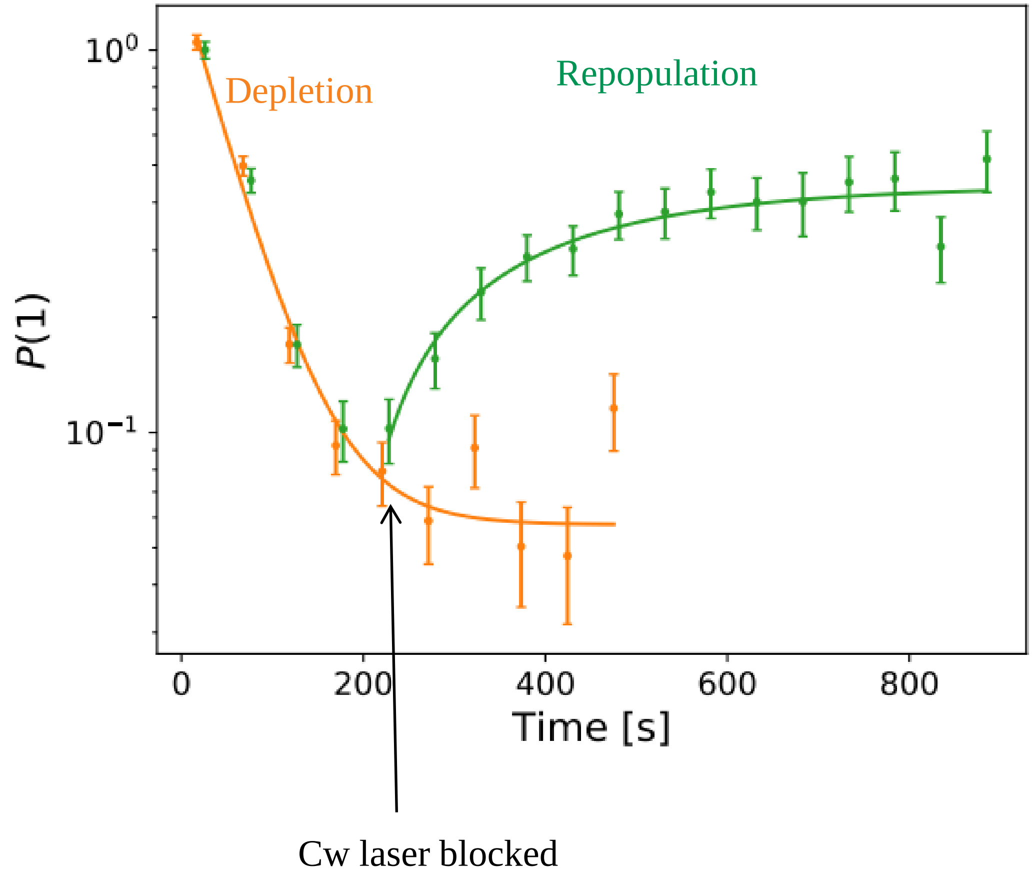


Next step: replace OH^- with OD^-

- Finer rotational splittings, longer lifetimes
- Depletion

$P(1) = 5.7 \pm 1.1\ \%$

$T = 7.4 \pm 0.4\ \mathrm{K}$



Rotational relaxation of molecular ions in DESIREE

- OH^-
  - H. T. Schmidt *et al*, Phys. Rev. Lett. **119**, 073001 (2017)
- OD^-
  - G. Eklund *et al*, Manuscript in preparation. 7.5 K by selective photodetachment
- CH^-
  - W. D. Geppert, M. K. Kristiansson, G. Eklund … work in progress (preliminary results)
    - Electronic excited $^1\Delta$ state lifetime. **14.5±0.8 s**

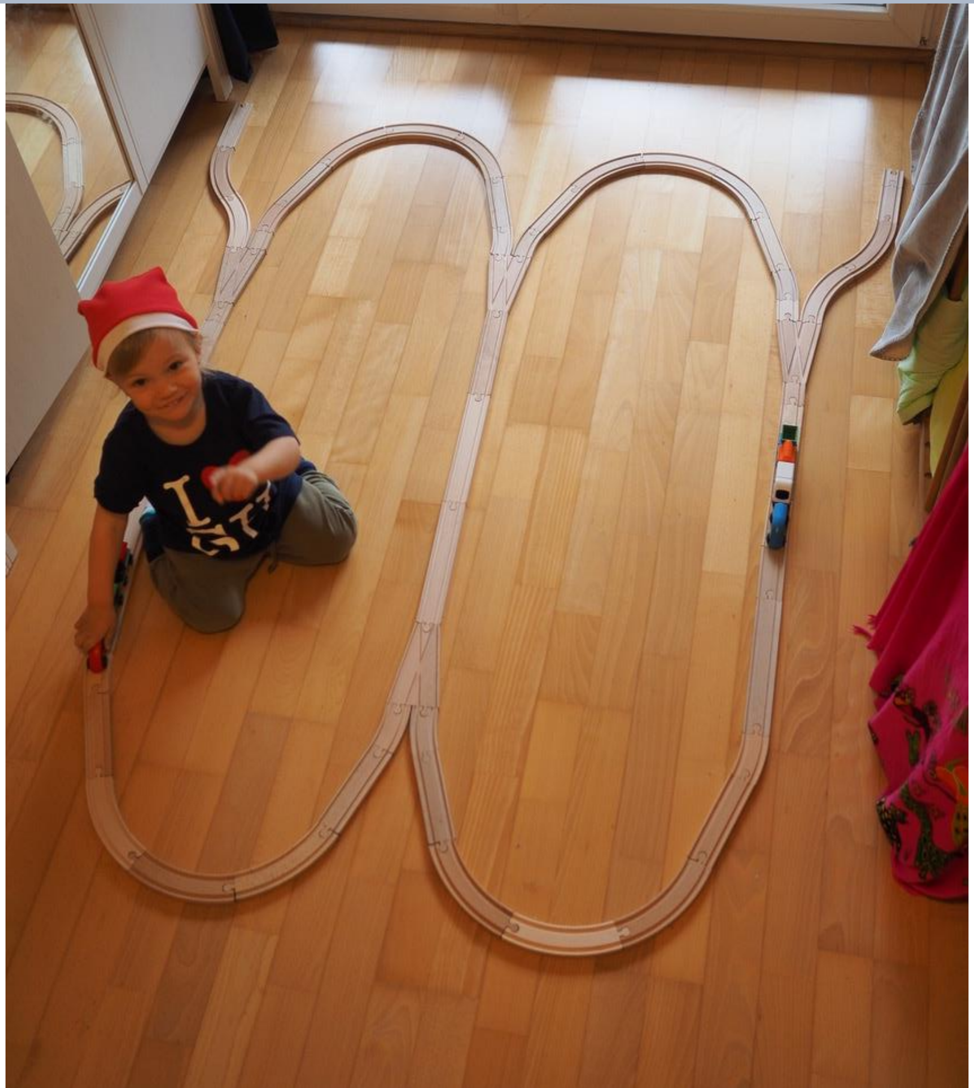
      (Okumura, M., *et al*. J. Chem. Phys. **85**, 1971 (1986): 5.8±0.8 s)
    - New value for electron affinity. **1.215±0.004 eV.**

      (Kasdan, A.,*et al*, Chem. Phys. Lett. **31**, 78 (1975): 1.239±0.007 eV;
      Goebbert, Daniel Chem. Phys. Lett. **515**, 19 (2012): 1.26±0.02 eV)

Outline

- A description of DESIREE
- Long storage time – cold ions
- Cold collisions??
- Hot stuff in a cold ring

DESIREE Principle – Merged beams



17 keV $^{7}Li^{+}$ // 6.5 keV D^{-}

Major deflectors (160°)

Minor deflectors (10°)

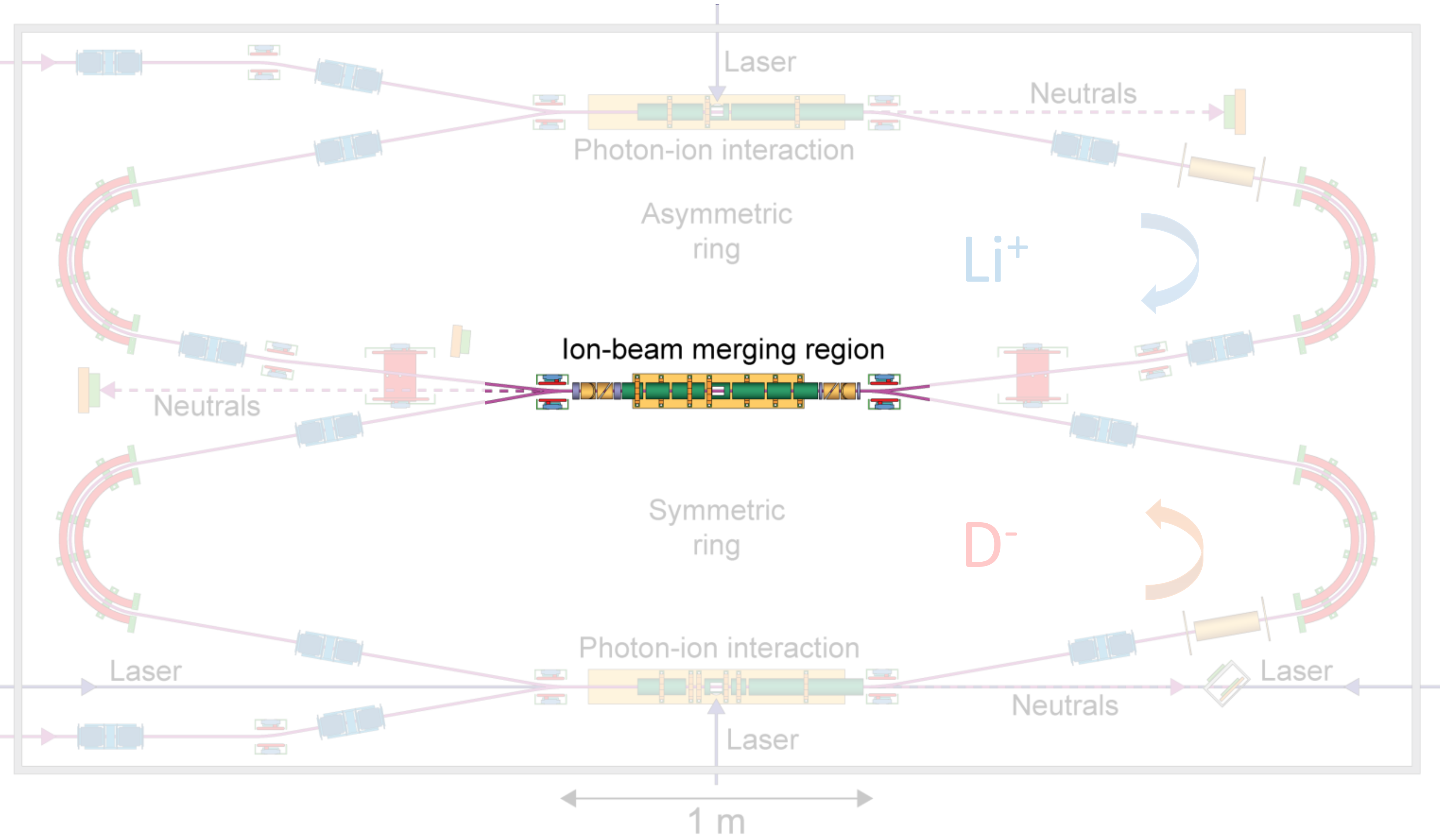
Position sensitive pickups

Focusing quadrupoles

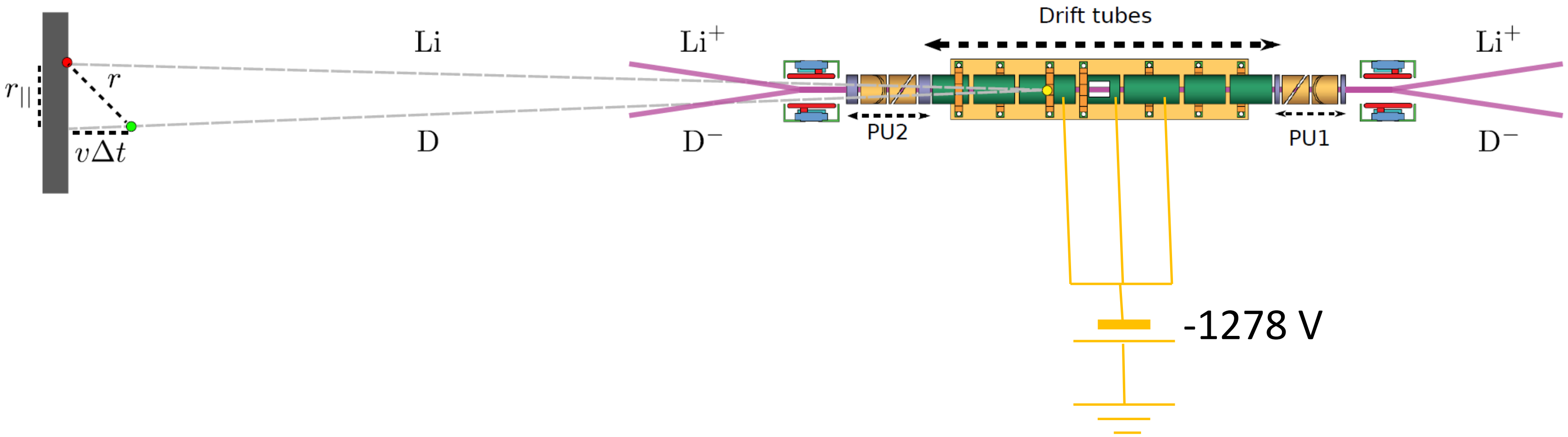
Overlap control deflectors

RF buncher

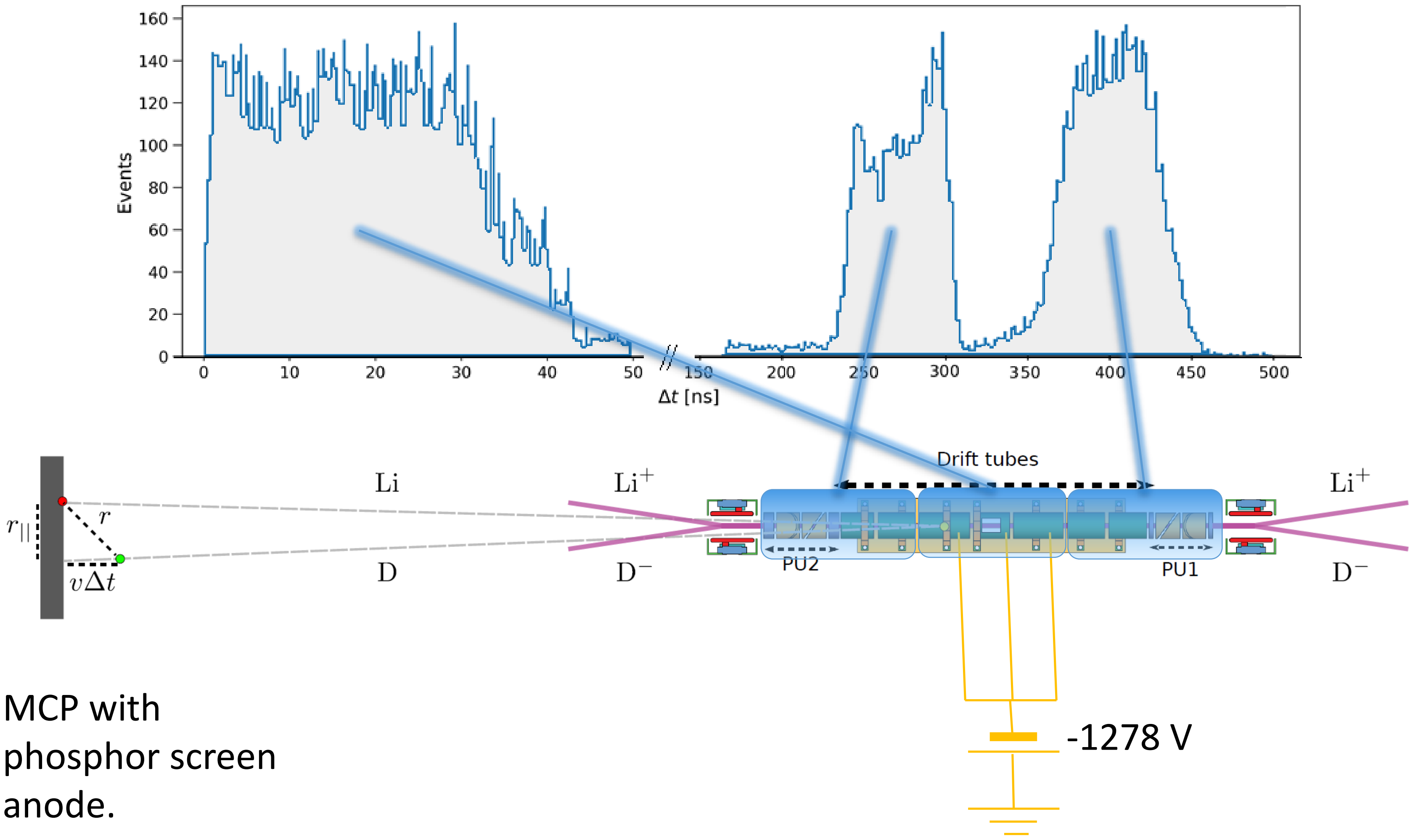
Detector



17 keV Li^+ // 6.5 keV D^-

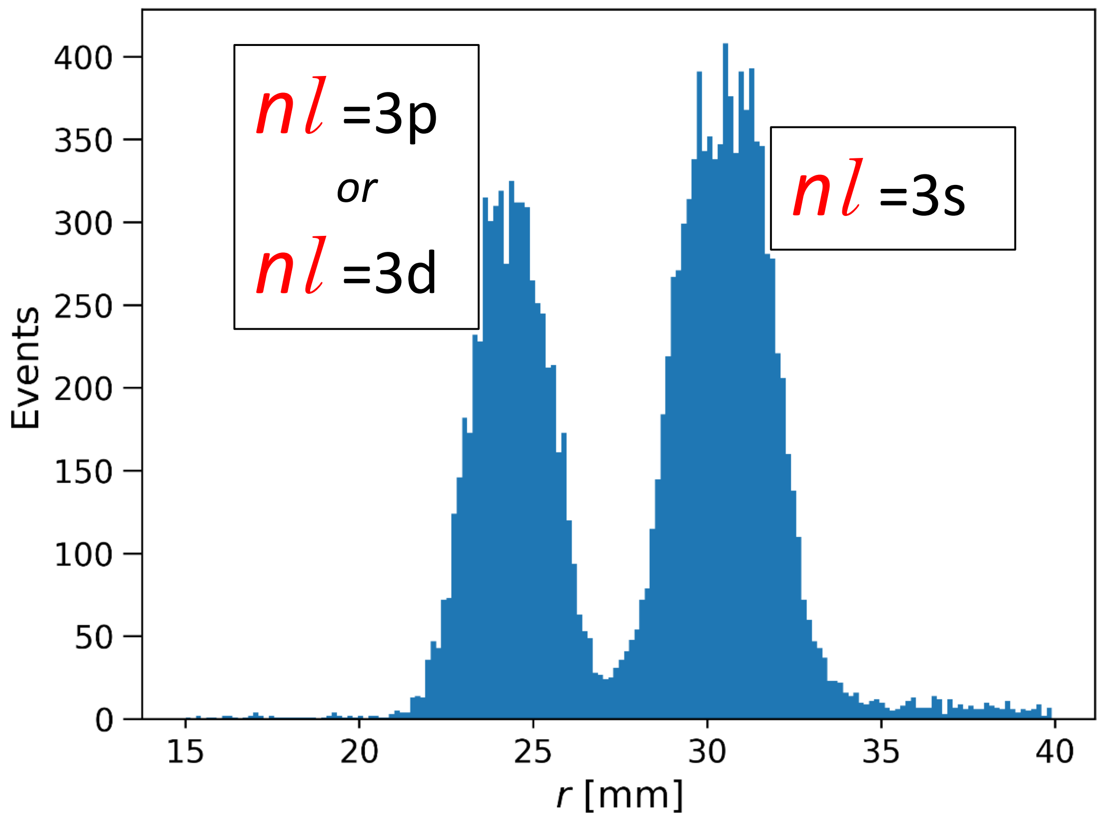


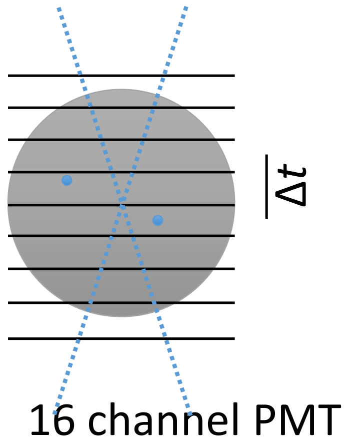
17 keV Li^+ // 6.5 keV D^-

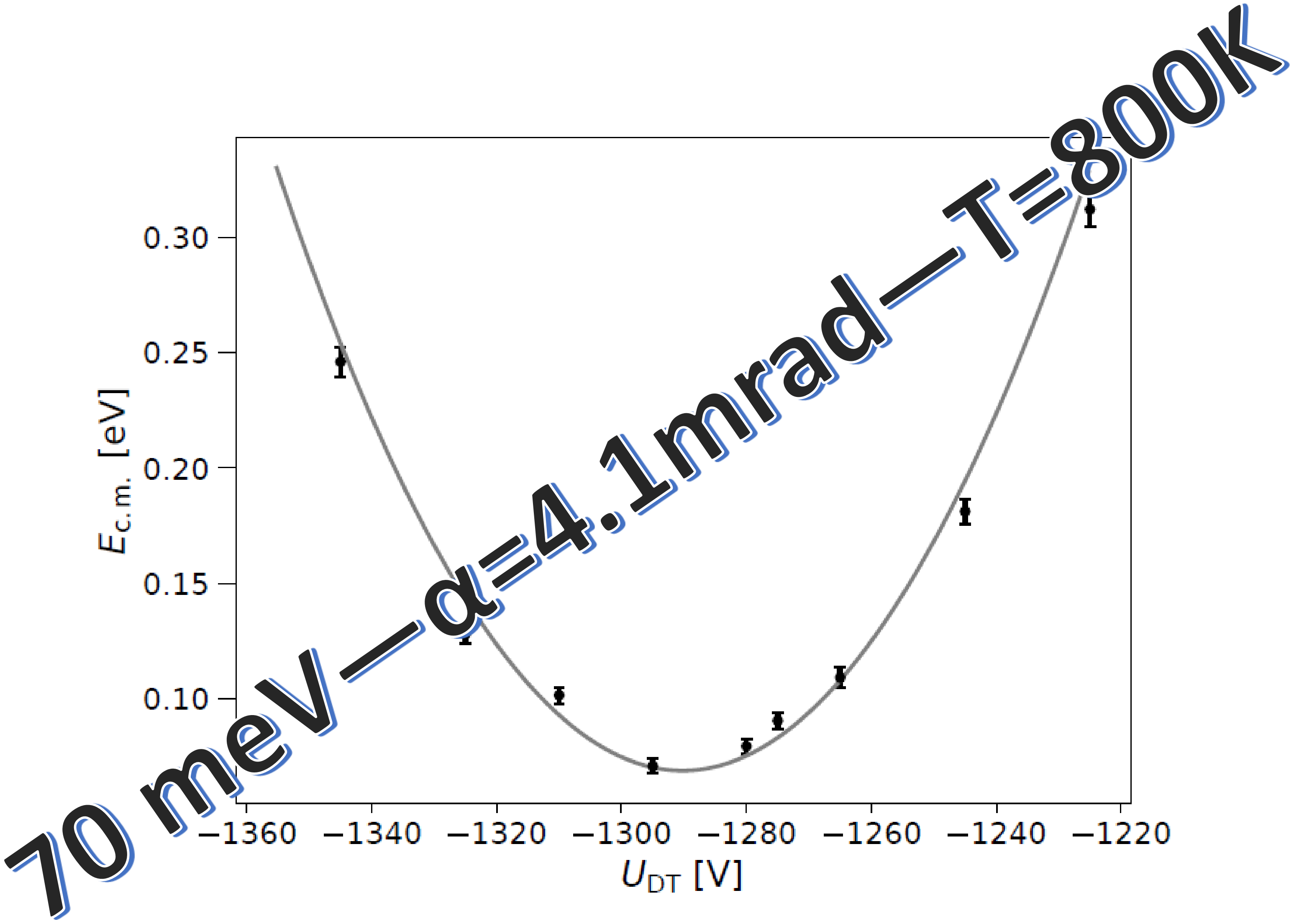


MCP with
phosphor screen
anode.

$$D^-(1s^2) + Li^+(1s^2) \rightarrow D(1s) + Li(1s^2 n l)$$






70 meV—α=4.1mrad—T=800K
$E_{c.m.}$ [eV]
0.30
0.25
0.20
0.15
0.10
−1360
−1340
−1320
−1300
−1280
−1260
−1240
−1220
U_{DT} [V]

Outline

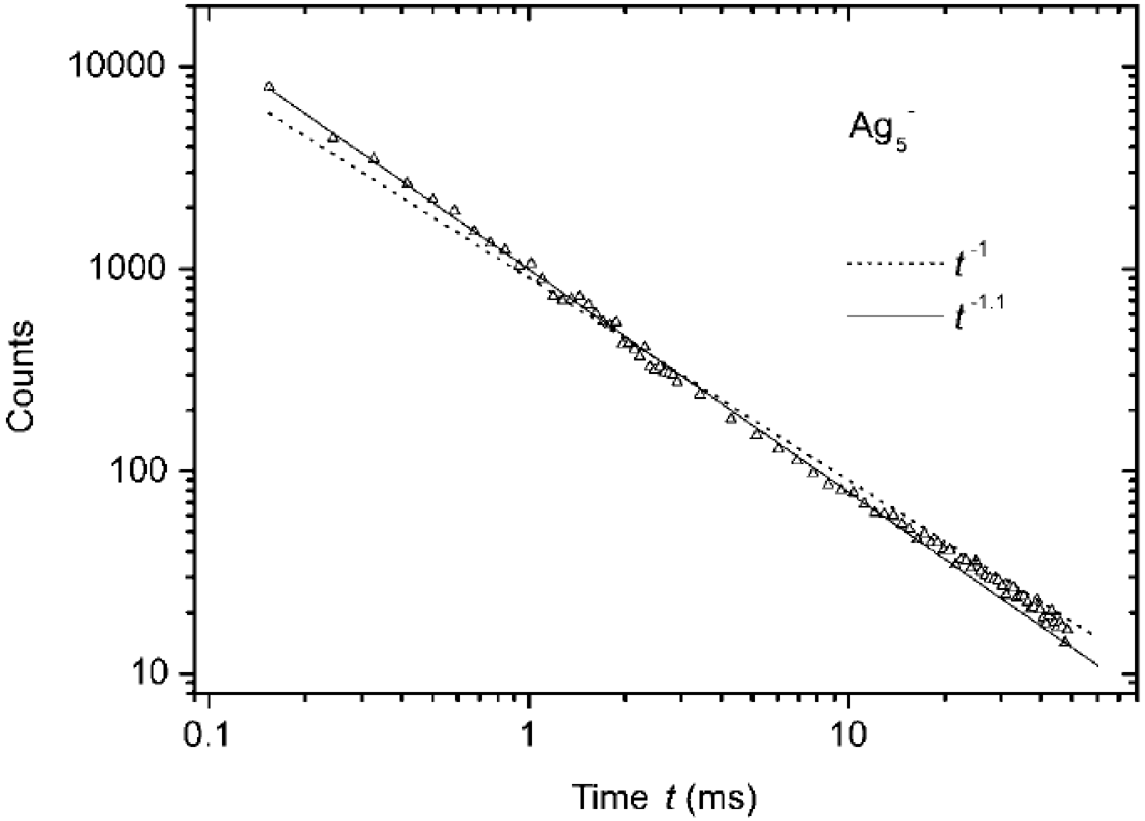
- A description of DESIREE
- Long storage time – cold ions
- Cold collisions??
- Hot stuff in a cold ring

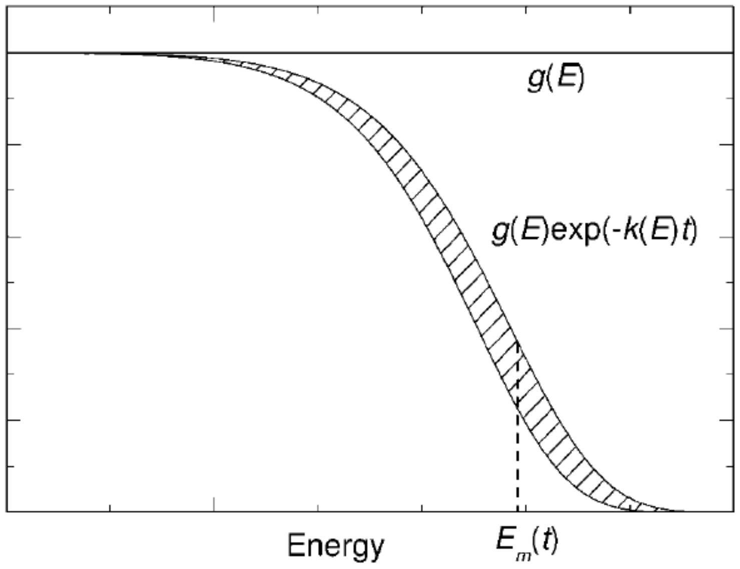
Metal cluster anion decays in DESIREE

- Copper,
  - K. Hansen *et al*, Phys. Rev. A **95**, 022511 (2017)
- Silver
  - E.K. Anderson *et al*, Phys. Rev. A **98**, 022705 (2018)
- Copper and silver dimers -- BO breakdown
  - E.K. Anderson *et al*, submitted to PRL (Nov 20, 2019)

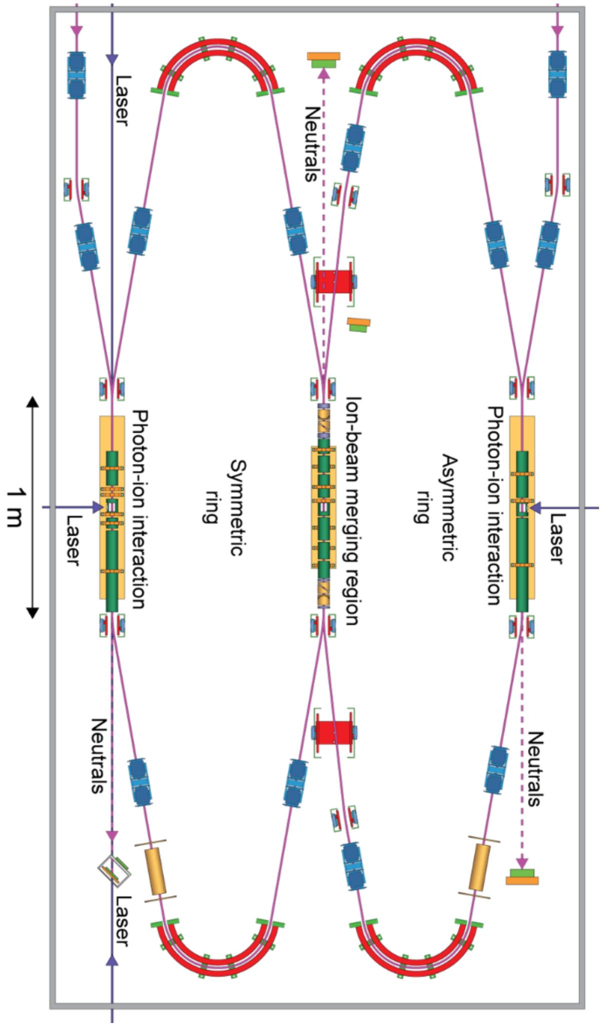
Observation of a $1/t$ Decay Law for Hot Clusters and Molecules in a Storage Ring

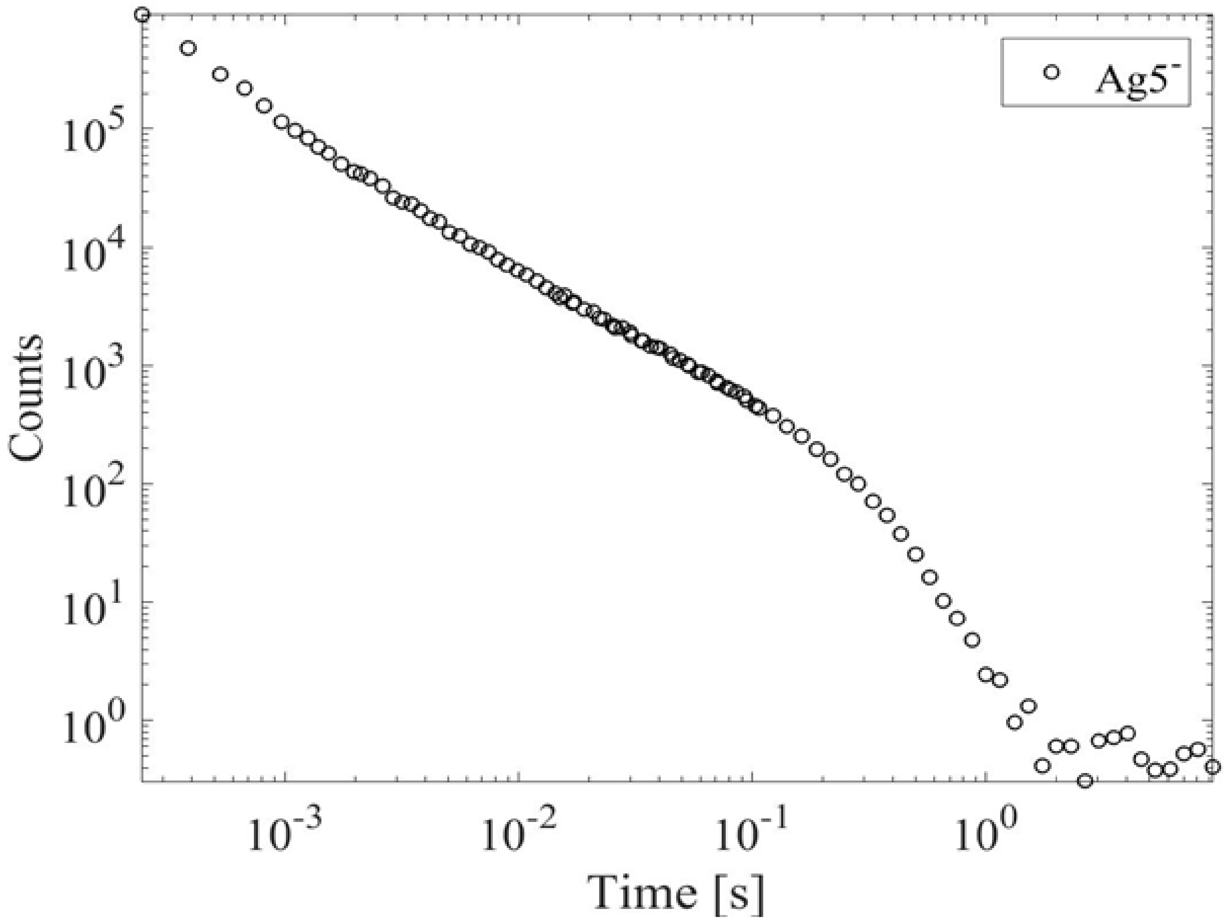
K. Hansen,[1] J. U. Andersen,[1] P. Hvelplund,[1] S. P. Møller,[2] U. V. Pedersen,[2] and V. V. Petrunin[1]
[1]Institute of Physics and Astronomy, University of Aarhus, DK-8000 Aarhus C, Denmark
[2]Institute of Storage Ring Facilities, University of Aarhus, DK-8000 Aarhus C, Denmark
(Received 26 April 2001; published 29 August 2001)





Cluster ions in cryogenic rings/traps – a new time scale!





- E.K. Anderson *et al*, Phys. Rev. A **98**, 022705 (2018)

Decay of highly rotationally and vibrationally excited silver dimer anions.

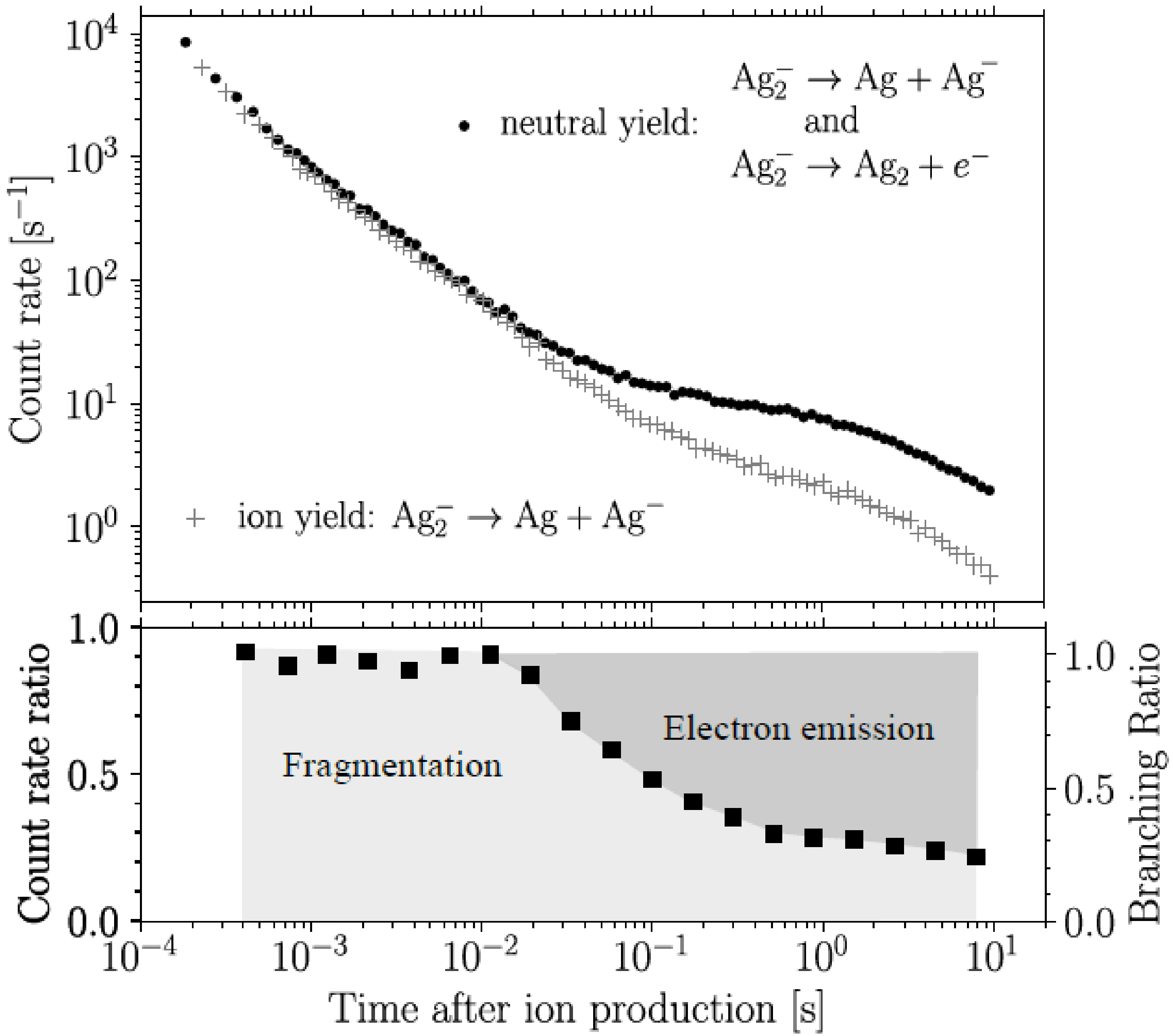
- Submitted Wednesday this week to PRL.

Spontaneous electron emission from hot silver dimer anions: Breakdown of the Born-Oppenheimer approximation

E. K. Anderson,[1,*] A. F. Schmidt-May,[2,1] P. K. Najeeb,[1] G. Eklund,[1] K. C. Chartkunchand,[3] S. Rosén,[1] Å. Larson,[1] K. Hansen,[4,5] H. Cederquist,[1] H. Zettergren,[1] and H. T. Schmidt[1,†]

[1] *Department of Physics, Stockholm University, AlbaNova, SE-106 91 Stockholm, Sweden*
[2] *Institut für Ionenphysik und Angewandte Physik, Universität Innsbruck, A-6020 Innsbruck, Austria*
[3] *Atomic, Optical, and Molecular Physics Laboratory, RIKEN Cluster for Pioneering Research Wako-shi, Saitama 351-0198, Japan*
[4] *Center for Joint Quantum Studies and Department of Physics, Tianjin University, 92 Weijin Road, Tianjin 300072, China*
[5] *Department of Physics, University of Gothenburg, 41296 Gothenburg, Sweden*

(Dated: November 20, 2019)



Count rate [s^{-1}]
10^4
10^3
10^2
10^1
10^0
neutral yield:
$Ag_2^- \rightarrow Ag + Ag^-$
and
$Ag_2^- \rightarrow Ag_2 + e^-$
ion yield: $Ag_2^- \rightarrow Ag + Ag^-$
Count rate ratio
1.0
0.5
0.0
Fragmentation
Electron emission
Branching Ratio
10^{-4}
10^{-3}
10^{-2}
10^{-1}
10^0
10^1
Time after ion production [s]

Previous observation of Fragmentation for t<15 ms

PRL **94,** 113201 (2005) PHYSICAL REVIEW LETTERS week ending 25 MARCH 2005

Nonthermal Power Law Decay of Metal Dimer Anions

J. Fedor,[1] K. Hansen,[2] J. U. Andersen,[3] and P. Hvelplund[3]

Institute for Ion Physics, Technikerstrasse 25, Leopold-Franzens Universität, A-6020 Innsbruck, Austria
[2]*Department of Physics, Gothenburg University, SE-41296 Gothenburg, Sweden*
[3]*Department of Physics and Astronomy, University of Aarhus, DK-8000 Aarhus C, Denmark*

(Received 6 October 2004; published 23 March 2005)

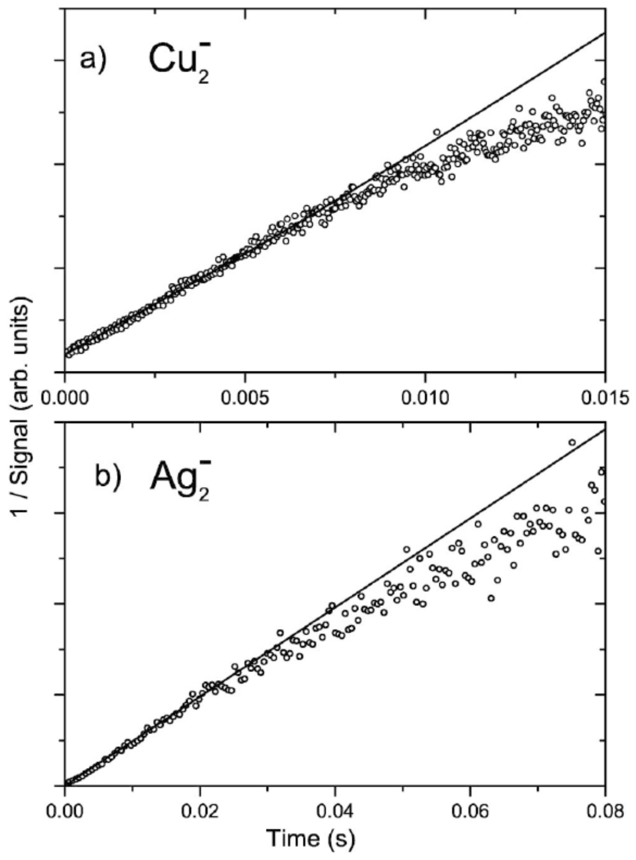


FIG. 1. The time dependence of the reciprocal of the signal in the neutrals detector for (a) Cu_2^- and (b) Ag_2^-. The counts are bunched into one point for each turn in the storage ring for Cu_2^-. For Ag_2^- three turns are bunched into one point. The offset in time for Cu is due to the finite time of extraction from the source. Also shown is the time dependence of the decay intensity (solid lines), calculated using Eq. (1) with identical population of all rotational and vibrational states, $\rho(v, L) = \text{const}$.

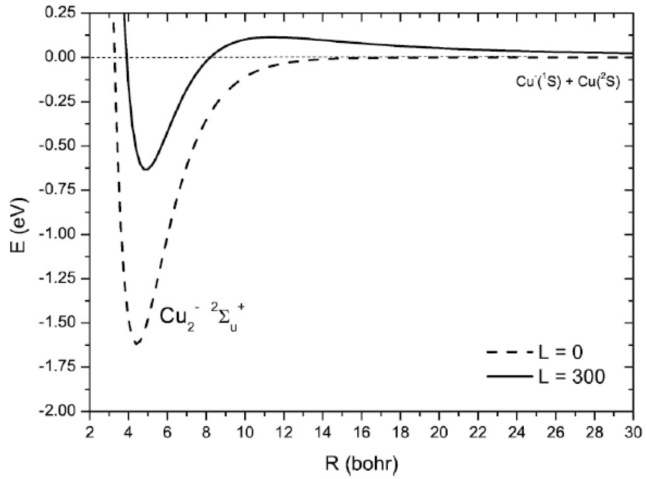
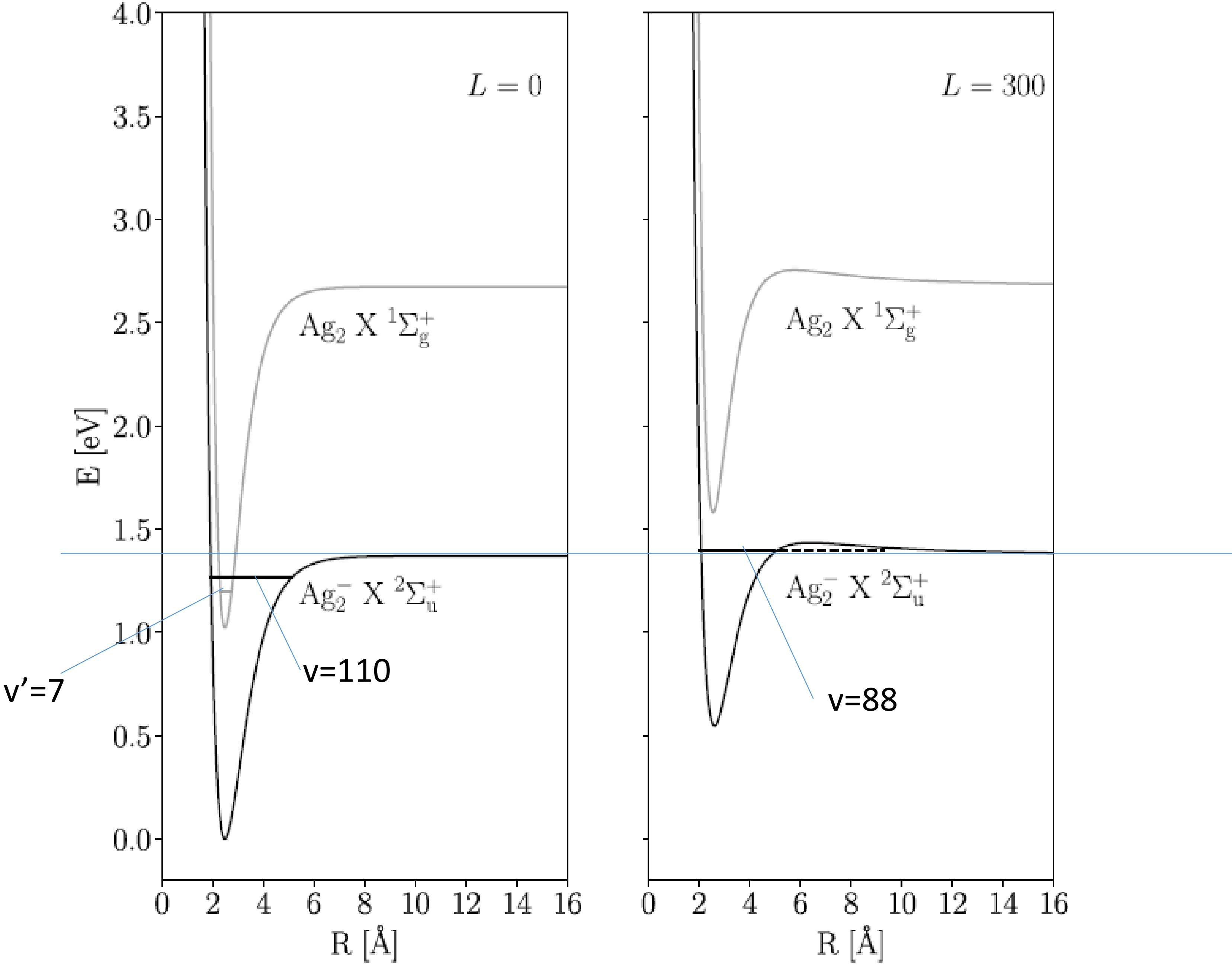


FIG. 2. Potential energy curve of the ground electronic state of Cu_2^- [9] (dashed curve). The solid line represents the effective potential at the rotational quantum number $L = 300$.

- Examples of ro-vibrational levels

- High L: Fragmentation

- Low L: electron emission

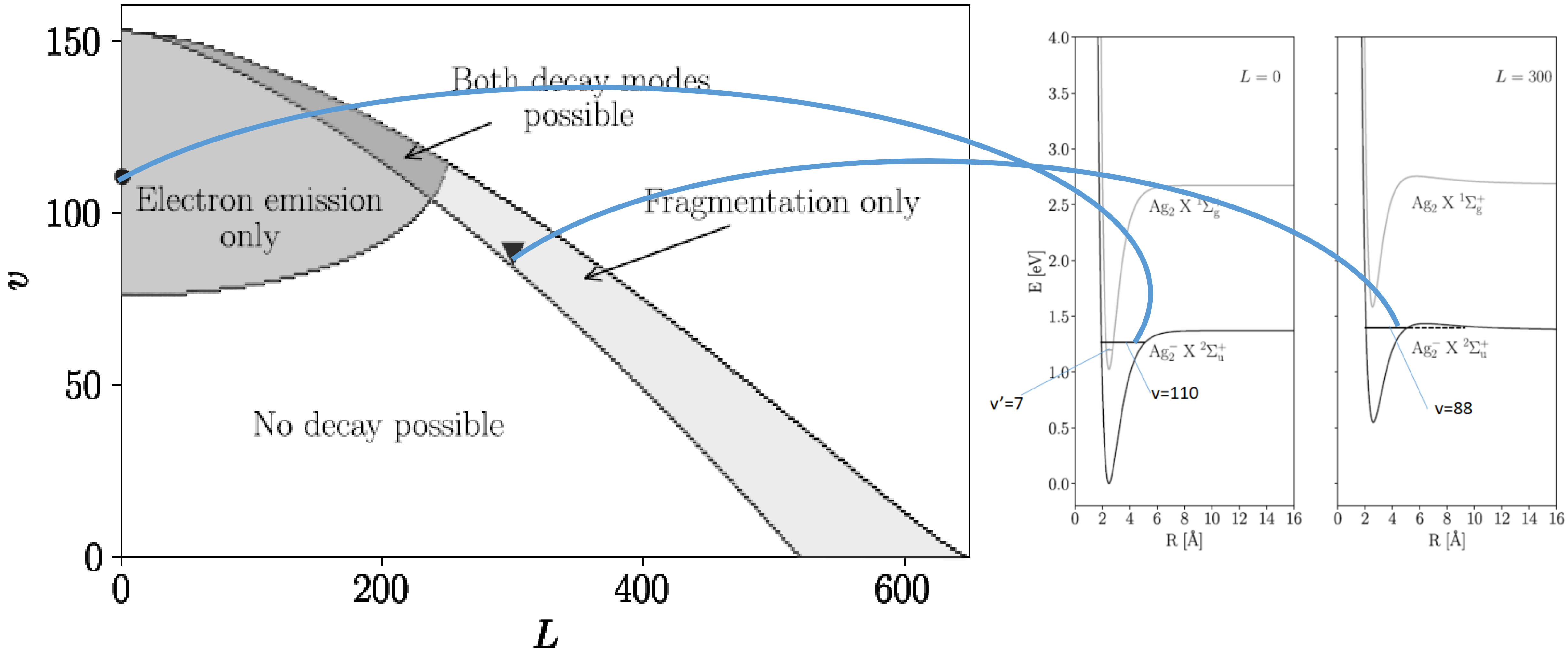


What we know

1. Ag_2^- is not infrared active (no E-dipole moment)
   - This means that the individual dimer anions remain in their given ro-vibrational level until they decay or are lost in a collision.
2. Some levels can decay via fragmentation and some via electron emission.
3. The time scales are varying tremendously.

Calculation of ro-vibrational energies

- Use Morse potentials with the proper constants and the centrifugal distortion.
- Solve the Schrödinger equation for the nuclear motion numerically.
- Check for every level whether or not it is energetically allowed to decay by electron emission and/or fragmentation.
  - For the question of electron emission it is assumed that the angular momentum is not changed in the process.



Both decay modes possible
Electron emission only
Fragmentation only
No decay possible
v
L
0
50
100
150
200
400
600
L = 0
L = 300
E [eV]
R [Å]
Ag_2 X $^1\Sigma_g^+$
Ag_2^- X $^2\Sigma_u^+$
v'=7
v=110
v=88

The electron emission

- From detailed balance arguments we find from the rate when e-emission start to dominate (1/(100 ms)=10 s^{-1}) that the cross section at 20 meV for attaching an electron to a neutral dimer in a low vibrational level by deposition of the energy into vibrational excitation is of the order of 10^{-9} $Å^2$.

$$k \sim \sigma \frac{gm}{\pi^2 \hbar^3} \varepsilon^2$$

V. Weisskopf, Phys. Rev. **52**, 295 (1937).
K. Hansen, *Statistical physics of nanoparticles in the gas phase*, Springer Series on Atomic, Optical, and Plasma Physics, Vol. 73 (Springer Dordrecht, 2013).

DESIREE references.

- Instrumental:
  - Design : **R.D. Thomas** *et al.*, Rev Scientific Instruments **82** 065112 (2011)
  - Commissioning: **H.T. Schmidt** *et al.*, Rev Scientific Instruments **84** 055115 (2013)
  - Electrospray/trap setup **N. deRuette** *et al.*, Rev Scientific Instruments **89** 075102 (2018)
- Atomic negative ions
  - S^-, Se^-, Te^-: **E. Bäckström** *et al.*, Phys. Rev. Lett. **114**, 143 003 (2015)
  - Ni^-: **M. Kaminska** *et al.*, Phys. Rev. A **93**, 012512 (2016)
  - Pt^-: **K. Chartkunchand** *et al.*, Phys. A **94**, 032501 (2016)
- Molecular anions
  - OH^-: **H.T. Schmidt** *et al*, Phys. Rev Lett. **119**, 073001 (2017)
- Metal cluster anions
  - Cu_n^-: **K. Hansen** *et al*, Phys. Rev A **95**, 022511 (2017)
  - Ag_n^-: **E.K. Anderson** *et al*, Phys. Rev A **98**, 022705 (2018)
- Cations
  - PAHs: **M. H. Stockett** *et al*, Faraday Discuss. **217**, 126 (2019)
- Carbon clusters
  - C_n^{2-}: **K. Chartkunchand** *et al.*, Rev Scientific Instruments **89** 033112 (2018)
  - C_n^-: **J. N. Bull** *et al.*, J. Chem. Phys **151** 114104 (2019)


Benjamin Gatchell
John Alexander
Michael Wolf
Gustav Eklund
Richard Thomas
Mikael Blom
Mikael Björkhage
Henning Zettergren
Henning Schmidt
Patrik Löfgren
Naoko Kono
MingChao Ji
PK Najeeb
Henrik Hartman Malmö University
Paul Barklem, Uppsala University
Åsa Larson
Wolf Geppert
Dag Hanstorp Gothenburg
Mark Stockett
Moa Kristiansson
Emma Anderson
Stefan Rosén
Linda Giacomozzi
Peter Reinhed
Nathalie de Ruette
Henrik Cederquist

International users/collaborators include:

Klavs Hansen, Tianjin University
Daniel Gibson/Wesley Walter, Denison University
Jerôme Bernard/Serge Martin, Université Lyon 1
Christine Joblin, Université Paul Sabatier, Toulouse
Robin Golser, Universität Wien
Hubert Gnaser Universität Kaiserslautern
Group of Paul Scheier, Universität Innsbruck
James Bull, Norwich University
Markus Schöffler, Frankfurt University
KC. Chartkunchand, RIKEN
Paola Bolognesi and Lorenzo Avaldi, University of Rome
Jeff Thomson et al, U. of Nevada, Reno
…

New COST Action

- CA18212 - Molecular Dynamics in the Gas Phase (MD-GAS)
- Starting date: 12 November 2019
- Three working groups:
  1. New high-performance instrumentation and experimental methods to study gas phase molecular dynamics **at ion-beam storage rings and traps**, at synchrotrons and X-ray facilities.
  2. Survival and destruction of molecules following their processing by heavy particles, electrons, or photons.
  3. Charge-, energy flow, and molecular growth processes in intermolecular and intracluster reactions.
- Open to COST members (38 countries). Non-COST members can join on the basis of mutual benefits

https://www.cost.eu/actions/CA18212