

# Heavy Molecular Ions in Electron Cooler Storage Rings

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Claude Krantz

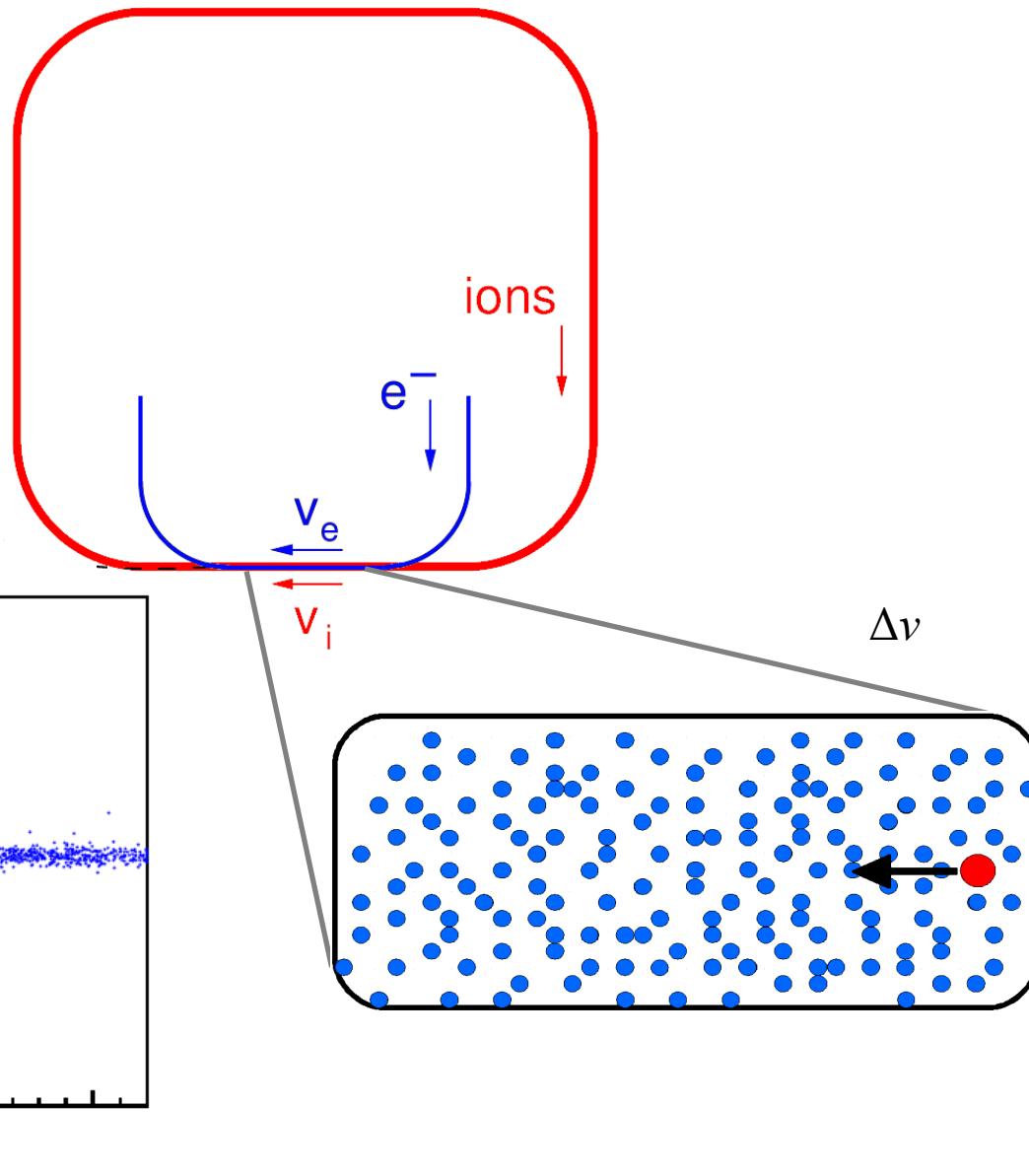
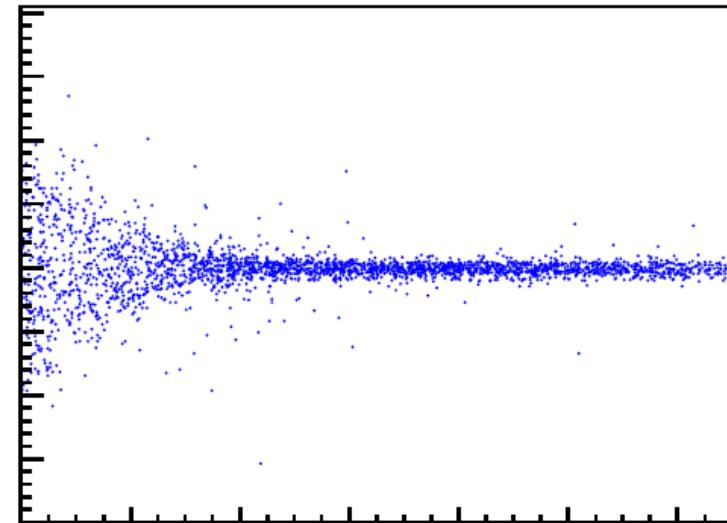
Max Planck Institute for Nuclear Physics





# e-Coolers

“Electron cooling”



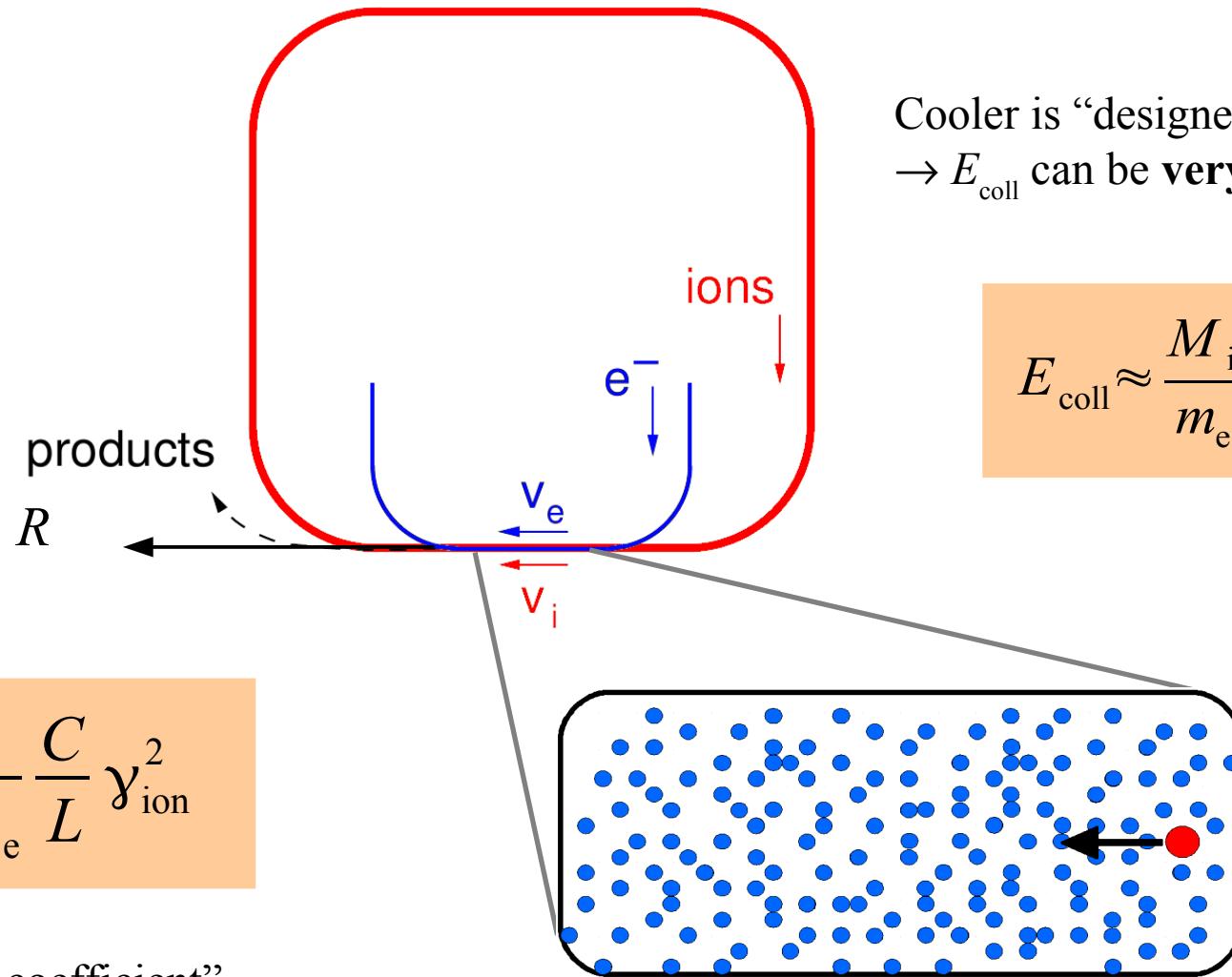


# e-Cooler: Low energy electron-ion collider

$$\alpha = \frac{R}{N_{\text{ion}} n_e} \frac{C}{L} \gamma_{\text{ion}}^2$$

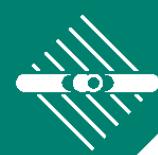
reaction “rate coefficient”

$$\alpha = \langle \sigma(v) v \rangle$$



Cooler is “designed” for  $v = 0$   
 $\rightarrow E_{\text{coll}}$  can be **very small** (meV!)

$$E_{\text{coll}} \approx \frac{M_i}{m_e} \frac{(e \Delta U)^2}{4 E_i}$$



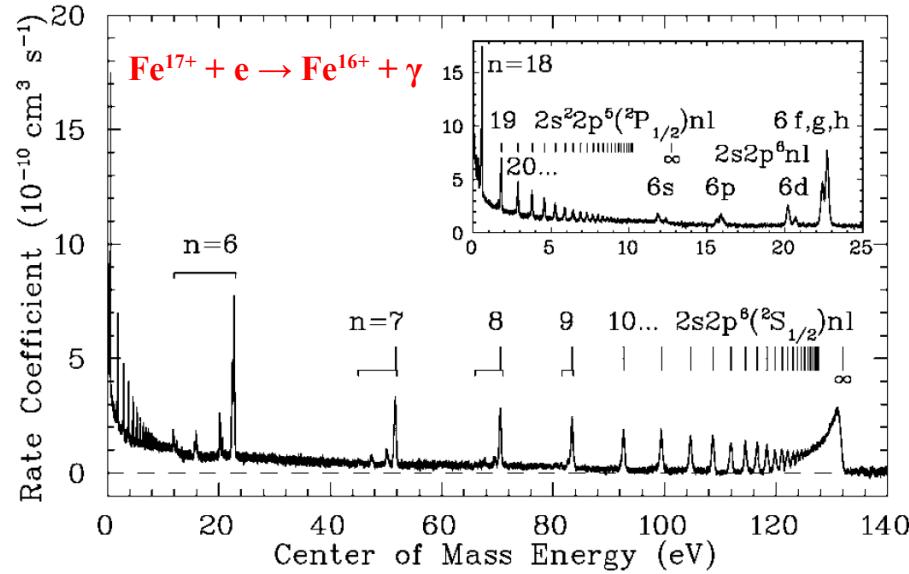
# e-Cooler: Low energy electron-ion collider

Recombination rates of  
HCl in astrophysical plasma

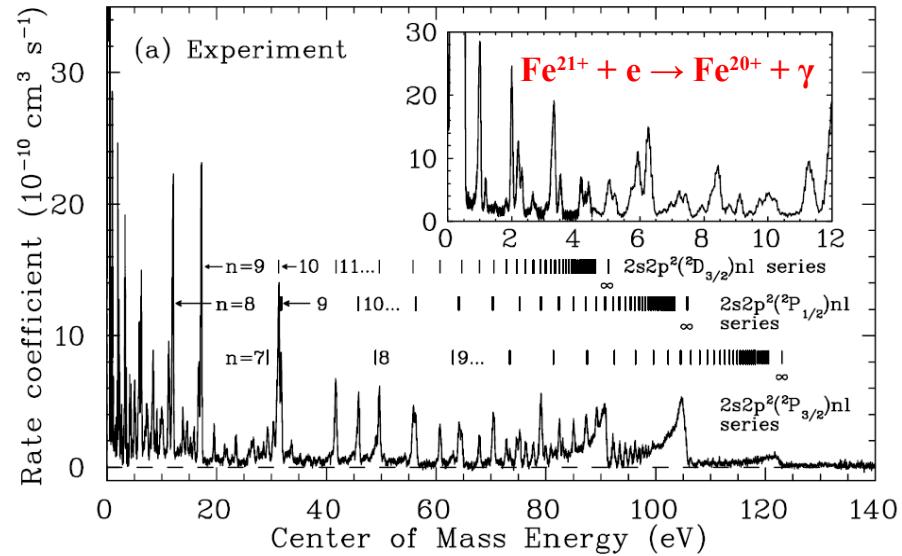
TSR (MPIK) 1988 - 2012



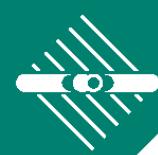
$$\alpha = \langle \sigma(v)v \rangle$$



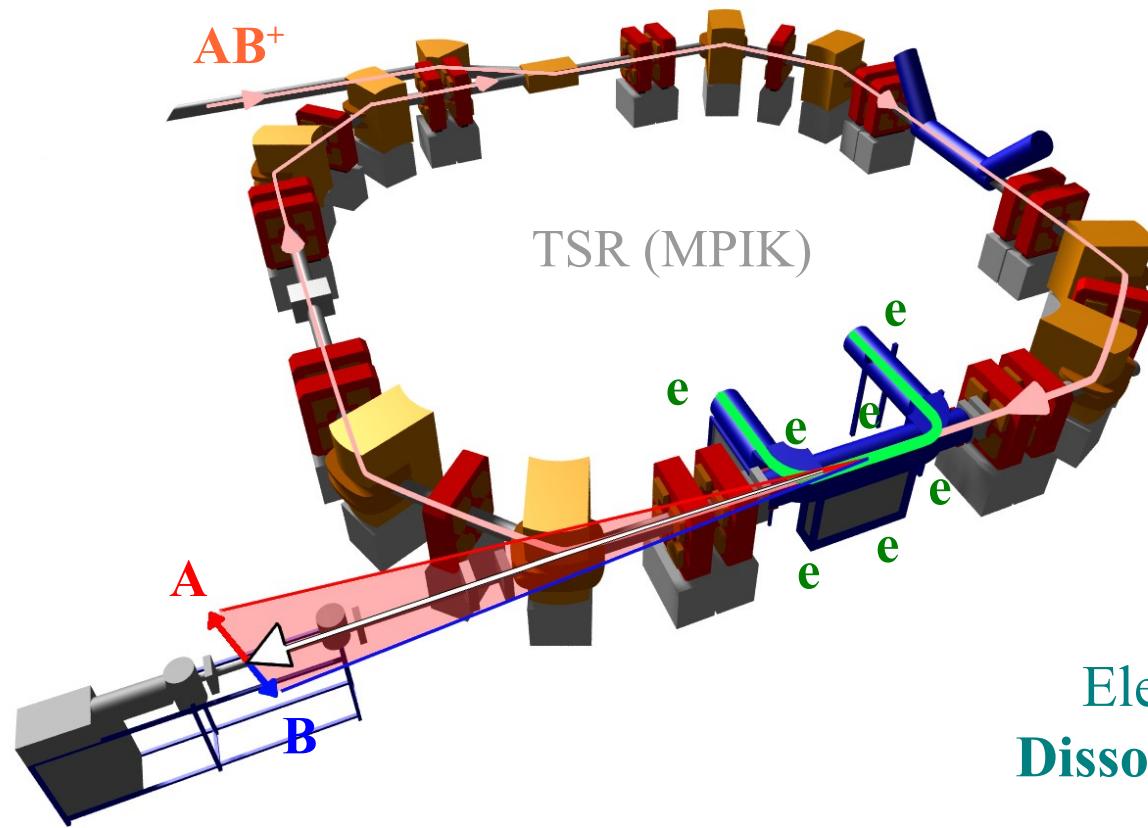
[Savin, ApJ 489 (1997)]



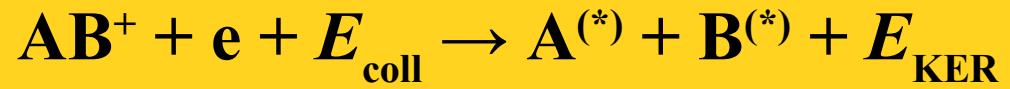
[Savin, ApJ Suppl. Ser. 147 (2003)]

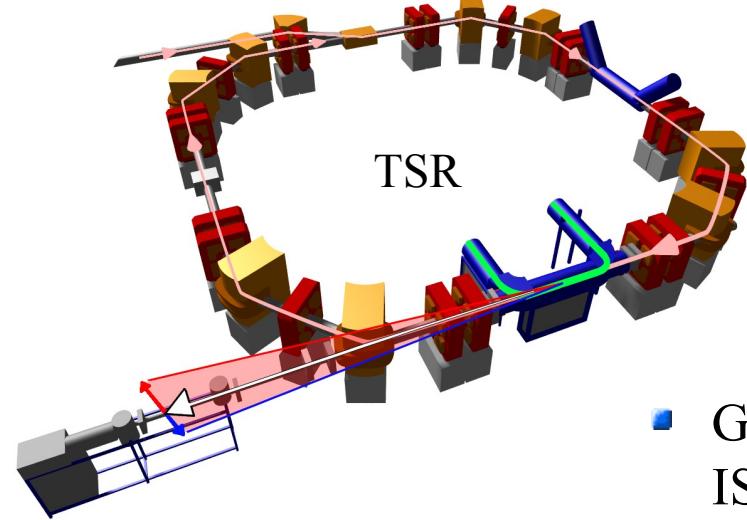


# Recombination of molecular ions

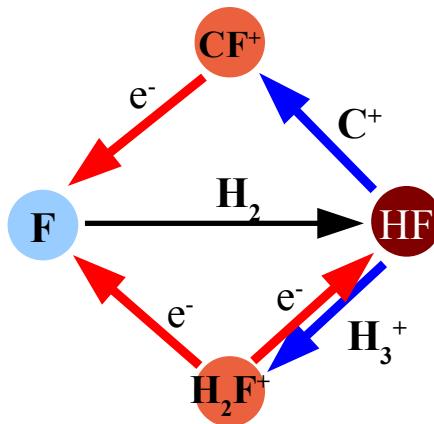


Electrons and molecular ions:  
**Dissociative Recombination (DR)**





# DR of molecular ions



- Gas-phase chemistry in ISC:  
 $\text{H}_3^+ + \text{e} \rightarrow \text{H} + \text{H} + \text{H}$

e.g. [Petrignani, PRA 83 (2011); Kreckel, PRA 82 (2010)]

- A source of energetic products in cold environments



[Buhr, PRL 105 (2010)]

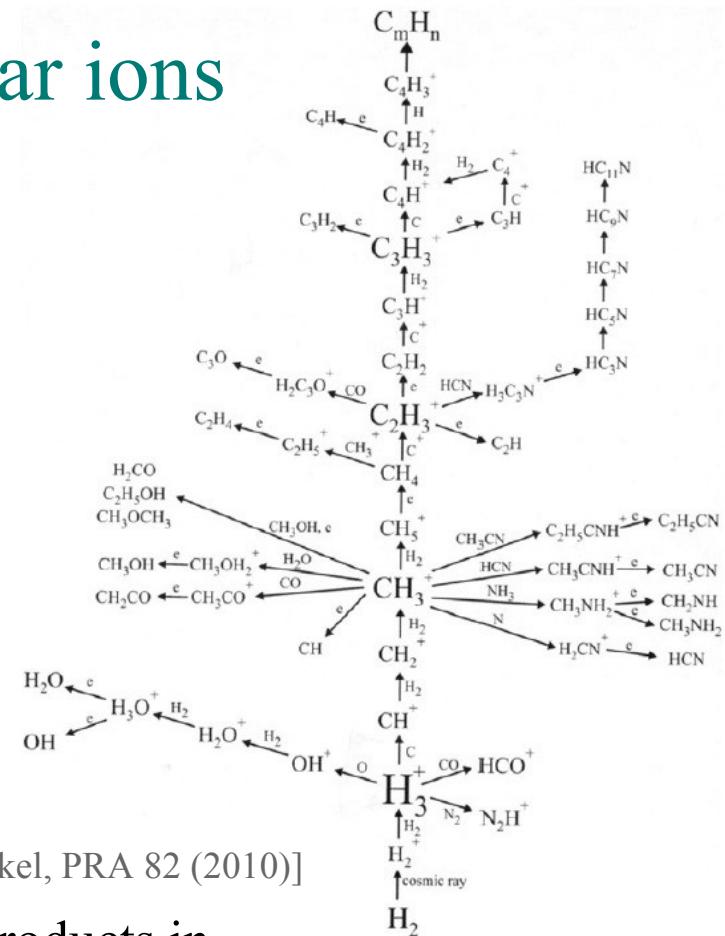


[Mendes, ApJ Lett. 746 (2012)]

- Molecular proxy for H<sub>2</sub>: HF

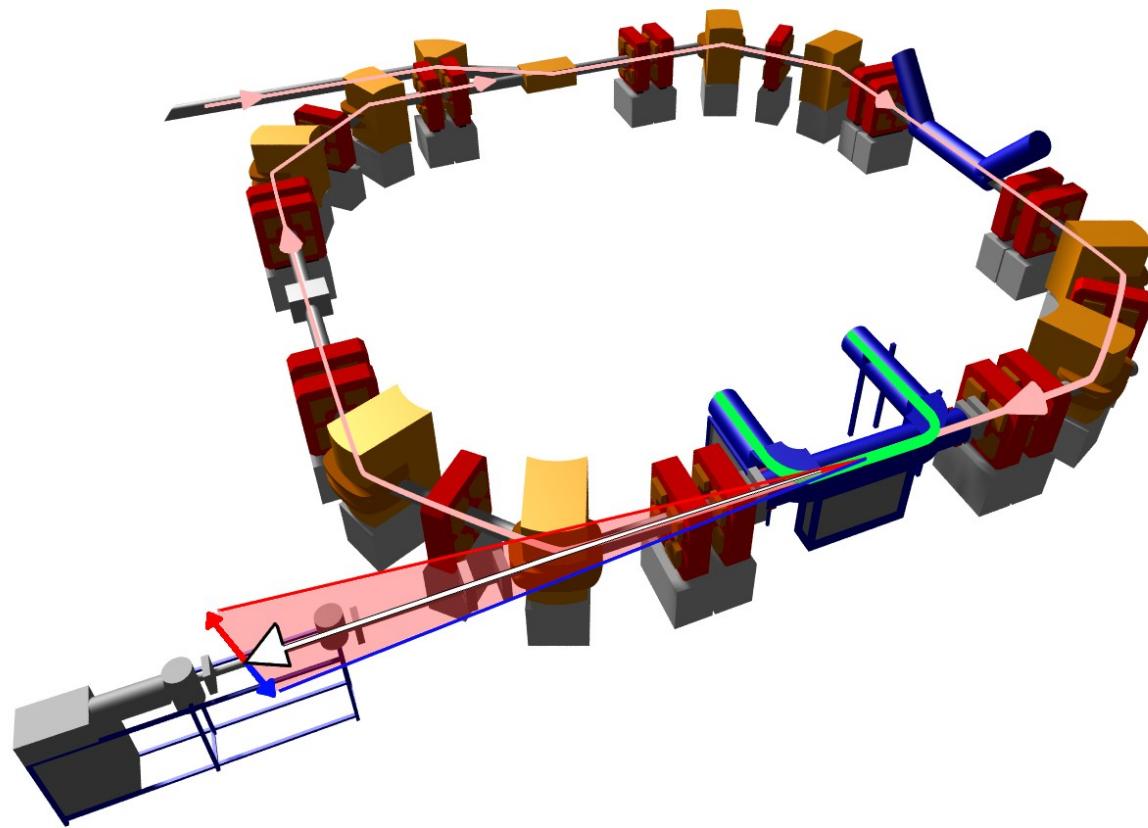


[Novotný, t.b.p.]



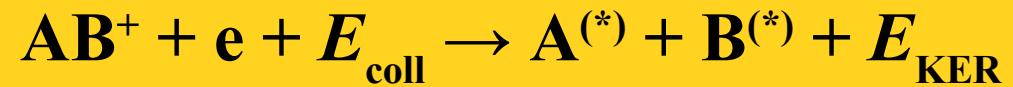


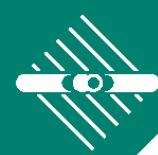
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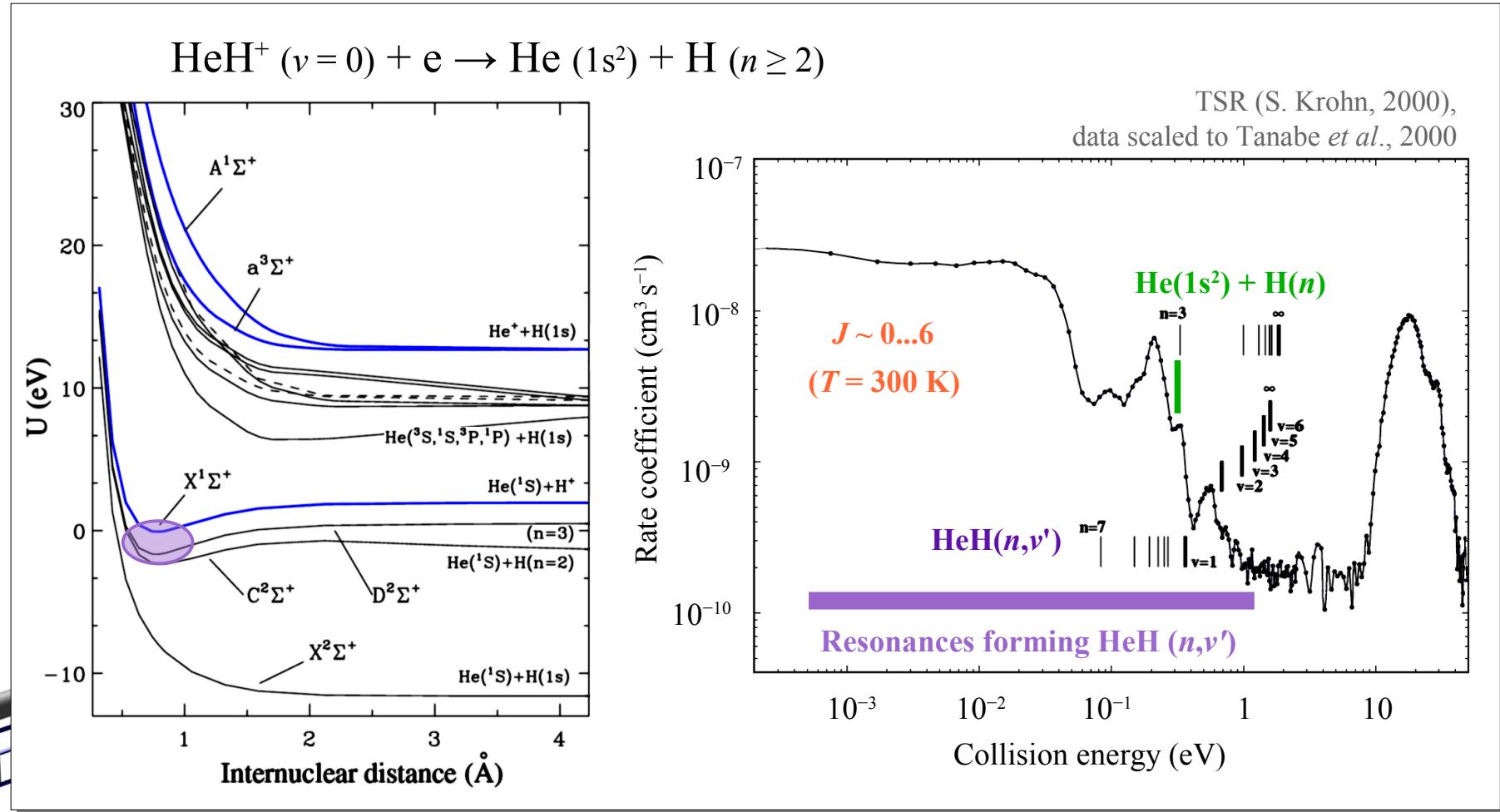
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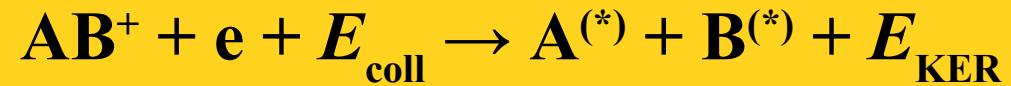


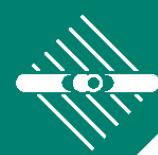
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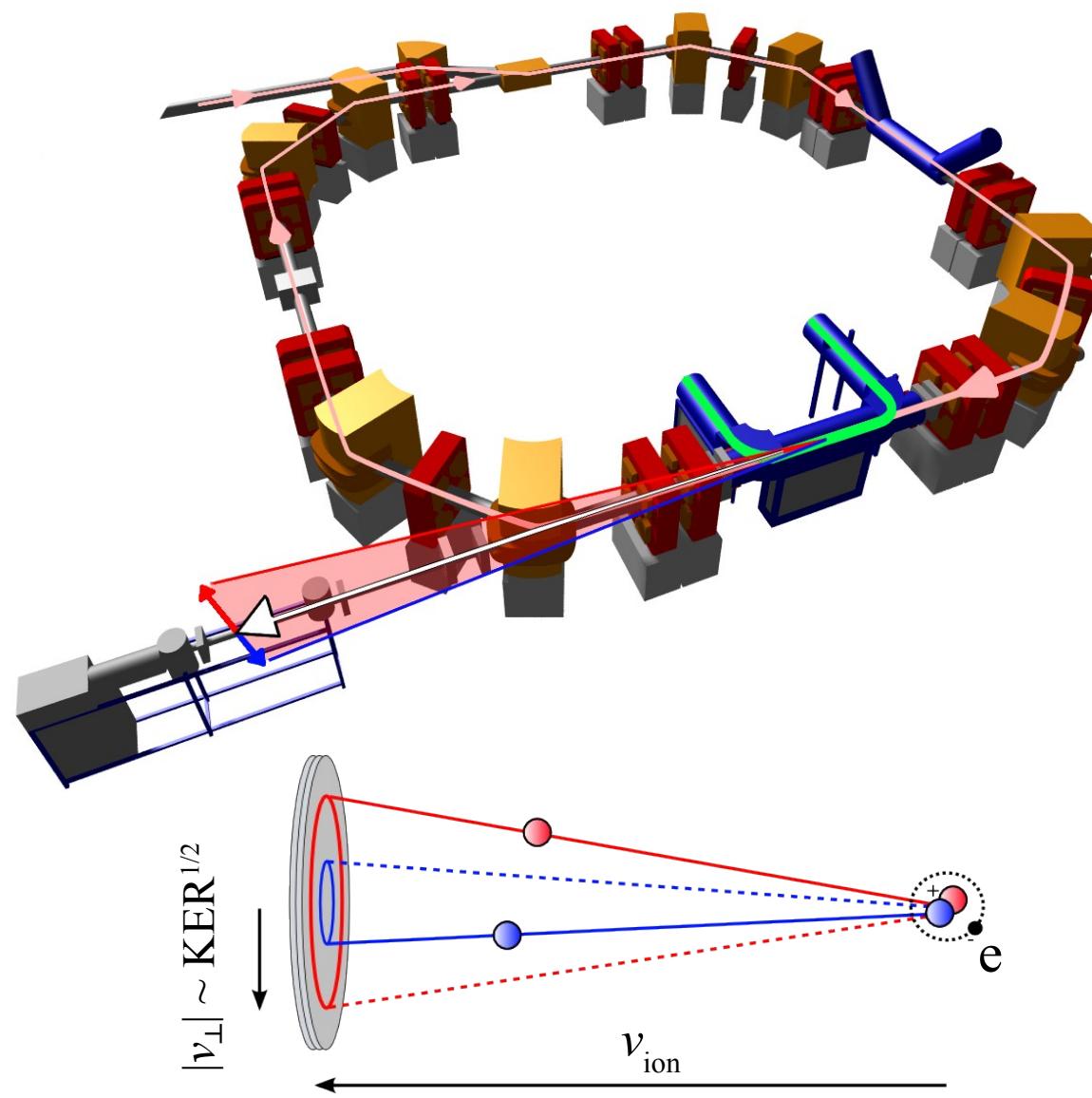
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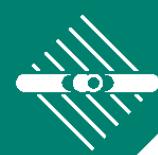
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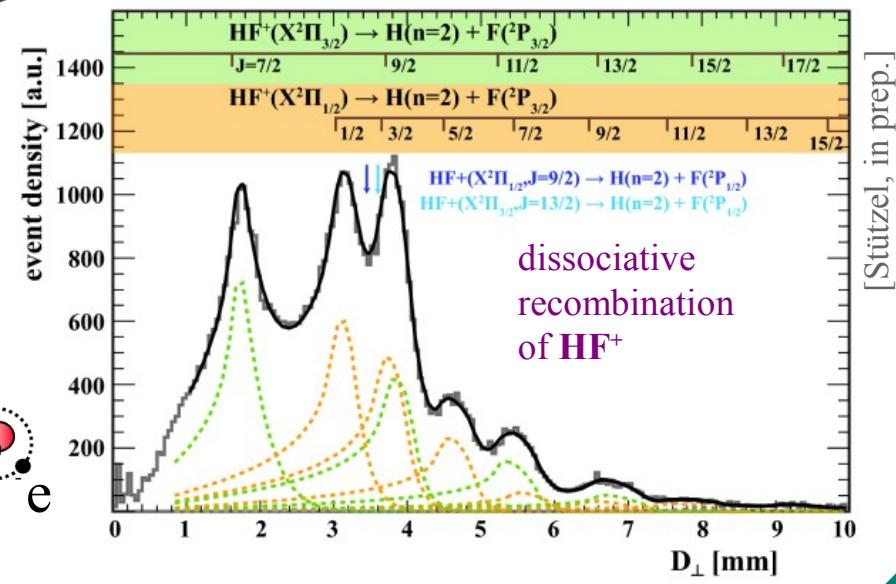
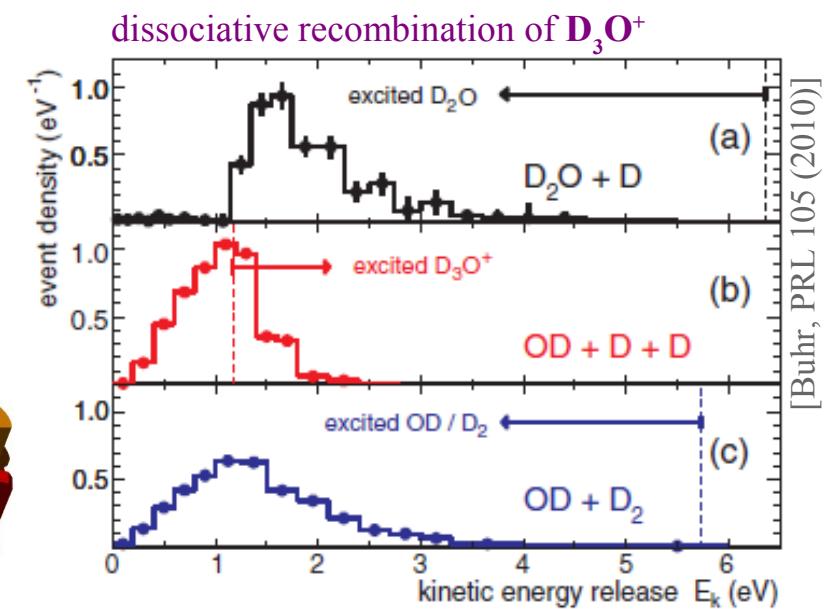
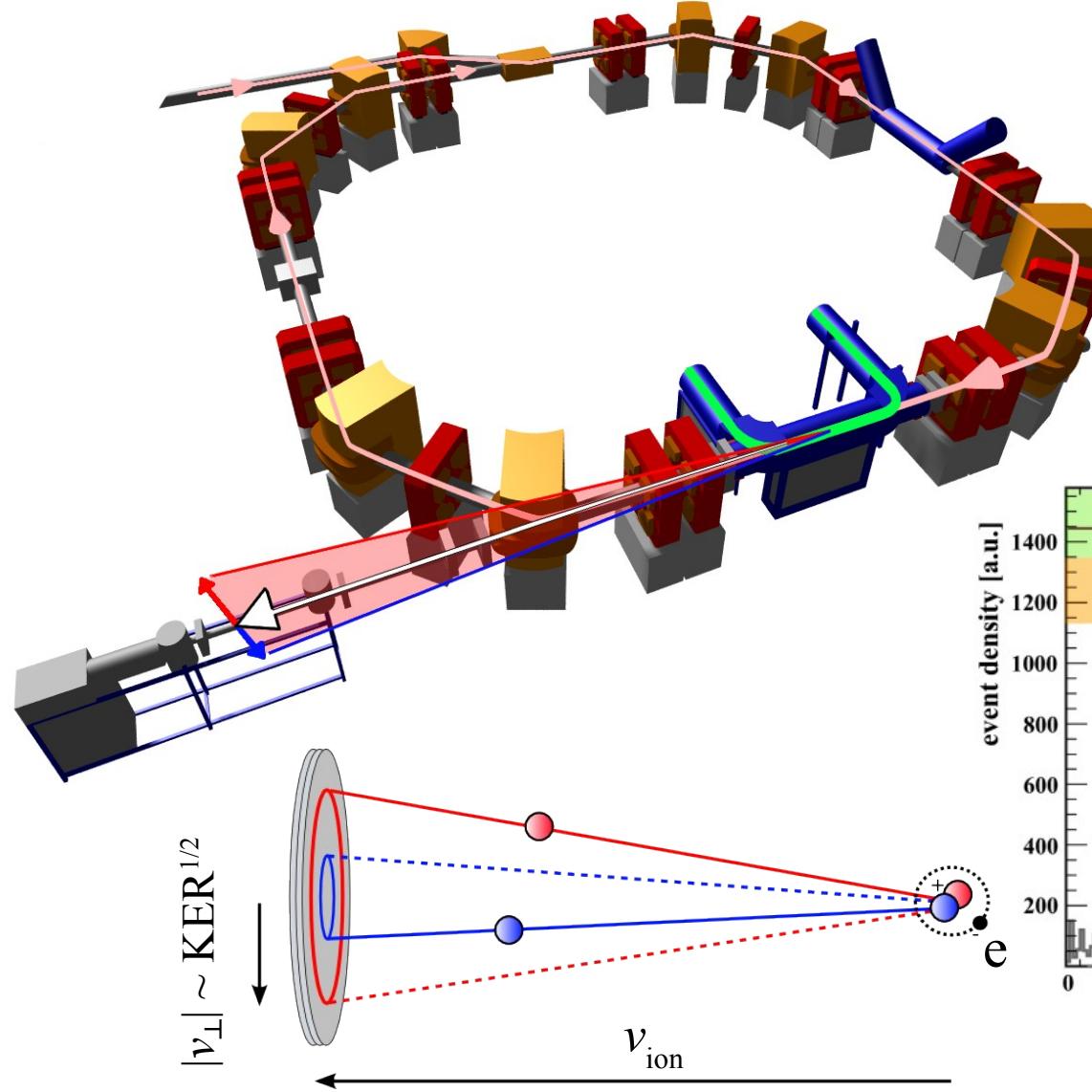


# DR of molecular ions



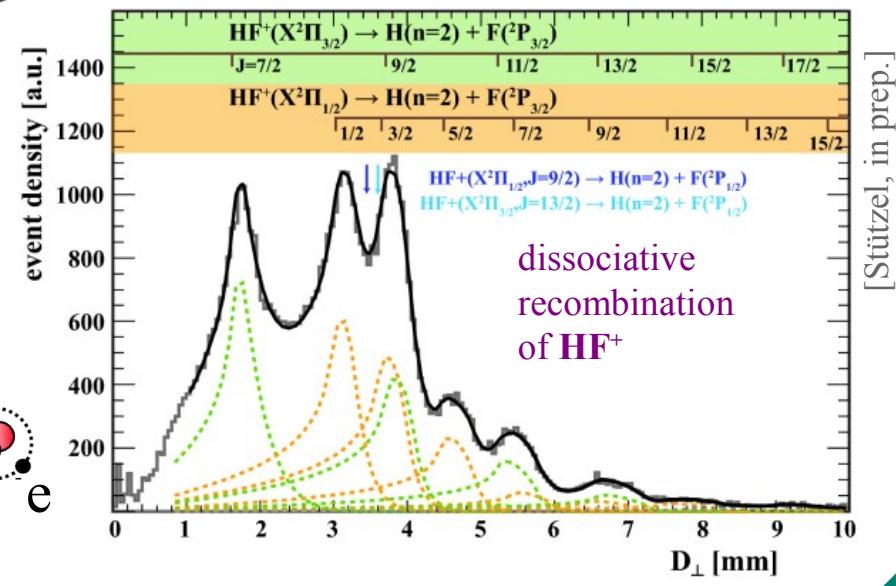
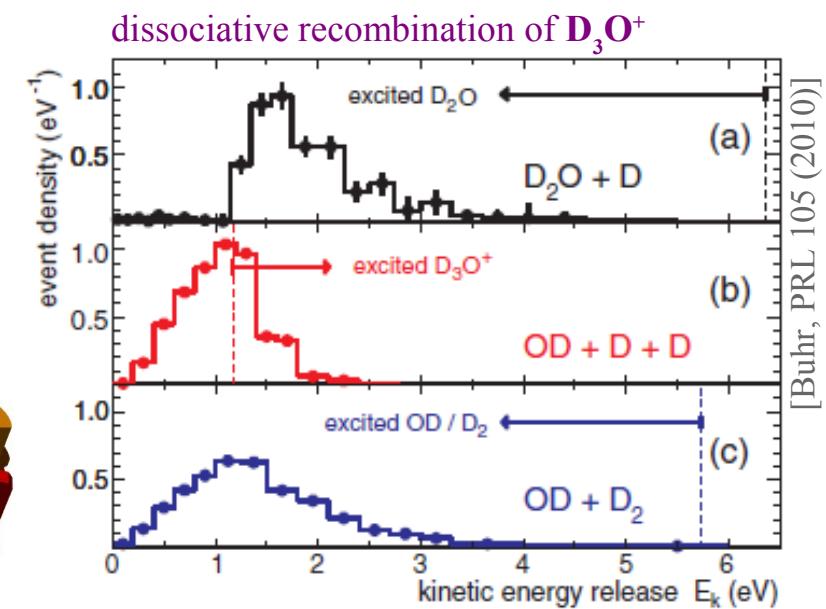
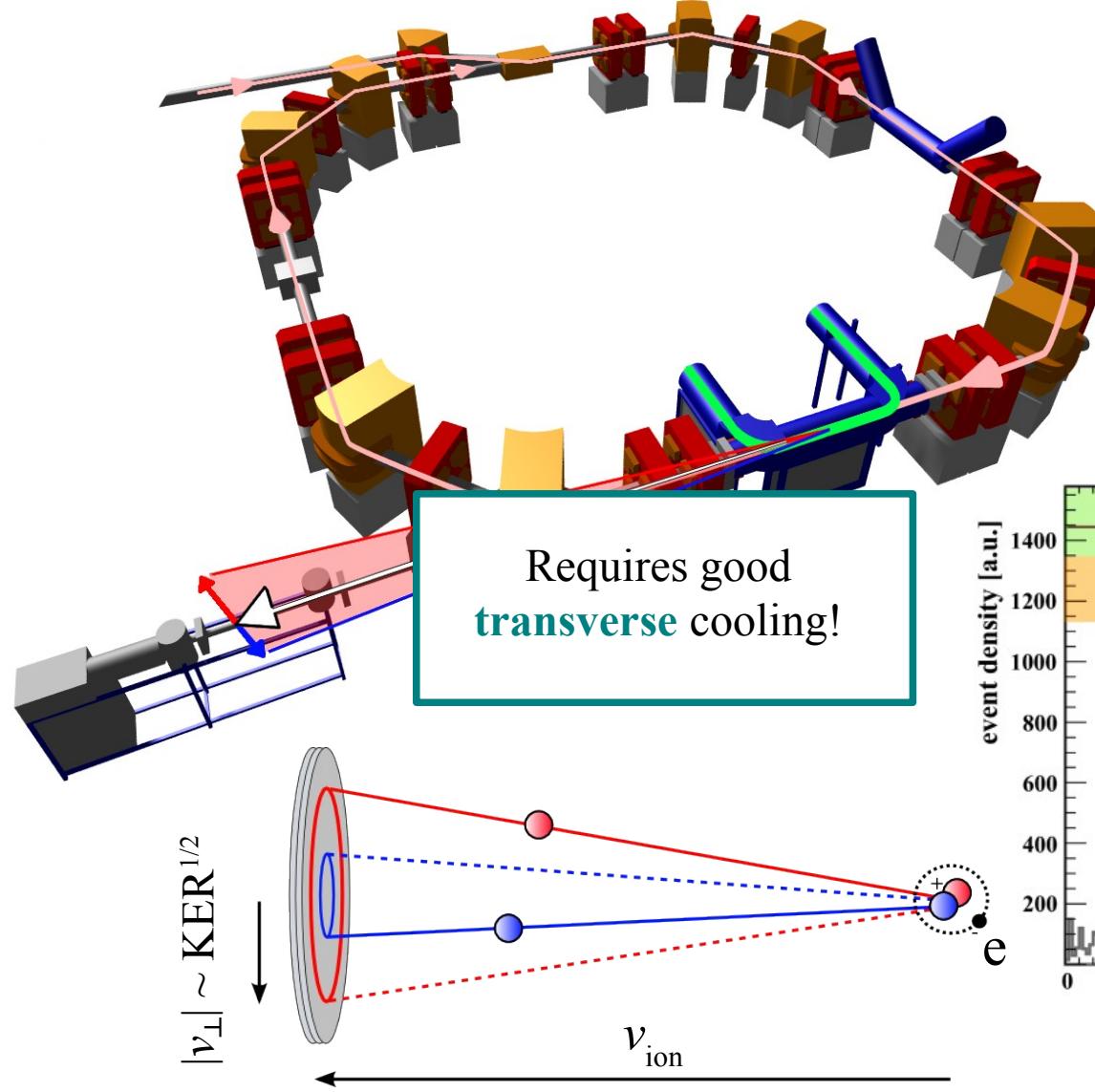


# DR of molecular ions



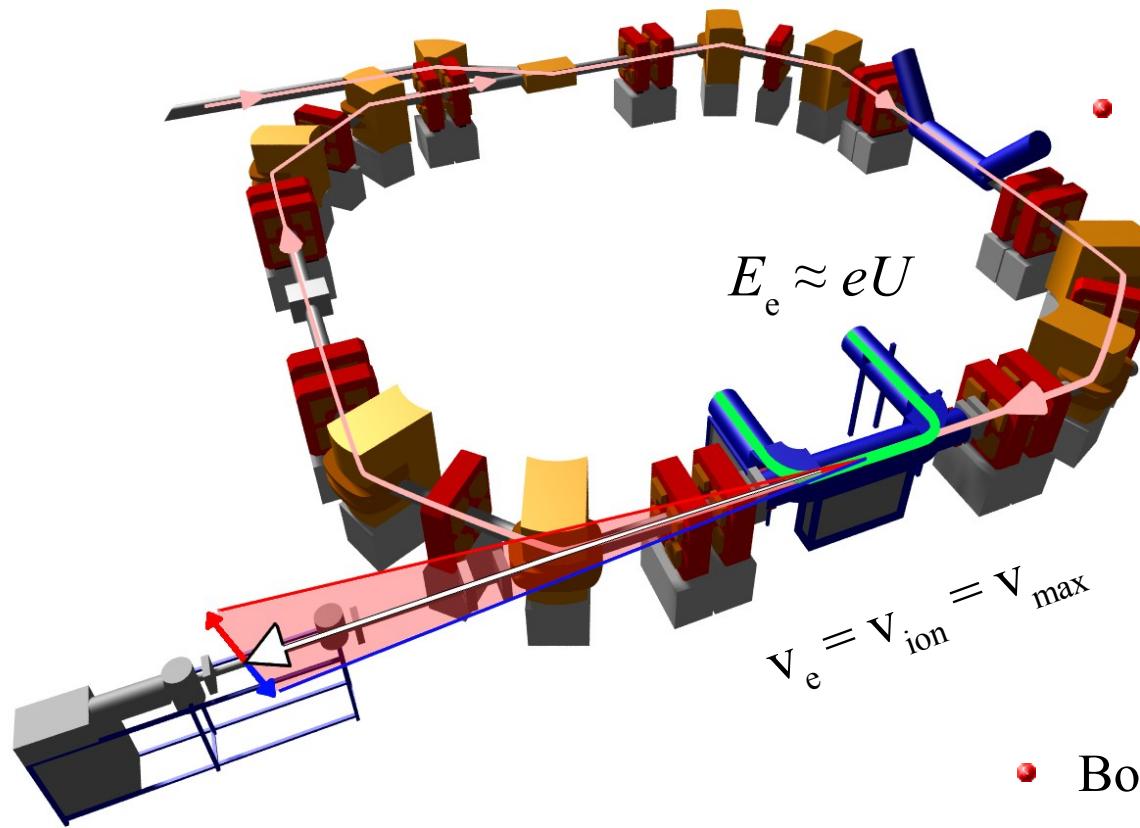


# DR of molecular ions





# Electron cooling molecular ions



- Maximum rigidity:  $r B_{\max}$   
for TSR:  $\approx 1.4$  Tm

- Maximum velocity:

$$v_{ion} = \frac{Z_{ion}}{M_{ion}} r B_{max} \rightarrow U \sim \frac{Z_{ion}^2}{M_{ion}^2}$$

e.g.  $\text{CHD}^+$  (15 u) :  $v_{ion} \sim 0.030 \text{ c}$ ,  $U \sim 230 \text{ V}$   
 $\text{D}_3\text{O}^+$  (22 u) :  $v_{ion} \sim 0.020 \text{ c}$ ,  $U \sim 110 \text{ V}$   
 $\text{DCND}^+$  (30 u) :  $v_{ion} \sim 0.015 \text{ c}$ ,  $U \sim 55 \text{ V}$   
 $\text{HCl}^+$  (36 u) :  $v_{ion} \sim 0.012 \text{ c}$ ,  $U \sim 40 \text{ V}$   
 $\text{D}_2\text{Cl}^+$  (39 u) :  $v_{ion} \sim 0.010 \text{ c}$ ,  $U \sim 31 \text{ V}$

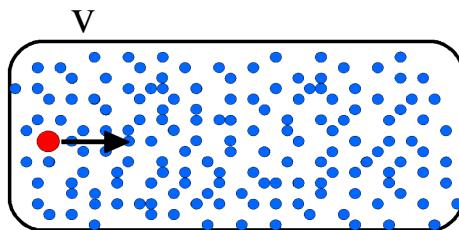
- Both **storage** and **electron cooling** of molecular ions are difficult!

... **short** storage times  
(res. gas losses)

... **long** e-cooling times



# Cooling force

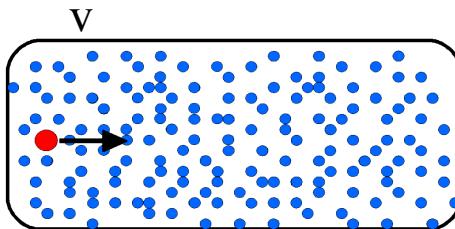


$$\vec{F} \sim -\frac{Z_{ion}^2 n_e}{v^2} \frac{\vec{v}}{|v|}$$



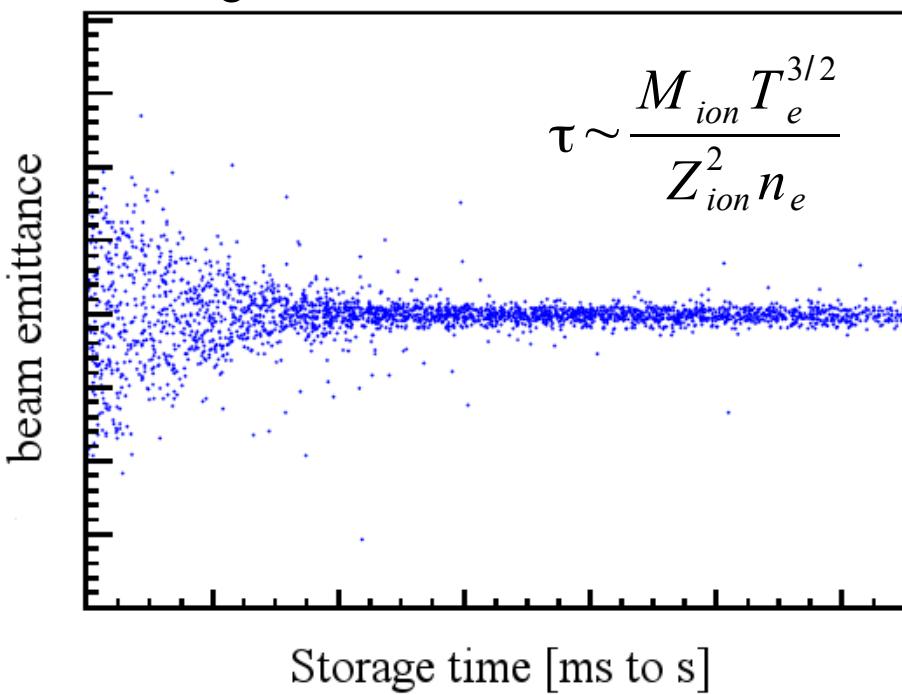


# Cooling force

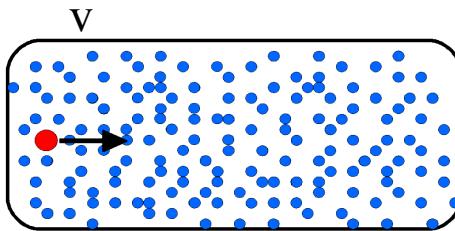


$$\vec{F} \sim -\frac{Z_{ion}^2 n_e}{v^2} \frac{\vec{v}}{|v|}$$

Cooling time

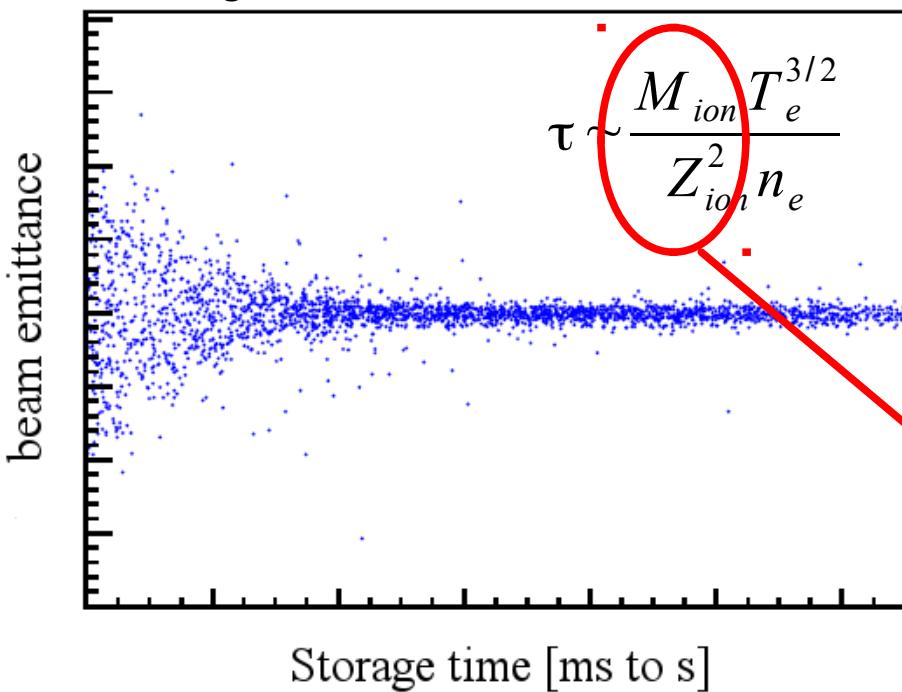


# Cooling force



$$\vec{F} \sim -\frac{Z_{ion}^2 n_e}{v^2} \frac{\vec{v}}{|v|}$$

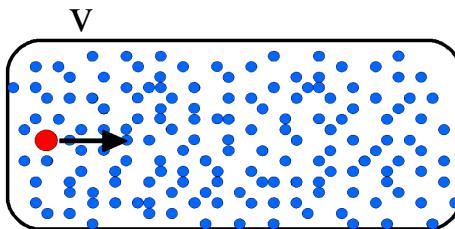
Cooling time



$$\tau \sim \frac{M_{ion} T_e^{3/2}}{Z_{ion}^2 n_e}$$

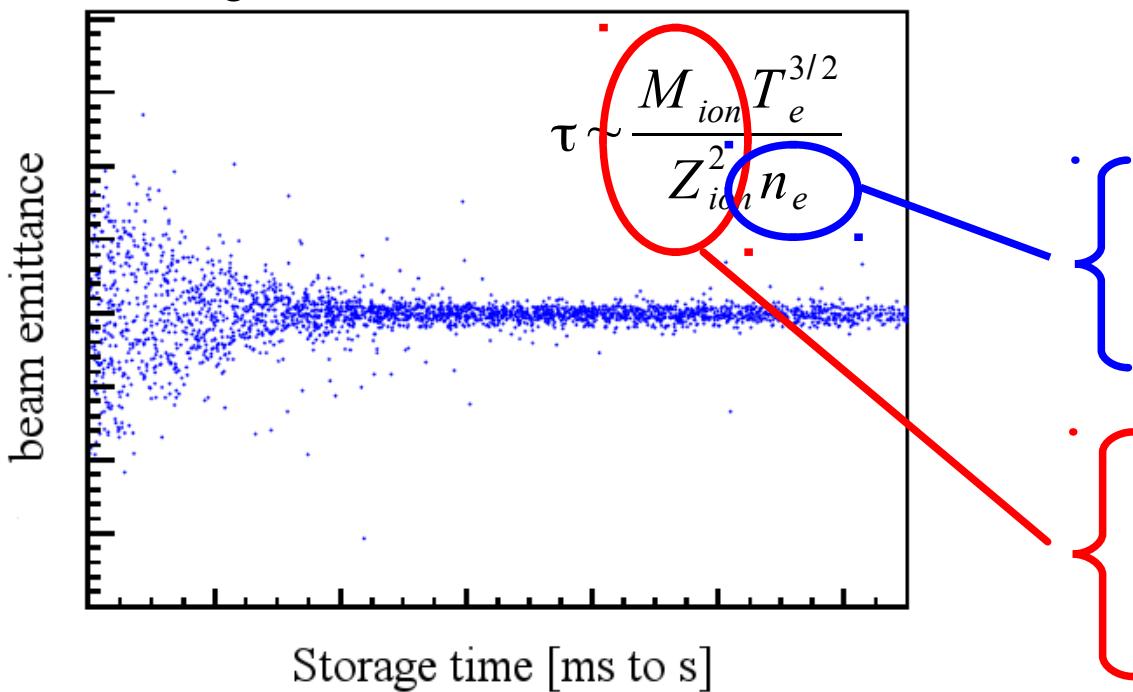
For molecular ions:  
 $M_{ion} \gg 10 \text{ u}$   
 $Z_{ion} = 1$

# Cooling force



$$\vec{F} \sim -\frac{Z_{ion}^2 n_e}{v^2} \frac{\vec{v}}{|v|}$$

Cooling time

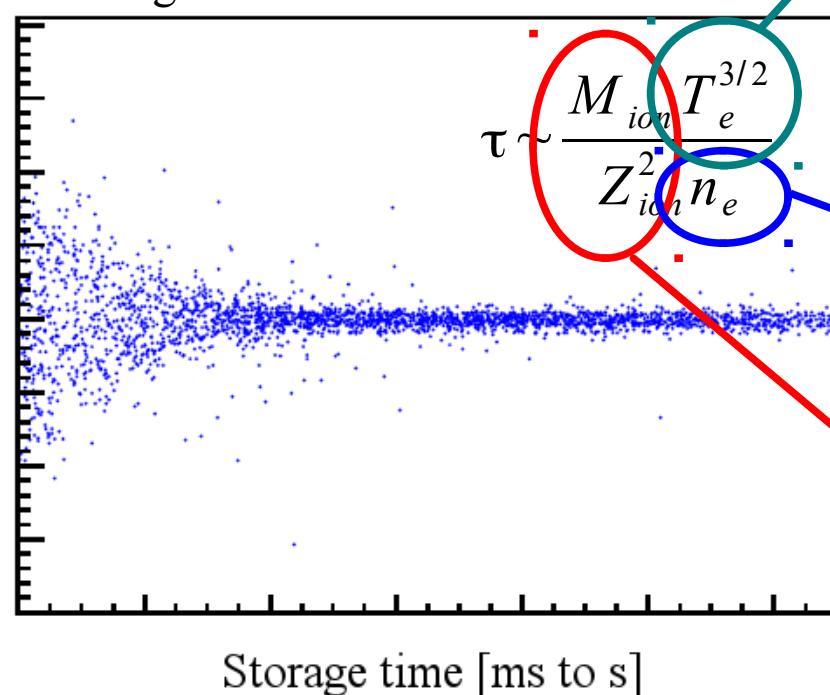
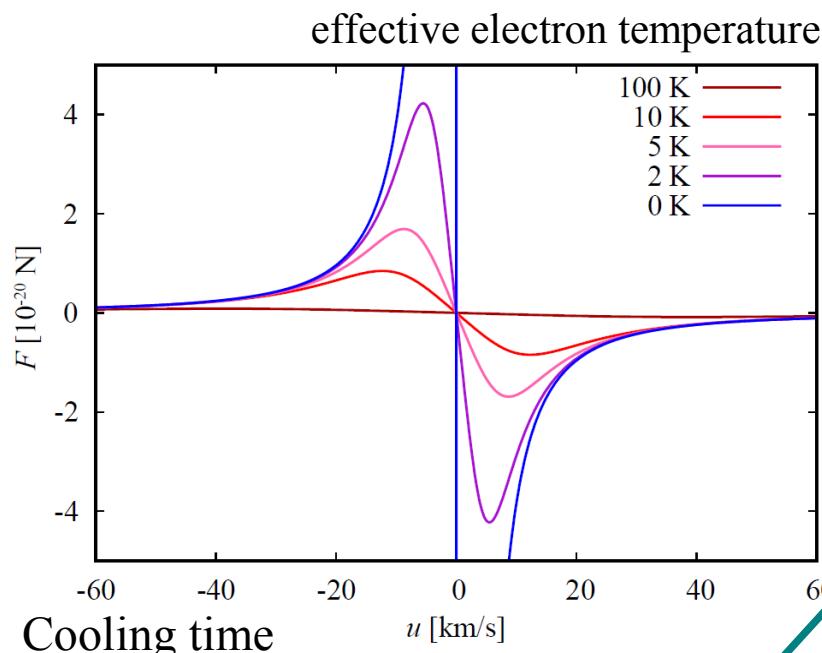


High beam **rigidity** (low  $v_{ion}$ ):  
 → low electron energy ( $\sim U_e$ )  
 → low  $n_e$  ( $I_{max} \sim U^{3/2}$ )

For **molecular** ions:  
 $M_{ion} \gg 10 \text{ u}$   
 $Z_{ion} = 1$



# Cooling force



Major impact on cooling force  
for transverse cooling (DR imaging!):

$$T_e \approx T_\perp$$

Improvement: Magnetic expansion

$$T_\perp \approx T_{\text{cath}} \frac{B_f}{B_i}$$

but: decreases also  $n_e$ !

Better: Start with low  $T_{\text{cath}}$  directly!

High beam rigidity (low  $v_{\text{ion}}$ ):

- low electron energy ( $\sim U_e$ )
- low  $n_e$  ( $I_{\text{max}} \sim U^{3/2}$ )

For molecular ions:

$$M_{\text{ion}} \gg 10 \text{ u}$$

$$Z_{\text{ion}} = 1$$



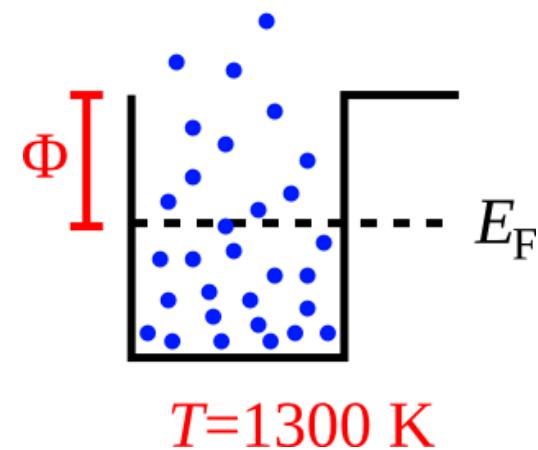
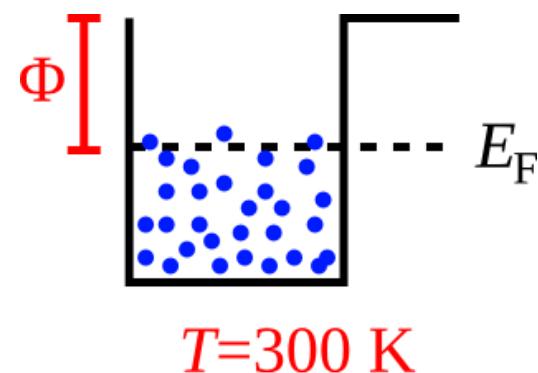
# Electron temperature

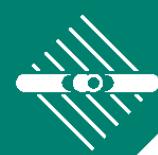
Thermionic cathodes:

$$J \sim T^2 \exp\left(\frac{-\Phi}{k_B T}\right)$$

established technology  
high  $J$  are possible ...

high electron- $T$   
( $k_B T > 100$  meV)





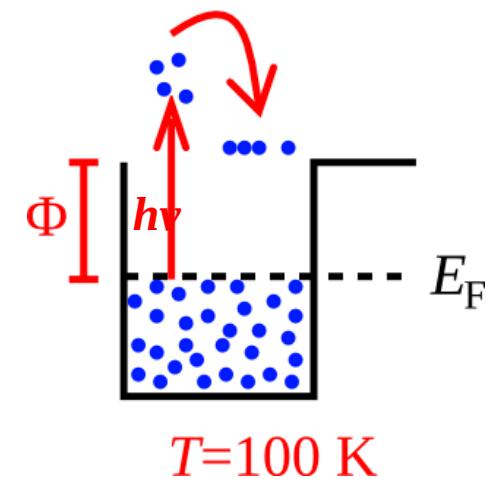
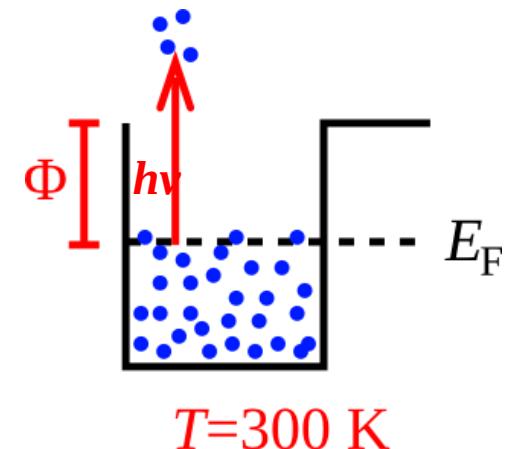
# Electron temperature

## Photocathodes:

- Electrons overcome  $\Phi$  by **absorbtion of photons** ( $h\nu > \Phi$ )
- Semiconductor *Negative Electron Affinity* (NEA) photocathodes: e's can **thermalise to states close to vacuum energy**.

$$T_e \sim T_{\text{cath}}$$

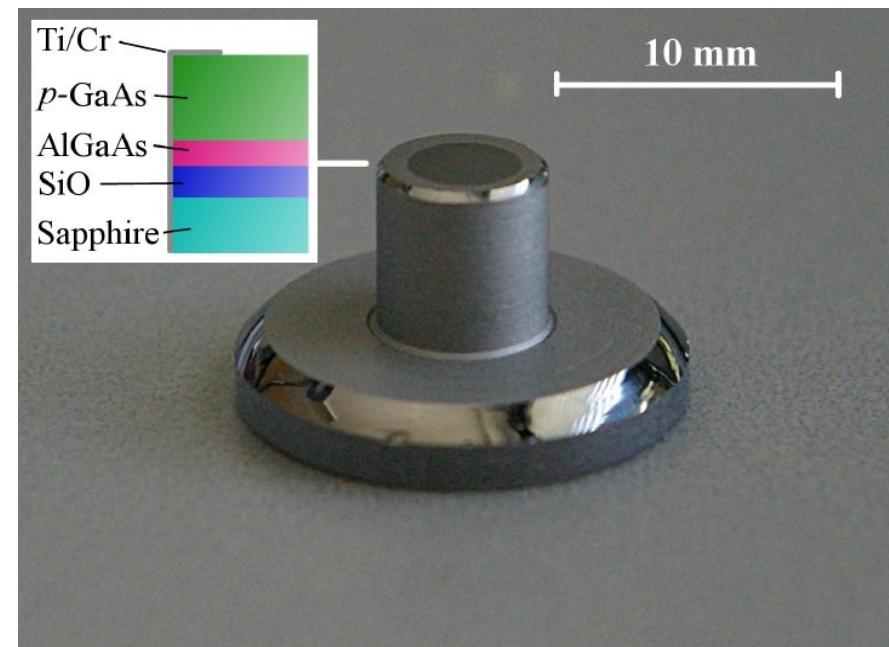
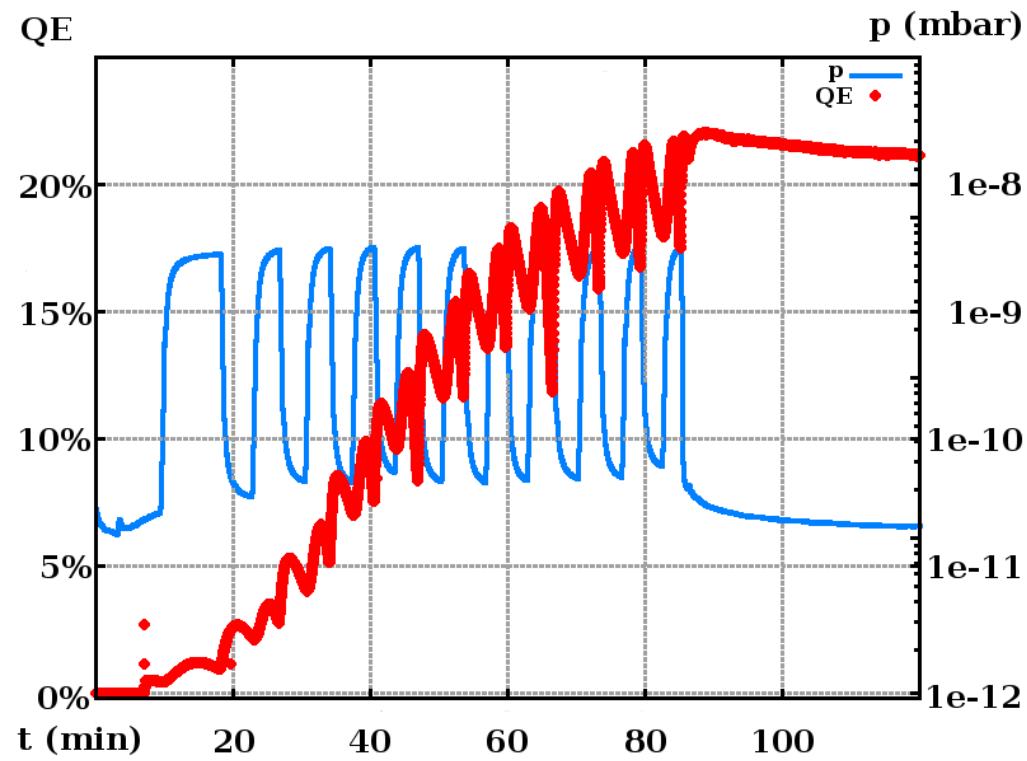
$$(k_B T \approx 10 \dots 24 \text{ meV})$$





# GaAs photo cathodes

NEA-activation by exposure  
to Cs and O<sub>2</sub>.

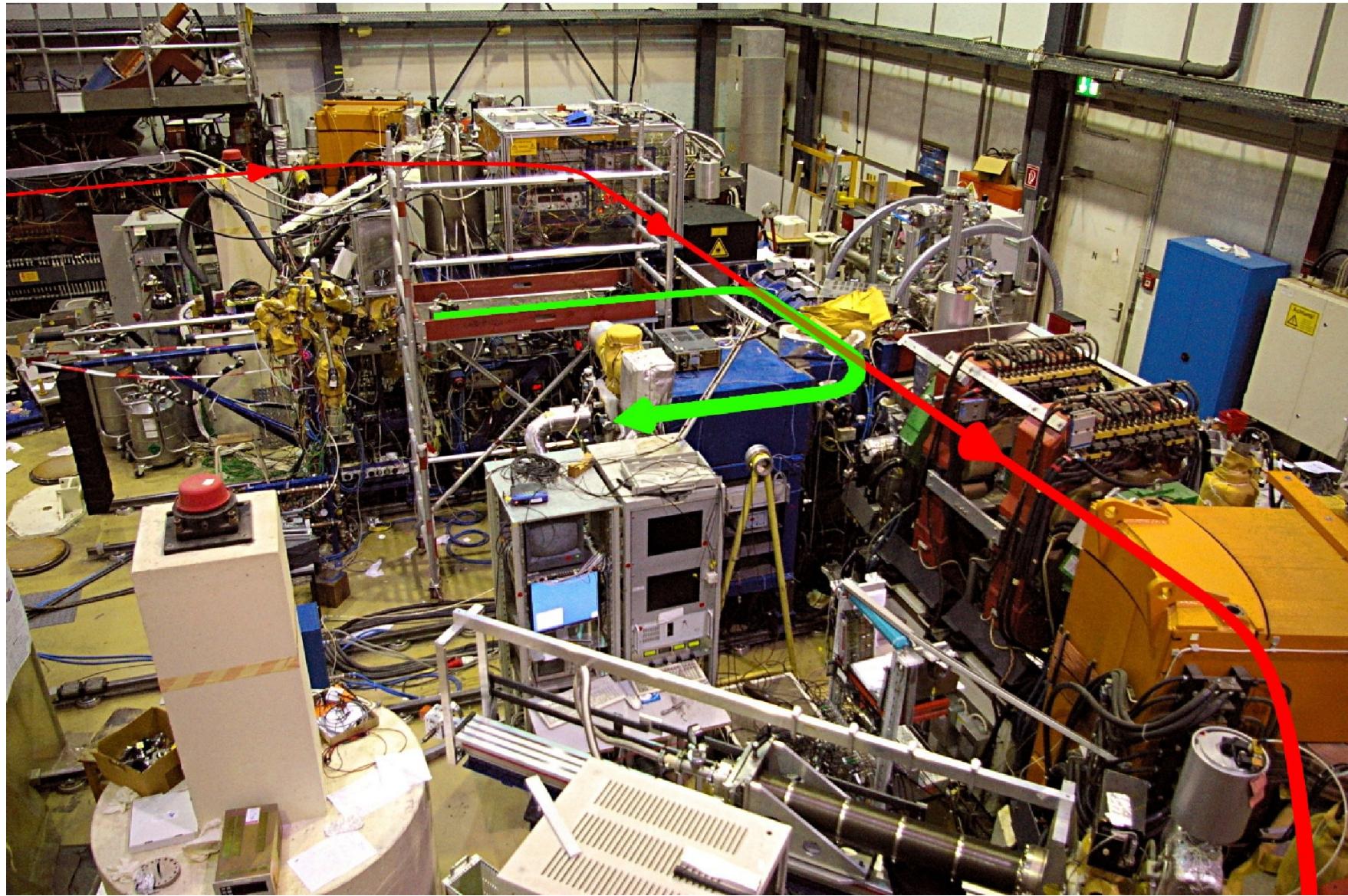


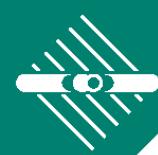
[A. S. Terekhov *et al.*, ISP Novosibirsk]

$k_B T_e = 10 \dots 24 \text{ meV}$   
 $I_e \leq 1 \text{ mA}$   
 $Q_{\max} \sim 100 \text{ C}$

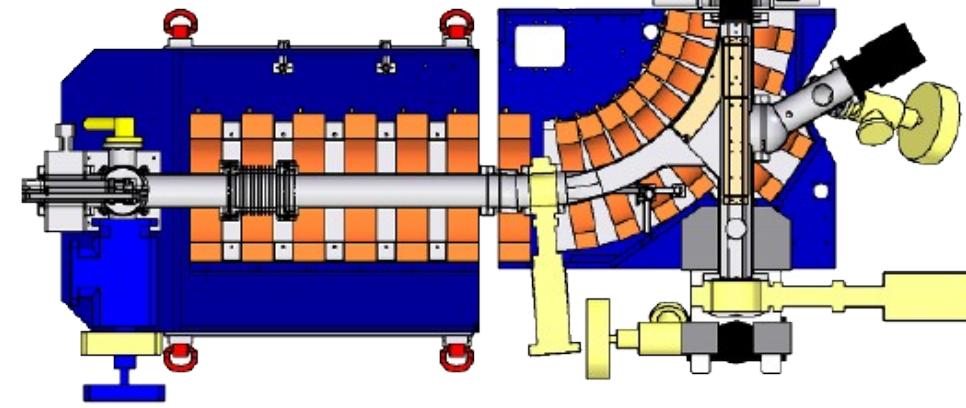
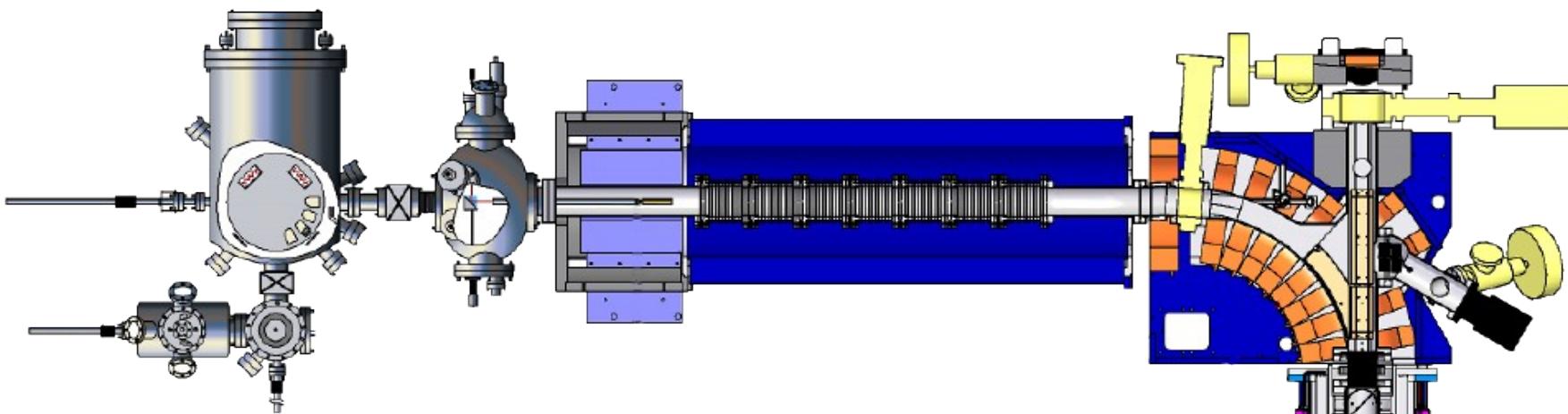


# Photo cathode electron cooler



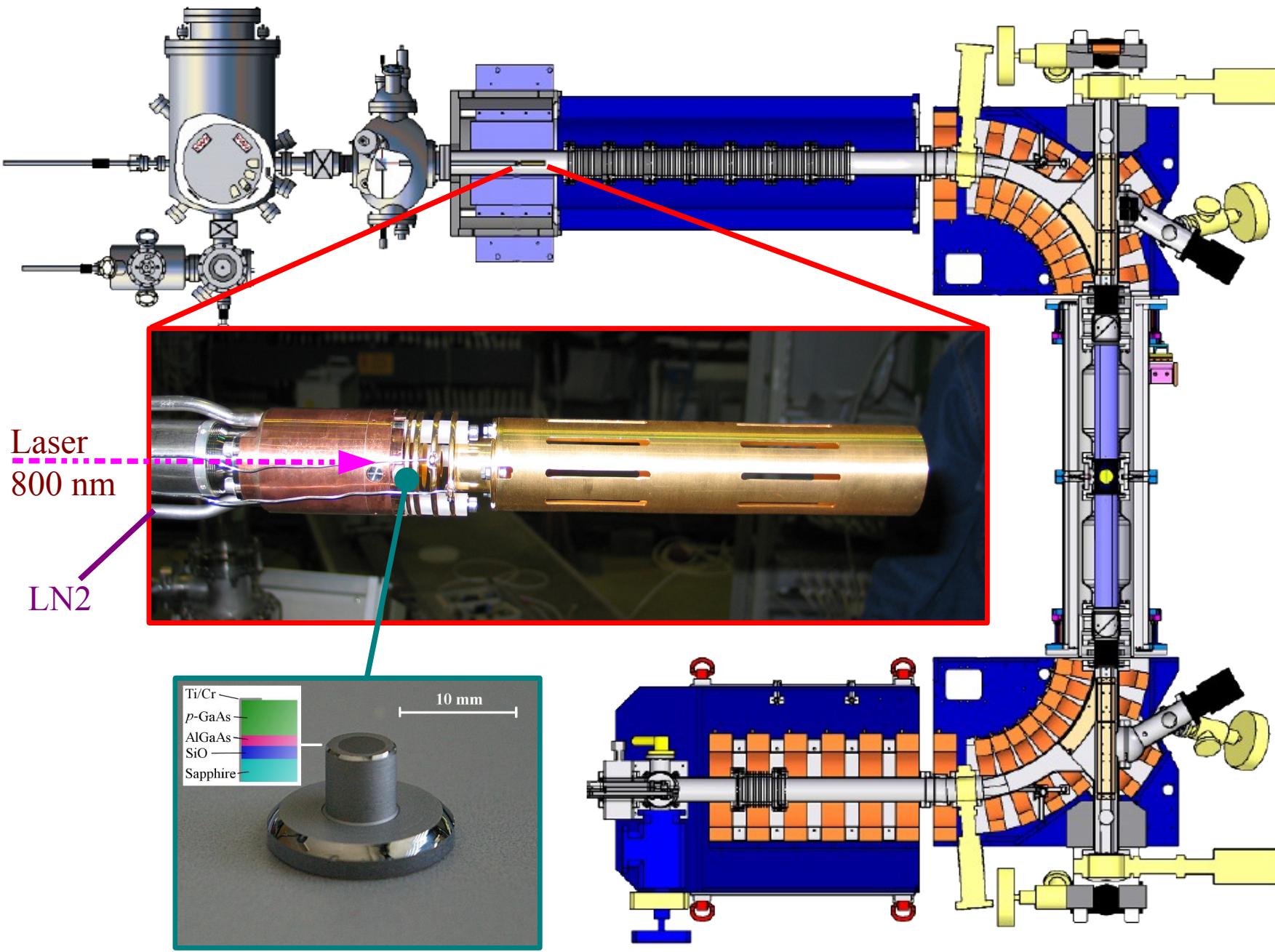


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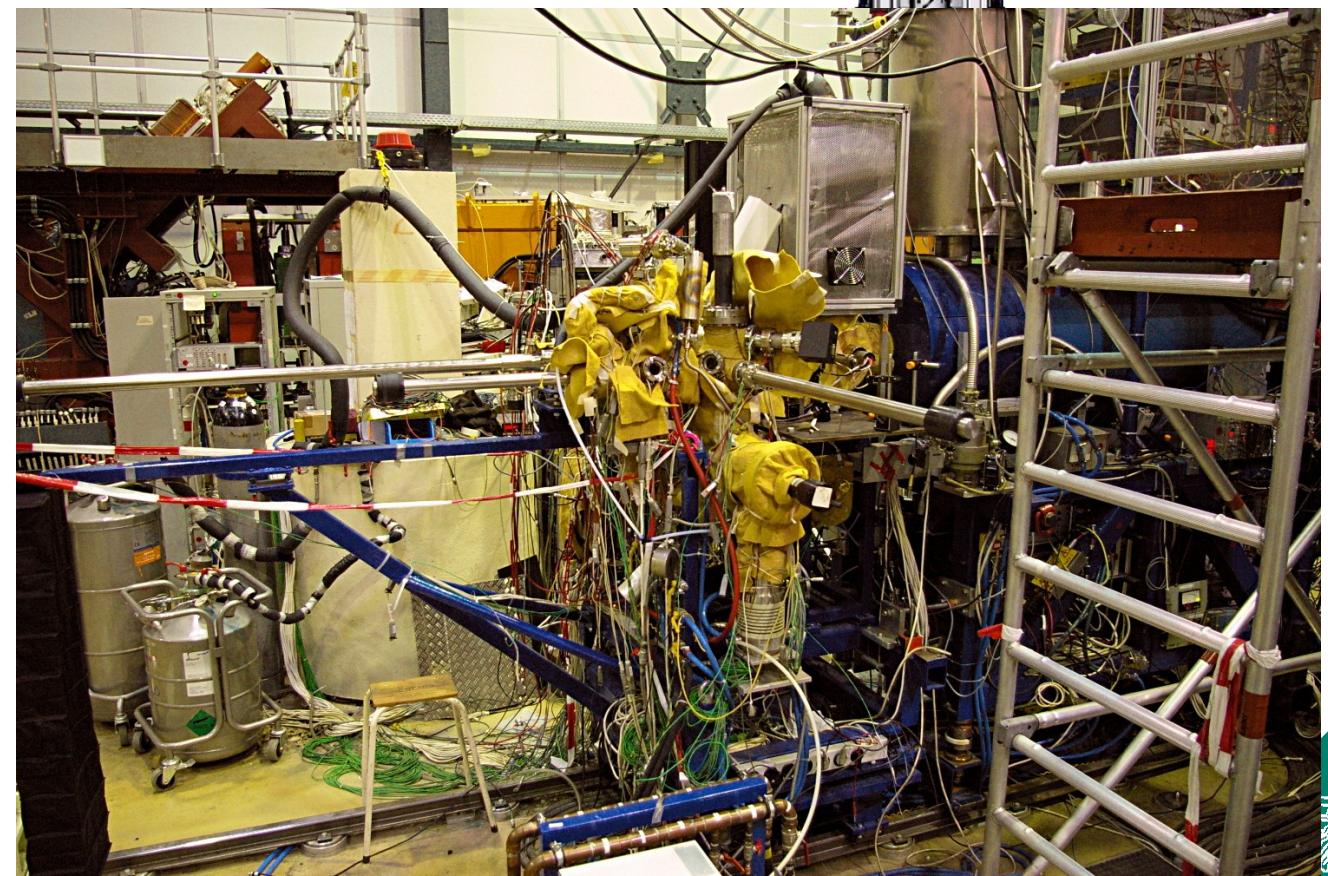
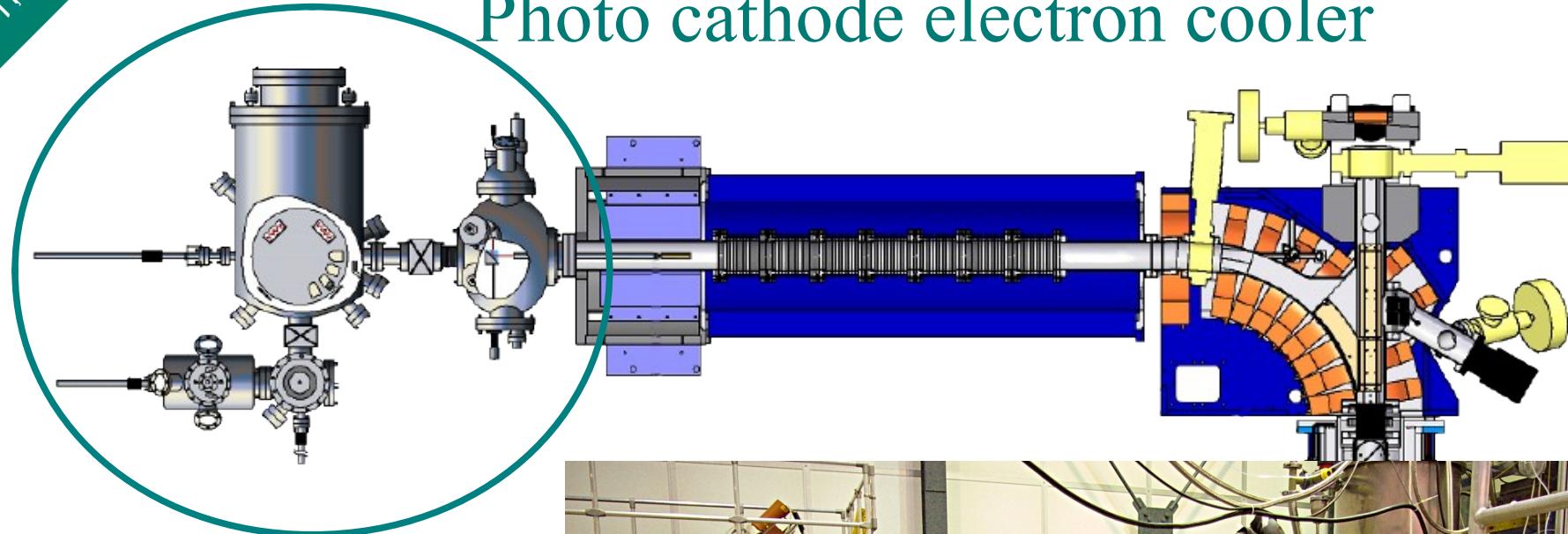


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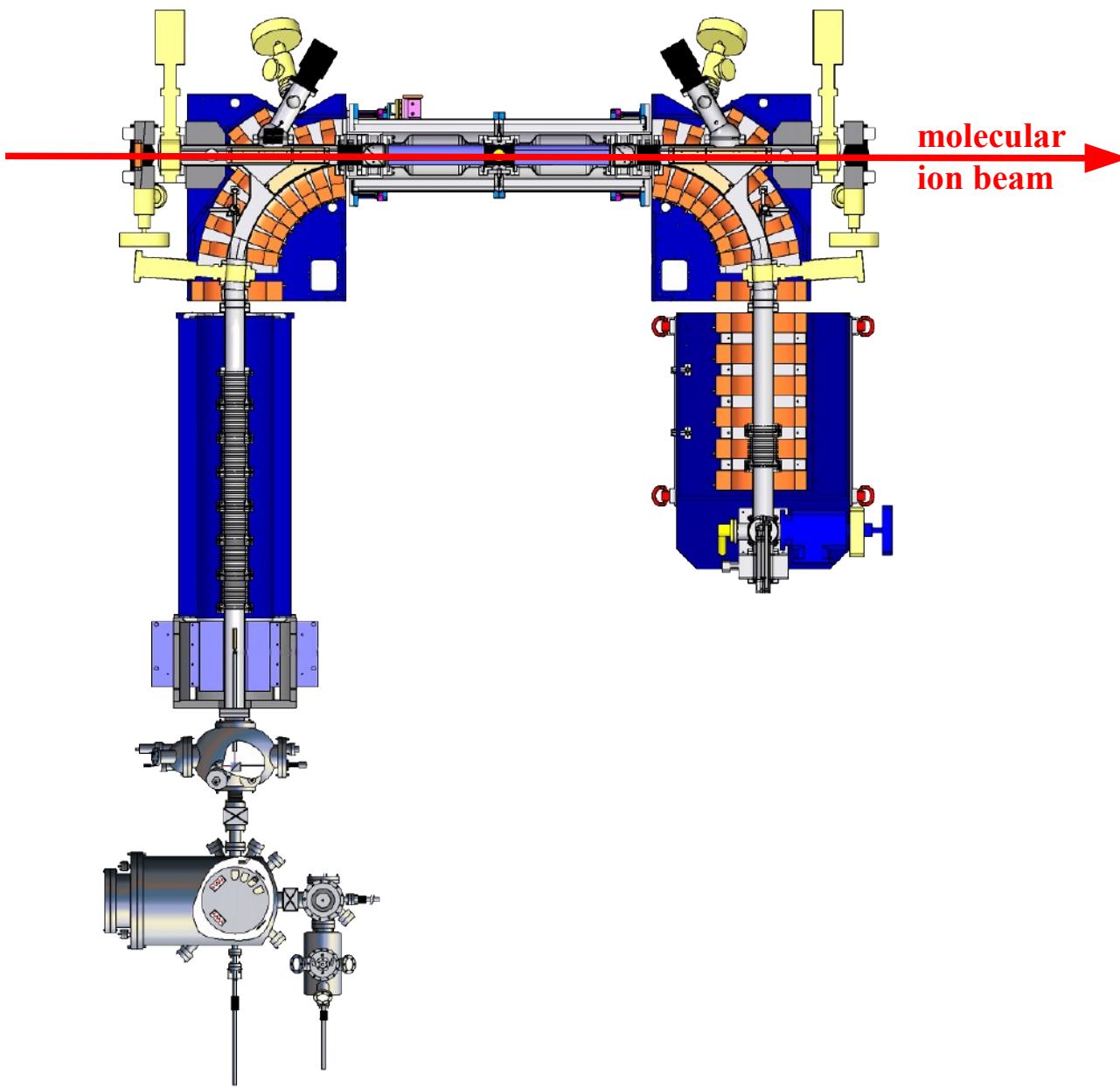


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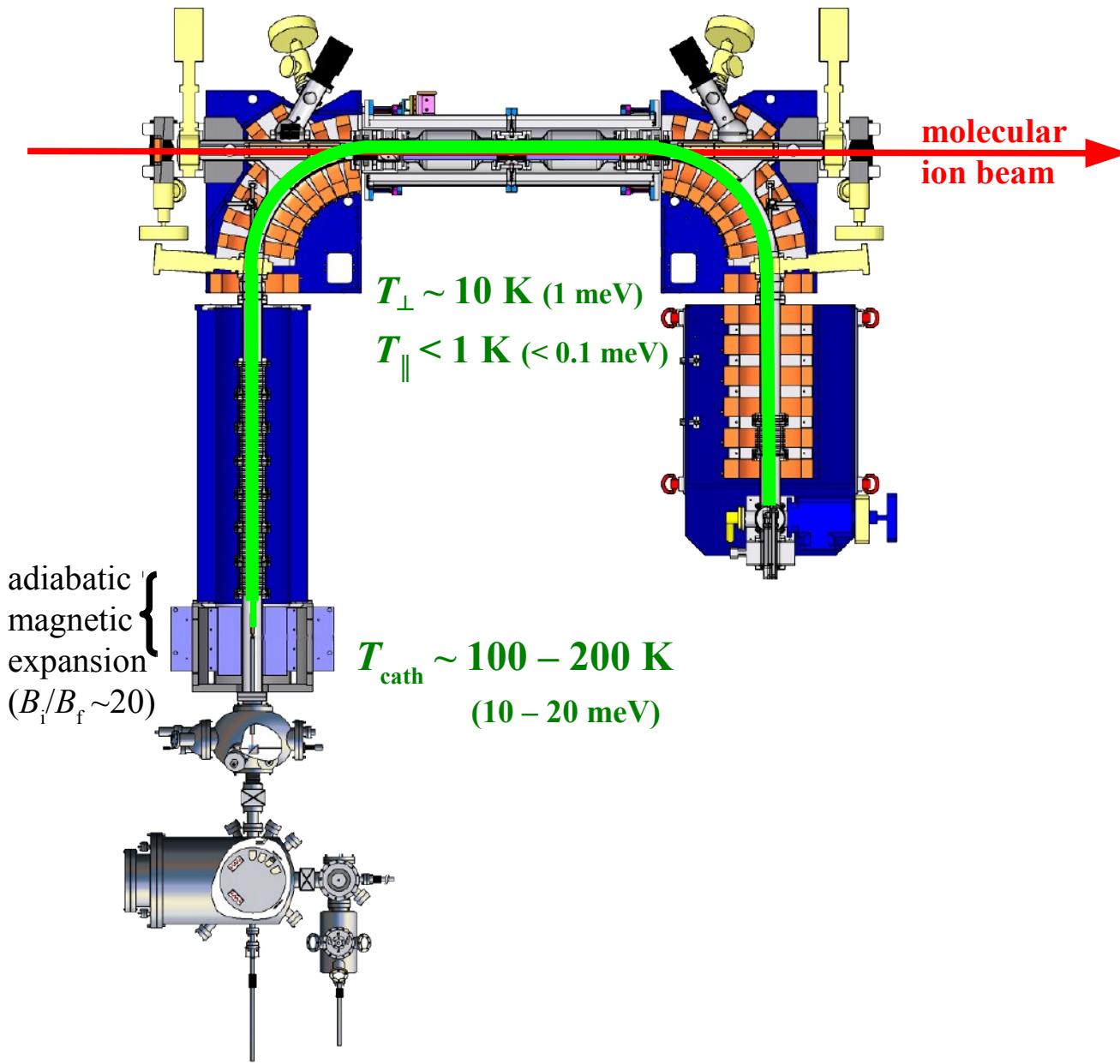


# Electron cooling at low velocity



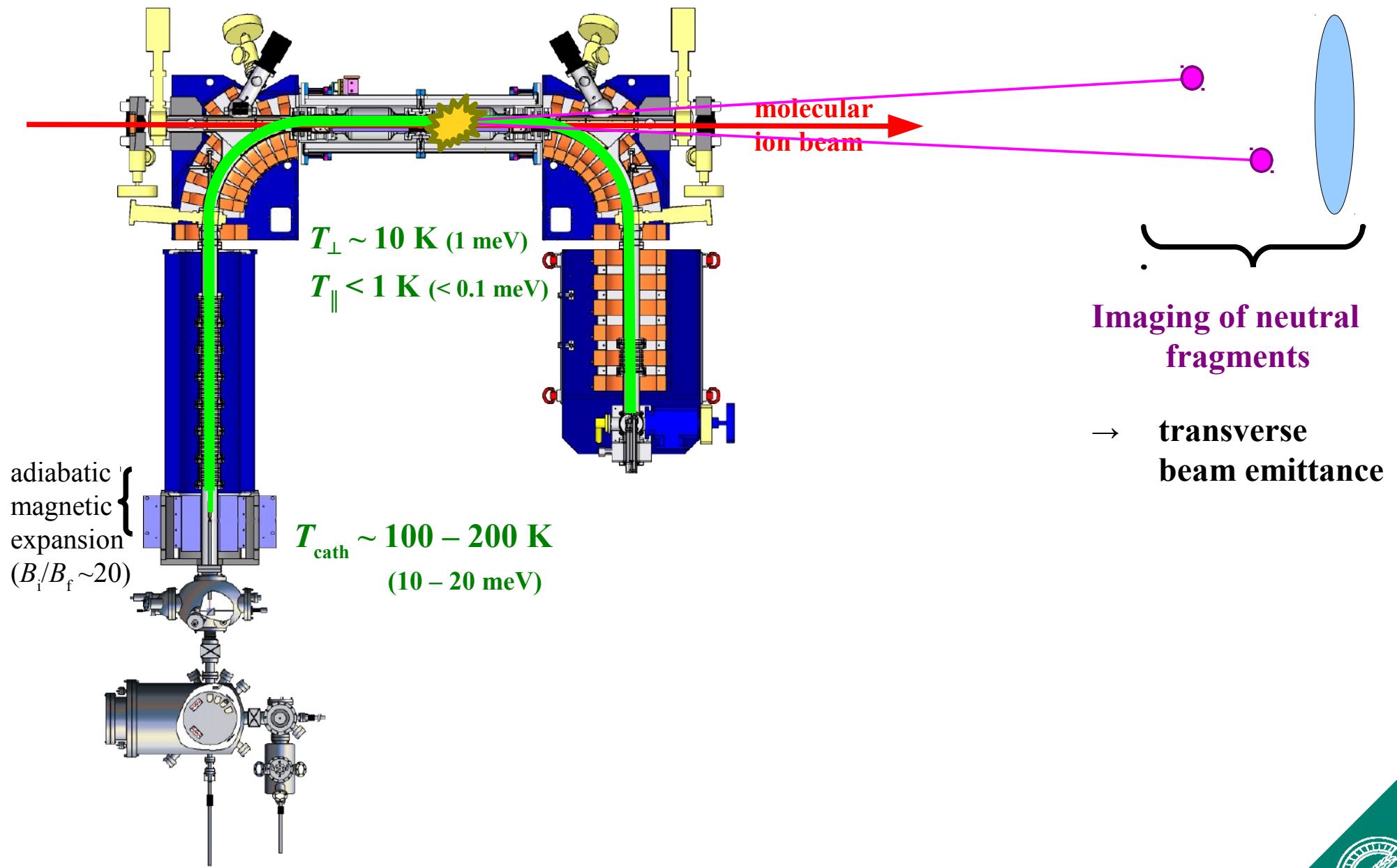


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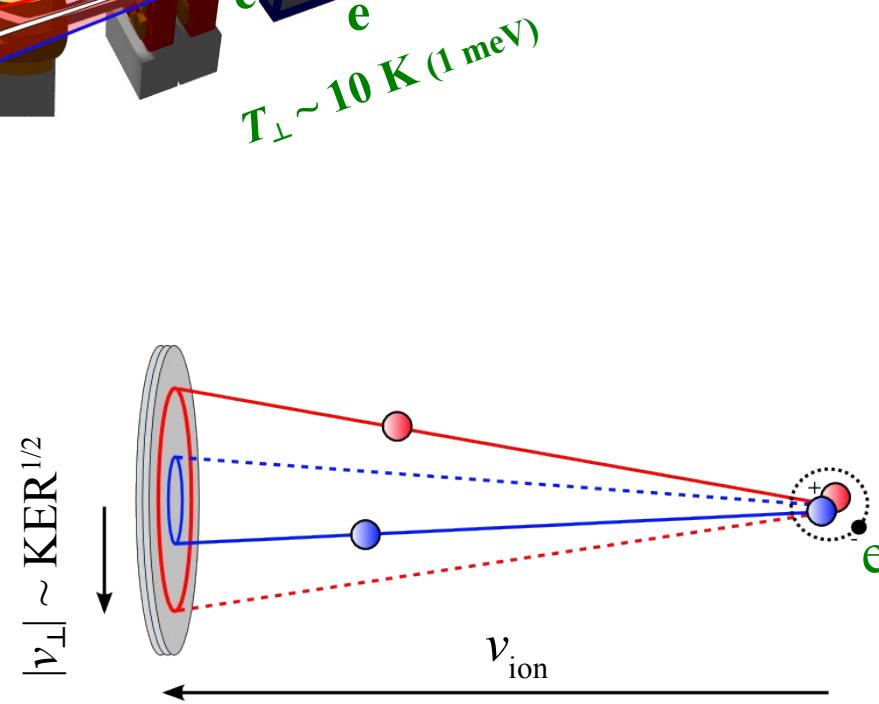
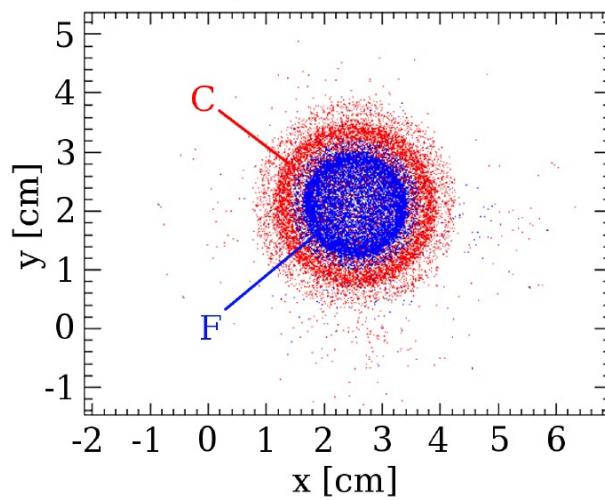
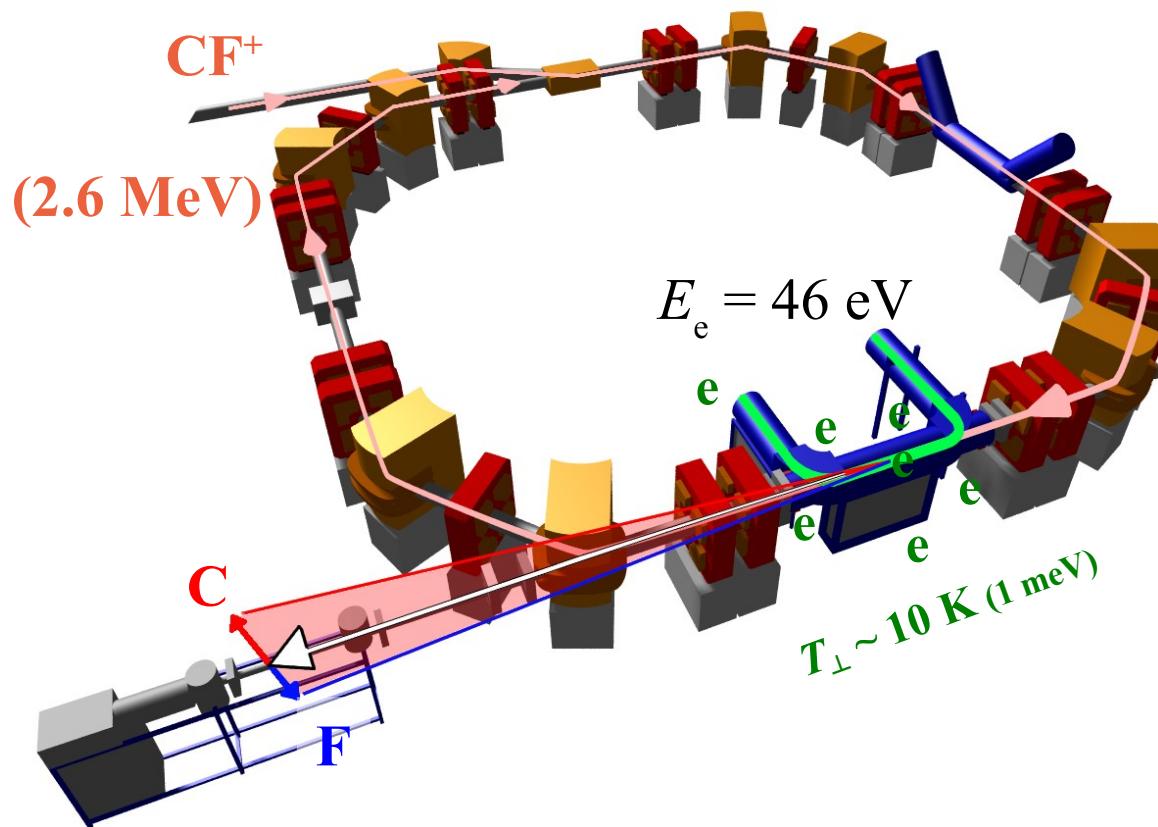


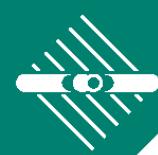
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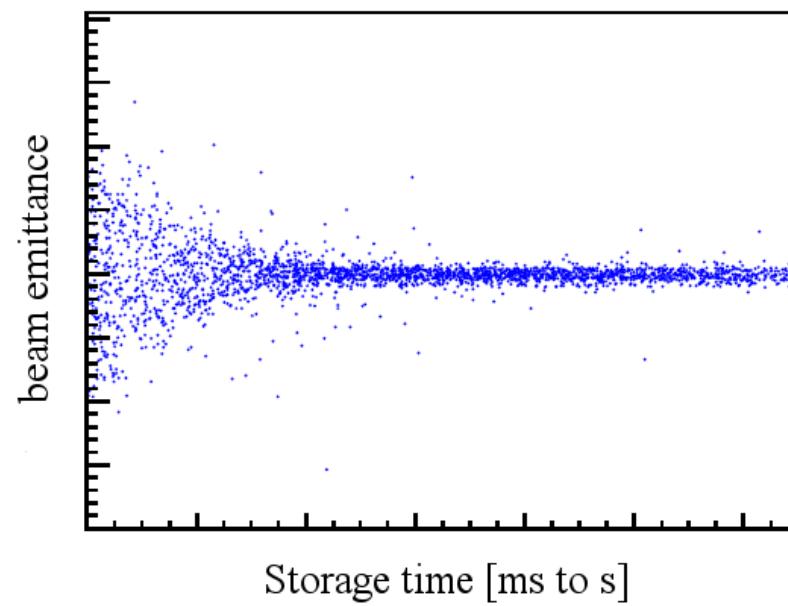
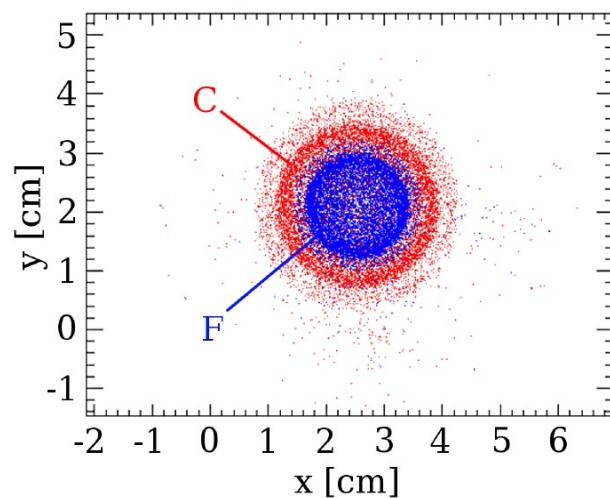
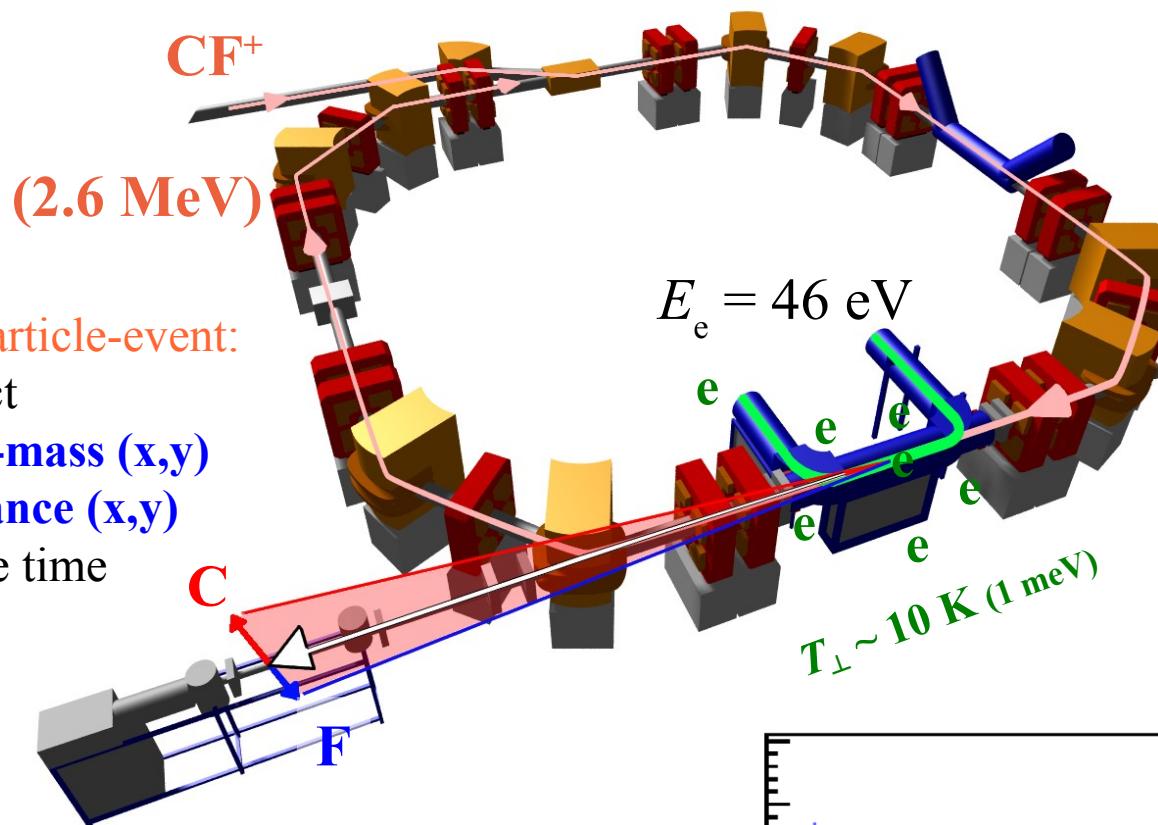
# Electron cooling at low velocity





# Electron cooling at low velocity

For each 2-particle-event:  
→ reconstruct  
**centre-of-mass (x,y)**  
→ get **emittance (x,y)**  
vs. storage time



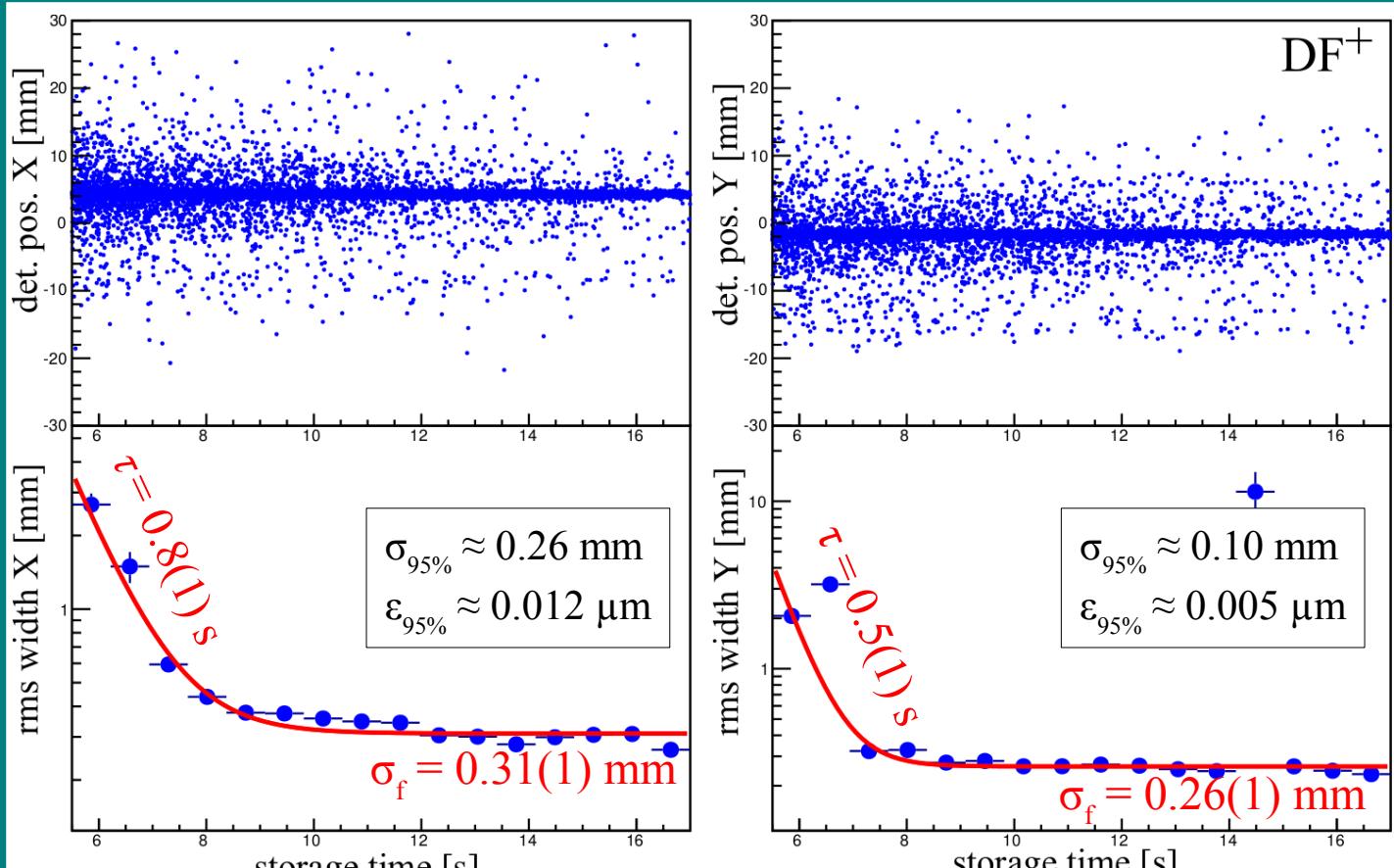


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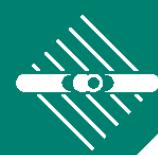
Ions	$M_{\text{ion}}$	$E_e(\text{cool.})$	$B\rho_{\text{TSR}}$ (Tm)
...			
$\text{H}_3^+$	3 u	735 eV	0.49
$\text{HD}_2^+$	5 u	327 eV	0.55
$\text{CHD}^+$	15 u	231 eV	1.37
$\text{HF}^+$	20 u	112 eV	1.28
$\text{DF}^+$	21 u	115 eV	1.31
$\text{D}_3\text{O}^+$	22 u	112 eV	1.36
$\text{DCND}^+$	30 u	56 eV	1.36
$\text{DCO}^+$	30 u	56 eV	1.36
$\text{N}_2\text{D}^+$	30 u	56 eV	1.36
$\text{CF}^+$	31 u	46 eV	1.27
$\text{HS}^+$	33 u	45 eV	1.33
$^{18}\text{O}^{16}\text{O}^+$	34 u	43 eV	1.34
$\text{H}^{35}\text{Cl}^+$	36 u	40 eV	1.32
$\text{D}_2^{35}\text{Cl}^+$	39 u	31 eV	1.31

# Electron cooling at low velocity

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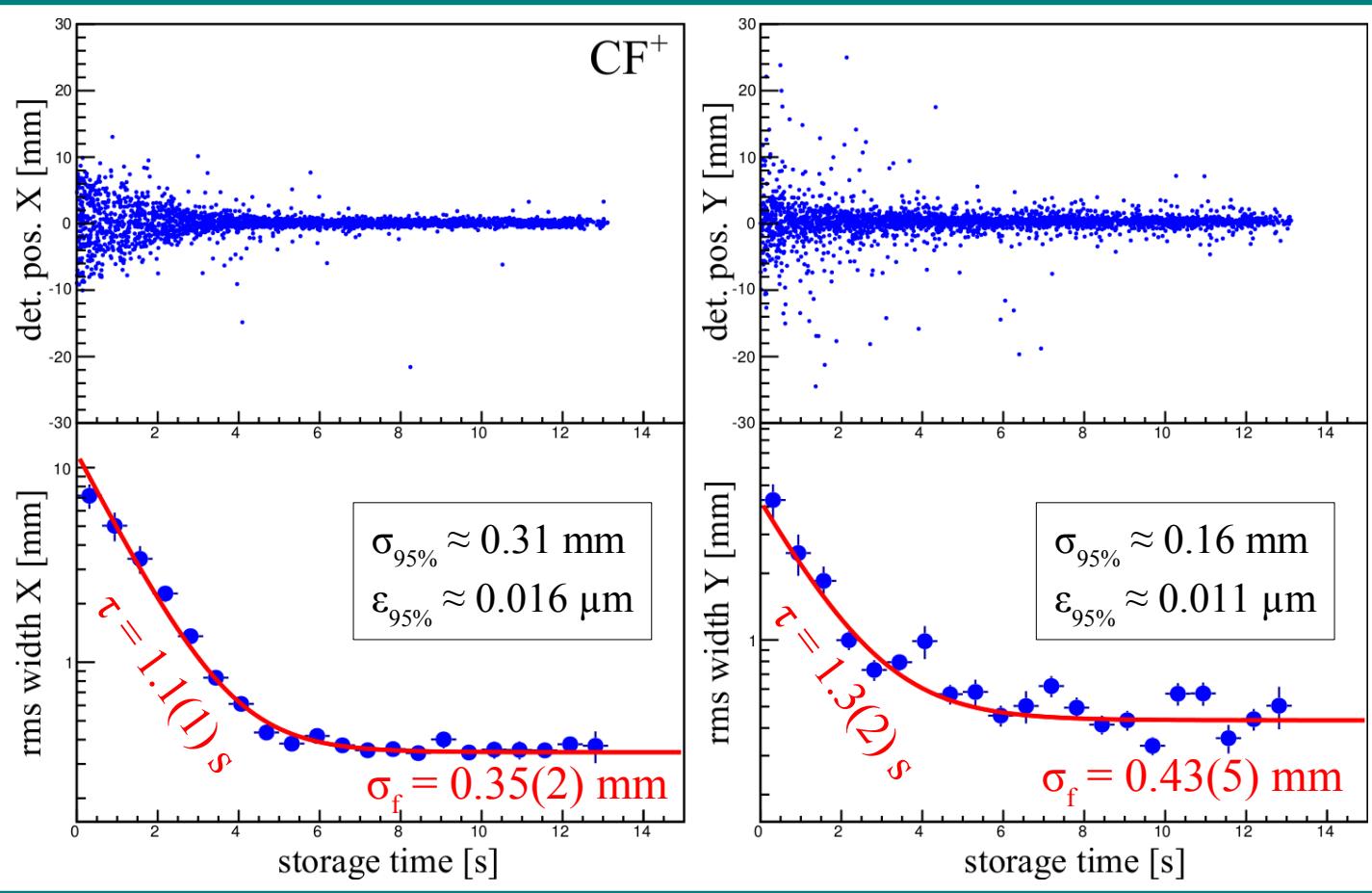


Preliminary!



# Electron cooling at low velocity

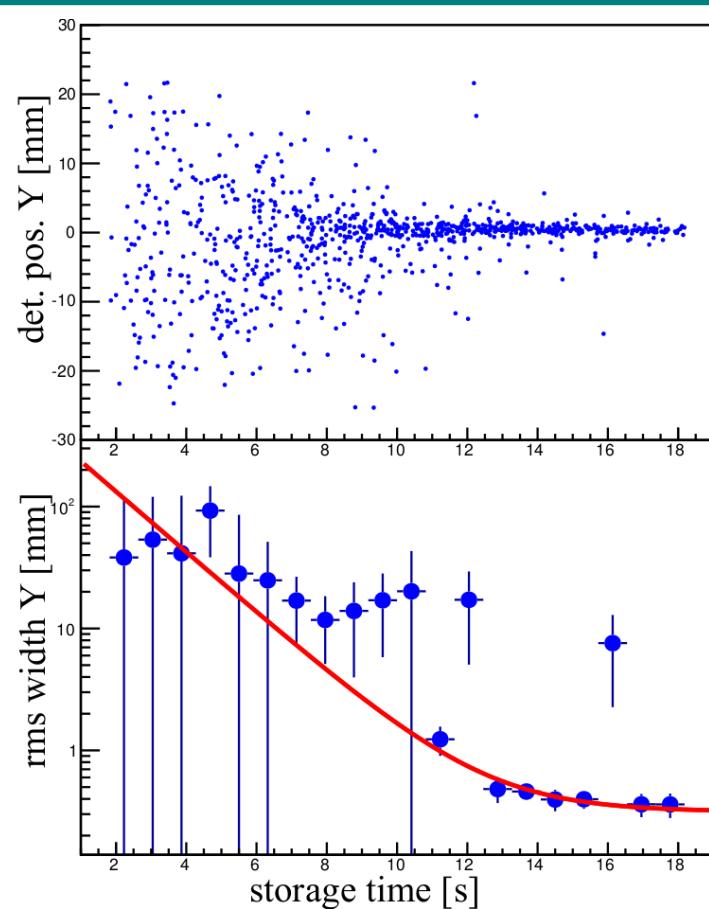
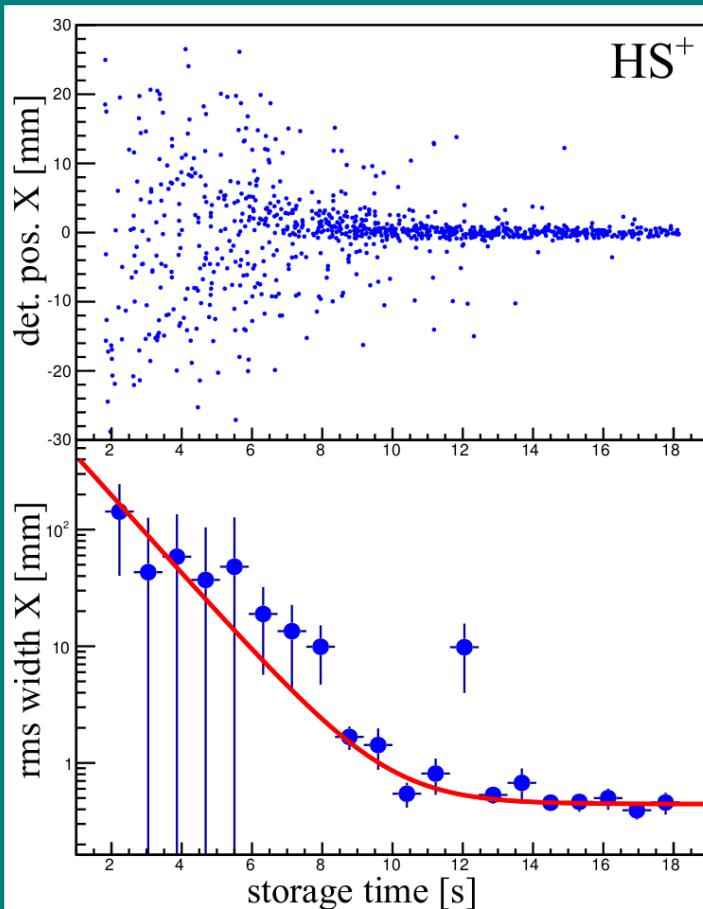
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$^{18}\text{O}^{16}\text{O}^+$	34 u	43 eV	
$\text{H}^{35}\text{Cl}^+$	36 u	40 eV	
$\text{D}_2^{35}\text{Cl}^+$	39 u	31 eV	



Preliminary!

# Electron cooling at low velocity

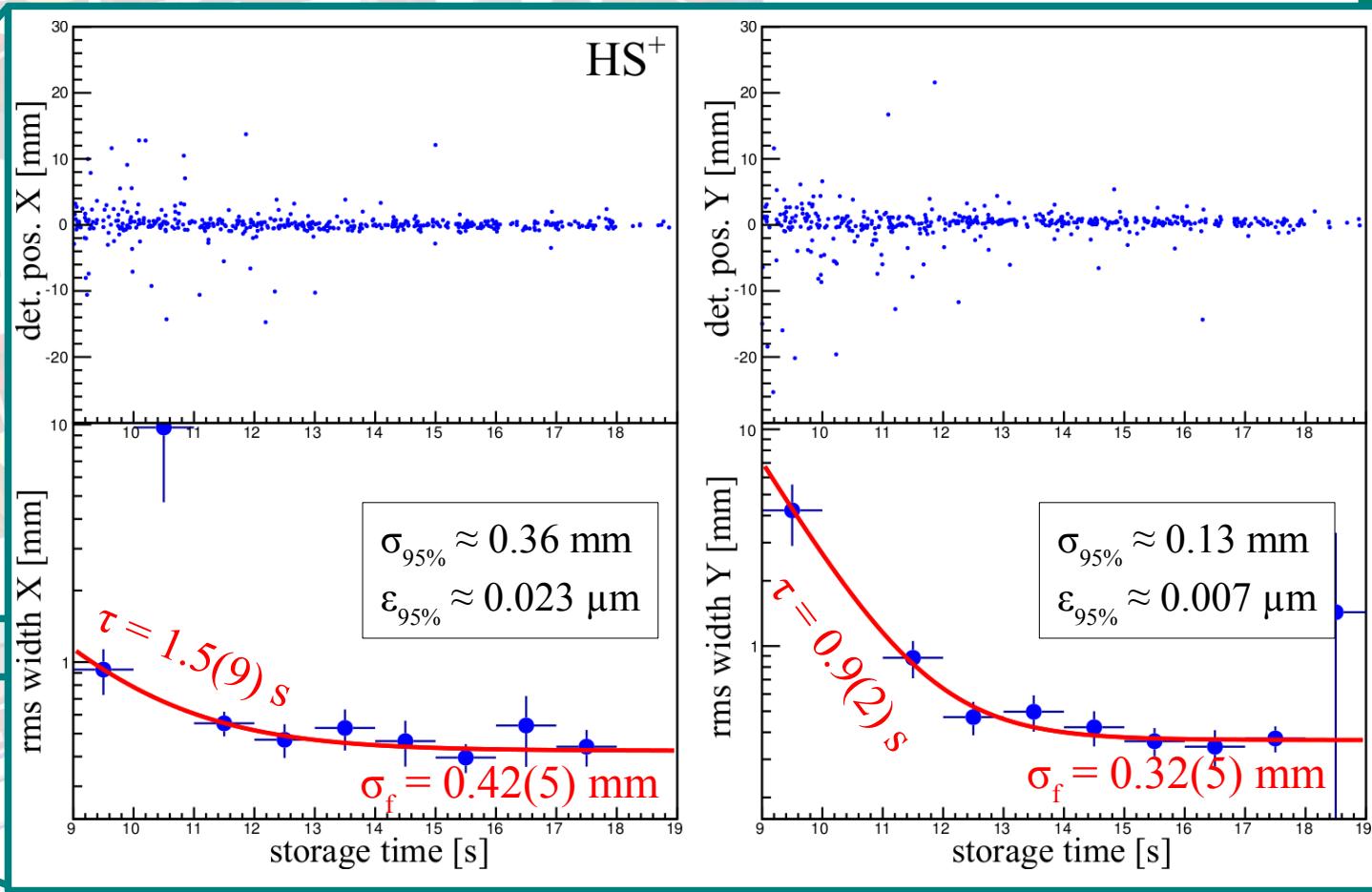
Ions	$M_{\text{ion}}$	$E_e$ (cool.)	$B\rho_{\text{TSR}}$ (Tm)
...			
$\text{H}_3^+$	3 u	735 eV	
$\text{HD}_2^+$	5 u	327 eV	
$\text{CHD}^+$	15 u	231 eV	
$\text{HF}^+$	20 u	112 eV	
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Preliminary!

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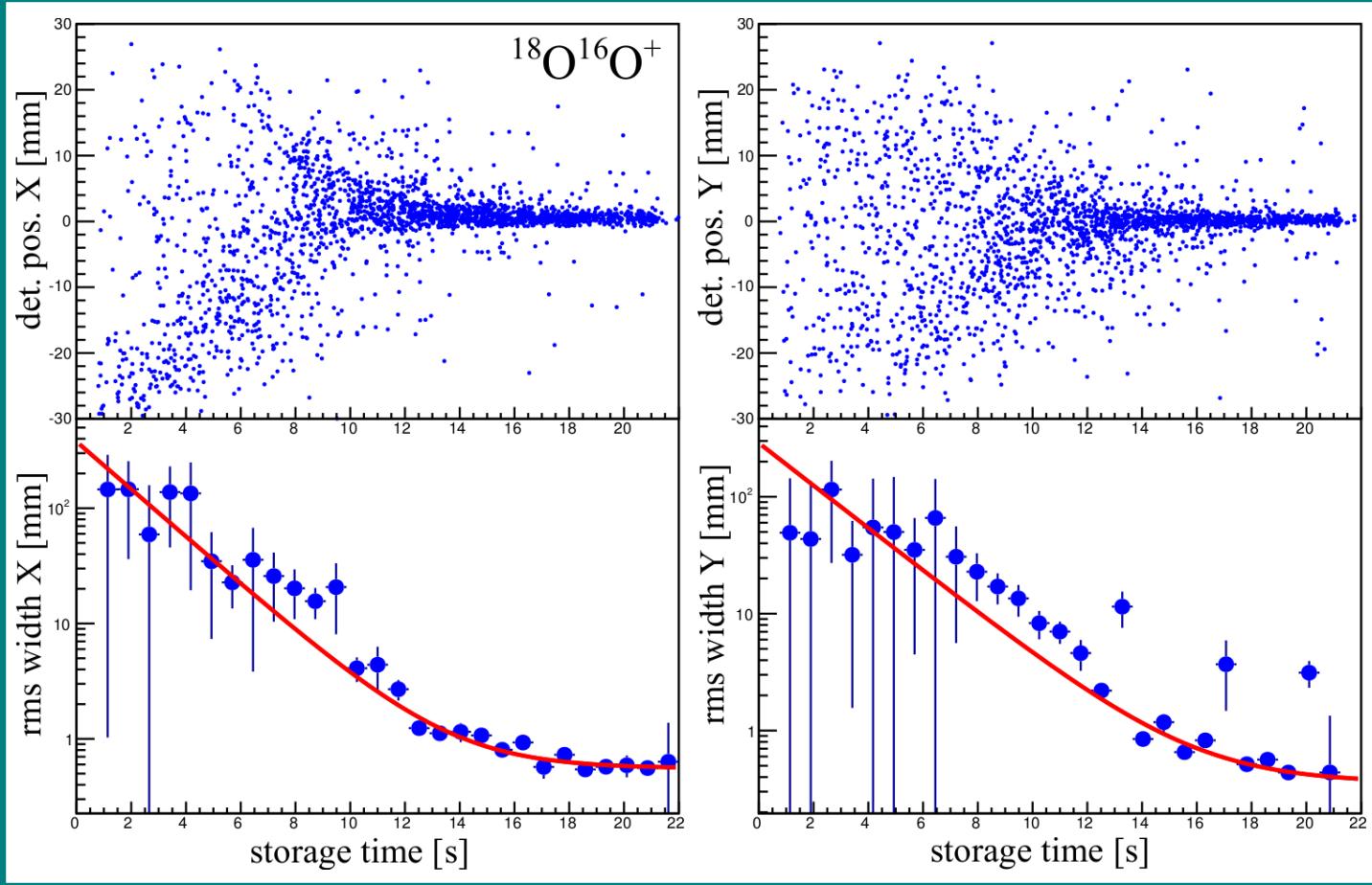
Preliminary!



# Electron cooling at low velocity

Ions       $M_{\text{ion}}$        $E_e(\text{cool.})$        $B\rho_{\text{TSR}} \text{ (Tm)}$

...			
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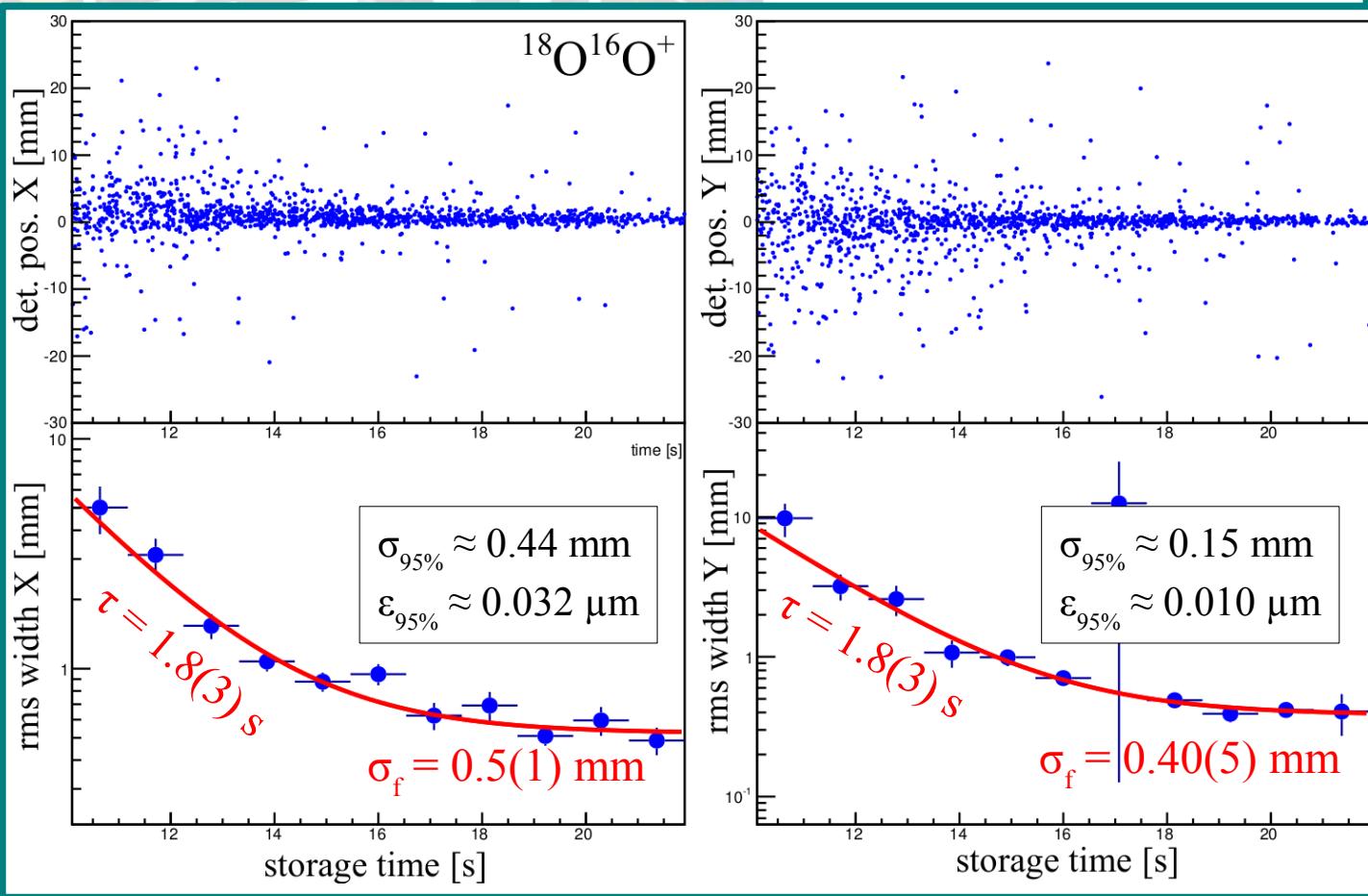
Preliminary!



# Electron cooling at low velocity

Ions	$M_{\text{ion}}$	$E_e(\text{cool.})$	$B\rho_{\text{TSR}} \text{ (Tm)}$
------	------------------	---------------------	-----------------------------------

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Preliminary!



# Electron cooling at low velocity

Ions	$M_{\text{ion}}$	$E_e(\text{cool.})$	$B\rho_{\text{TSR}}$ (Tm)	$I_e$ (mA)	$n_e$ (cm $^{-3}$ )	$\tau_{\text{cool}}$ (s)	“t <sub>cool</sub> ” (s)	$\varepsilon_{\text{fi}}$ ( $\mu\text{m}$ )
...								
H <sub>3</sub> <sup>+</sup>	3 u	735 eV	0.49					
HD <sub>2</sub> <sup>+</sup>	5 u	327 eV	0.55					
CHD <sup>+</sup>	15 u	231 eV	1.37					
HF <sup>+</sup>	20 u	112 eV	1.28	0.3	$2.1 \cdot 10^6$	1.4(1)	3	$\leq 0.008$
DF <sup>+</sup>	21 u	115 eV	1.31	0.3	$2.1 \cdot 10^6$	0.7(2)	3	$\leq 0.012$
D <sub>3</sub> O <sup>+</sup>	22 u	112 eV	1.36					
DCND <sup>+</sup>	30 u	56 eV	1.36					
DCO <sup>+</sup>	30 u	56 eV	1.36	0.3	$3.0 \cdot 10^6$		4	
N <sub>2</sub> D <sup>+</sup>	30 u	56 eV	1.36	0.2	$2.0 \cdot 10^6$		8	
CF <sup>+</sup>	31 u	46 eV	1.27	0.27	$3.0 \cdot 10^6$	1.2(2)	4	$\leq 0.016$
HS <sup>+</sup>	33 u	45 eV	1.33	0.24	$2.7 \cdot 10^6$	1.4(3)	10	$\leq 0.023$
<sup>18</sup> O <sup>16</sup> O <sup>+</sup>	34 u	43 eV	1.34	0.22	$2.5 \cdot 10^6$	1.8(3)	8	$\leq 0.032$
H <sup>35</sup> Cl <sup>+</sup>	36 u	40 eV	1.32	0.22	$2.6 \cdot 10^6$		12	
D <sub>2</sub> <sup>35</sup> Cl <sup>+</sup>	39 u	31 eV	1.31	0.2	$2.7 \cdot 10^6$		10	

Preliminary!



# Electron cooling at low velocity

Ions	$M_{\text{ion}}$	$E_e$	$n_e (\text{cm}^{-3})$	$\tau_{\text{cool}} (\text{s})$	“ $t_{\text{cold}}$ ” (s)	$\varepsilon_{\text{fi}} (\mu\text{m})$
...						
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$\text{HD}_2^+$	5 u	32 eV				
$\text{CHD}^+$	15 u	231 eV	1.37			
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We are able to **cool** even the **heaviest** (slowest) molecular ions within  $\sim 10$  s.

Preliminary!



# Electron cooling at low velocity

Ions	$M_{\text{ion}}$	$E_e(\text{co})$	$n_e(\text{cm}^{-3})$	$\tau_{\text{cool}}(\text{s})$	“ $t_{\text{cold}}$ ” (s)	$\varepsilon_{\text{fi}}(\mu\text{m})$
...						
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**Cooling rates agree with  $T_e = 10 \dots 20 \text{ K}$   
(lots of uncertainties ... )**

Preliminary!

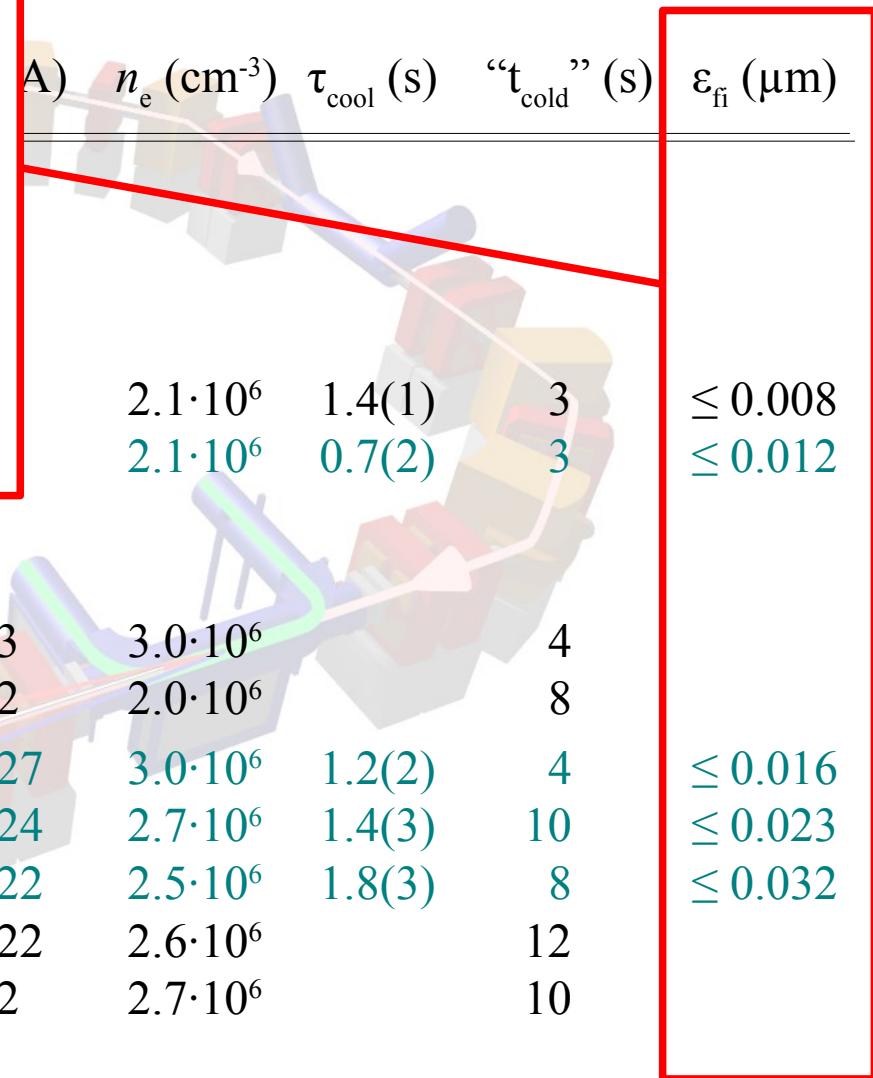


# Electron cooling at low velocity

Ions	$M_{\text{ion}}$	$E_e$ (eV)	Transverse momentum spread corresponds to ~ 40 ... 70 K
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Transverse momentum spread corresponds to  
~ 40 ... 70 K

(roughly what one expects from IBS modeling at given  $\tau_{\text{cool}}$ .)



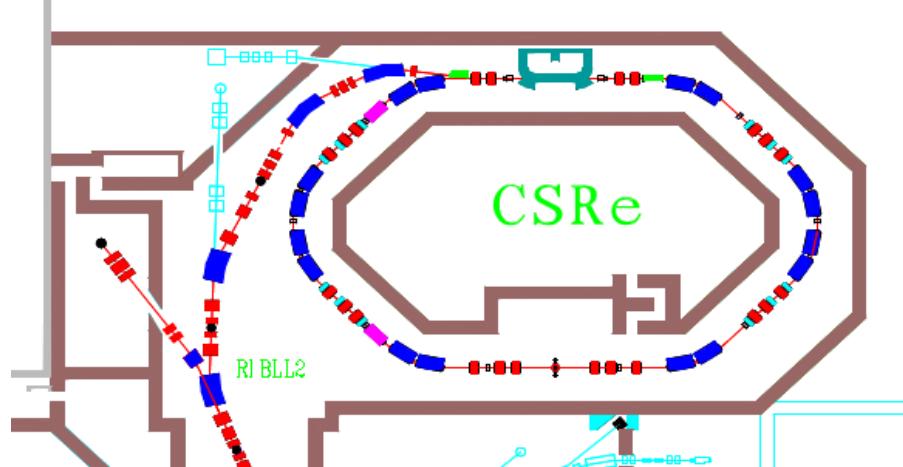
Preliminary!



# How to reach even higher mass?

Solution 1: Use a bigger storage ring

CSRe (“Cooler Storage Ring”, IMP)  
**9.4 Tm**



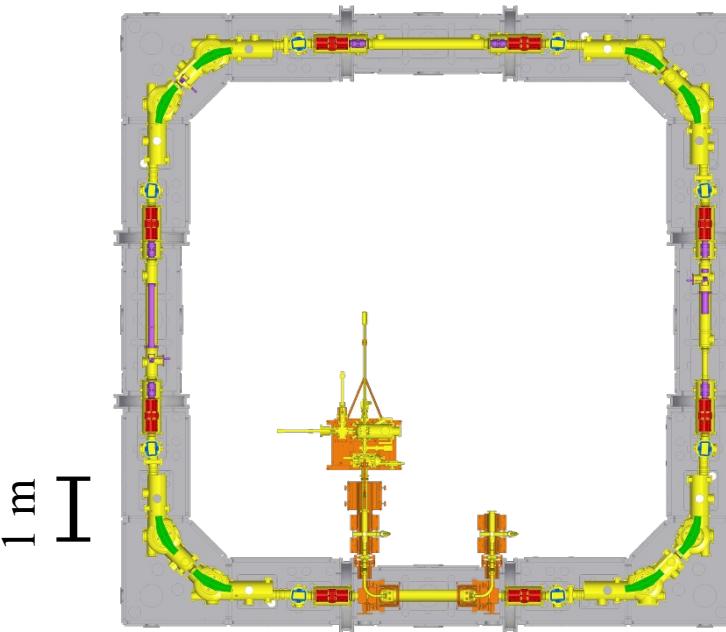
Molecular ions research facility,  
Lanzhou, China.

$$\rightarrow M_{\text{ion}}/Z_{\text{ion}} \sim 200$$

Solution 2: Store (much!) longer

CSR (“Cryogenic Storage Ring”, MPIK)

beam line at  $\sim 10$  K  
 $\rightarrow 10^{-13}$  mbar  
 $\rightarrow 100 \times$  longer ion lifetimes



$$M_{\text{ion}}/Z_{\text{ion}} \geq 160 \text{ (with e-cooling!)}$$

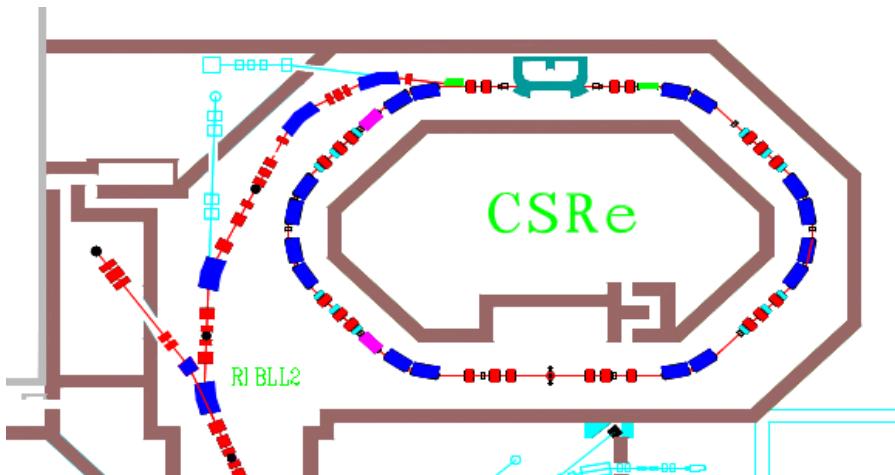
**Added value: IR-radiation-free!**



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$$M_{\text{ion}}/Z_{\text{ion}} \geq 160 \text{ (with e-cooling!)}$$

**Added value: IR-radiation-free!**



# CSR Electron Cooler

**Technically challenging:** Cooler must be contained in the **CSR cryostat**  
(bakeable,  $10^{-13}$  mbar, 10 K)

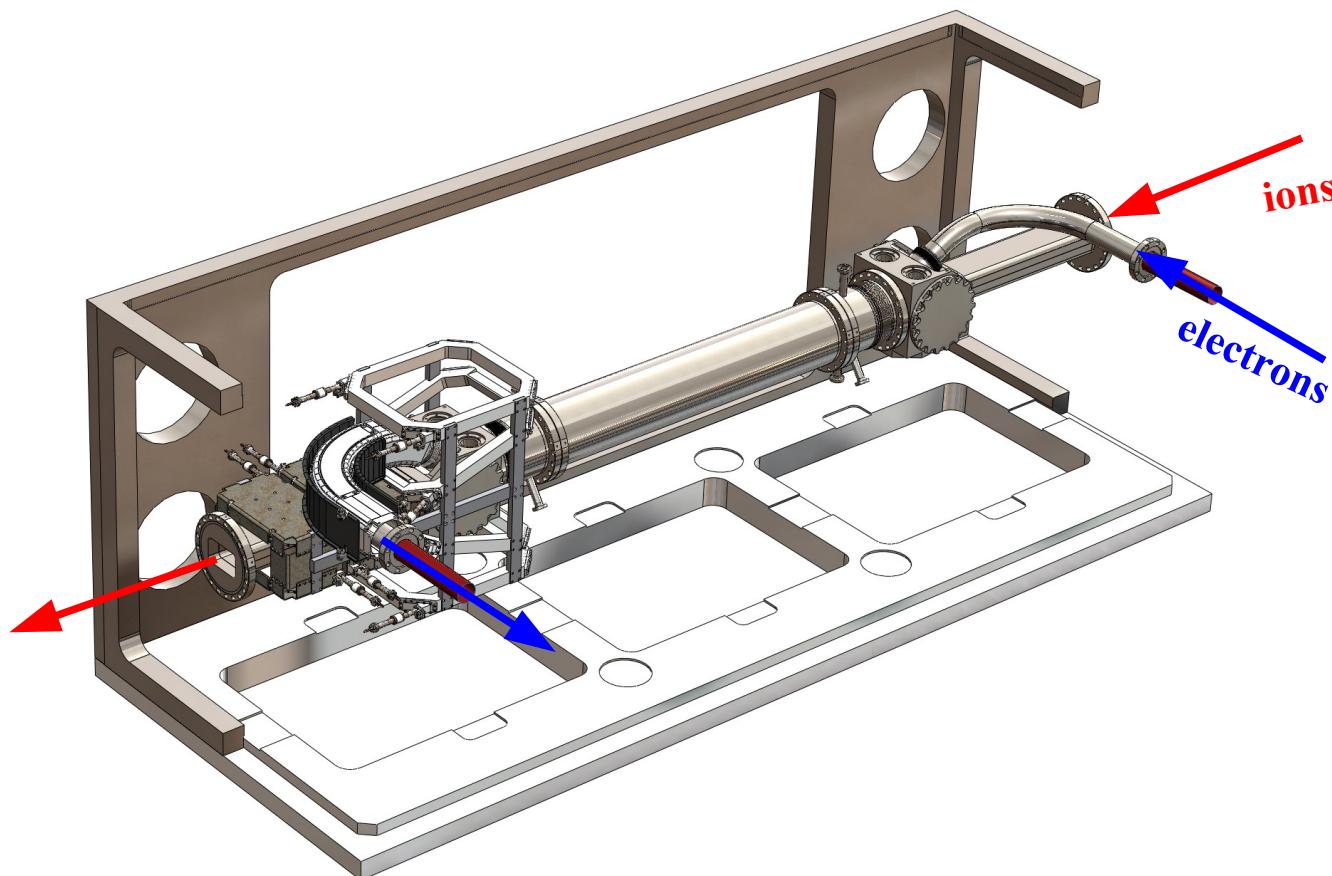
**CSR energy limit:** → Need **very slow** electrons

$$E_{\text{ion}}/Z_{\text{ion}} = 300 \text{ keV}$$

160 eV for  $p^+$

< 20 eV for most mol. ions

1 eV for  $M_{\text{ion}} = 160 \text{ u}$

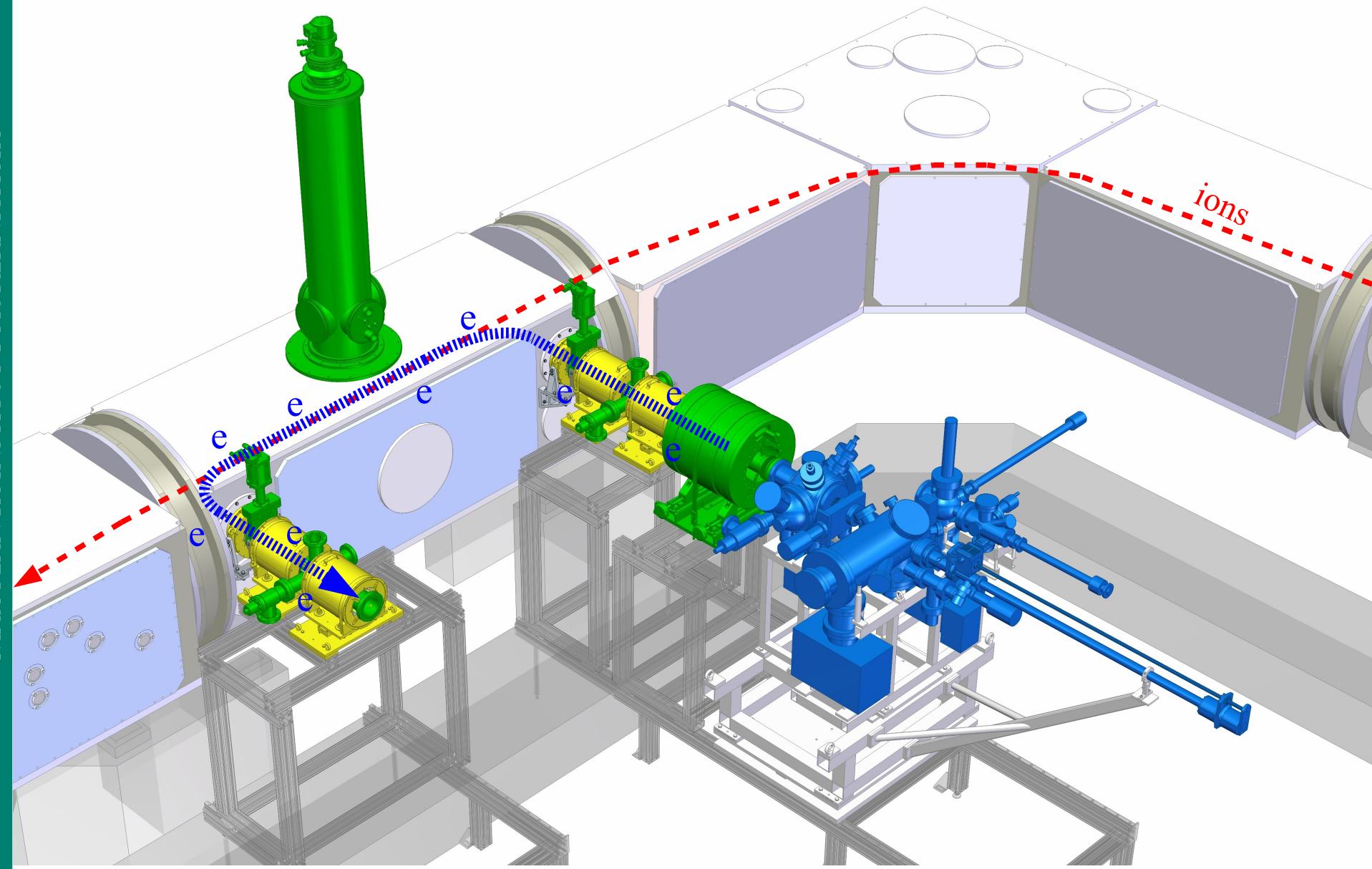


600  
km/s



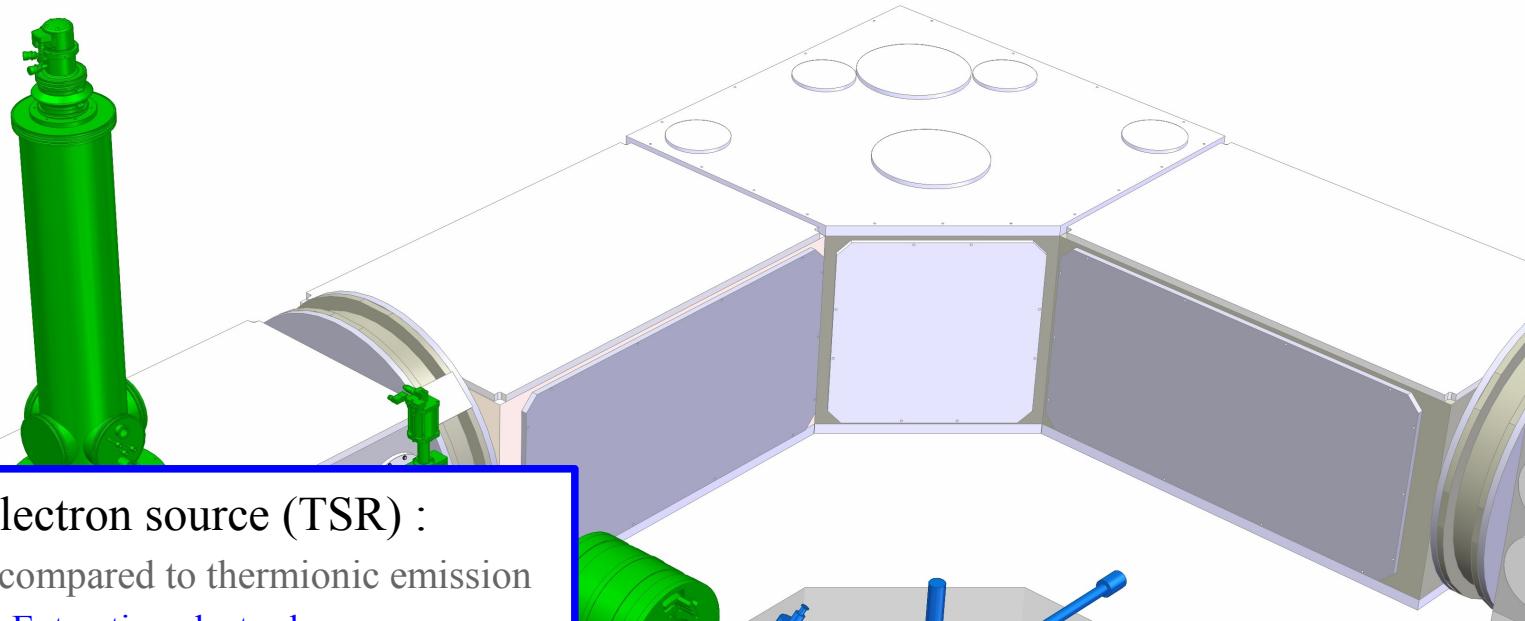
# CSR Electron Cooler

MAX-PLANCK-INSTITUT FÜR KERNPHYSIK



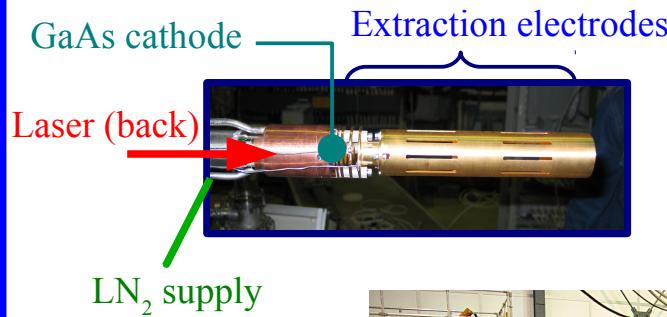


# CSR Electron Cooler



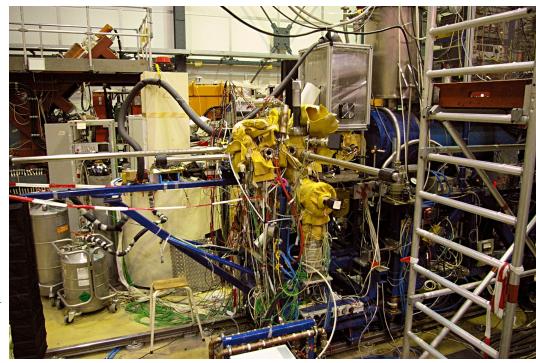
Photocathode electron source (TSR) :

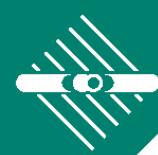
~ 10 x lower  $T_e$  compared to thermionic emission



$$k_B T_e \sim 1 \text{ meV}$$
$$n_e \sim 10^5 \text{ cm}^{-3} @ 1 \text{ eV}$$

TSR E-Target  
photocathode e-gun  
( $T_e \sim 10 \text{ K}$ )





## Summary

Slow molecular ion beams are fun but challenging.

---

e-cooling of them requires cold, slow electron beams.  
GaAs photo-cathodes are ideal sources.

---

The photocathode e-cooler at TSR has cooled molecular ions up to mass 39 to emittances of  $\sim 0.01 \mu\text{m}$  in a few seconds.

---

A low energy (1 eV) e-cooler based on the same emitter is being build for the electrostatic Cryogenic Storage Ring.





# Thank you!

**Max-Planck-Institut  
für Kernphysik,  
Heidelberg**



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Arno Becker

Stephen Vogel

Manfred Grieser

Andreas Wolf

C. K.

**Institute of Semiconductor  
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Aleksandr Terekhov

Aleksandr Jaroshevich

**Columbia Astrophysics Lab,  
New York**



Oldřich Novotný

Daniel W. Savin

**Justus-Liebig Universität,  
Gießen**



Kaija Spruck

Stefan Schippers



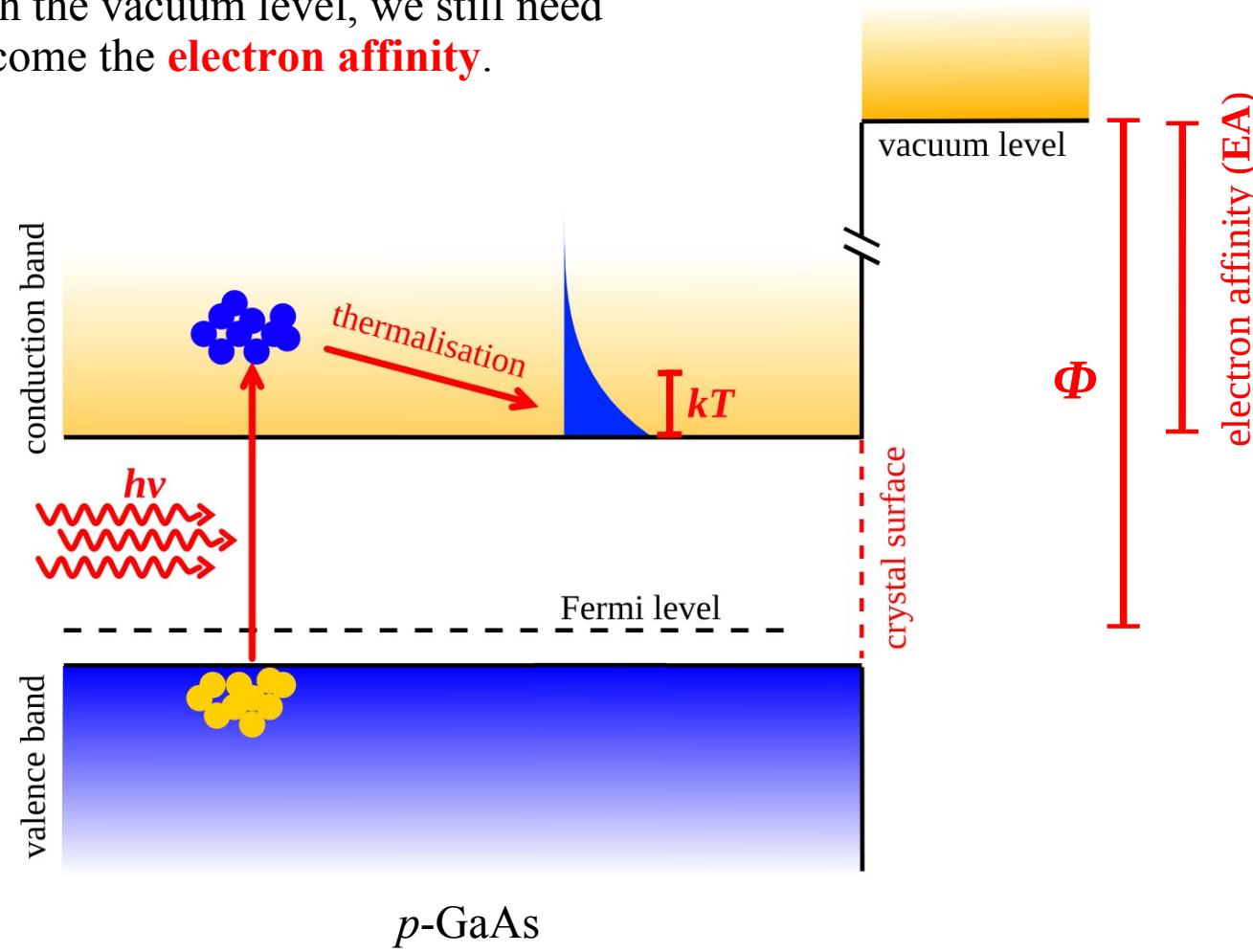




# GaAs photo cathodes

- Negative Electron Affinity:

- To reach the vacuum level, we still need to overcome the **electron affinity**.

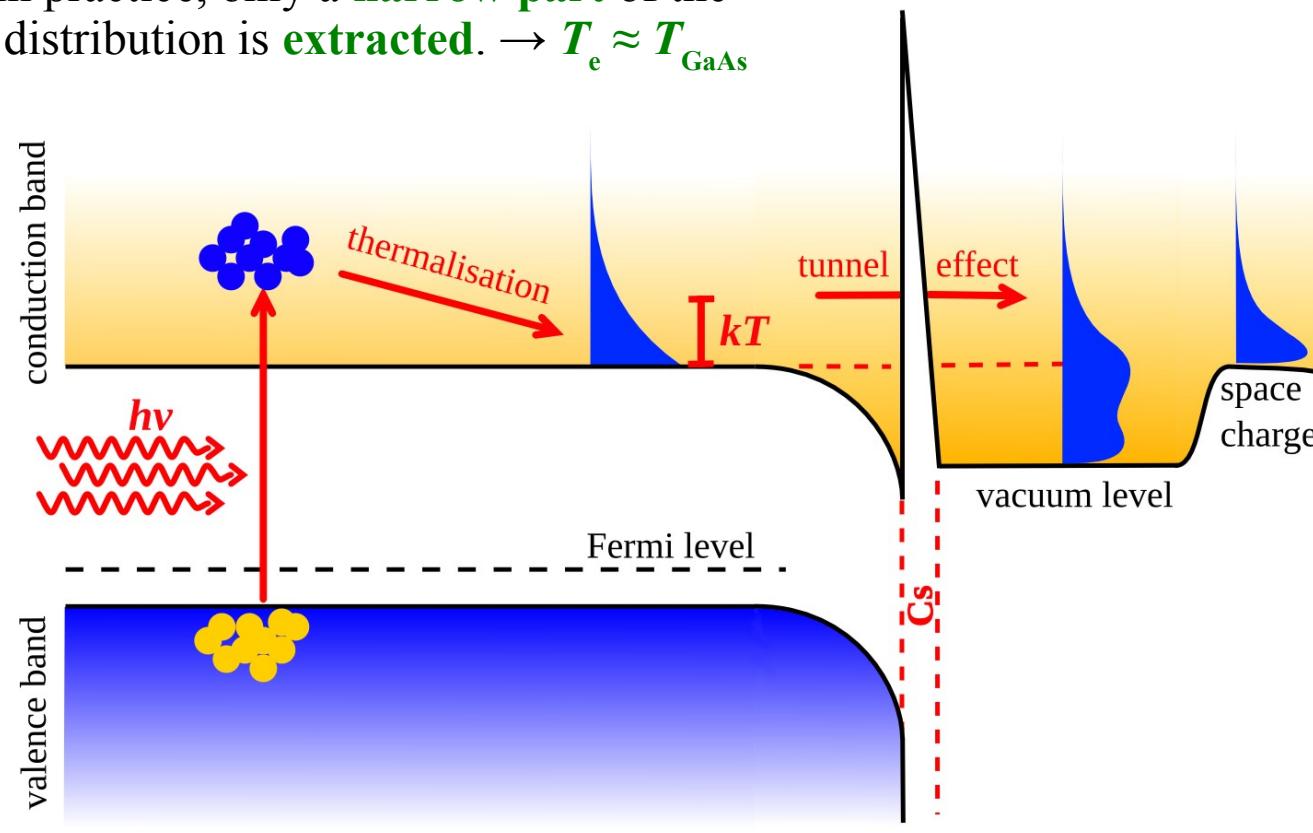




# GaAs photo cathodes

## ■ Negative Electron Affinity:

- electron-phonon **scattering broadens** the electron energy distribution ...
- ... but in practice, only a **narrow part** of the energy distribution is **extracted**.  $\rightarrow T_e \approx T_{\text{GaAs}}$



$p$ -GaAs



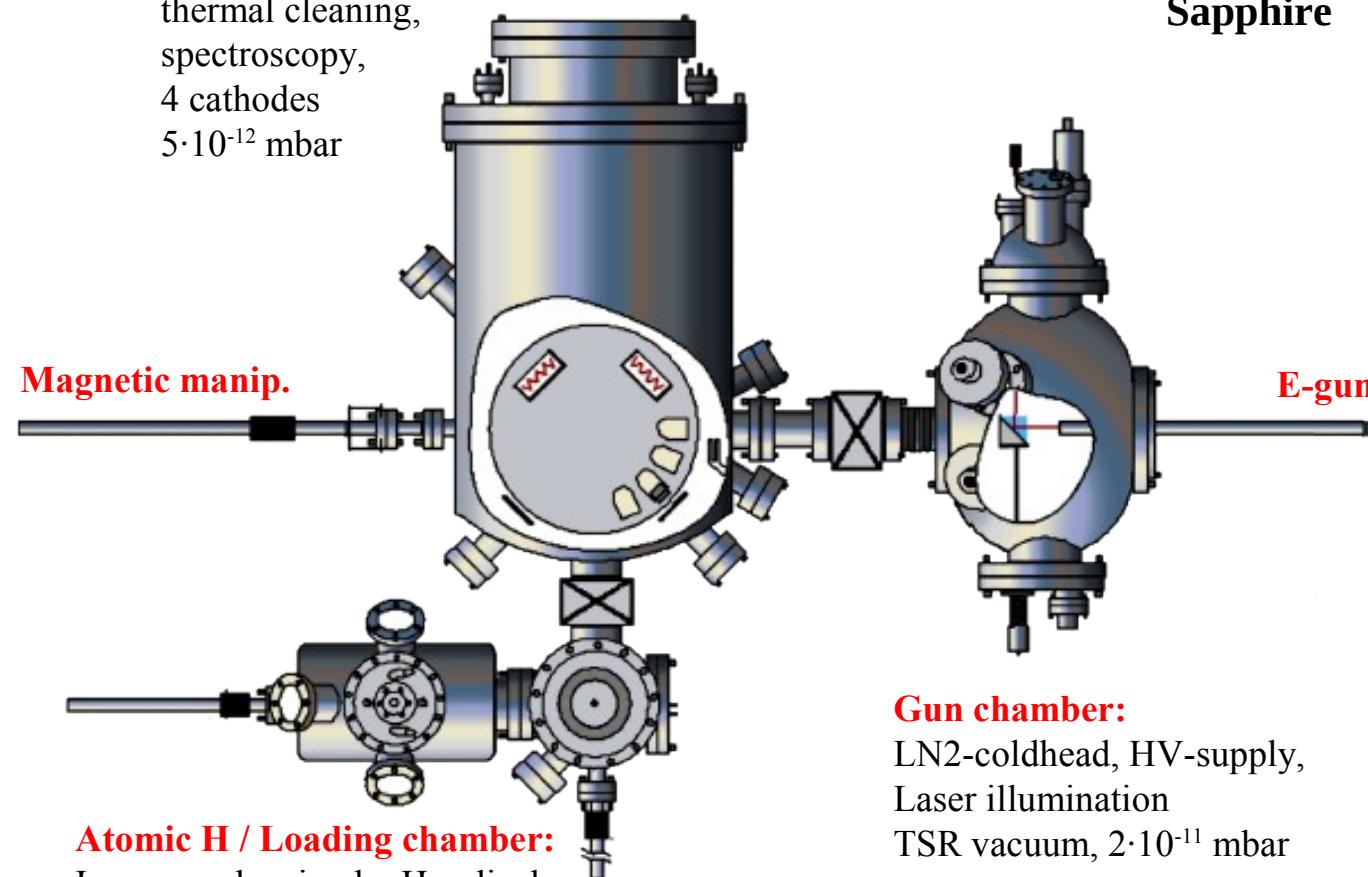
# Photo-cathode electron cooler

## Photocathode setup



### Preparation chamber:

Cs/O activation,  
thermal cleaning,  
spectroscopy,  
4 cathodes  
 $5 \cdot 10^{-12}$  mbar



### Atomic H / Loading chamber:

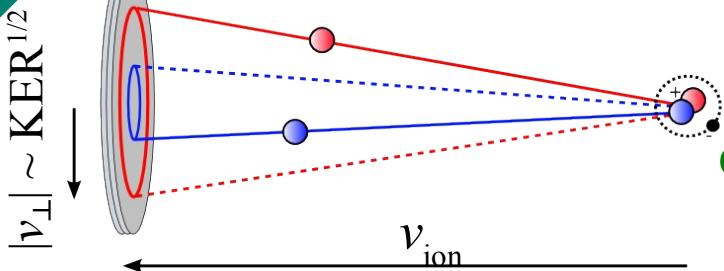
In-vacuo cleaning by H radicals,  
air-lock for cathode loading,  
 $10^{-9}$  mbar

### Gun chamber:

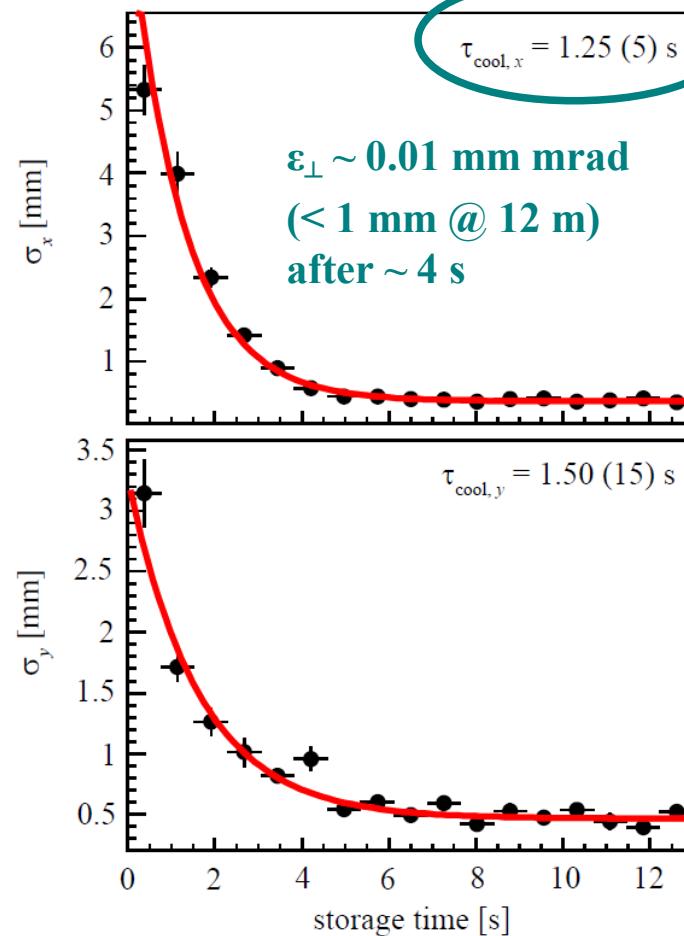
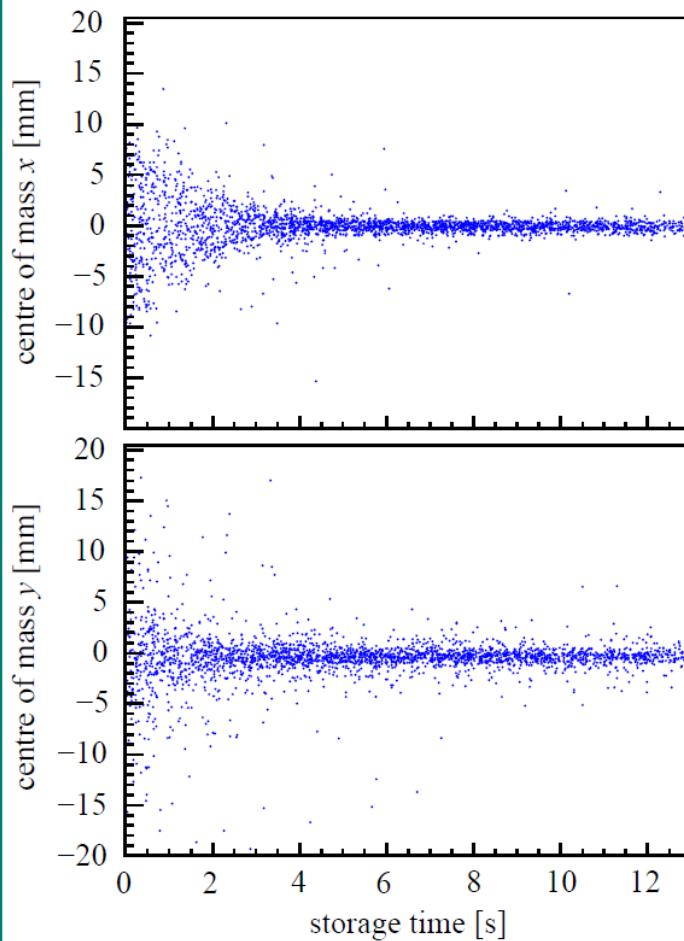
LN<sub>2</sub>-coldhead, HV-supply,  
Laser illumination  
TSR vacuum,  $2 \cdot 10^{-11}$  mbar



# Electron cooling at low velocity



Cooling  $\text{CF}^+$  at  $E_e = 46 \text{ eV}$



$$\tau \sim \frac{M_{\text{ion}} T_e^{3/2}}{Z_{\text{ion}}^2 n_e}$$

fits with

$$T_{e,\perp} = 15(3) \text{ K}$$

We need a cold electron source!

~~expected for thermionic electron cooler:  
( $T_{\perp} \sim 100 \text{ K}$ )~~  
 $\tau \sim 30 \text{ s}$   
(longer than ion lifetime)





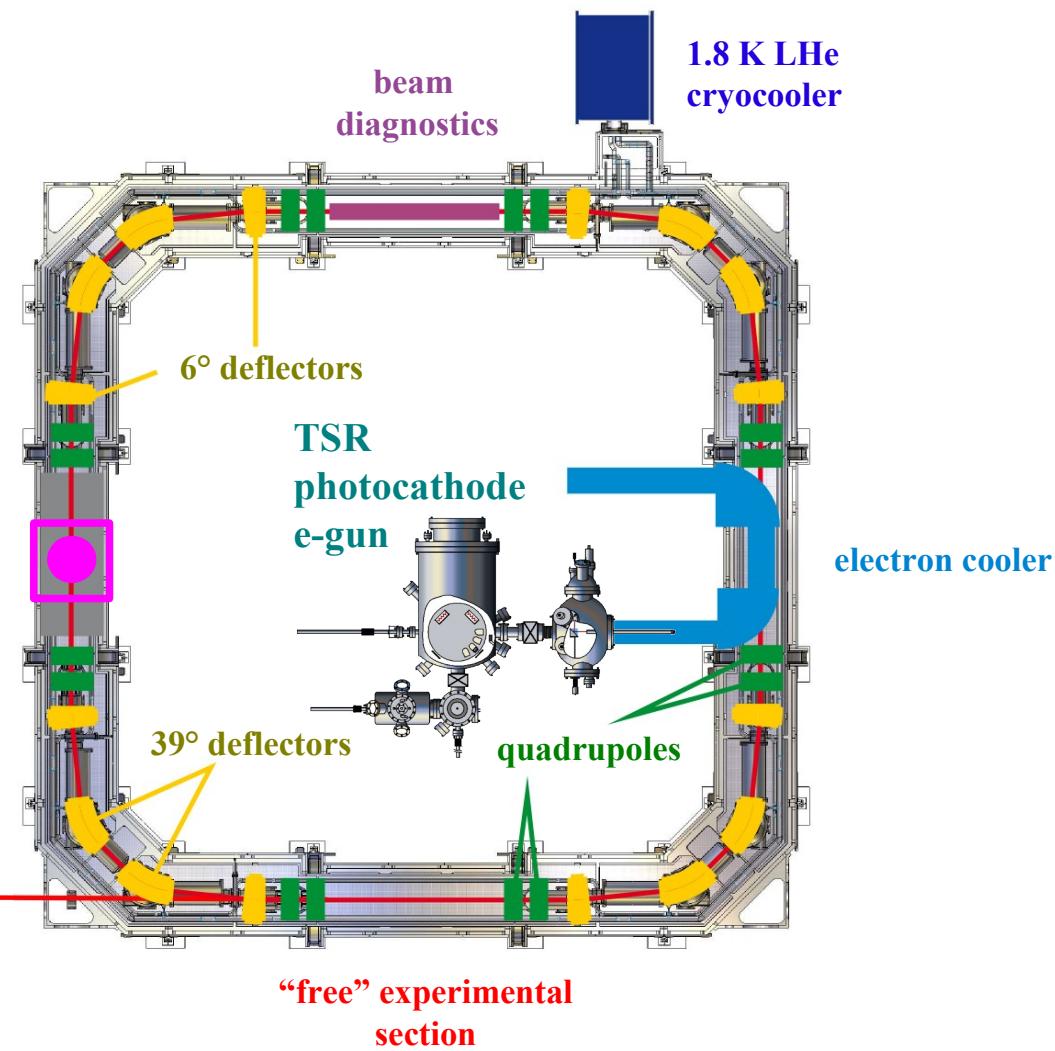
# The Cryogenic Storage Ring

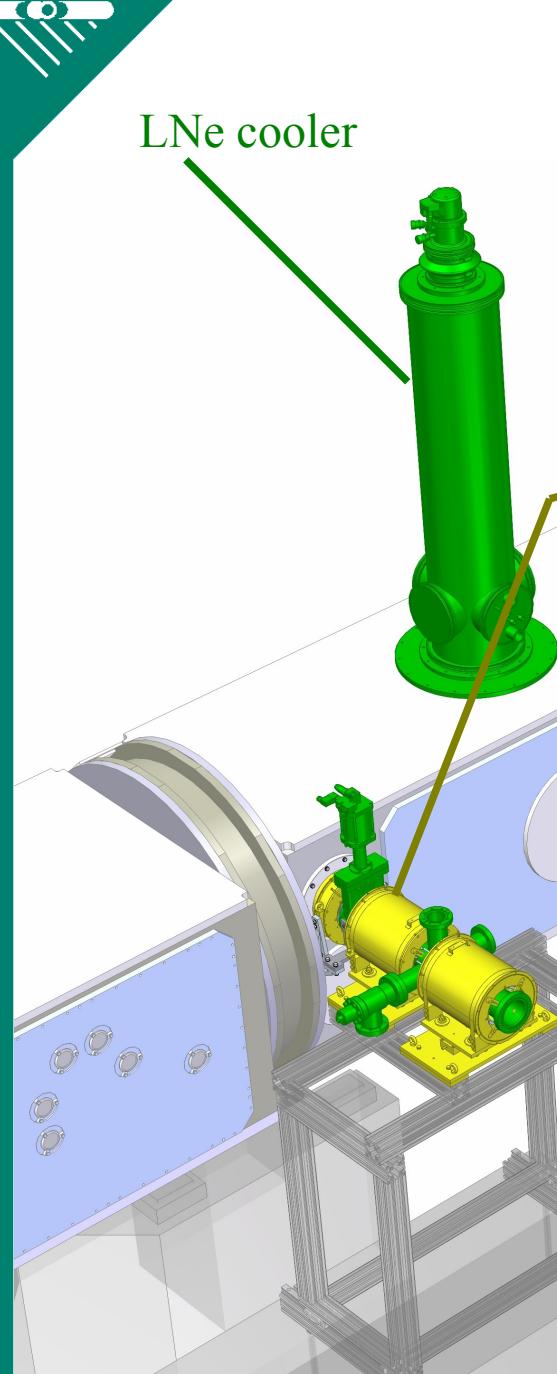
A fully-featured next generation storage ring

circumference:	35 m
beam energy:	300 keV/q
temperature:	10 ... 300 K
res. gas press. (@ < 10 K):	$10^{-13}$ mbar RTE (~ 1000 cm <sup>3</sup> )
ion masses (for $q = 1$ e)	
no cooling:	1 ... "∞" u
with cooling:	1 ... 160 u

gas jet target  
+ reaction microscope

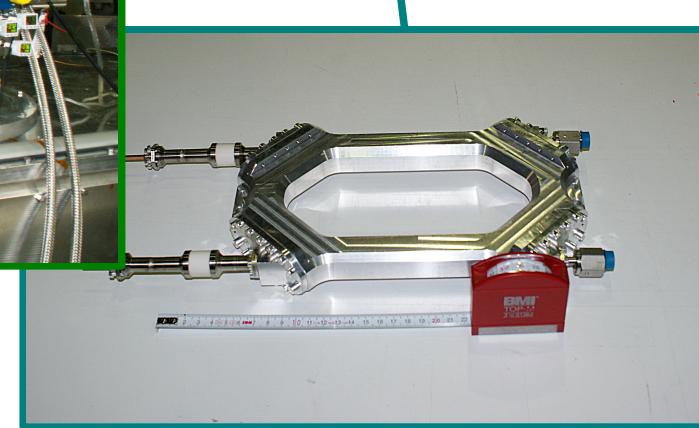
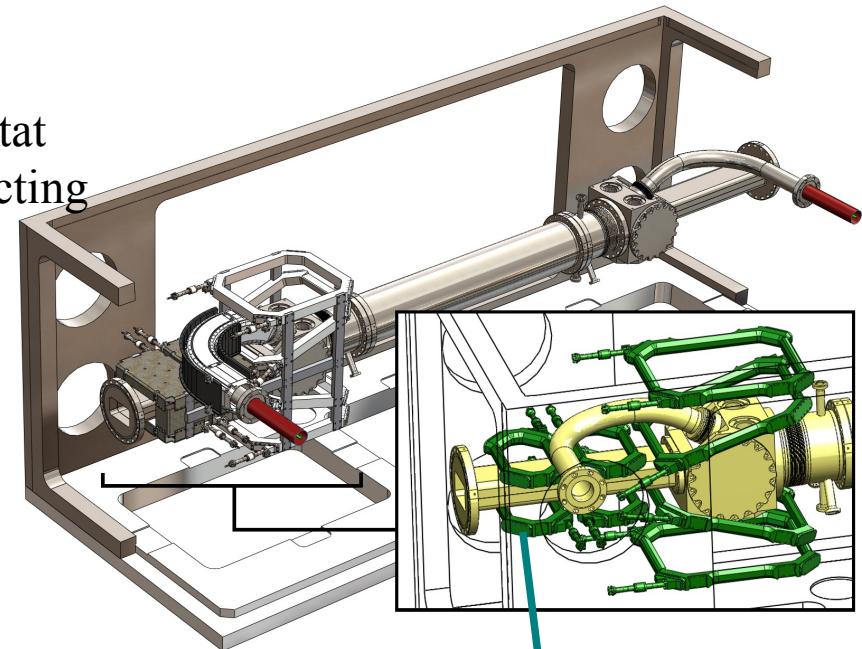
Injection  
(20 ... 300 keV/q)





LNe Cooler:  
**Guiding field** inside cryostat  
is provided by superconducting  
(HTS) coils.

A ~32 K LNe cooler for  
the HTS coils to has





# The CSR electron cooler

Electron energy: towards 1 eV and below ...

- Calibration of  $E_e$  against cathode potential taking beam **space charge** and **work function** differences into account

- Current:  
few  $\mu\text{A}$  at  $E_{\text{cool}} = 1 \text{ eV}$

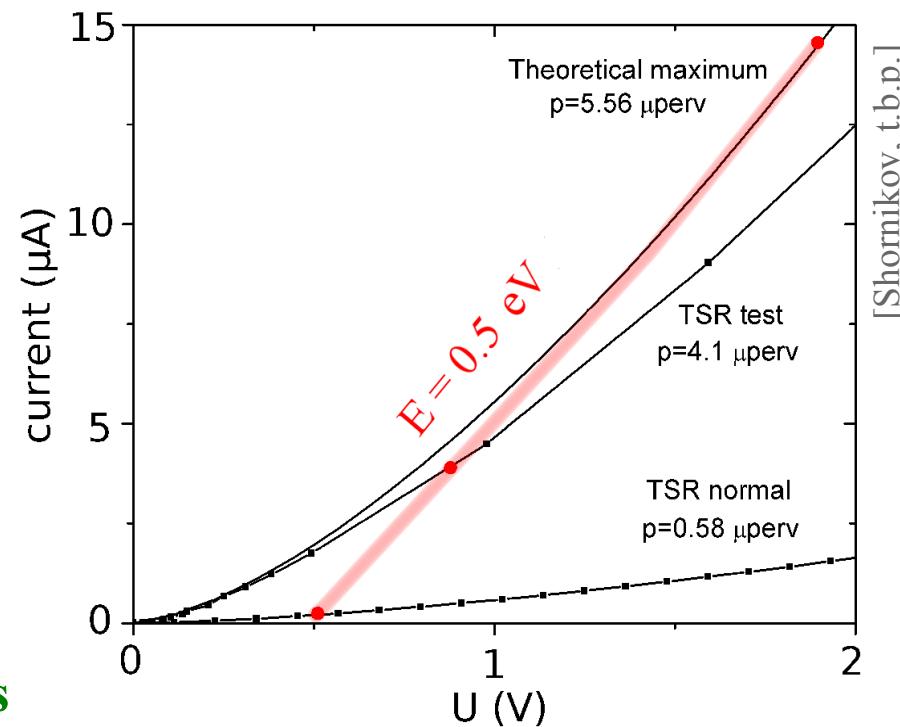
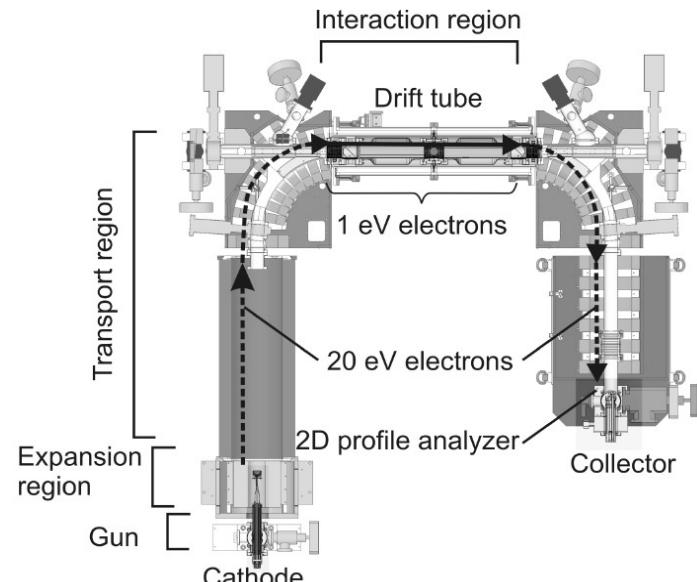
$$n_e \sim 10^5 \text{ cm}^{-3}$$

- Cooling times

$$\tau \sim \frac{M_{\text{ion}} T_e^{3/2}}{Z_{\text{ion}}^2 n_e}$$

**up to  $\sim 100 \text{ s}$  ...**

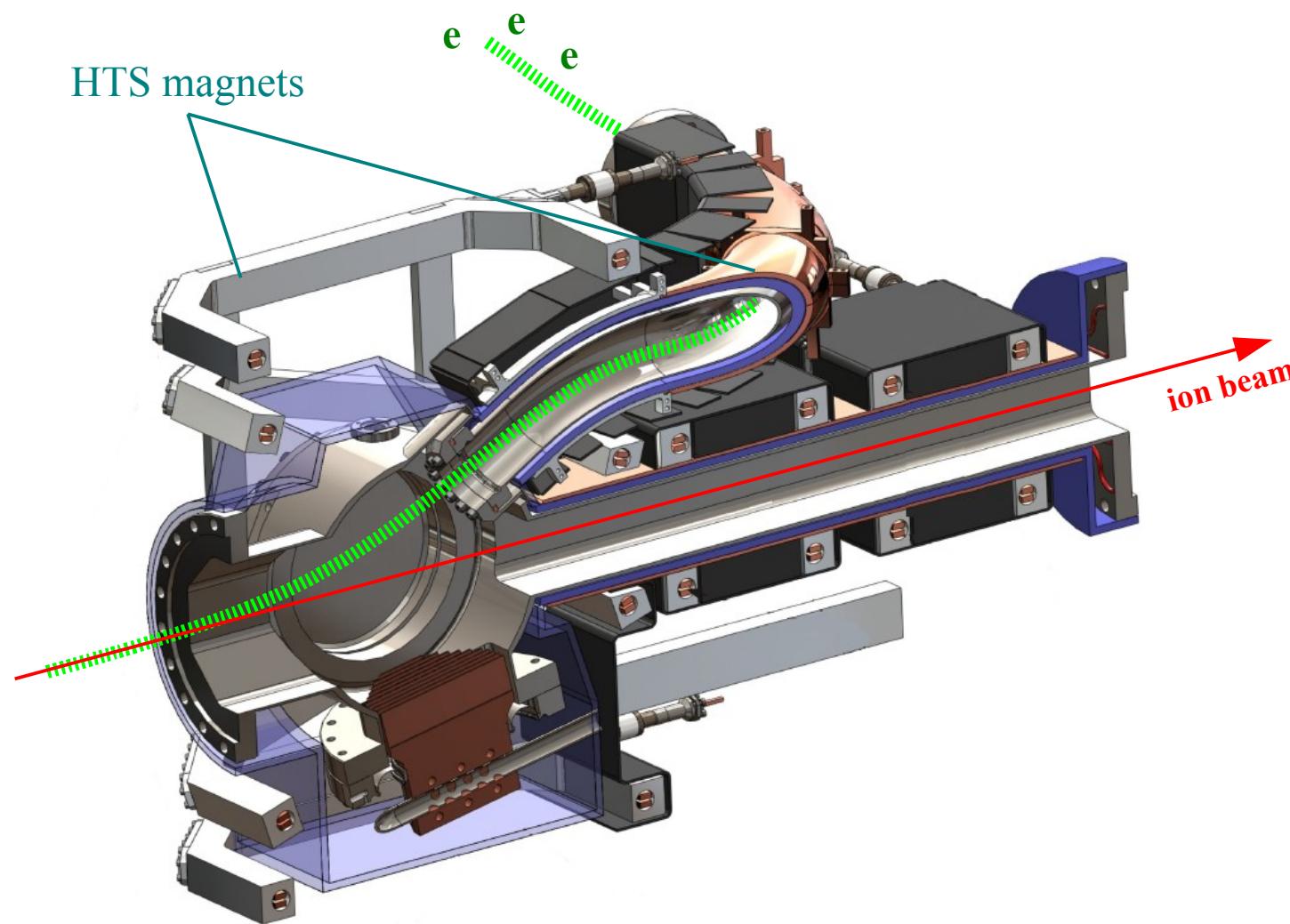
**... but: ion lifetime  $\sim 1000 \text{ s}$**





# The CSR electron cooler

Magnetic guiding field



Need  $B = 250$  G for adiabatic transport

Magnets + chambers  
cryogenic

HT superconductors  
(no heating of CSR)

Cryogen: LNe