Perspectives of Fast-Beam Molecular Photofragmentation at FLASH and FLASH II

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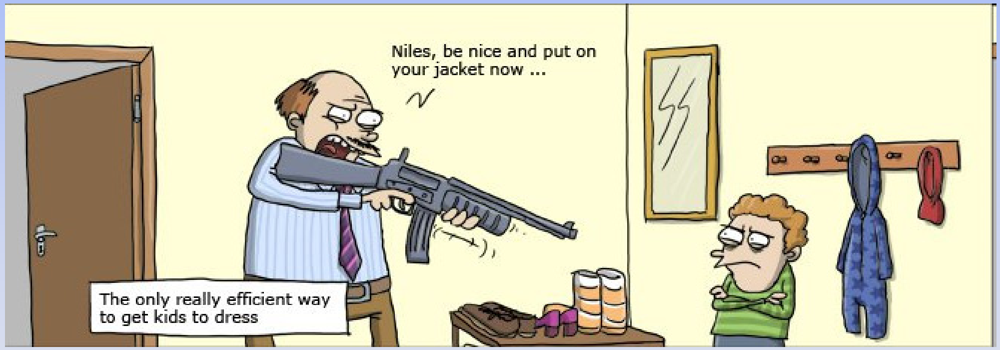

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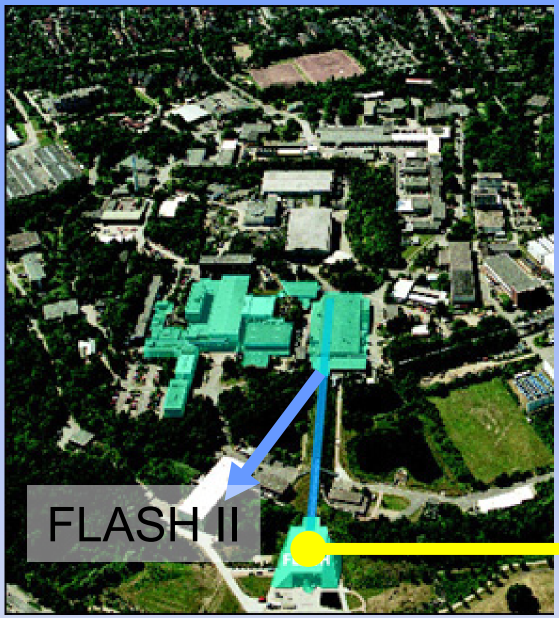
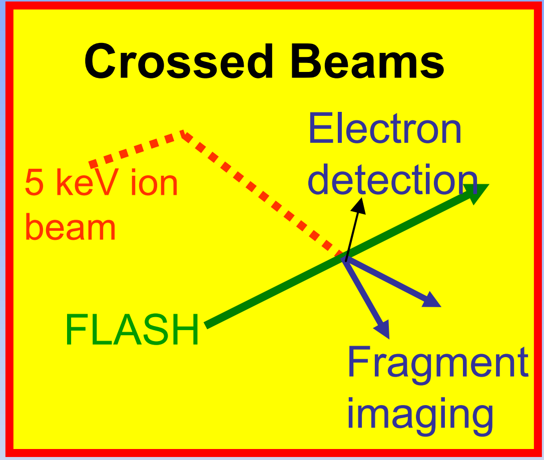
New Science Opportunities at FLASH, DESY, October 12-14, 2011

Fast beam molecular photofragmentation

→ Intense XUV light source
→ Accelerated (fast) ion beams



$AB^q + \gamma \rightarrow$



TIFF

Trapped
Ion
Fragmentation
FEL

	FLASH	FLASH II
Wavelength	4 - 50 nm (~ 310-25 eV)	4 - 80 nm (310-15 eV)
Intensity	10^{12}-10^{14} photons/pulse	10^{12}-10^{14} photons/pulse
Pulse length	10-50 fs	<200 fs
Rep. Rate	>50 pulses/train	>50 pulses/train
Wavelength scan	difficult	feasible

Does FLASH II provide new possibilities in this field ?

Who are we ?

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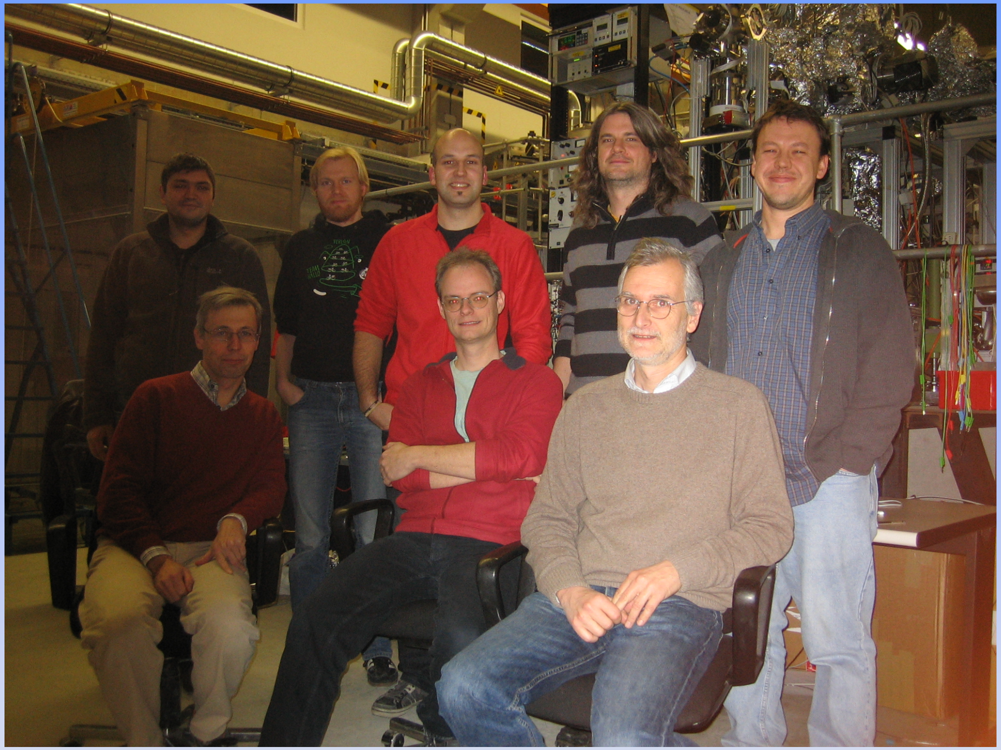

HASYLAB, DESY at Hamburg, Germany
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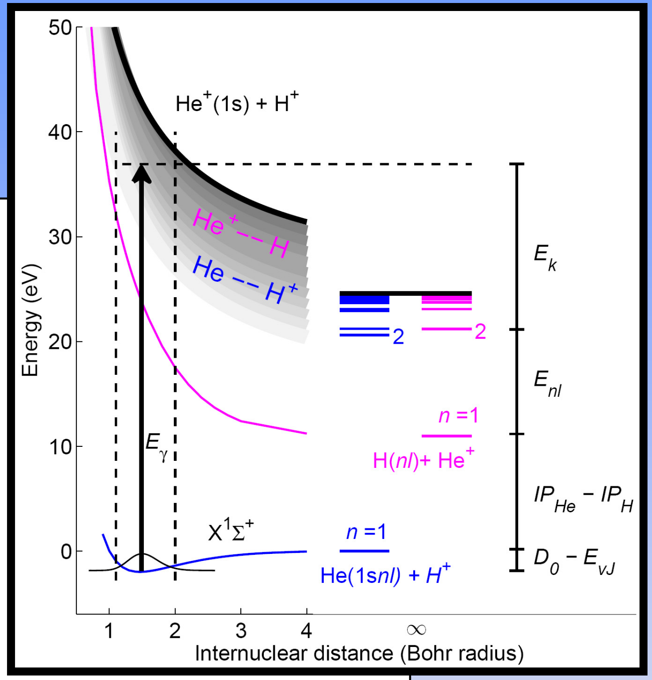
Holger Weigelt
Rolf Treusch
+ + +

Dynamics of excited states of molecular ions

- Single photon absorption



$AB^q + \gamma$

$\rightarrow [AB^q]$ Absorption

Energy dissipation

$\rightarrow A(p_A, q_A) + B^+(p_B, q_B) + e^-(p_e)$

Processes in molecular ions
>10 eV - experimentally almost unexplored

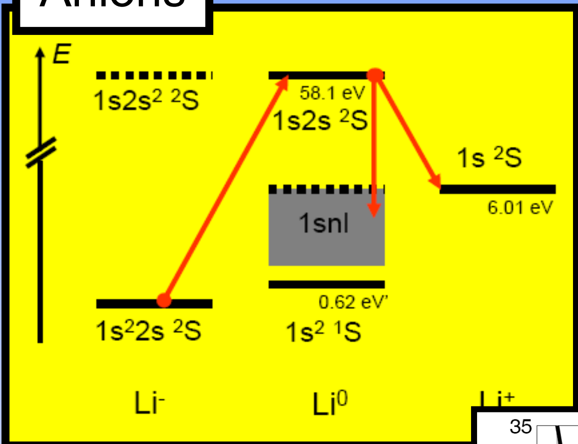
Excitation energy (λ) → excited state physics

~10-200 eV → inner + outer valence (L) shell : FLASH + FLASH II

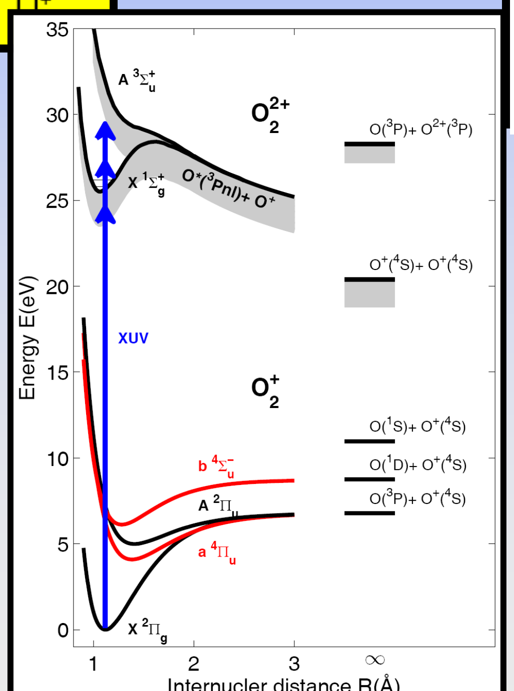
~250-3000 eV → inner (K) shell : XFEL ..

Physics of molecular ions beyond 10 eV excitation → a new world to be investigated ..

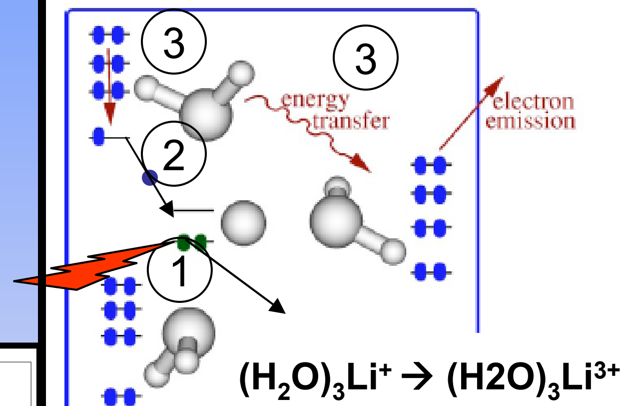
Anions



Dynamics in small molecular systems



New mechanisms
- ICD, ETMD



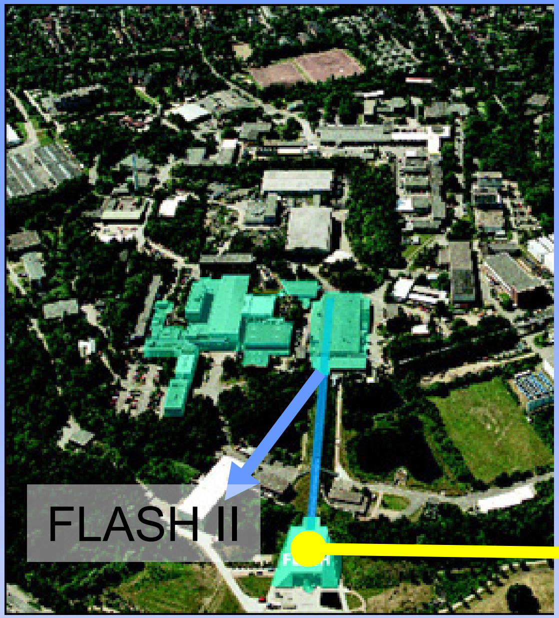
Müller *et al.* 2005

Laboratory astrophysics

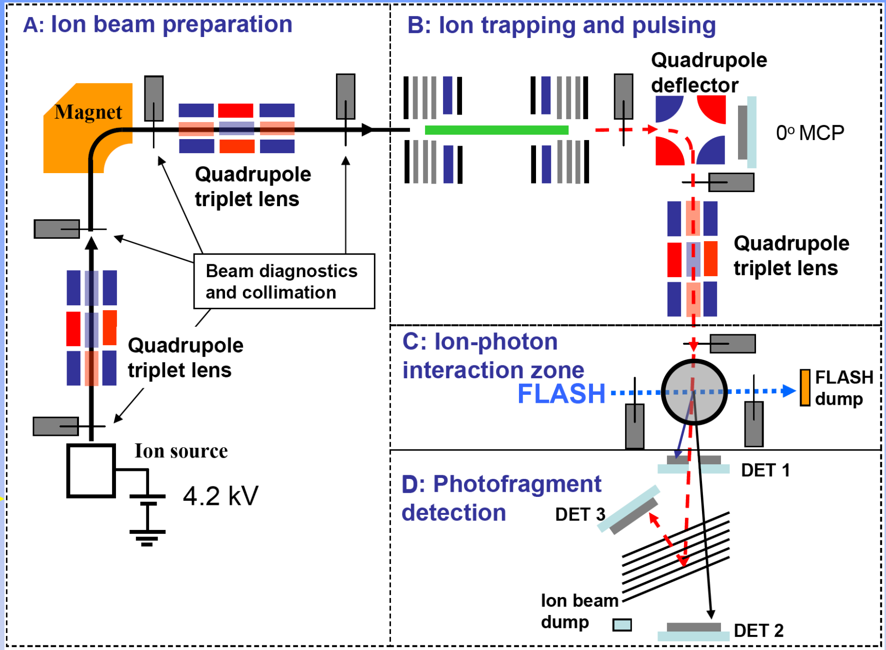
Molecular gas under XUV radiation

Infra red spectrum from of the planetary nebula NGC 7027, Hubbel telescope

The TIFF experiment at FLASH



Trapped Ion Fragmentation FEL



Free Electron Lasers

High intensity

Pulsed time structure

High rep rate

→feasible reaction rates
→feasible detection + coincidence

Advantages of fast ion beam methods:

1. Fast ion beams
   → target preparation,
   → universal access to all fragments

2. Ion storage devices
   → preparation/characterization of initial state

3. Momentum imaging
   → kinematically complete experiments

Combining FELs and fast ion beams

Ion traps:
- Control/characterization of initial state

FLASH/FLASH II
- Access to the highly excited states

$AB^q + \gamma$

$\rightarrow [AB^q]$ Absorption

Energy dissipation

$\rightarrow A(p_A, q_A) + B^+(p_B, q_B) + e^-(p_e)$

Momentum imaging – kinematically complete:
- Revealing details on the fragmentation pathways

Our studies at FLASH so far

→ **Small systems HeH^+, H_3O^+, $H^+(H_2O)_{2\text{-}4}$ …**

→ **Stepwise implementation of ion beam methods as FLASH developed**
ion cooling, electron spectroscopy, complete fragment detection

Outline

I. Examples of results & developments at TIFF

→ TIFF experiment & developments

→ Simple molecules : HeH^+ -photodissociation

→ Small water clusters : $H^+(H_2O)_{n=1-4}$
photoionization + dissociation

II. Perspectives for TIFF at FLASH

→ Anions

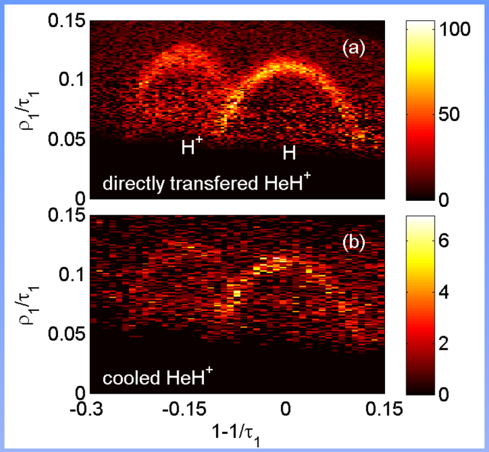
→ Di-cations

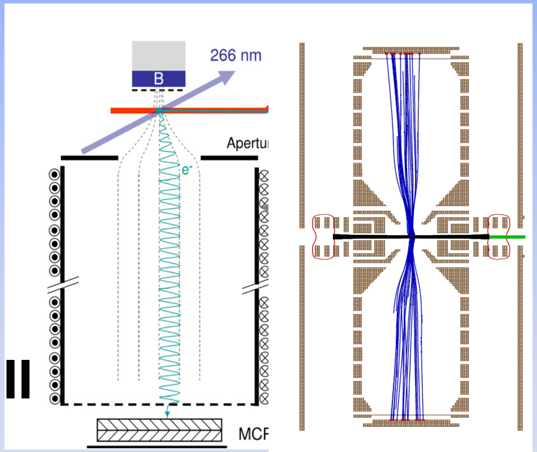
→ Molecular astrophysics, ex. hydrides

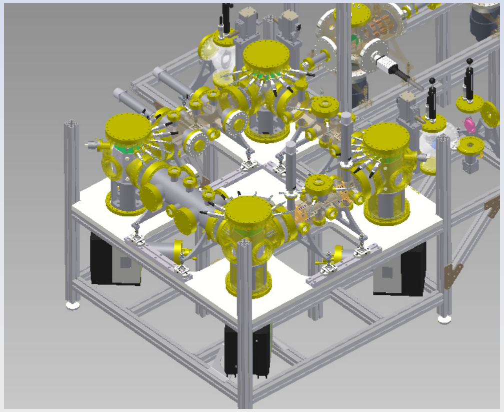
III. Perspectives / developments for FLASH/FLASH II

→ Experiments at longer wavelengths

→ Developments: Ion cooling in storage ring

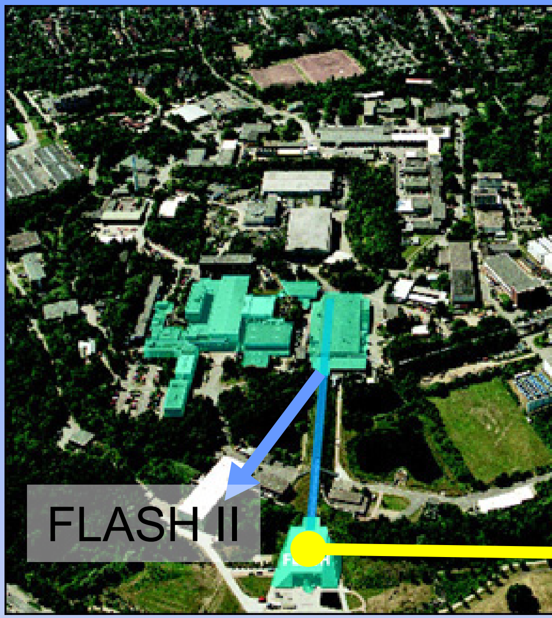




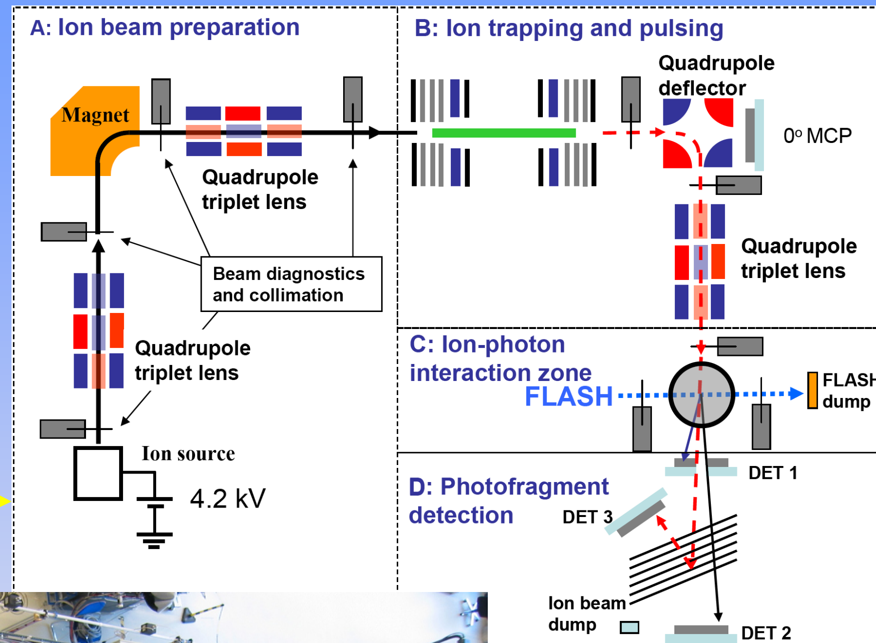


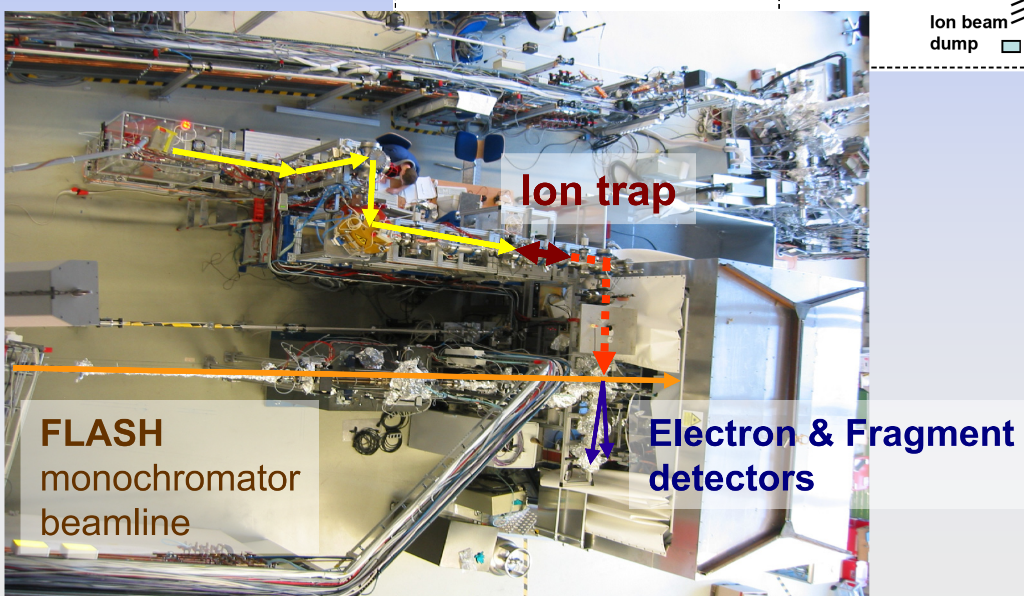
The TIFF experiment at FLASH

The TIFF experiment at FLASH

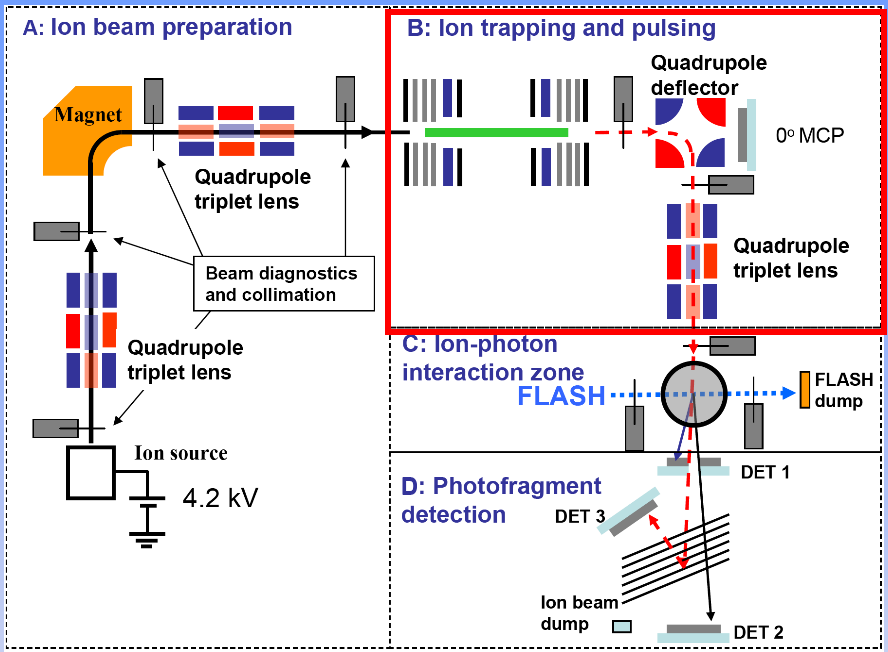


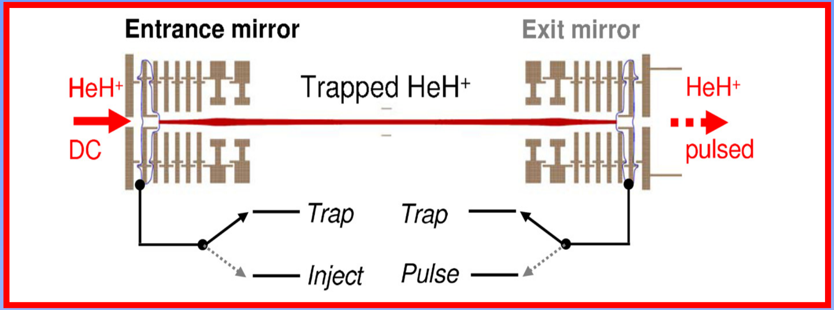
Trapped Ion Fragmentation FEL

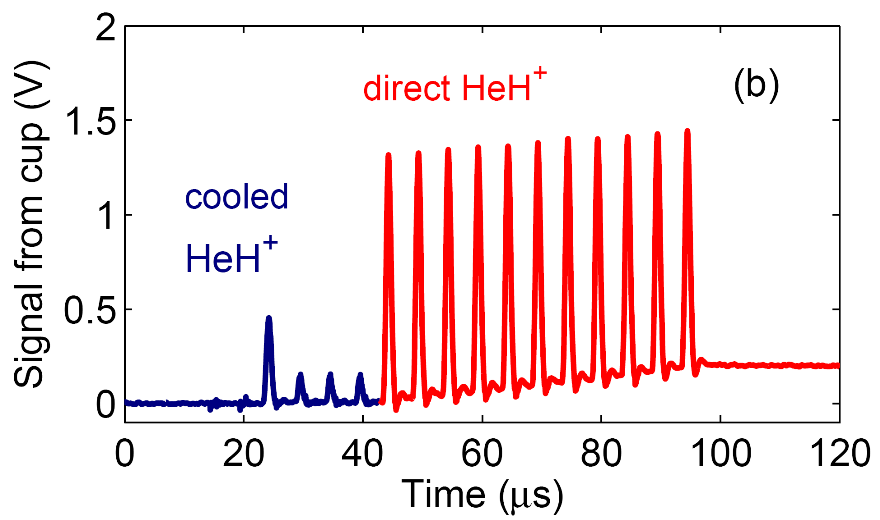




The TIFF experiment at FLASH

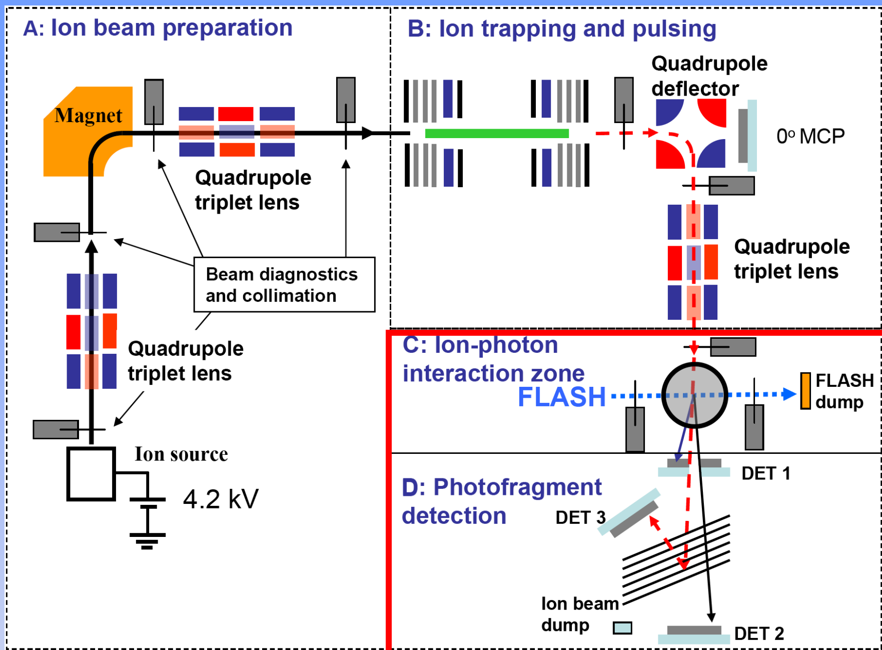


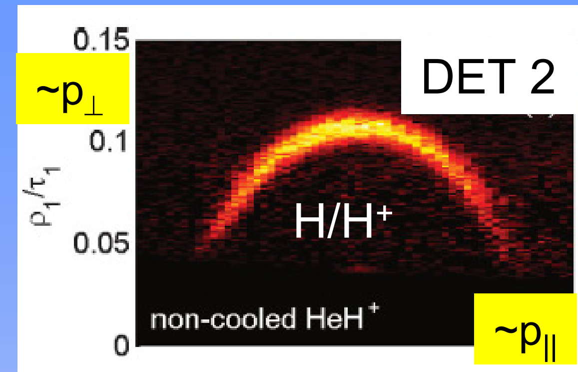


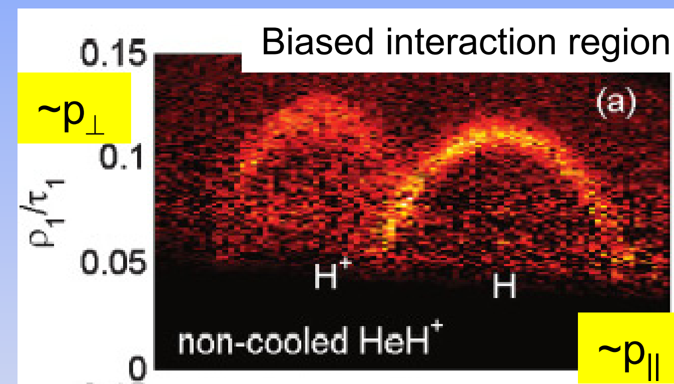


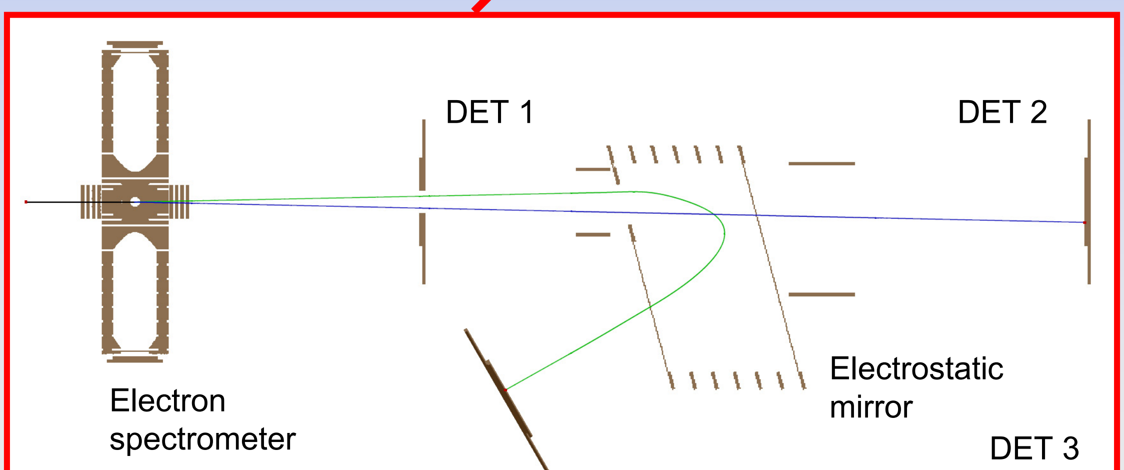
→ Vibrational cooling of molecular ions
→ Matching timestructure of ion pulses and FLASH pulses

The TIFF experiment at FLASH









→Detection of all fragments
- → Electrons
- → Charge fragments
- → Neutral fragmenets

→Biased interaction region
- → Channel identification

Examples of results from TIFF at FLASH

FLASH

→ Intense pulses in the XUV (30 μJ →)
→ High repetition rate (10Hz x 50 pulses = 500 pulse/sec)

The TIFF experiment

→ Fast dilute target of molecular ions
→ Cold ions by electrostatic ion trapping
→ Momentum Imaging of fragments

HeH^+ + 32 nm → $[HeH^+]^*$ → $He + H^+$
→ $He^+ + H$

valence excitation
+ dissociation

$H^+(H_2O)_n$ + 13.5 / 35 nm / 59 nm → break-up

valence ionisation
+ dissociation

small n (= 1 – 2) → release of protons
large n (= 3 – 4) → many body break-up

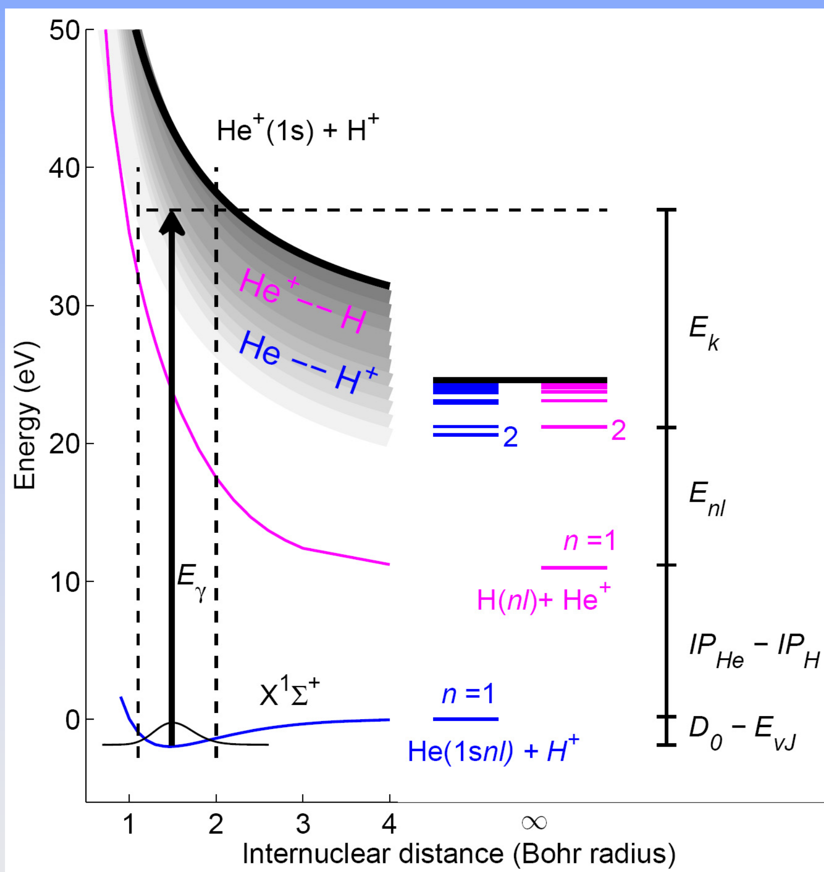
Examples from beamtime August 2011: H_3O^+, H_2O-H-OH_2

HeH$^+$ photodissociation at 32 nm

→ what can we do with momentum imaging of cold ions ?

HeH$^+$ + 32 nm → **[HeH$^+$(Σ/Π)]***

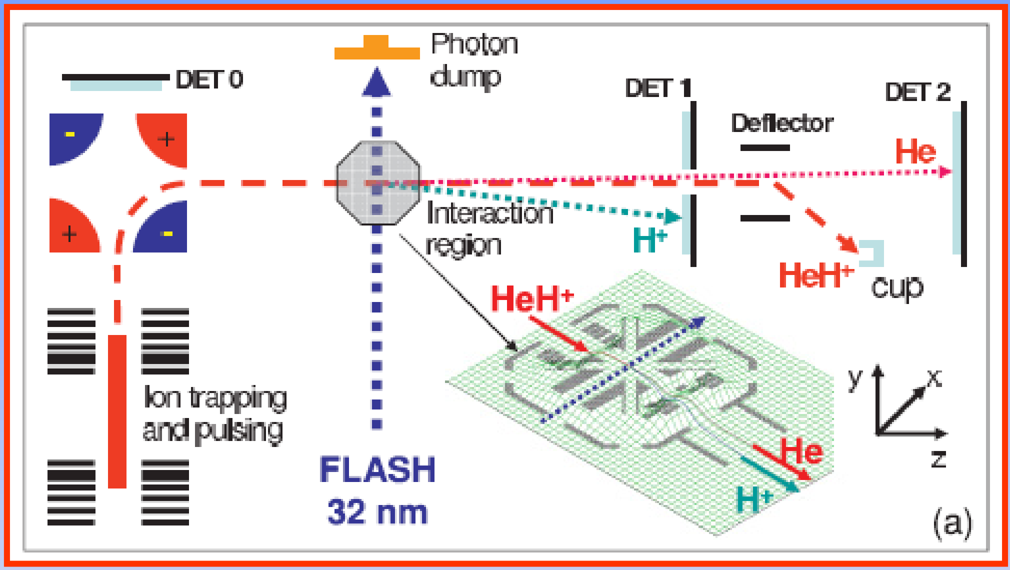
He(1s*nl*) + H$^+$ **He$^+$(1s) + H(*nl*)**

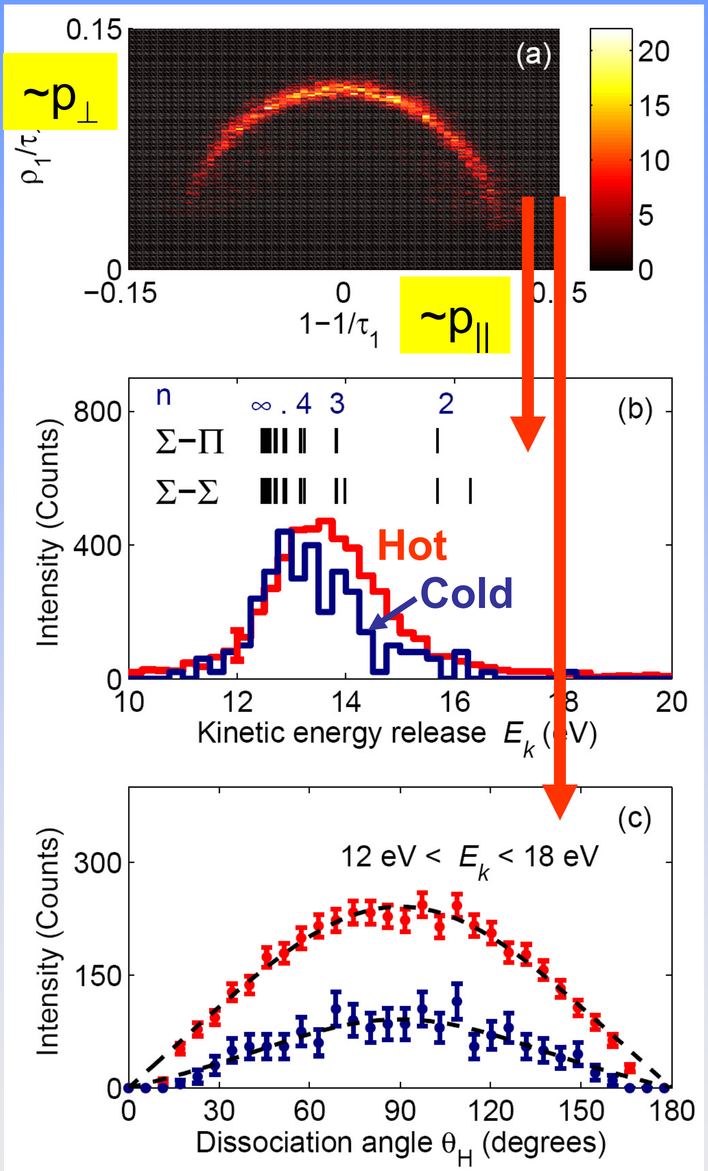


- Dominating reaction channels ?
  He + H$^+$ or He$^+$ + H
- Dominating absorption states ?
  Σ-Σ versus Σ-Π
- Dominating fragment states ?
  H(*nl*), He(1s*nl*) – which *n* ?
- Importance of vibrational excitation ?

HeH$^+$ photodissociation at 32 nm

→ the **He(1s*nl*) + H$^+$** channel





Coincidence between DET1 + DET2

→Isolates He + H$^+$ from He$^+$ + H

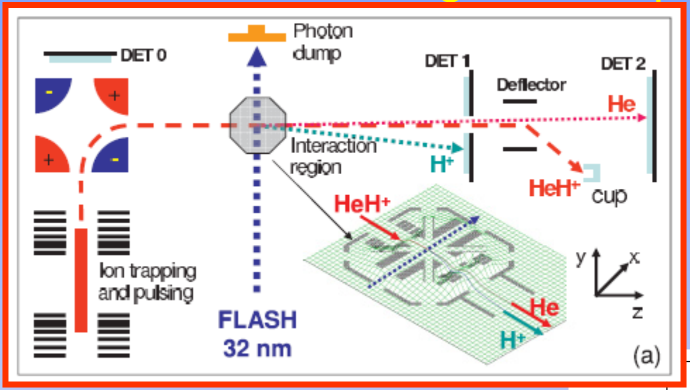
Theory:

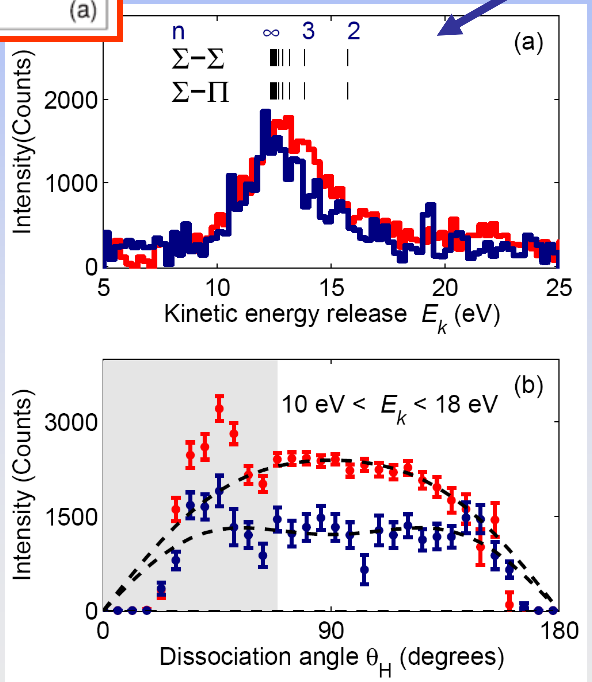
I. Dumitriu and A. Saenz, J. Phys. B ***42****,165101 (2009) / K. Sodoga et al., Phys. Rev. A* ***80****, 033417 (2009)*

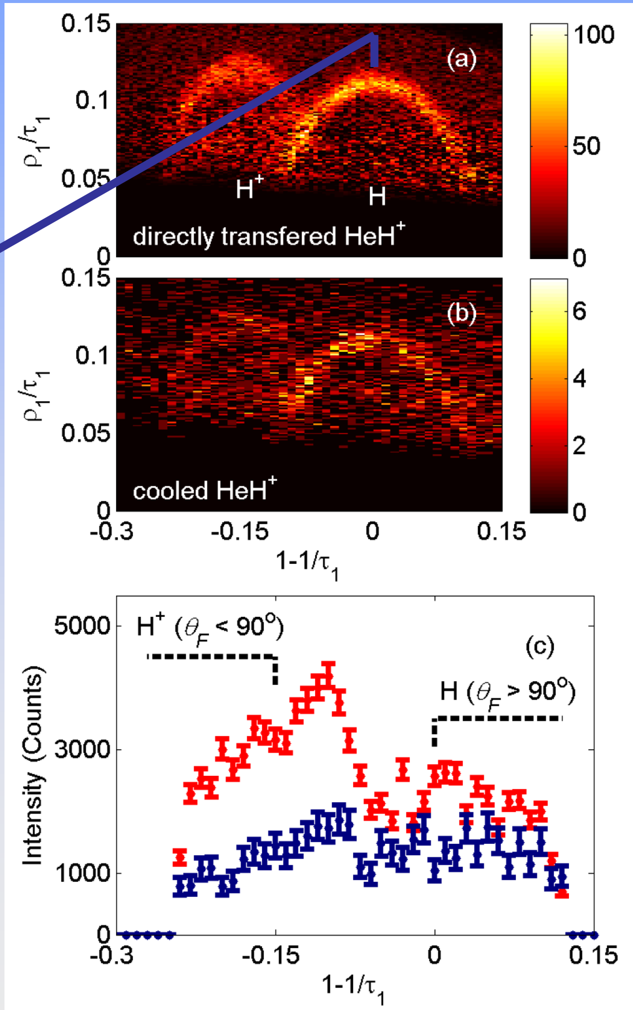
HeH^+ photodissociation at 32 nm

→ the **$He^+(1s)$ + H(*nl*)** channel

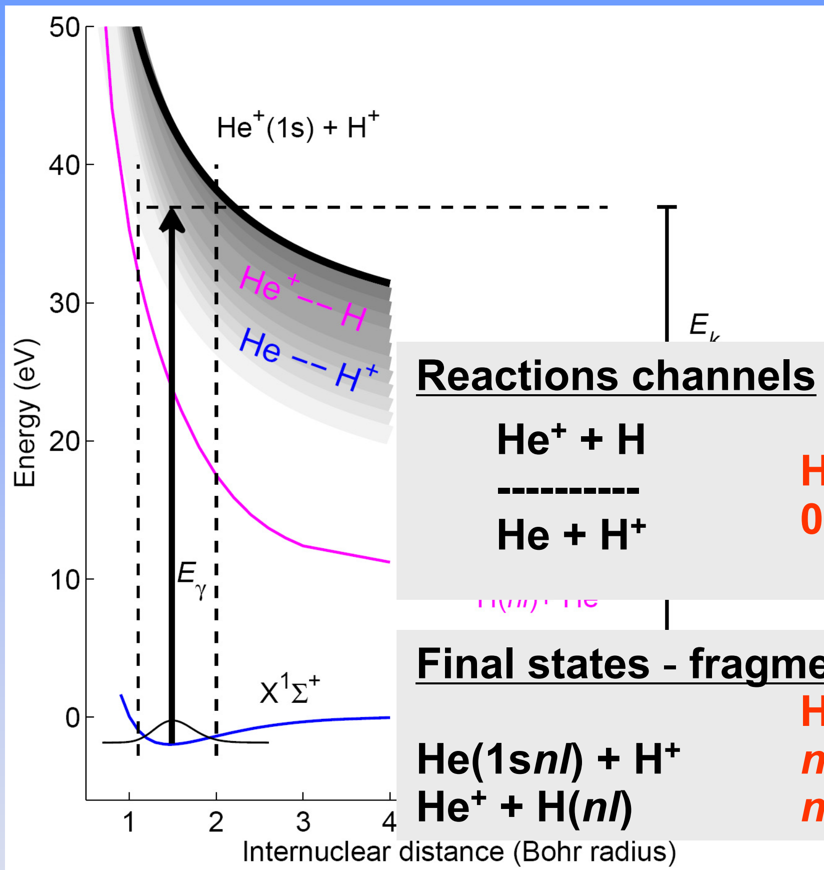
Biased interaction region → separate fragments in TOF







HeH$^+$ photodissociation at 32 nm
– Quantitative results



Reactions channels ← **Imaging with biased interaction region**

	HOT ions	COLD ions (v=0)	Theory
He$^+$ + H / He + H$^+$	0.96 ± 0.11	1.70 ± 0.48	~ 1.6

Final states - fragment excitation ← **Final kinetic energy**

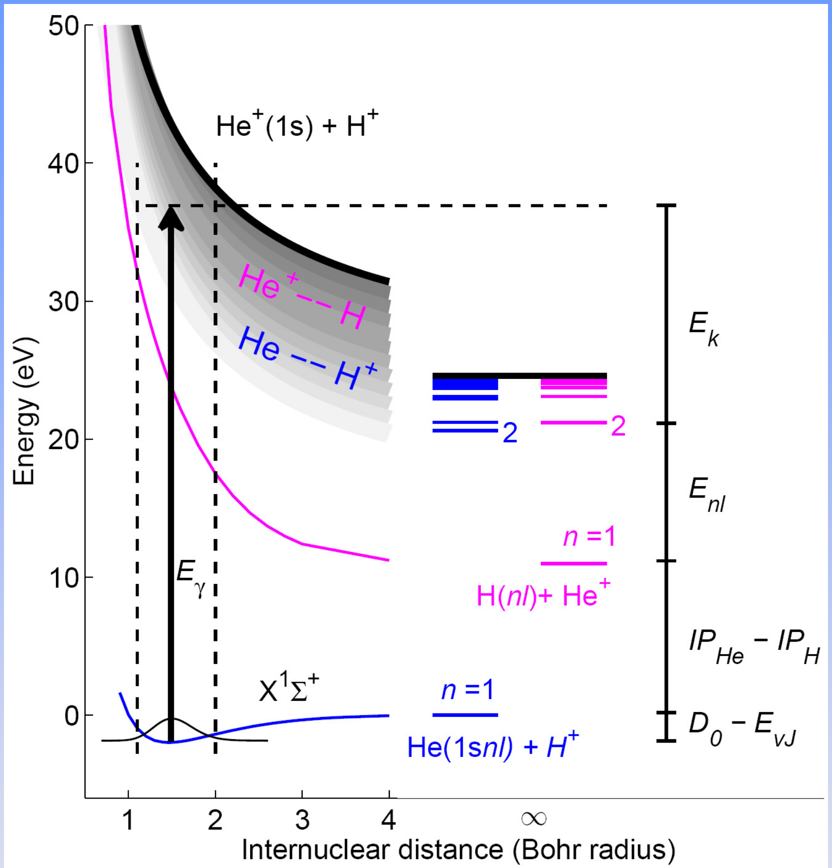
	HOT ions	COLD ions (v=0)	Theory
He(1s*nl*) + H$^+$	*n* > 3 - 4	*n* > 3 - 4	?
He$^+$ + H(*nl*)	*n* > 3 - 4	*n* > 3 - 4	?

Absorption states ← **Angular distributions**

	HOT ions	COLD ions (v=0)	Theory
He + H$^+$:	Σ - 30 ± 2 %	Σ - 24 ± 6 %	~ 30 %
	Π - 70 ± 2 %	Π - 76 ± 6 %	~ 70 %
He$^+$ + H :	Σ - 38 ± 3 %	Σ - 50 ± 3 %	~ 15 %
	Π - 62 ± 1 %	Π - 50 ± 5 %	~ 85 %

Pedersen et al. PRL ***98****, 223202 (2007) / Pedersen et al. PRA* ***82****, 023415 (2010)*

HeH^+ photodissociation at 32 nm - summary



What did we learn about the physics of dissociating HeH^+ ?

HeH^+ photodissociation prefers the higher excited states in the dissociation process

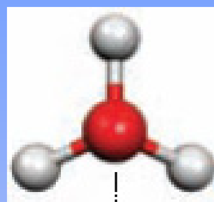
→ Non-adiabatic interactions ?

→ Density of states ?

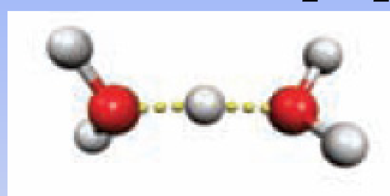
*Pedersen et al. PRL **98**, 223202 (2007) / Pedersen et al. PRA **82**, 023415 (2010)*

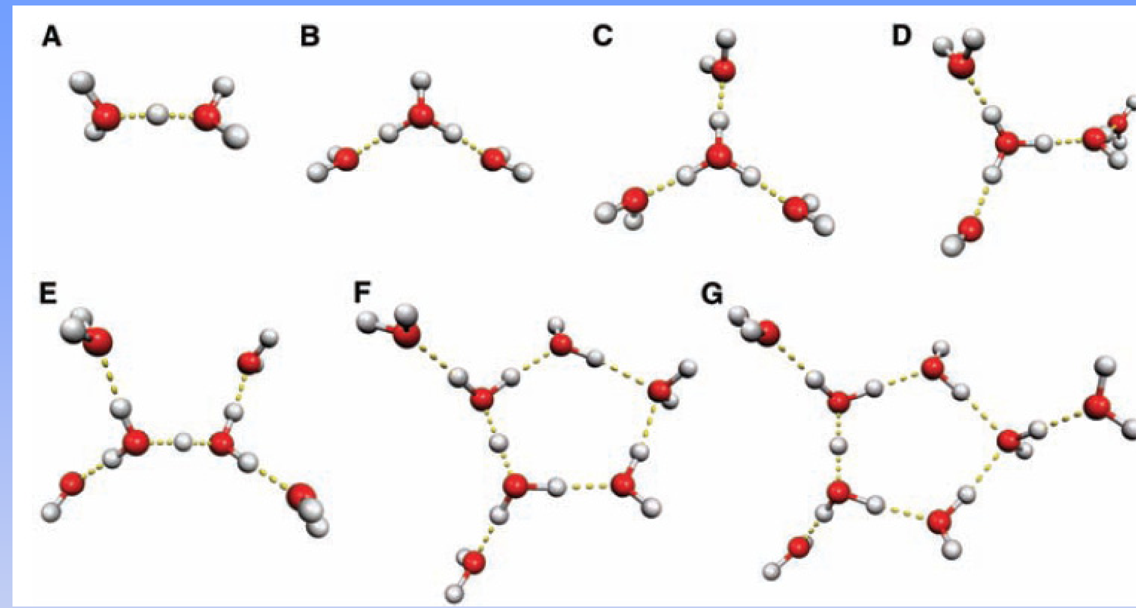
Small water cluster cations

Eigen form H_3O^+



Zundel form $H^+(H_2O)_2$





J. M. Headrick et al., Science 308, 1765 (2005)

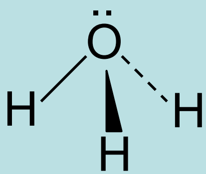
→ Building blocks for proton solvation in water and for isolated protonated water clusters

→ Water clusters ions in the Earth's ionospere

A lot of studies in the IR range

Almost no knowledge on water cluster ion fragmentation under ionizing radiation

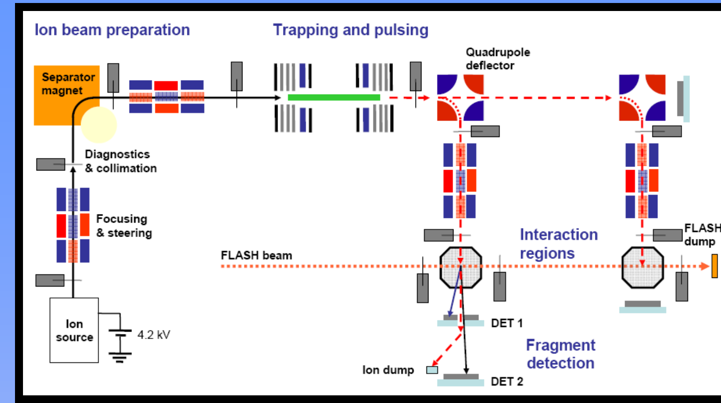
H_3O^+ + 13.5 nm studied at TIFF
- first measurements 2009



+ 13.5 nm (92 eV)

→ $H_2O^+ + H^+$ $\quad 0.37 \pm 0.18 \cdot 10^{-18}$ cm^2

→ $OH + H^+ + H^+$



$OH^+ + H + H^+$ (small)

~~**$O^+ + H_2 + H^+$**~~

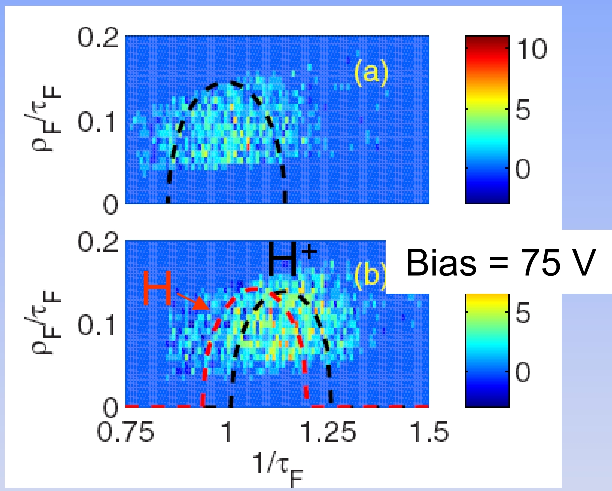
~~**$OH^+ + H_2^+$**~~

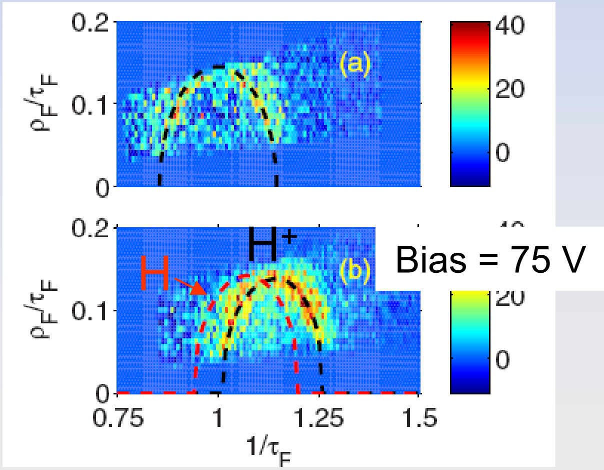
~~**$O + H_2^+ + H^+$**~~

~~**$O^+ + H_2^+ + H$**~~

~~**$O + H + H^+ + H^+$**~~

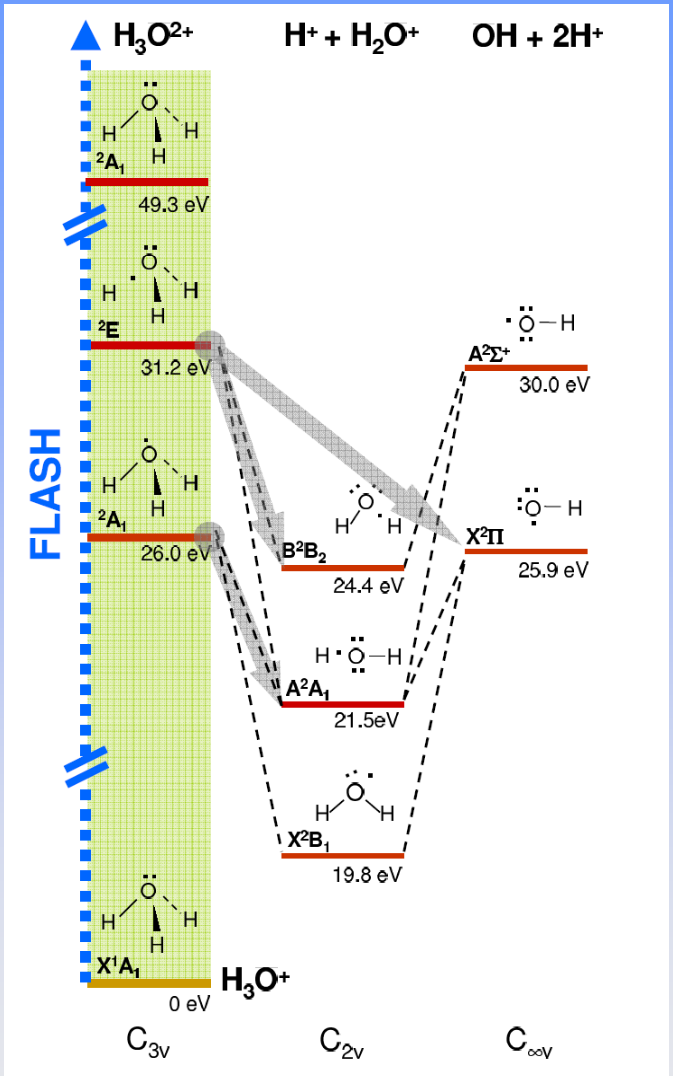
~~**$O^+ + H + H + H^+$**~~



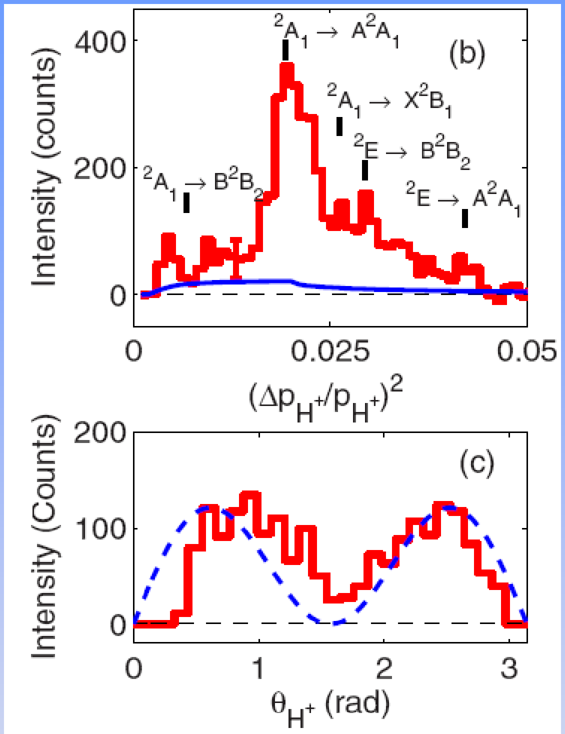


*Pedersen et al. PRA **80**, 012707 (2009)*

H_3O^+ + 13.6 nm at FLASH



$\rightarrow H_2O^+ + H^+$



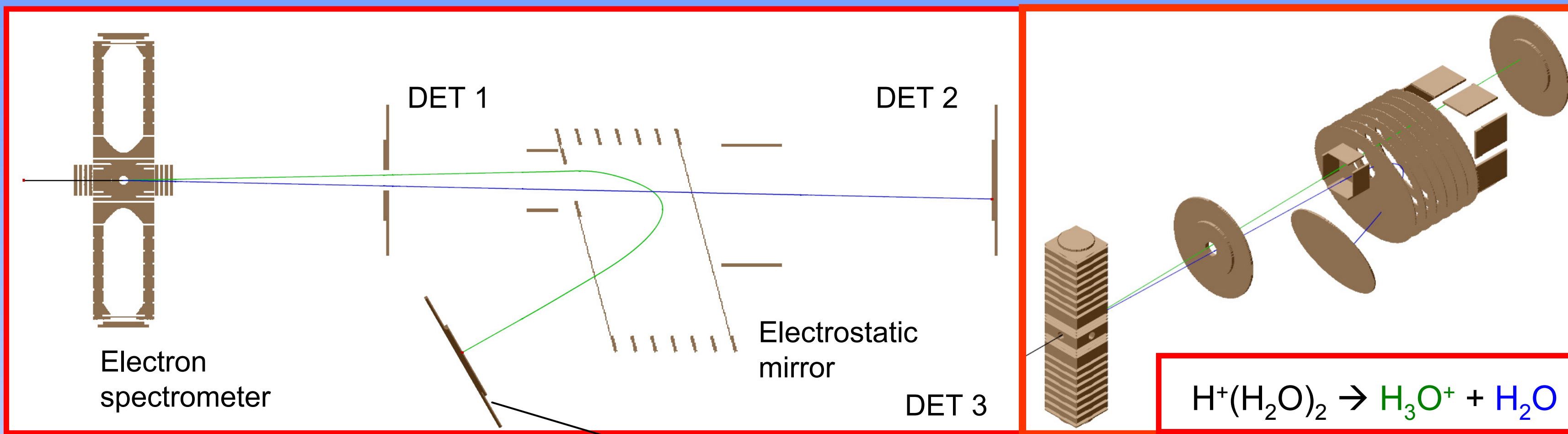
Major pathways identified

$\rightarrow OH + H^+ + H^+$	0.6	$0.37\pm0.18\cdot10^{-18}$ cm^2
$\rightarrow \left[H_2O^+\right]^* + H^+$	1	

*Pedersen et al. PRA **80**, 012707 (2009)/Lammich et al. PRL **105**, 253003 (2010)*

A more careful look at water cluster ions – August 2011

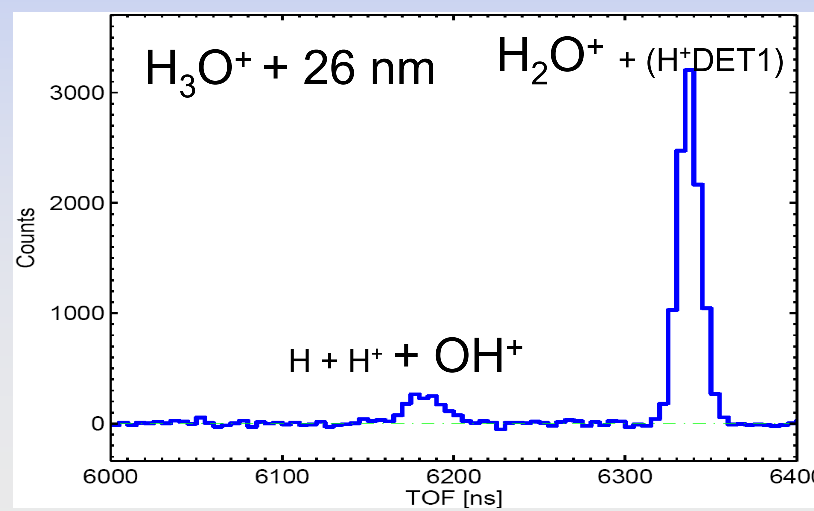
$H^+(H_2O)_n$ + 26nm (58 eV)



Achievements 2010-11

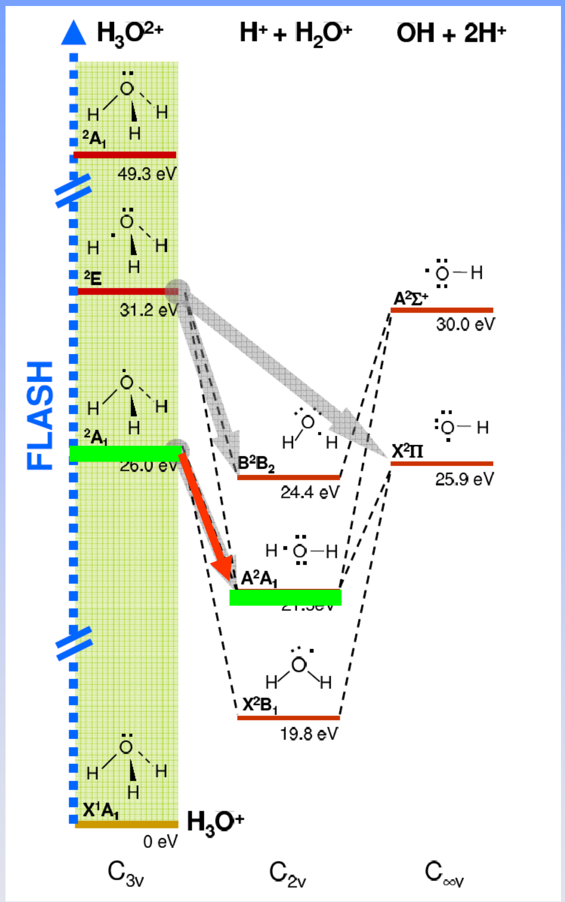
+ photoelectron spectroscopy
  + careful FEL guiding
  + careful stray light shielding
    → counting of electrons possible

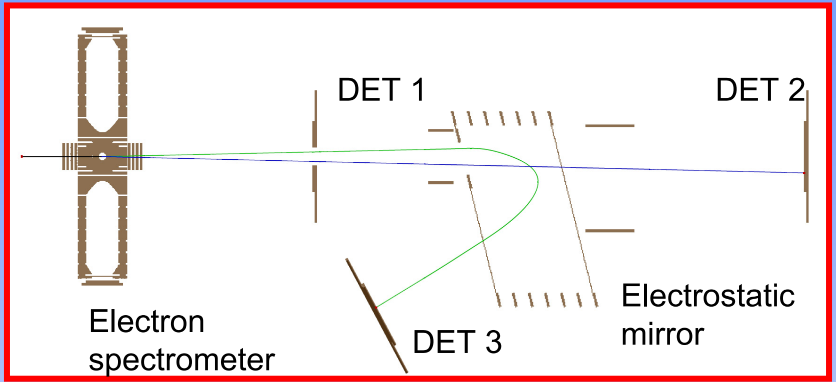
+ detection of all fragments (DET 3)
    → almost background free

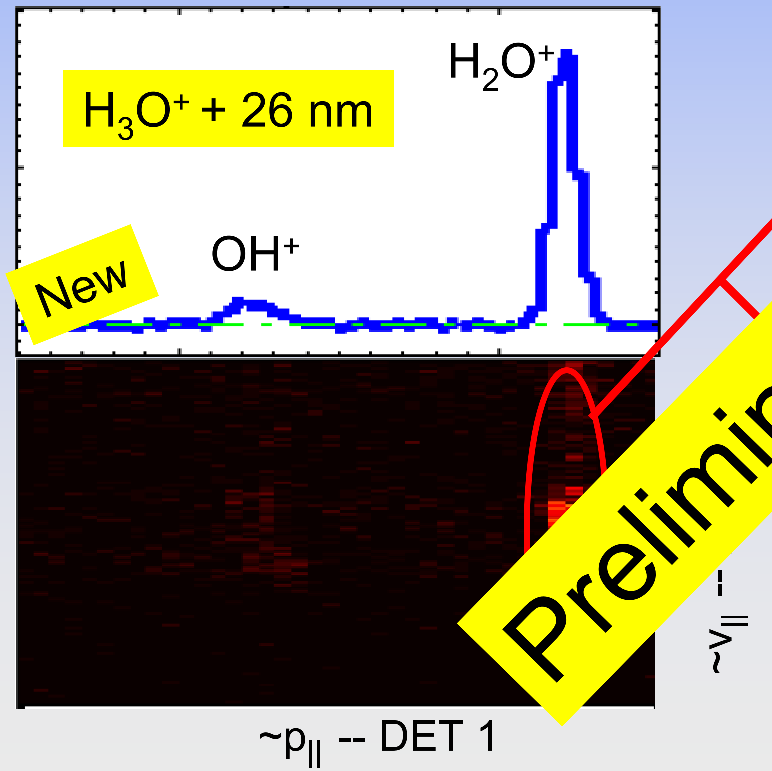


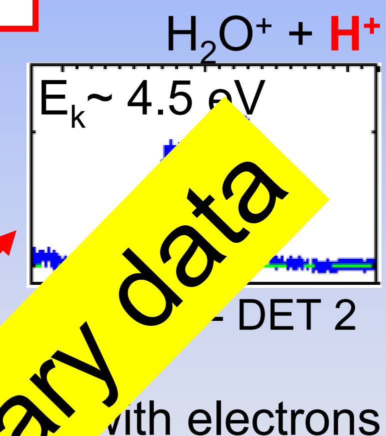
A more careful look at water cluster ion – August 2011

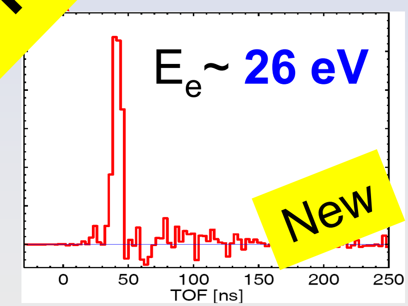
$H^+(H_2O)_n$ + 26nm (58 eV) – complete characterization of fragmentation





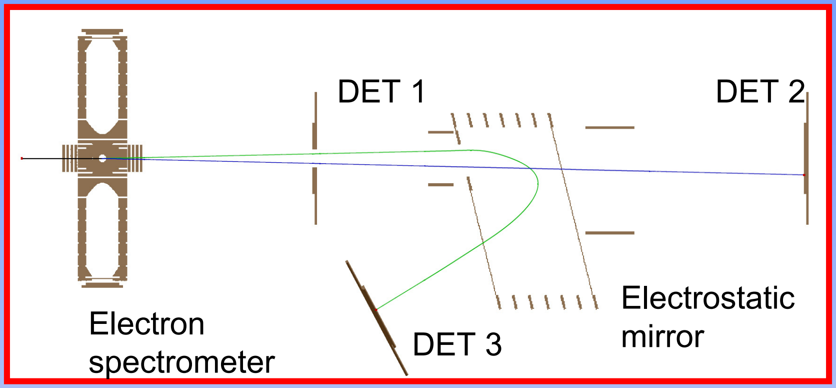






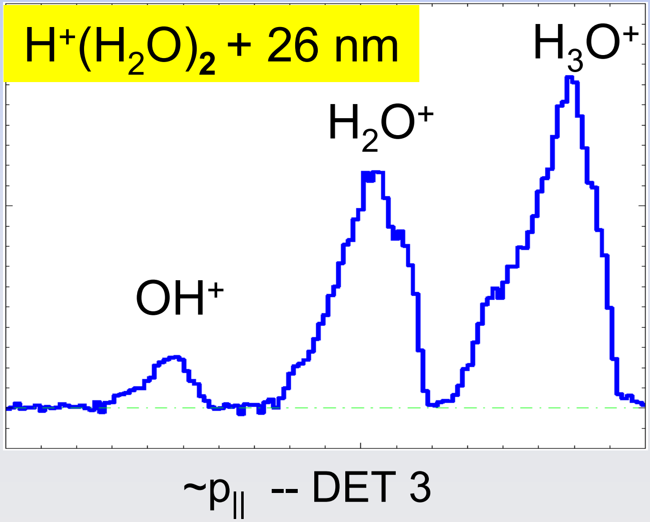
A more careful look at water cluster ions – August 2011

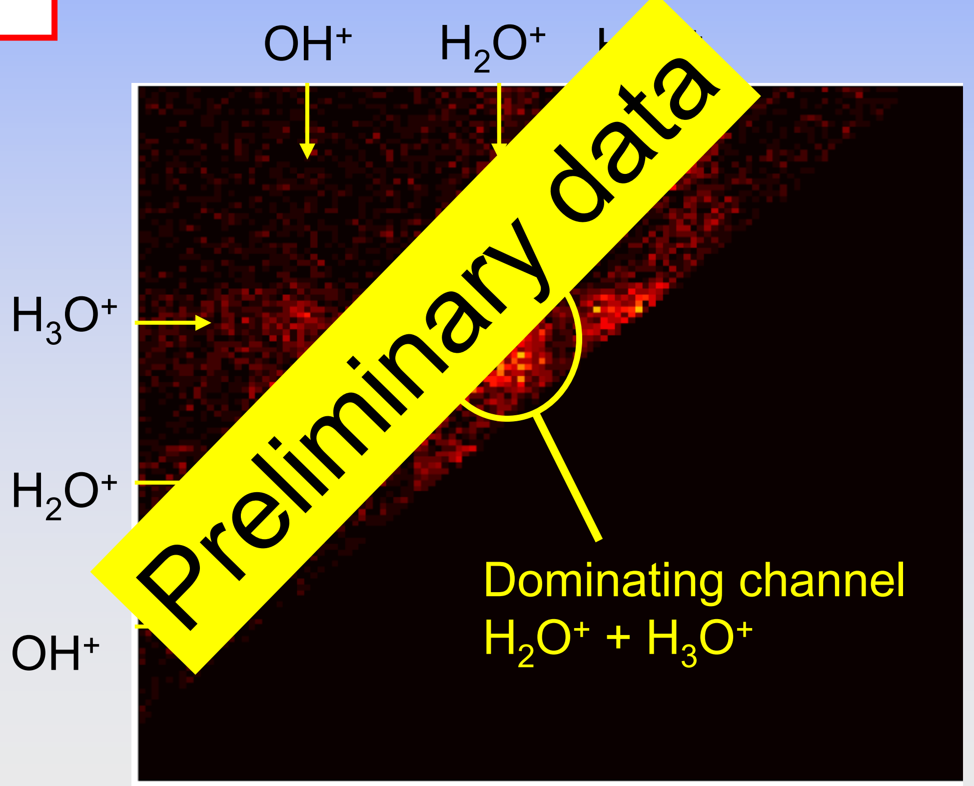
$H^+(H_2O)_n$ + 26nm (58 eV) – complete characterization of fragmentation



$H^+(H_2O)_\mathbf{2}$ + 26 nm

Analysing charged fragments



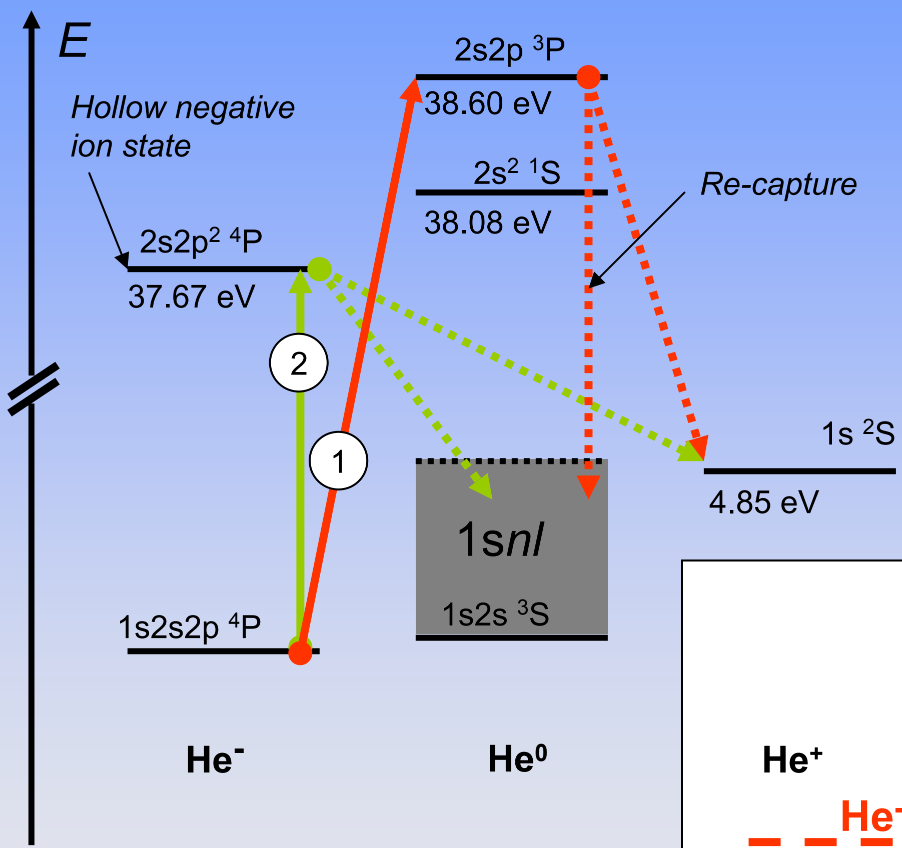


Perspectives for TIFF at FLASH

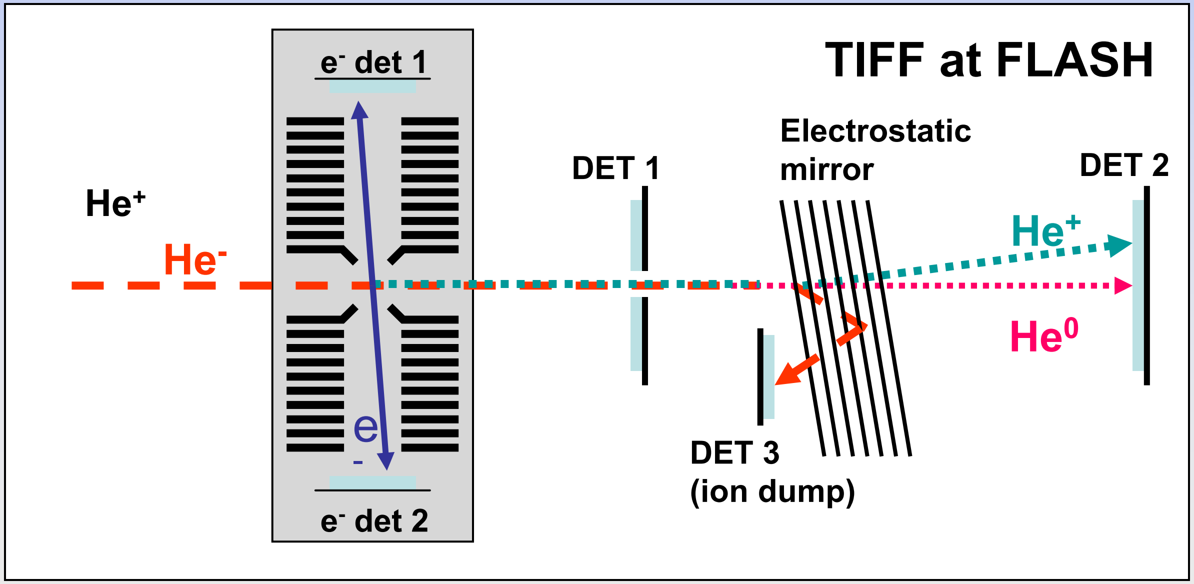
Anions
Di-cations
Molecular astrophysics, ex. hydrides

Perspectives for TIFF at FLASH

- Atomic anions (proposal 2011) + molecular anions (ex. vinylidene H_2CC^-)



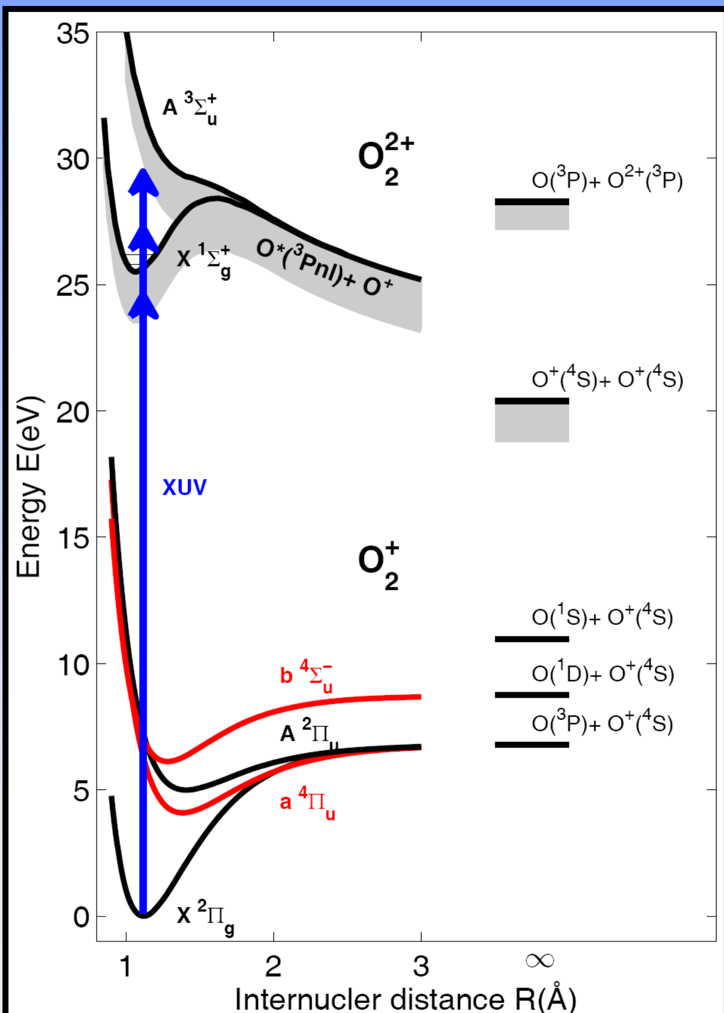
Dynamics of processes dominated by electron correlation studied in detail



N. Berrah et al. 2002,
R. Bilodeau et al. 2004

Perspectives for TIFF at FLASH

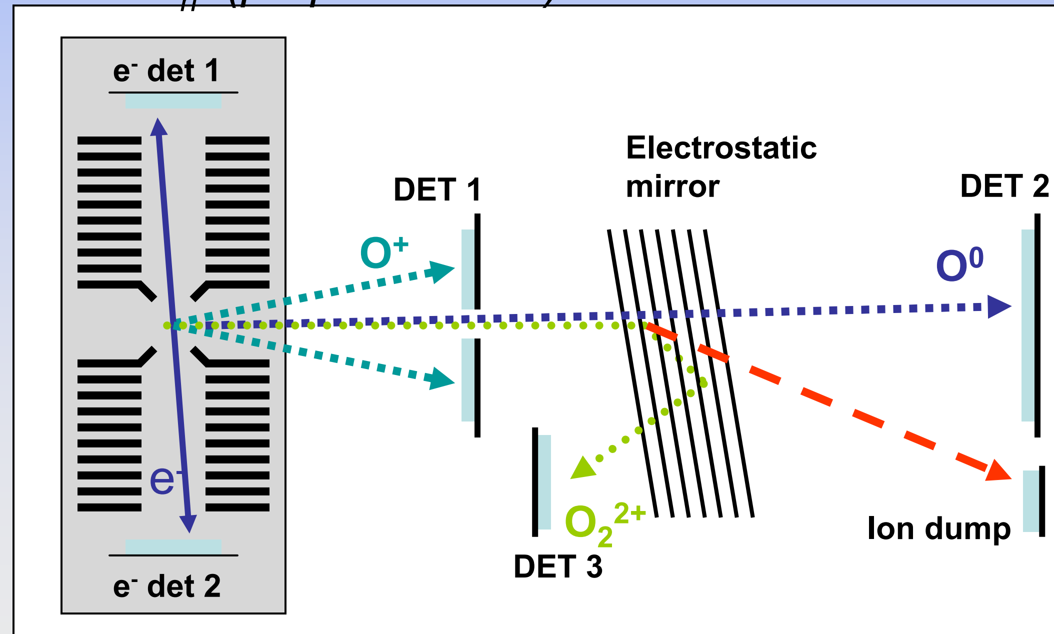
- Di-cations (proposal 2011)



Competion between excitation and ionization near di-cationic states

Link to multiphoton ionization starting from neutral O_2^+

Equivalent dynamics also in polyatomic systems ex. NH_n^+ (proposal 2011)



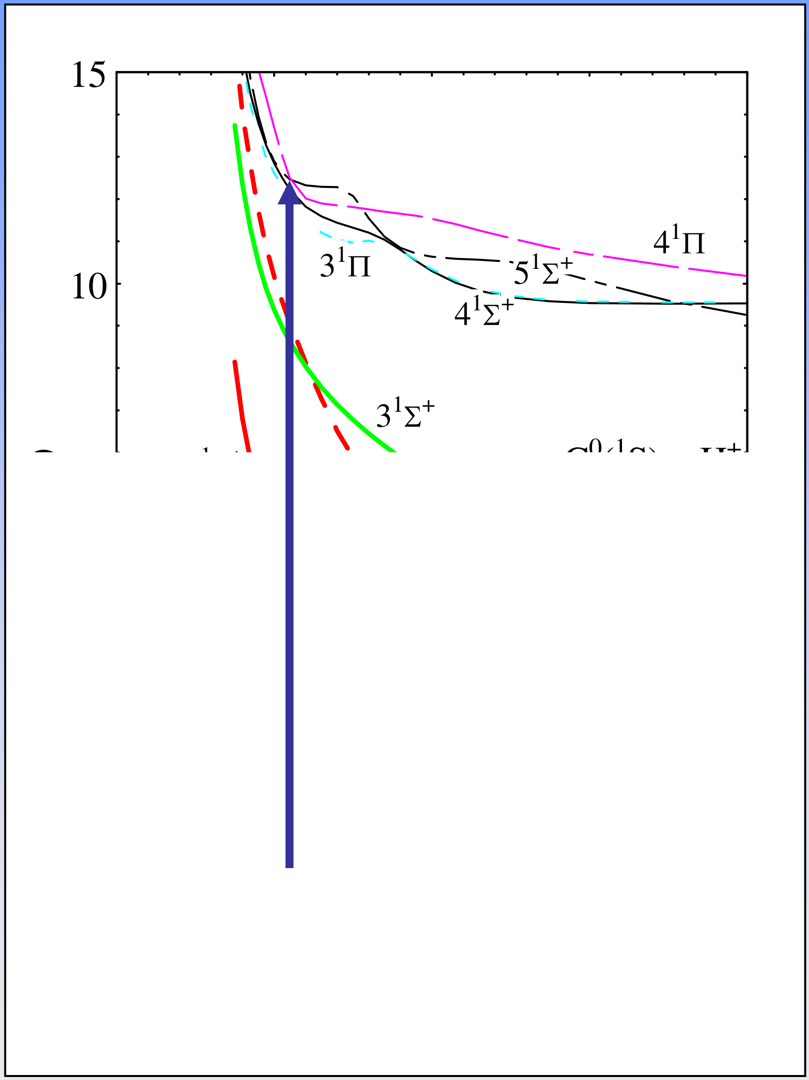
Perspectives / developments for FLASH and **FLASH II**

→ Experiments at longer wavelengths
→ Developments: Ion cooling in mini-storage ring

	FLASH	FLASH II
Wavelength	4 - 50 nm (~ 310-25 eV)	4 - 80 nm (310-15 eV)
Intensity	10^{12}-10^{14} photons/pulse	10^{12}-10^{14} photons/pulse
Pulse length	10-50 fs	<200 fs
Rep. rate	>50 pulses/train	>50 pulses/train
Wavelength scan	difficult	feasible

Perspective for FLASH II

Important systems absorb 10-20 eV, ex: **CH^+** - laboratory astrophysics



<u>Model chemistry of interstellar clouds CH^+</u>

Formation

$C^+ + H_2 \rightarrow CH^+ + H - 0.369$ eV

Destruction

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

$CH^+ + H_2 \rightarrow CH_2^+ + H$

$CH^+ + e^- \rightarrow C + H$

$CH^+ + VUV \rightarrow C^+ + H$

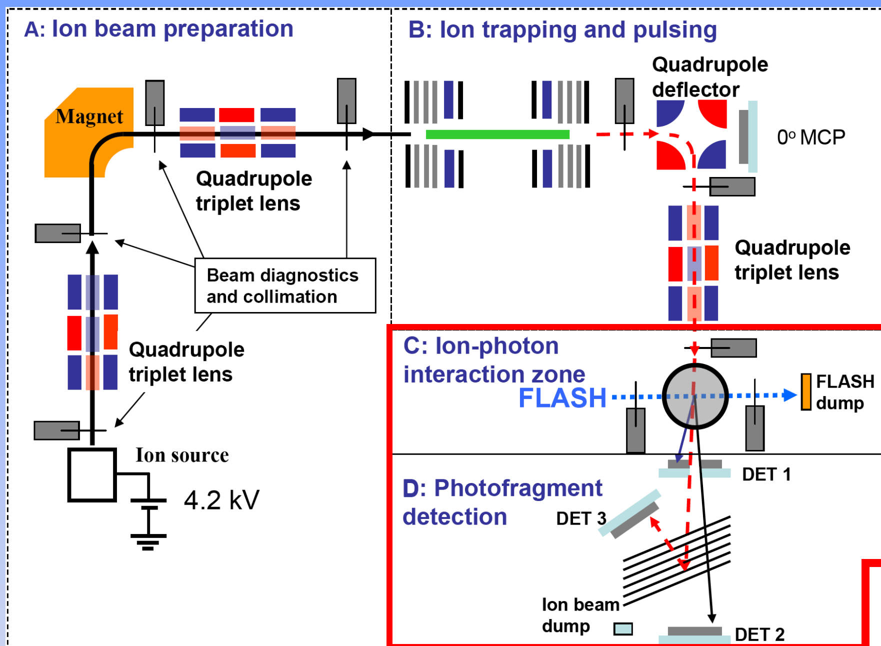
$C + H^+$

Is VUV photodissociation is a significant destruction mechanism for CH^+ in diffuse interstellar clouds ?

*Theory: Kirby et al.,ApJ **239**, 885 (1980)*

Perspective for FLASH + FLASH II

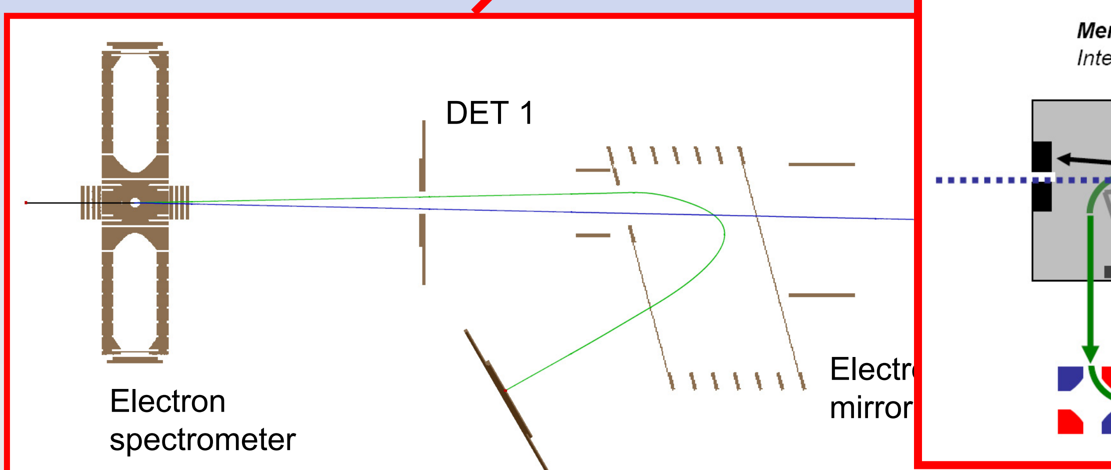
- Ion beam facility with integrated ion storage ring

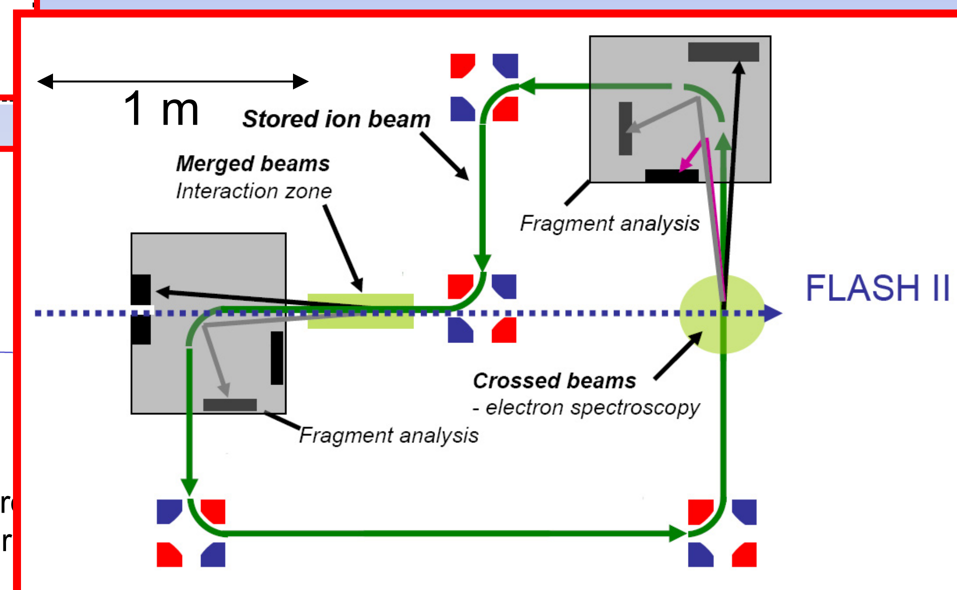


Cold ions:
Ion pulsing/extraction → storage ring

To take full advantage of the high rep. rate of FLASH and FLASH II experiments should be done directly on stored ions

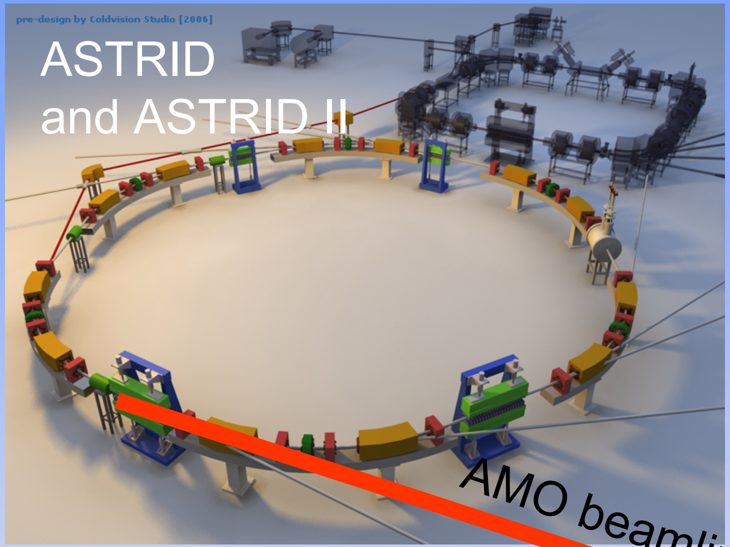
Crossed AND merged beams





Perspective for FLASH + FLASH II

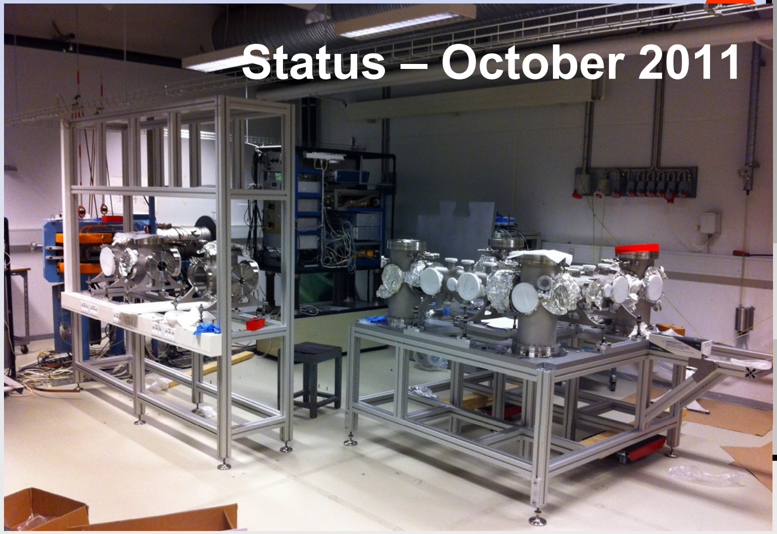
- an ion beam facility with storage ring under construction at ASTRID II

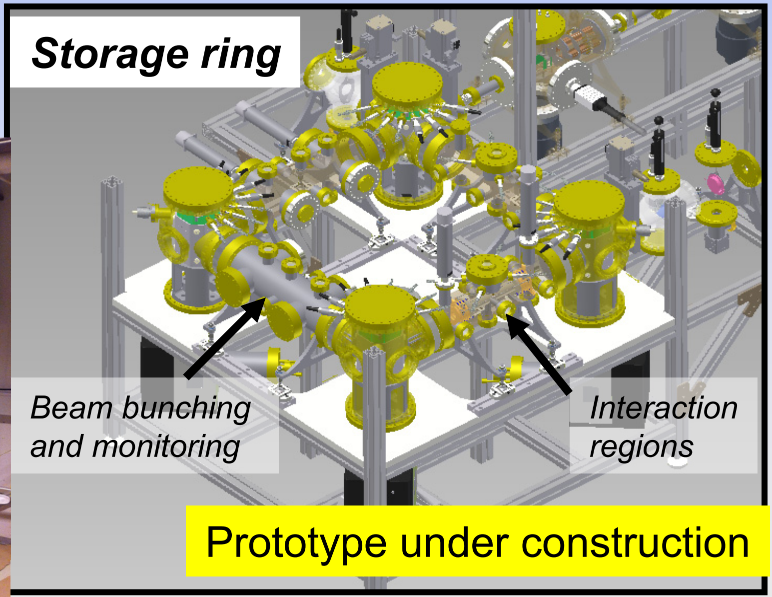


- ca. 5-250 eV
- 10^{15} photons/sec/0.1%BW

Complementary to FLASHs !

- **Small molecules**
- **Laboratory astrophysics**
- **Biophysics**

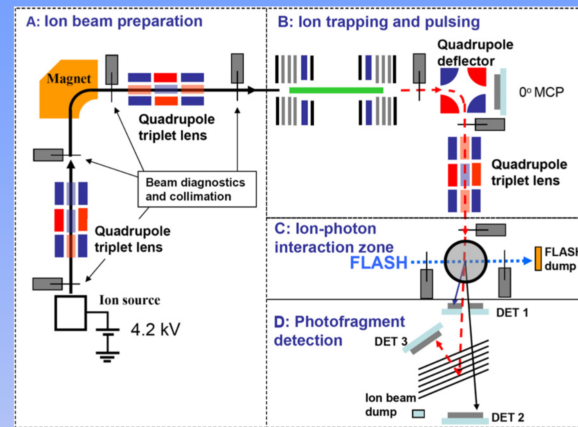


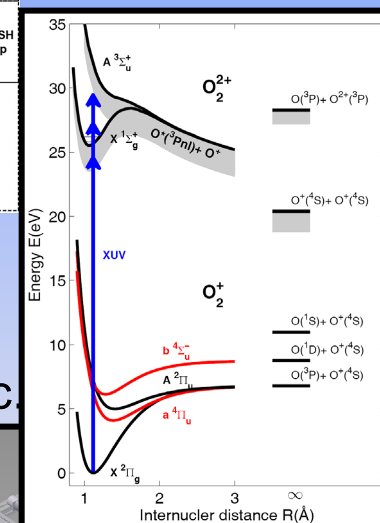


Summary

Achieved results from TIFF at FLASH

- Photodissociation
  HeH^+
- Photoionization + dissociation :
  Water cluster $H^+(H2O)_n$
  + photoelectrons
  + all fragments





Perspectives

- Several upcoming possibilities at FLASH (anions, di-cations, etc.
- FLASH II – opens the *long* (..80 nm) wavelength region
  ex. CH^+
- A new ion beam facility should include a storage ring to
  take full advantage of the high rep. rate of FLASH II