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Low energy ionization, charge transfer and reactive collisions

for ion source and edge plasma chemistry

X. Urbain

Experimentalists' Network Meeting IAEA November 2018

Merged Ion Beams

- Low temperature & high resolution
- Diatomic systems
- Triatomic systems

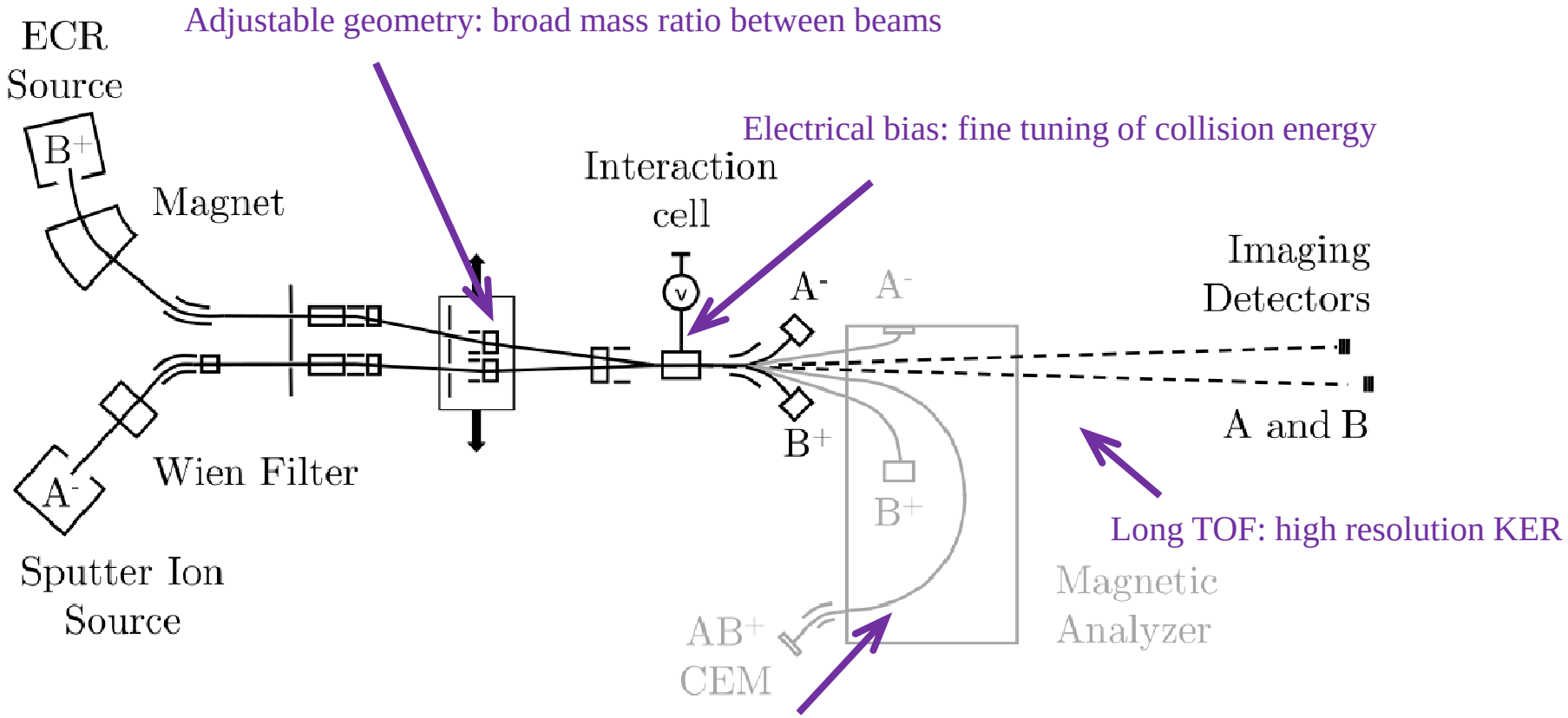
Crossed Electron-Ion Beams

- Animated beam method
- Excited atoms
- Molecular ions

Ion Beam Gas Target Measurements

- Deceleration
- Vibrational diagnostics

MERGED BEAMS SET-UP



For a review on merged beams: Phaneuf *et al.* (1999) *Reports on Progress in Physics*, *62*, 1143

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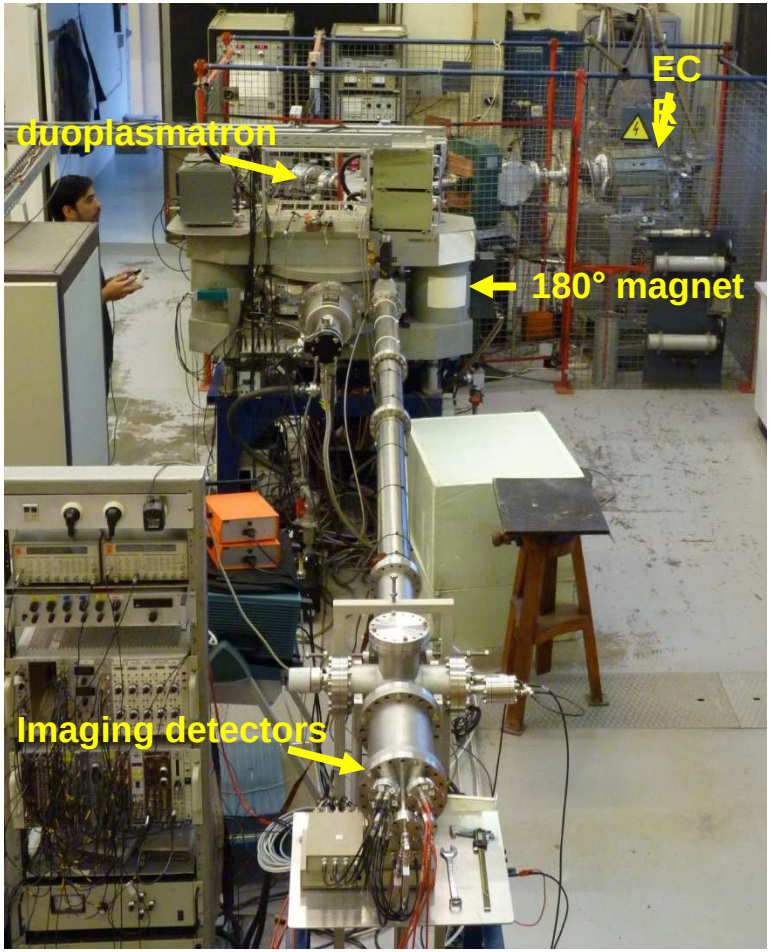
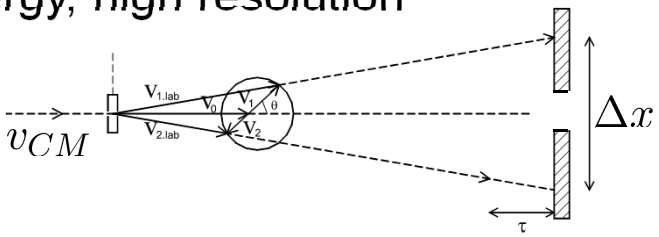
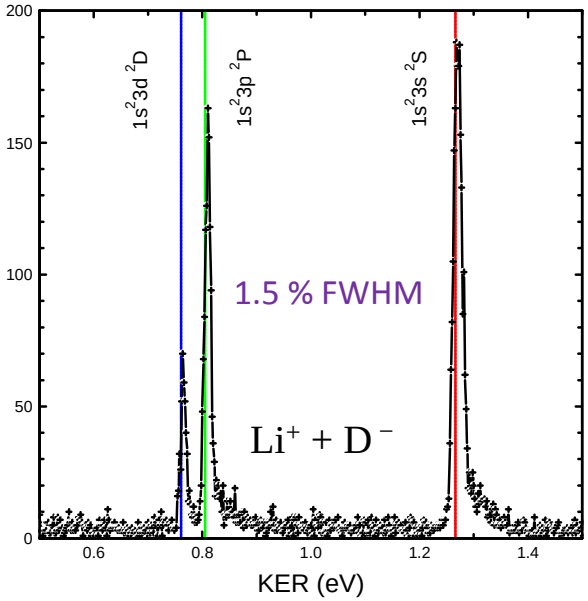
MERGED BEAM SET-UP : low collision energy, high resolution



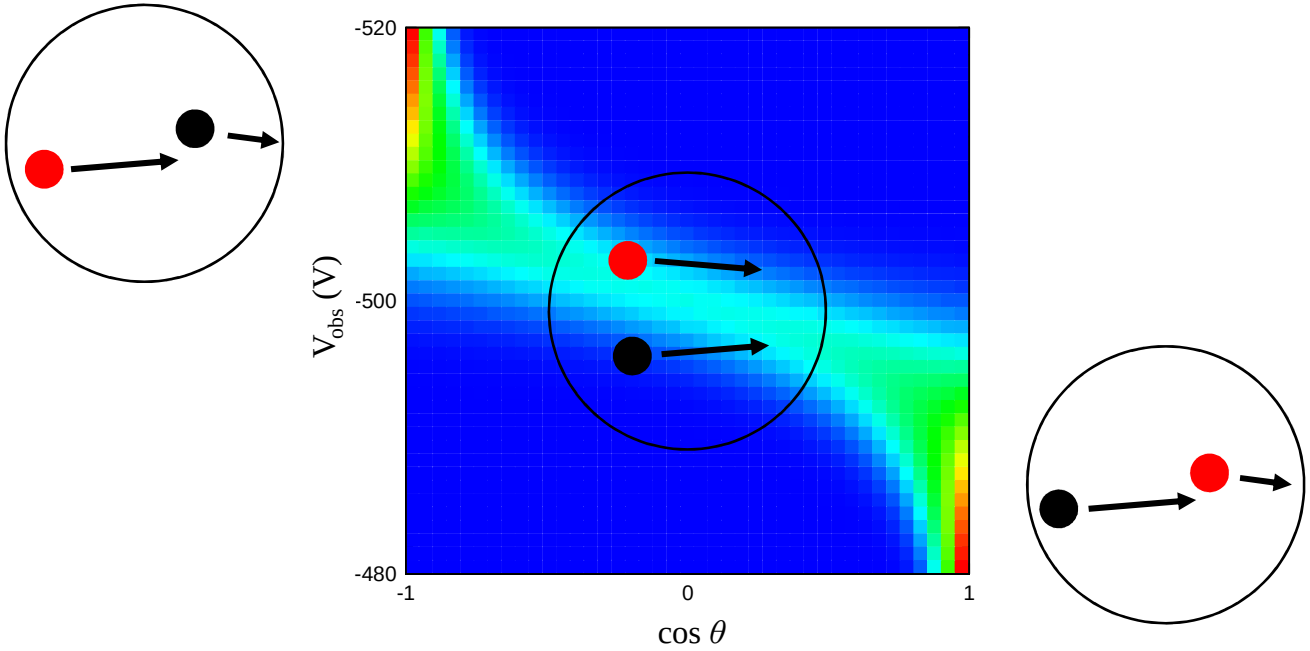


$$KER = \frac{E}{4L^2}\left[\Delta x^2 + v_{CM}^2 \Delta t^2\right]$$

$$L = 320 \pm 4(2)\ \text{cm}$$



(AN)ISOTROPY : The collision axis flips by π when moving across zero detuning



ENERGY RESOLUTION: longitudinal and transverse temperature // e$^-$-ion collisions

$$f(v_d,\vec{v}) = \frac{m}{2\pi k T_\perp}\sqrt{\frac{m}{2\pi k T_{||}}}\exp\left[-\frac{m v_\perp^2}{2\pi k T_\perp} - \frac{m(v_{||}-v_d)^2}{2\pi k T_{||}}\right],\ v_{||} \simeq |v|(1 \pm \Delta E/2E)\,,\ v_\perp \simeq |v|\sin\theta$$

12 keV C^+ and C^- beams, 5 eV dispersion, 1 mrad angular spread: $T_{||} \simeq 8\,K\,,\quad T_\perp \simeq 70\,K$

Low energy anion-cation reactions

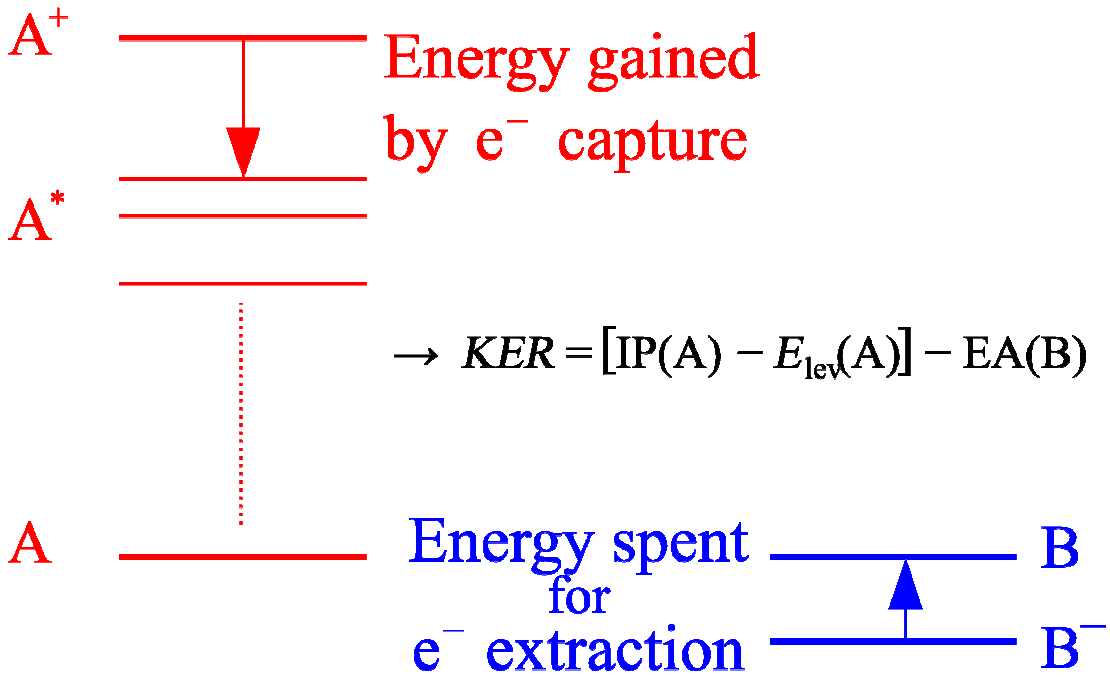
$A^+ + B^- \rightarrow A^* + B$	Mutual Neutralization (MN)
$\rightarrow A + B^+ + e$	Transfer Ionization (TI)
$\rightarrow A^- + B^+$	Charge Exchange
$\rightarrow AB^+ + e$	Associative Ionization (AI)

e.g. $H_2^+ + O^-$ Le Padellec *et al.*, J. Chem. Phys. **124**, 154304 2006

Additional reactions in polyatomic systems

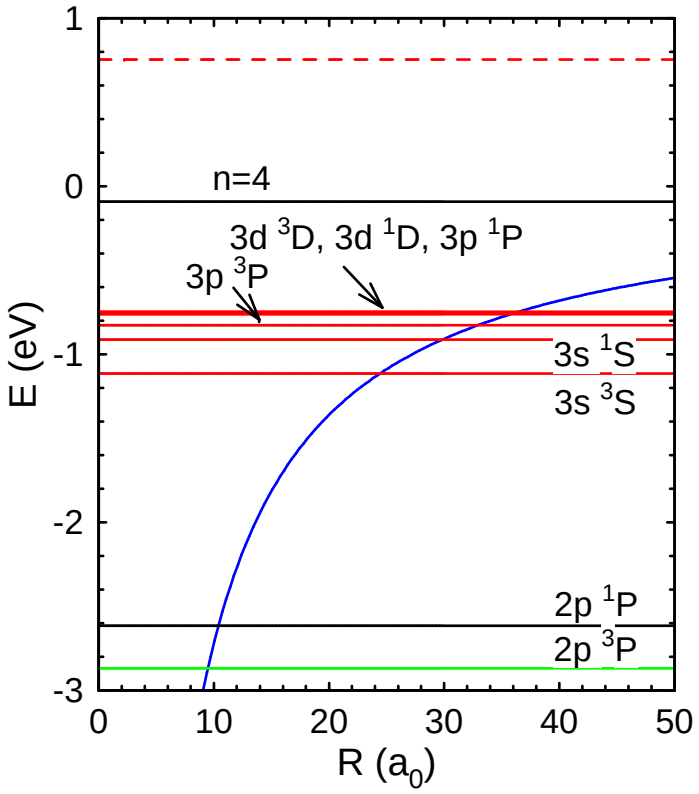
$AB^+ + C^- \rightarrow AC^* + B$	Reactive Neutralization
$\rightarrow AC^+ + B + e$	Reactive Ionization (RI)
$\rightarrow ABC + h\nu$	Radiative Association

$A^+ + B^- \rightarrow A^* + B + \text{Kinetic Energy Release}$

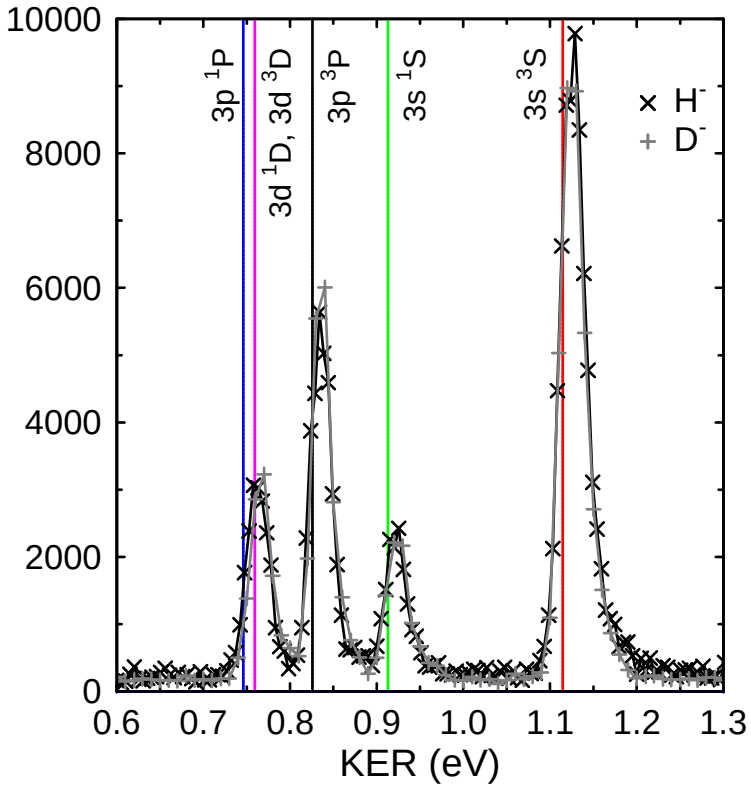


KER SPECTRUM: Partial cross sections e.g. $He^+ + H^- \rightarrow He\ (1snl\ ^{1,3}L) + H$

kinetic energy release → state identification

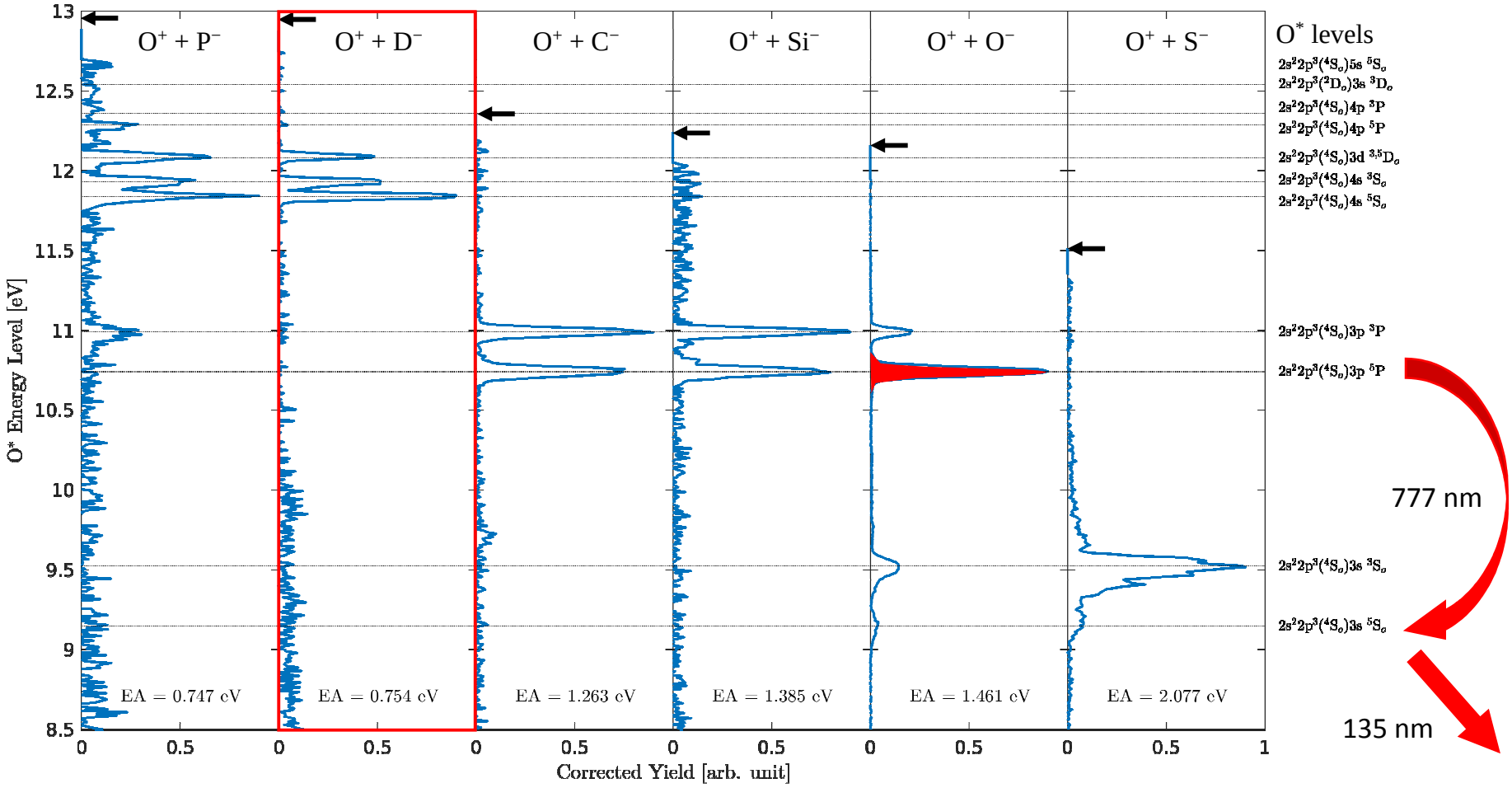


relative peak area → branching ratio



Theory: Larson *et al*. (2016) *Phys. Rev.* A **94**, 022709

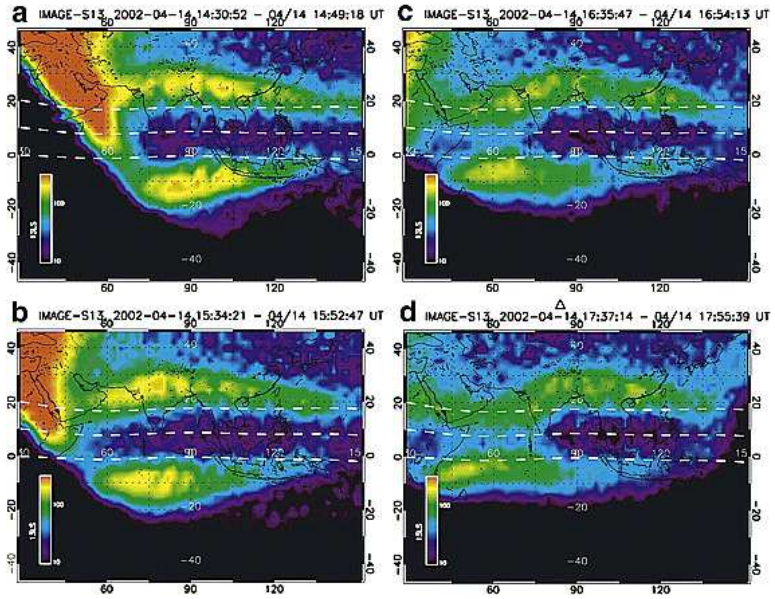
$O^+ + X^-$ $E_{level} = IP\,(A) - EA(B) - KER$ N. de Ruette *et al*. (2018) Phys. Rev. Letters **121**, 083401



Negative Ions in Earth's ionosphere: UV tropical nightglow of O I (135 nm)

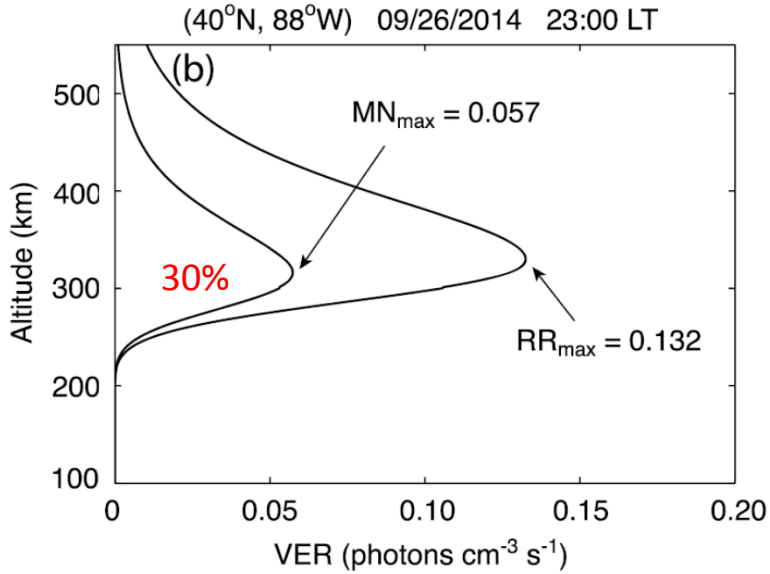
$$O^+ + O^- \rightarrow O\,(^5P) + O$$

R. E. Olson, J. R. Peterson, and J. Moseley (1970) *Geophys. Res.*, 76, 2516, 197



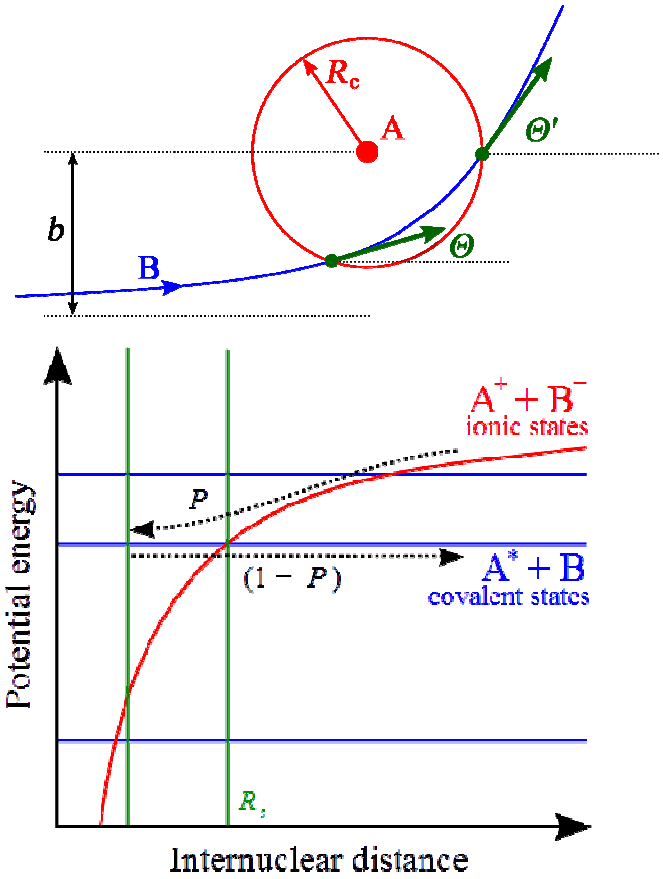
Longitudinal structure of the equatorial anomaly in the nighttime ionosphere observed by IMAGE/FUV

E. Sagawa *et al.* (2005) *J. Geophys. Res.*, 110, A11302



J. Qin et al. (2015) *J. Geophys. Res. Space Physics*, 120, 10116

Multistate Curve Crossing Model: electron 'hops' from B to A^+



- Total transfer probability (*Landau-Zener*)

$$\sim 2P(1-P)$$

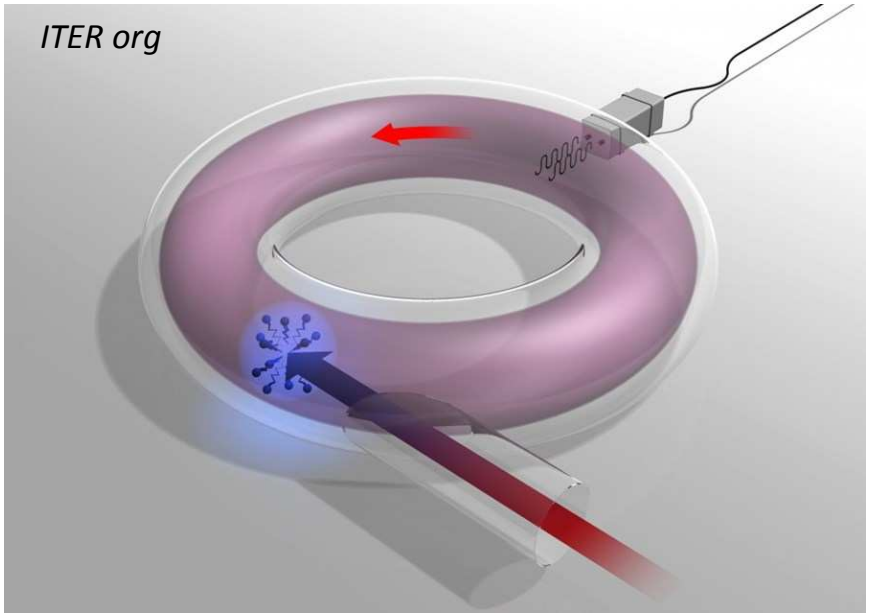
$$P \sim 1 - \exp\left\{-\frac{2\pi H_{12}^2}{v_c\, a}\right\}$$

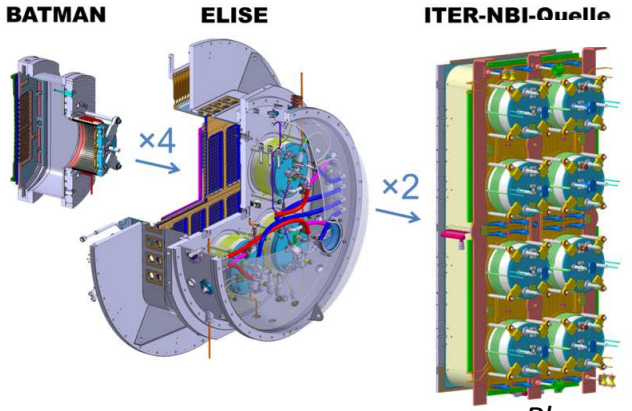
- H_{12} matrix element evaluated using Landau-Herring asymptotic method

Chibisov and Janev (1988) *Phys. Rep.* **166**, 1

→ transition probability window that mostly depends on covalent state potential energy

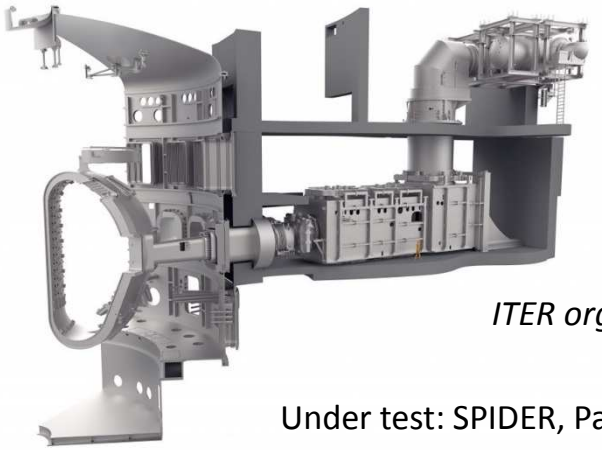
Negative Ions in Fusion Research





MPI Plasmaphysik

extraction area of 1.9 × 0.9 m^2
ion current of 40 A, heating power 16.5 MW



ITER org

Under test: SPIDER, Padua, Italy

High energy neutral beams: negative ions preferred

electron stripping >> electron capture at E > 100 keV

D^- yield limited by volume recombination (MN) with D^+/D_2^+

U. Fantz, P. Franzen, D. Wünderlich (2012) *Chem. Phys.* **398**, 7

NEGATIVE ION SOURCES: destruction of H^- by MN with parent cation

$H_2 + e^- \rightarrow H_2(B,C) + e^- \rightarrow H_2(v) + photon$

$H_2(v) + e^- \rightarrow H + H^-$ dissociative electron attachment (DEA)

$H_2^+ + H^- \rightarrow H_2^{(*)} + H^{(*)}$ **mutual neutralization** (MN) *merged beams*

$H^+ + H^- \rightarrow H + H^*$

DETACHED PLASMA: Molecular assisted recombination

$H_2\,(v) + H^+ \rightarrow H_2^+(v) + H$ **charge transfer** *decelerated beam*

Urbain *et al.*(2013) *Phys. Rev. Letters* **111**, 203201 (H_2(v=0))

$H_2^+(v) + e^- \rightarrow H + H^*$ dissociative recombination *storage ring*

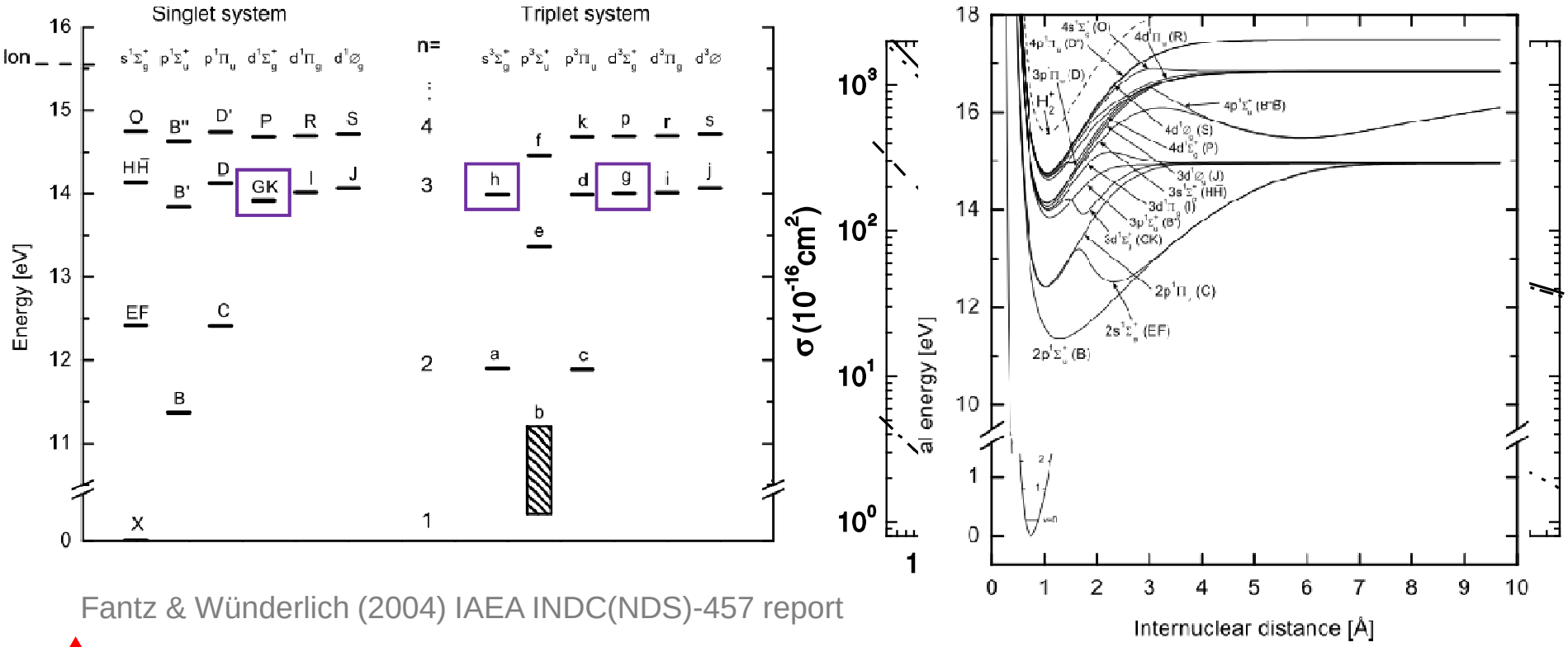
Amitay *et al.*(1998) *Science* **281**, 75 & (1999) *Phys. Rev. A* **60**, 3769 (HD^+)

$H_2^+ + H^-$

Calculations: Landau-Herring asymptotic method

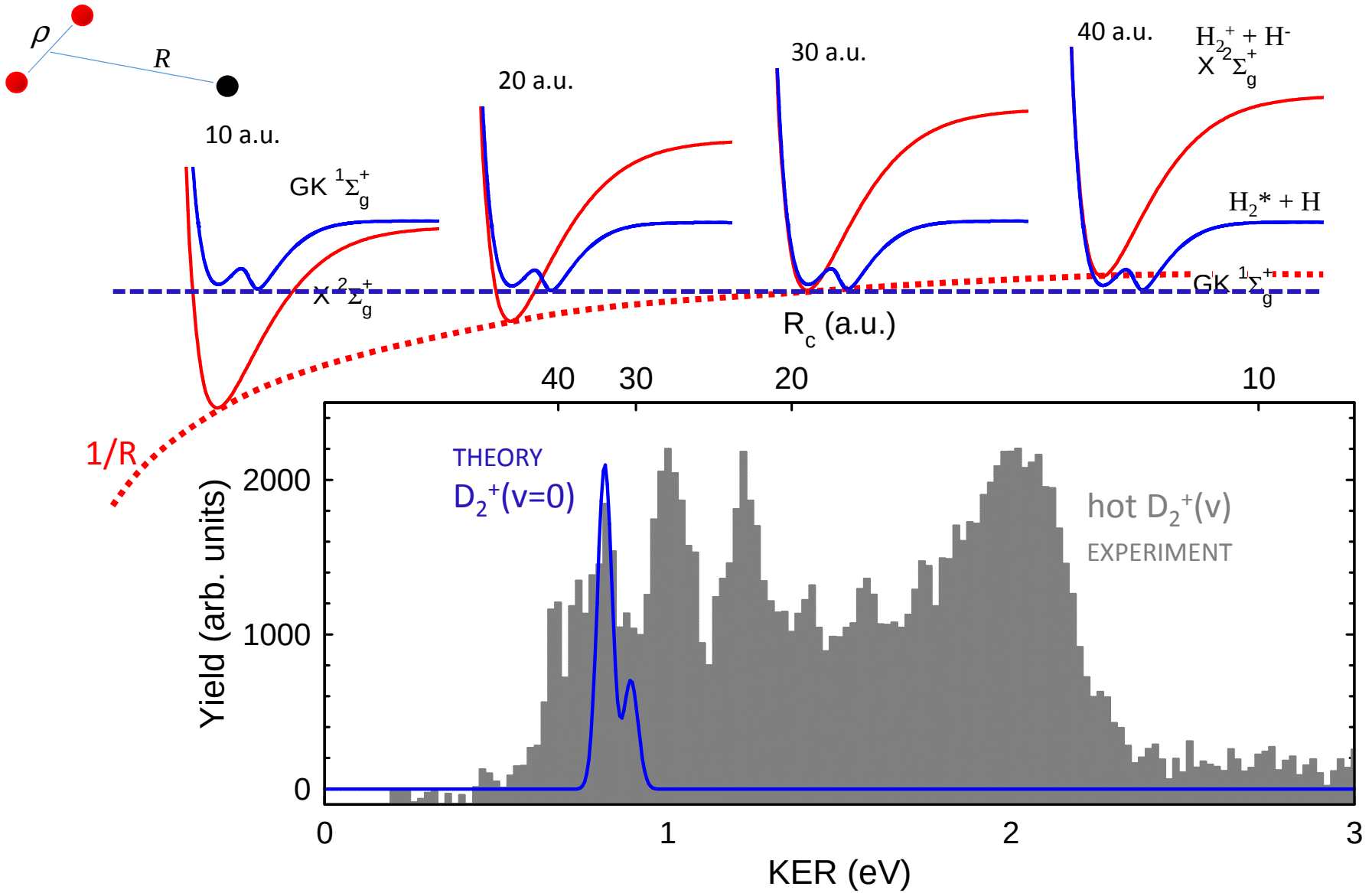
C. L. Liu, J. G. Wang, and R. K. Janev (2006) *J. Phys. B: At. Mol. Opt. Phys*. 39, 1223

Atom-like description: electron capture to Rydberg state of H_2 – diagonal FCF



Fantz & Wünderlich (2004) IAEA INDC(NDS)-457 report

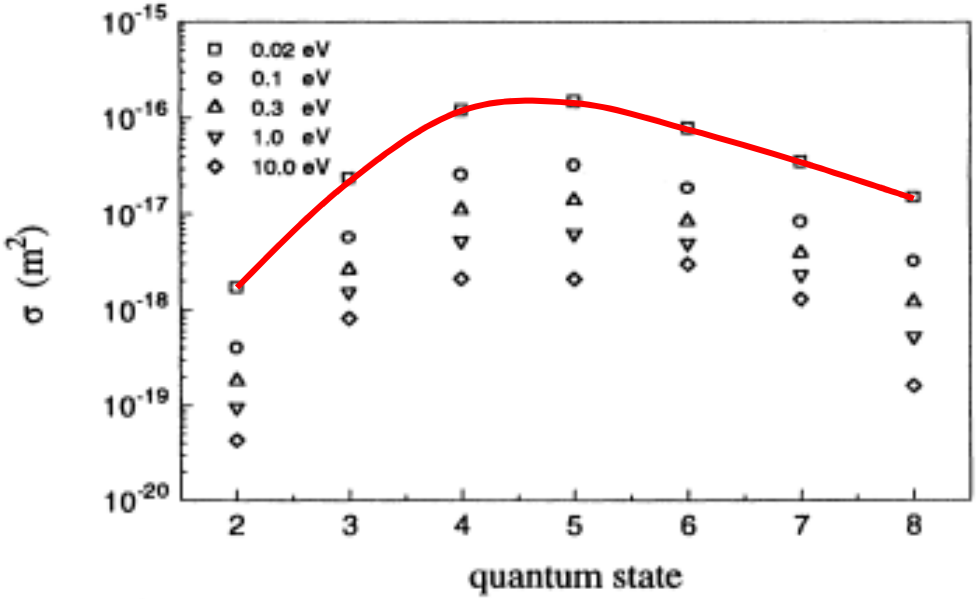
Restriction to Σ_g for collinear geometry ($D_{\infty h}$) – Σ_g, Π_u, Δ_g for T-shape geometry (C_{2v})

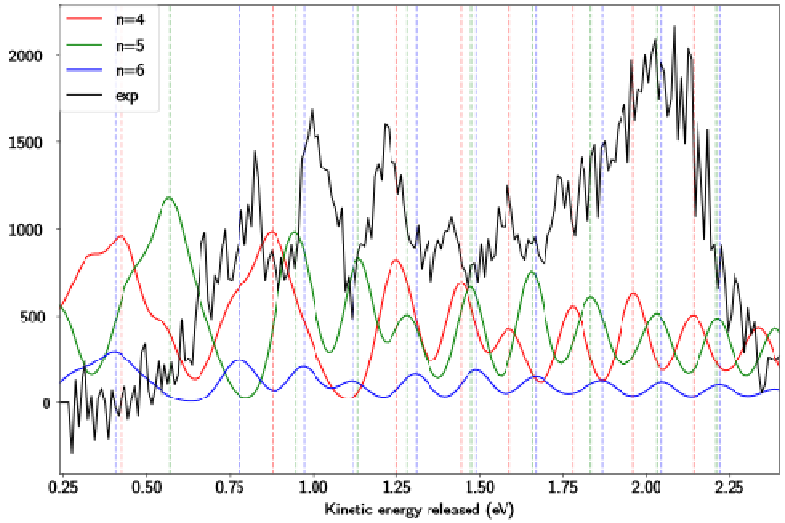

ρ
R
10 a.u.
20 a.u.
30 a.u.
40 a.u.
H2+ + H-
X 2Σg+
GK 1Σg+
H2* + H
GK 1Σg+
X 2Σg+
Rc (a.u.)
40
30
20
10
1/R
THEORY
D2+(v=0)
hot D2+(v)
EXPERIMENT
Yield (arb. units)
2000
1000
0
0
1
2
3
KER (eV)

$H_2^+ + H^-$ Calculations under assumption: $H_2^+ + H^- \rightarrow H_2 + H(n)$

M. J. J. Eerden, M. C. M. van de Sanden, D. K. Ortobaev & D. .C. Schram (1995)
Phys. Rev. A **51**, 3362

Predicted propensity towards n=5





No evidence for direct atomic excitation in our data

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Merged Ion Beams

- Low temperature & high resolution
- Diatomic systems
- Triatomic systems

Crossed Electron-Ion Beams

- Animated beam method
- Excited atoms
- Molecular ions

Ion Beam Gas Target Measurements

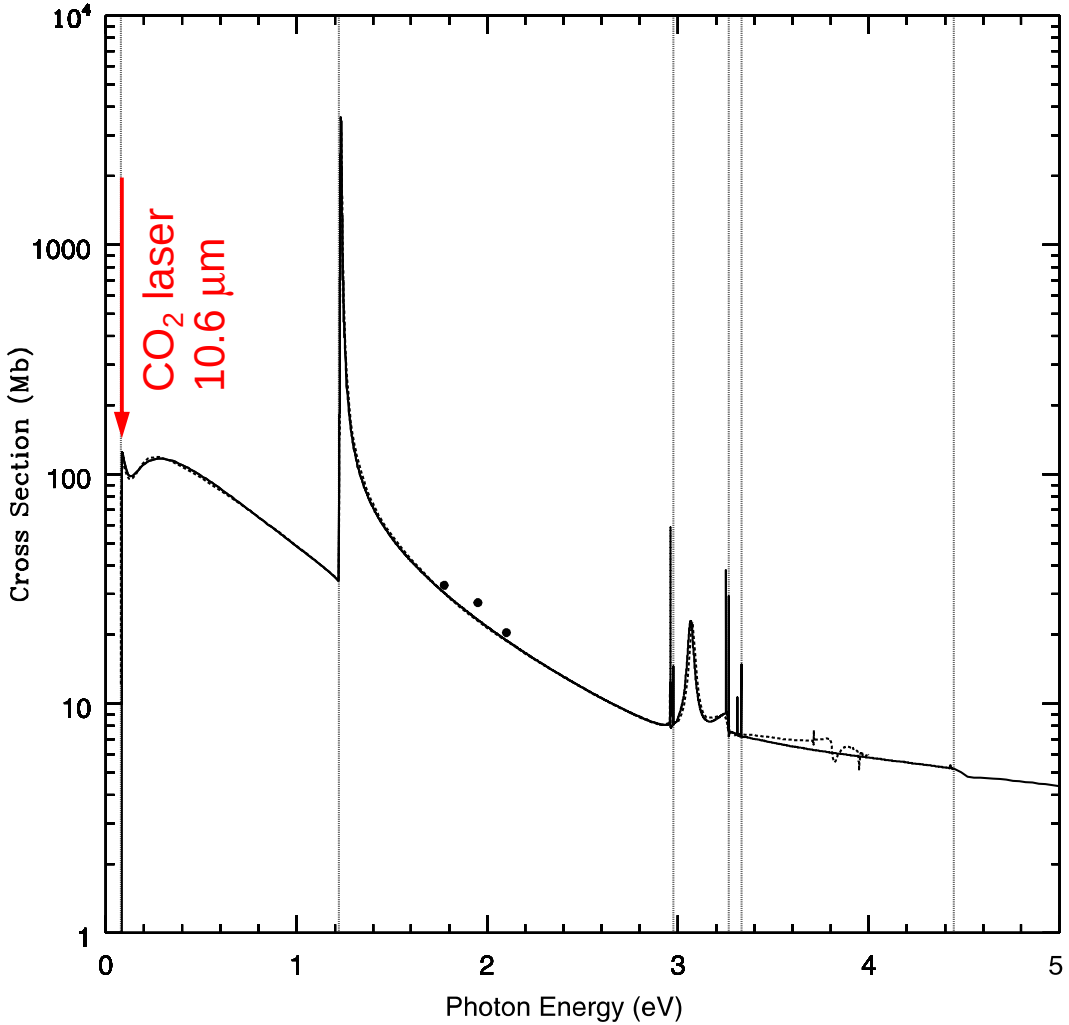
- Deceleration
- Vibrational diagnostics

Electron impact ionization of excited helium atoms

How to get a pure He(1s2s ^{3}S) beam?

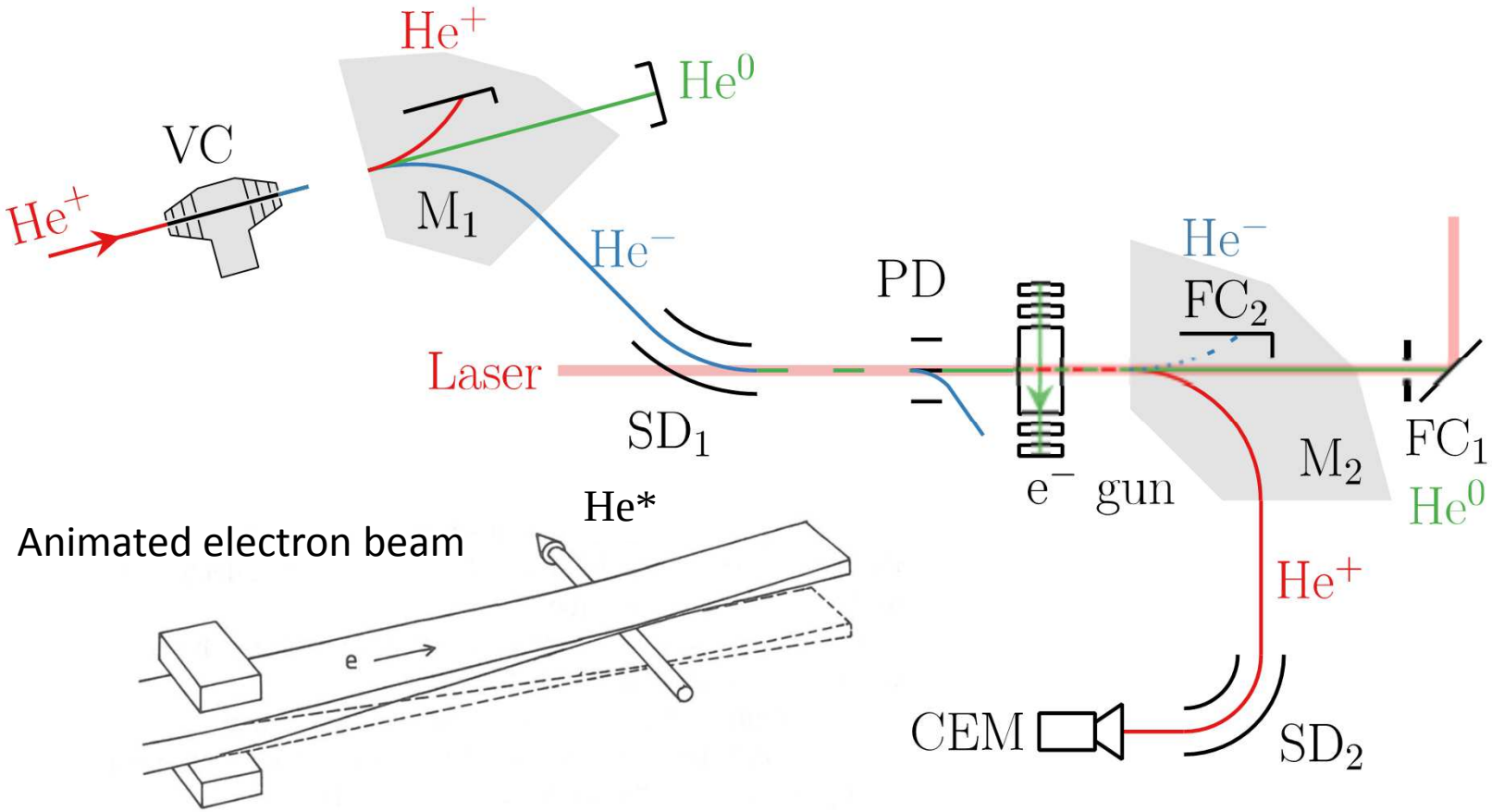
1) $He^+ + Na \rightarrow He^-$ (1s2s2p ^{4}P)

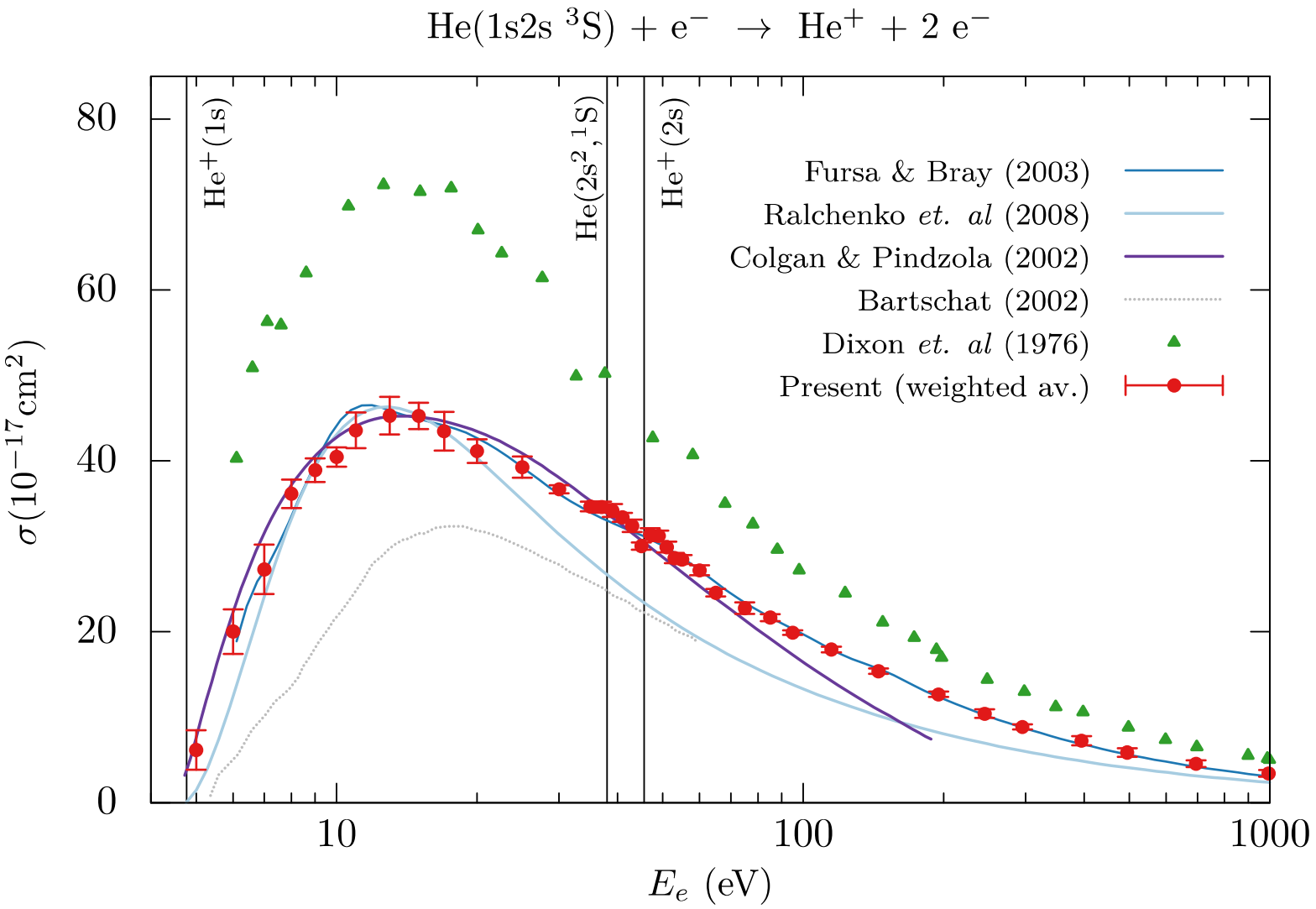
2) He^-(1s2s2p ^{4}P) + hν
→ He (1s2s ^{3}S) + e^- : 50% efficiency!



New beam line on crossed ion-electron beam setup

J. Lecointre, D. S. Belic, H. Cherkani-Hassani, J. J. Jureta, and P. Defrance (2006) J. Phys. B **39**, 3275





M. Génévriez, J. J. Jureta, P. Defrance, and X. Urbain (2017) Phys. Rev. A **96**, 010701(R)

Dissociative excitation and ionization of molecular helium ions by electron impact

Helium operation

symmetric collisions: rapid thermalization

$He^* + He^{++}/He^+/He/He^* \rightarrow He^+ + He^{++}/He^+/He/He^*$

molecular ions (divertor region): well-known in technical plasmas

$He^* + He^* \rightarrow He_2^+ + e^-$

$He + He + He^+ \rightarrow He_2^+ + He$

$\rightarrow$ **molecular ions appear in denser, colder regions**

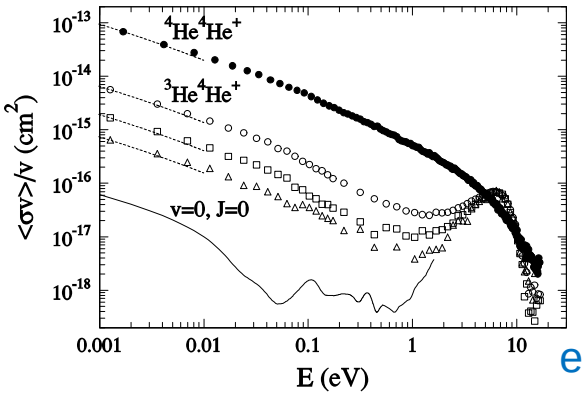
+ destruction mechanisms

dissociative recombination with electrons

$He_2^+ + e \rightarrow He + He$ *extremely sensitive to plasma temperature and formation mechanism*

Pedersen et al. (2005) Phys. Rev. A **72**, 012712

X. Urbain *et al*. (2005) J. Phys. B: At. Mol. Opt. Phys. **38**, 43



electron impact dissociation/ionization

$He_2^+ + e \rightarrow He^+ + He + e$ *unknown*

Merged Ion Beams

- Low temperature & high resolution
- Diatomic systems
- Triatomic systems

Crossed Electron-Ion Beams

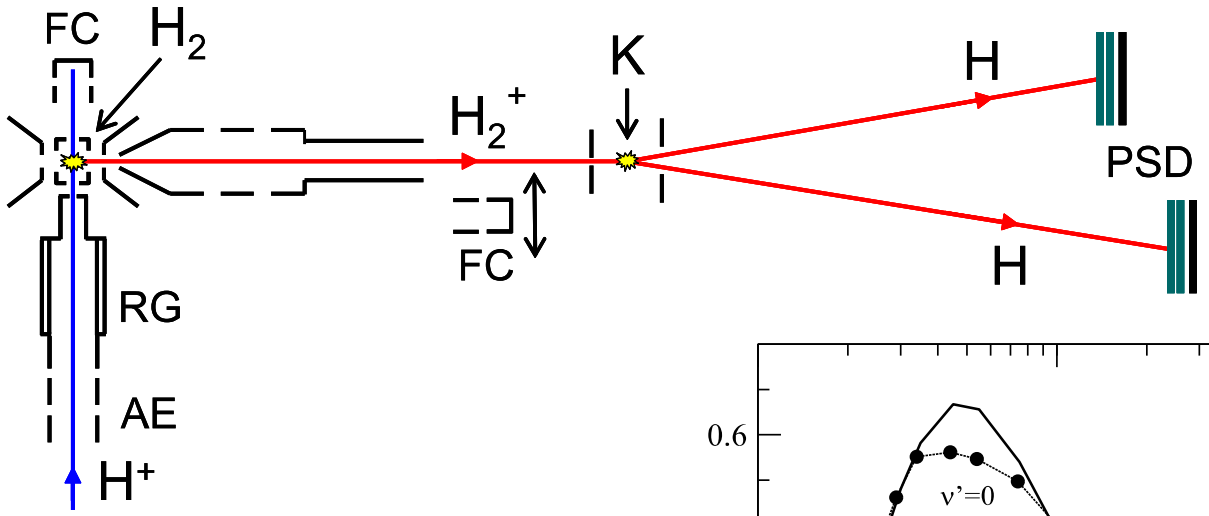
- Animated beam method
- Excited atoms
- Molecular ions

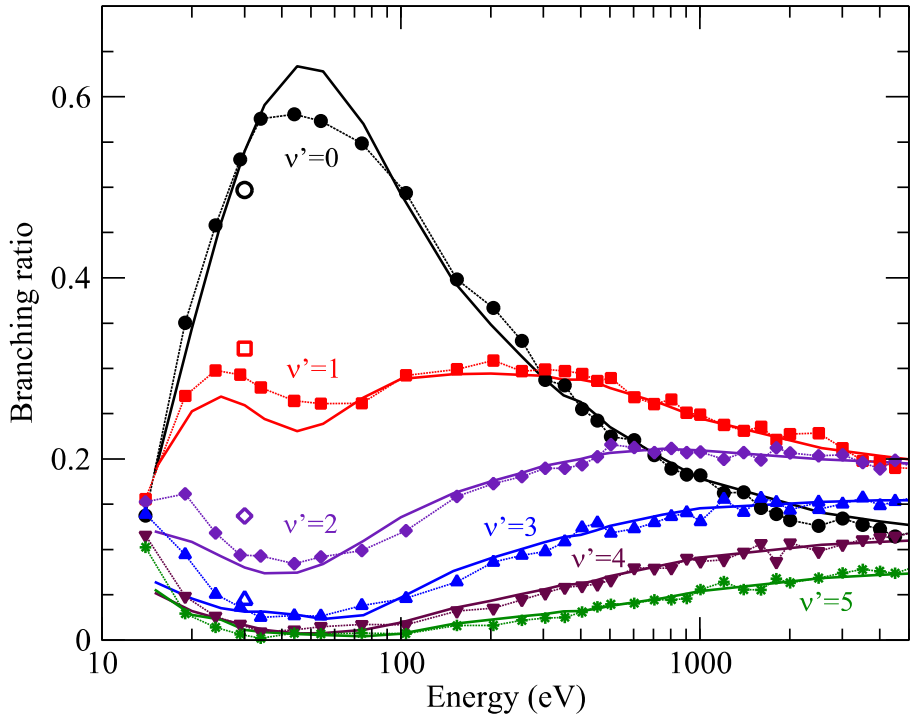
Ion Beam Gas Target Measurements

- Deceleration
- Vibrational diagnostics

Vibrationally resolved charge transfer between slow ions and molecules

Urbain et al. (2013) Phys. Rev. Letters 111, 203201





SUMMARY

Merged Ion Beams

- Full resolution of initial and final states through 3D imaging
- Preliminary results of state-resolved differential cross sections
- total cross sections measurements affected by excessive background

Crossed Electron-Ion Beams

- Animated beam method perfectly suited for absolute measurements
- Excited atoms from photodetachment ($O(^1D)$ in preparation)
- Molecular ions with known vibrational excitation needed

Ion Beam Gas Target Measurements

- Deceleration 10 eV and above (guided beams under study)
- Vibrational diagnostics: works for a few diatomics (H_2,O_2, HeH)

  more general if Rydberg target, as tested with CO and N_2

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