Electron-cation and atom-molecule reactive collisions in the cold regions of the fusion plasmas

Ioan F. Schneider
Université du Havre, LOMC-UMR-6294
Visitor at Université Paris-Sud, LAC-UPR-3321

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Results of / collaboration with:

F. O. Waffeu Tamo, F. Lique, S. Niyonzima, D. Backodissa, M. Lanza (Le Havre),

Ch. Jungen, J. Zs. Mezei, O. Dulieu, J. Robert (Orsay),

O. Motapon (Douala), K. Chakrabarti (Kolkatta), N. Pop (Timisoara),

X. Urbain (Louvain), A. Wolf (Heidelberg),…

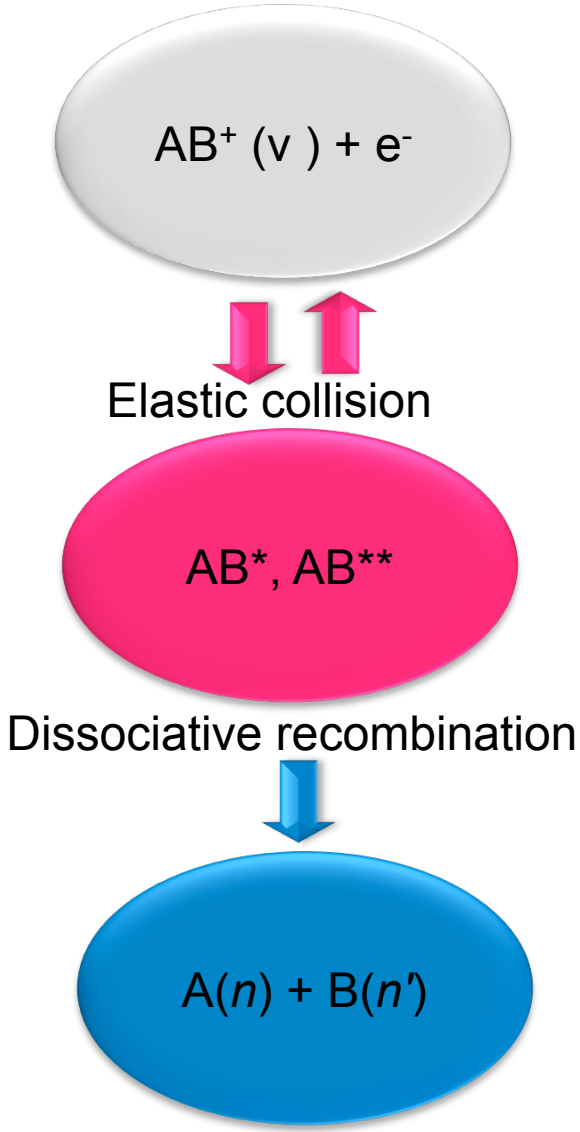
2012/09/24: Cadarache ADAS

e^-/MOLECULAR CATION collisions

2012/09/24: Cadarache ADAS

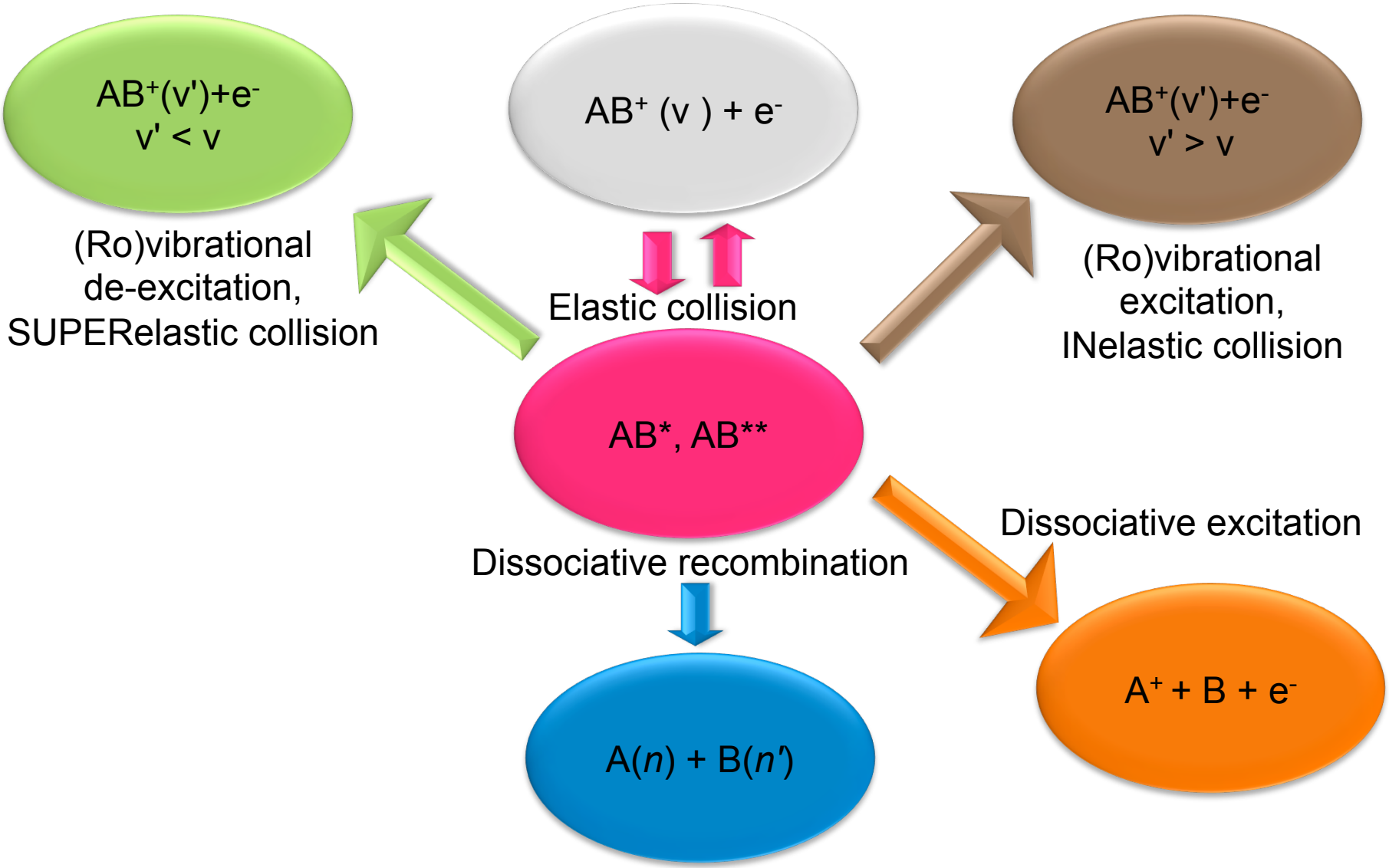
Electron/molecular cation reactive collisions

Rich dynamics, many continua, unified treatment



Electron/molecular cation reactive collisions

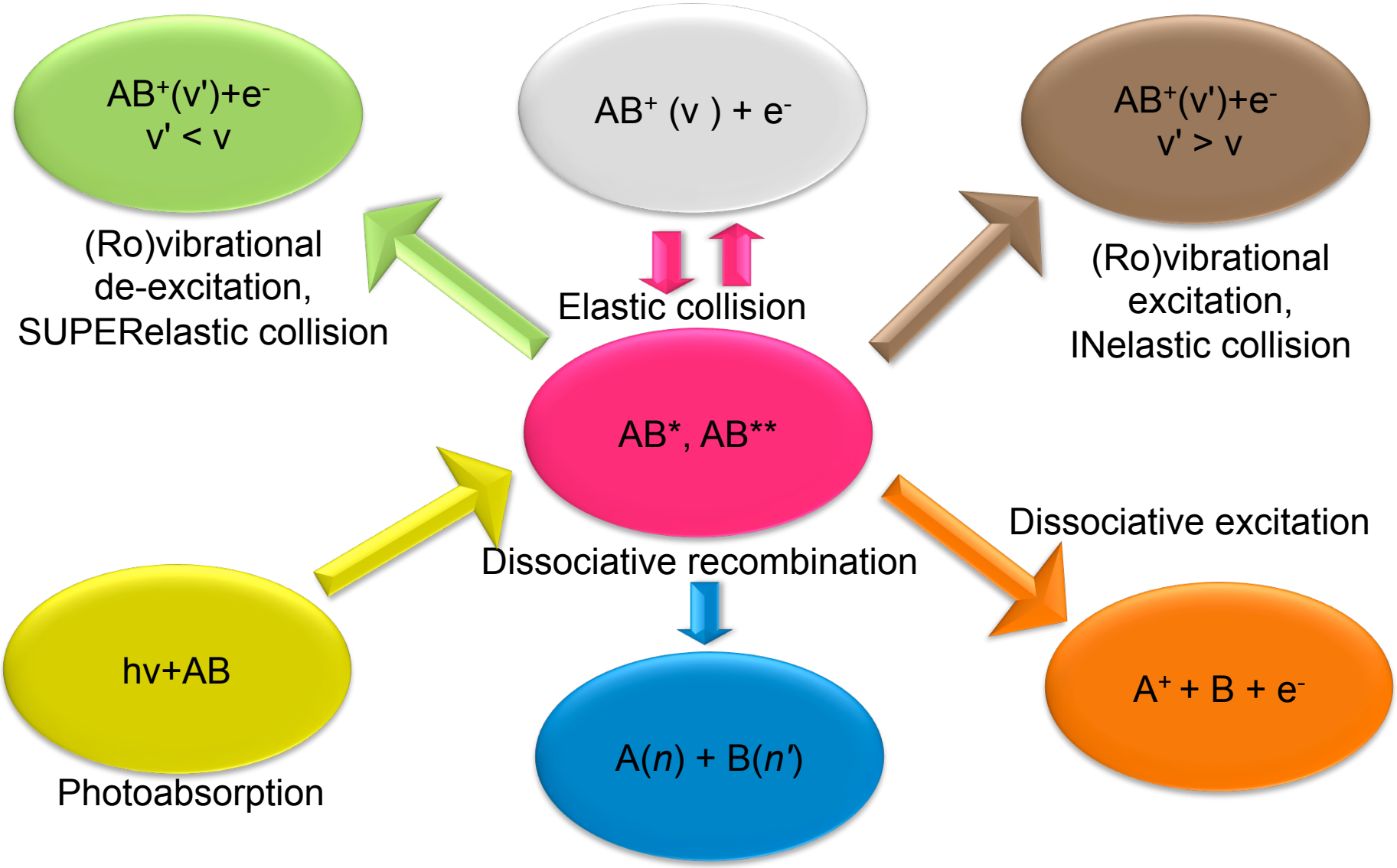
Rich dynamics, many continua, unified treatment



Electron/molecular cation reactive collisions

AND photoabsorption by neutral molecules

Rich dynamics, many continua, unified treatment



Electron/molecular cation reactive collisions

Main THEORETICAL approach: MQDT

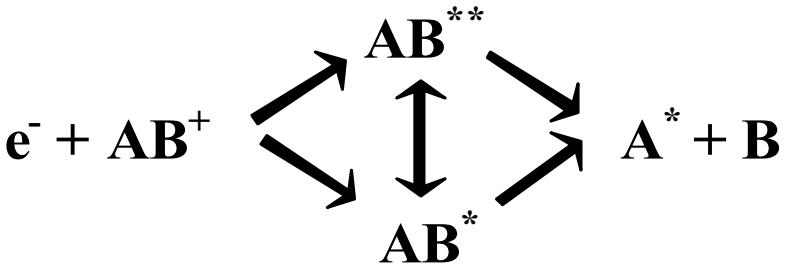
Multichannel Quantum Defect Theory

Seaton (1958-1983), Fano, Jungen, Greene, Giusti -Suzor (1970-…),…

Quantum Interferences

DIRECT

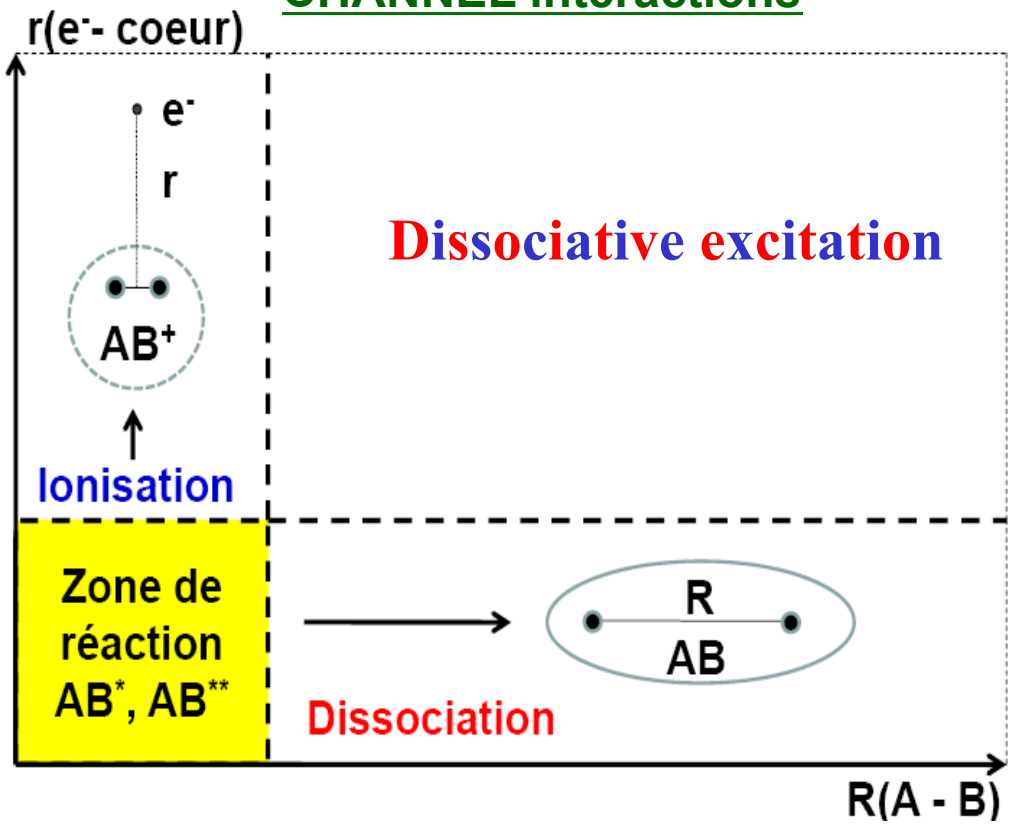
Dissociative state



Bound (Rydberg) state

INDIRECT

CHANNEL interactions



VERY ACCURATE computations: rotational & vibrational interactions

H_2^+

et isotopomers

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Electron/molecular cation reactive collisions

RESULTS: MQDT computations and comparison with experiment

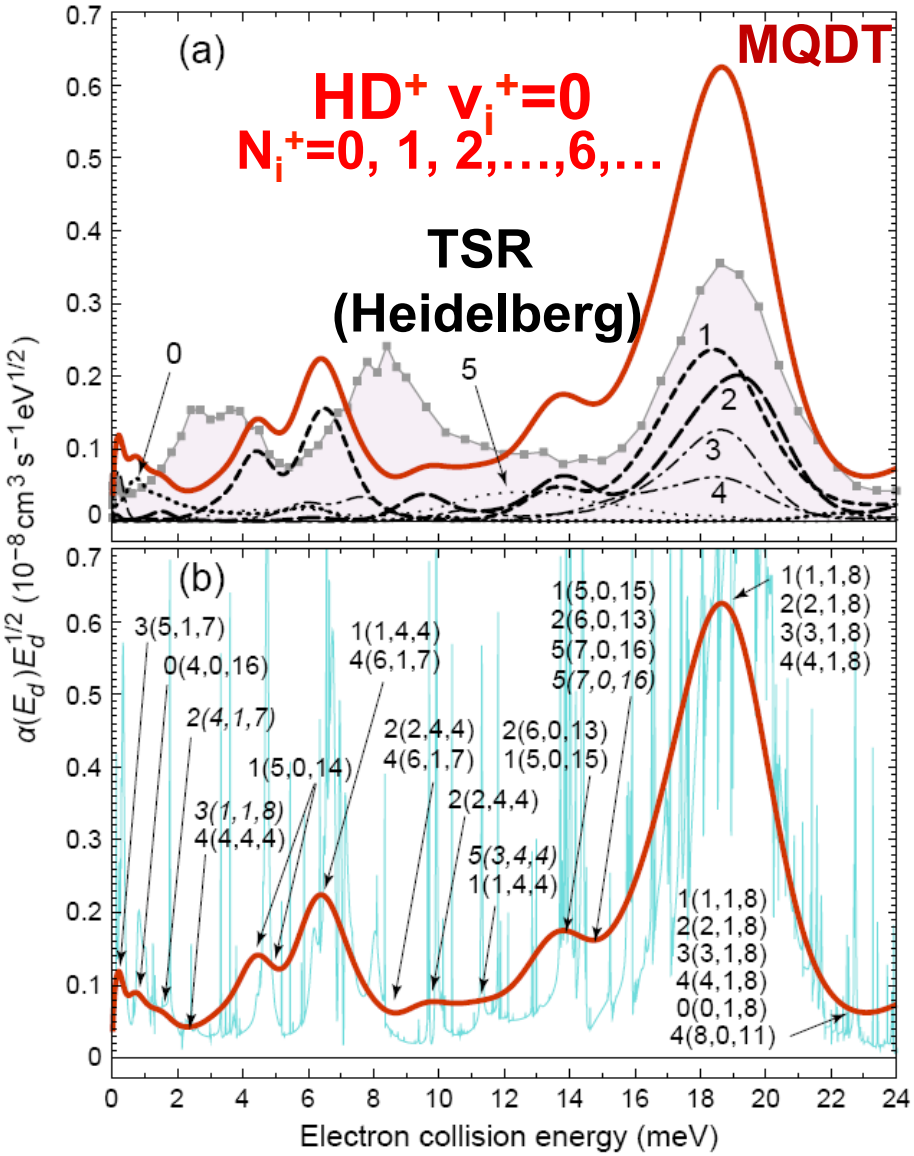
Decisive role of ROTATIONAL effects

Assignment of resonances in dissociative recombination of HD^+ ions: high-resolution measurements compared with accurate computations

Physical Review **A 84** 022710 (2011)

LOMC, MPIK, UCL (Louvain)

2011



2012/09/24: Cadarache ADAS

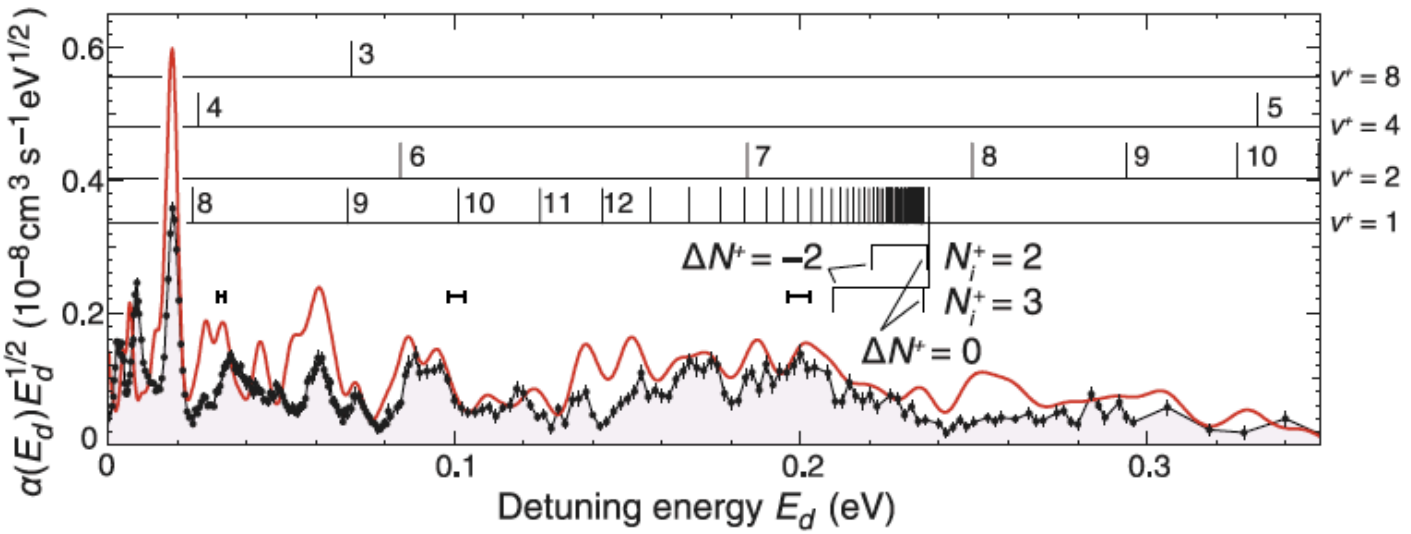


FIG. 10. (Color online) Reduced DR rate coefficients for the DR of $HD^+(^2\Sigma_g^+, v_i^+ = 0)$ from experiment (black dots and thin line) and MQDT theory (thick solid curve) after collision energy convolution and initial state averaging as in Fig. 9. Prediction bars: Rydberg resonance energies (labeled by principal quantum numbers n; zero quantum defect) below the energy thresholds for vibrational excitation to levels v^+ as indicated, assuming $\Delta N^+ = 0$ in the resonance formation. Near the $v^+ = 1$ threshold, its energetic shifts due to molecular rotation are indicated for initial states $N_i^+ = 2,3$ and the cases $\Delta N^+ = 0$ and $\Delta N^+ = -2$. Horizontal bars: FWHM experimental collision energy spread in the respective energy regions (see Sec. III).

The SAME

Rydberg resonances

(related to CLOSED channels !)

have an enhancing role in the PHOTOIONIZATION of H_2

Photoabsorption by neutral molecules

Ch. Jungen's Global-MQDT method

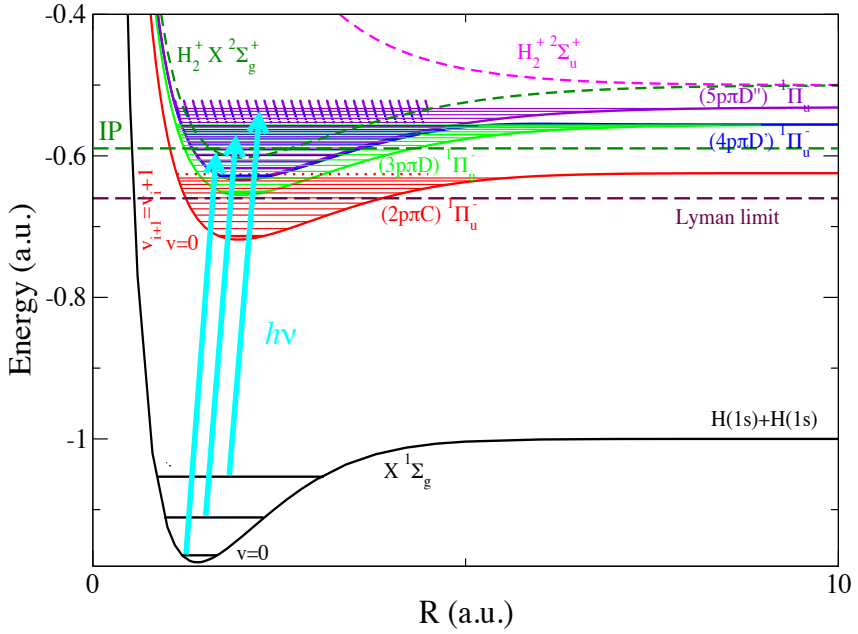
- Photoabsorption, spontaneous emission probability

$$\mathrm{H}_2(N'', v'') + h\nu \rightarrow \mathrm{H}_2(N', v')$$

$$\mathrm{H}_2(N', v') \rightarrow \mathrm{H}_2(N'', v'') + h\nu$$

- Photoionization

$$\mathrm{H}_2(N'', v'') + h\nu \longrightarrow \mathrm{H}_2^+(N', v') + e^-$$

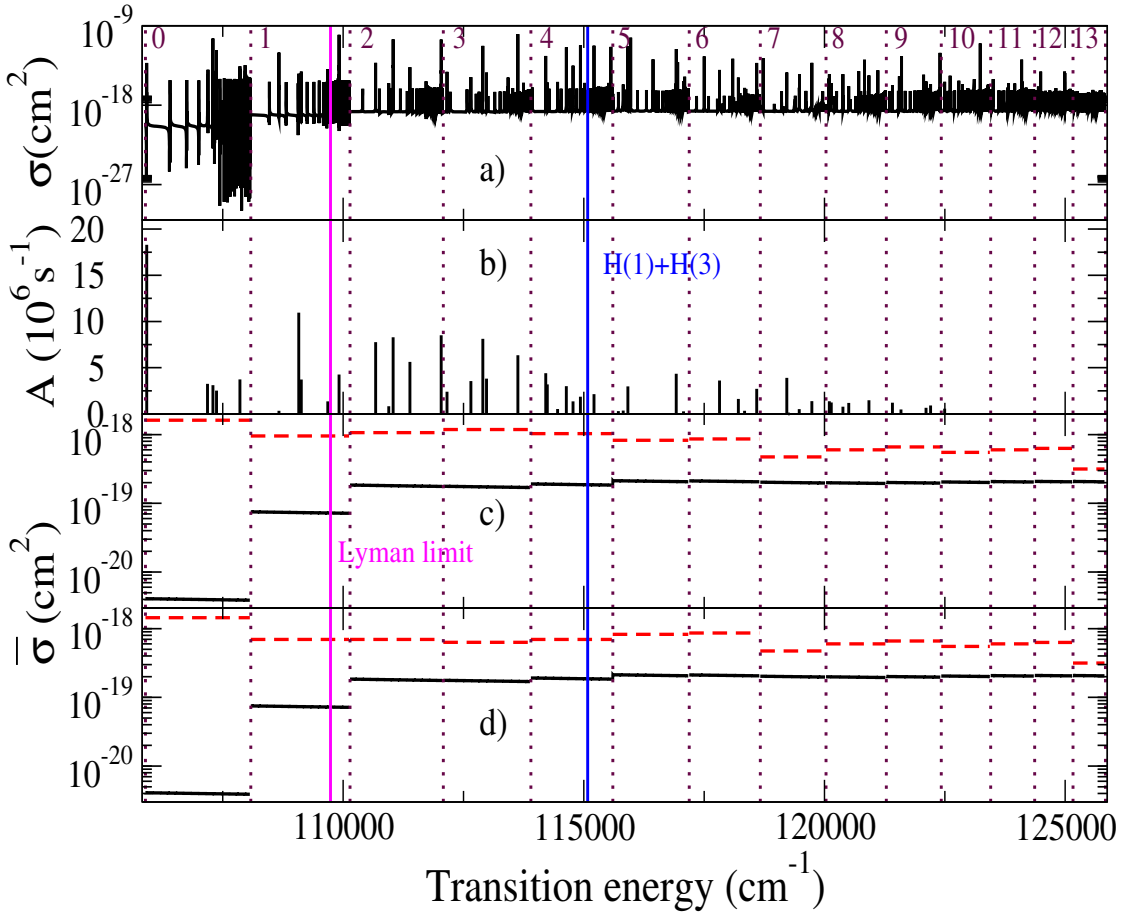


Photoabsorption by neutral molecules

Ch. Jungen's Global-MQDT method

Resonances in photoionisation: Cross sections for vibrationally excited H_2
J. Zs. Mezei, I. F. Schneider, E. Roueff et Ch. Jungen
Physical Review **A 85** 043411 (2012).

Photoionization



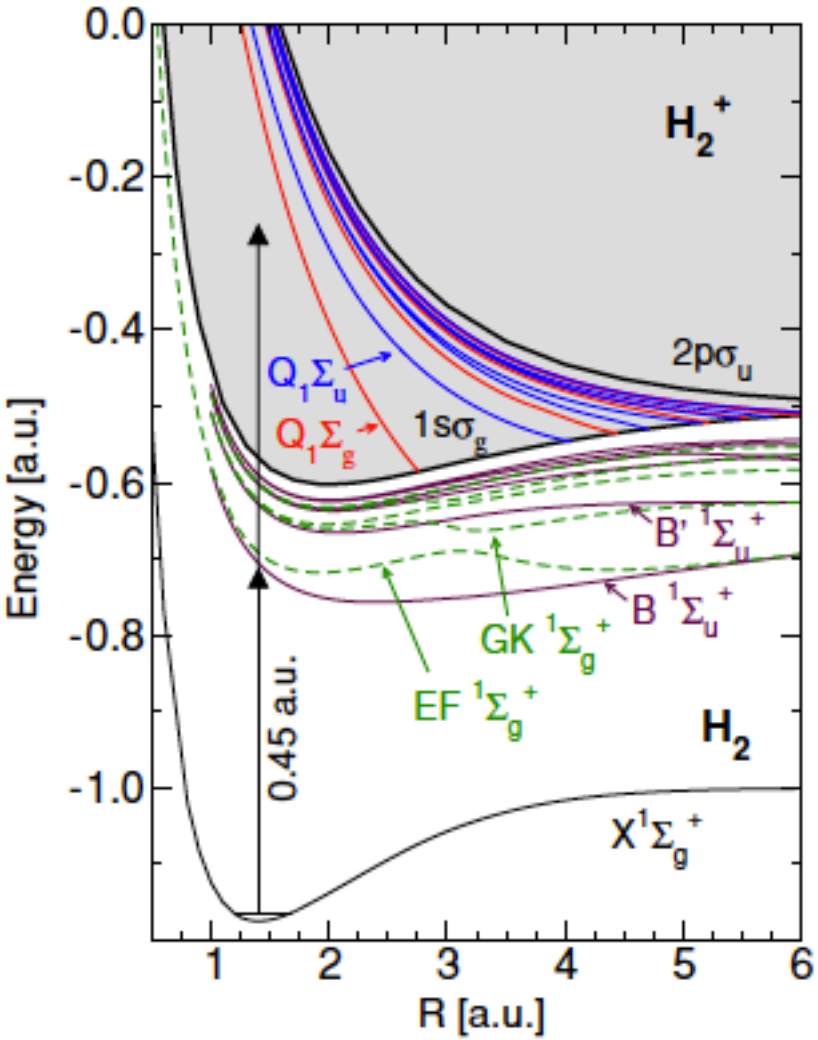
$Q(1),\ v'' = 5$

H_2

Spectroscopy & reactivity perspectives
(Jungen, Mezei, Schneider)

* Adapt the Global-MQDT method to the study of the Dissociative Recombination

* Study the resonant role of the mono-excited Rydberg H_2 states in two-photon absorption (collaboration with D. Dowek, F. Martin)



QUICK & APPROXIMATE, but REALISTIC estimations

Interstelar molecular clouds

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H_3^+
Dissociative Recombination

$$H_3^+ + e^- \Rightarrow H_2 + H,\ H + H + H$$

DR of H_3^+: … a «long-lived» mistery since the 1970's

2001 theoretical breakthrough:

DR in H_3^+ is an indirect process mediated by the Jahn-Teller mode in the p-wave channel

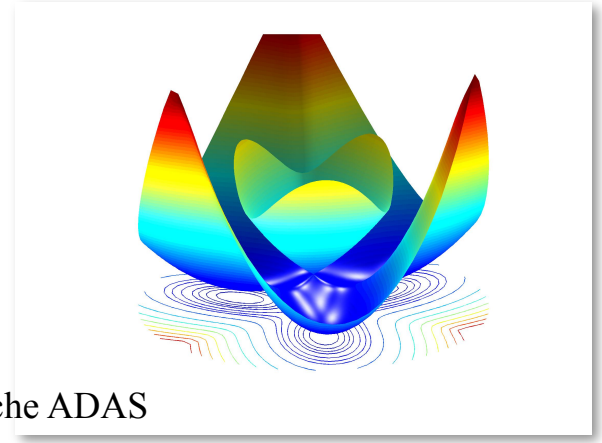
since then sophisticated large-scale calculations by the same group

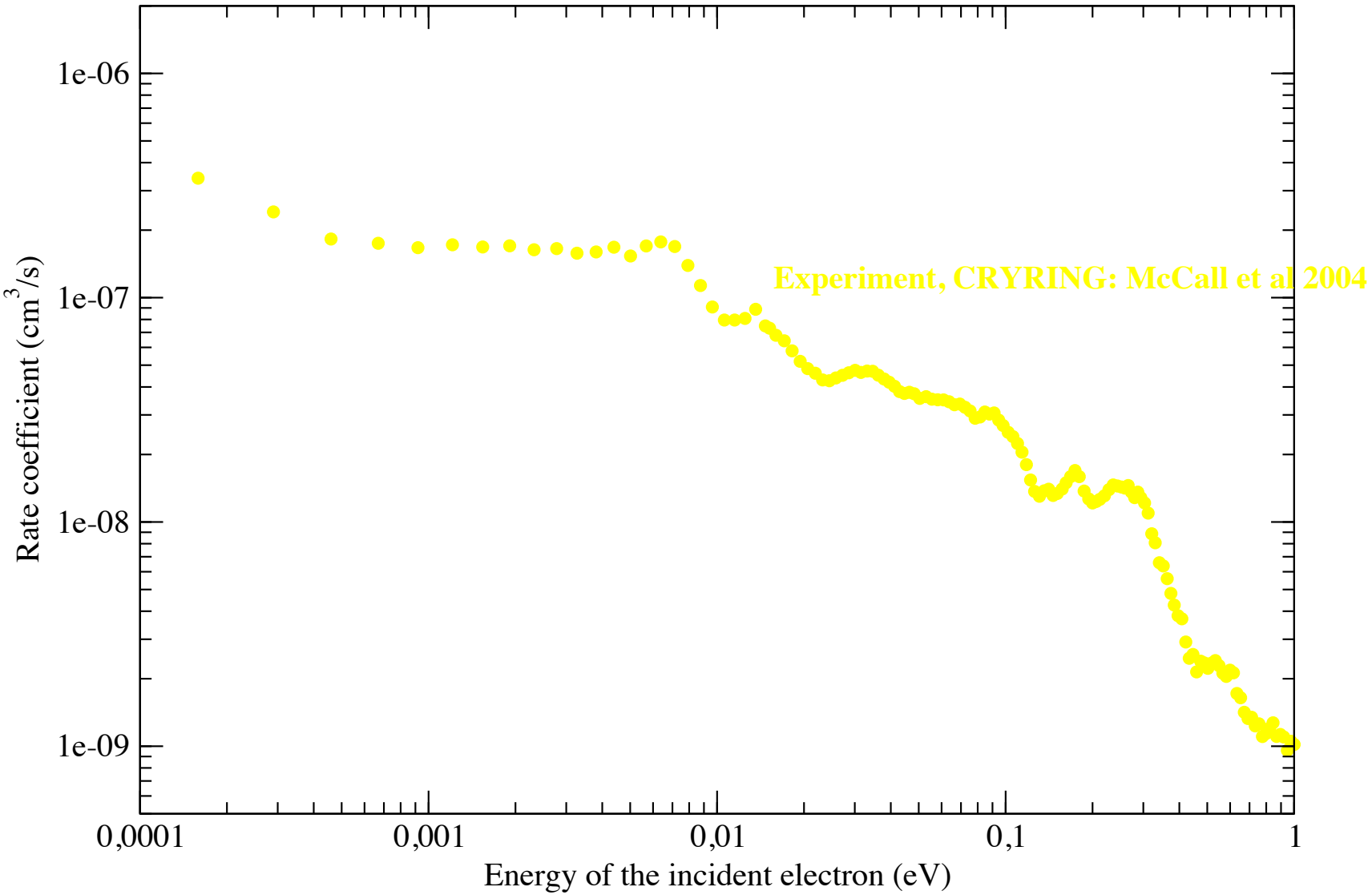
letters to nature

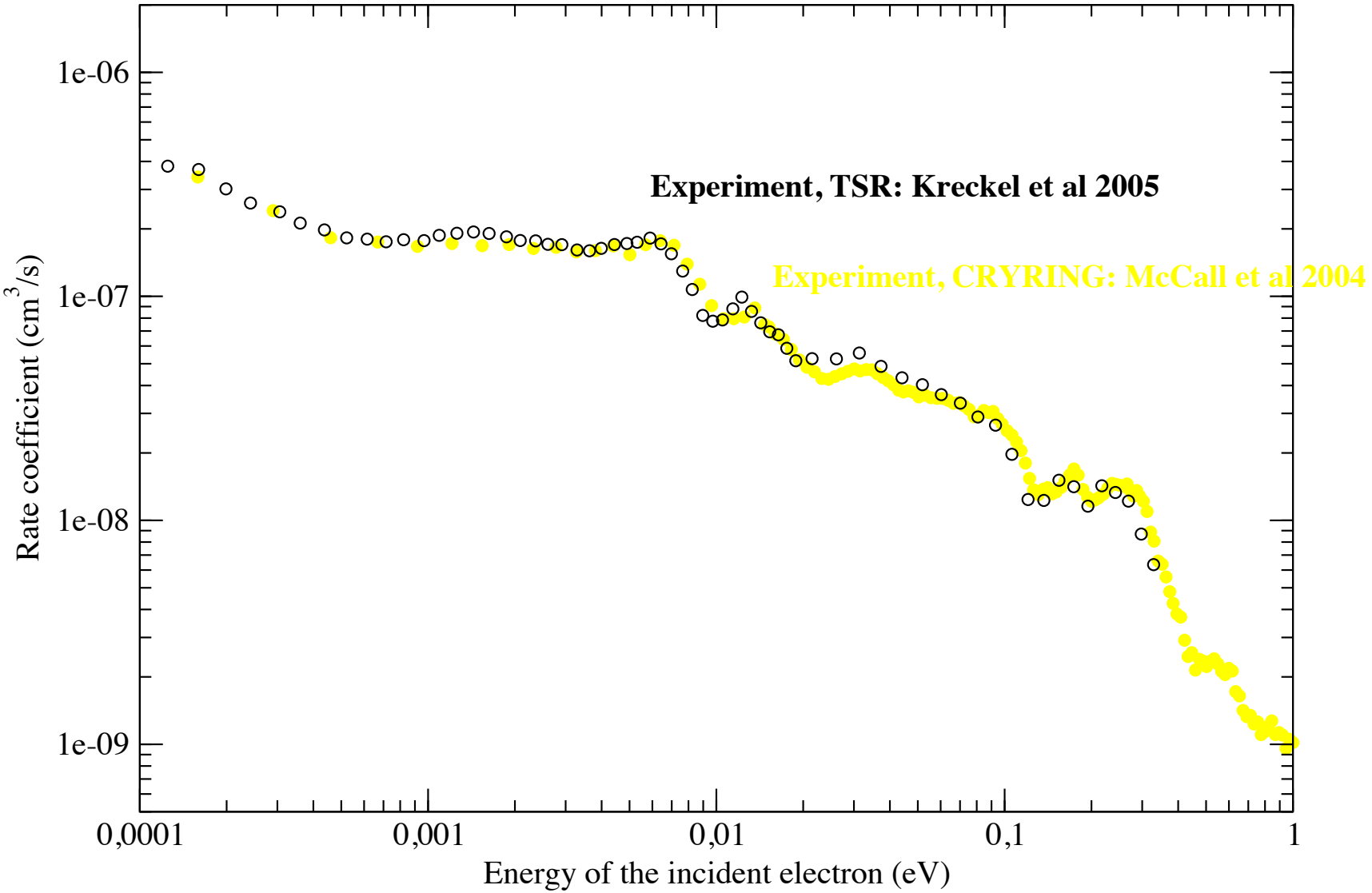
Mechanism for the destruction of H_3^+ ions by electron impact

V. Kokoouline*, Chris H. Greene* & B. D. Esry†

** Department of Physics and JILA, University of Colorado, Boulder, Colorado 80309-0440, USA*
† Department of Physics, Cardwell Hall, Kansas State University, Manhattan, Kansas 66506, USA




1e-06
1e-07
1e-08
1e-09
Rate coefficient (cm3/s)
0,0001
0,001
0,01
0,1
1
Energy of the incident electron (eV)
Experiment, CRYRING: McCall et al 2004


Experiment, TSR: Kreckel et al 2005
Experiment, CRYRING: McCall et al 2004
Rate coefficient (cm^3/s)
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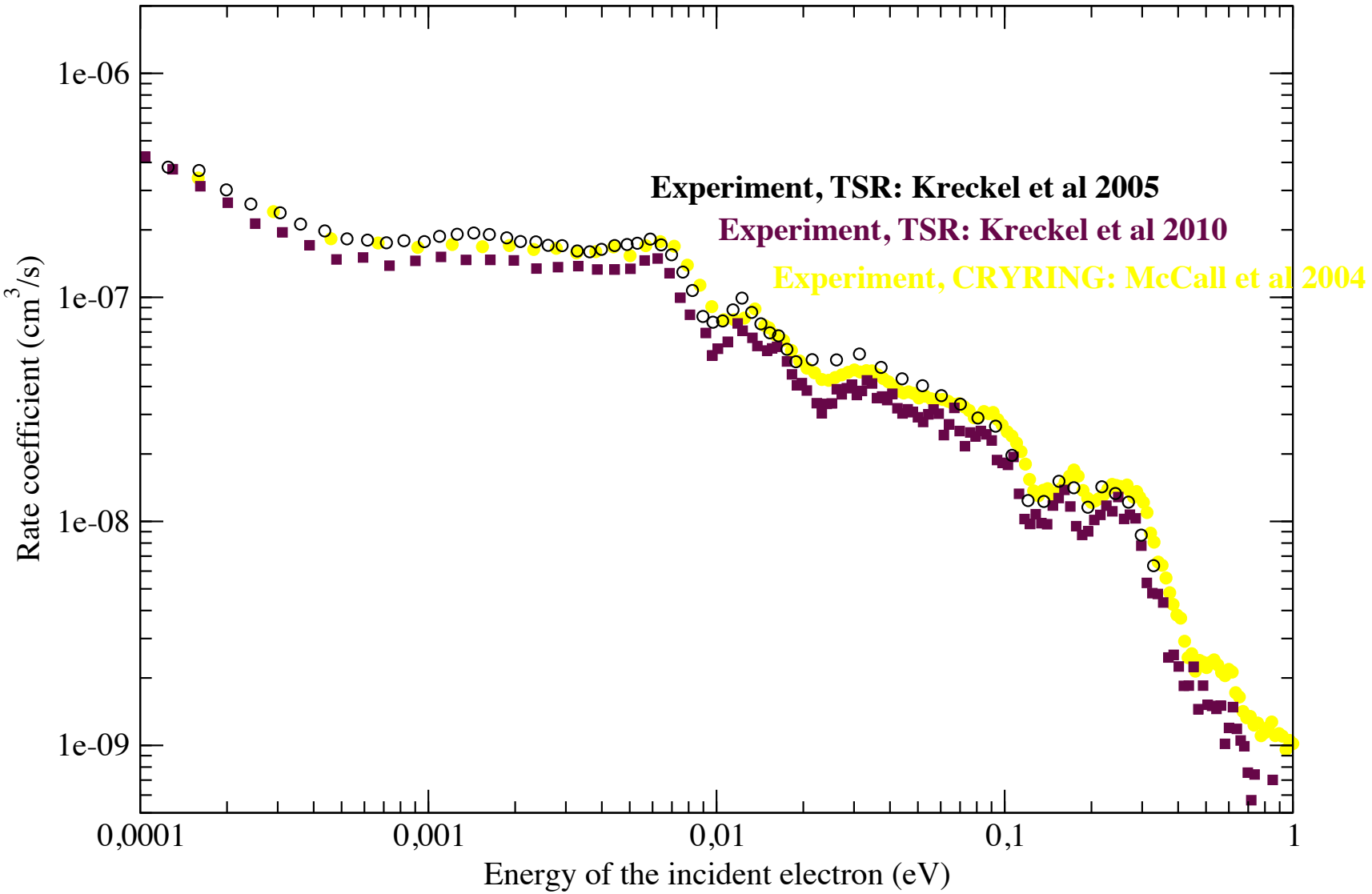
1e-07
1e-08
1e-09
0,0001
0,001
0,01
0,1
1
Energy of the incident electron (eV)


Experiment, TSR: Kreckel et al 2005
Experiment, TSR: Kreckel et al 2010
Experiment, CRYRING: McCall et al 2004
Rate coefficient (cm3/s)
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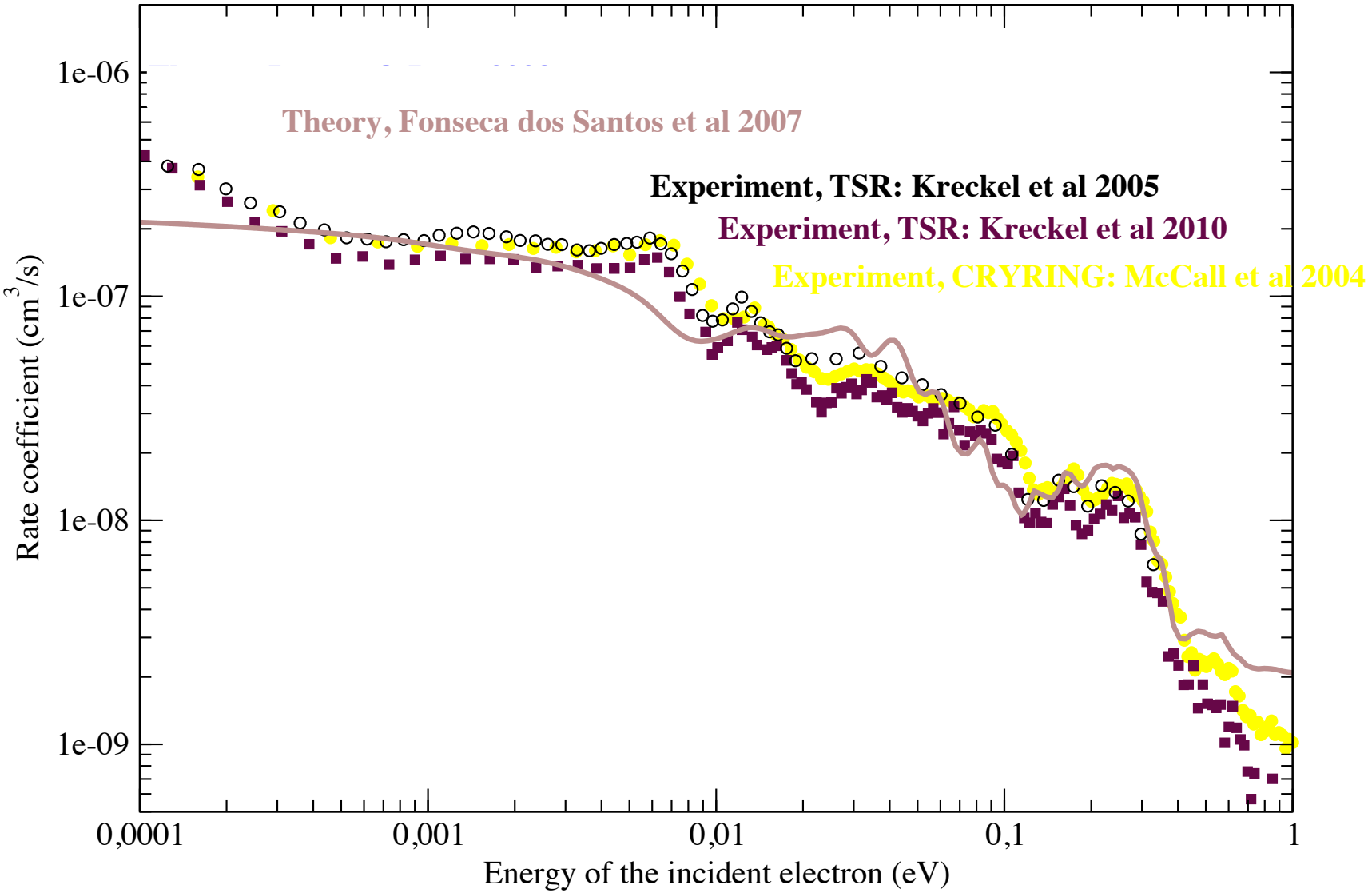
1e-07
1e-08
1e-09
0,0001
0,001
0,01
0,1
1
Energy of the incident electron (eV)


Theory, Fonseca dos Santos et al 2007
Experiment, TSR: Kreckel et al 2005
Experiment, TSR: Kreckel et al 2010
Experiment, CRYRING: McCall et al 2004
Rate coefficient (cm^3/s)
1e-06
1e-07
1e-08
1e-09
0,0001
0,001
0,01
0,1
1
Energy of the incident electron (eV)

A spectroscopic approach of the Dissociative Recombination

Jungen & Pratt 2008-2010

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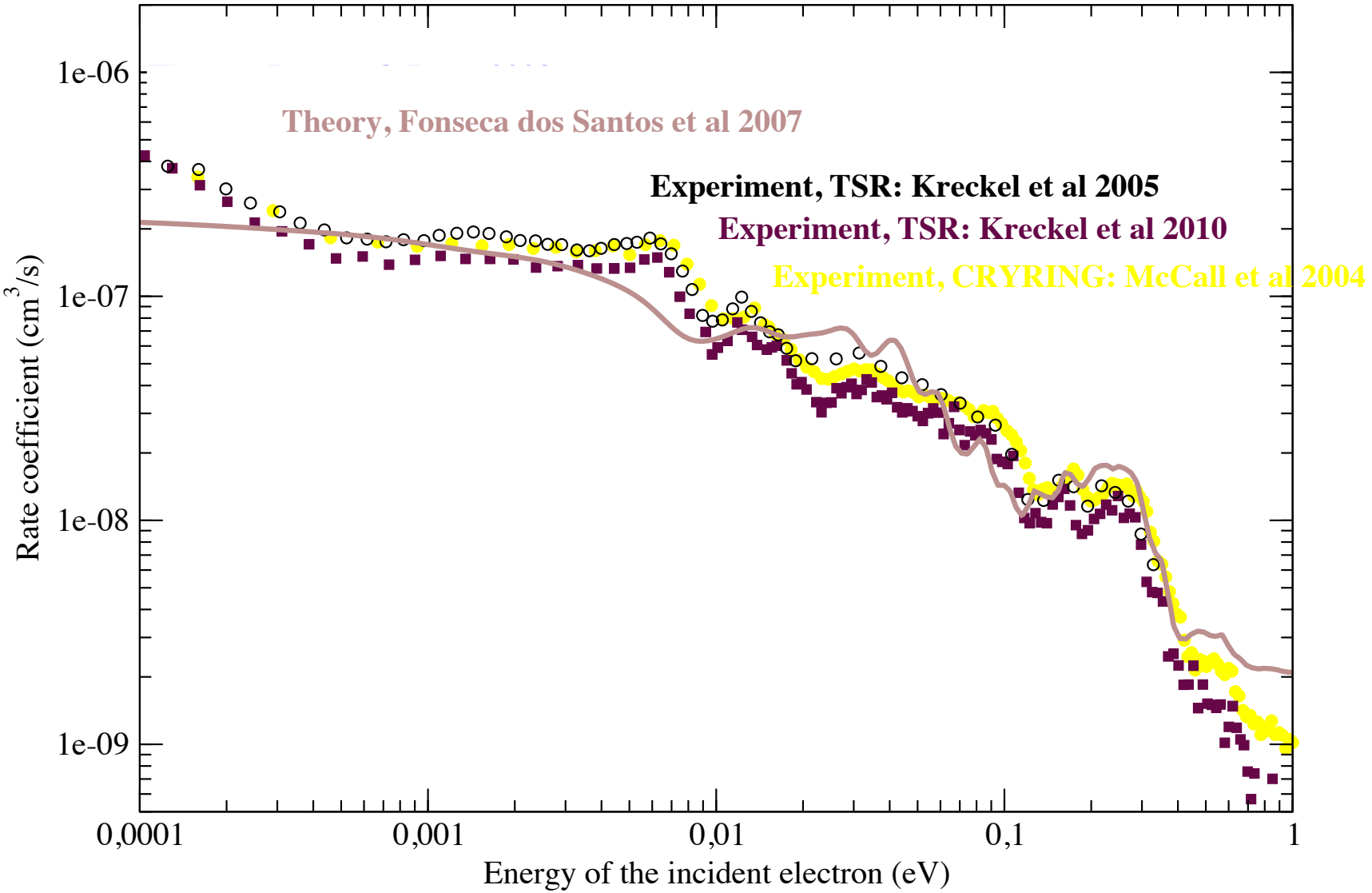
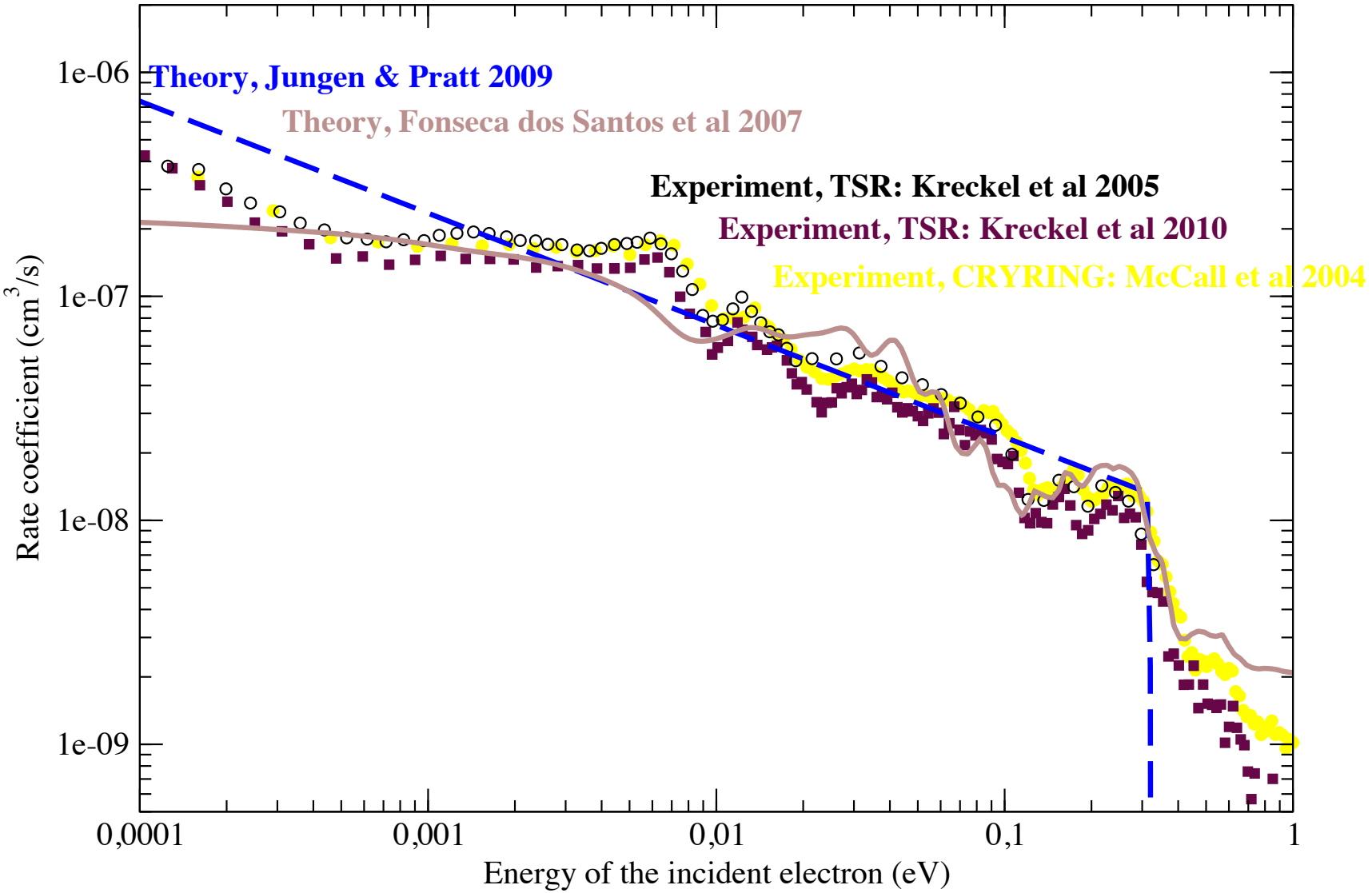
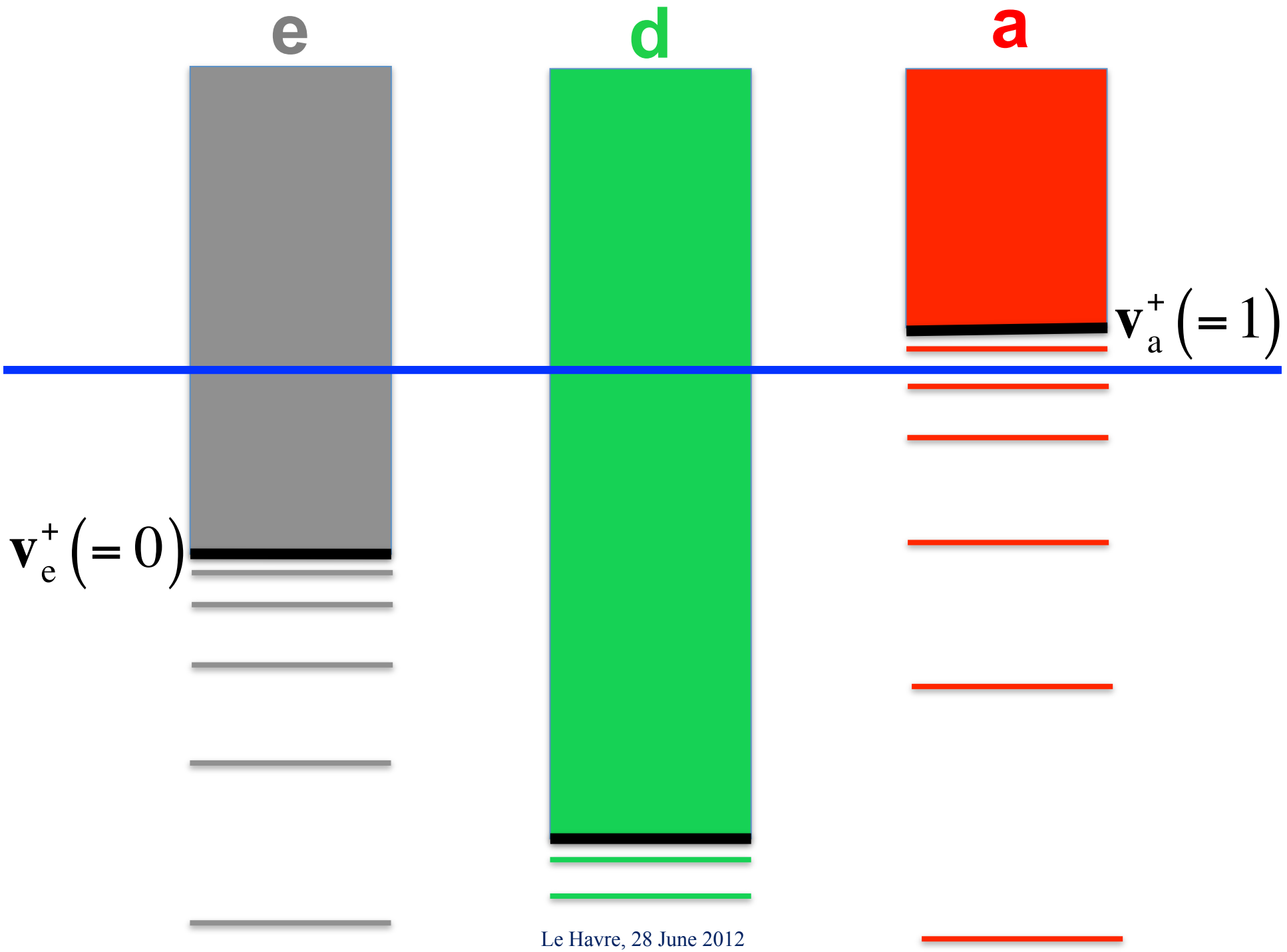
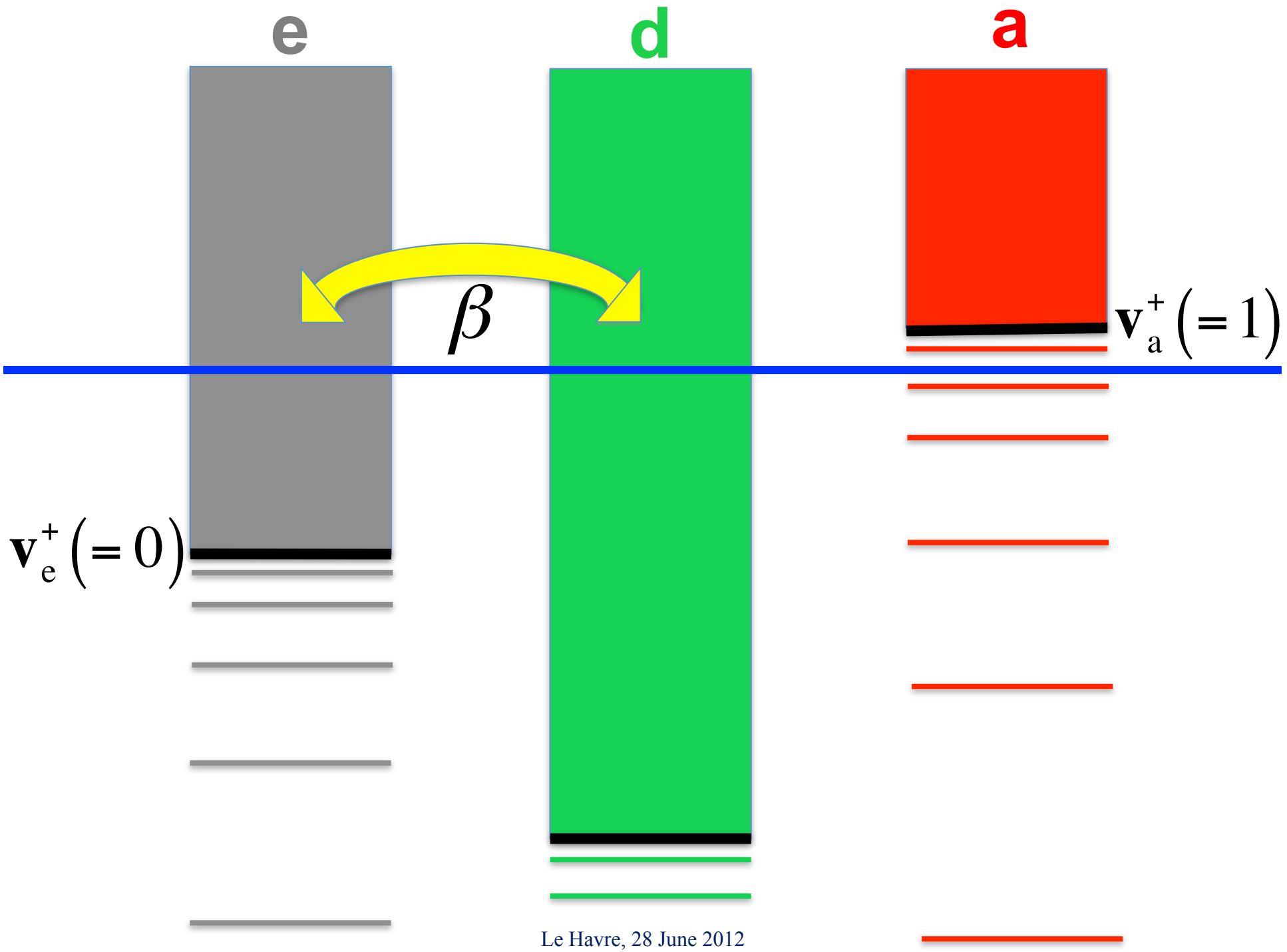

Theory, Fonseca dos Santos et al 2007
Experiment, TSR: Kreckel et al 2005
Experiment, TSR: Kreckel et al 2010
Experiment, CRYRING: McCall et al 2004
Rate coefficient (cm^3/s)
1e-06
1e-07
1e-08
1e-09
0,0001
0,001
0,01
0,1
1
Energy of the incident electron (eV)

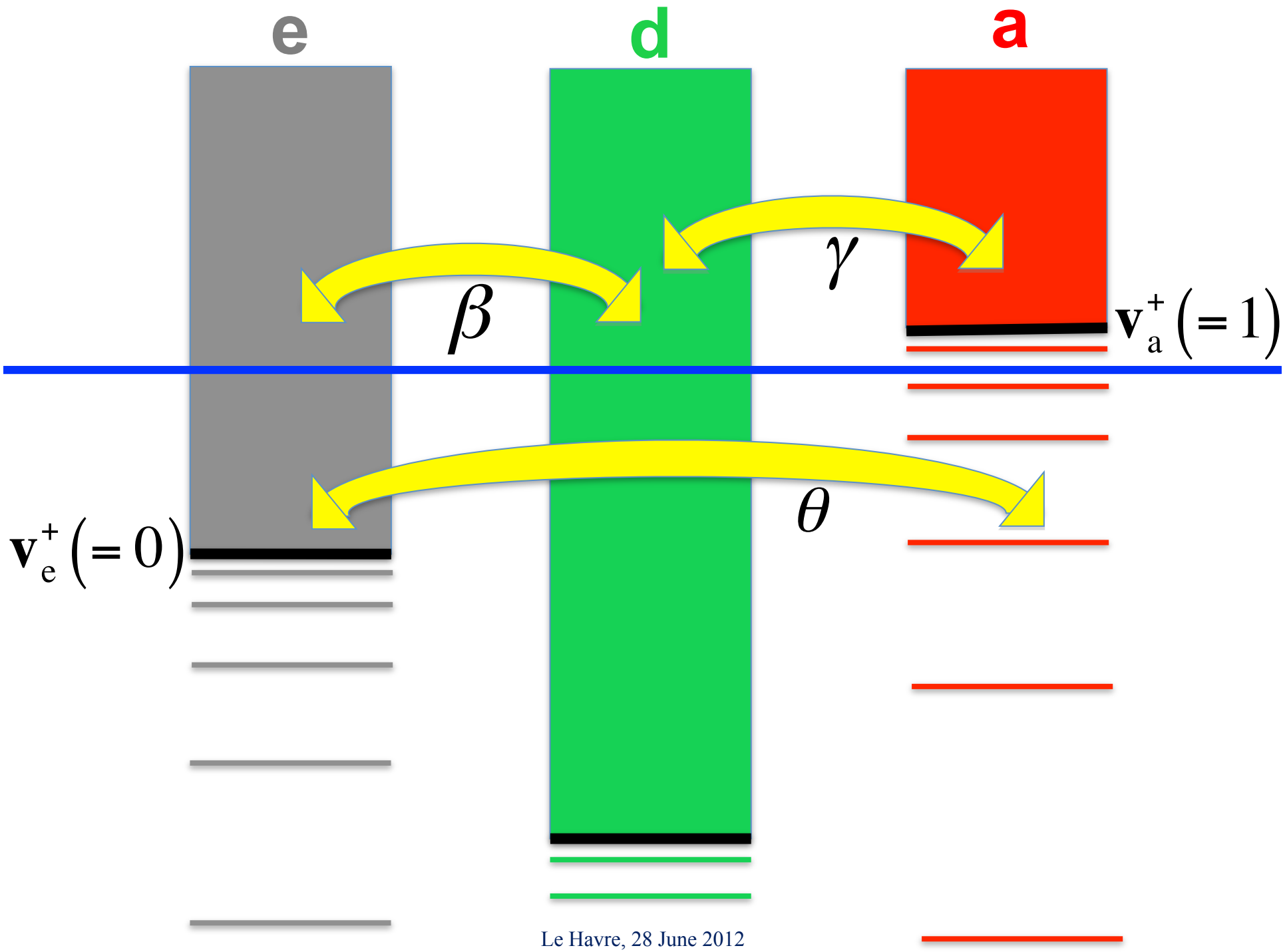

Theory, Jungen & Pratt 2009
Theory, Fonseca dos Santos et al 2007
Experiment, TSR: Kreckel et al 2005
Experiment, TSR: Kreckel et al 2010
Experiment, CRYRING: McCall et al 2004
1e-06
1e-07
1e-08
1e-09
Rate coefficient (cm3/s)
0,0001
0,001
0,01
0,1
1
Energy of the incident electron (eV)

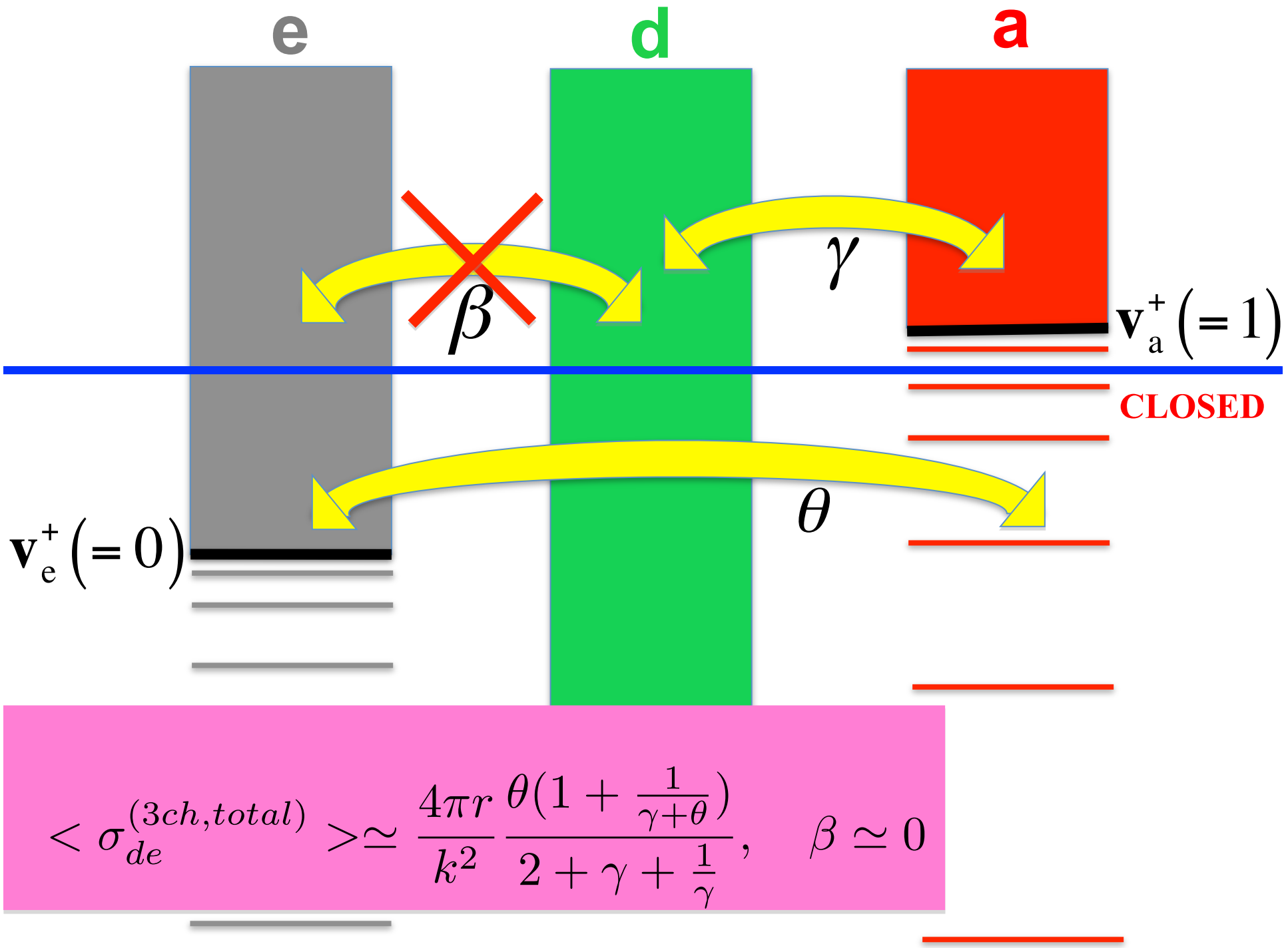
Refined model:
3-channels
(MINIMUM number of channels)

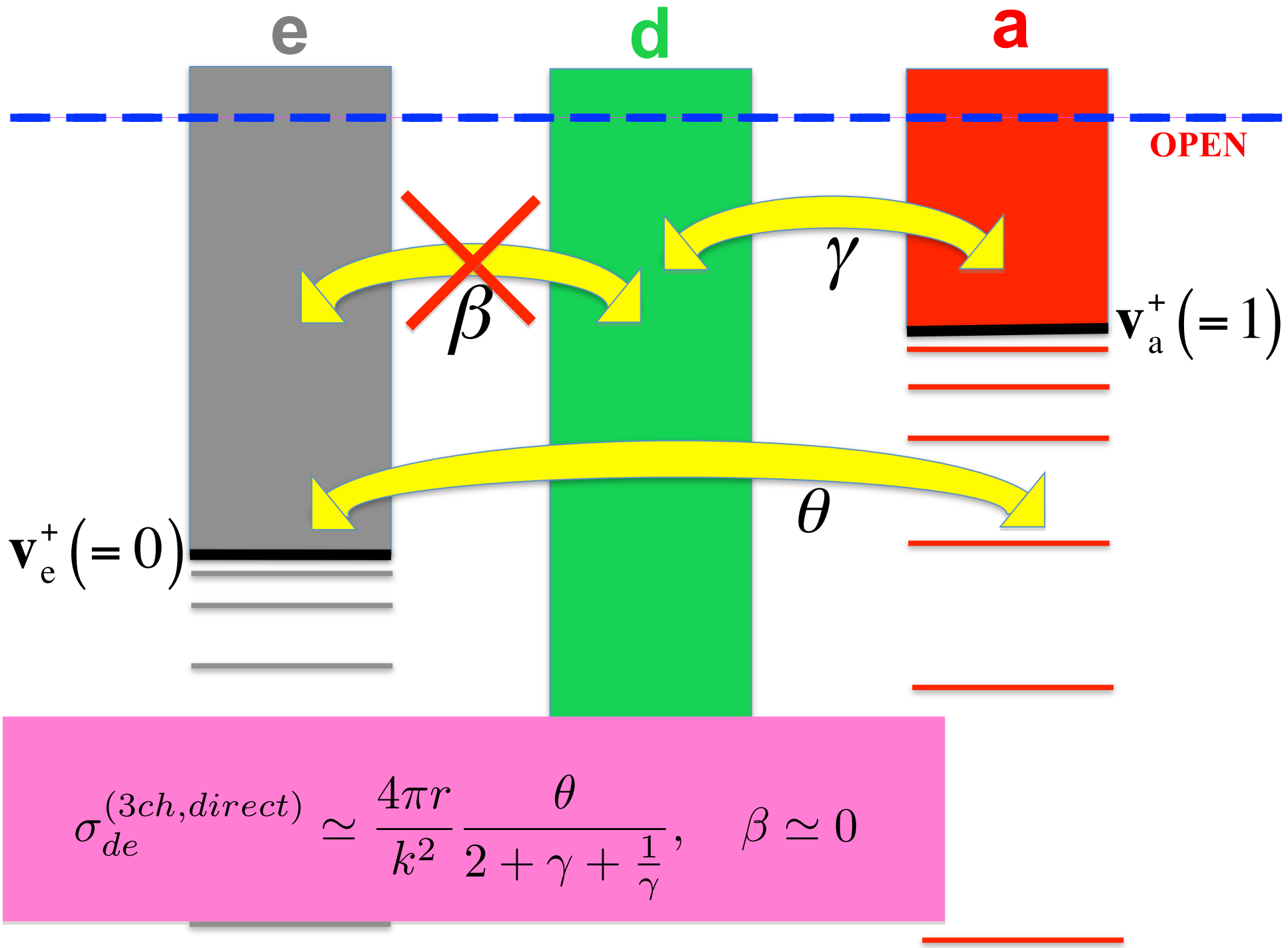
Schneider, Pop, Lique, Jungen 2010-2012


e
d
a
$\mathbf{v}_a^+(=1)$
$\mathbf{v}_e^+(=0)$

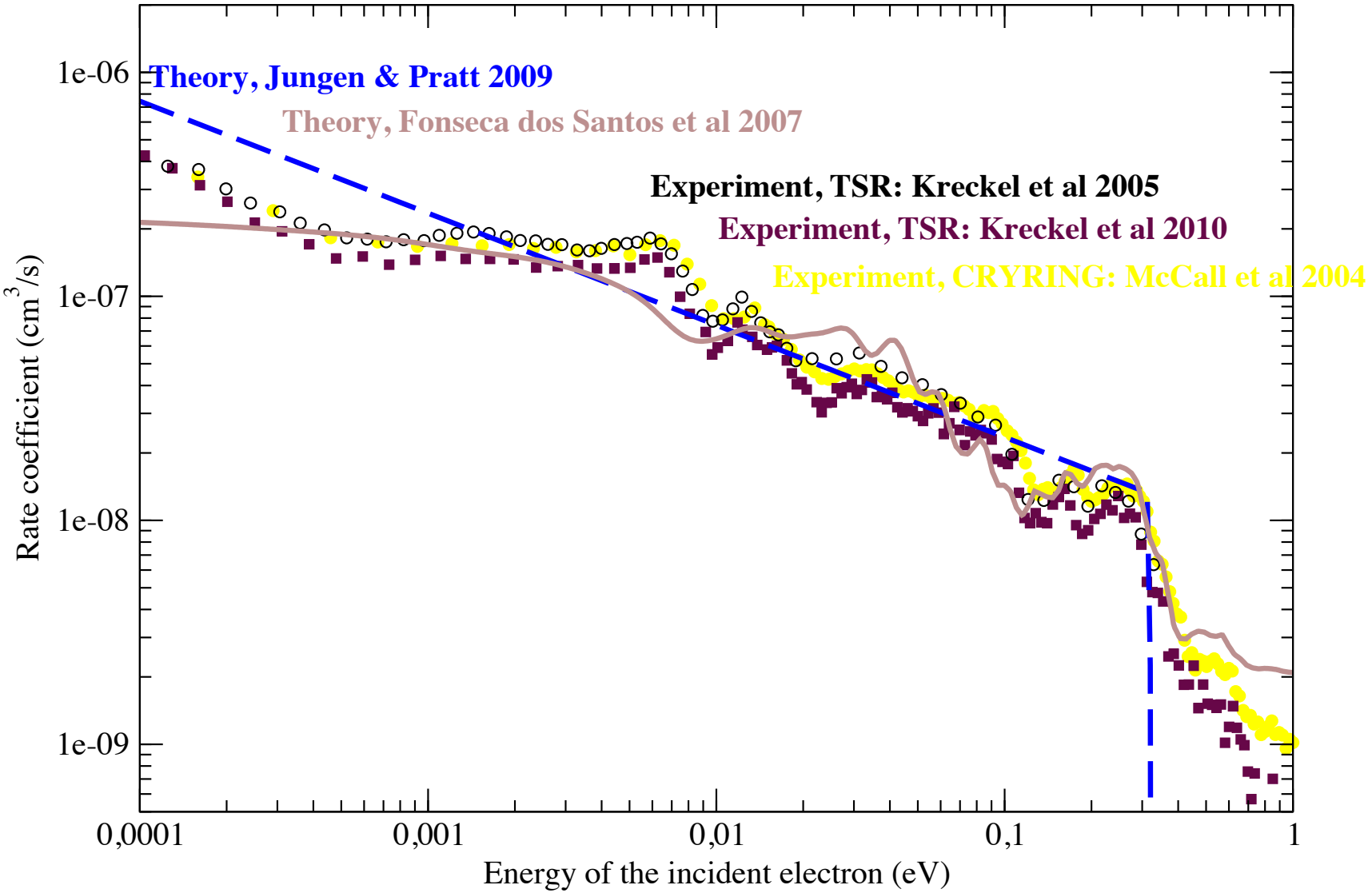

e
d
a
β
$\mathbf{v}_a^+(=1)$
$\mathbf{v}_e^+(=0)$

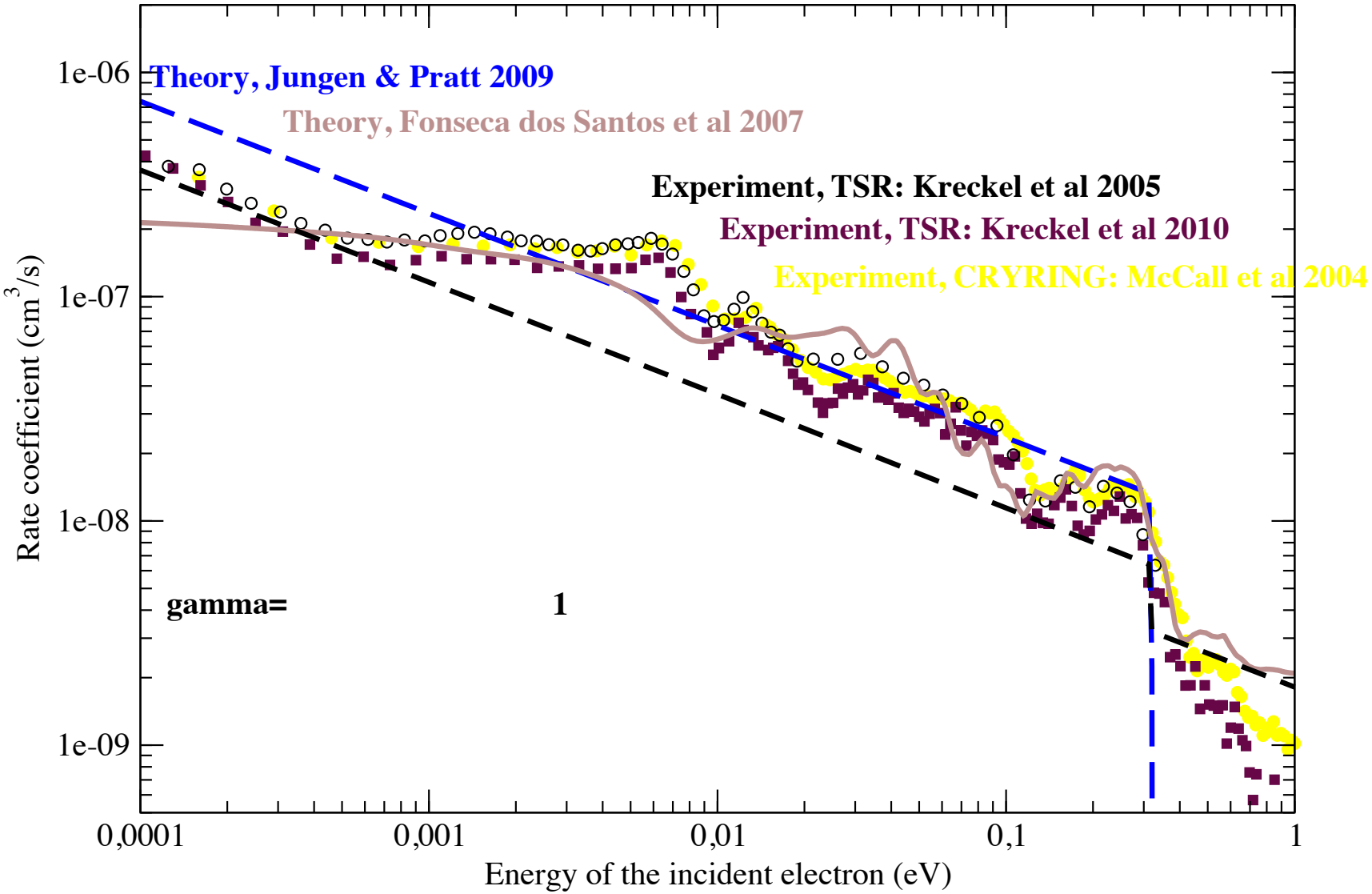

e
d
a
β
γ
θ
$\mathbf{v}_a^+(=1)$
$\mathbf{v}_e^+(=0)$

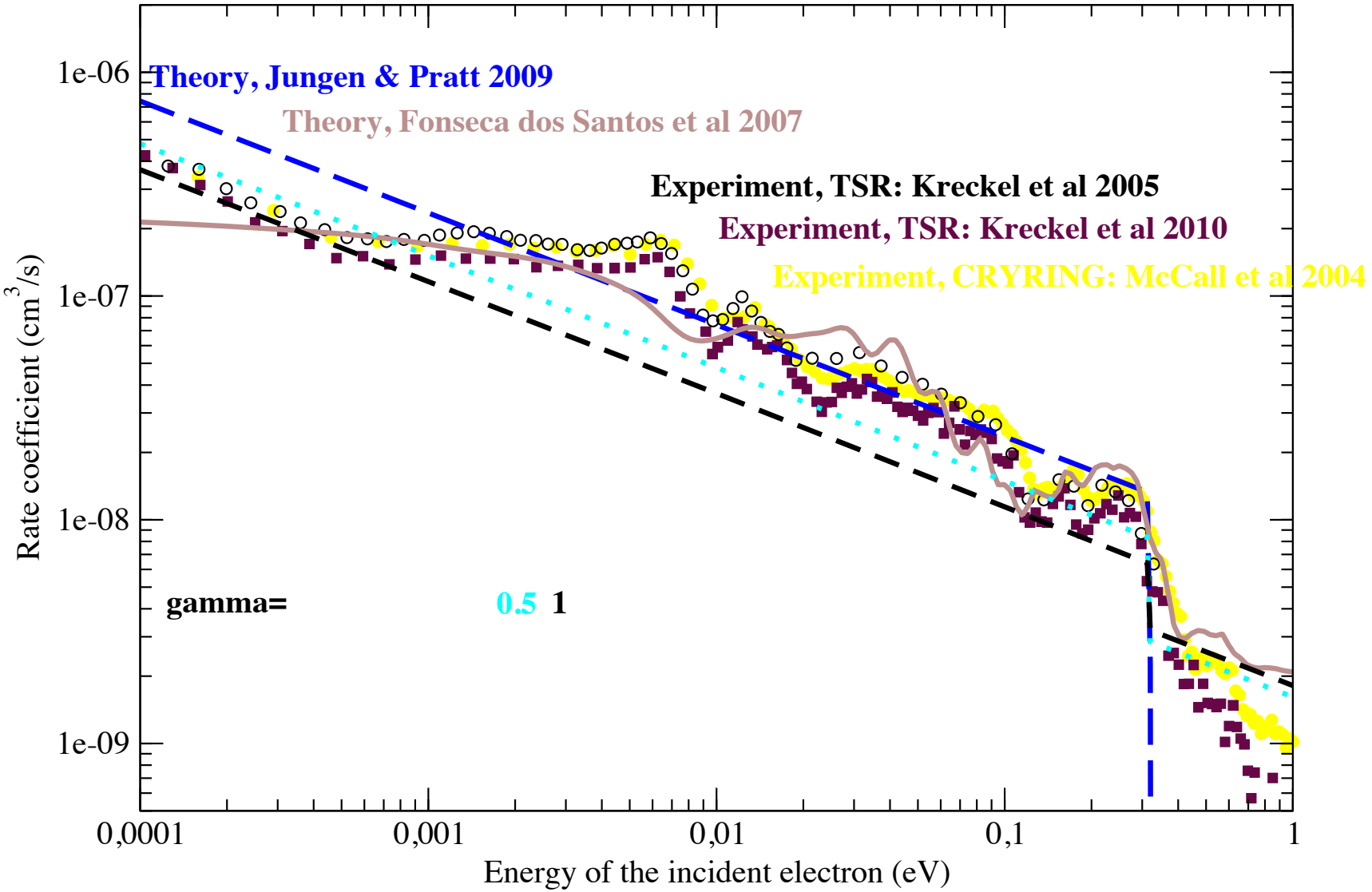

e
d
a
γ
β
$\mathbf{v}_a^+(=1)$
CLOSED
θ
$\mathbf{v}_e^+(=0)$
$< \sigma_{de}^{(3ch,total)} > \simeq \frac{4\pi r}{k^2} \frac{\theta(1+\frac{1}{\gamma+\theta})}{2+\gamma+\frac{1}{\gamma}}, \quad \beta \simeq 0$

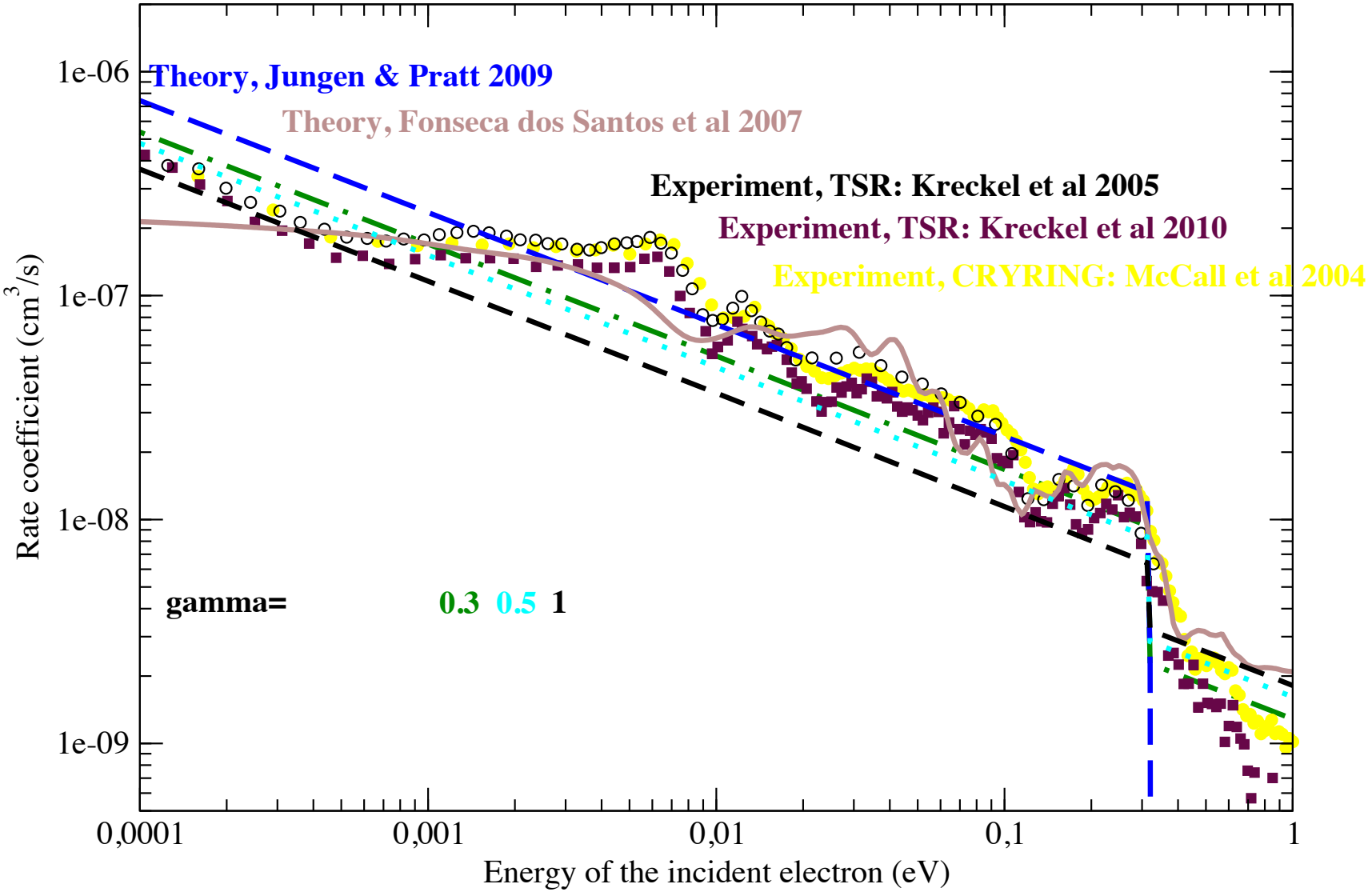


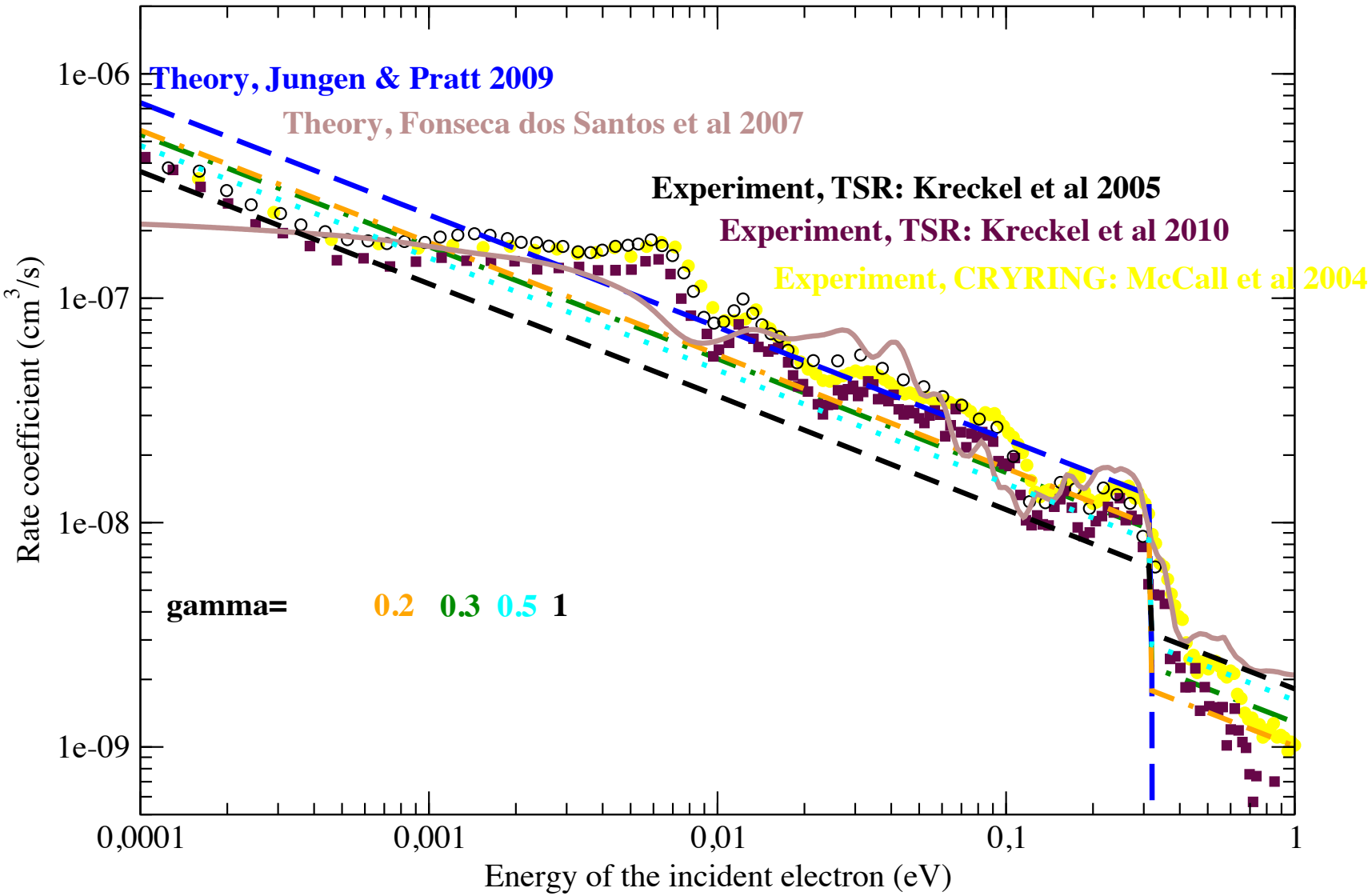
$$\sigma_{de}^{(3ch,direct)} \simeq \frac{4\pi r}{k^2} \frac{\theta}{2+\gamma+\frac{1}{\gamma}}, \quad \beta \simeq 0$$

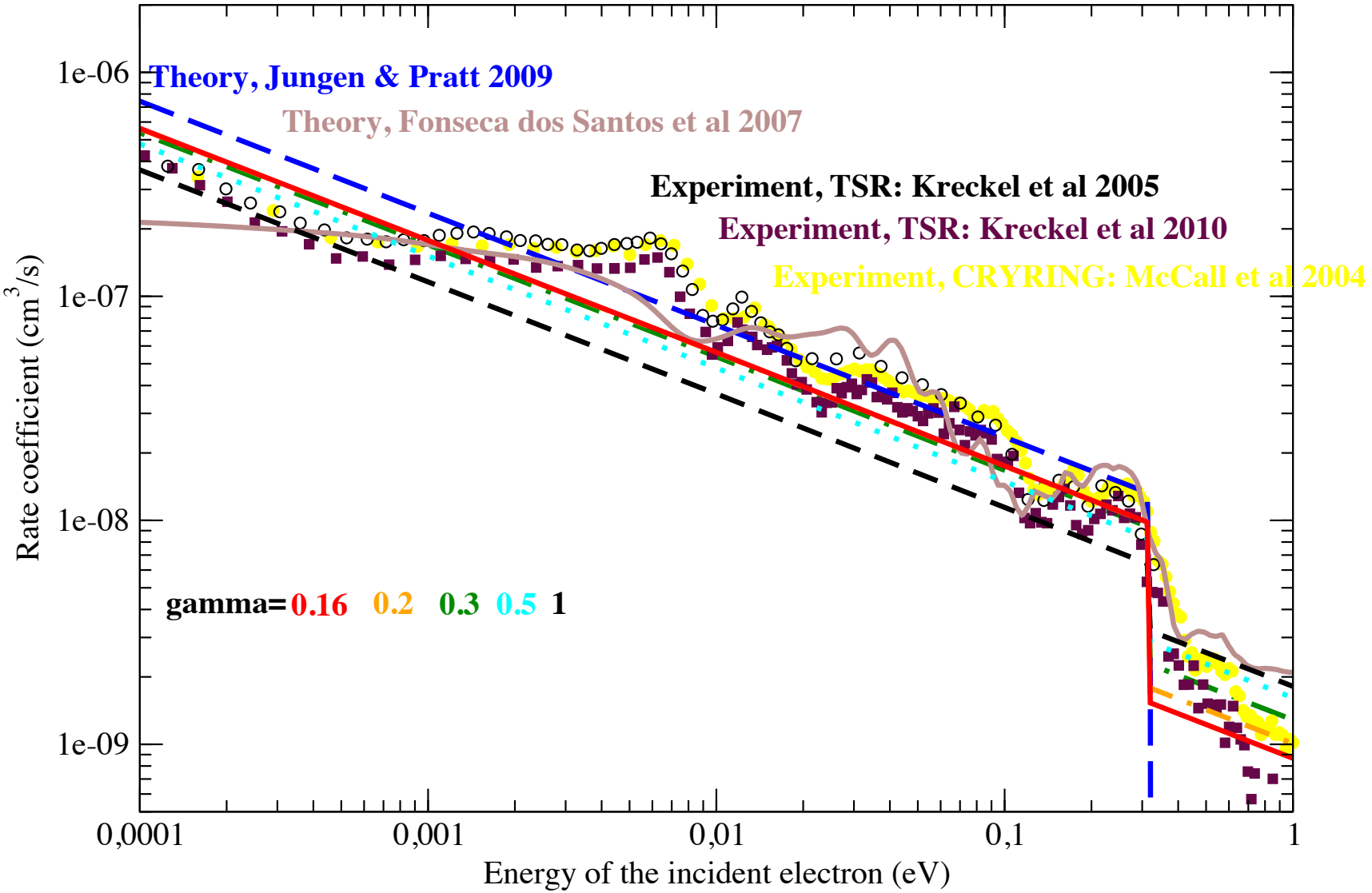

Theory, Jungen & Pratt 2009
Theory, Fonseca dos Santos et al 2007
Experiment, TSR: Kreckel et al 2005
Experiment, TSR: Kreckel et al 2010
Experiment, CRYRING: McCall et al 2004
1e-06
1e-07
1e-08
1e-09
Rate coefficient (cm3/s)
0,0001
0,001
0,01
0,1
1
Energy of the incident electron (eV)


Theory, Jungen & Pratt 2009
Theory, Fonseca dos Santos et al 2007
Experiment, TSR: Kreckel et al 2005
Experiment, TSR: Kreckel et al 2010
Experiment, CRYRING: McCall et al 2004
gamma= 1
1e-06
1e-07
1e-08
1e-09
Rate coefficient (cm3/s)
0,0001
0,001
0,01
0,1
1
Energy of the incident electron (eV)


Theory, Jungen & Pratt 2009
Theory, Fonseca dos Santos et al 2007
Experiment, TSR: Kreckel et al 2005
Experiment, TSR: Kreckel et al 2010
Experiment, CRYRING: McCall et al 2004
gamma=
0.5
1
1e-06
1e-07
1e-08
1e-09
Rate coefficient (cm3/s)
0,0001
0,001
0,01
0,1
1
Energy of the incident electron (eV)


Theory, Jungen & Pratt 2009
Theory, Fonseca dos Santos et al 2007
Experiment, TSR: Kreckel et al 2005
Experiment, TSR: Kreckel et al 2010
Experiment, CRYRING: McCall et al 2004
gamma=
0.3
0.5
1
1e-06
1e-07
1e-08
1e-09
Rate coefficient (cm3/s)
0,0001
0,001
0,01
0,1
1
Energy of the incident electron (eV)


Theory, Jungen & Pratt 2009
Theory, Fonseca dos Santos et al 2007
Experiment, TSR: Kreckel et al 2005
Experiment, TSR: Kreckel et al 2010
Experiment, CRYRING: McCall et al 2004
gamma= 0.2 0.3 0.5 1
Rate coefficient (cm3/s)
1e-06
1e-07
1e-08
1e-09
0,0001
0,001
0,01
0,1
1
Energy of the incident electron (eV)

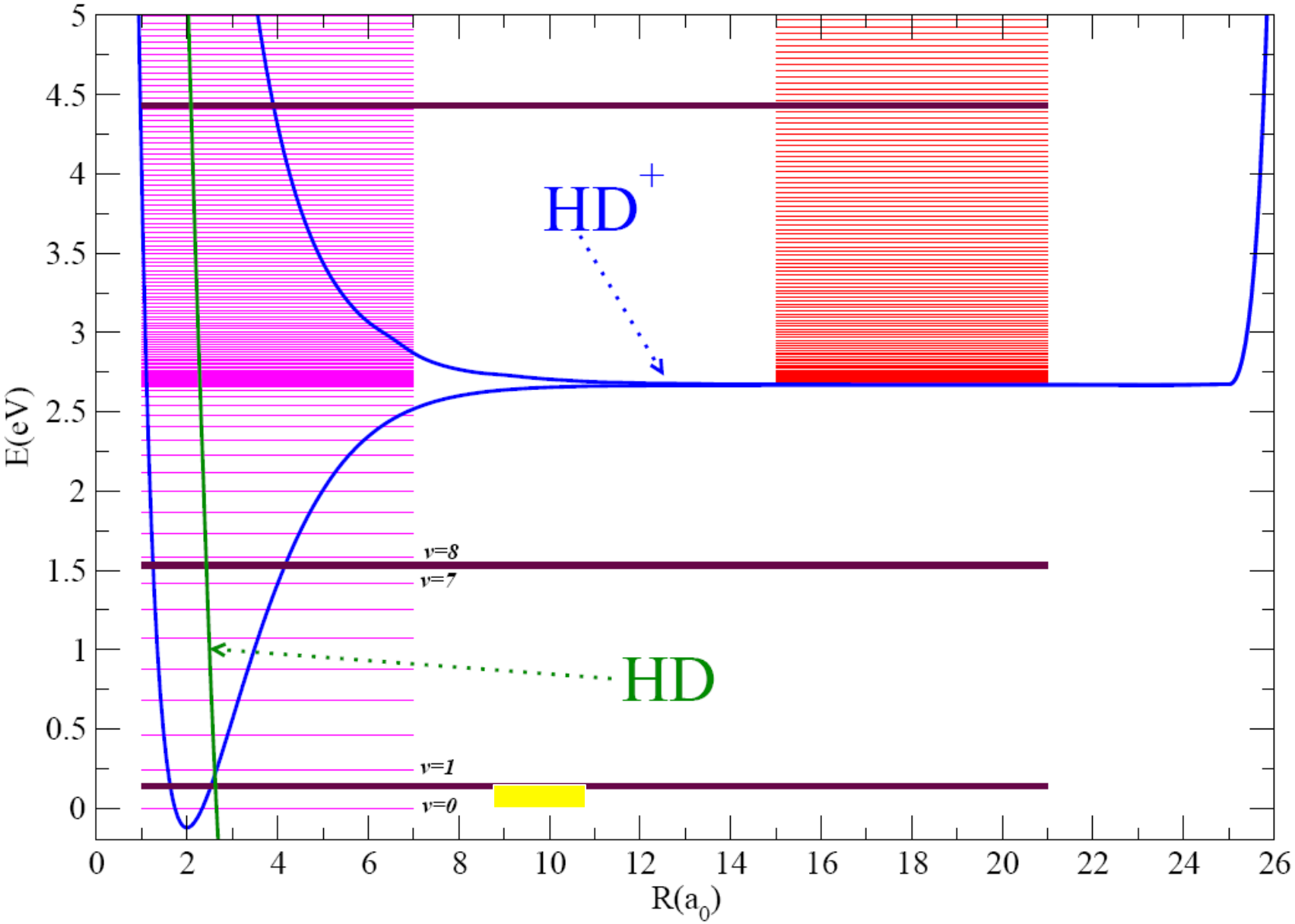

Theory, Jungen & Pratt 2009
Theory, Fonseca dos Santos et al 2007
Experiment, TSR: Kreckel et al 2005
Experiment, TSR: Kreckel et al 2010
Experiment, CRYRING: McCall et al 2004
gamma= 0.16 0.2 0.3 0.5 1
Rate coefficient (cm3/s)
1e-06
1e-07
1e-08
1e-09
0,0001
0,001
0,01
0,1
1
Energy of the incident electron (eV)

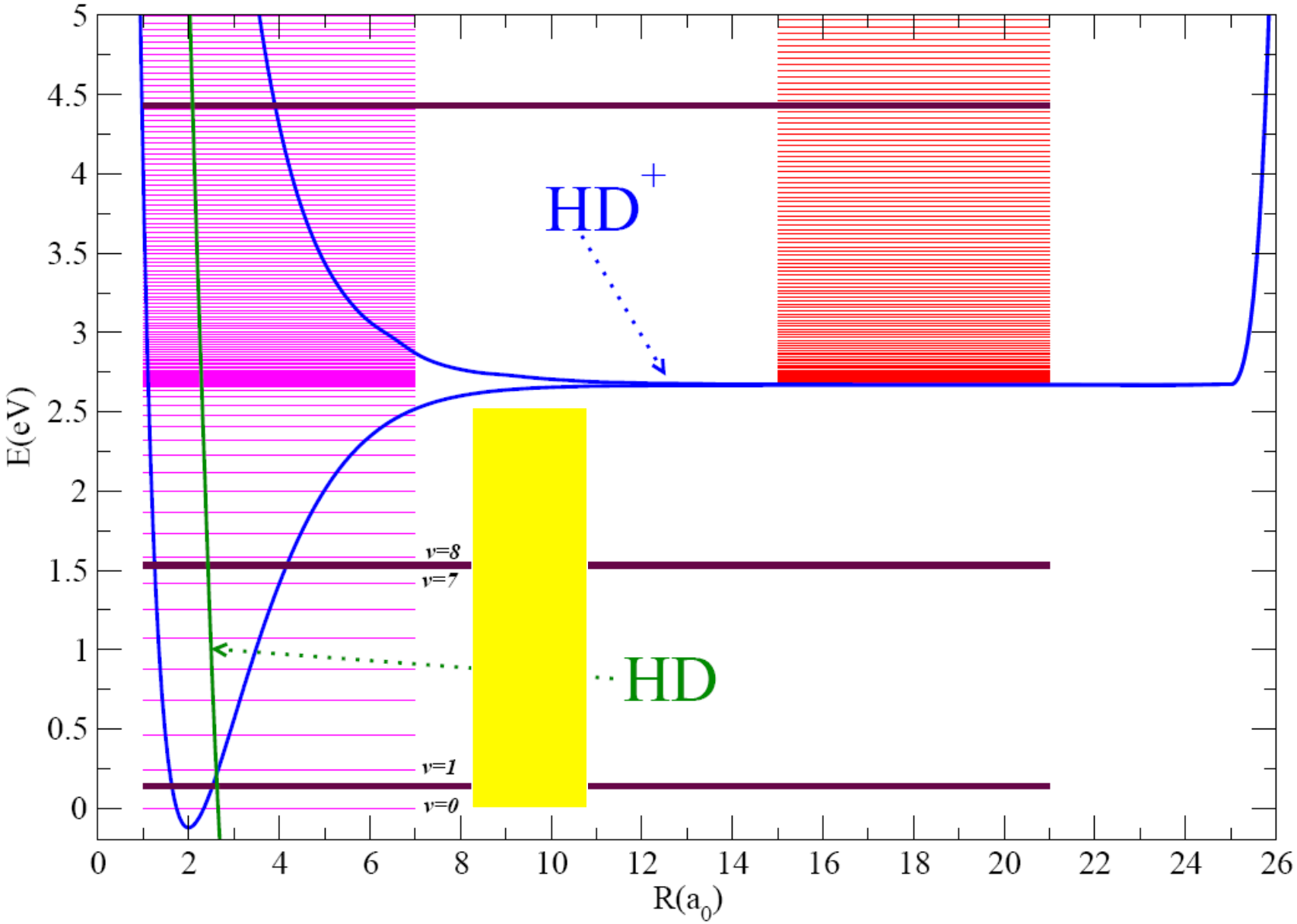
ACCURATE computations: VIBRATIONAL interactions

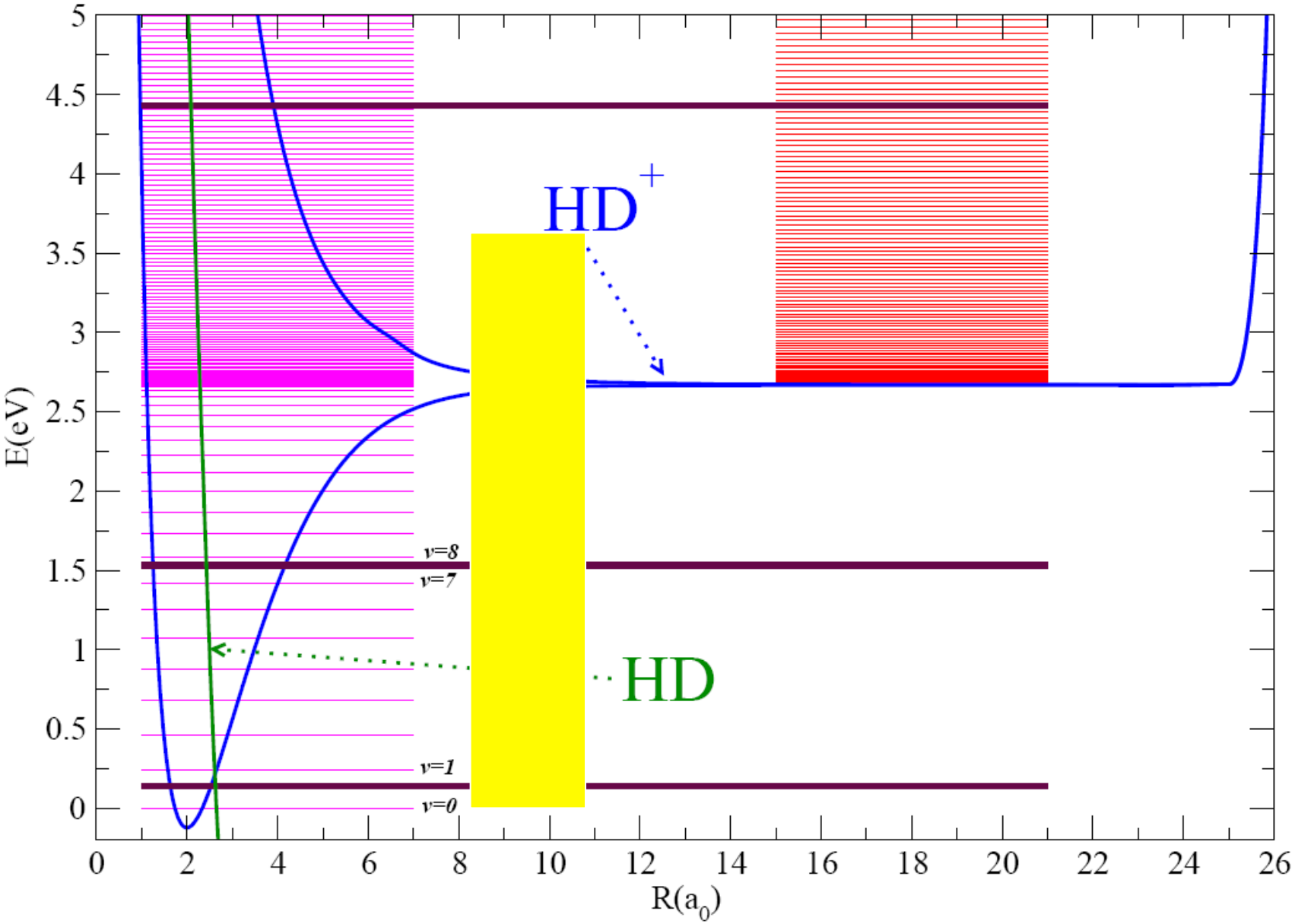
H_2^+

et isotopomers

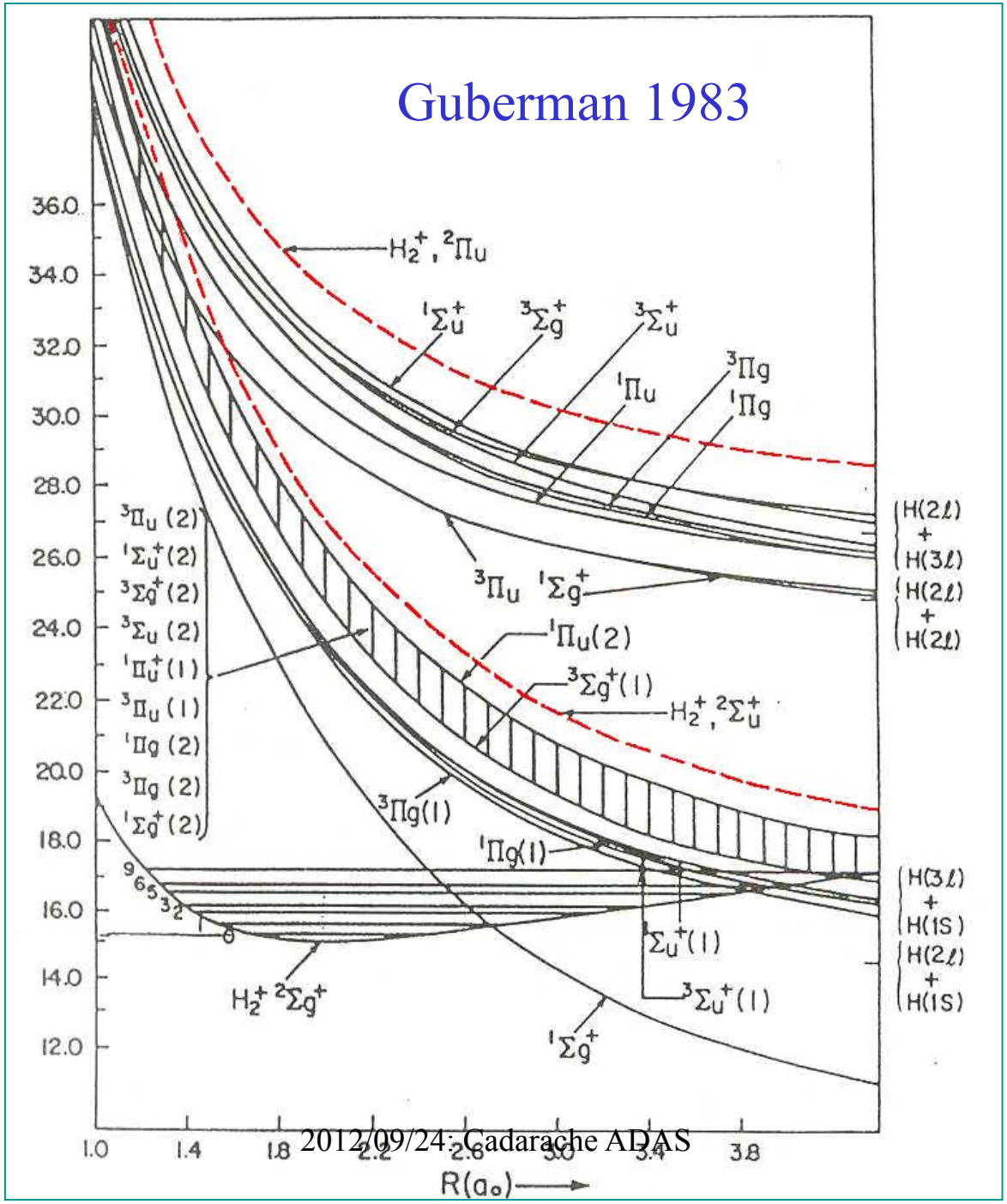
2012/09/24: Cadarache ADAS

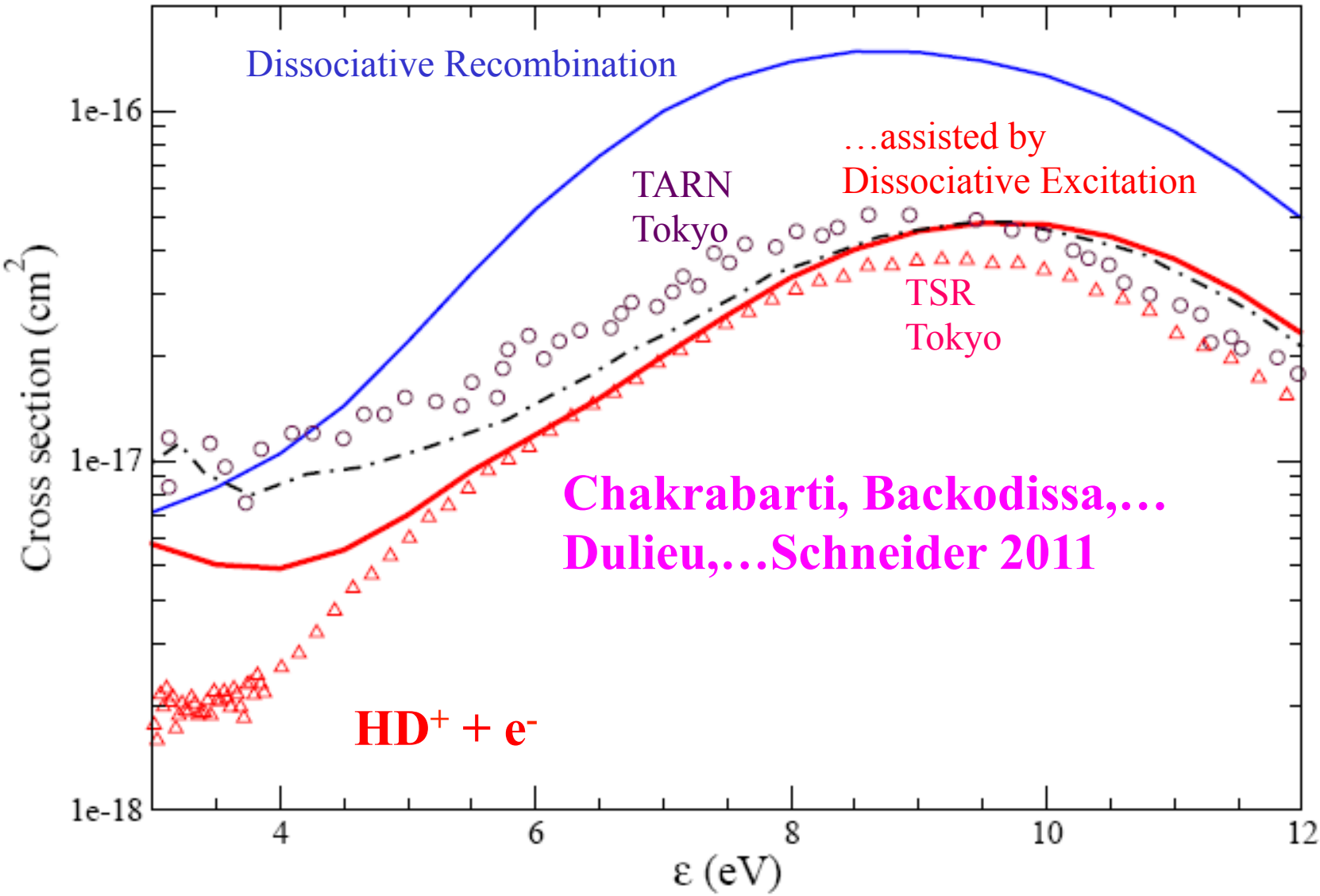

E(eV)
R(a0)
HD+
HD
v=8
v=7
v=1
v=0
0
0.5
1
1.5
2
2.5
3
3.5
4
4.5
5
0
2
4
6
8
10
12
14
16
18
20
22
24
26


E(eV)
R(a0)
HD+
HD
v=8
v=7
v=1
v=0


HD+
HD
v=8
v=7
v=1
v=0
E(eV)
R(a0)
0
0.5
1
1.5
2
2.5
3
3.5
4
4.5
5
0
2
4
6
8
10
12
14
16
18
20
22
24
26

Guberman 1983





Dissociative recombination of high-energy electrons with diatomic molecular cations: application to H_2^+ and HD^+ ions

2012/09/24: Cadarache ADAS

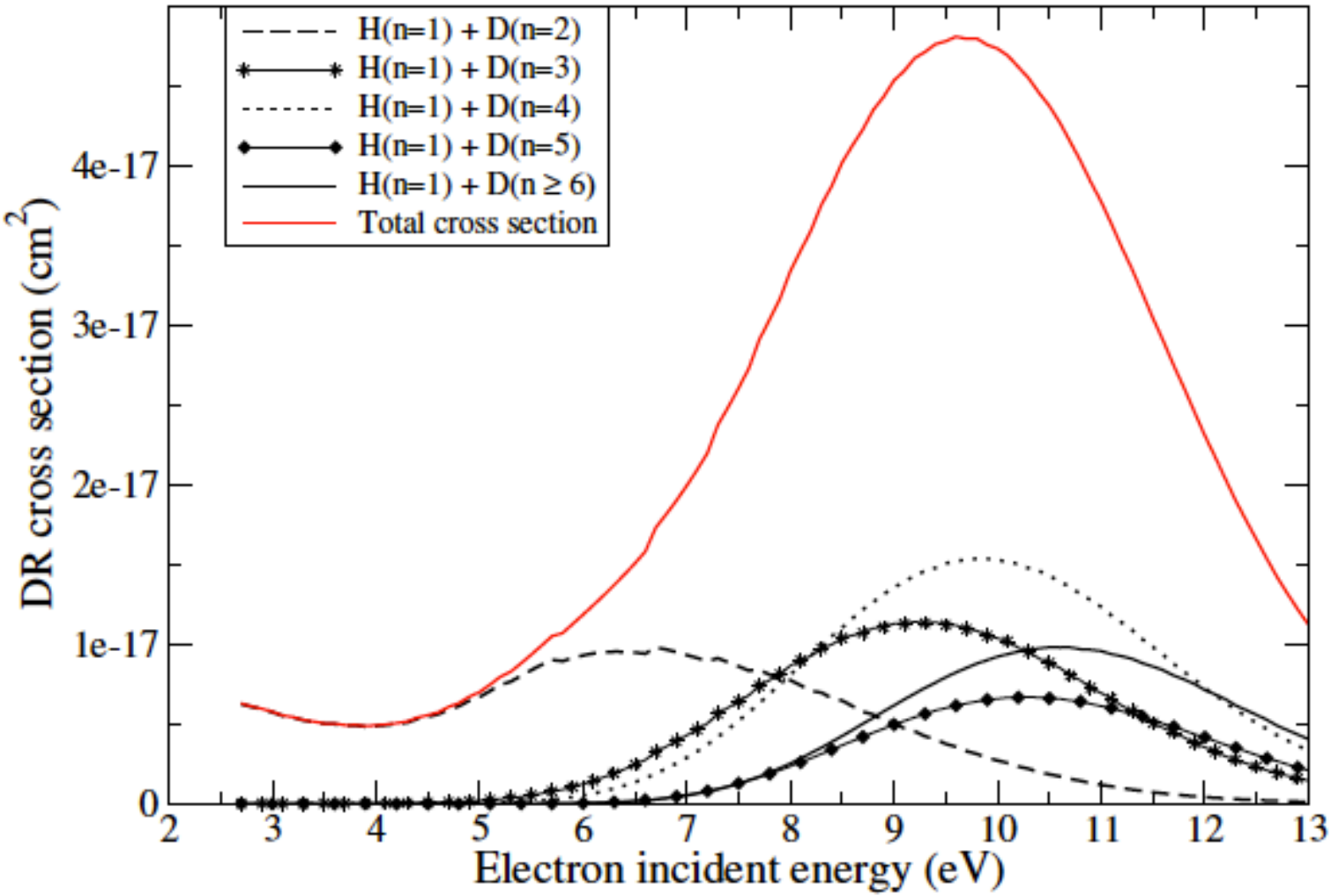


FIG. 6: (Colour online) Dissociative recombination of ground state HD^+ ion, contributions of each pair of atomic states resulting from dissociation. 'H(n=1) + D(n=2,3,4,...)' stands for 'D(n=1) + H(n=2,3,4,...)' too.

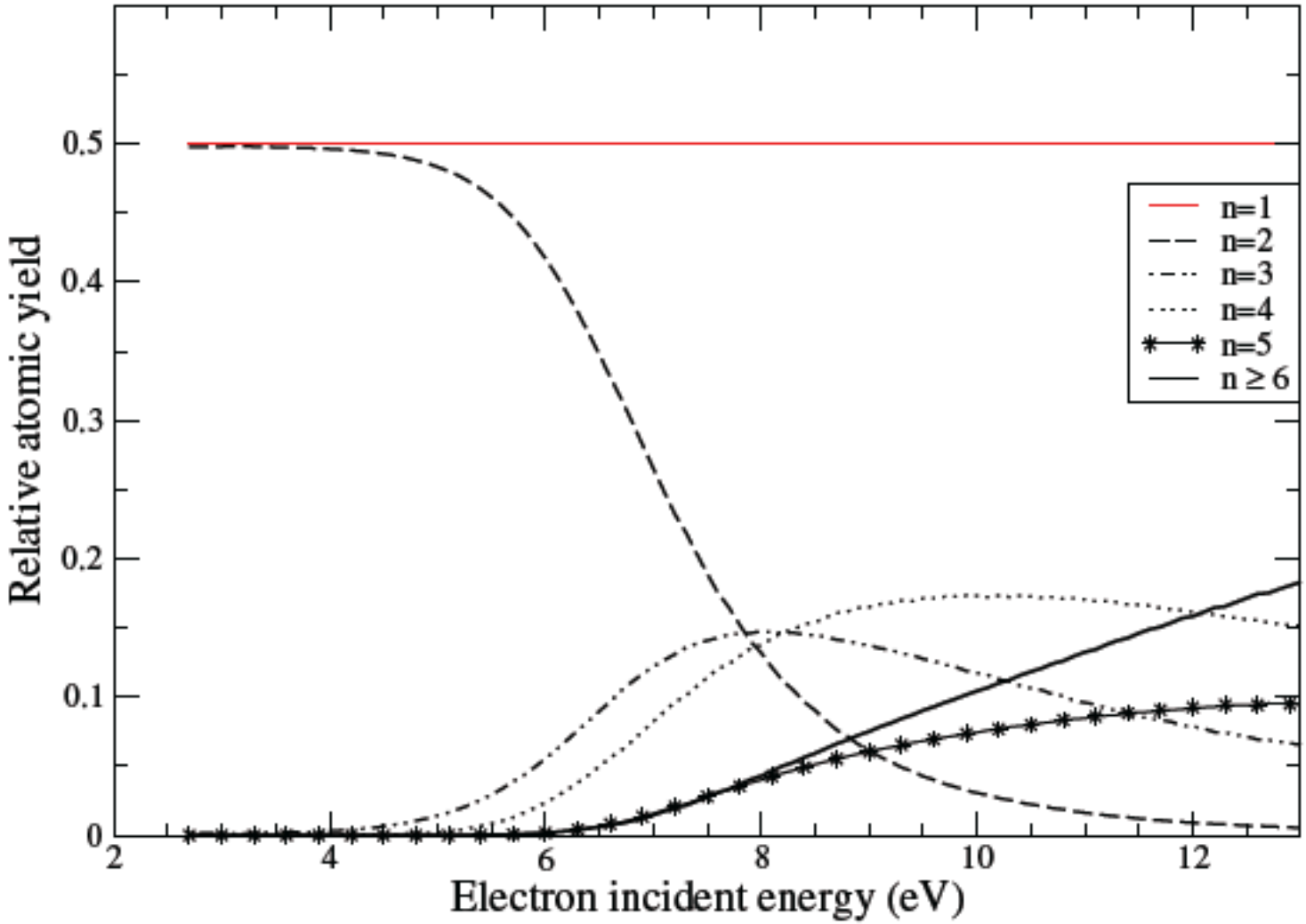


FIG. 7: (Colour online) Dissociative recombination of ground state HD^+ ion, relative atomic H(n) or D(n) final states yields.

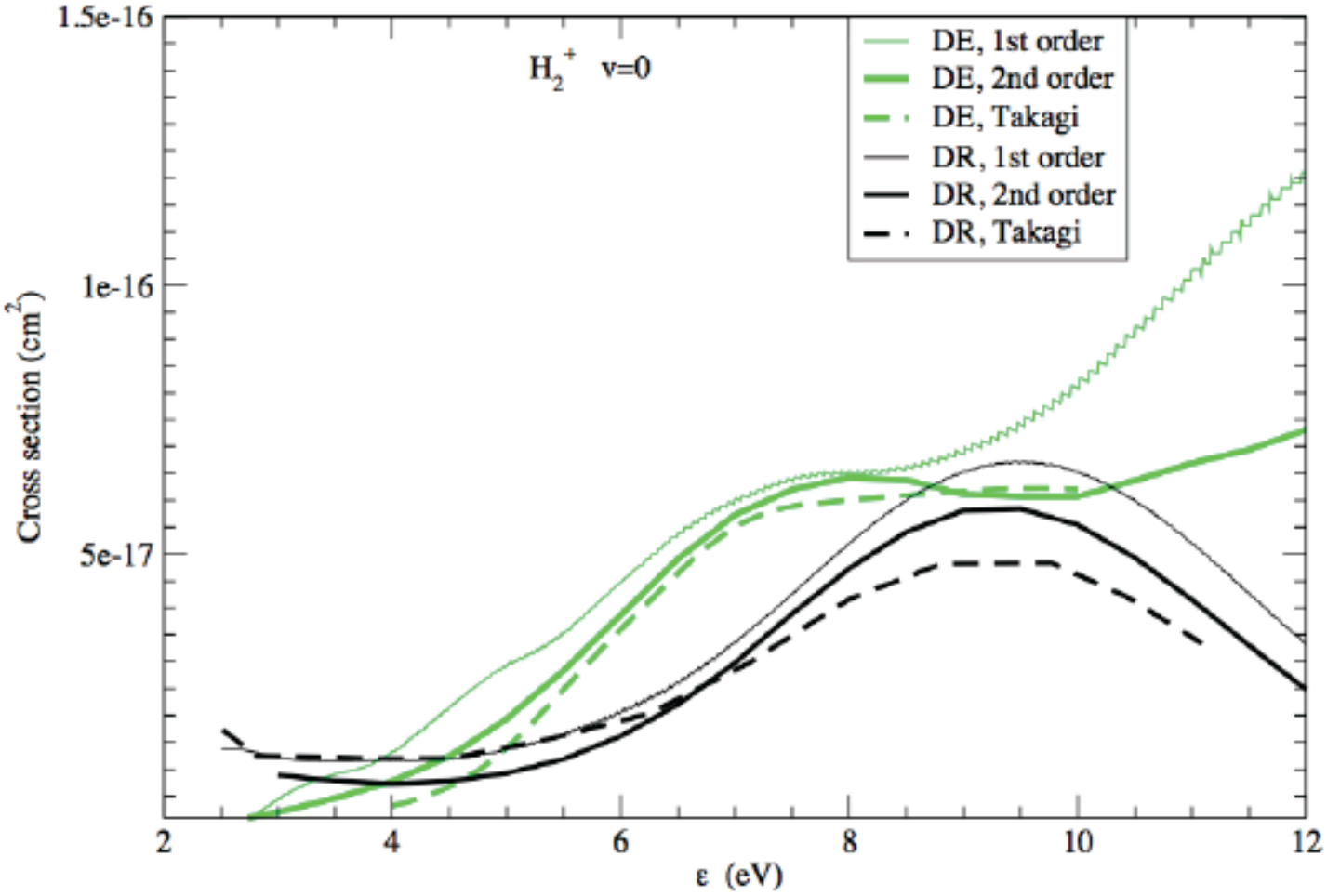


FIG. 8: (Colour online) Dissociative recombination - black curves - and dissociative excitation - green curves - of the H_2^+ molecular ion. Thin/thick continuous curves: first order/second order calculations. Dashed curves: theoretical results of Takagi [22].

The same doubly-excited Rydberg states:

IOP PUBLISHING JOURNAL OF PHYSICS B: ATOMIC, MOLECULAR AND OPTICAL PHYSICS

J. Phys. B: At. Mol. Opt. Phys. **44** (2011) 215203 (12pp) doi:10.1088/0953-4075/44/21/215203

Collisional production of fast metastable hydrogen atoms from cold H_2: toward twin atoms

Aline Medina[1,2], G Rahmat[1], C R de Carvalho[1,2], Ginette Jalbert[1,2], F Zappa[3], R F Nascimento[2,4], R Cireasa[1,5], N Vanhaecke[1], Ioan F Schneider[6], N V de Castro Faria[1,2] and J Robert[1]

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[2] Instituto de Física, UFRJ, Cx. Postal 68528, Rio de Janeiro, RJ 21941-972, Brazil
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2012/09/24: Cadarache ADAS

H_2 dissociation

- **H_2 beam (supersonic nozzle beam)**

+

- **Pulsed electron beam**

- **H (2s) + H (2s) → correlated pair of metastable states with opposite velocity and correlated spin polarization (Twin Atoms)**

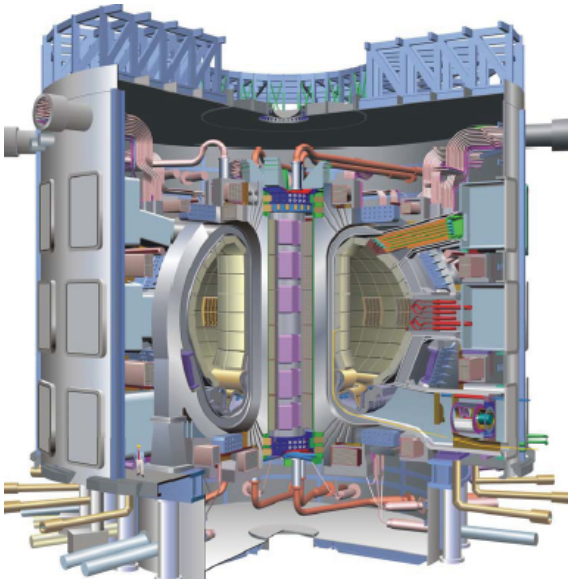
BeH^+

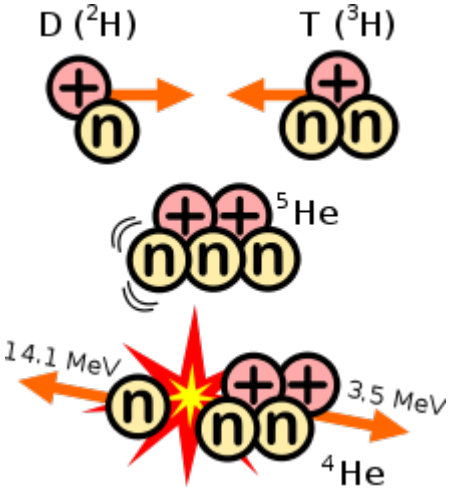
et isotopomers

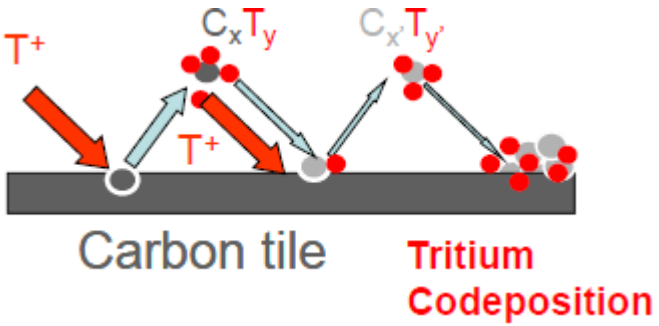
2012/09/24: Cadarache ADAS

Dissociative recombination, dissociative excitation and vibrational excitation at the walls of the fusion plasma devices (ITER project)

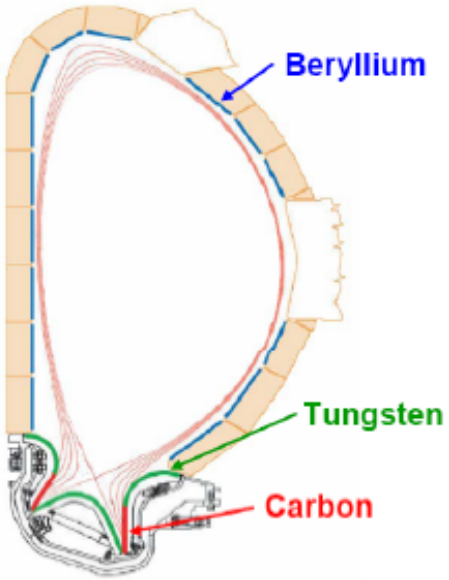
Fuel: H, D, T







Walls coated with Be



Molecules: BeH/BeH^+, BeC/ BeC^+, CH/CH^+, CH_4/CH_4^+, CH_2 /CH_2^+, H_2^+, HeH^+, and isotopomers (H→D, H→T)

IOP PUBLISHING PLASMA PHYSICS AND CONTROLLED FUSION

Plasma Phys. Control. Fusion **54** (2012) 035012 (11pp) doi:10.1088/0741-3335/54/3/035012

State-to-state electron impact cross sections for BeH^+ molecular ions in ITER-like fusion edge plasmas with Be walls

R Celiberto[1,2], R K Janev[3,4] and D Reiter[4]

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[2] Institute of Inorganic Methodologies and Plasmas, CNR, 70125 Bari, Italy
[3] Macedonian Academy of Sciences and Arts, PO Box 428, 1000 Skopje, Macedonia
[4] Institute of Energy and Climate Research - Plasma Physics, Forschungszentrum Jülich GmbH
Association EURATOM-FZJ, Partner in Trilateral Euregio Cluster, 52425 Jülich, Germany

2012/09/24: Cadarache ADAS

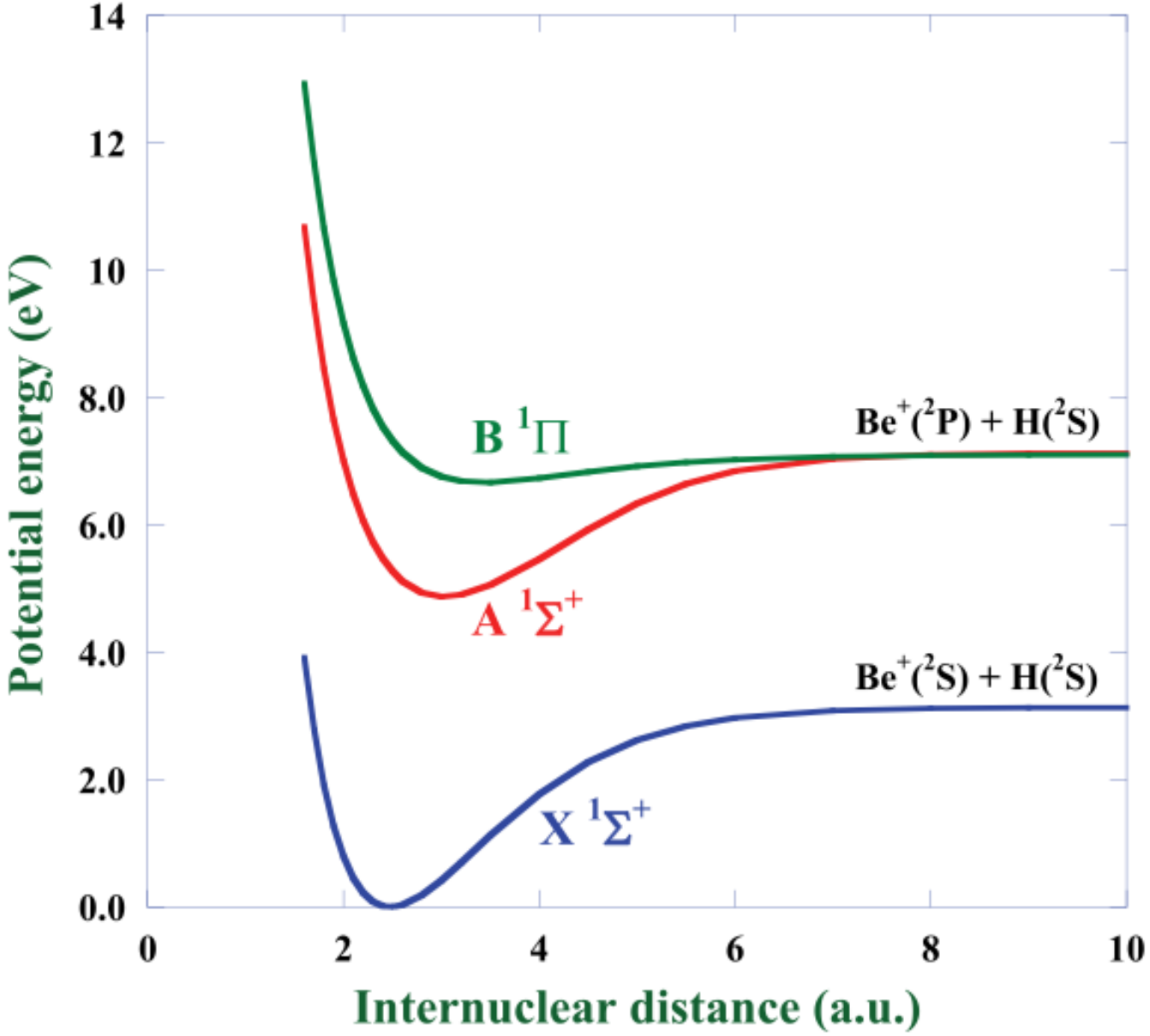


Figure 1. BeH^+ potential energy curves, as a function of the internuclear distance, for the electronic states $X\ ^1\Sigma^+$, $A\ ^1\Sigma^+$ and $B\ ^1\Pi$ correlating with the atomic states indicated in the figure [18].

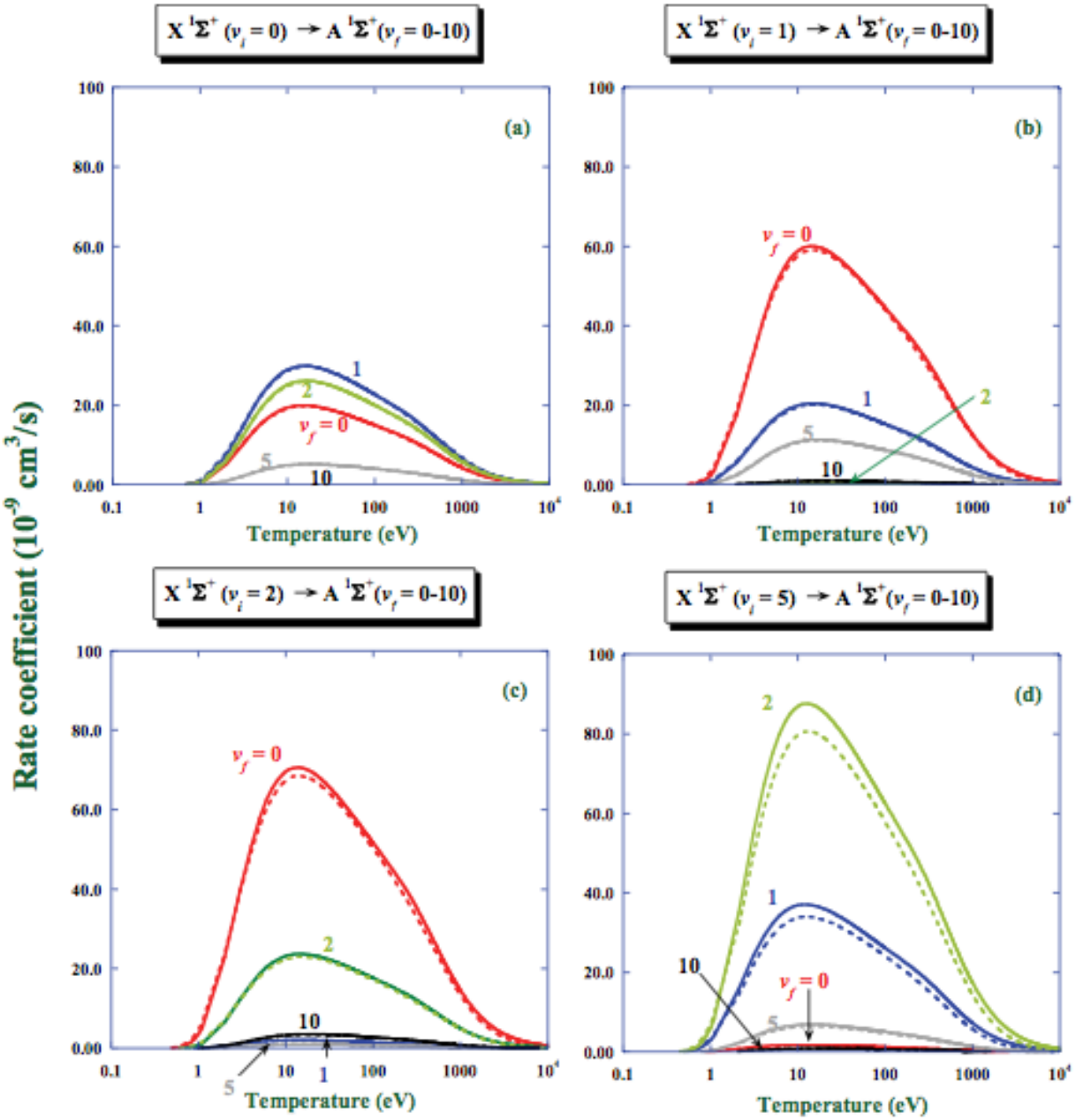
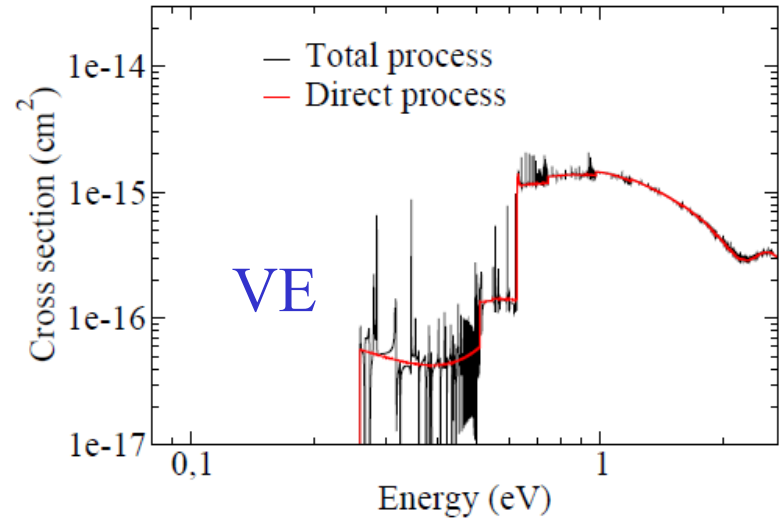
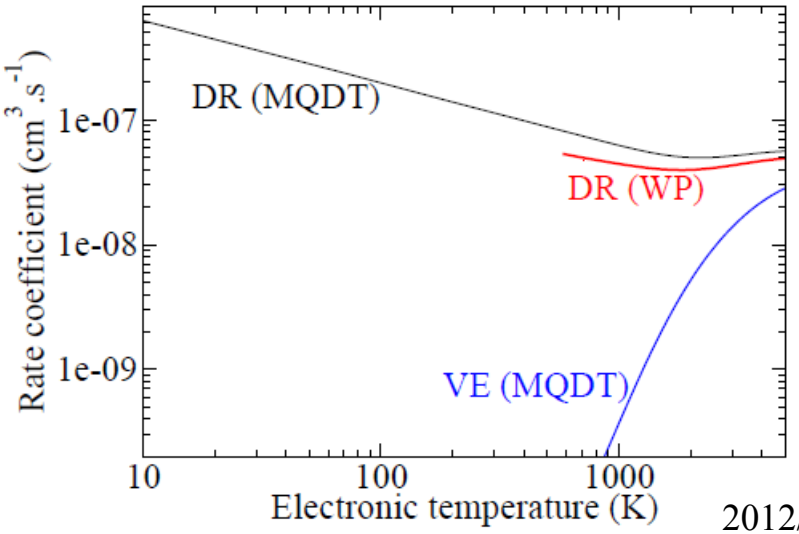


Figure 7. Rate coefficients for the $X(v_i) \rightarrow A(v_f)$ transitions involving the indicated vibrational levels. Full lines: calculated data; dashed lines: results of the scaling formula, equation (8). The horizontal and vertical scales are kept the same for easy comparison.

BeH/BeH^+

Niyonzima *et al* 2012, collaboration with A. Larson & A. Orel





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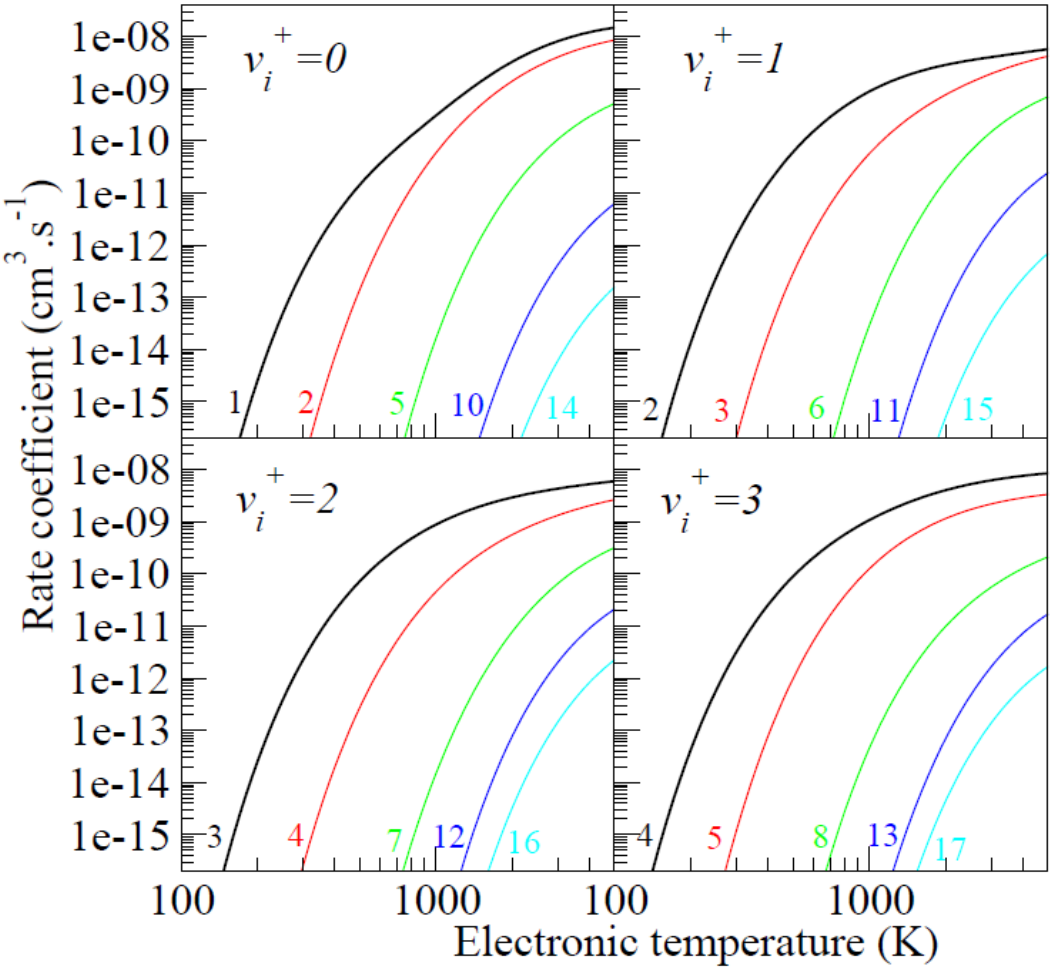
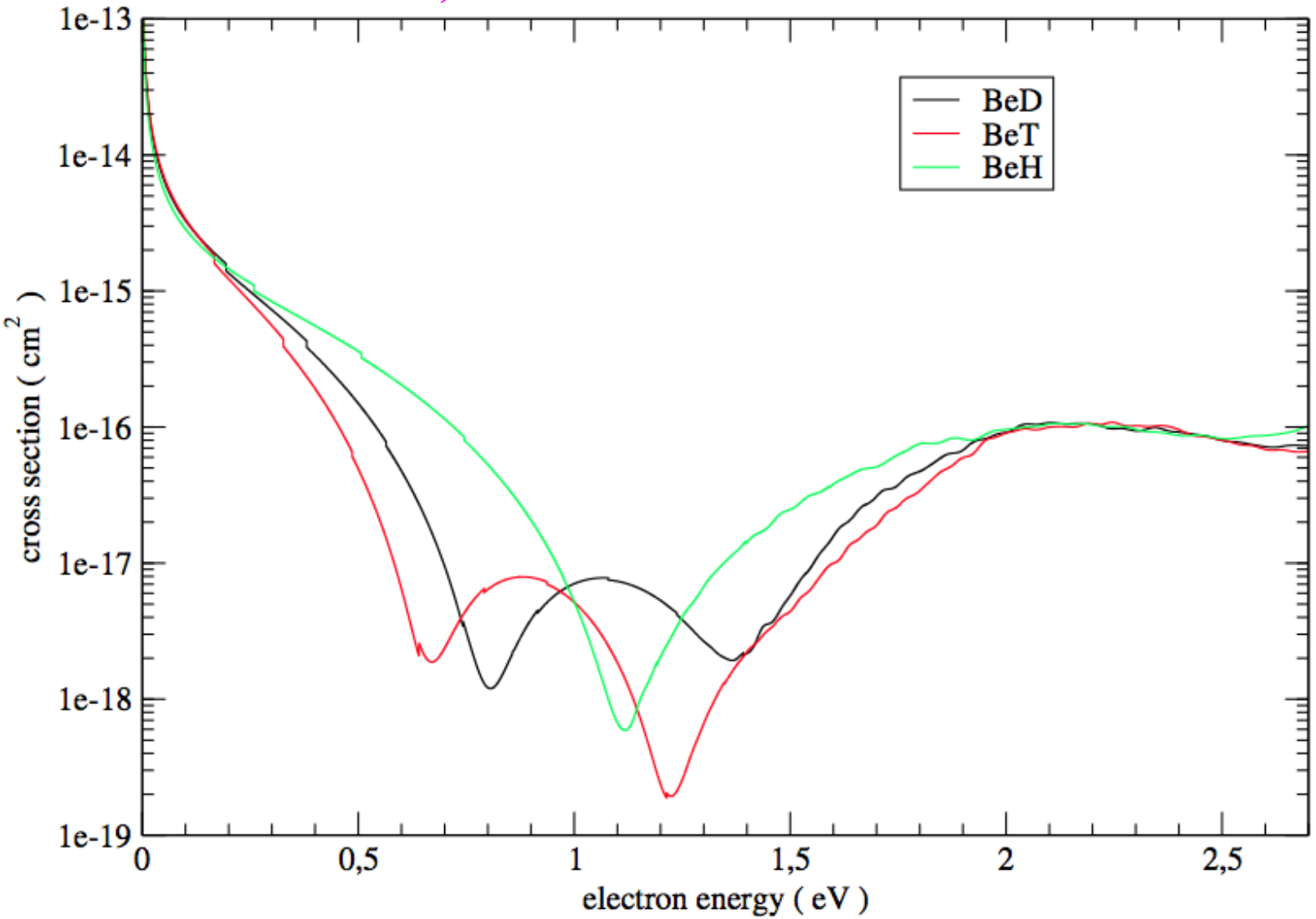


Isotopic effects in direct dissociative recombinaison of BeH^+ for $^2\Pi$

Lanza *et al* 2012, collaboration with A. Larson & A. Orel



ATOM/MOLECULE collisions:

The role of the REACTIVITY

J. Phys. B: At. Mol. Opt. Phys. **32** (1999) 2415–2431. Printed in the UK PII: S0953-4075(99)00027-9

Elastic and vibrationally inelastic slow collisions: $H + H_2$, $H^+ + H_2$

Predrag S Krstić and David R Schultz

Physics Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831-6373, USA

Received 9 December 1998

Abstract. We report on a comprehensive study of the scattering of hydrogen atoms on the ground electronic surface of hydrogen molecules in the range of centre of mass energies 0.1–100 eV. Differential and integral elastic cross sections, the related transport cross sections, and vibrationally inelastic cross sections starting from both ground and excited vibrational states, are calculated using a fully quantal, coupled-channel approach in a truncated vibrational basis set, while the rotational dynamics of H_2 is treated with the infinite order sudden approximation prescription. For comparison and to highlight the major physical mechanisms revealed in these collisions, a parallel study is carried out for scattering of protons on hydrogen molecules.

INSTITUTE OF PHYSICS PUBLISHING PLASMA SOURCES SCIENCE AND TECHNOLOGY

Plasma Sources Sci. Technol. **15** (2006) S62–S66 doi:10.1088/0963-0252/15/2/S08

State-to-state cross sections for H_2 and its isotopic variants

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[2] Dipartimento di Ingegneria Civile ed Ambientale, Politecnico di Bari, Italy
[3] Dipartimento di Chimica, Università di Bari, Italy

Received 5 October 2005, in final form 27 February 2006
Published 24 April 2006
Online at stacks.iop.org/PSST/15/S62

Abstract
State-resolved cross sections for electron–H_2 and H–H_2 collision-induced processes have been calculated, using semiclassical and quasiclassical approaches, respectively. Corresponding results for the deuterium system are presented, introducing mass-scaling relations.

2012/09/24: Cadarache ADAS

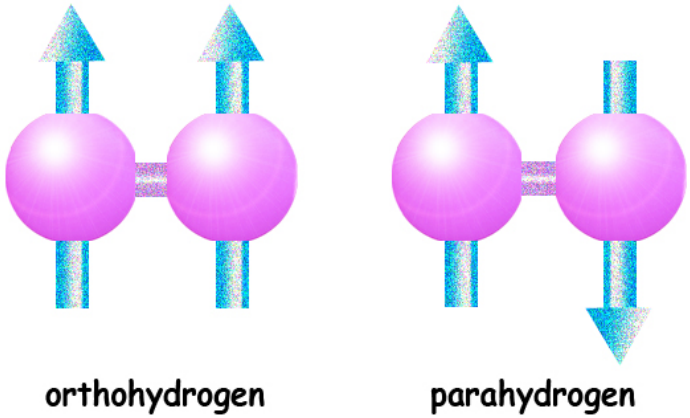
Rotational excitation of H_2 by H: reactivity via ortho-para transitions

Beyond the rigid rotor model (proton exchange)

ortho-para-H_2 ratio in plasmas

H_2 :

ortho/para ratio : key parameter for the physical chemistry of hydrogen dominated plasmas



H_2 is a homonuclear molecule (no dipole moment) ->
Ro-vibrational **transitions with Δj=+/- 2**
Inelastic collisions do not change ortho/para ratio

⇩

ortho-para-H_2 conversion in plasmas ?

H + p-H_2 -> H + o-H_2 $H^+ + p\text{-}H_2 \rightarrow H^+ + o\text{-}H_2$ $H_3^+ + p\text{-}H_2 \rightarrow H_3^+ + o\text{-}H_2$

H_3: three undistinguishable protons, symetry D_{3h}

- Wavefunctions properly antisymmetrized with the interchange of the protons (Miller 1969)
- Post-symetrization (protons as distinguishable) :

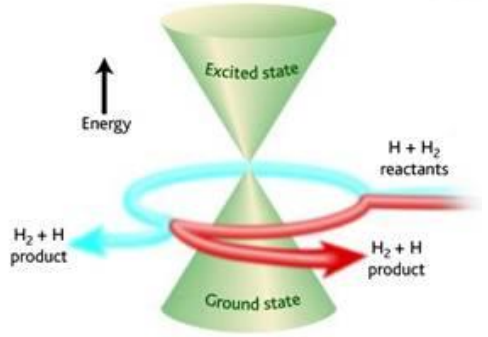
Rotational excitation of H_2 by H

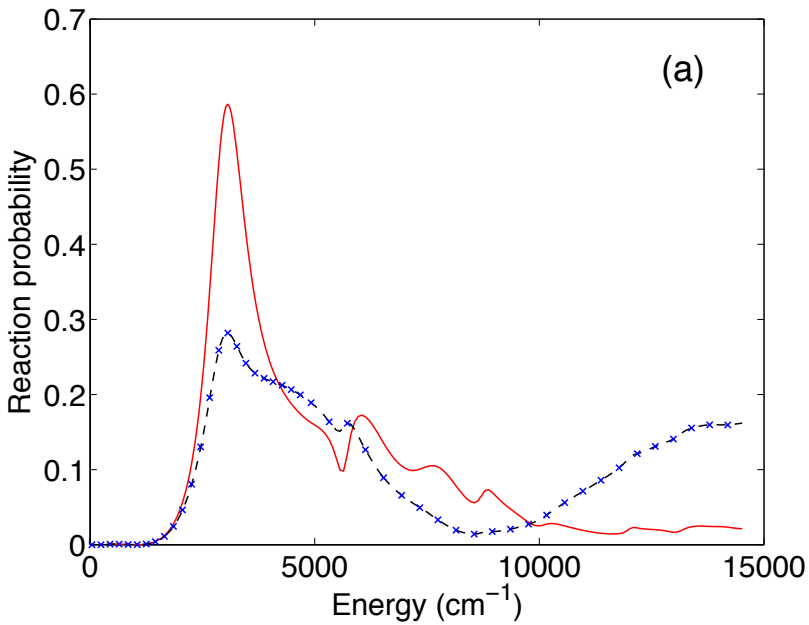
Geometric phase (GP) effect and postsymmetrisation validation

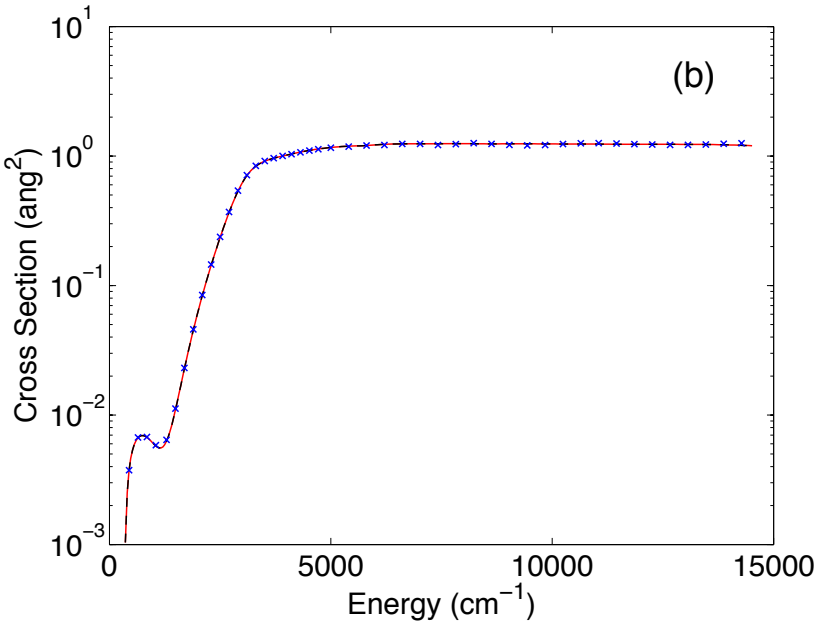
Geometric phase effect:
the ground electronic state of H_3 conically intersects the first excited state of H_3: sign change of the electronic wavefunction

⇩

Sign change of the diffusion matrice

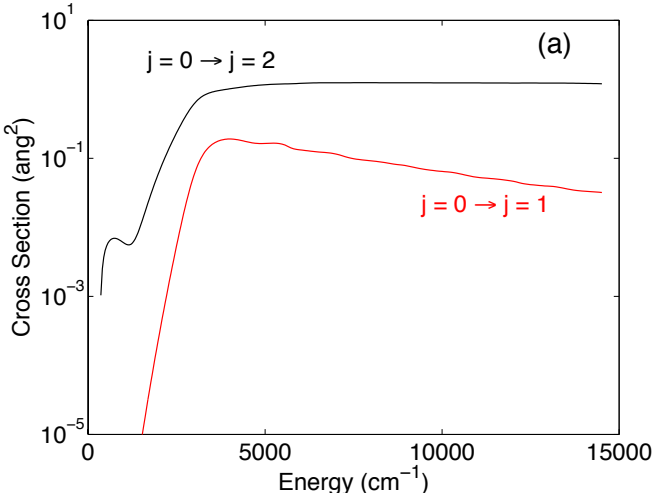


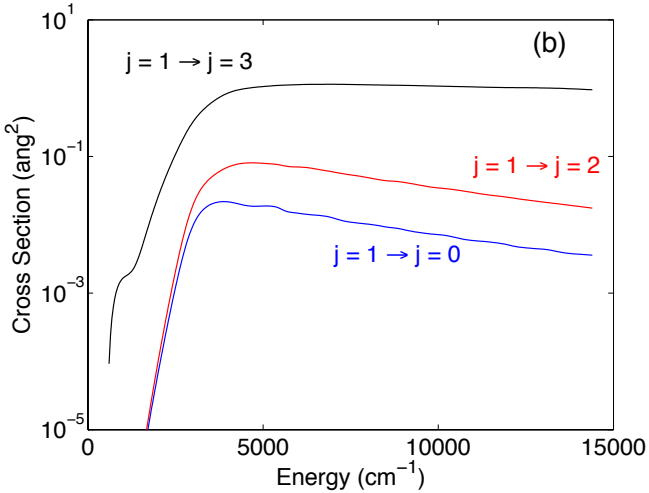




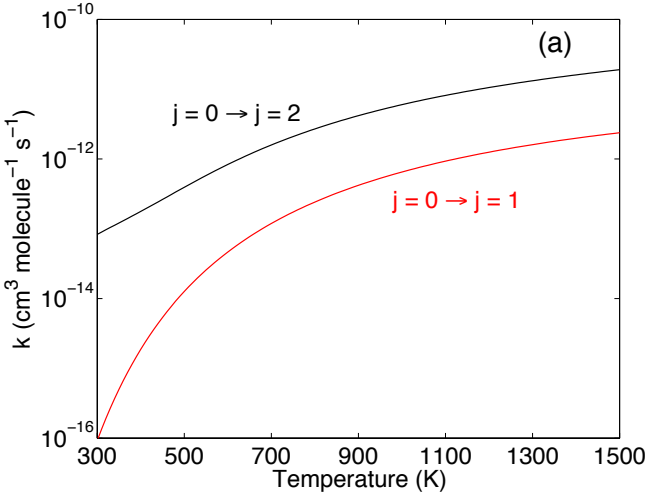
(a) Probablilities for the $H+H_2(j=0)\rightarrow H+H_2(j'=2)$ reaction for J = 0.
(b) $H+H_2(j=0)\rightarrow H+H_2(j'=2)$ cross section: Dashed and continuous lines refer to the calculations excluding and including the GP respectively. Cross indicate the results obtained using the method of Miller (1969)

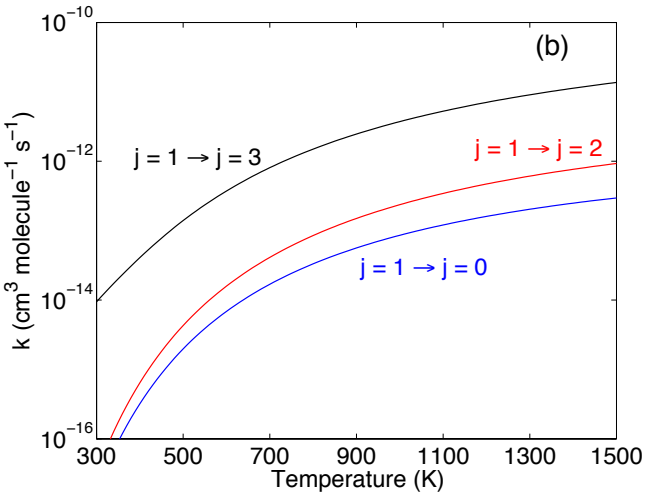
Rotational excitation of H_2 by H





Collisional enegy dependence of integral cross sections for the rotational excitation of p-H_2 (a) and o-H_2 (b) by H



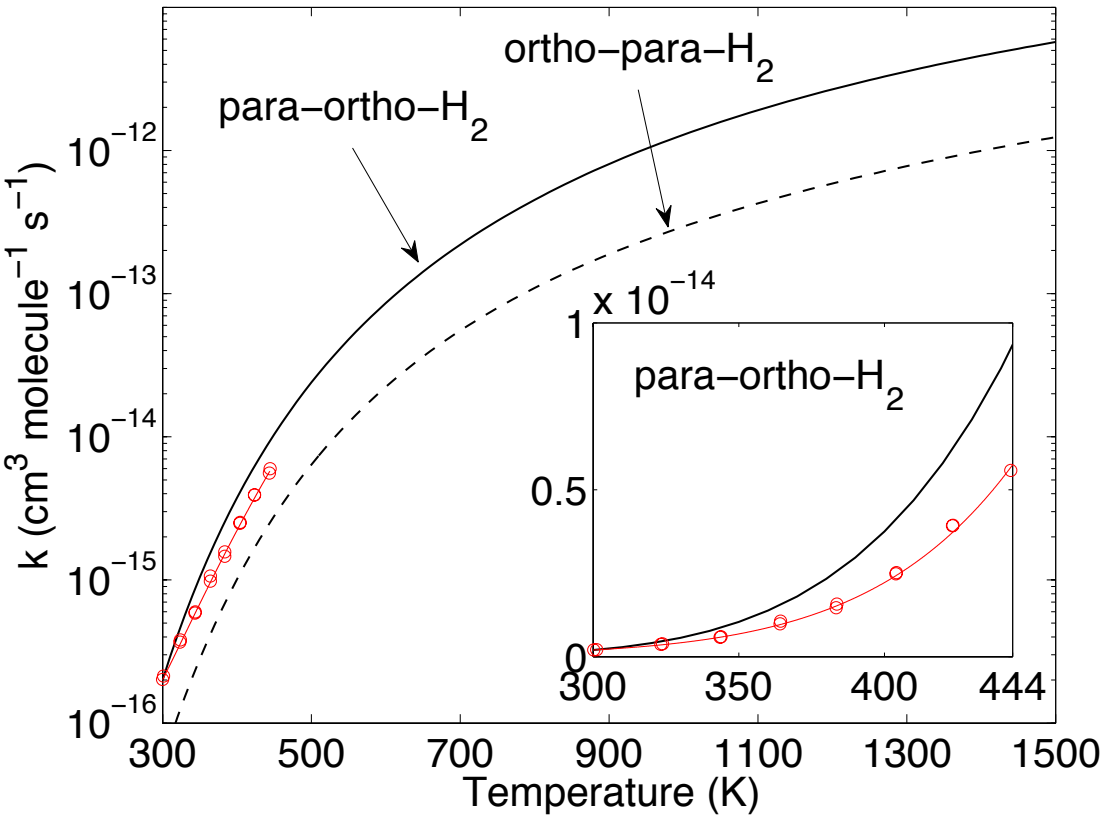


Temperature dependence of the rate coefficients for the rotational excitation of p-H_2 (a) and o-H_2 (b) by H

- The p-o-H_2 et o-p-H_2 transitions are slower than the transitions conserving nuclear spin
- Only one order of magnitude of difference for high temperature (T > 1000 K)
- Important process for hot plasmas

Rotational excitation of H_2 by H

Comparison with experiment



Temperature dependence of the rate coefficients for the p–o-H_2 and o–p-H_2 conversion. The line with circles indicates the experimental results of Schulz & Le Roy (1965).

Thermal average for the o-p-H_2 and p-o-H_2 conversion rates :

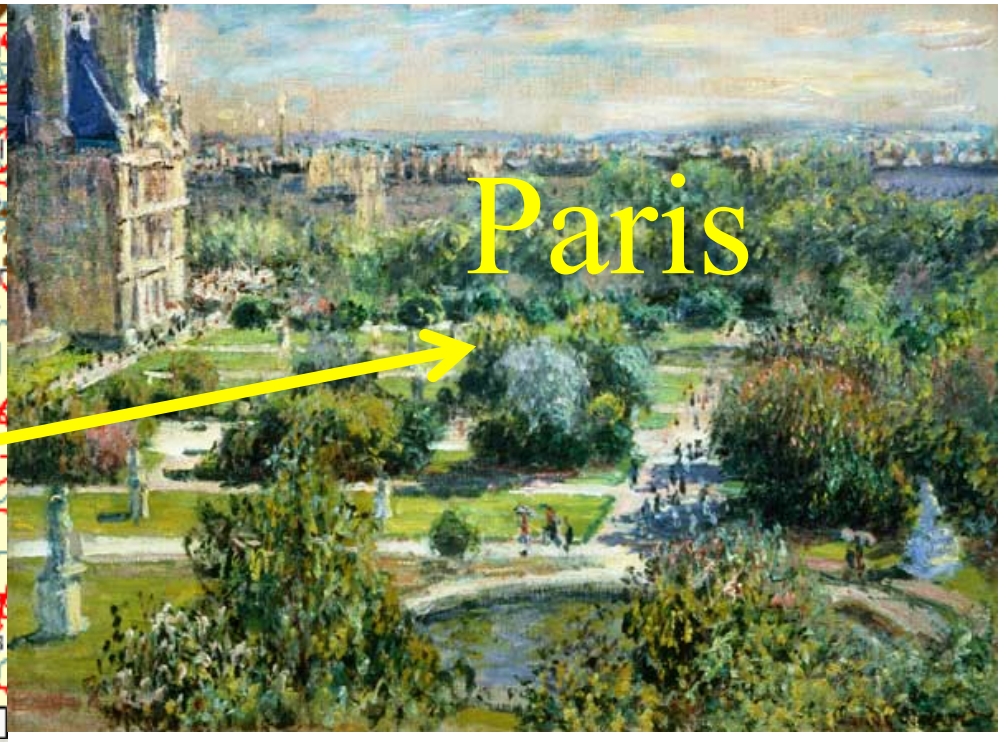
$$\bar{k}(j=0-8) = \frac{\sum_{j=0}^{8}(2j+1)e^{-\epsilon_j/kT}k(j)}{Q}$$

$$Q = \sum_j (2j+1)e^{-\epsilon_j/kT}.$$

- The rates increase rapidly with increasing temperature
- o-p-H_2 < p-o-H_2
- Very good agreement between theory and experiments

⇩

Accurate understanding of o-p-H_2 and p-o-H_2 conversion process



Coming:

9th International Conference on Dissociative Recombination: Theory, Experiment & Appliccations

7-12 july 2013, Paris

I. F. Schneider, O. Dulieu, J. Robert