Photofragmentation of H_2 and low-energy H^+/H^- collisions

J. Zs. Mezei, I. F. Schneider, Ch. Jungen, A. Larson

IAEA-Consultancy Meeting-Be2019
Wien, 06.06.2019.

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- Theoretical framework and mechanisms
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    - Photoionisation of H_2
    - Photodissociation of H_2
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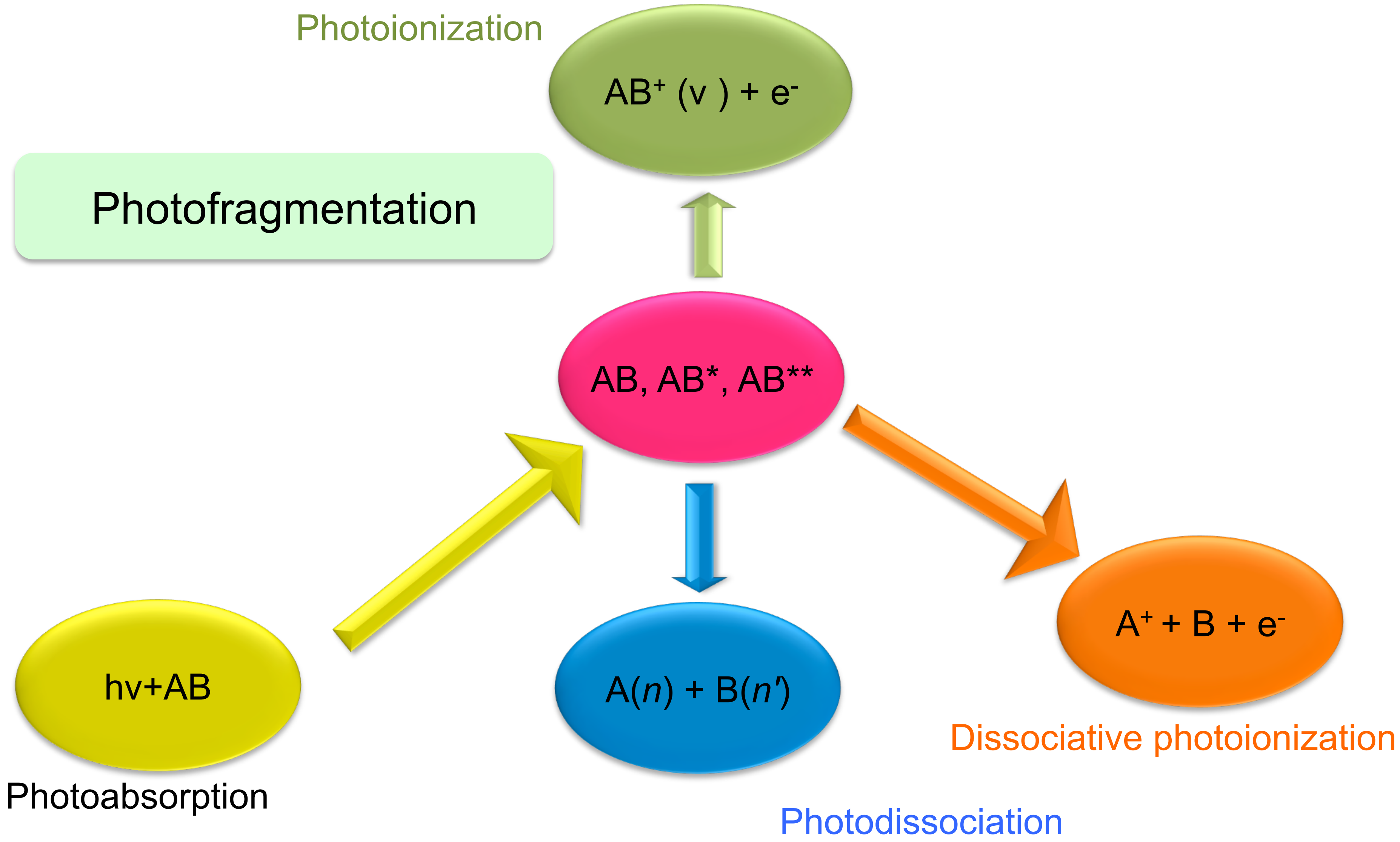
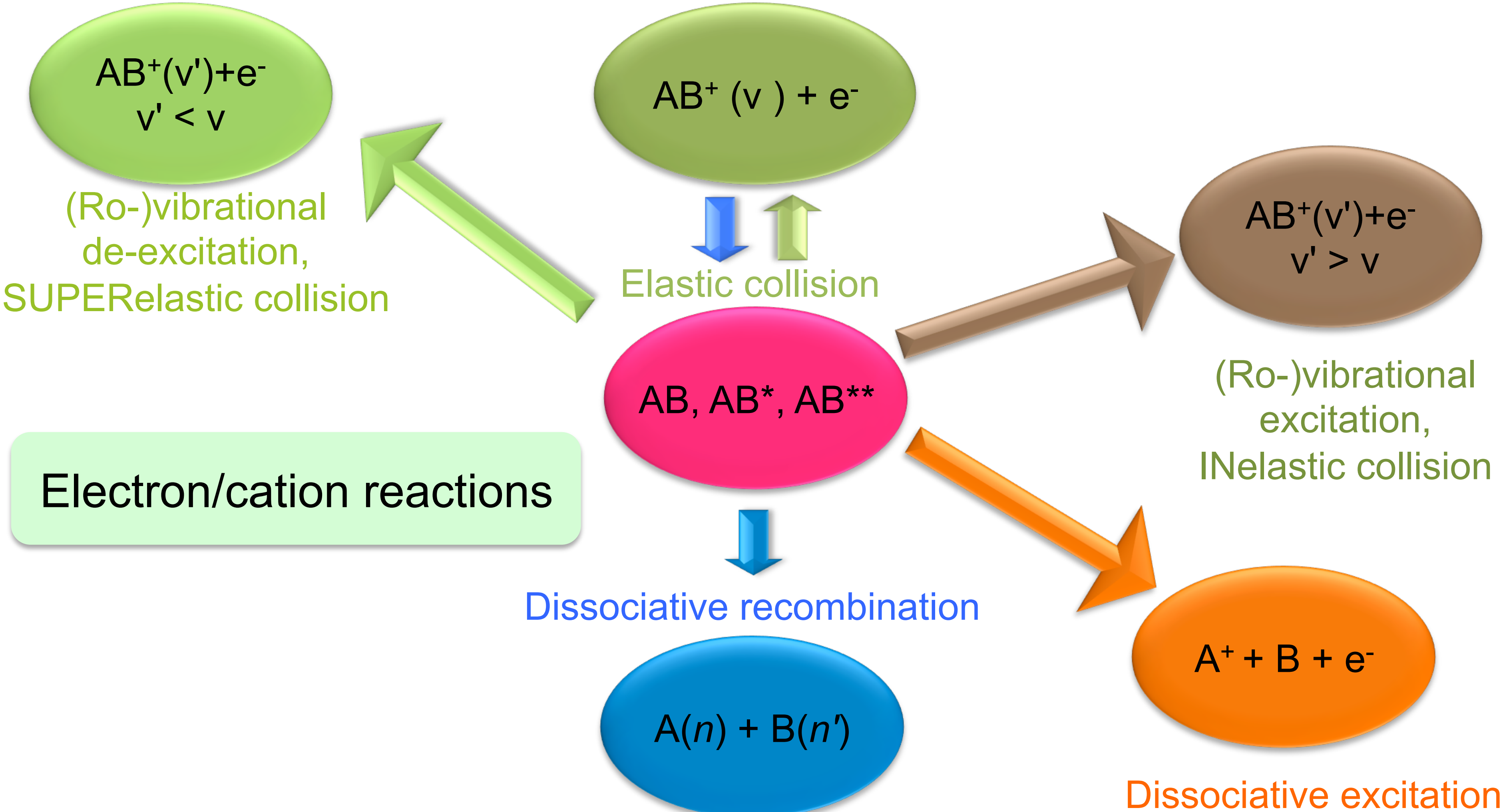
- Conclusions and future plans

Introduction: Elementary processes



Introduction: Elementary processes



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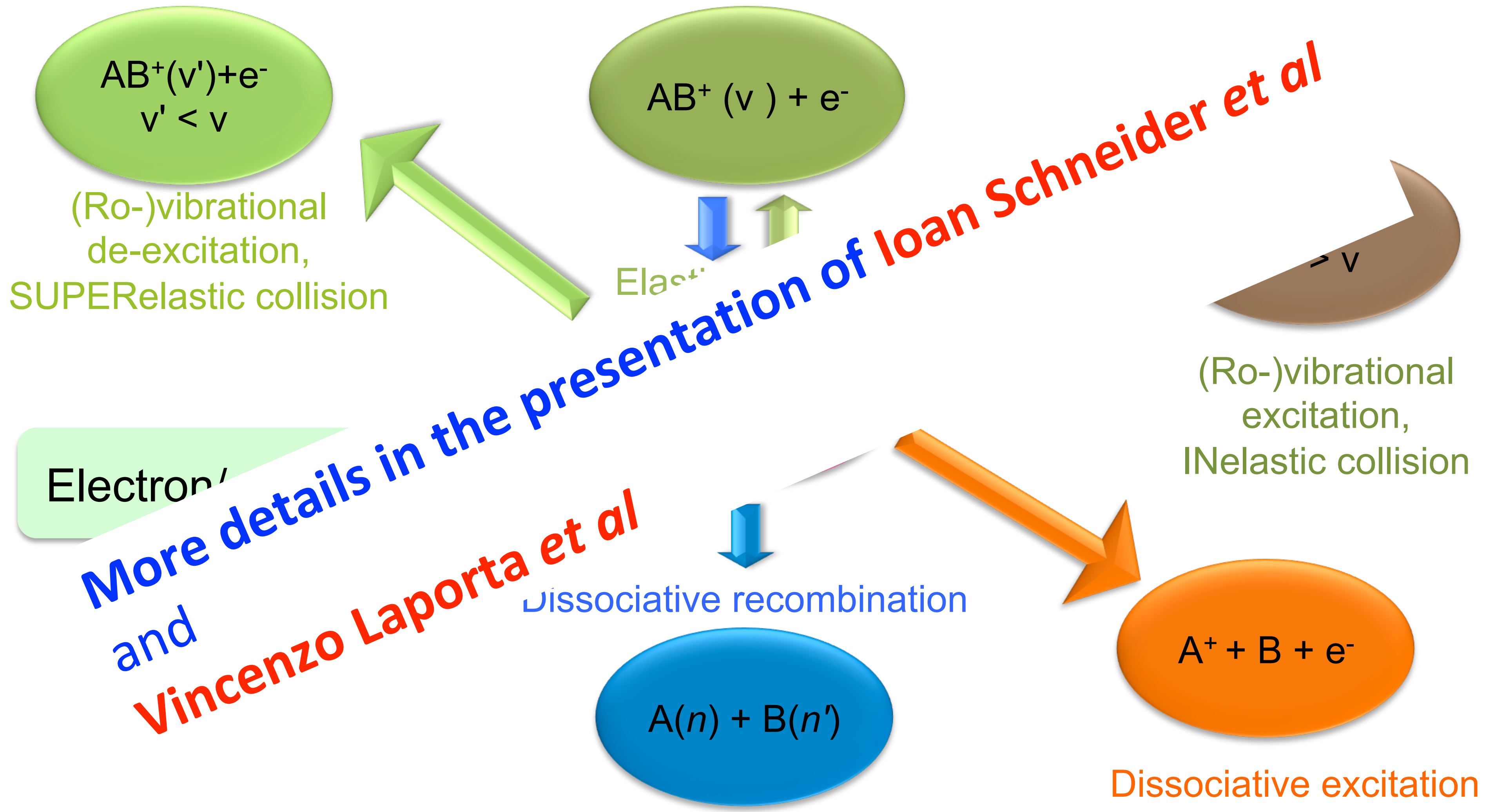

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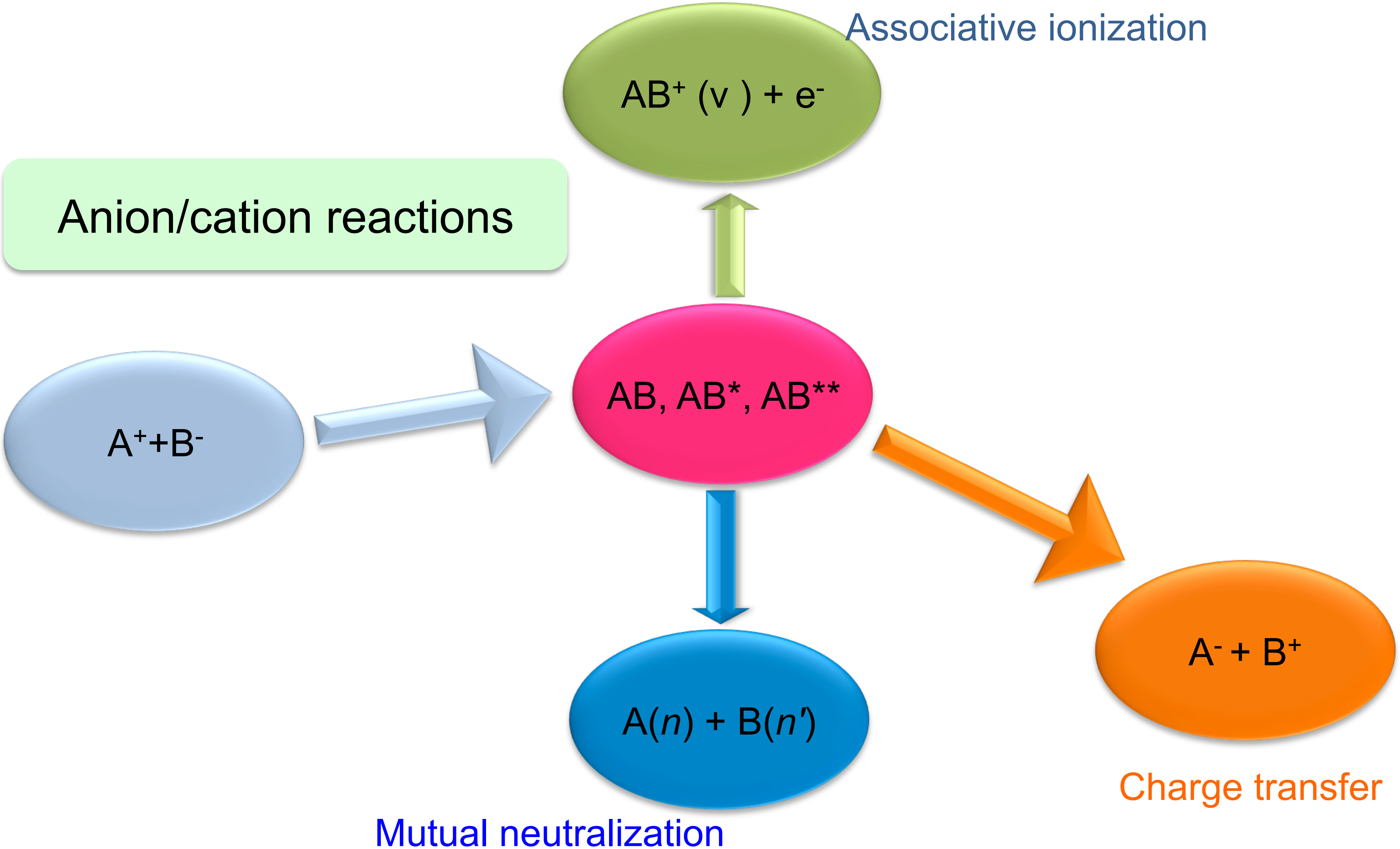

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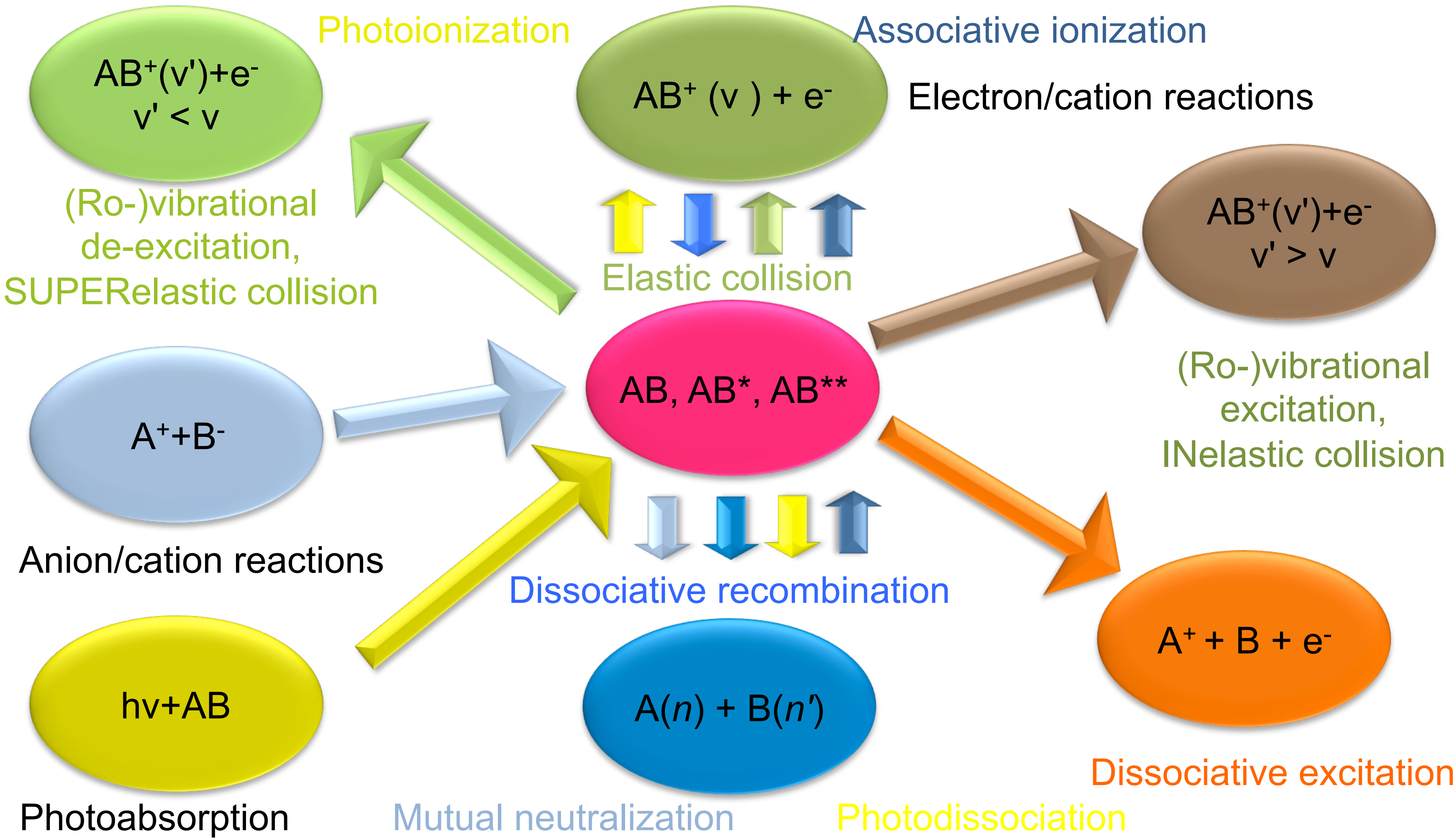
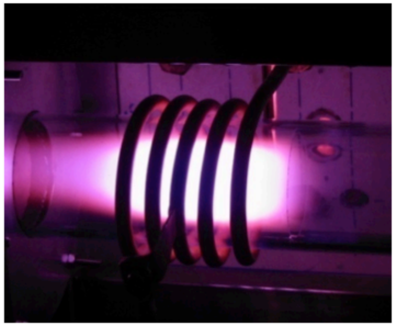


Introduction: Cold ionized media

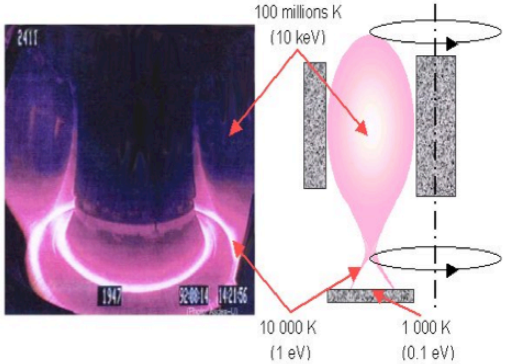
Interstellar molecular clouds

Planetary atmospheres

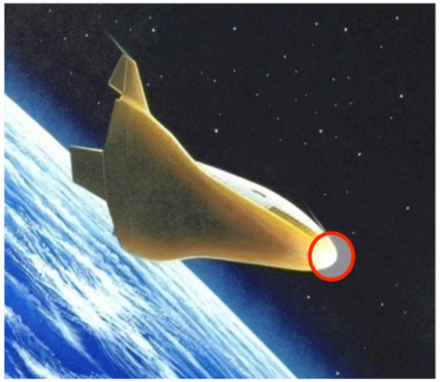
Cold laboratory plasmas



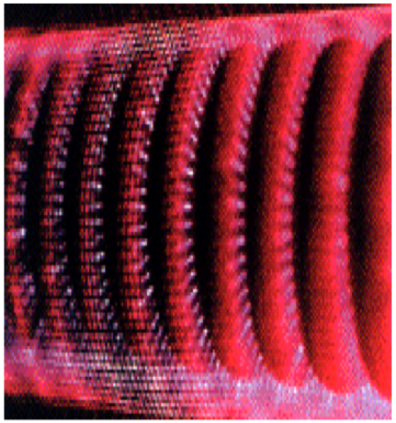
At the wall of the fusion devices (ITER) project



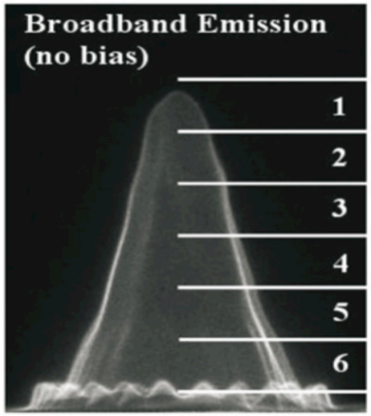
Hypersonic entry of spacecrafts



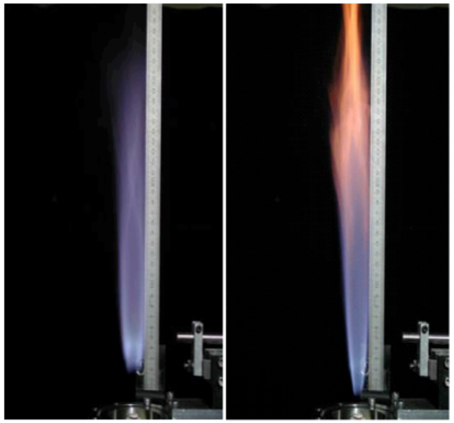
Plasma-assisted depollution



Electric-field-assisted combustion



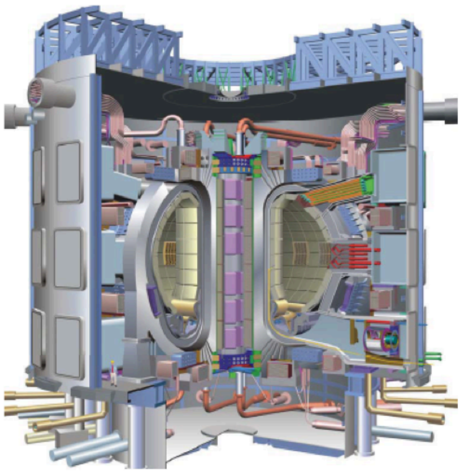
Plasma-assisted-combustion



Introduction: H_2 - ITER

b. Plasma de fusion thermonucléaire (JET, ITER)

Combustibles: H, D,



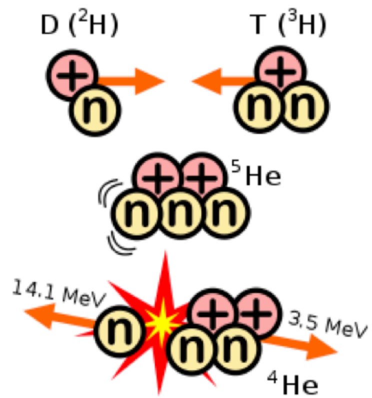
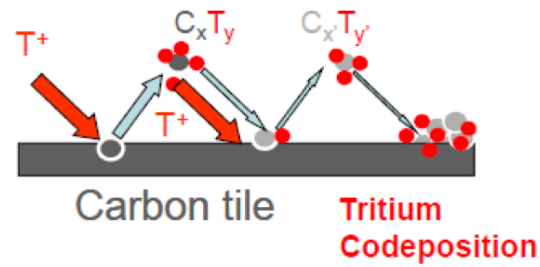
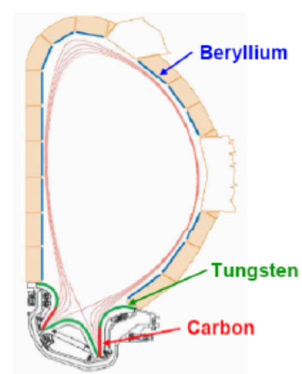


Fig.1. Coupe schématique de l'ITER

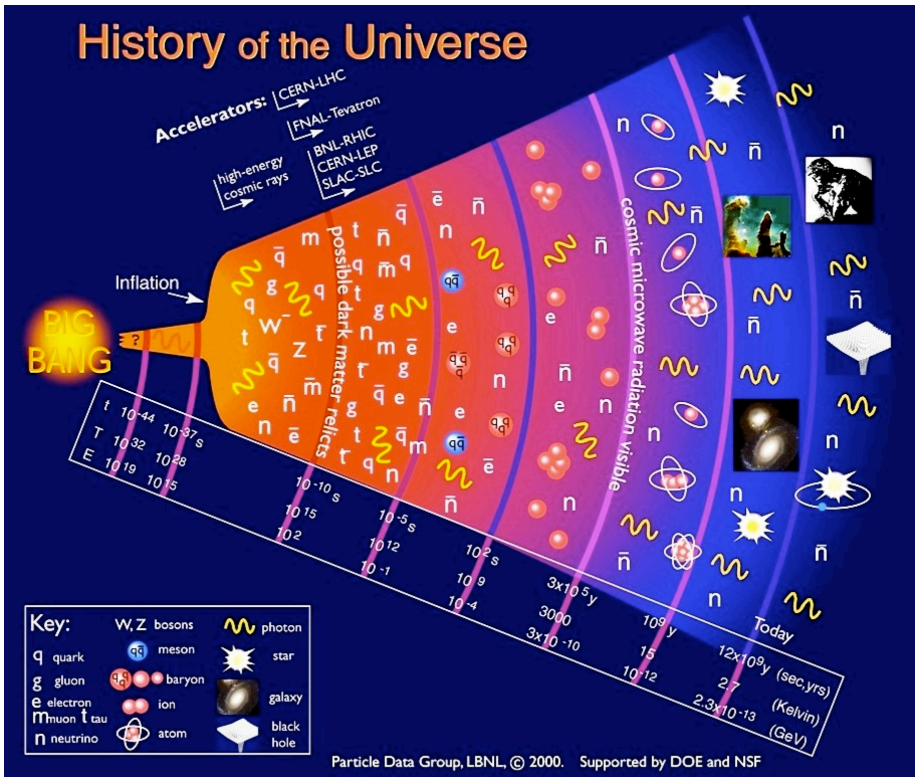
Formation des molécules: BeH/ BeH^+, BeC/BeC^+, CH/CH^+, CH_4/CH_4^+, CH_2 /CH_2^+, H_2^+, HeH^+,et isotopologues (H→D, H→T)

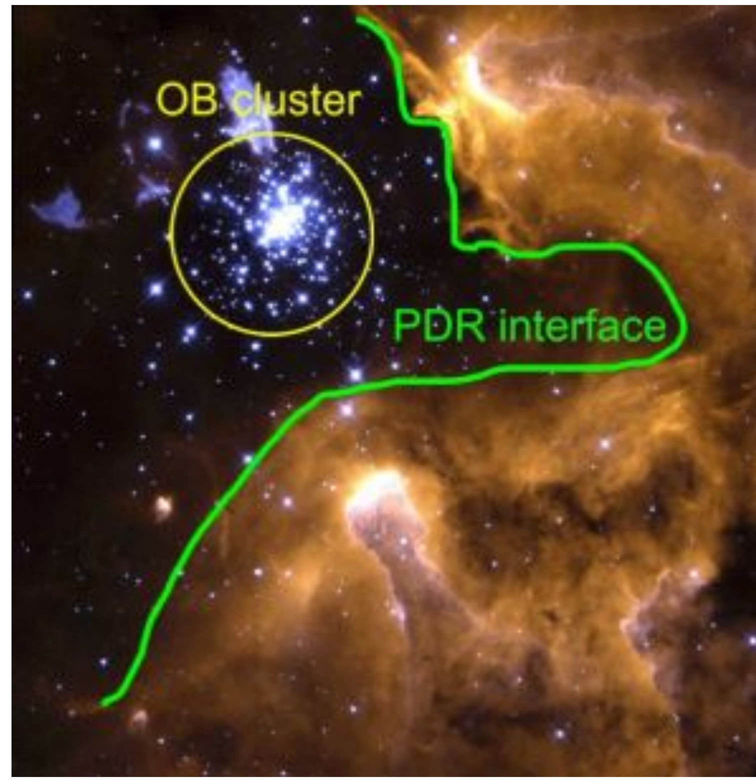


Be: matériau de parois dans les dispositifs de fusion.

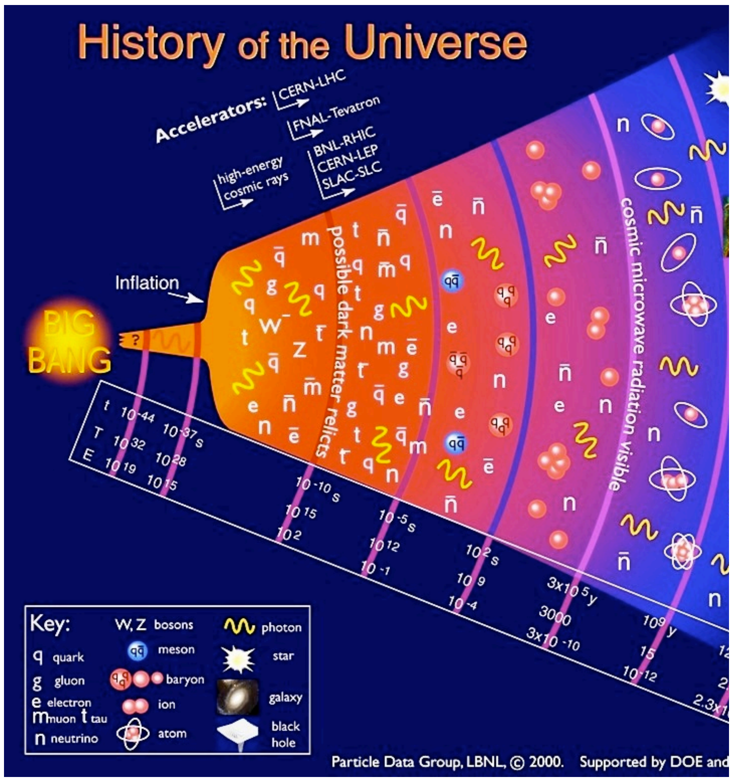


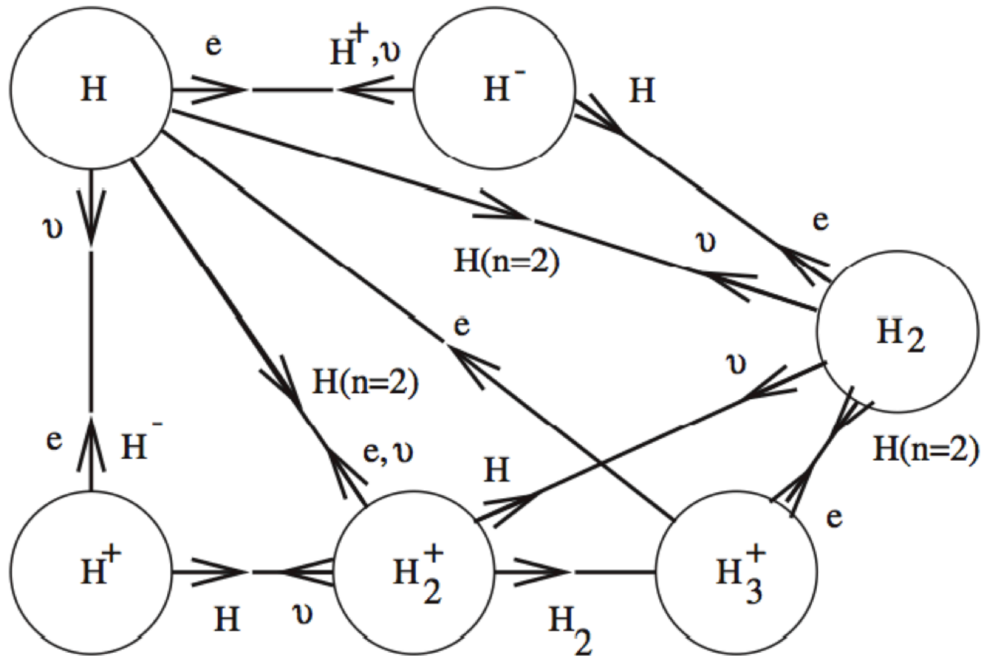
Introduction: H_2 (most abundant molecule)





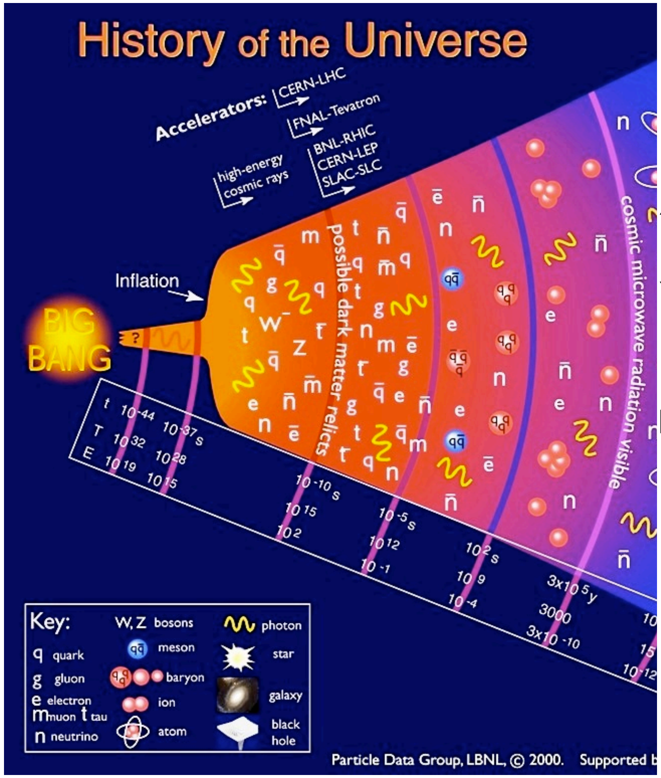
Introduction: H_2 (basic building block of astrochemistry)

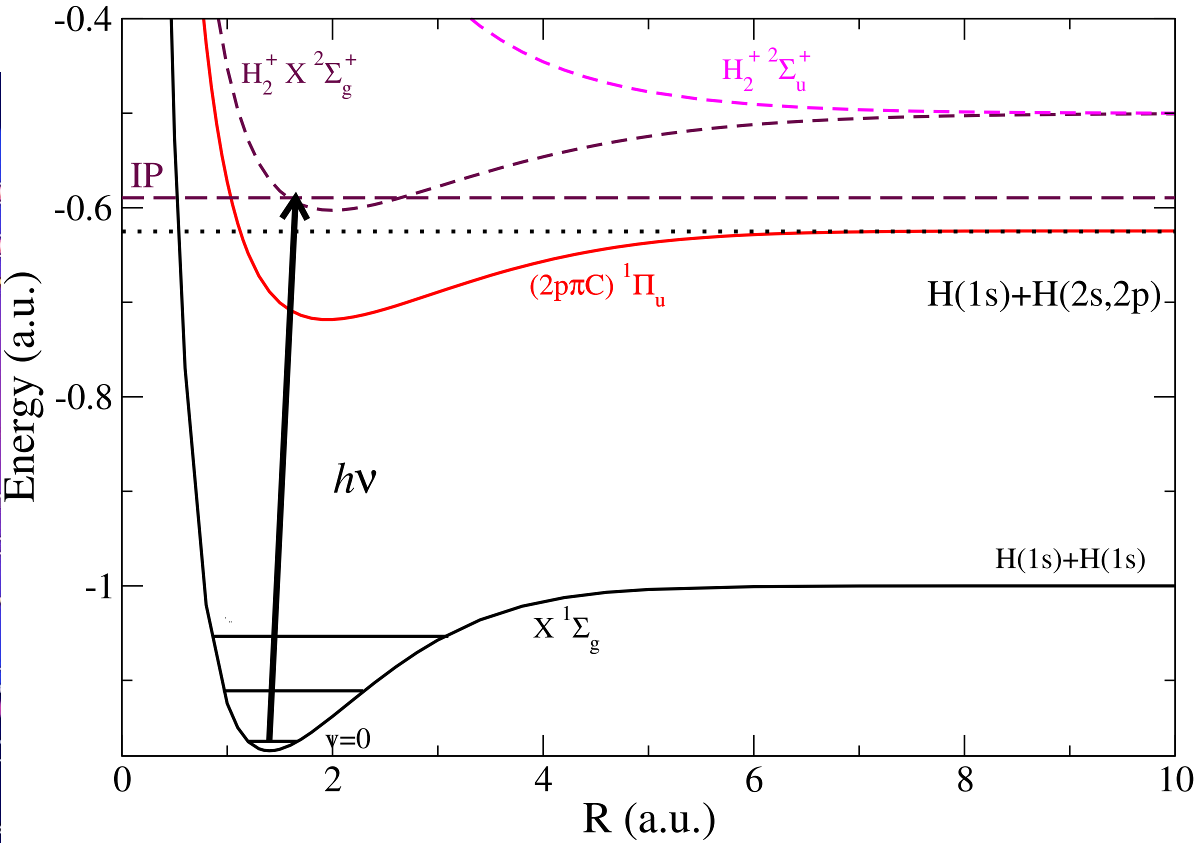




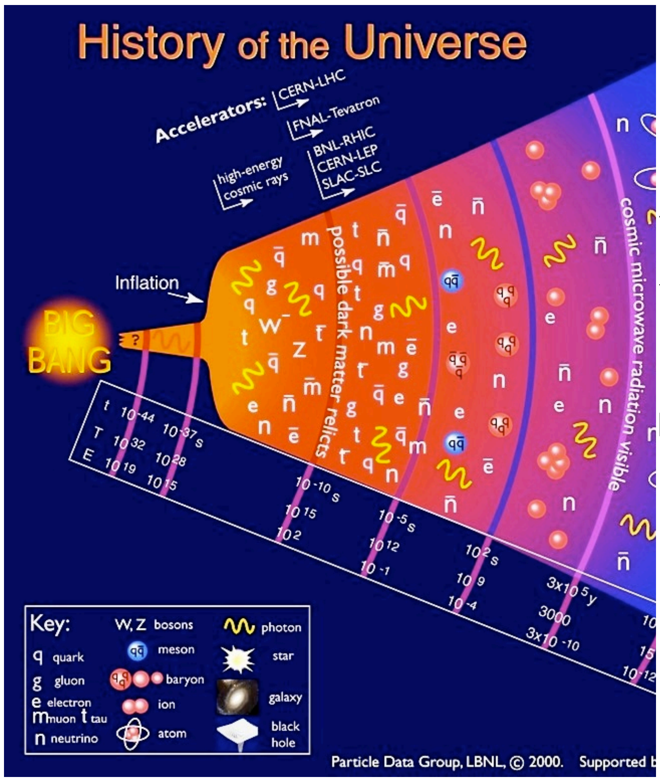
S Lepp, PC Stancil and A Dalgarno, J. Phys. B **35** R1-R24 (2002)

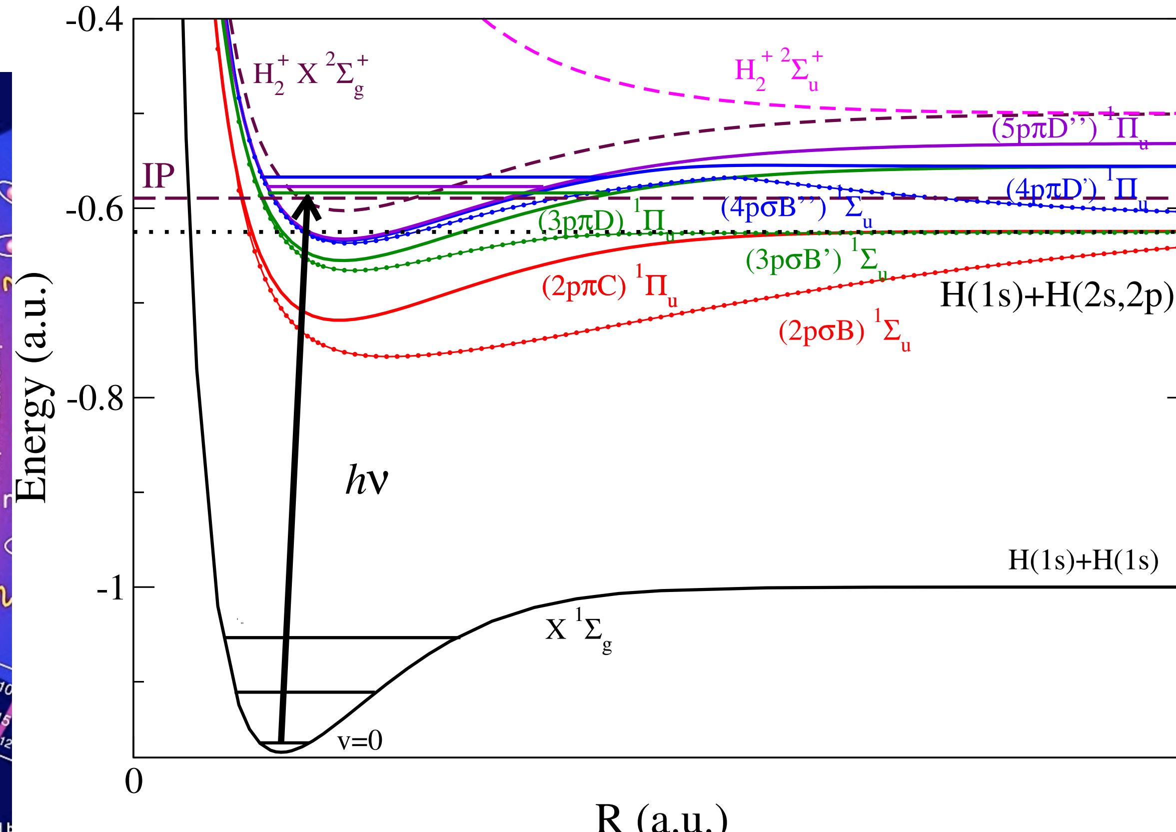
Introduction: H_2 (kinetic modeling: **direct** vs autoionisation)





Introduction: H_2 (kinetic modeling: direct vs **autoionisation**)

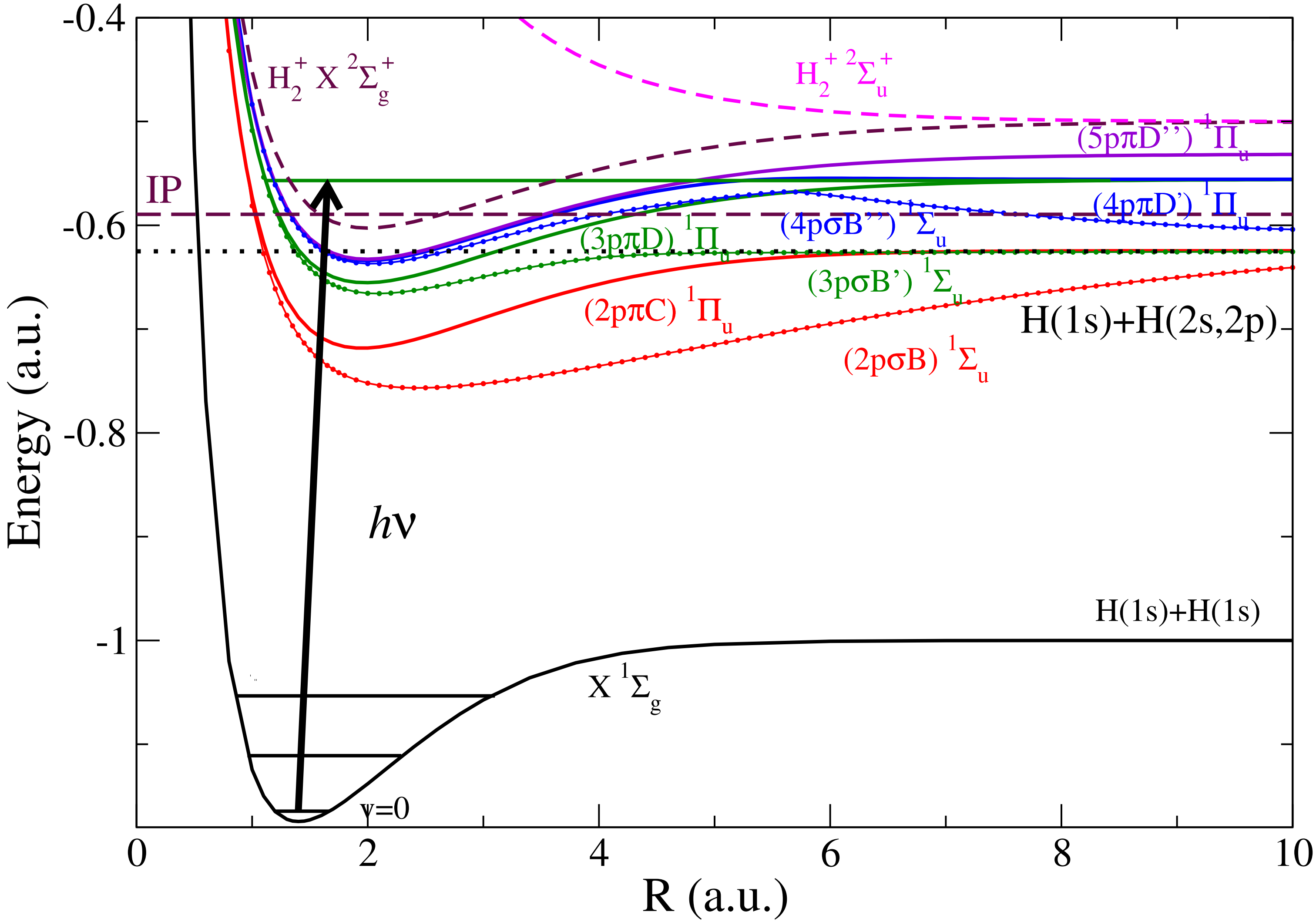




Photofragmentation: Reactive collisions

$$h\nu + \mathrm{AB}(v_i, N_i) \rightarrow \mathrm{AB}^*, \mathrm{AB}^{**} \rightarrow \begin{cases} \mathrm{AB}(v_f, N_f) & PhE \\ \mathrm{A} + \mathrm{B} & PhD \\ \mathrm{AB}^+(v_f^+, N_f^+) + e^- & PhI \end{cases}$$

Photofragmentation: Reactive collisions



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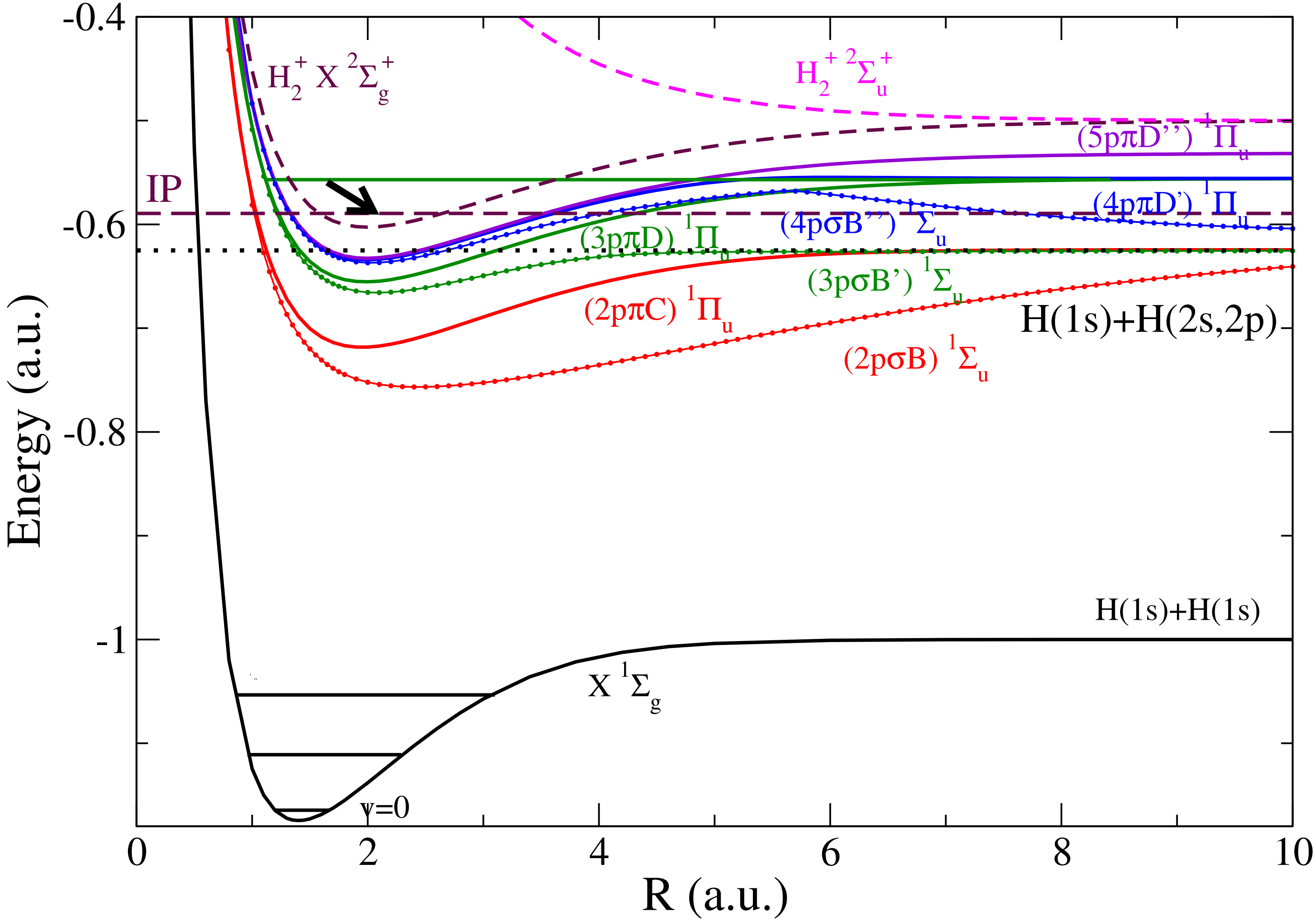

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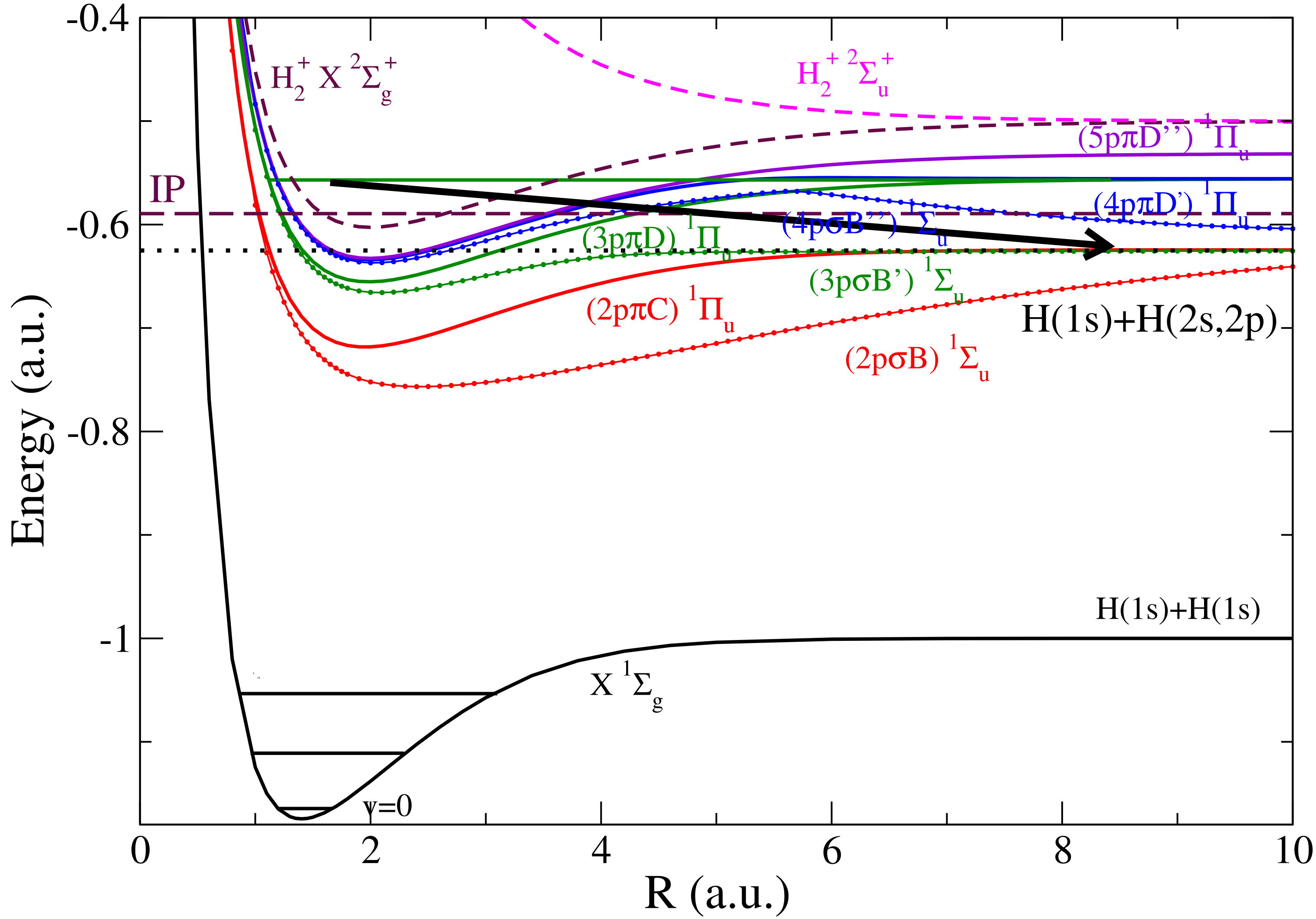
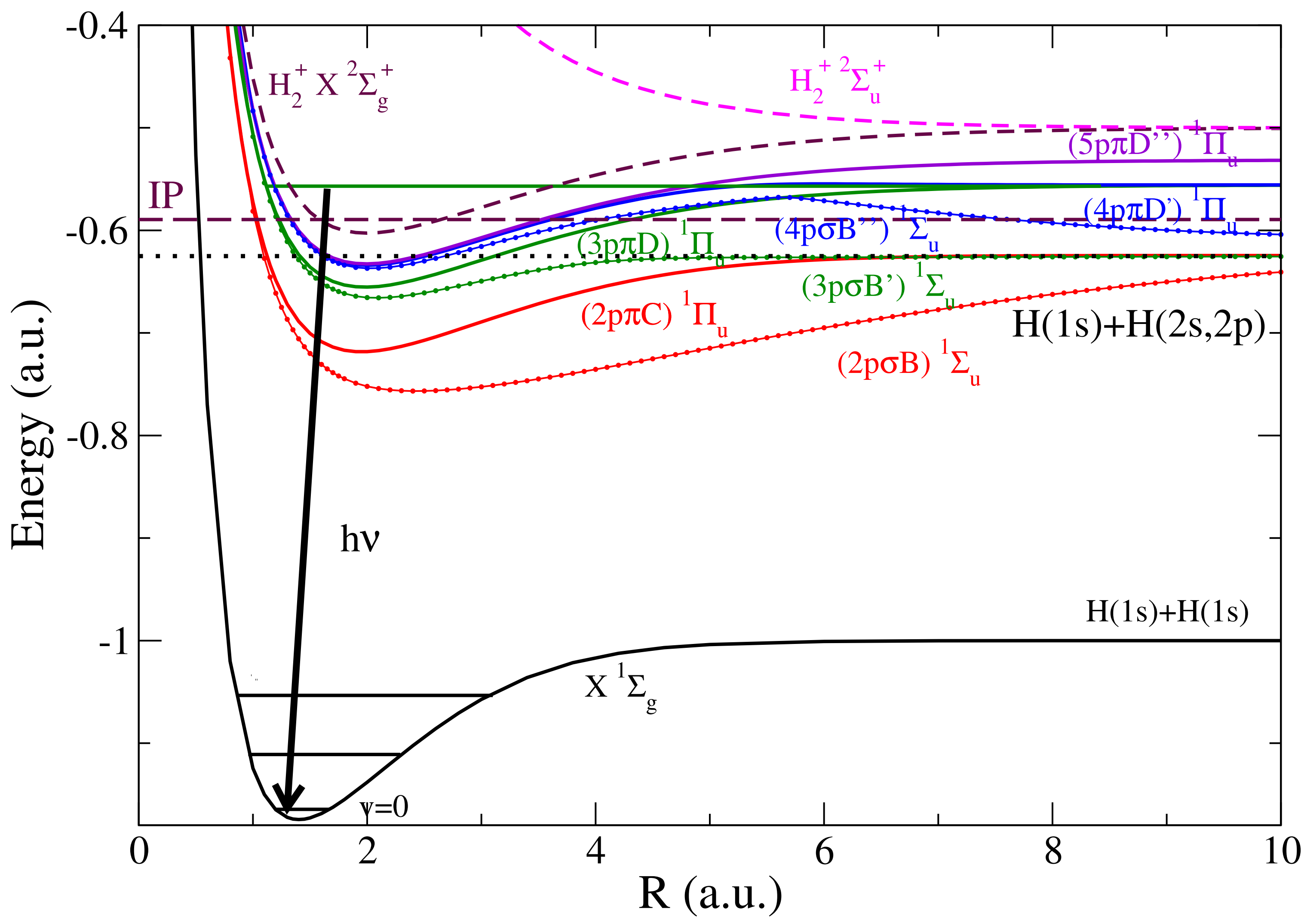


Photofragmentation: Reactive collisions



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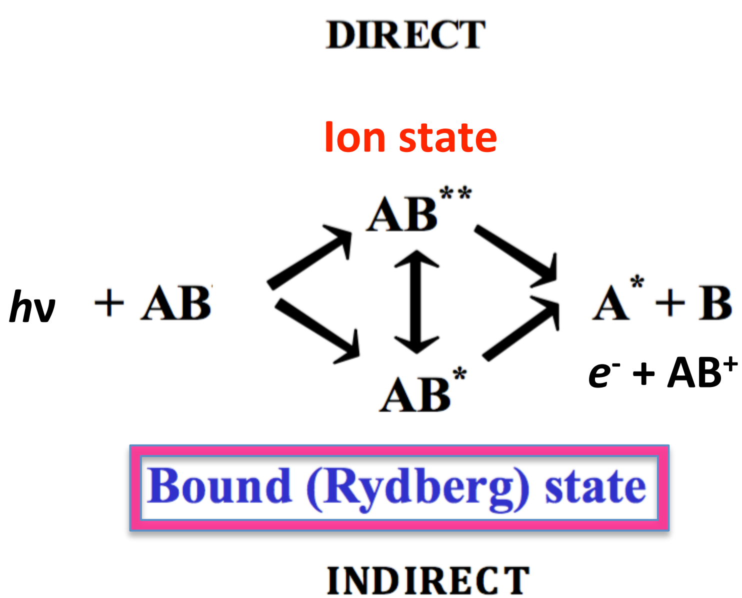
Theoretical approach: MQDT

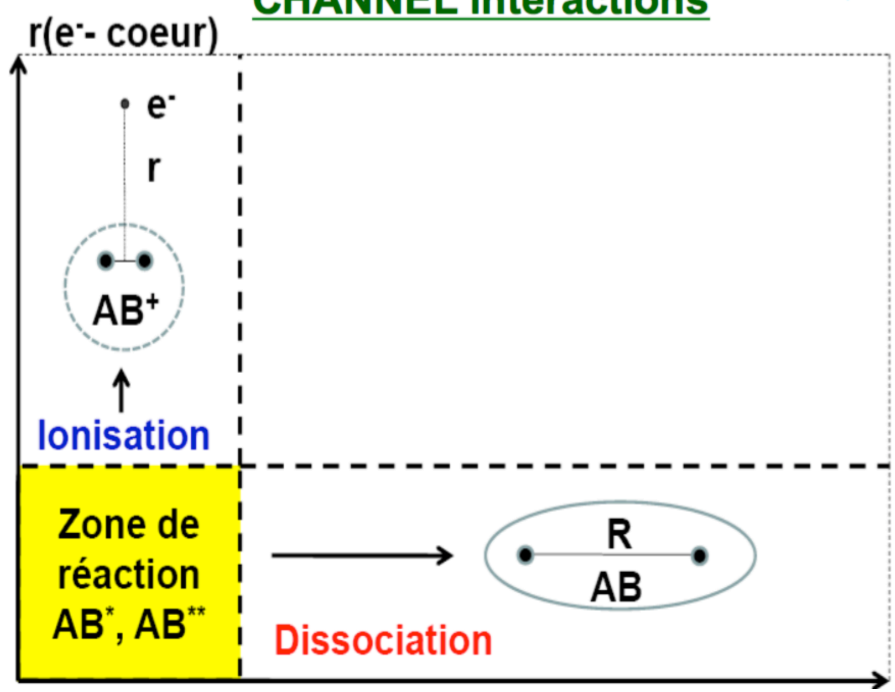
Multichannel Quantum Defect Theory

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

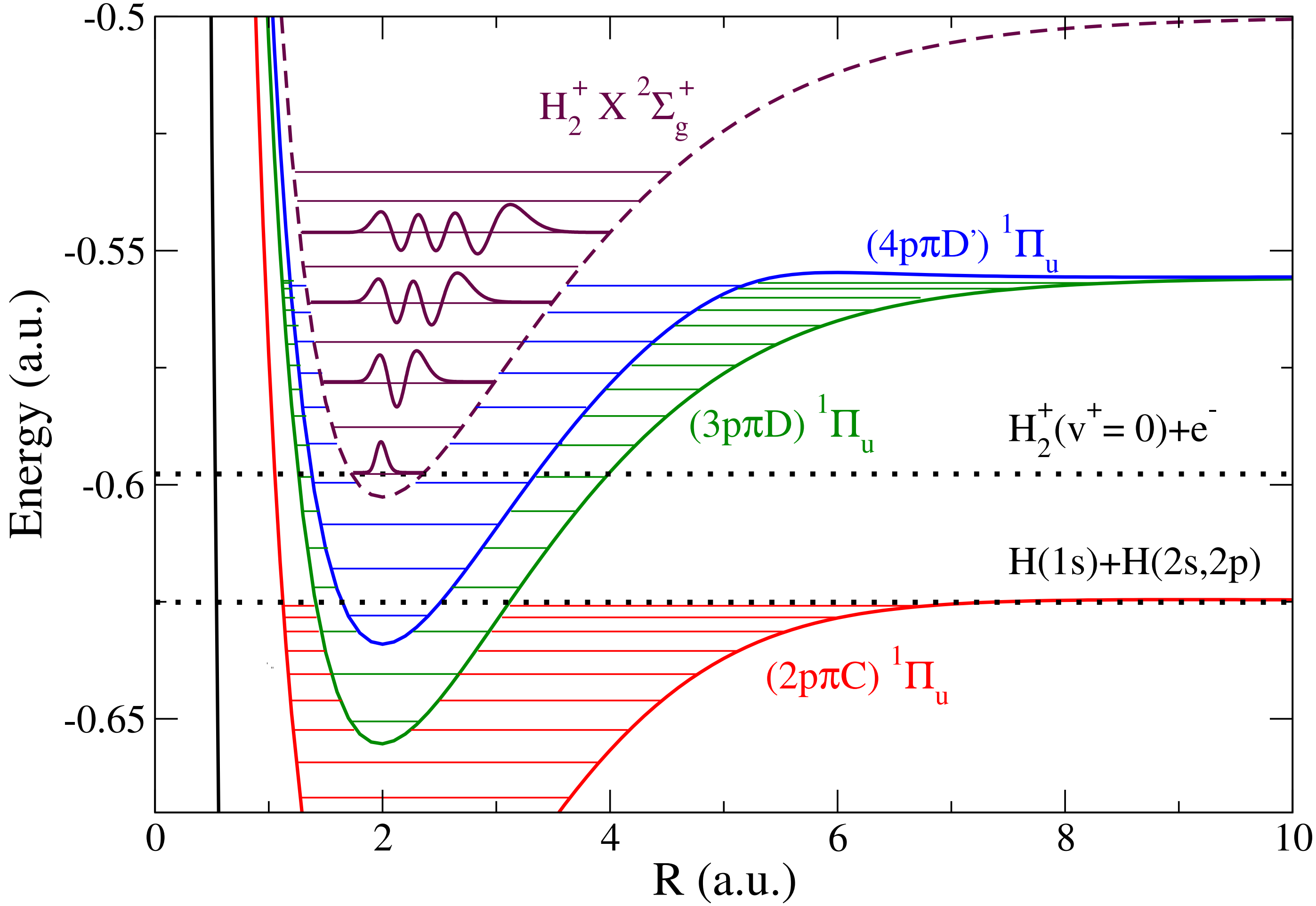
Quantum Interferences



CHANNEL interactions

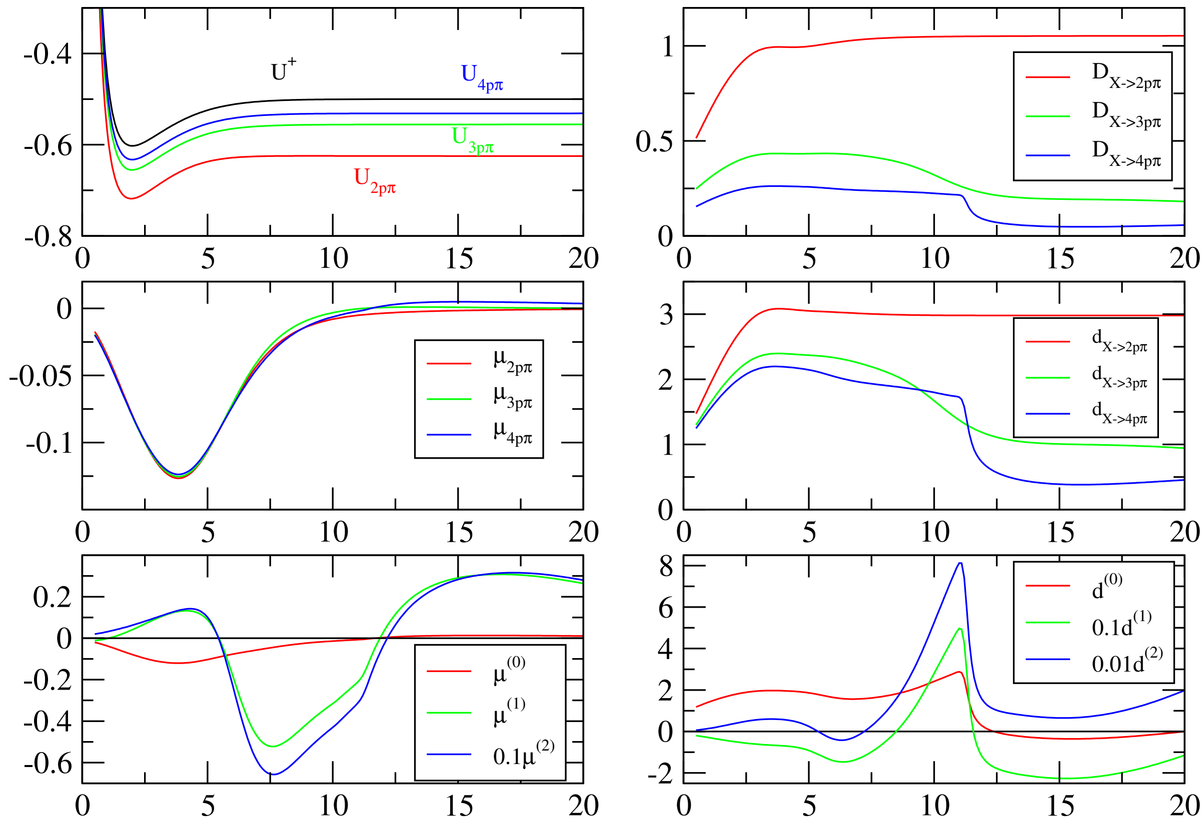


1. # MQDT: Molecular data



Wolniewicz *et al*

2. MQDT: Molecular data



- Rydberg equation

$$U_{np\pi}(R) = U^{+}(R) - \frac{1}{2\left[n - \mu_{np\pi}(R)\right]^{2}},$$

- Energy dependent quantum defect

$$\mu(\epsilon, R) = \mu^{(0)}(R) + \epsilon\mu^{(1)}(R) + \frac{1}{2}\epsilon^{2}\mu^{(2)}(R)] + \frac{m}{M}\mu^{\mathrm{spec}}(R).$$

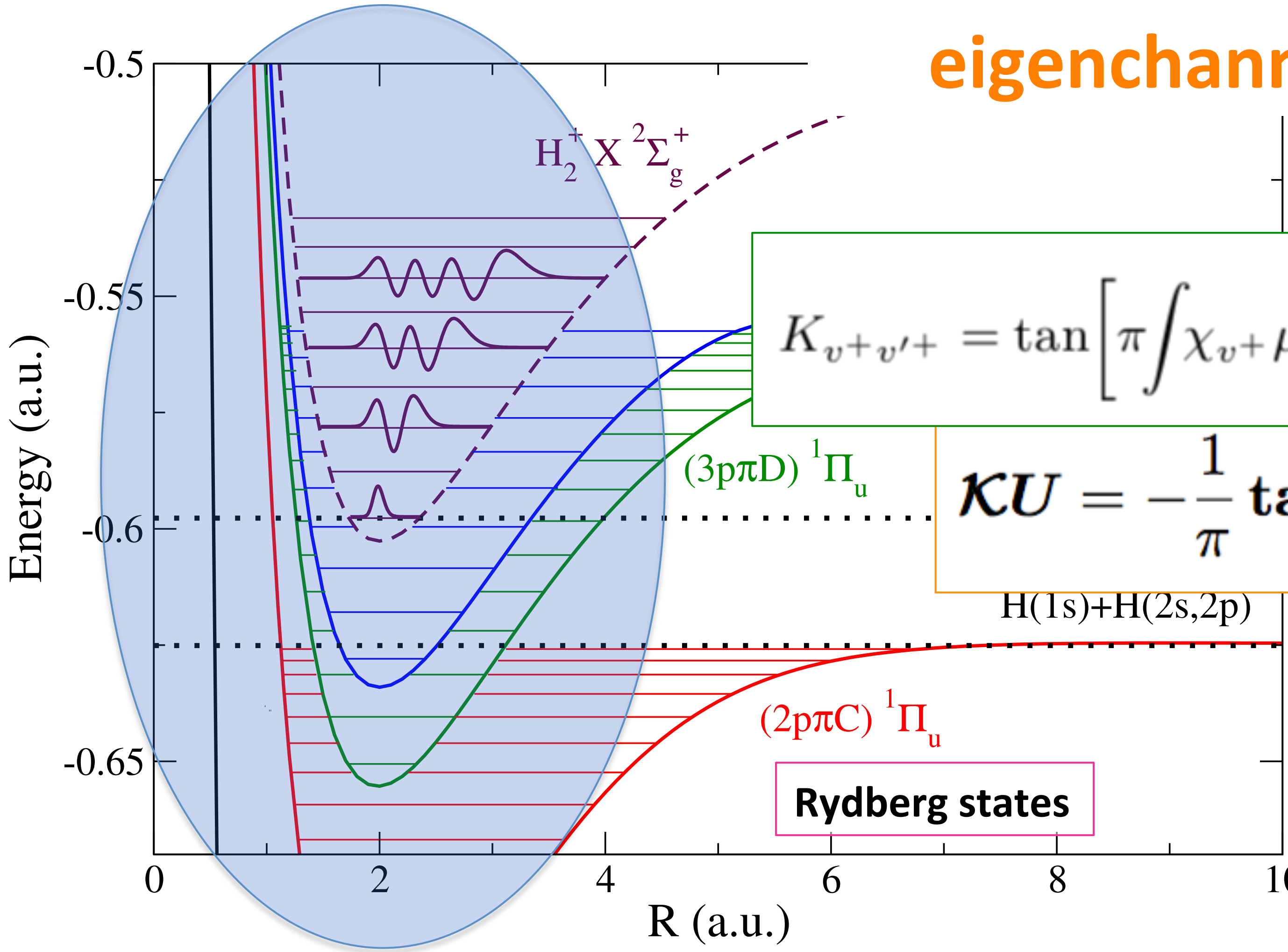
- Energy dependent dipole moment

$$d(\epsilon, R) = d^{(0)}(R) + \epsilon d^{(1)}(R) + \frac{1}{2}\epsilon^{2}d^{(2)}(R),$$

3. MQDT: The formalism

Reaction zone

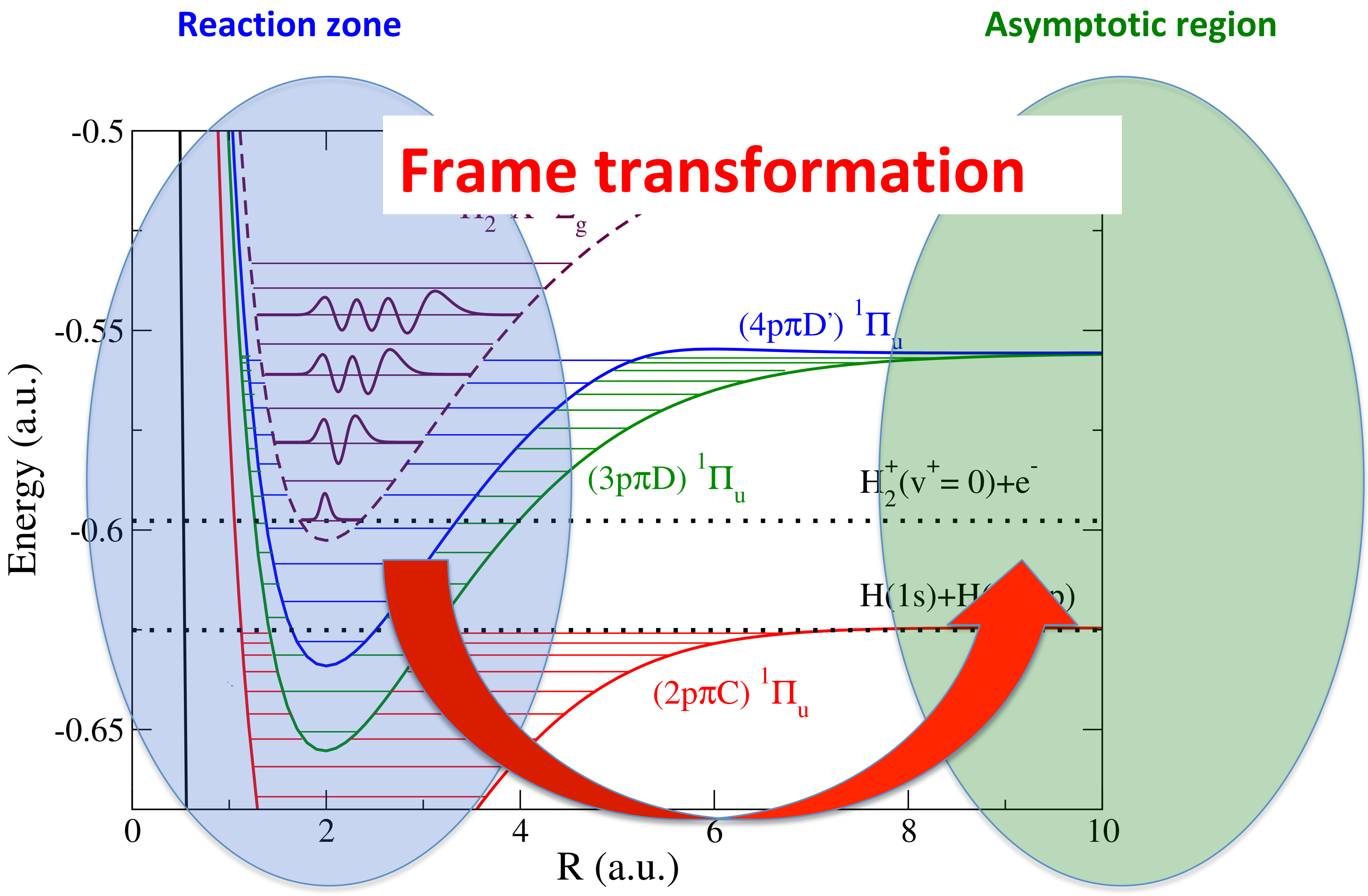
K-matrix + eigenchannels



$$K_{v^+v'^+} = \tan\left[\pi \int \chi_{v^+} \mu(R) \chi_{v'^+} dR\right]$$

$$\boldsymbol{\mathcal{K}U} = -\frac{1}{\pi}\tan(\boldsymbol{\eta})\boldsymbol{U},$$

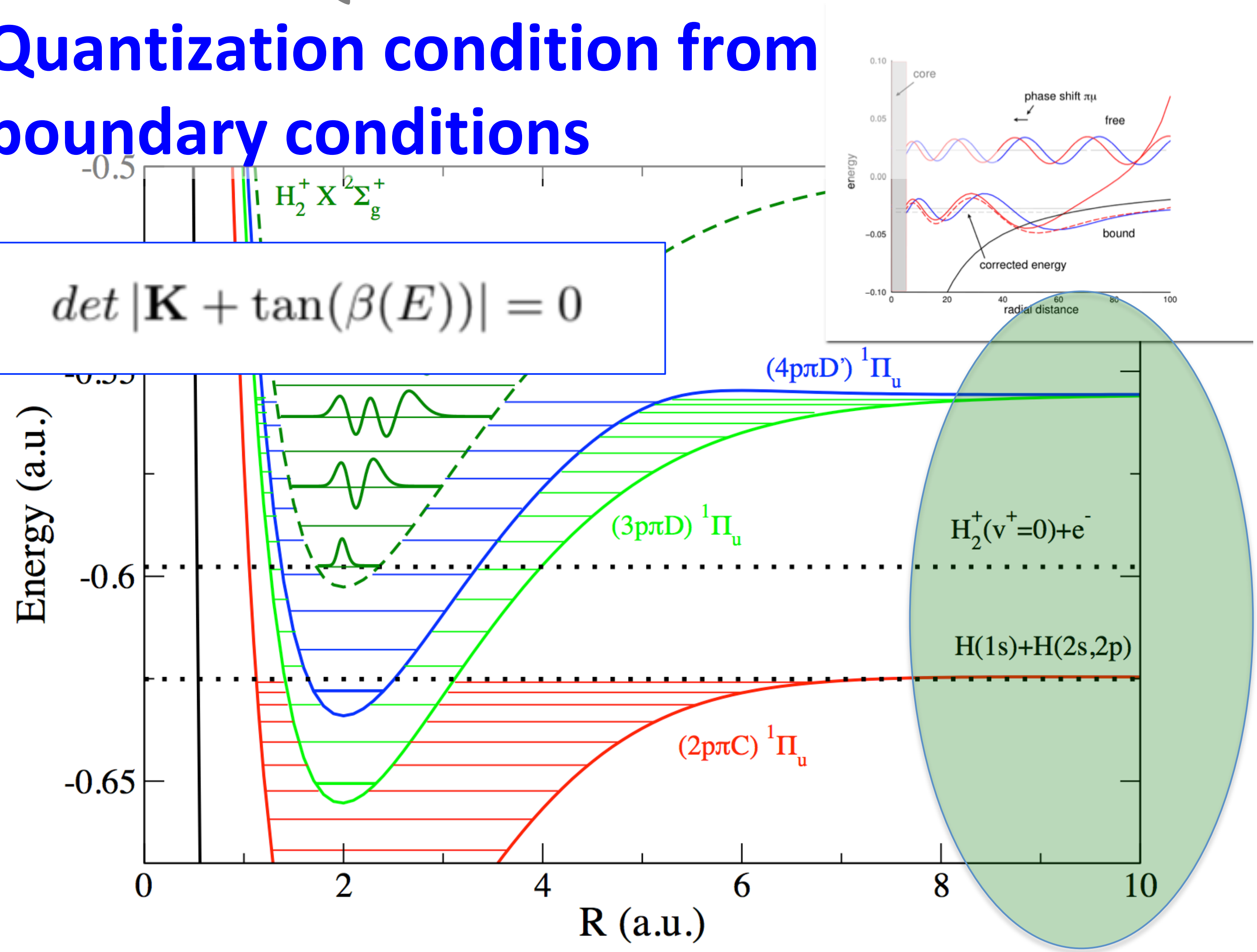
4. MQDT: The formalism



5. MQDT: The formalism

Quantization condition from boundary conditions

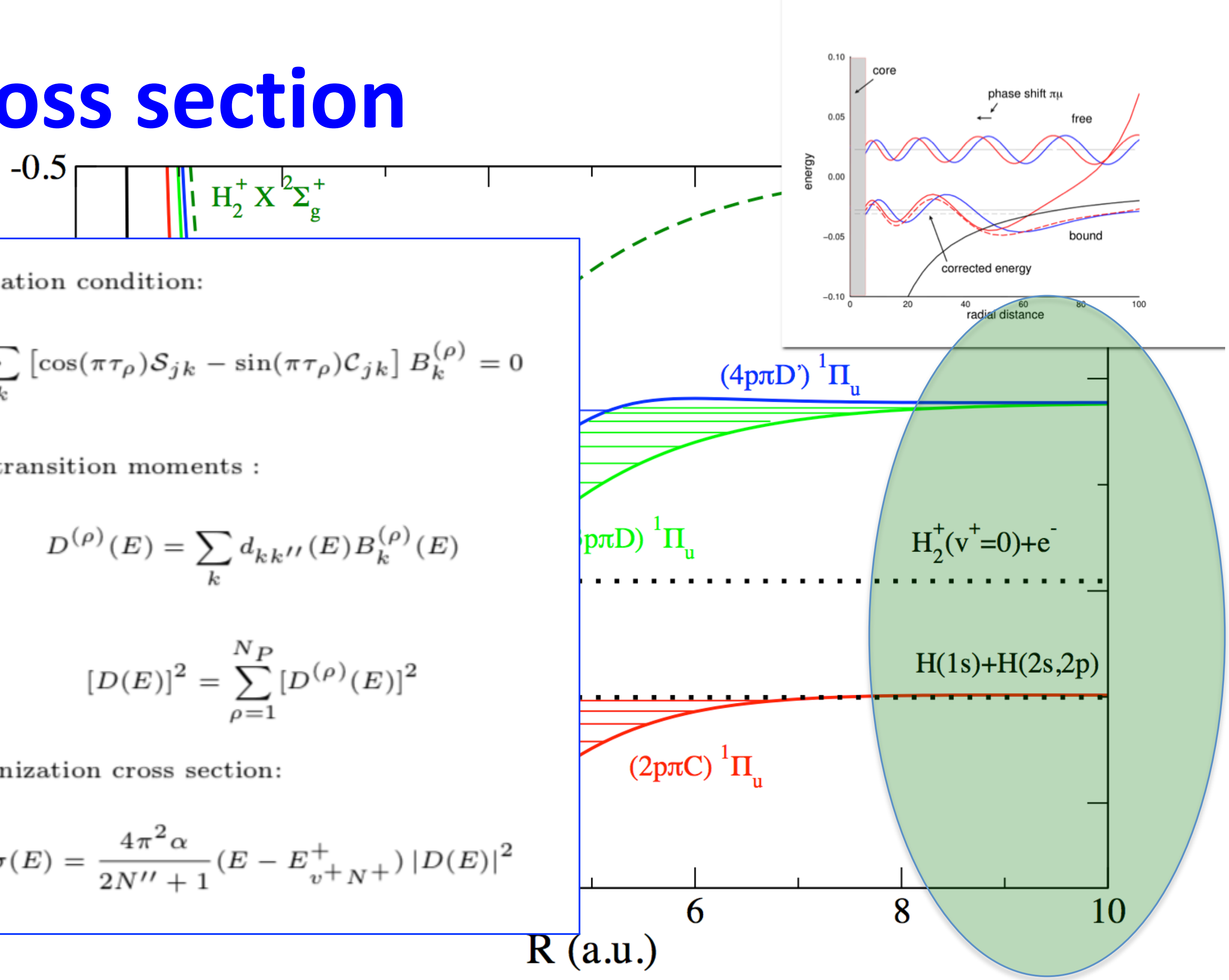
$$det\left|\mathbf{K}+\tan(\beta(E))\right|=0$$



Asymptotic region

6. MQDT: The formalism

Cross section



- Quantization condition:

$$\sum_k \left[\cos(\pi\tau_\rho)\mathcal{S}_{jk} - \sin(\pi\tau_\rho)\mathcal{C}_{jk}\right] B_k^{(\rho)} = 0$$

- Dipole transition moments :

$$D^{(\rho)}(E) = \sum_k d_{kk''}(E) B_k^{(\rho)}(E)$$

$$[D(E)]^2 = \sum_{\rho=1}^{N_P} [D^{(\rho)}(E)]^2$$

- Photoionization cross section:

$$\sigma(E) = \frac{4\pi^2\alpha}{2N''+1}(E - E^+_{v^+N^+})\,|D(E)|^2$$

Asymptotic region

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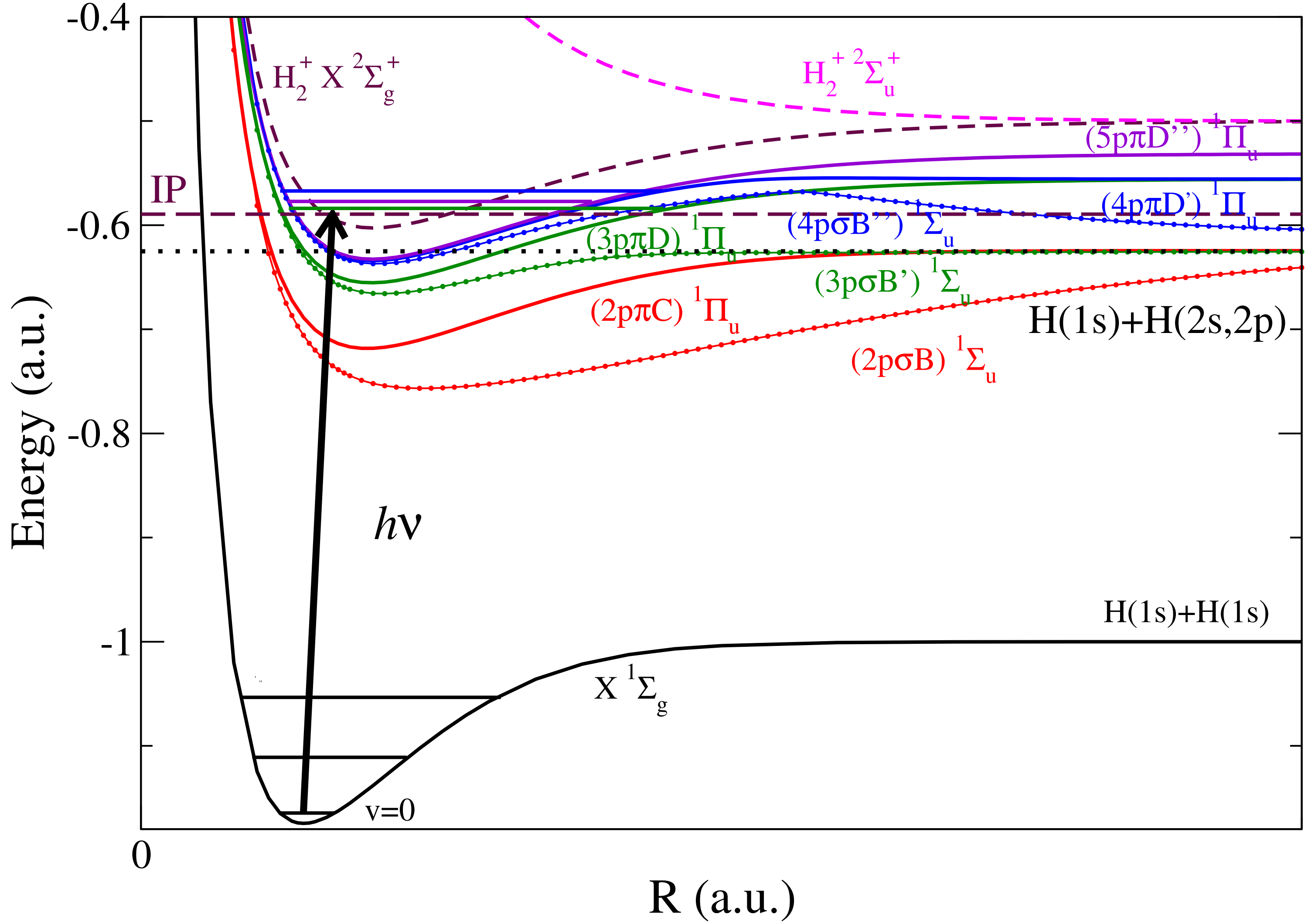
Results: H_2 – Photoionization



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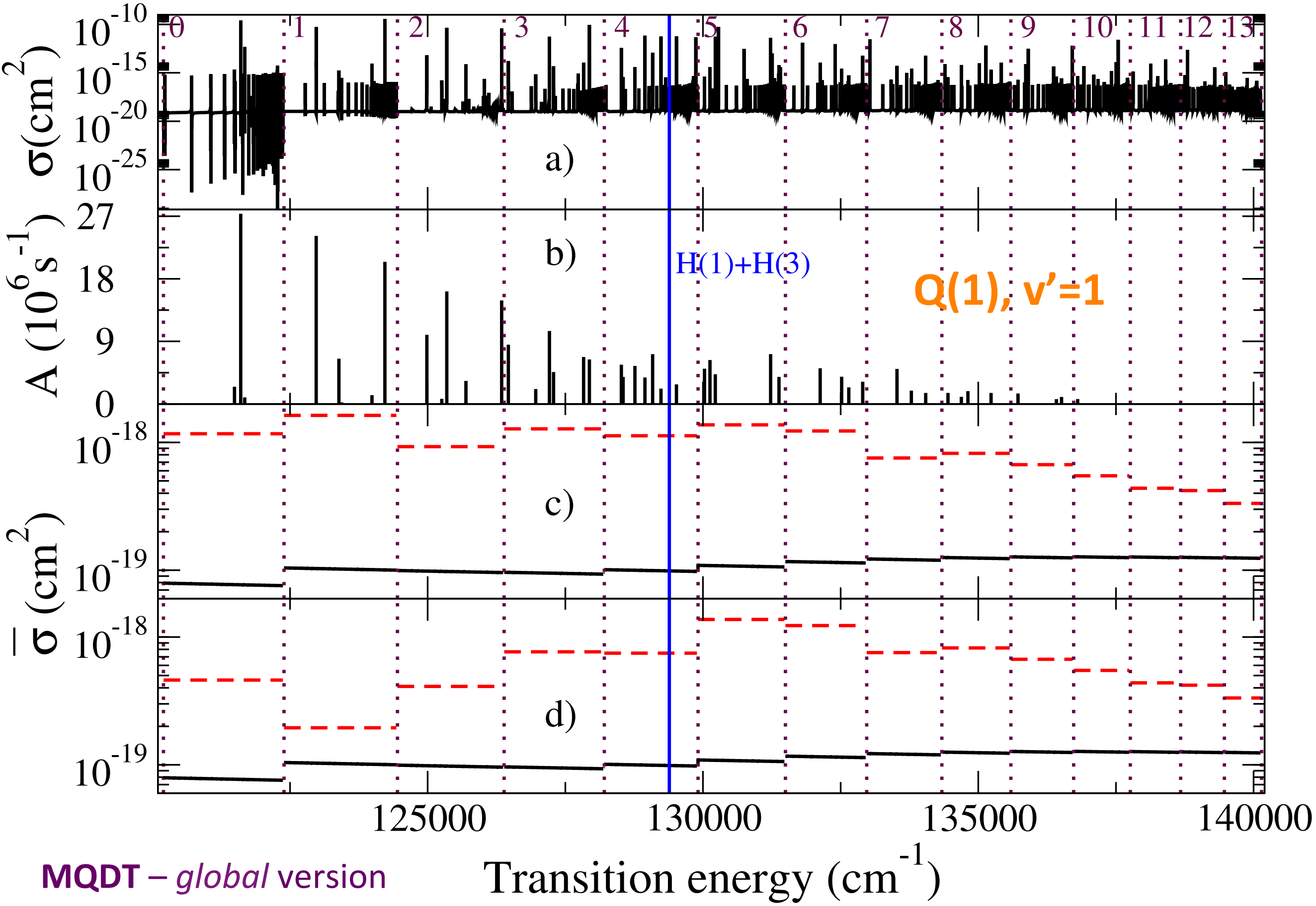

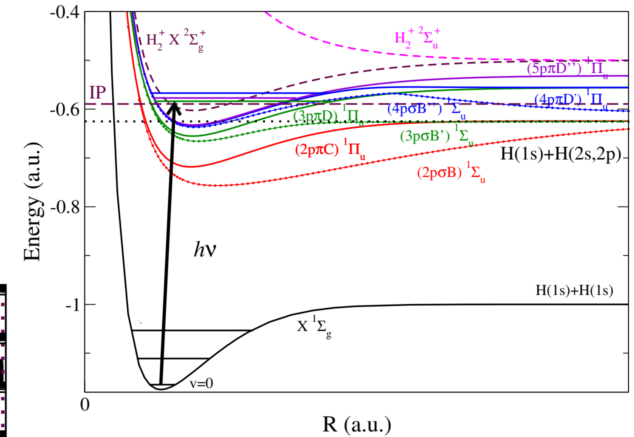

MQDT – *global* version

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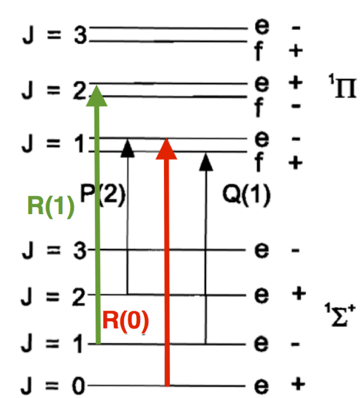

Resonances in photoionization: Cross sections for vibrationally excited H_2

J. Zs. Mezei,[1,2,*] I. F. Schneider,[1,†] E. Roueff,[3] and Ch. Jungen[2,‡]

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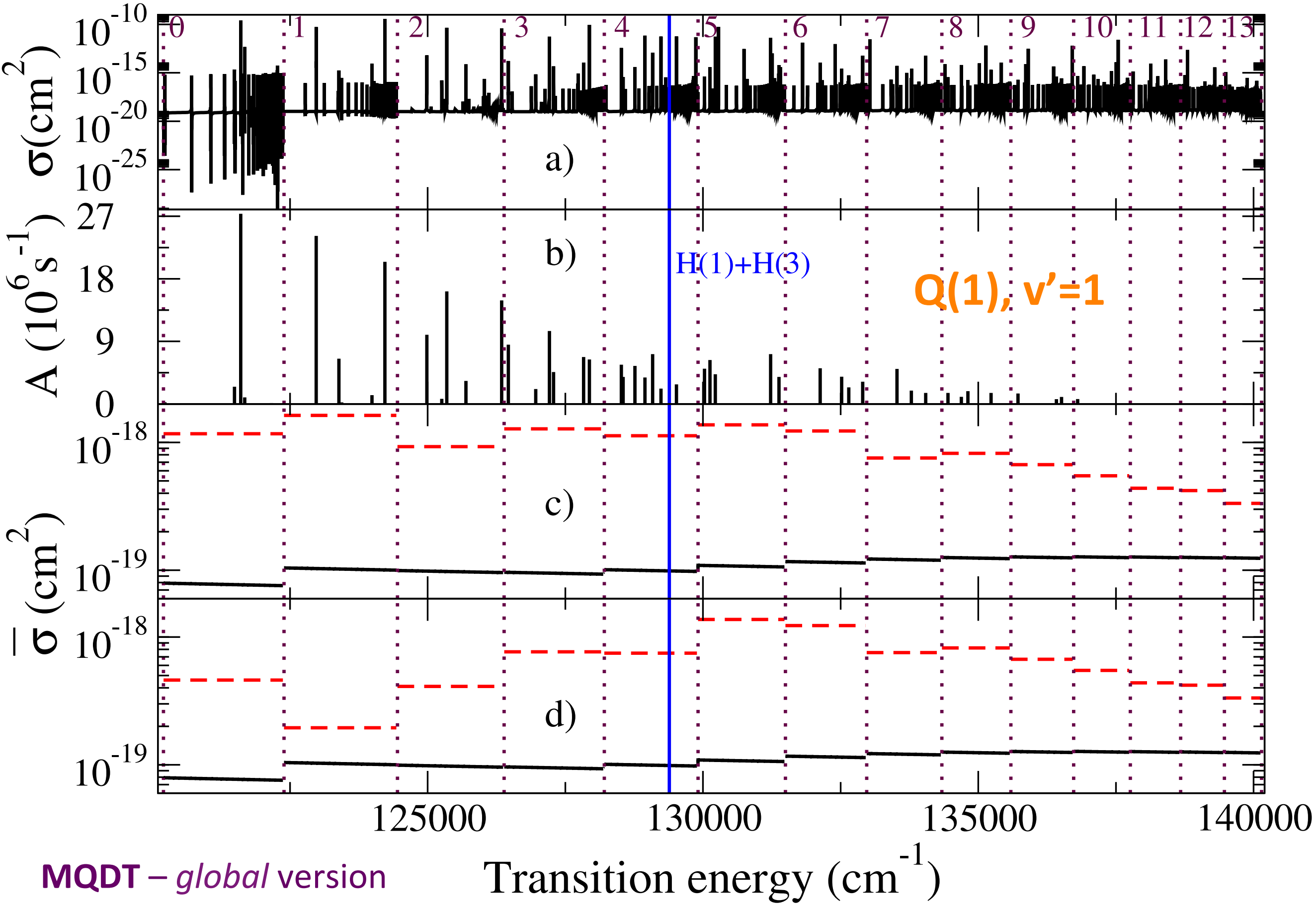
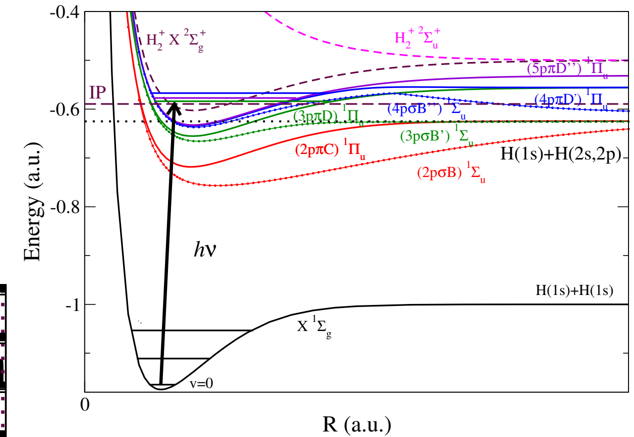
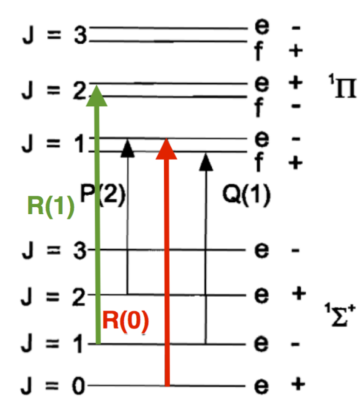




a) Photoionization profile



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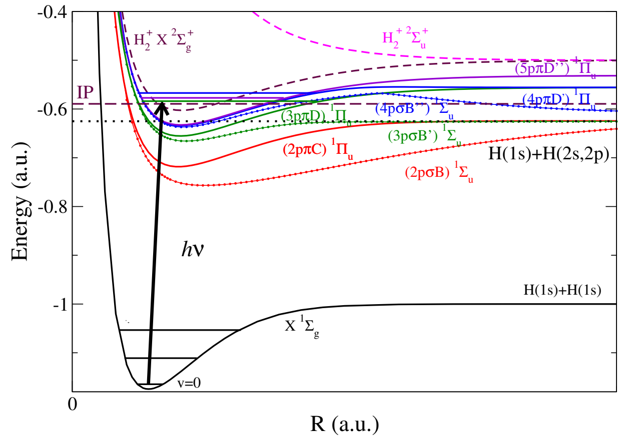

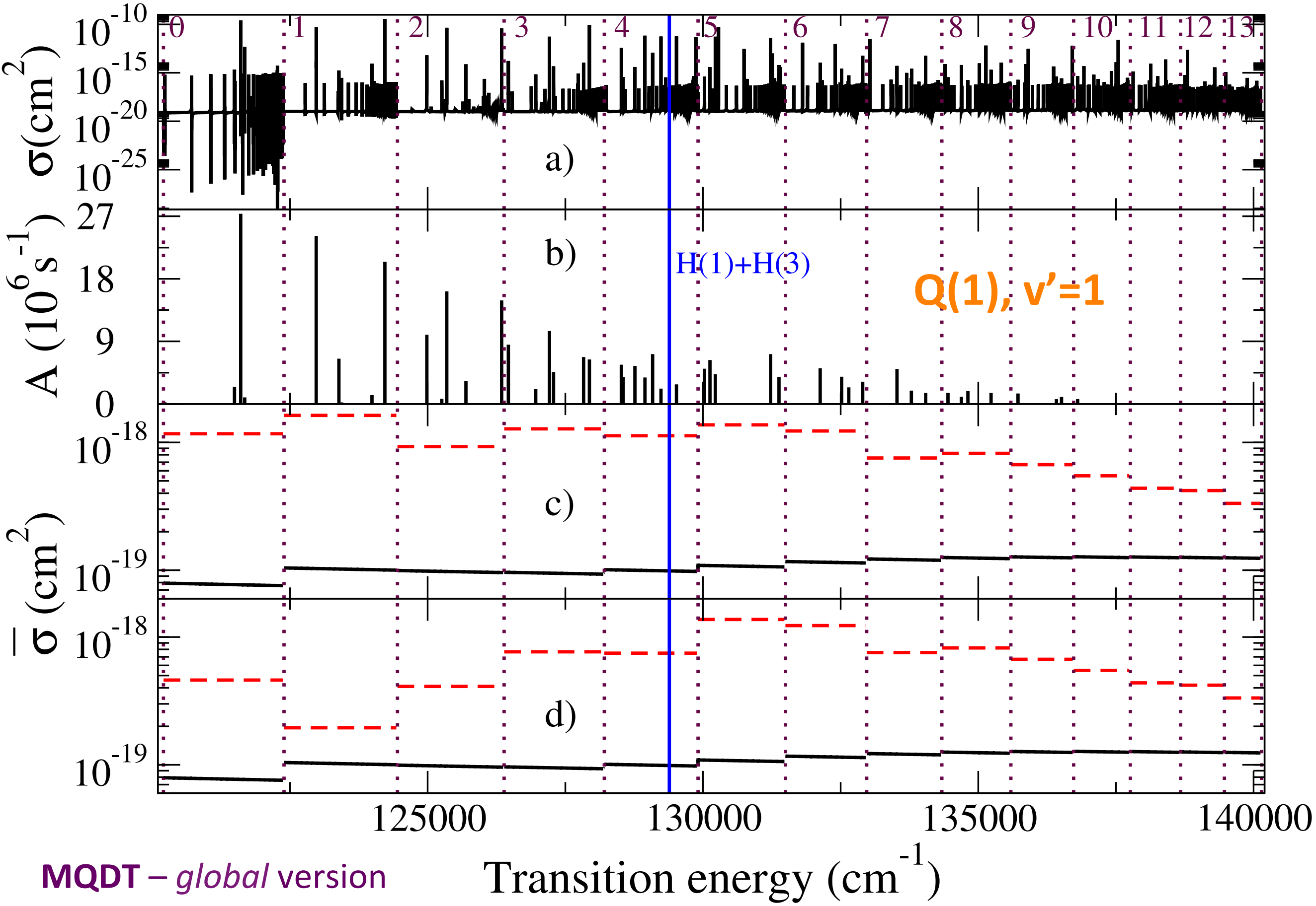

a) Photoionization profile

b) Most intense lines

MQDT – *global* version

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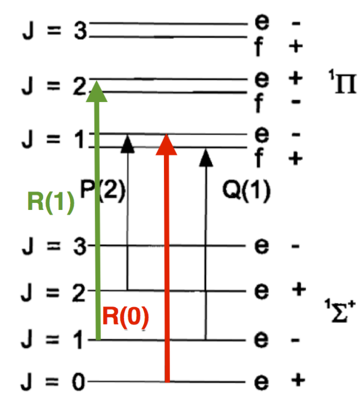

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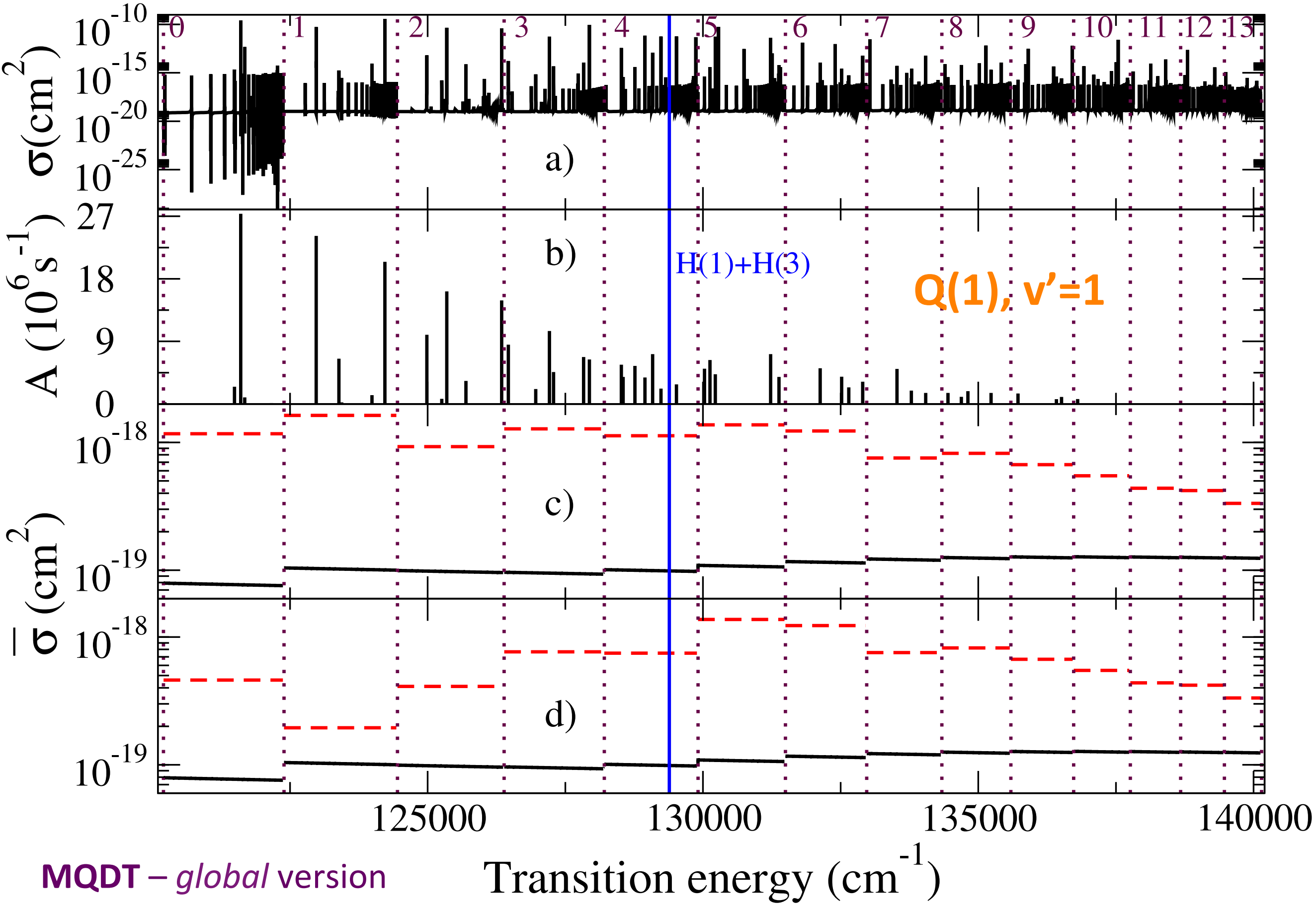
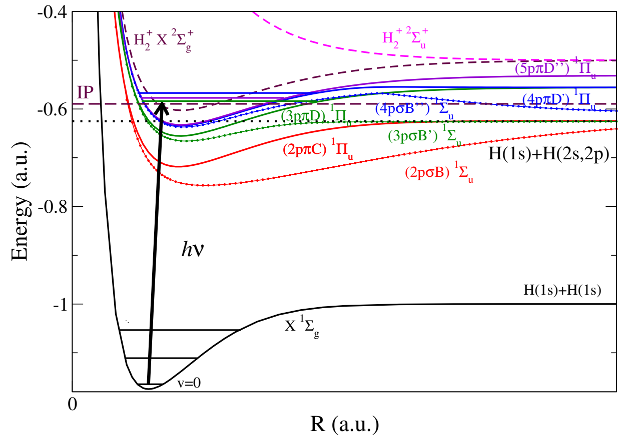
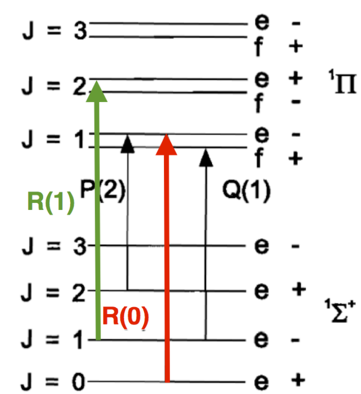




a) Photoionization profile

b) Most intense lines

c,d) Average photoionization cross section



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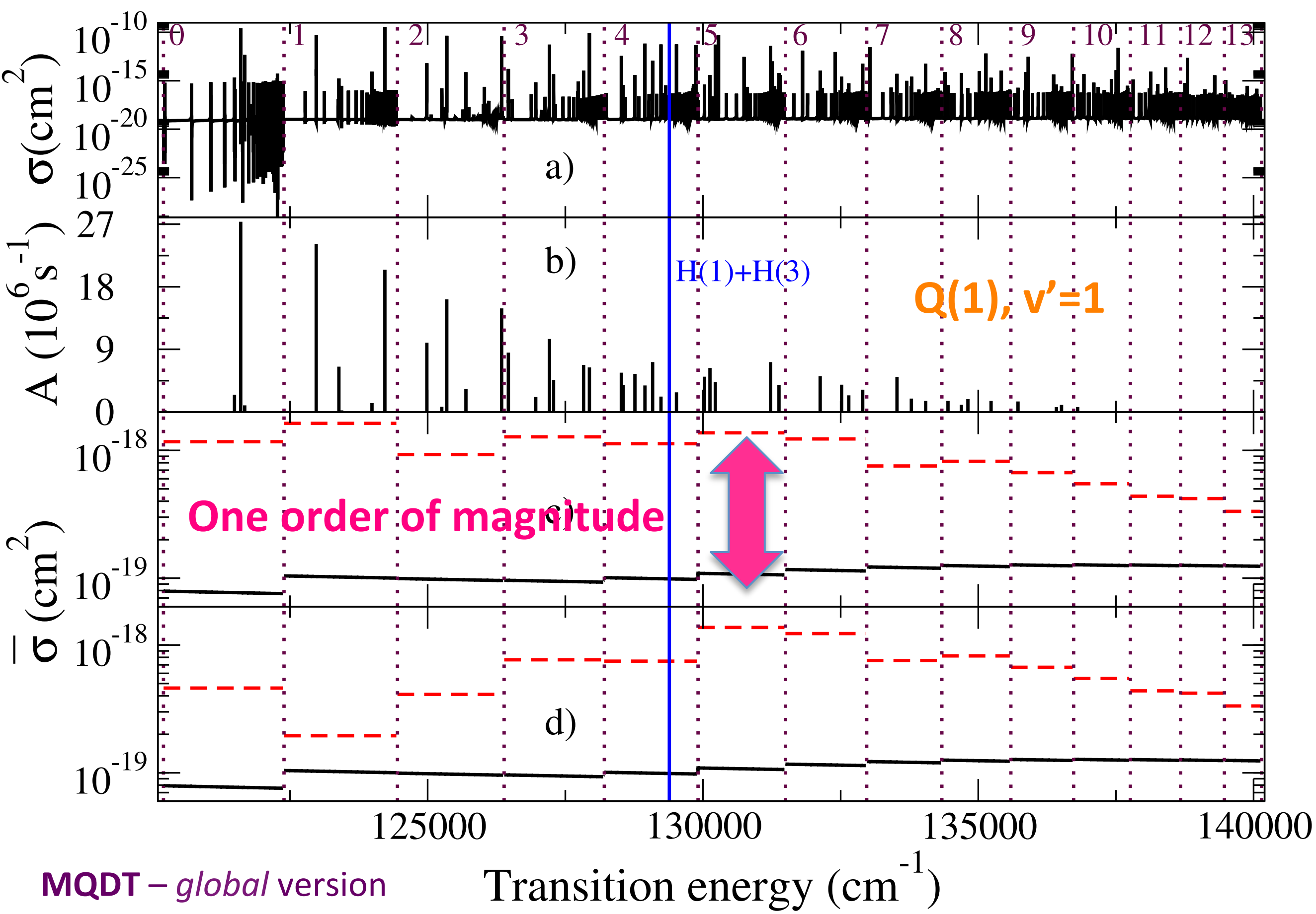

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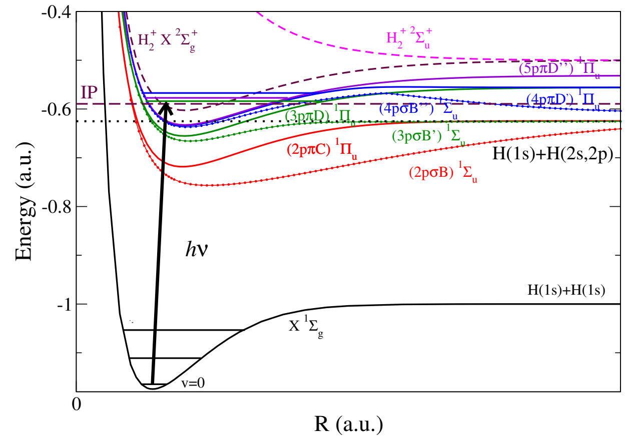
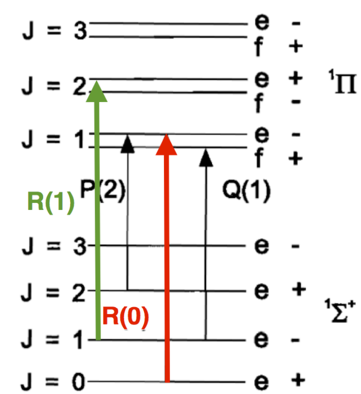


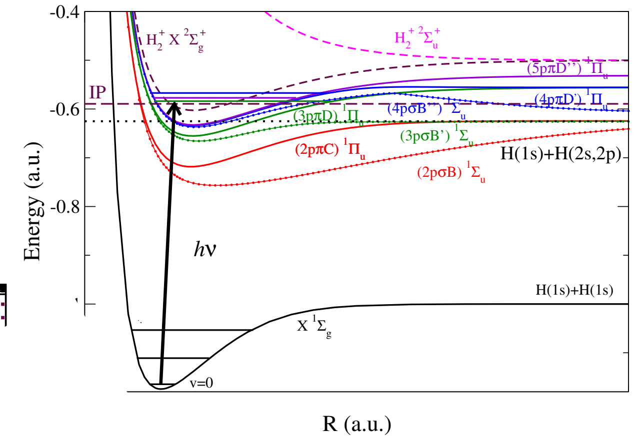
a) Photoionization profile

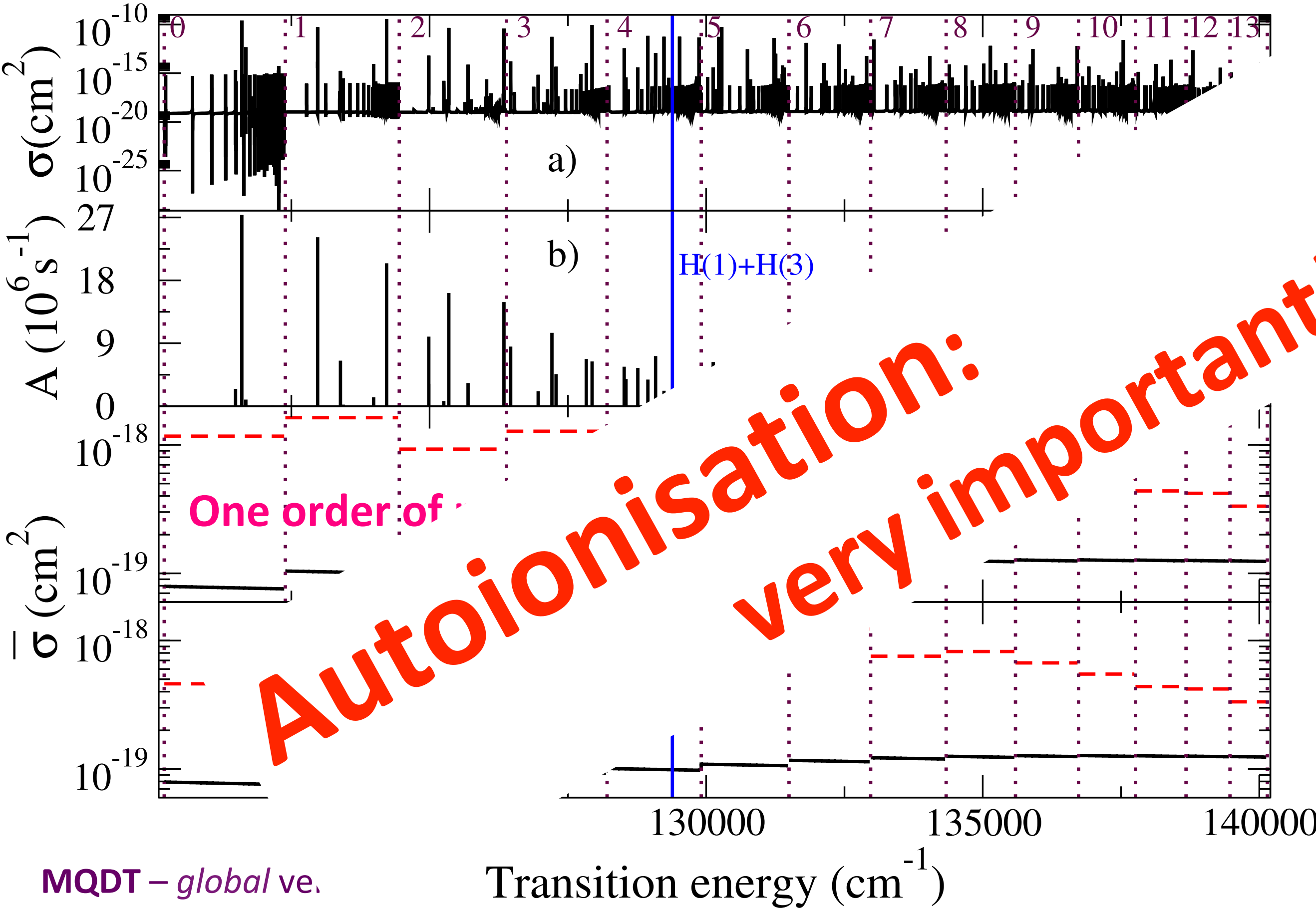
b) Most intense lines

c,d) Average photoionization cross section



Results: H$_2$ - Photoionisation





Autoionisation: very important!!!

…otoionization profile

…, Most intense lines

c,d) Average photoionization cross section

MQDT – *global* ve…

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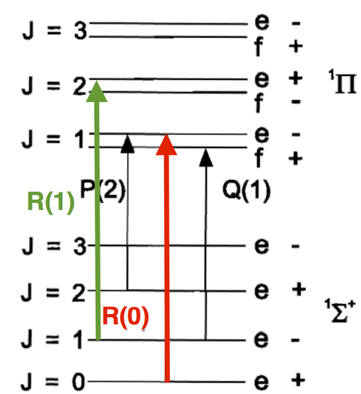
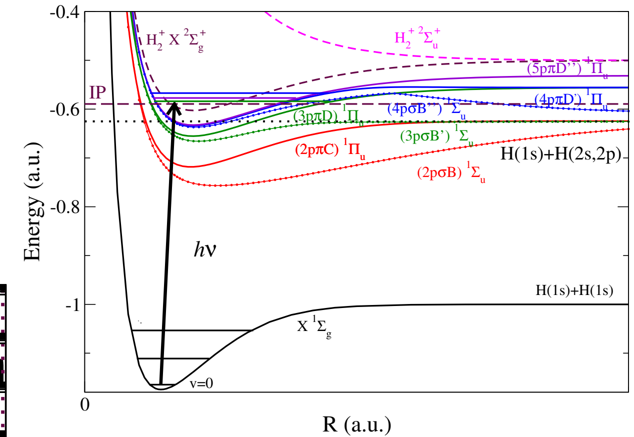
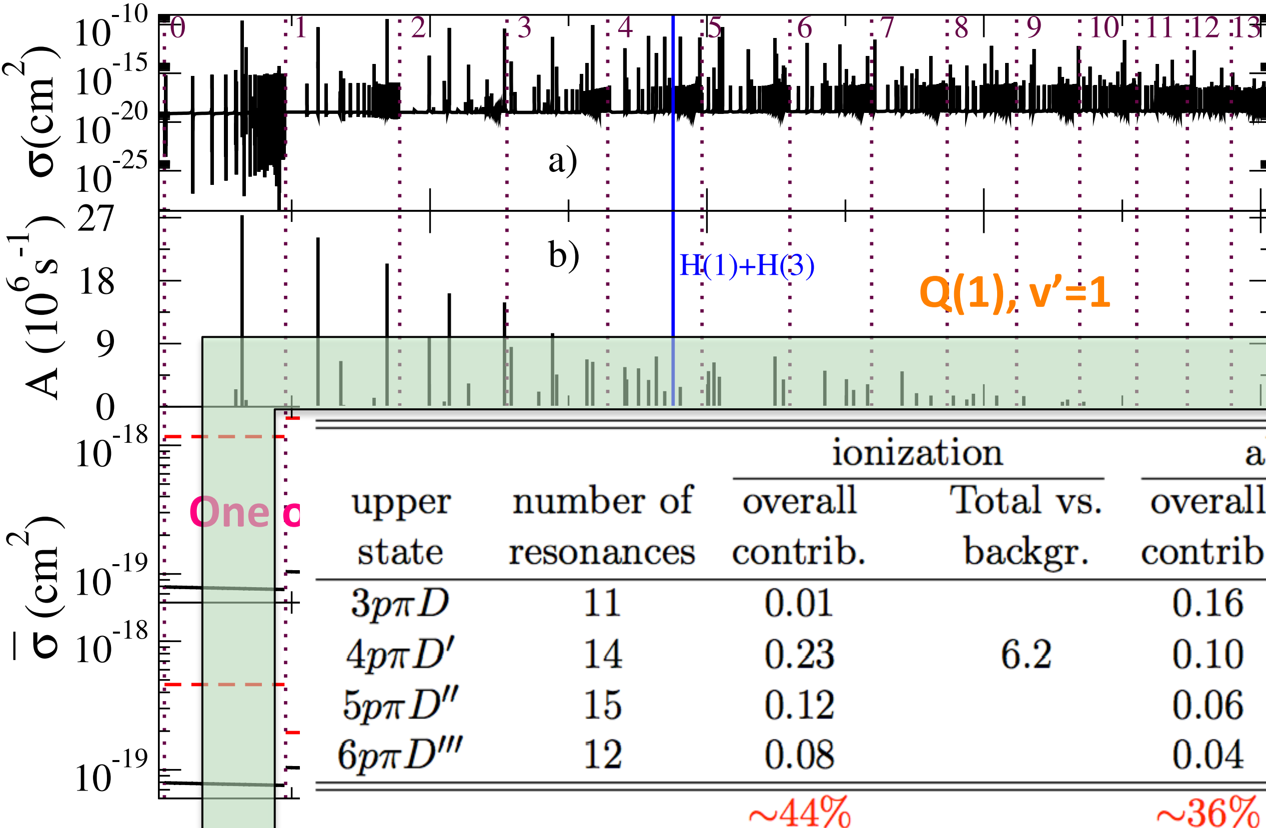


Results: H_2 - Photoionisation



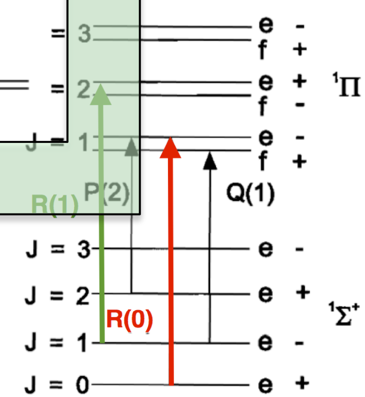


One c

upper state	number of resonances	ionization overall contrib.	ionization Total vs. backgr.	absorption overall contrib.	absorption Total vs. backgr.
$3p\pi D$	11	0.01		0.16	
$4p\pi D'$	14	0.23	6.2	0.10	8.1
$5p\pi D''$	15	0.12		0.06	
$6p\pi D'''$	12	0.08		0.04	
		~44%		~36%	

MQDT – *global* version

Transition energy (cm^{-1})

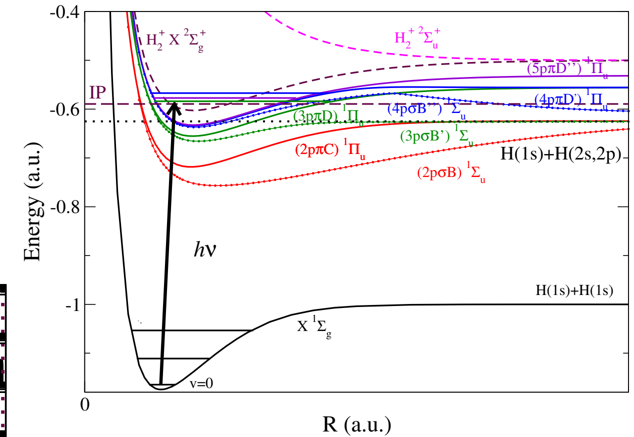


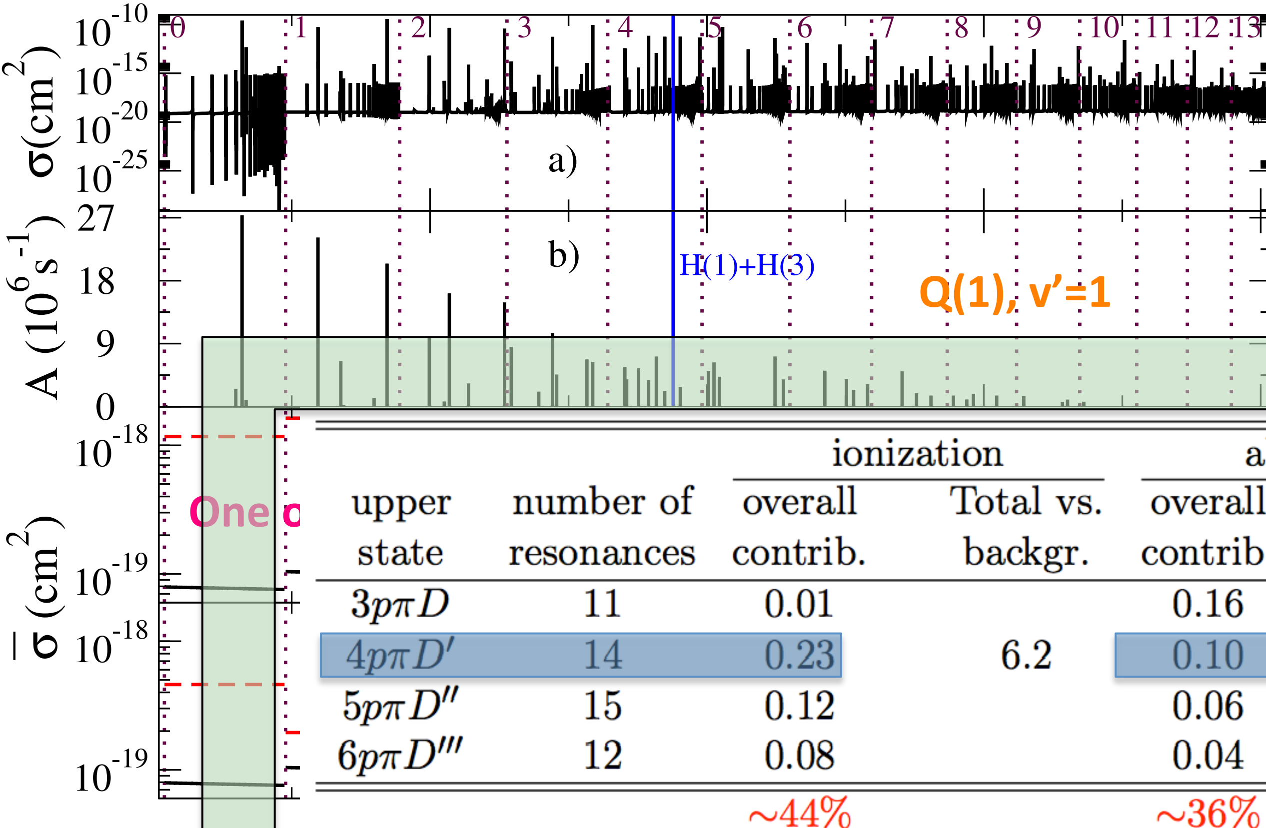
PHYSICAL REVIEW A **85**, 043411 (2012)

Resonances in photoionization: Cross sections for vibrationally excited H_2

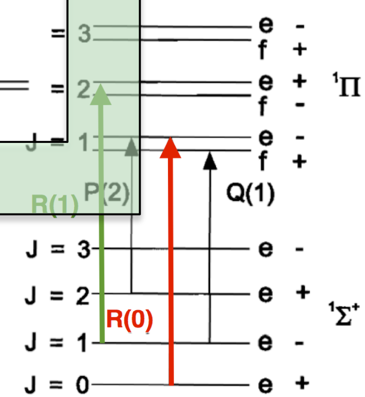
J. Zs. Mezei,[1,2,*] I. F. Schneider,[1,†] E. Roueff,[3] and Ch. Jungen[2,‡]

Results: H_2 - Photoionisation





upper state	number of resonances	ionization: overall contrib.	ionization: Total vs. backgr.	absorption: overall contrib.	absorption: Total vs. backgr.
$3p\pi D$	11	0.01		0.16	
$4p\pi D'$	14	0.23	6.2	0.10	8.1
$5p\pi D''$	15	0.12		0.06	
$6p\pi D'''$	12	0.08		0.04	
		~44%		~36%	



MQDT – *global* version

PHYSICAL REVIEW A **85**, 043411 (2012)

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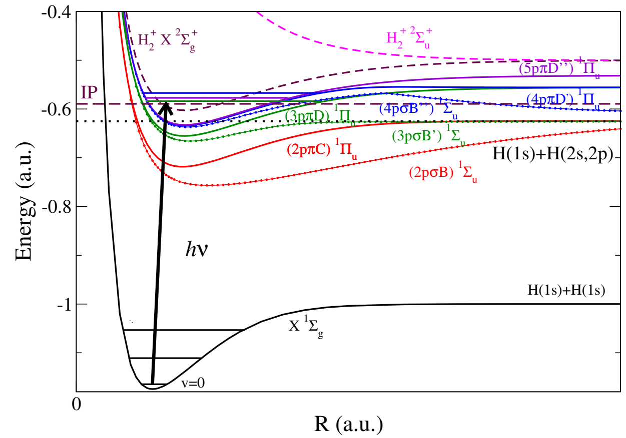

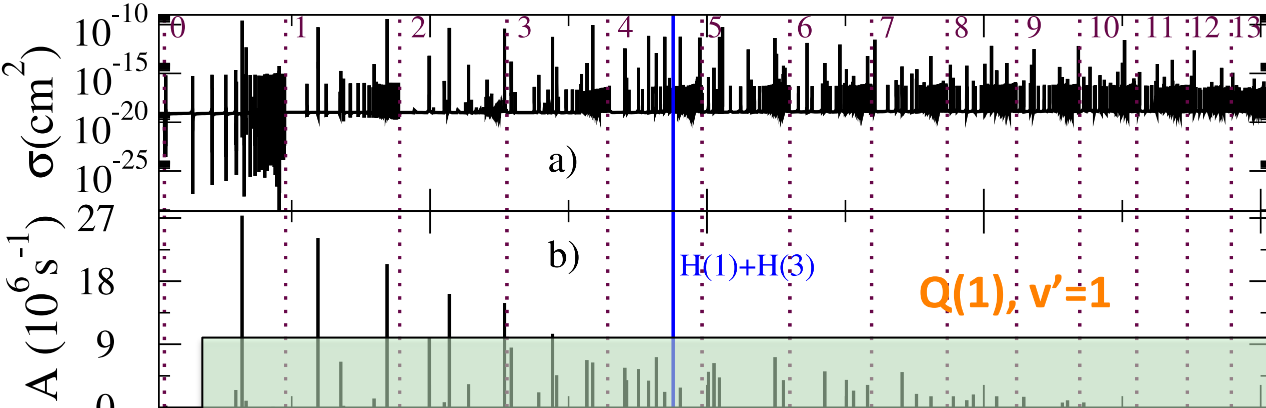

Q(1), v'=1

One c

upper state	number of resonances	ionization overall contrib.	ionization Total vs. backgr.	absorption overall contrib.	absorption Total vs. backgr.
$3p\pi D$	11	0.01		0.16	
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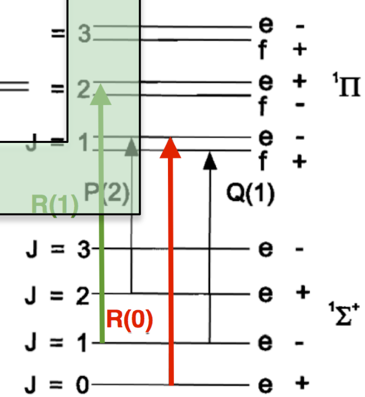


MQDT – *global* version

Transition energy (cm^{-1})

PHYSICAL REVIEW A **85**, 043411 (2012)

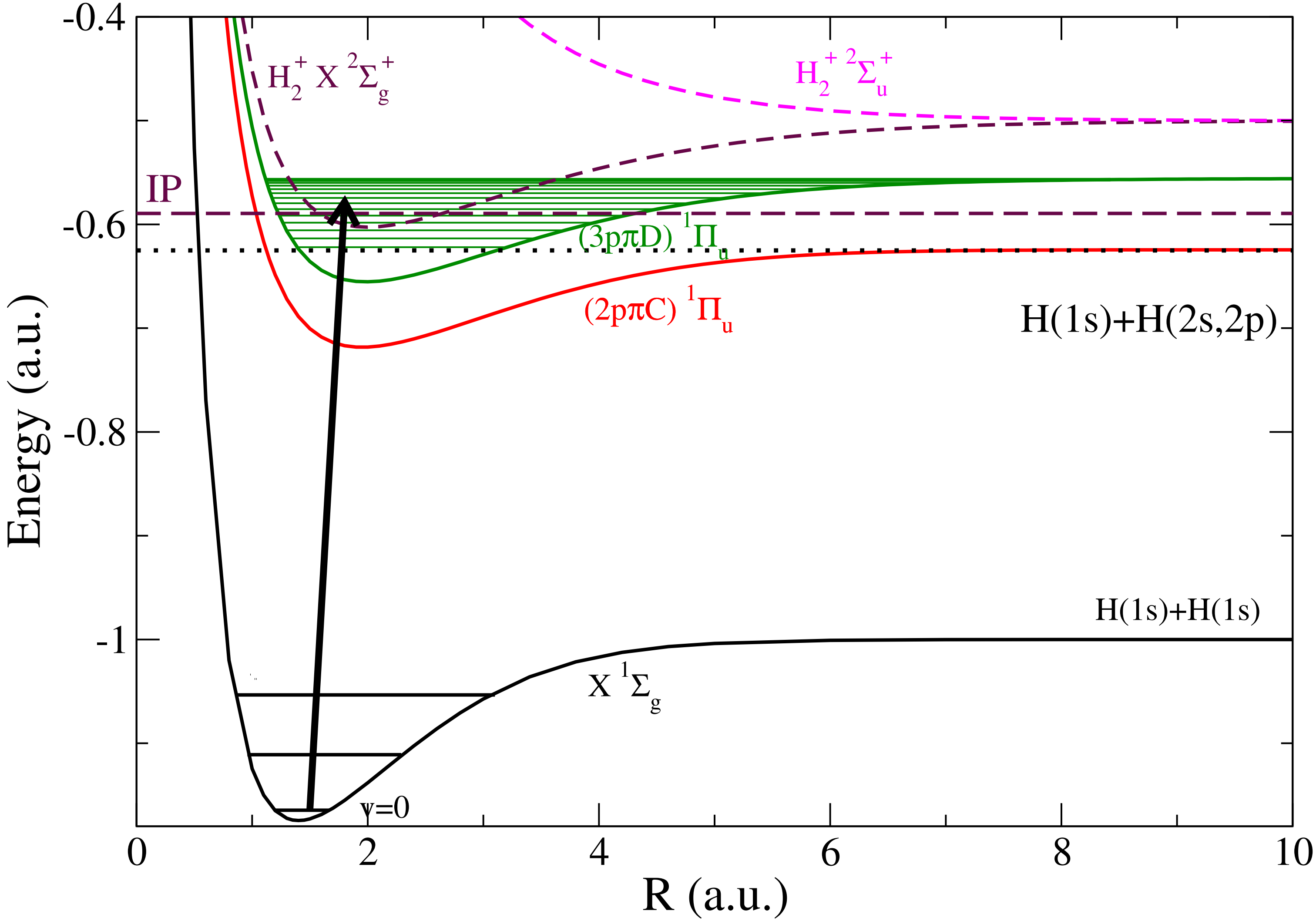
Resonances in photoionization: Cross sections for vibrationally excited H_2

J. Zs. Mezei,[1,2,*] I. F. Schneider,[1,†] E. Roueff,[3] and Ch. Jungen[2,‡]

Overview

- Introduction and motivations
- Theoretical framework and mechanisms
- Results
  - Photofragmentation
    - Photoionisation of H_2
    - Photodissociation of H_2
  - Anion-cation collisions
- Conclusions and future plans

Results: H_2 – Photodissociation



Results: H_2 – Photodissociation - *experiments*

THE JOURNAL OF CHEMICAL PHYSICS **133**, 144317 (2010)

Synchrotron vacuum ultraviolet radiation studies of the $D\,^1\Pi_u$ state of H_2

G. D. Dickenson,[1] T. I. Ivanov,[1] M. Roudjane,[2,a)] N. de Oliveira,[2] D. Joyeux,[2,3] L. Nahon,[2] W.-Ü. L. Tchang-Brillet,[4] M. Glass-Maujean,[5] I. Haas,[6] A. Ehresmann,[6] and W. Ubachs[1,b)]

- absorption spectra in the 74 – 94 nm range (115000 – 135000 cm^{-1})
- limited by Doppler broadening at 100 K (0.6 cm^{-1}), FT resolution: 0.35 cm^{-1}
- estimated accuracy: 0.06 cm^{-1} (Q-transition); $\sim$ 0.7 cm^{-1} (R-transition)

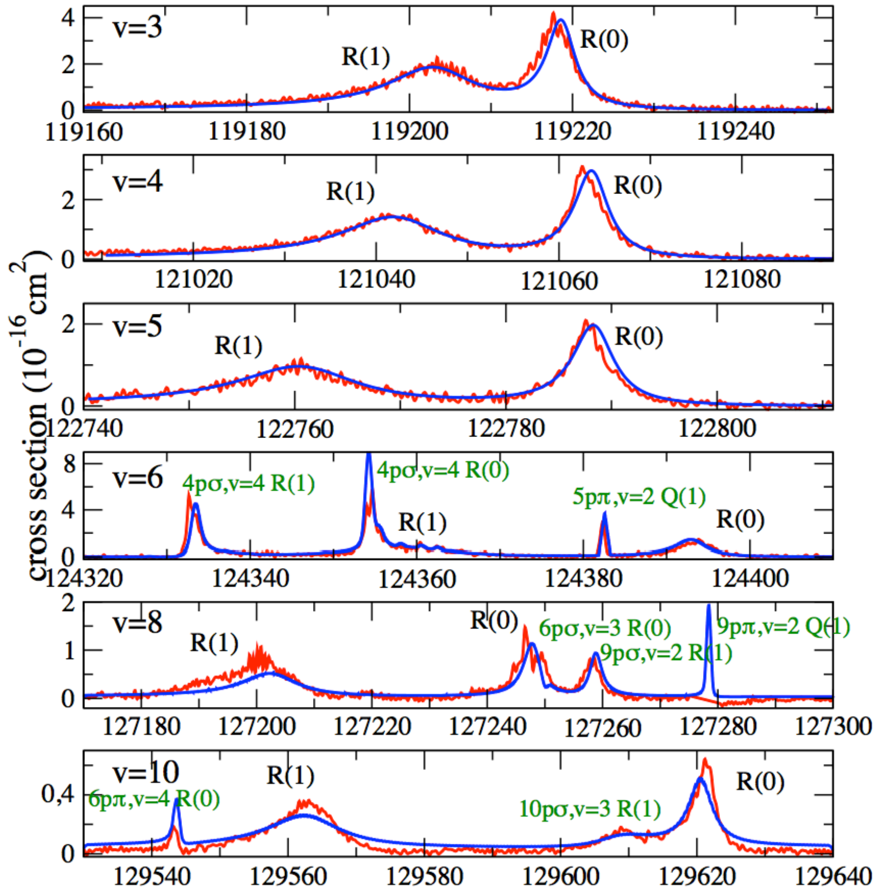
DESIRS beamline SOLEIL

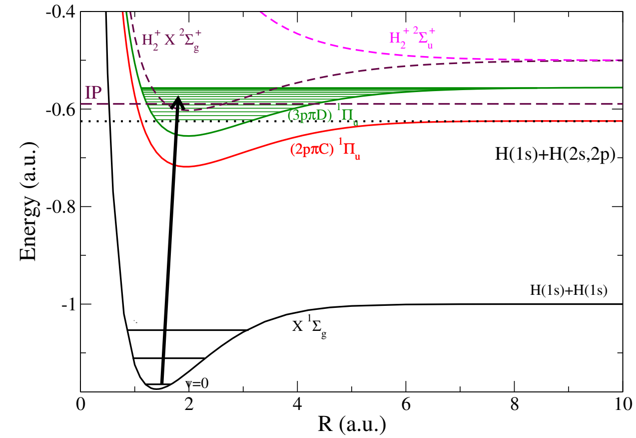
- Fourier transform spectrometer in the VUV
- high resolution and high absolute accuracy

U125/2 beamline BESSY II

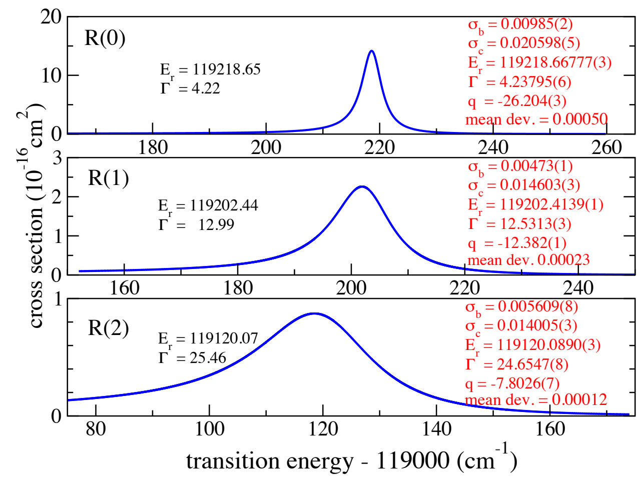
- 10m normal incidence scanning monochromator
- ionization, dissociation and fluorescence detection
- spectral resolution 2 cm^{-1}
- absolute intensity measurements

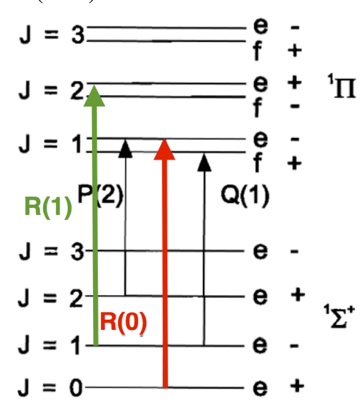
Results: H_2 - Photodissociation





Fano profiles



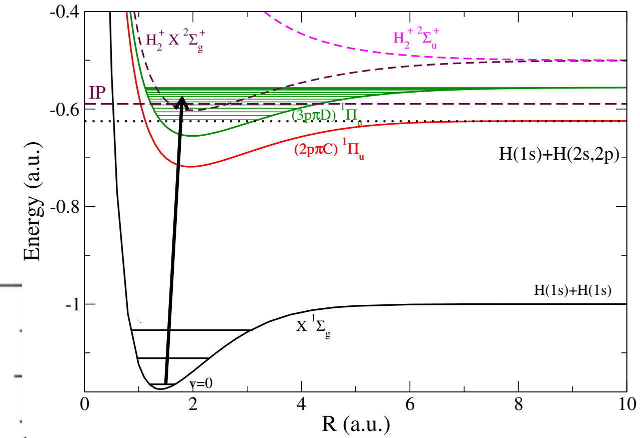


THE JOURNAL OF CHEMICAL PHYSICS **141**, 064305 (2014)

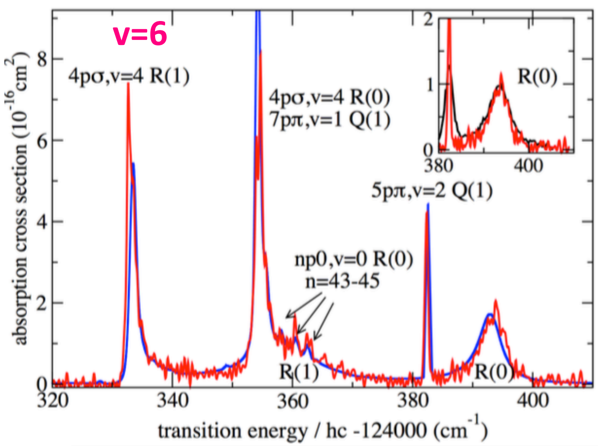
Resonances in photoabsorption: Predissociation line shapes in the $3p\pi\, D\,^1\Pi_u^+ \leftarrow X\,^1\Sigma_g^+$ system in H_2

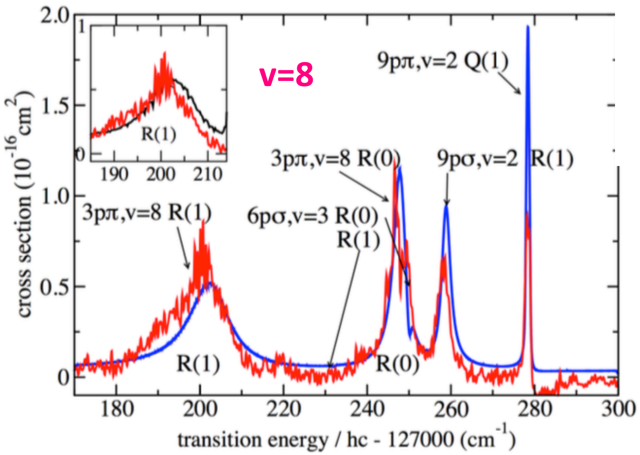
J. Zs. Mezei,[1,2,a)] I. F. Schneider,[2] M. Glass-Maujean,[3] and Ch. Jungen[1,4,b)]

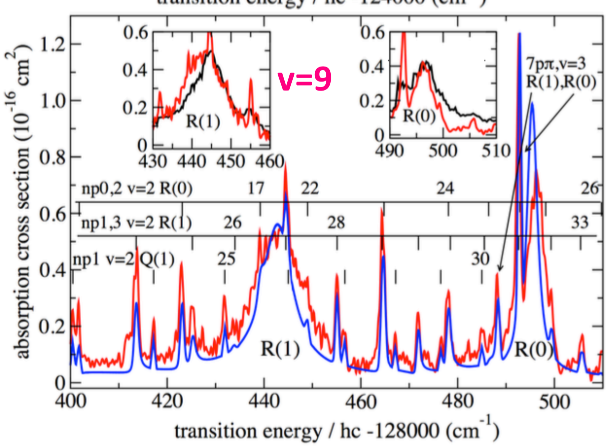
Results: H_2 - Photodissociation

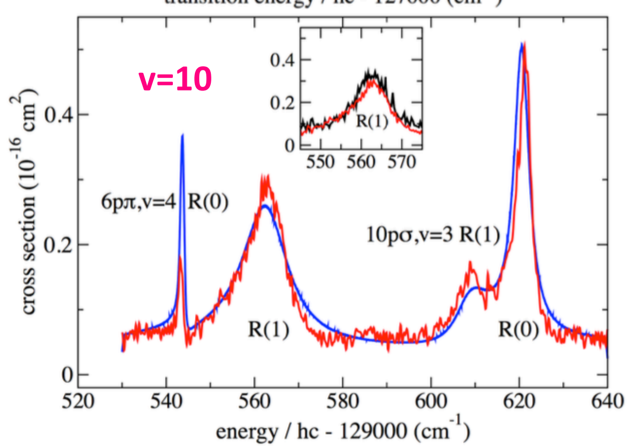


Very high spectral resolution (~0.7 cm^{-1})

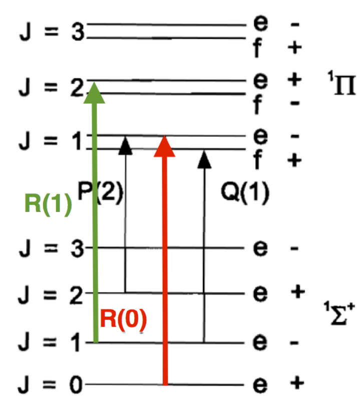








Very good agreement with the measurements performed on **SOLEIL** - Dickenson *et al*, JCP (2010) - and **BESSY** – Glass-Maujean *et al*, PRA (2012) - synchrotrons

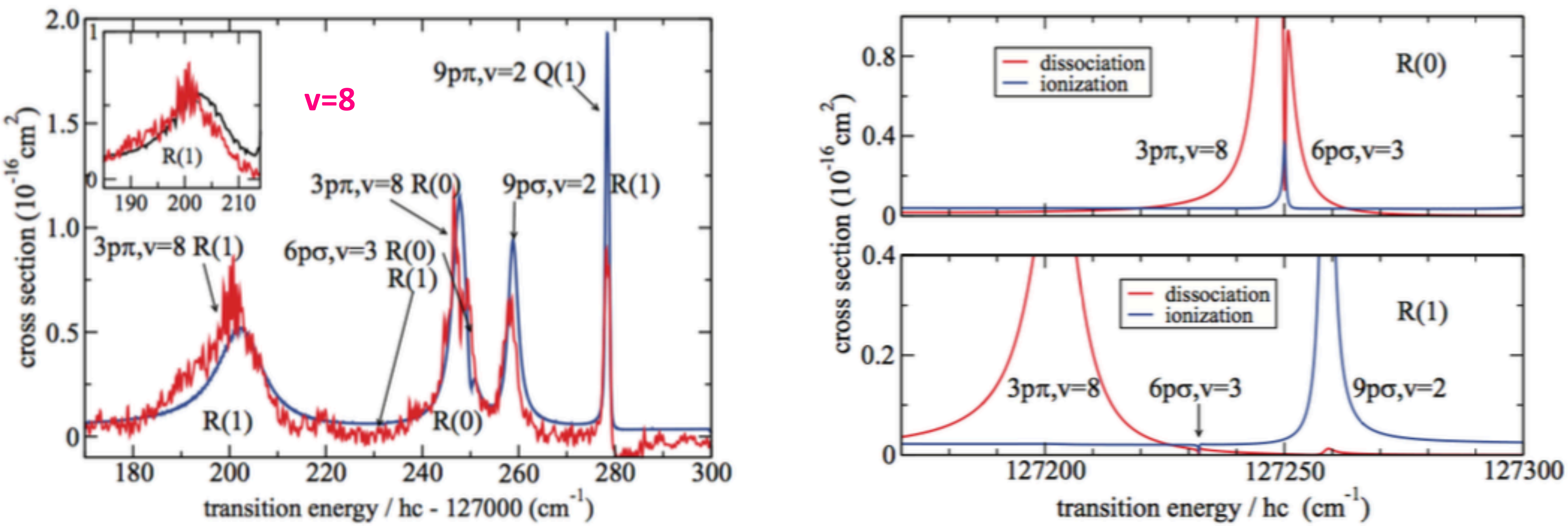


MQDT – *global* version: I+D

Resonances in photoabsorption: Predissociation line shapes in the $3p\pi\, D\,^1\Pi_u^+ \leftarrow X^1\Sigma_g^+$ system in H_2

J. Zs. Mezei,[1,2,a)] I. F. Schneider,[2] M. Glass-Maujean,[3] and Ch. Jungen[1,4,b)]

Results: H_2 – Photodissociation - competition



THE JOURNAL OF CHEMICAL PHYSICS **141**, 064305 (2014)

Resonances in photoabsorption: Predissociation line shapes in the $3p\pi\, D\, ^1\Pi_u^+ \leftarrow X^1\Sigma_g^+$ system in H_2

J. Zs. Mezei,[1,2,a)] I. F. Schneider,[2] M. Glass-Maujean,[3] and Ch. Jungen[1,4,b)]

Results: H_2 – Photoabsorbtion - *experiments*

THE JOURNAL OF CHEMICAL PHYSICS **144**, 084303 (2016)

CrossMark

Absorption, autoionization, and predissociation in molecular hydrogen: High-resolution spectroscopy and multichannel quantum defect theory

M. Sommavilla,[1] F. Merkt,[1] J. Zs. Mezei,[2,3] and Ch. Jungen[3,a)]
[1]*Laboratorium für Physikalische Chemie, ETH-Zürich, 8093*
[2]*Laboratoire Ondes et Milieux Complexes, UMR 6294 CNRS*
BP 540, F-76058 Le Havre, France
[3]*Laboratoire Aimé Cotton du CNRS, Bâtiment 505, Universit*

(Received 16 December 2015; accepted 21 January 201

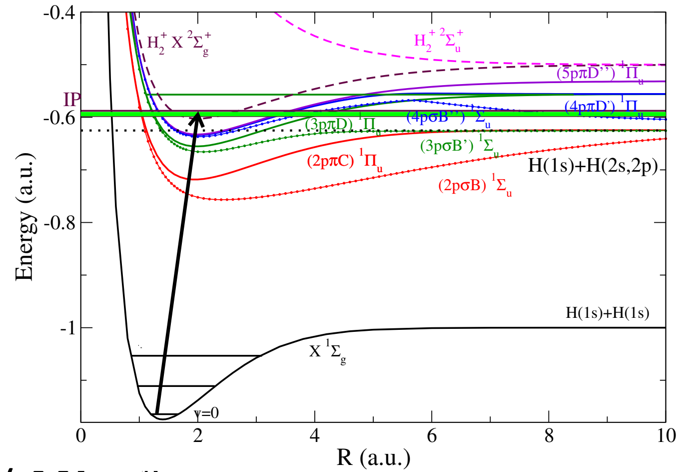
INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS B: ATOMIC, MOLECULAR AND OPTICAL PHYSICS
J. Phys. B: At. Mol. Opt. Phys. **35** (2002) 3901–3921 PII: S0953-4075(02)38026-X

High-resolution laser absorption spectroscopy in the extreme ultraviolet

M Sommavilla, U Hollenstein, G M Greetham and F Merkt

Absorption and photoionization spectra of H_2 have been recorded at a resolution of 0.09 and 0.04 cm^{-1}, respectively, between 125 600 cm^{-1} and 126 000 cm^{-1}. The observed Rydberg states belong to series ($n = 10 – 14$) converging on the first vibrationally excited level of the X $^2\Sigma_g^+$ state of H_2^+, and of lower members of series converging on higher vibrational levels. The observed resonances are characterized by the competition between autoionization, predissociation, and fluorescence. The unprecedented resolution of the present experimental data leads to a full characterization of the predissociation/autoionization profiles of many resonances that had not been resolved previously. Multichannel quantum defect theory is used to predict the line positions, widths, shapes, and intensities of the observed spectra and is found to yield quantitative agreement using previously determined quantum defect functions as the unique set of input parameters. © *2016 AIP Publishing LLC.* [http://dx.doi.org/10.1063/1.4941920]

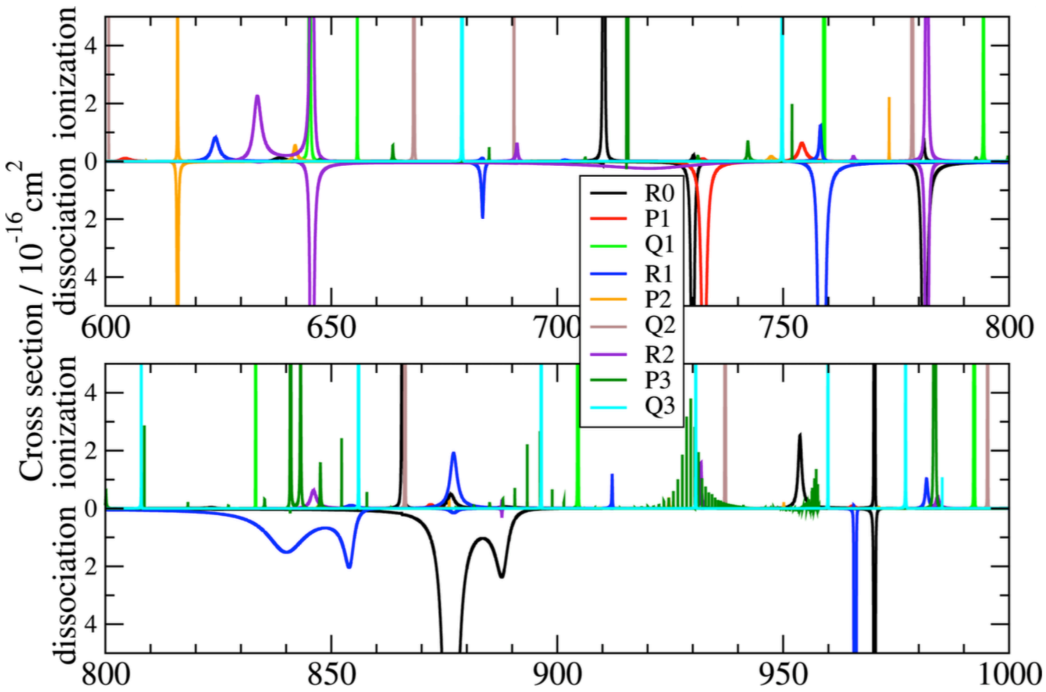
Results: H_2 - Photoabsorption



Doppler-free (~0.04 cm⁻¹)

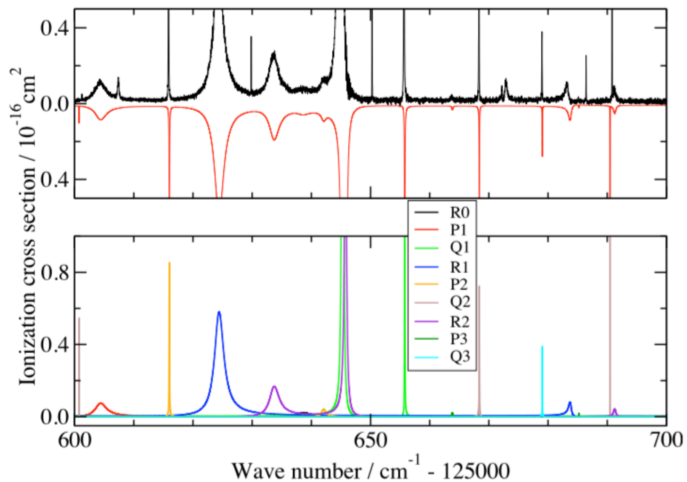
VUV-laser based spectroscopy

$v^+=0$



$v^+=1$

MQDT

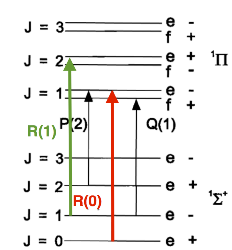


MQDT – *global* version: I+D

THE JOURNAL OF CHEMICAL PHYSICS **144**, 084303 (2016)

Absorption, autoionization, and predissociation in molecular hydrogen: High-resolution spectroscopy and multichannel quantum defect theory

M. Sommavilla,[1] F. Merkt,[1] J. Zs. Mezei,[2,3] and Ch. Jungen[3,a)]



Overview

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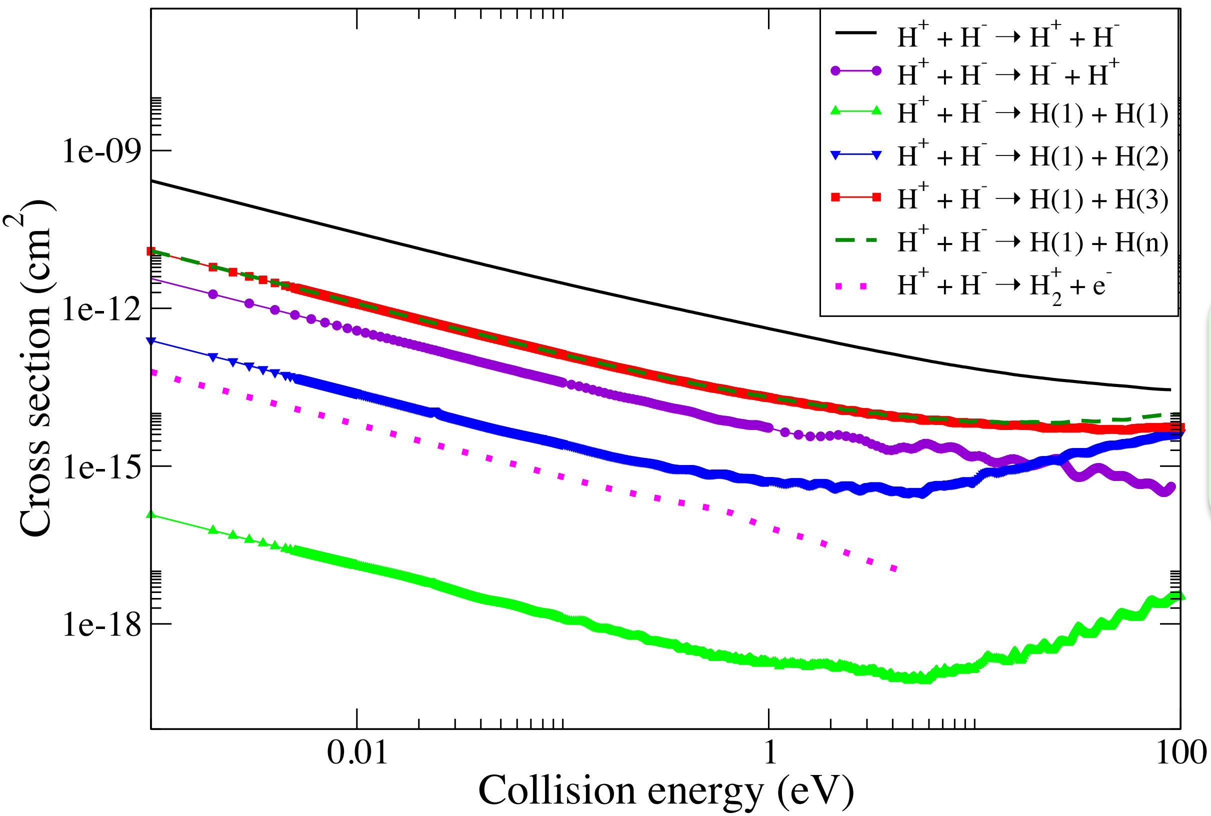
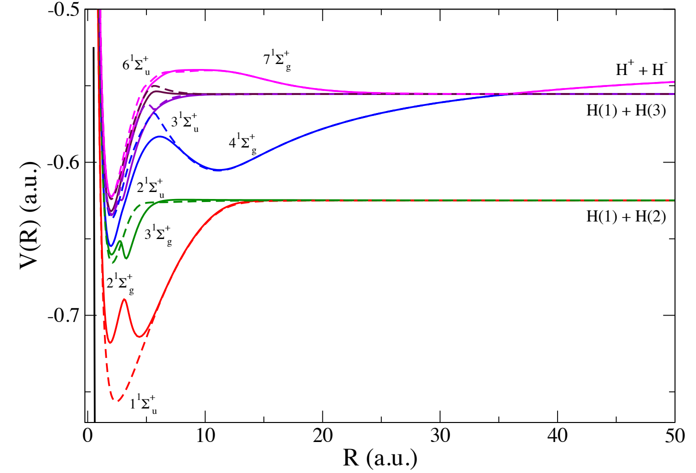
    - Photodissociation of H_2
  - Anion-cation collisions
- Conclusions and future plans

Anion-cation: Reactive collisions

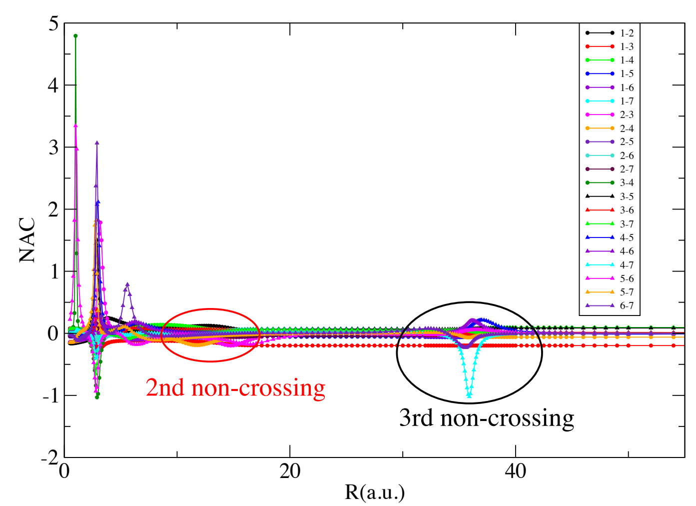
$$A^+ + B^- \rightarrow \begin{cases} A^+ + B^- & \mathrm{EC} \\ A^- + B^+ & \mathrm{CT} \\ A(n) + B(m) & \mathrm{MN} \end{cases}$$

Results: low energy $H^+ + H^-$





- **PES**: **Molpro** et **Wolniewicz** *et al.* – Strict diabatisation
- **Nuclear dynamics**: Molecular close coupling method

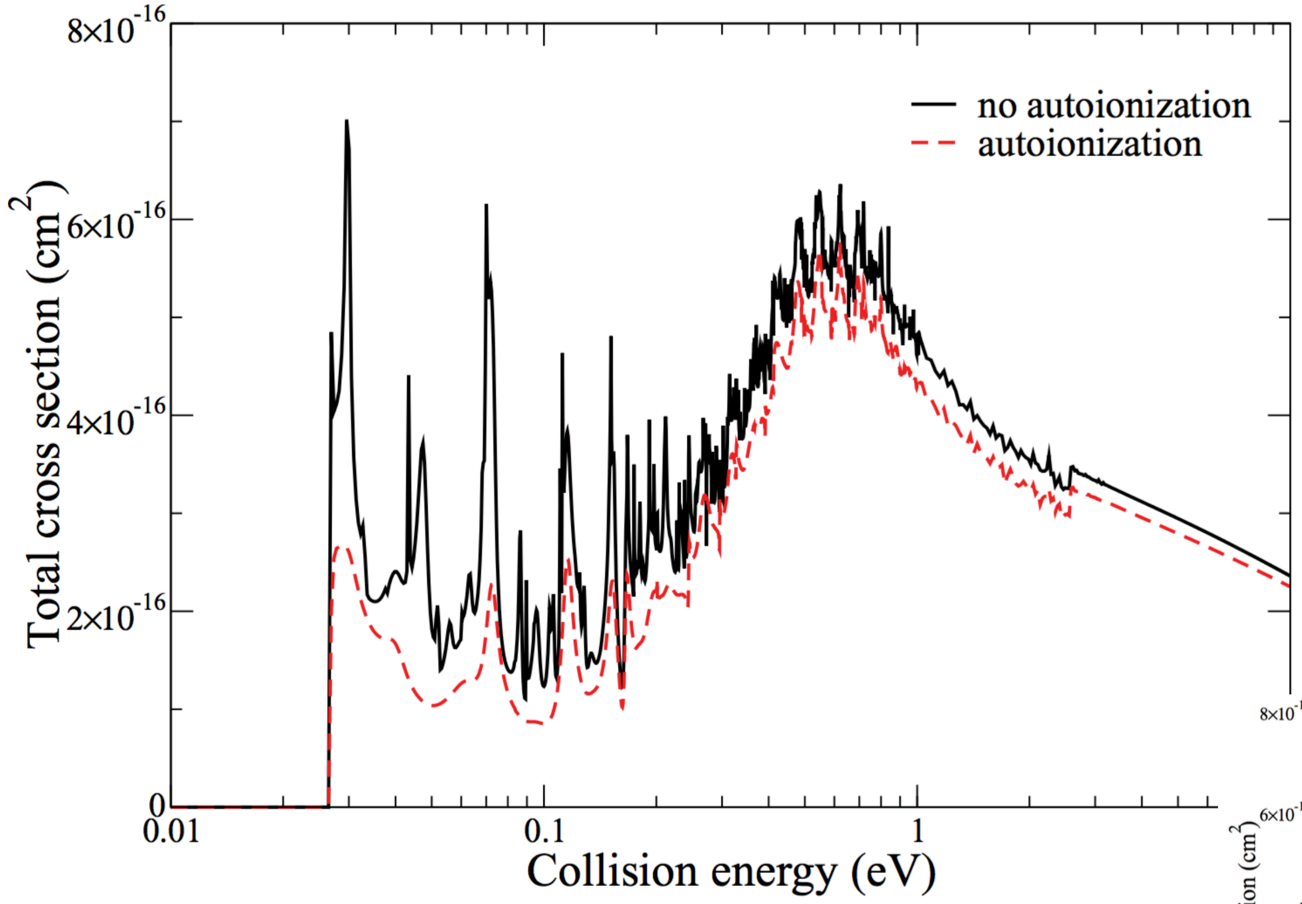


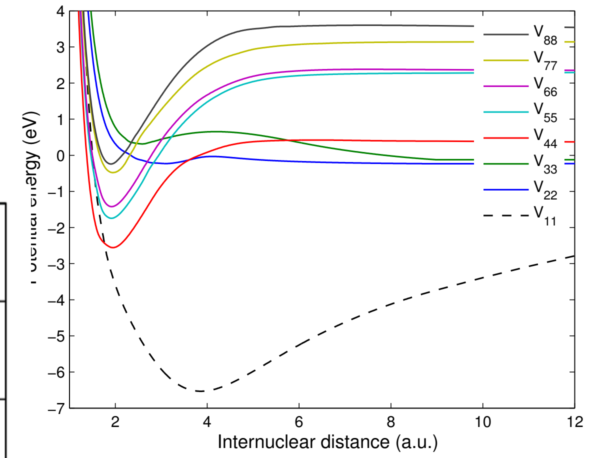
PHYSICAL REVIEW A **82**, 014701 (2010)

Double charge transfer in low-energy $H^+ + H^-$ collisions

J. Zs. Mezei,* M. Stenrup, N. Elander, and Å. Larson
Department of Physics, Stockholm University, AlbaNova University Center, S-106 91 Stockholm, Sweden

Results: low energy $H^+ + F^-$





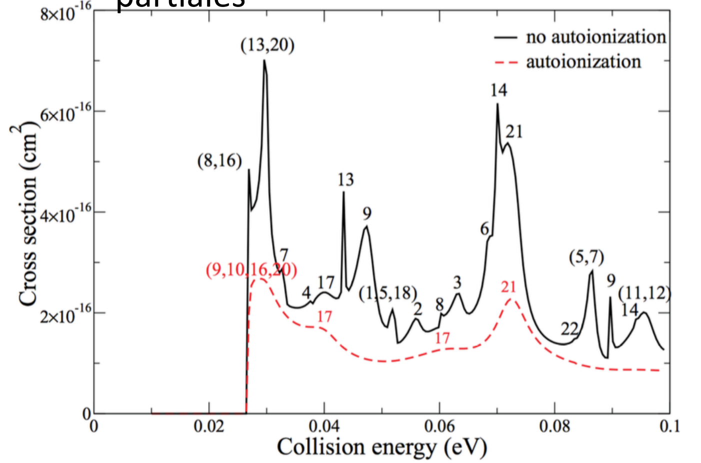
J. B. Ross *et al*: PRA **80**, 012501 (2009):

PES – MRCI + Kohn variational method

quasidiabatisation (2x2 rotations)

L'autoionisation !!!

Résonances: les indexes des ondes partiales



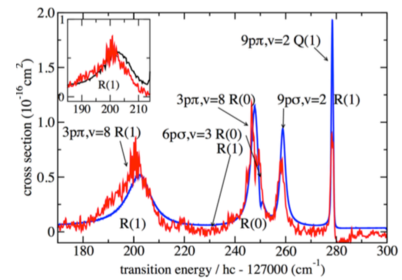
PHYSICAL REVIEW A **84**, 012703 (2011)

Mutual neutralization in low-energy $H^+ + F^-$ collisions

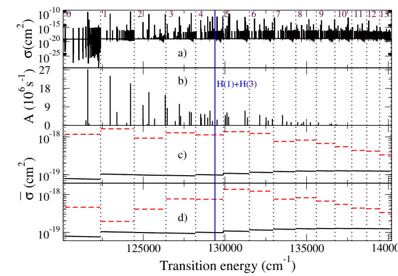
J. Zs. Mezei,[1,2,3] J. B. Roos,[3] K. Shilyaeva,[4,3] N. Elander,[3] and Å. Larson[3,*]

Conclusions

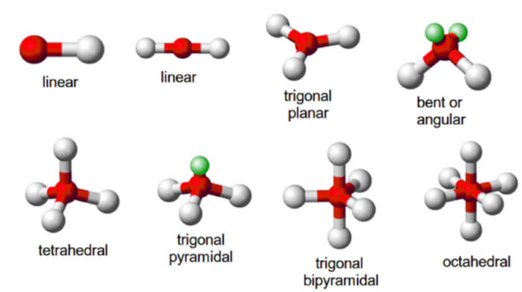
- MQDT: state-to-state calculations



- Temporary captures into super-excited states: **HUGE RESONANT EFFECTS**



- Full study on H_2 and other systems (di- and poly-atomics)



TRIANGLE DE LA PHYSIQUE

International Atomic Energy Agency
IAEA

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ANR

PROGRAMME BLANC
Acronyme SUMOSTAI
EDITION 2009

SUperexcited MOlecular STates of Astrophysical Importance : dissociative recombination and spectroscopy

Thank you for the attention!

NEMZETI KUTATÁSI, FEJLESZTÉSI ÉS INNOVÁCIÓS HIVATAL
AZ INNOVÁCIÓ LENDÜLETE

AZ NKFI ALAPBÓL MEGVALÓSULÓ PROJEKT

2018-1.2.1-NKP

Investigation of atomic and molecular processes initiated by ultrashort photon and electron pulses at ELI-ALPS, development of method and instrument

OTKA grant K128621