Max-Planck-Institut
für Plasmaphysik

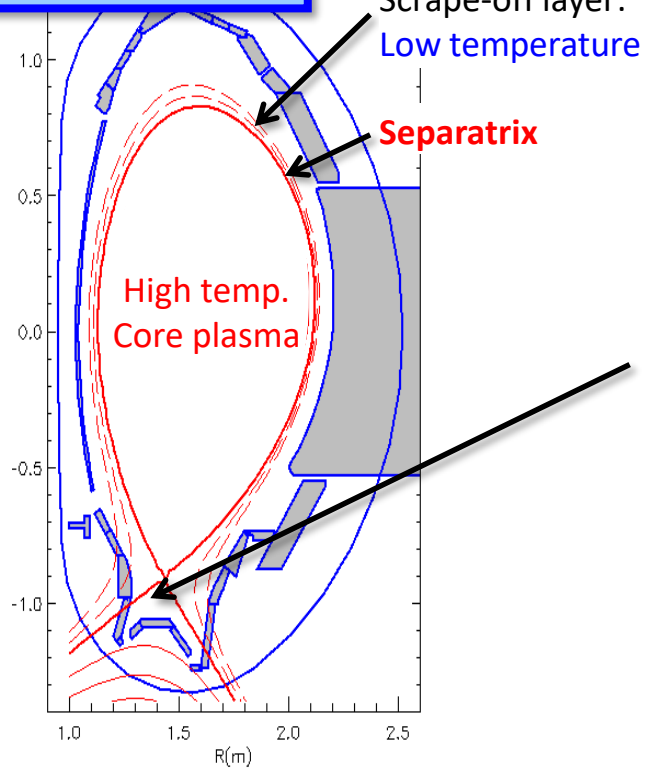
# Collisional radiative models for atomic and molecular hydrogen in the edge plasma of fusion devices

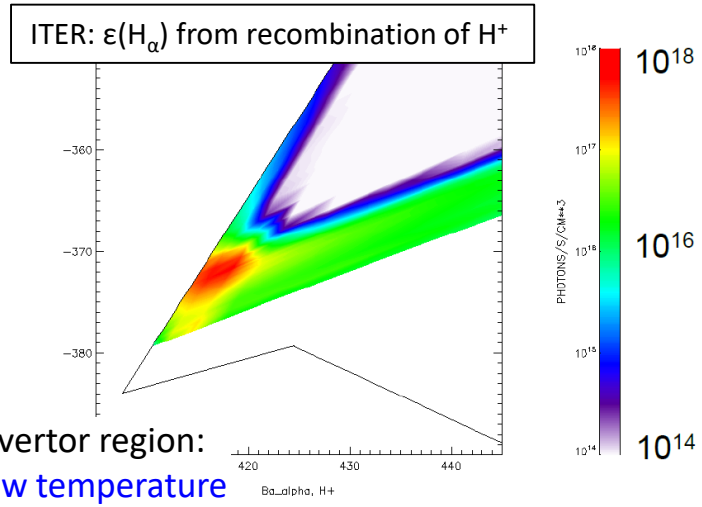
D. Wünderlich and U. Fantz

# Edge plasma of fusion devices.

Plasma edge of fusion device

Wide parameter range
$T_e$=50 eV … below 1 eV
$n_e$=$10^{18}$ $m^{-3}$ … $10^{21}$ $m^{-3}$
Ionizing → recombining




Picture by courtesy of D. Reiter

- Relevant gases: $H_2$, $D_2$ and/or $T_2$.
- High relevance of atomic and also molecular processes, especially in the detached case.
- Molecular assisted recombination.
- Impurity seeding ($N_2$, Ar, …)
  ⇒ enhanced radiation losses.
- Optical thickness.

# Molecules in the divertor.

… and their role for recombination and ionization of the plasma.

**Electron Ion Recombination EIR**

$H^+ + e^- + e^- \rightarrow H^* + e^-$ 3-body rec. and/or

$H^+ + e^- \rightarrow H^* + h\nu$ radiative recombination

**Plasma ionization**

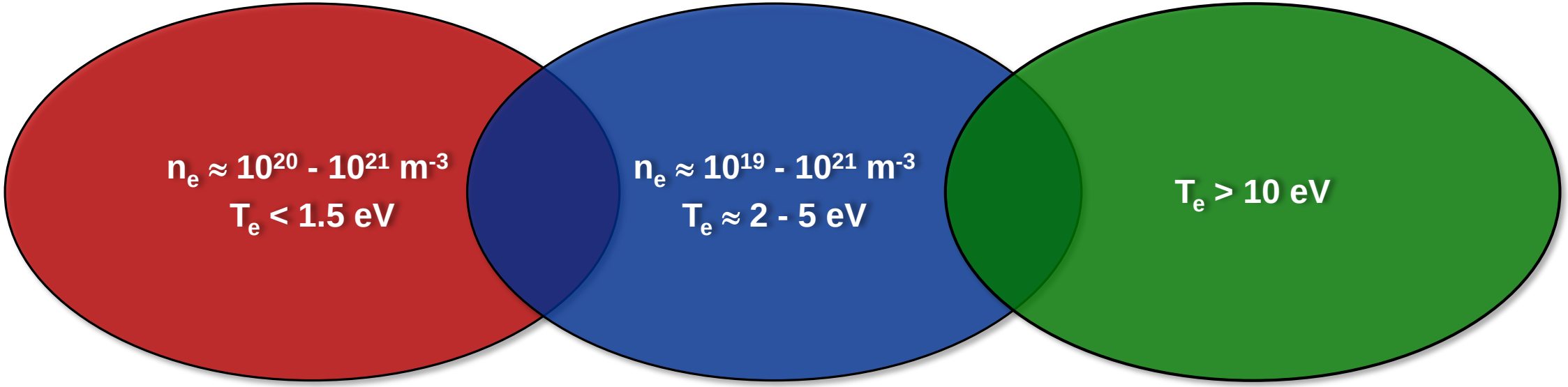
$H + e^- \rightarrow H^+ + 2e^-$ ionization of atoms and

$H_2(v) + e^- \rightarrow H_2^+ + 2e^-$ ionization of molecules

**Molecular Assisted Recombination MAR**

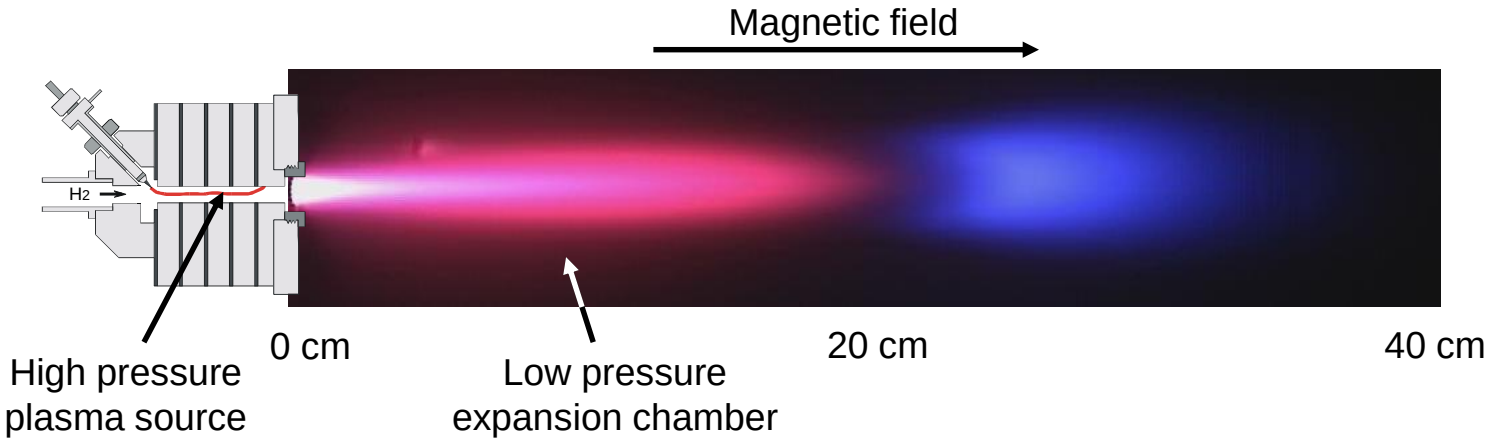
$\mathbf{H_2(v)} + H^+ \rightarrow H_2^+(v) + H$ charge exchange with

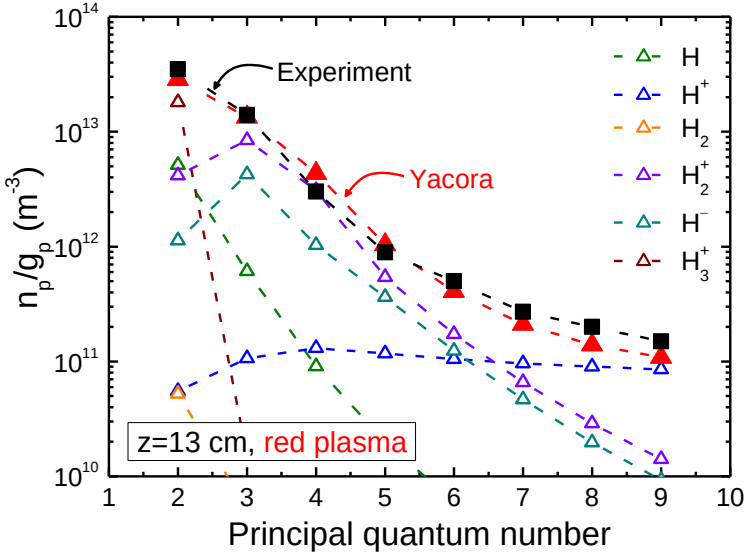
$H_2^+(v) + e^- \rightarrow H + H^*$ dissociative recombination



# Transition ionizing – recombining plasma.

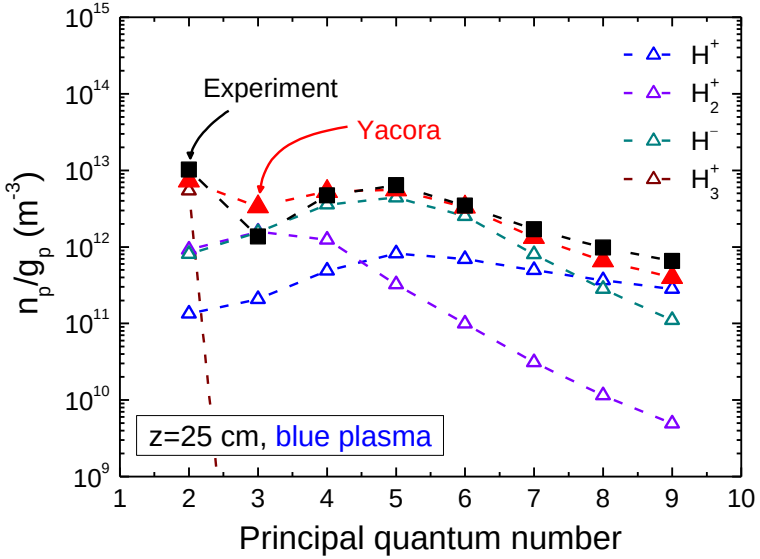
**Magnetized plasma expansion at TU/e**




**Excellent agreement experiment ↔ Yacora for red and blue plasma**

- Correct prediction of overpopulation of n=4,5 and 6.
- Mutual neutralization of $H_2^+$ with $H^-$: the two reaction channels proposed by Janev and Eerden exist simultaneously.
- Recombination of $H_3^+$ has to be taken into account (in order to fulfill quasi neutrality).

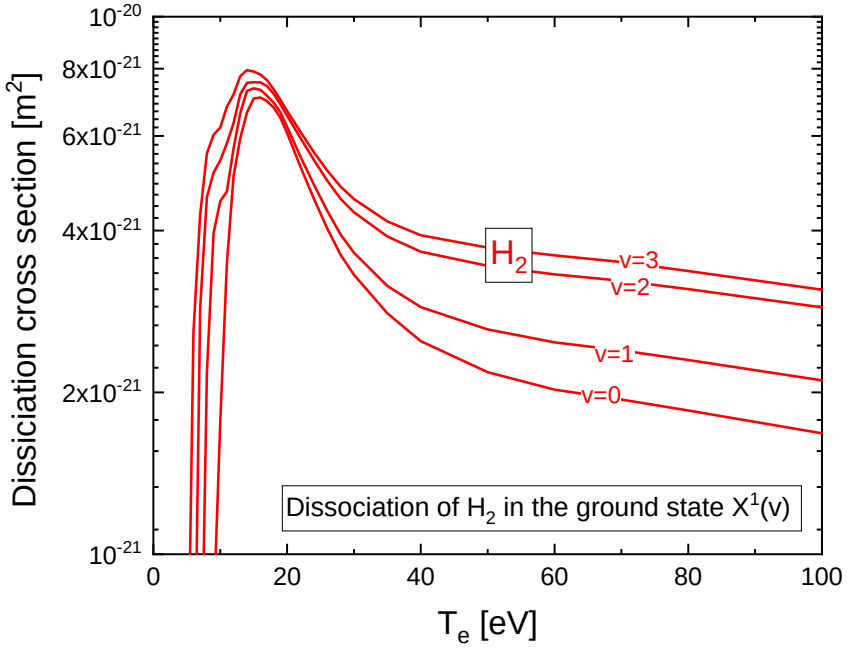



W.E.N. van Harskamp, Phys. Rev. E 83, 2011, 036412
R. Janev et al, JÜL-4105, 2003
M.J.J. Eerden at al, Phys. Rev. A 51, 1995, 3362

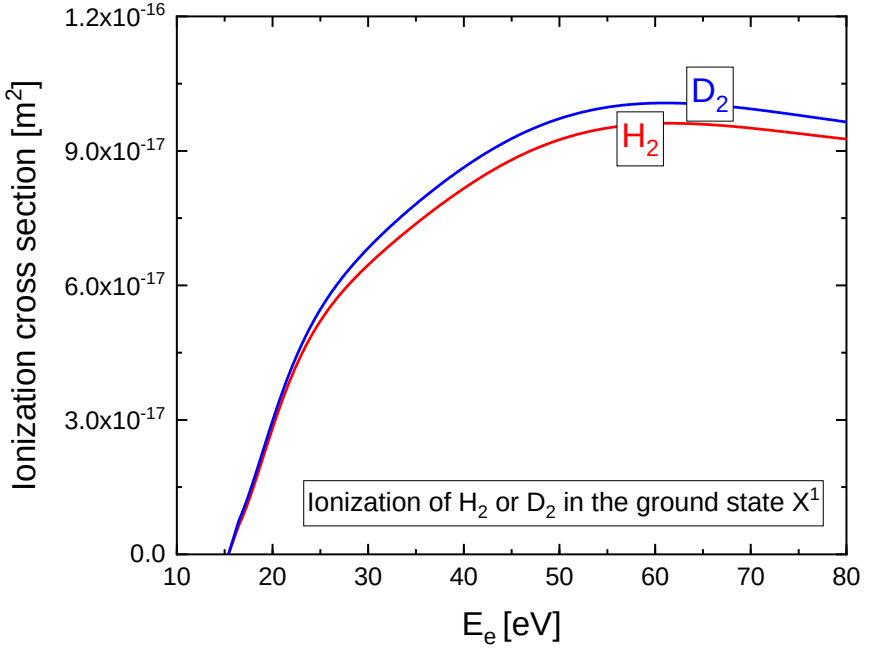
# Isotope effect and ro-vibrational excitation.

Isotope effect mainly caused by:

- More tight vibrational and rotational energy level spacing in $D_2$.
- Impact on wave functions and threshold energies.




L. Scarlett et al, Atoms 7, 2019, 75

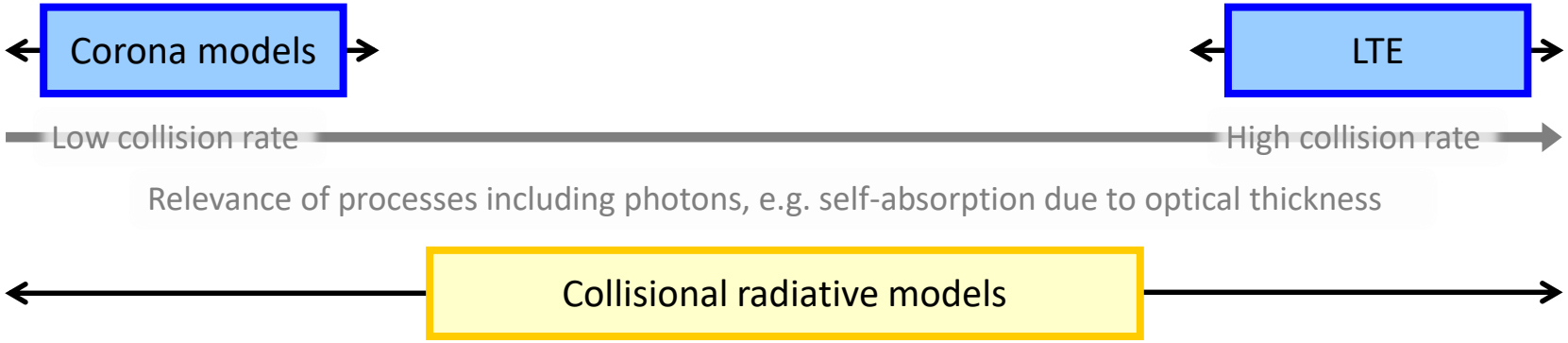



J.S. Yoon et al, J. Phys. Chem. Ref. Data 37, 2008, 913
J.S. Yoon et al, Rep. Prog. Phys. 73, 2010, 116401

# Population models for fusion edge plasmas.

Population models

- Predict population densities in dependence of plasma parameters ($T_e$, $n_e$, ground state densities).
- Main field of application: plasma diagnostics.




Collisional radiative models

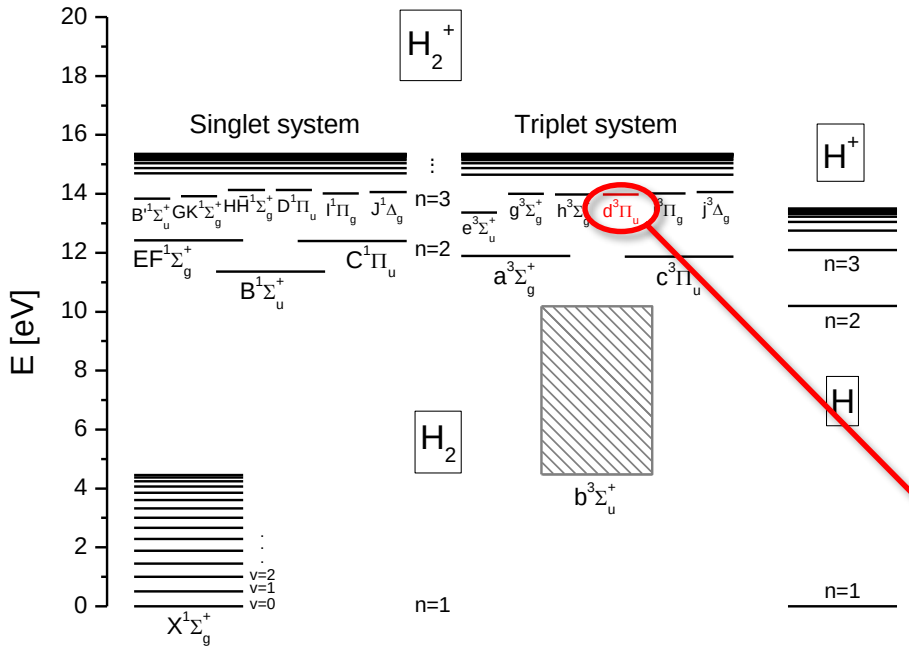
Balance all relevant exciting and de-exciting reactions.

⇒ Needed: extensive data base of reaction probabilities.

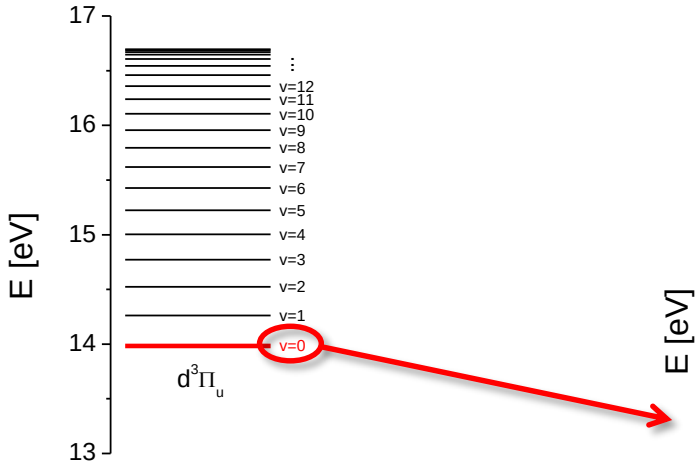
⇒ Drastically increased complexity for molecules (vibrational and rotational excitation).

Error bar of model results directly correlates with the quality of the used input data.

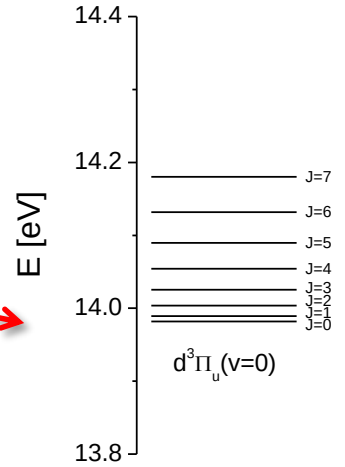
# CR models for molecular hydrogen: structure of $H_2$.




Electronic excitation




Vibrational excitation




Rotational excitation

Large number of excited states

**Needed:**

Probabilities for all reactions interconnecting all excited states

# CR models for molecular hydrogen: input cross sections.

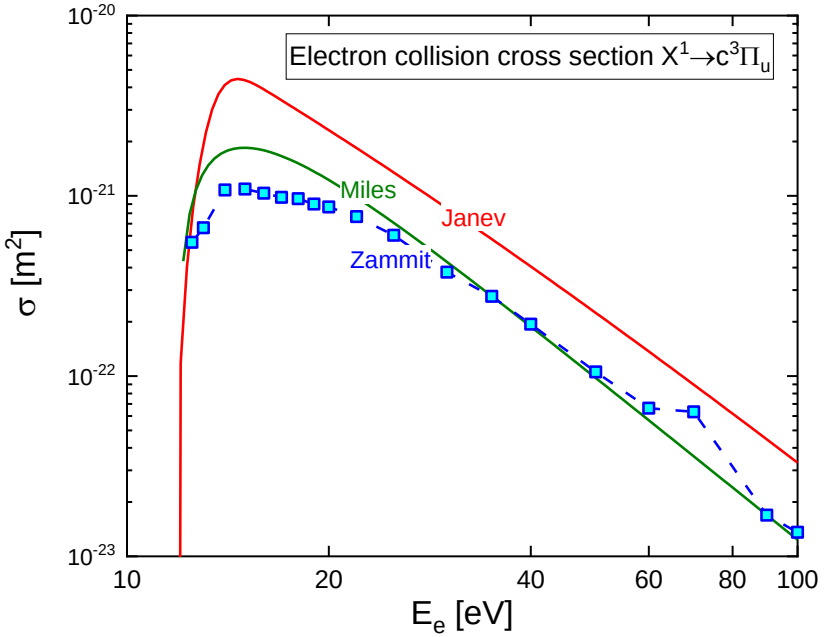
Electron collision excitation processes in the $H_2$ molecule:

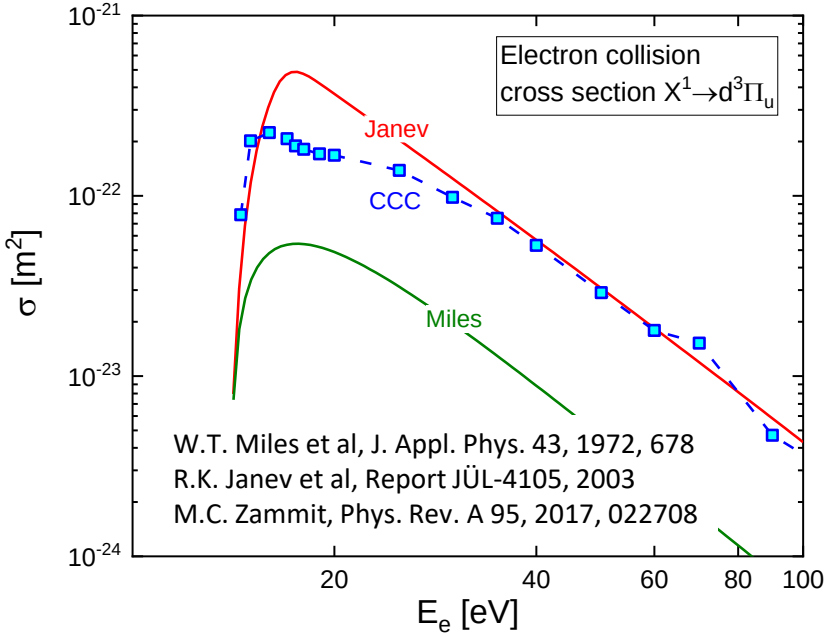
- Data collections by Tabata (2000) and Yoon (2008): only a few reactions.
- Calculations by Celiberto (2001): only a few transitions, but isotope effect.
- Miles: semi empiric cross sections, 1972.
- Janev: summary of recent measurements and calculations, 2003.

} Significant inconsistencies

- New consistent data set: MCCC (Molecular Convergent Close-Coupling), fully quantum-mechanical.
  Since 2017 close collaboration with the Group of D. Fursa, Curtin University, Australia.

T. Tabata et al, ADNDT 76, 2000,1
J.S. Yoon et al, J. Phys. Chem. Ref. Data 37, 2008, 913
R. Celiberto et al, ADNDT 77, 2001, 161



# MCCC cross sections for $H_2$ CR models.

MCCC allows for the first time using in CR models a comprehensive set of input cross sections for electron-impact excitation:

- Excitation from ground state to n=2 and n=3.

  ***previously: significant inconsistencies.***

- Excitation within n=2.

  ***previously: σ for hydrogen-like ions.***

- Excitation from n=2 to n=3.

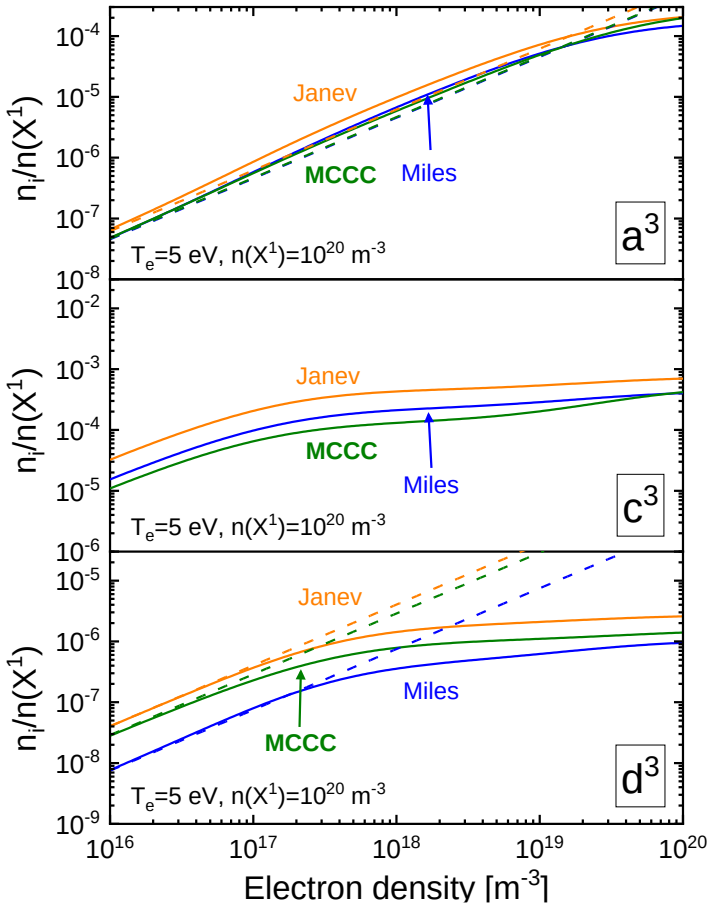
  ***previously: scaled σ for the hydrogen atom.***

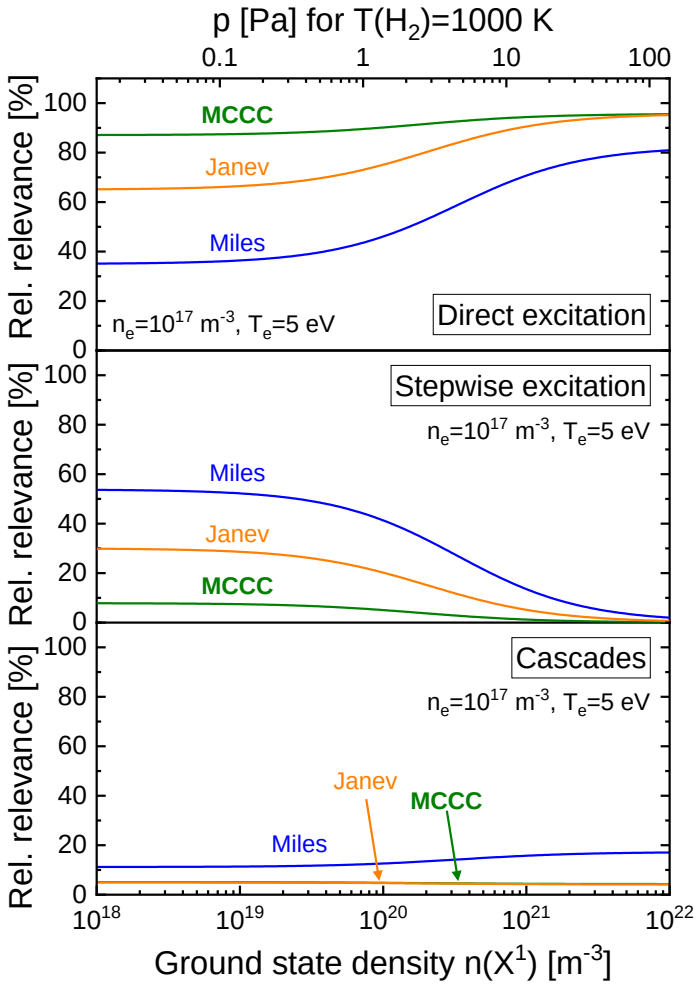
Significant differences in results based on MCCC cross sections and the previously used data.

D. Wünderlich et al, J. Phys. D 54, 2021, 115201

Yacora CR model for the electronic states of the triplet system in $H_2$

W.T. Miles at al, J. Appl. Phys. 43, 1972, 678.
R. Janev et al, Report JÜL-4105, 2003.



# Validation of the MCCC cross sections for $H_2$.

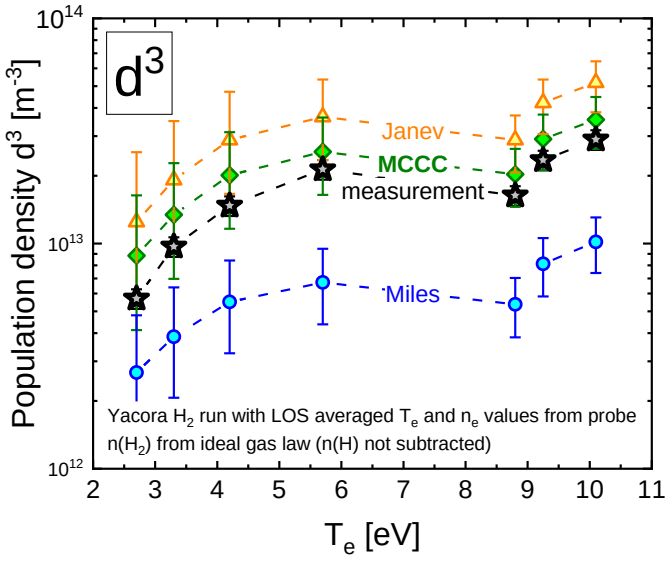
**Yacora CR model for the triplet system** used for validating MCCC cross sections:

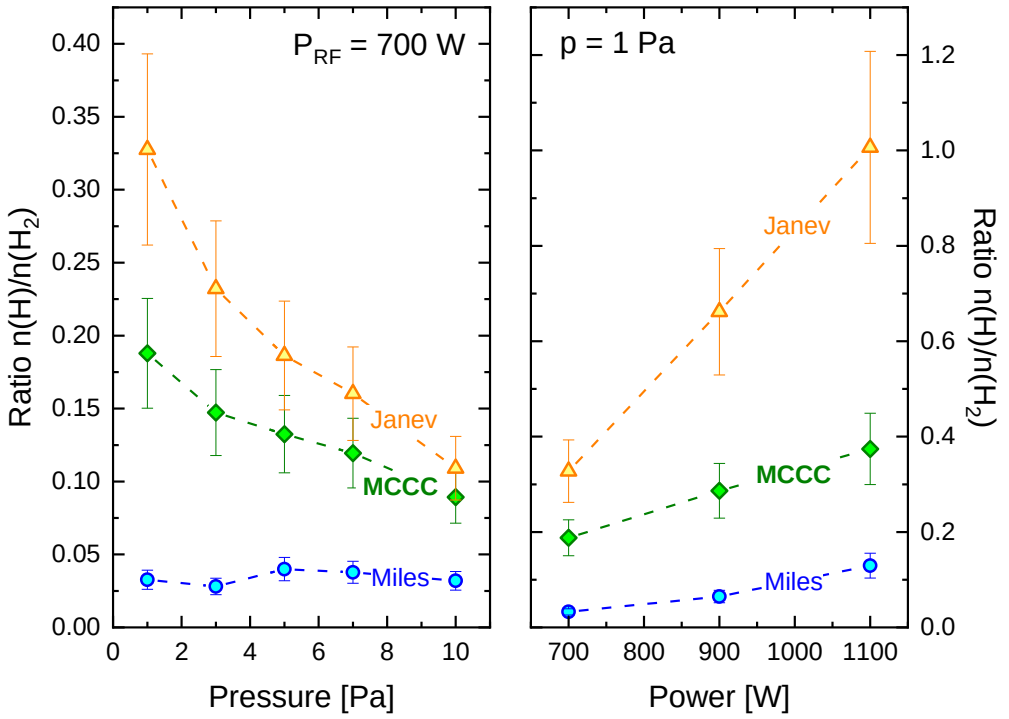
- Low pressure, low-temperature lab experiment.
- $T_e$ and $n_e$ from Langmuir probe.
- Low electron densities ($<10^{17}$ $m^{-3}$).

Several excited states: agreement measurement ↔ CR model much better than using previously available cross sections.

Application to plasma diagnostics:

- Strong impact on results.
- Example: linear effect on density ratio $n(H)/n(H_2)$, but affects also particle fluxes, …




D. Wünderlich et al, J. Phys. D 54, 2021, 115201

# Ionization cross section for $H_2$ and isotopomeres.

**2011:** comprehensive set of cross sections for ionization of molecular ground state $X^1$ and excited states, based on the semi-classical Gryzinski method.

Successful validation vs. experimental and theoretical data for $X^1$.

Two possible reaction channels:

Non-dissociative ionization:

$$\mathrm{H}_2(p', v') + e^- \rightarrow \mathrm{H}_2^+\left(^2\Sigma_\mathrm{g}^+, v''\right) + 2e^-$$

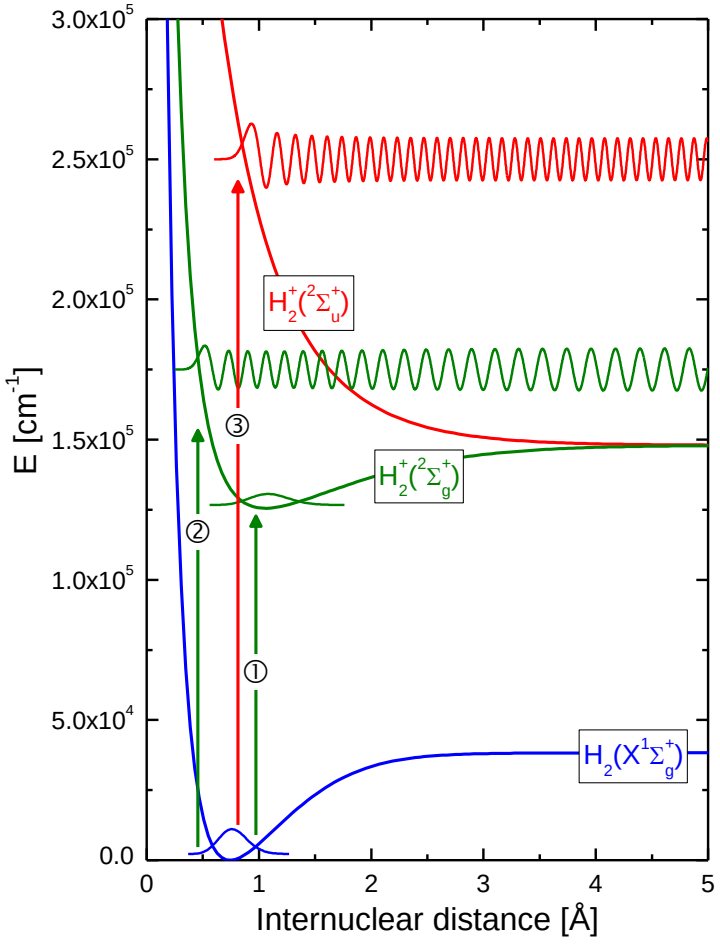
Dissociative ionization:

$$\mathrm{H}_2(p', v') + e^- \rightarrow \mathrm{H}_2^+\left(^2\Sigma_\mathrm{g}^+, E''\right) + 2e^- \rightarrow \mathrm{H} + \mathrm{H}^+ + 2e^-$$

$$\mathrm{H}_2(p', v') + e^- \rightarrow \mathrm{H}_2^+\left(^2\Sigma_\mathrm{u}^+, E''\right) + 2e^- \rightarrow \mathrm{H} + \mathrm{H}^+ + 2e^-$$

Needed for calculation of cross sections:

Franck Condon factors $q_{v'v''}^{p'p''}$ and Franck Condon densities…



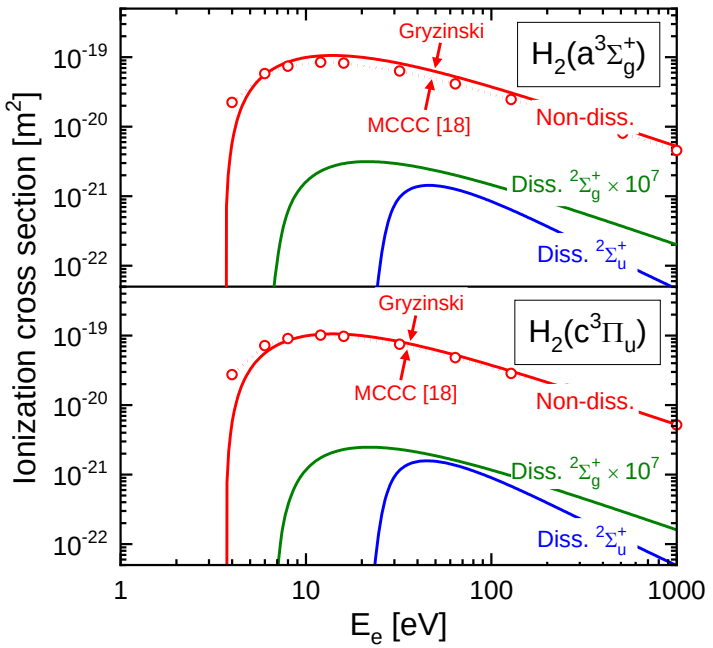

D. Wünderlich, Chem. Phys. 390, 2011, 75

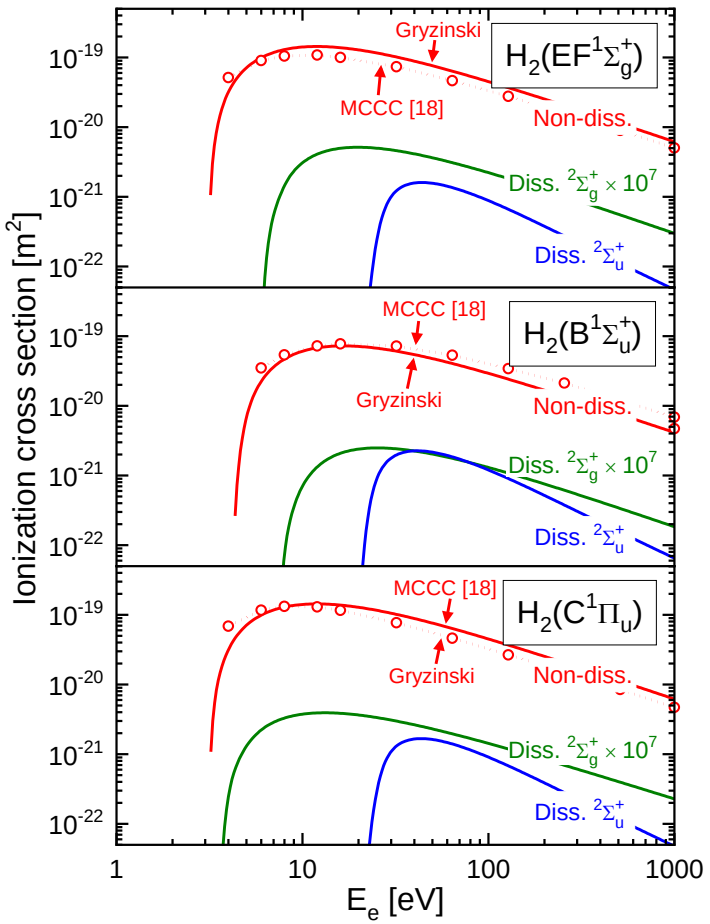
# Ionization cross sections: results.

**2021:** extension of the existing set of Gryzinski ionization cross sections:

- Cross sections for all isotopomeres of molecular hydrogen: $H_2$, $D_2$, $T_2$, HD, HT, DT.
- Removed a calculation error (affecting the cross sections for excited states).
- Validation also for excited states of $H_2$ versus MCCC.

Very good agreement with MCCC: semi-classical Gryzinski ionization cross sections are a good alternative if better methods are not available



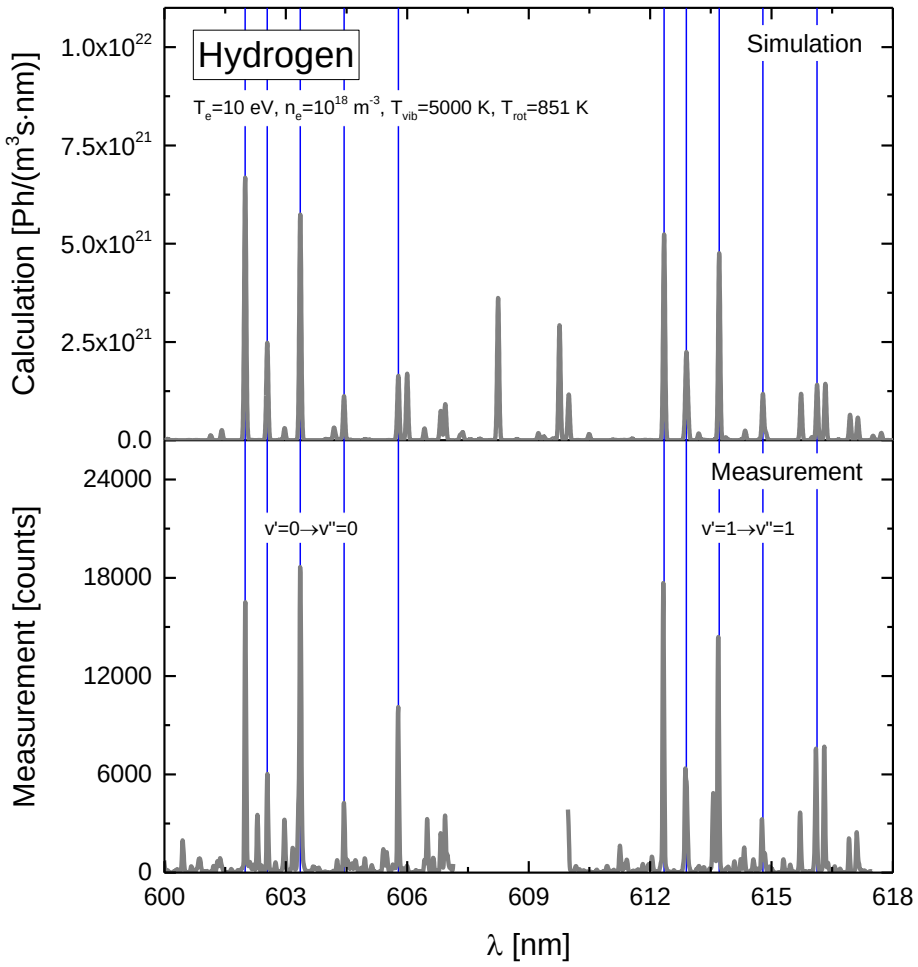

- Outlook: combine Gryzinski with MCCC for determining dissociative ionization cross sections with a high-accuracy.

D. Wünderlich, accepted by At. Data Nucl. Data Tables, 2021

# Ro-vibrationally resolved models.

**Fulcher band ($d^3\Pi_u \rightarrow a^3\Sigma_g^+$), the most intense transition of $H_2$ in the visible range**

- Ro-vibrationally resolved CR model: huge amount of input data needed (Cross sections, transition probabilities).
- First step: ro-vibrationally resolved corona model.
- Position of the lines and general structure well described by the model ✓

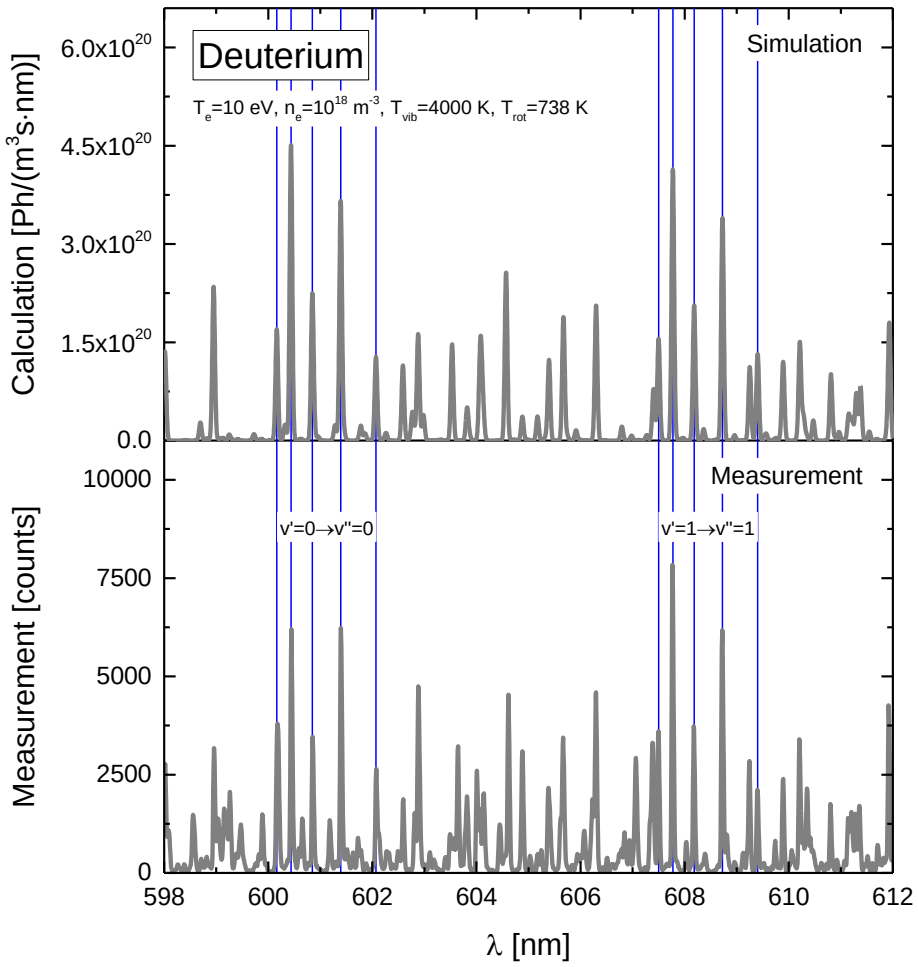



D. Wünderlich, Atoms 4, 2016, 26

# Ro-vibrationally resolved models.

**Fulcher band ($d^3\Pi_u \rightarrow a^3\Sigma_g^+$), the most intense transition of $H_2$ in the visible range**

- Ro-vibrationally resolved CR model: huge amount of input data needed (Cross sections, transition probabilities).
- First step: ro-vibrationally resolved corona model.
- Position of the lines and general structure well described by the model ✓
- Isotope effect hydrogen ↔ deuterium.




D. Wünderlich, Atoms 4, 2016, 26

# Ro-vibrationally resolved models.

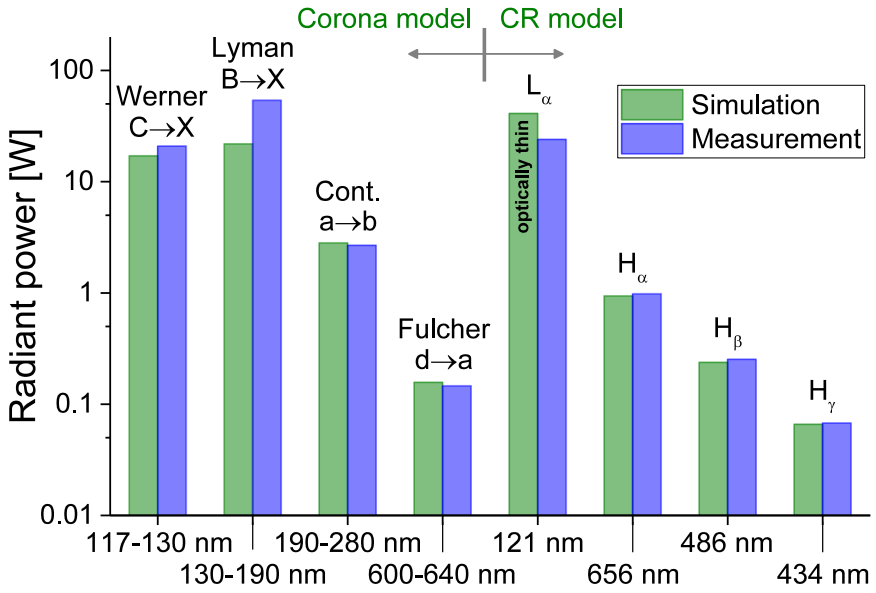
**Fulcher band ($d^3\Pi_u \rightarrow a^3\Sigma_g^+$), the most intense transition of $H_2$ in the visible range**

- Ro-vibrationally resolved CR model: huge amount of input data needed (Cross sections, transition probabilities).
- First step: ro-vibrationally resolved corona model.
- Position of the lines and general structure well described by the model ✓
- Isotope effect hydrogen ↔ deuterium.

**Similar models available also for the resonant Werner ($C^1\Pi_u \rightarrow X^1\Sigma_g$) and Lyman ($B^1\Sigma_u \rightarrow X^1\Sigma_g$) bands**

- Good agreement with measurements at an ICP experiment, f=13.56 MHz, P<600 W, p<10 Pa.
- Photon fluxes ≈ ion fluxes, atomic fluxes much higher

Extension to fully ro-vibrationally resolved CR models now is on the agenda thanks to the MCCC cross sections for $H_2$.




D. Wünderlich, Atoms 4, 2016, 26

# Yacora on the Web: online CR models for He, H and $H_2$.

Yacora on the Web provides online access to selected Yacora CR models

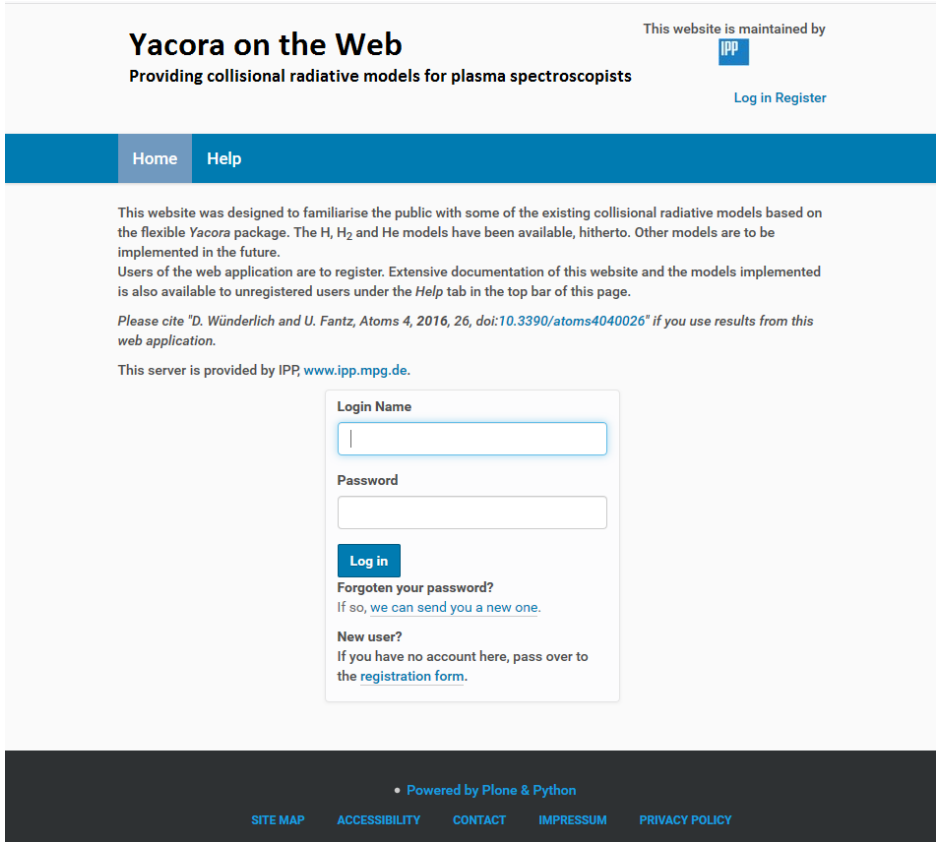
www.yacora.de

Aims and features of Yacora on the Web

- Making public collisional radiative models based on Yacora in a user friendly environment.

- Available up to now: models for H, $H_2$ and He.

- Very simple registration (self registration).
- Extensive documentation available (also for anonymous users).
- Web application based on Plone and Python.




D. Wünderlich, J. Quant. Spectrosc. Radiat. Transfer. 240, 2020, 106695

# Conclusions and ongoing work.

Achieved: significant steps towards comprehensive CR models for the hydrogen molecule in the edge plasma

**Next steps regarding models for $H_2$:**

- Isotope effect
- Ro-vibrationally resolved CR models (huge amount of data!)

} Based on MCCC cross sections

**Still an open point is the connection molecule – atom:**

Cross sections for dissociative recombination, … used in the model are based on simple assumptions.

**And there is still room for improvements regarding the atom!**

- Cross sections for direct excitation had to be “smoothed” over principal quantum numbers.
- Full set of input data for CR models up to high quantum numbers, over broad energy range and from one data source?