MAX-PLANCK-INSTITUT FÜR PLASMAPHYSIK

Vibrationally and ro-vibrationally resolved collisional radiative modelling of molecular hydrogen: current status and outlook

D. Wünderlich, R. Bergmayr, U. Fantz

EUROfusion

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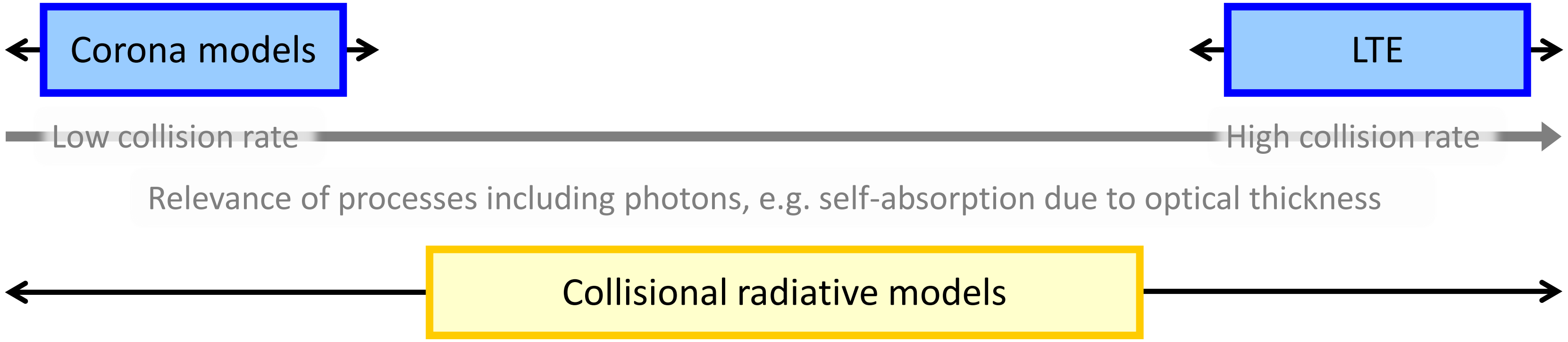
Population modelling

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.

$\Rightarrow$ Needed: extensive data base of reaction probabilities.

$\Rightarrow$ Drastically increased complexity for molecules (vibrational and rotational excitation).

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

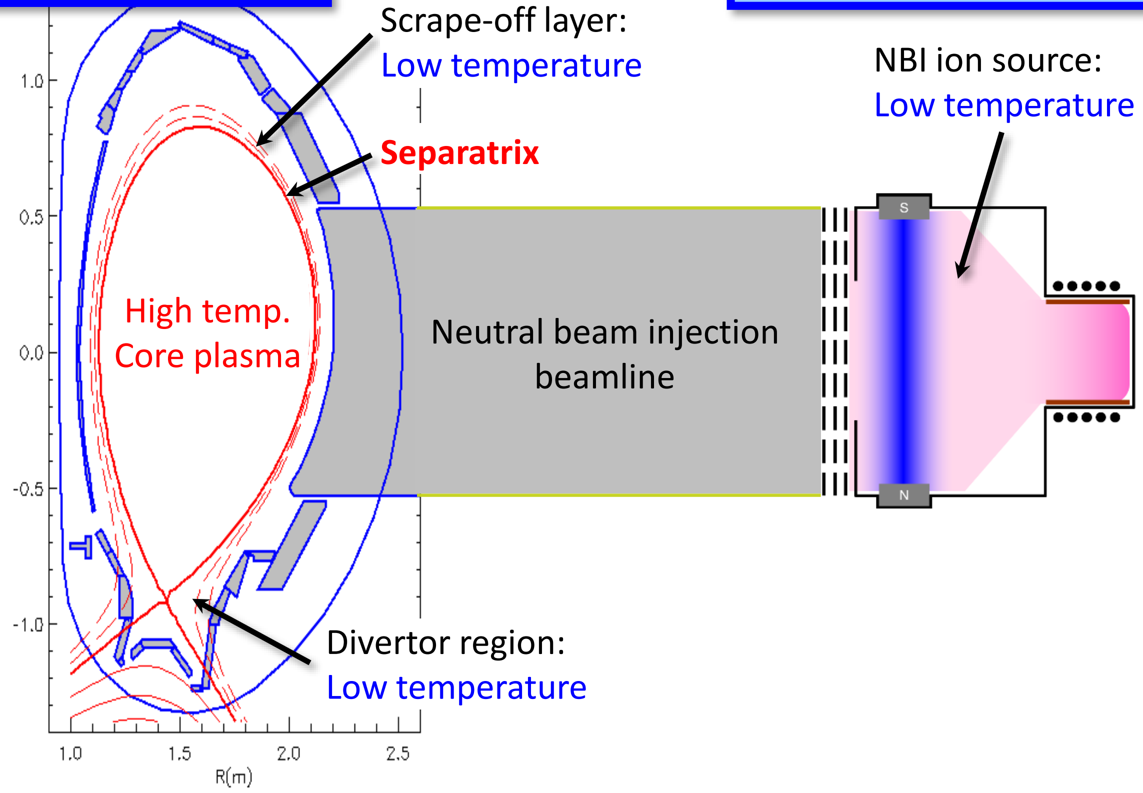
Plasmas for fusion: parameter variations over a wide range

Tokamak plasma edge

T_e=50 eV … below 1 eV
n_e=10^{18} m^{-3} … 10^{21} m^{-3}

NBI ion source plasma

T_e= ≈10 eV … ≈1 eV
n_e=10^{18} m^{-3} … 10^{17} m^{-3}



Both cases:

- Wide parameter range.
- Transition ionizing → recombining plasma.

(Possible) relevance of…

- Different plasma regimes.
- Isotopes of H_2.
- Optical thickness.
- Plasma-wall interaction

Application of population models:

- Interpreting diagnostics results.
- Used in transport codes.

Needed as input: atomic and molecular data (reaction probabilities).

The role of molecules for changing plasma parameters...

… 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

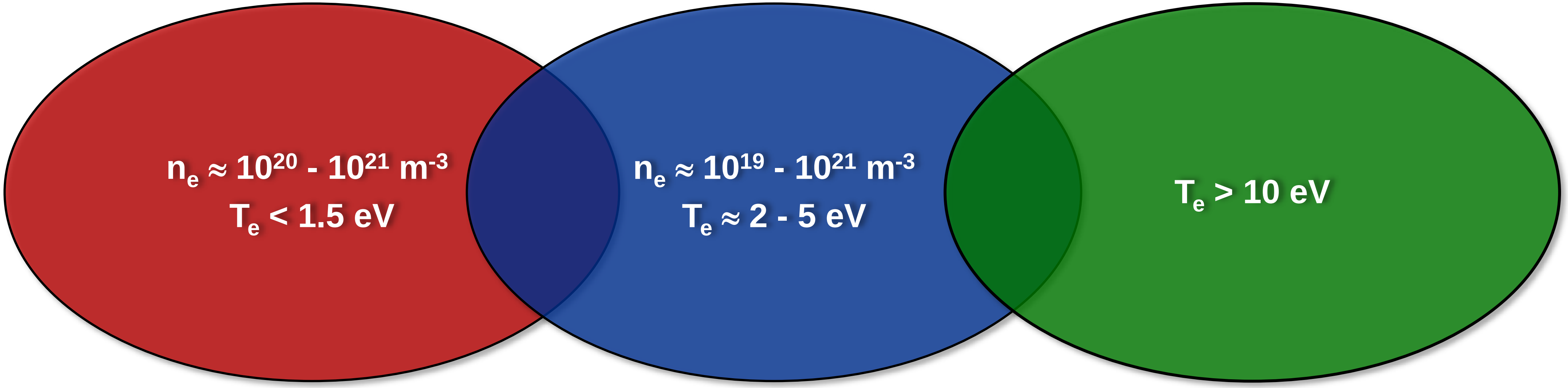
$H + e^- \rightarrow H^+ + 2e^-$ ionization of atoms and

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

Molecular Assisted Recombination MAR

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

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



Yacora: a flexible solver for Collisional Radiative models

Yacora[1] is a flexible (0D)-solver for Collisional Radiative models:

- Used and improved for more than 20 years.
- Almost all available CR models are relevant for application in plasmas used for fusion:

[1]: D. Wünderlich et al, Atoms 4, **2016**, 26

CR model for…		# states	Comment
H_2	Electronic states only	33	Issues with some cross sections, well benchmarked
	Some vibrational states	214	Issues with some cross sections, well benchmarked
	Vib-rot resolved	>626	Extended Corona models for different transitions
H		44	Coupling to H^+, H_2^+, H_3^+, H^-, very well benchmarked
He		19	Very well benchmarked
Ar		16	Well benchmarked
Ar^+		84	Only a collection of input data, not benchmarked
N_2	Electronic states only	6	Includes energy transfer to metastable Ar
C_2	Vibrationally resolved	80	
CH	Vibrationally resolved	29	
Cs		11	Includes MN H^- with Cs^+, well benchmarked

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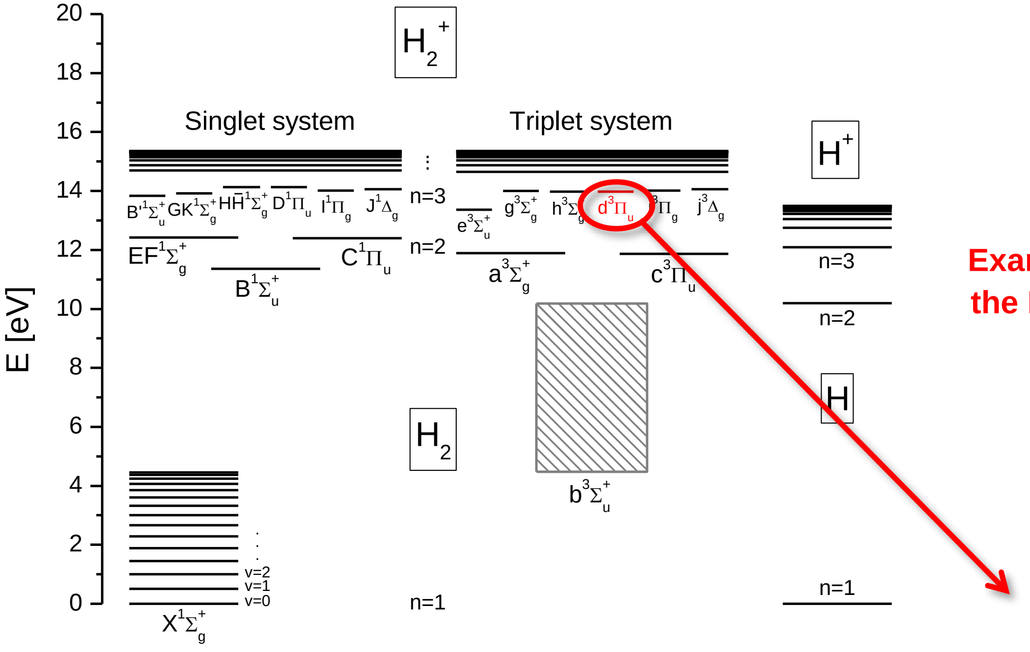
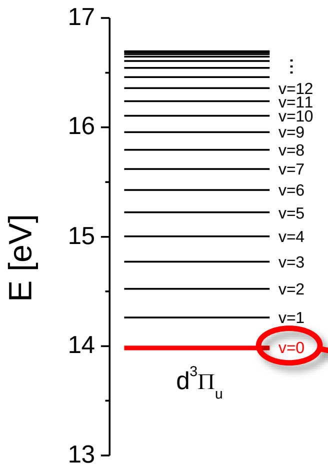
Energy levels within the hydrogen molecule



Electronic excitation

Example: d³ = upper level of the Fulcher band transition.

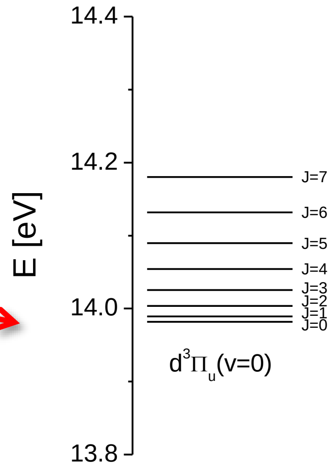


Vibrational excitation

Large number of excited states

Needed:

Probabilities for all reactions interconnecting all excited states



Rotational excitation

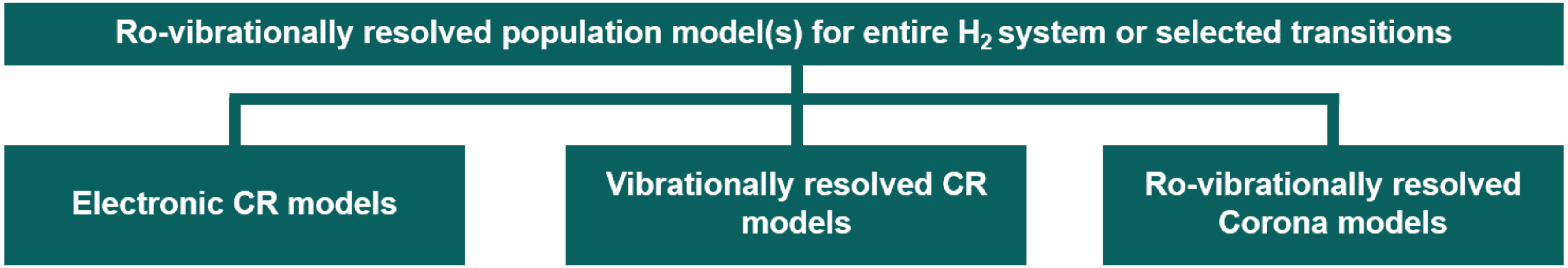
Combined application of different models for H_2

Wish list for a population model for molecular hydrogen:

- Can describe emission bands ro-vibrationally resolved .
- Ability to describe impact of ro-vibrational excitation on reaction kinetics.
- Availability for hydrogen and deuterium over a broad range of plasma regimes.

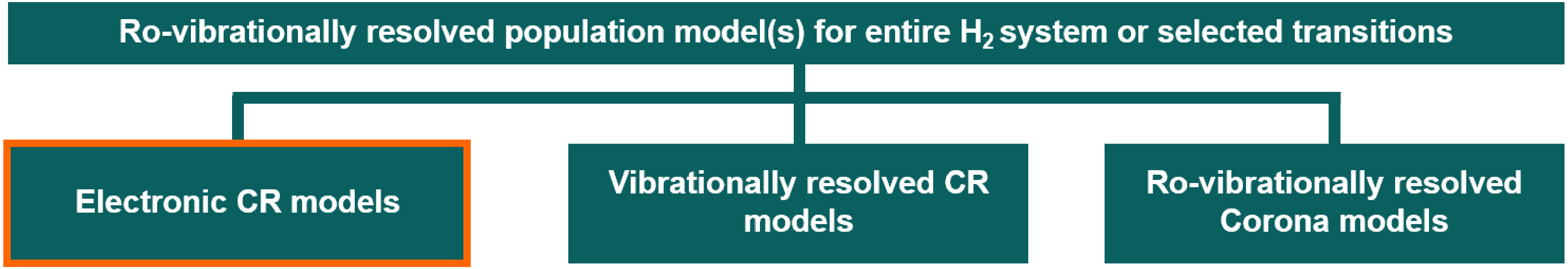
⇒ Ro-vibrationally resolved models needed!

- Available input data base (reaction probabilities) not sufficient for constructing such models.
- Use a stepwise approach with the final aim to construct a ro-vibrationally resolved extended Corona model:



- Effort of transferring the model to D_2, T_2, … increases drastically with the level of detail. For example electronic CR models can be directly applied to the isotopomeres of H_2.

Overview of the presentation



Electronic model: overview

Main aims of the electronic model:

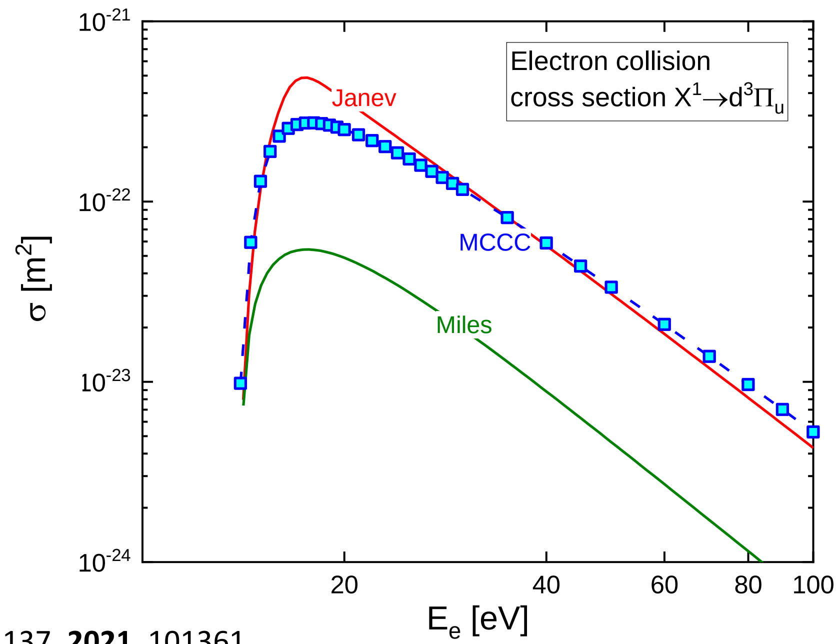
- Up to now, this model is a working horse for evaluating diagnostics results (scaled to the full emission band).
- Check, which reactions have to be included into an extended ro-vibrationally resolved Corona model.
- Validation of the existing reaction probabilities. Check new input data. Validate procedures for creating new data, e.g. by scaling.

Status prior to 2017:

Significant inconsistencies even in the reaction probabilities for the most simple excitation reaction, namely electron collision from the ground state X^1:

- Data collections by [2] and [3]: only a few reactions.
- Calculations by [4]: only a few transitions, but isotope effect.
- Miles[5]: semi empiric cross sections, 1972.
- Janev[6]: summary of measurements and calculations, 2003.

[2]: T. Tabata et al, ADNDT 76, **2000**, 1
[3]: J.S. Yoon et al, J. Phys. Chem. Ref. Data 37, **2008**, 913
[4]: R. Celiberto et al, ADNDT 77, **2001**, 161



[5]: W.T. Miles et al, J. Appl. Phys. 43, **1972**, 678
[6]: R.K. Janev et al, Report JÜL-4105, 2003

Since 2017:

MCCC (Molecular Convergent Close-Coupling), fully quantum-mechanical[7].
Close collaboration with the Group of D. Fursa, Curtin University, Australia.

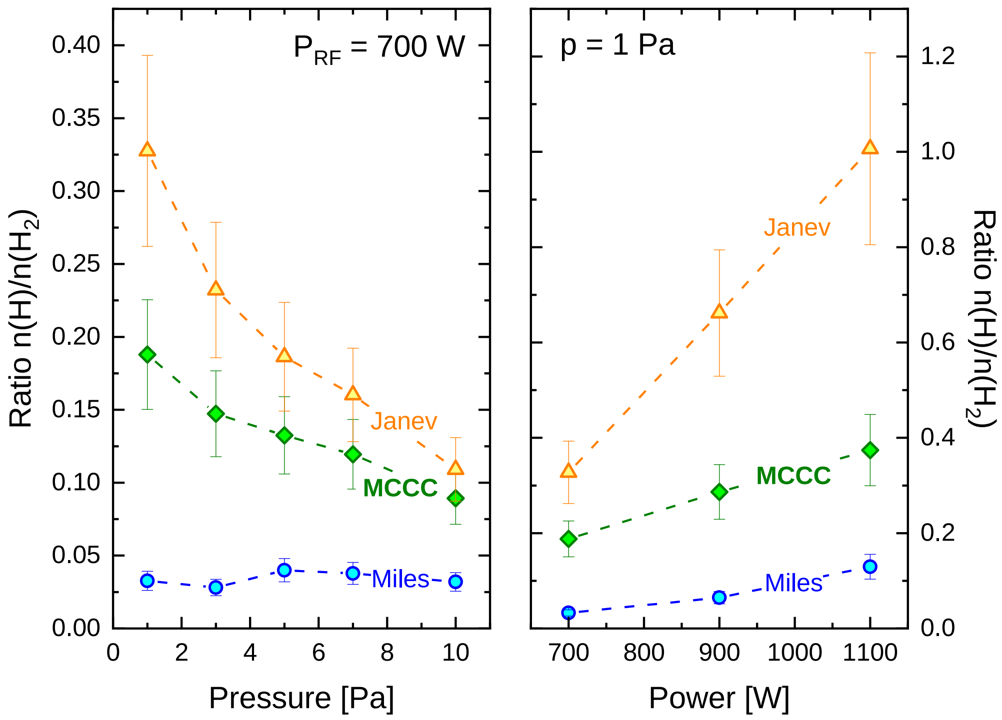
[7]: L. Scarlett et al, ADNDT 137, **2021**, 101361

Electronic model: MCCC cross sections

MCCC cross sections implemented in the electronically resolved Yacora model:

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

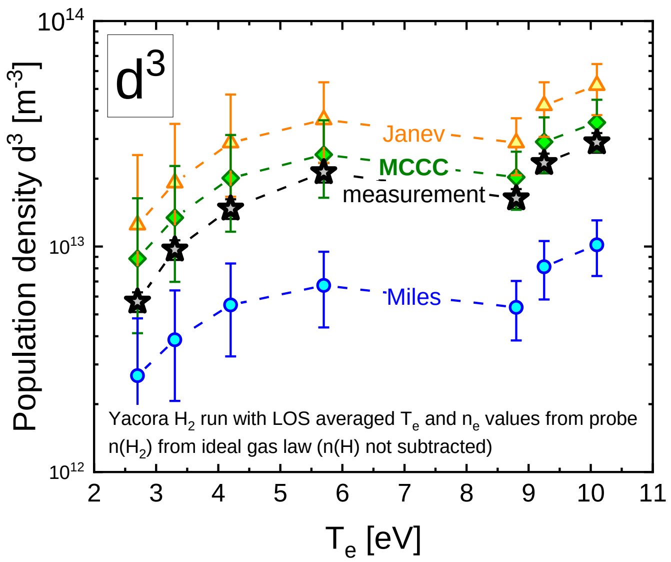
Several excited states: agreement measurement ↔ CR model much better than using previously available cross sections[8].



[8]: D. Wünderlich et al, J. Phys. D 54, **2021**, 115201

Application to plasma diagnostics:

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

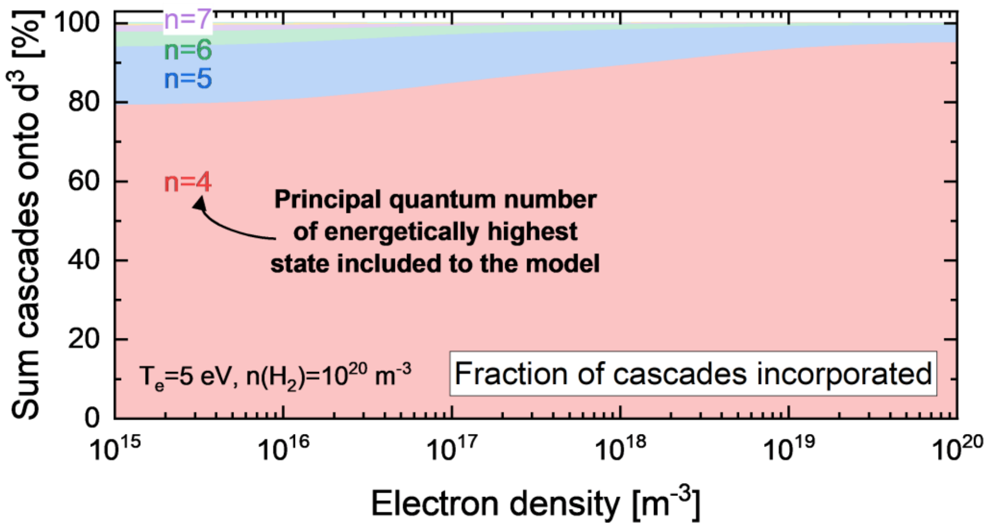


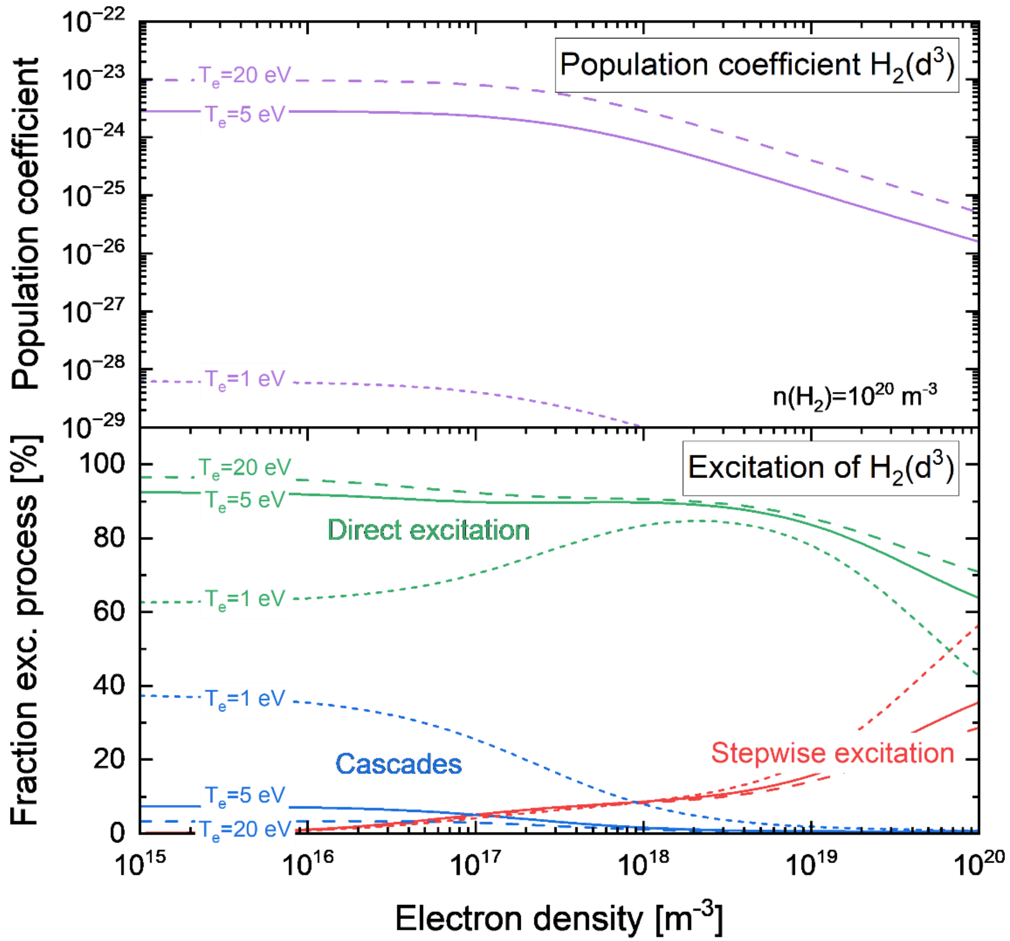
Electronic model: investigations towards the full model(s)

Relevance of different excitation channels:

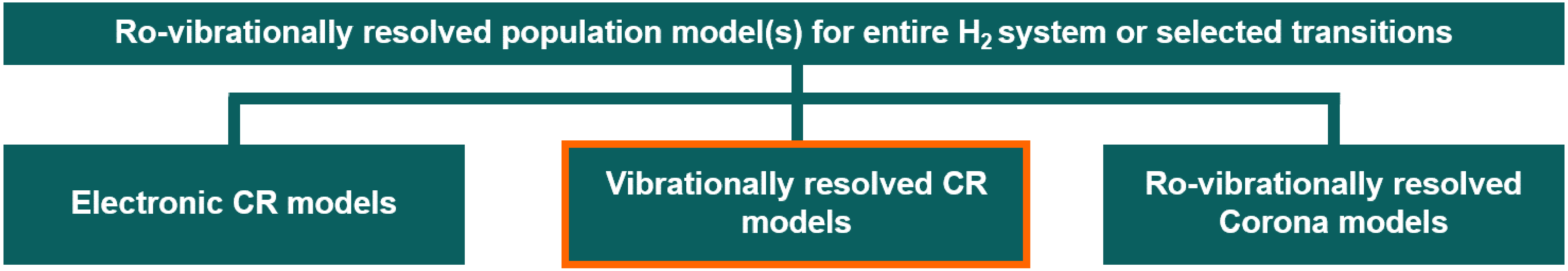
- Can strongly vary during plasma parameter variations, e.g. the onset of plasma detachment with T_e decrease and n_e roll-over.
- Impact of individual states, as example cascades from energetically higher levels.

⇒ An extended ro-vibrationally resolved Corona model may be sufficient for fulfilling the wish list.





Overview of the presentation



Vibrationally resolved model

Main aims of the vibrationally resolved CR model:

- Gain insight in the reaction dynamics inside the manifold of vibrational states of the ground state X^1.
- Test-case for the final ro-vibrationally resolved Corona model(s).

Status prior to 2022[1]:

- The model included a limited set of reactions interconnecting the states $X^1(v)$ and some electronically excited states.
- No self-consistent description of $n(X^1,v)$ possible, T_{vib} used as input parameter for the model.

[1]: D. Wünderlich et al, Atoms 4, **2016**, 26

Since 2022[8]:

- Updated input cross sections for processes (involving X^1) already implemented into the previous model.
- Additional reaction channels identified and added.
- Aim: self-consistent determination of the vibrational ground state distribution.
- Still an issue: processes like vibrational re-distribution at surfaces not included due to missing reaction probabilities.

[8]: R. Bergmayr et al, Eur. Phys. J. D 77, **2023**, 302

Vibrationally resolved model: list of included reactions

Reaction		Prior to 2022	Since 2022
Electron Impact (De-) Excitation (EIE)	e + $H_2(v)$ → e + $H_2(v')$	Buckman 1985	Janev 2015
Electron Impact Dissociation (EID)	e + $H_2(v)$ → e + $H_2(n>1)$ → e + H + H	Celiberto 1999	MCCC
Dissociative Attachment (DA)	e + $H_2(v)$ → H + H^-	Bardsley 1979	Horacek 2004
Charge Exchange (CX)	H^+ + $H_2(v)$ → H + H_2^+	Janev 2008	v=0: Urbain 2013 + Holliday 1971 v>0: Errea 2007, Janev 2003
Non-Dissociative Ionization (NDI)	e + $H_2(v)$ → e + e + H_2^+	Wünderlich 2021	Wünderlich 2021
Radiative Recombination (RR)	e + H_2^+ → $H_2(v)$	Sawada 1995	Sawada 1995
3-body Recombination (C3PR)	e + e + H_2^+ → e + $H_2(v)$	Sawada 1995	Sawada 1995
Transitions via B and C	Electron Impact (De-)Excitation Spontaneous Emission	Celiberto 2001, Janev 2003 Fantz 2006	MCCC Fantz 2006
Dissociative Ionization (DI)	e + $H_2(v)$ → e + e + H + H^+	-	Wünderlich 2021
Proton Impact (De-) Excitation (PIE)	H^+ + $H_2(v)$ → H^+ + $H_2(v')$	-	Krstic 2002 + 2005 ($\Delta v=4$)
Proton Impact Dissociation (PID)	H^+ + $H_2(v)$ → H^+ + H + H H^+ + $H_2(v)$ → H + H^+ + H	-	Janev 2003
H- Associative Detachment (H-AD)	H^- + H → $H_2(v')$ + e	-	Cizek 1998
Hydrogen Atom Impact Dissociation (HAID)	H + $H_2(v)$ → H + 2 H	-	Esposito 1999 rates
Hydrogen Molecule Impact Excitation (HMIE)	VT: $H_2(v)$ + $H_2(w)$ → $H_2(v\pm1)$ + $H_2(w)$ VV: $H_2(v)$ + $H_2(w+1)$ → $H_2(v+1)$ + $H_2(w)$	-	Matveyev 1995 rates
H_3^+ Dissociative Recombination (H3+DR)	e + H_3^+ → H + $H_2(v')$	-	Janev 2015 + Strasser 2001
Hydrogen Atom Impact (De-) Excitation (HAIE)	H + $H_2(v=0)$ → H + $H_2(v'>0)$	-	Janev 2003

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H- Associative Detachment (H-AD)	$H^- + H \rightarrow H_2(v') + e$	-	Cizek 1998
Hydrogen Atom Impact Dissociation (HAID)	$H + H_2(v) \rightarrow H + 2\,H$	-	Esposito 1999 rates
Hydrogen Molecule Impact Excitation (HMIE)	VT: $H_2(v) + H_2(w) \rightarrow H_2(v\pm1) + H_2(w)$ VV: $H_2(v) + H_2(w+1) \rightarrow H_2(v+1) + H_2(w)$	-	Matveyev 1995 rates
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Hydrogen Atom Impact (De-) Excitation (HAIE)	$H + H_2(v=0) \rightarrow H + H_2(v'>0)$	-	Janev 2003

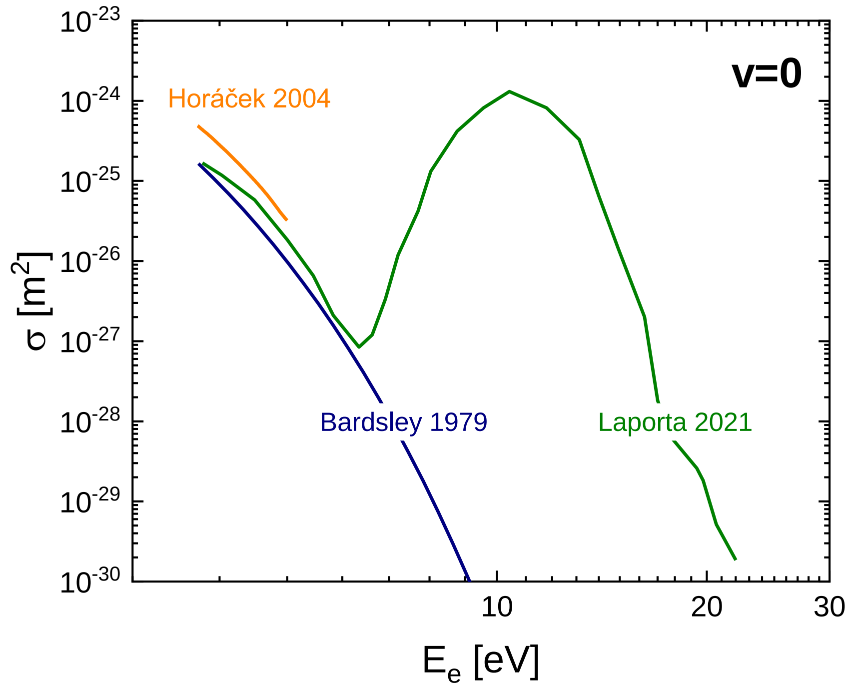
Vibrationally resolved model: data needs

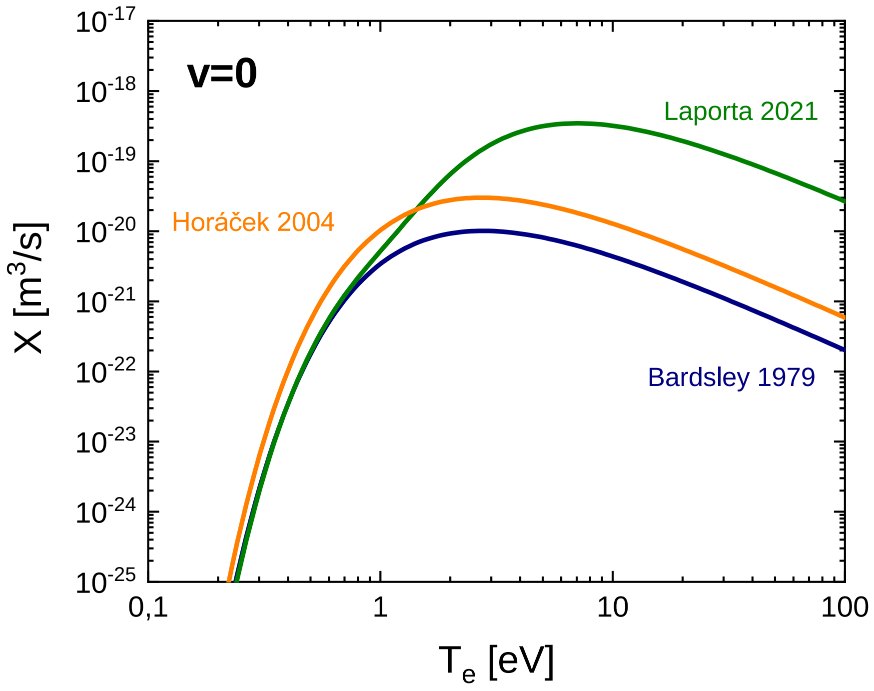
The dissociative attachment process is a good example:

$$e^- + H_2(v) \rightarrow H_2^- \rightarrow H(1s) + H^-$$

- Several reaction channels possible:
  via the resonances $H_2^-(X^2\Sigma_u^+)$, $H_2^-(B^2\Sigma_g^+)$ and $H_2^-(C^2\Sigma_g^+)$.
- Previously used data[9] included only the channel via $H_2^-(X^2\Sigma_u^+)$.
- The most recent cross sections[10] consider the other resonances. But available only for a few $H_2(v)$.
- Use instead cross sections from[11]: only for the $H_2^-(X^2\Sigma_u^+)$ resonance but available for $H_2(v=0...13)$.

[9]: J. N. Bardsley and J. M. Wadehra, Phys. Rev. A 20, **1979**, 1398
[10]: V. Laporta et al, Plasma Phys. Contr. Fus. 63, **2021**, 085006
[11]: J. Horáček et. al, Phys. Rev. A 70, **2004**, 052712





Overview of the presentation

Ro-vibrationally resolved population model(s) for entire H_2 system or selected transitions

- **Electronic CR models**
- **Vibrationally resolved CR models**
- **Ro-vibrationally resolved Corona models**

Ro-vibrational model: overview

Main aims of the ro-vibrationally resolved Corona model:

- Planned to be an integral part of evaluation diagnostics results over a broad range of plasma regimes.
- Model results can be directly compared with diagnostics results; no scaling needed.
- Enables diagnostics access to parameters like T_{gas} and T_{vib} than can be of high relevance for the reaction kinetics.

Status prior to 2022:

- Extended version of vibrationally resolved models for selected transitions, most prominently the Fulcher band.

Since 2022:

- Critical check of the input data, update to (vibrationally resolved) MCCC cross sections.
- Treatment of the non-diagonal rotational transitions by a simple scaling factor[12].
  Planned use of ro-vibrationally resolved cross sections when available (collaboration Group of D. Fursa).
- Extensive benchmark to different plasma experiments.

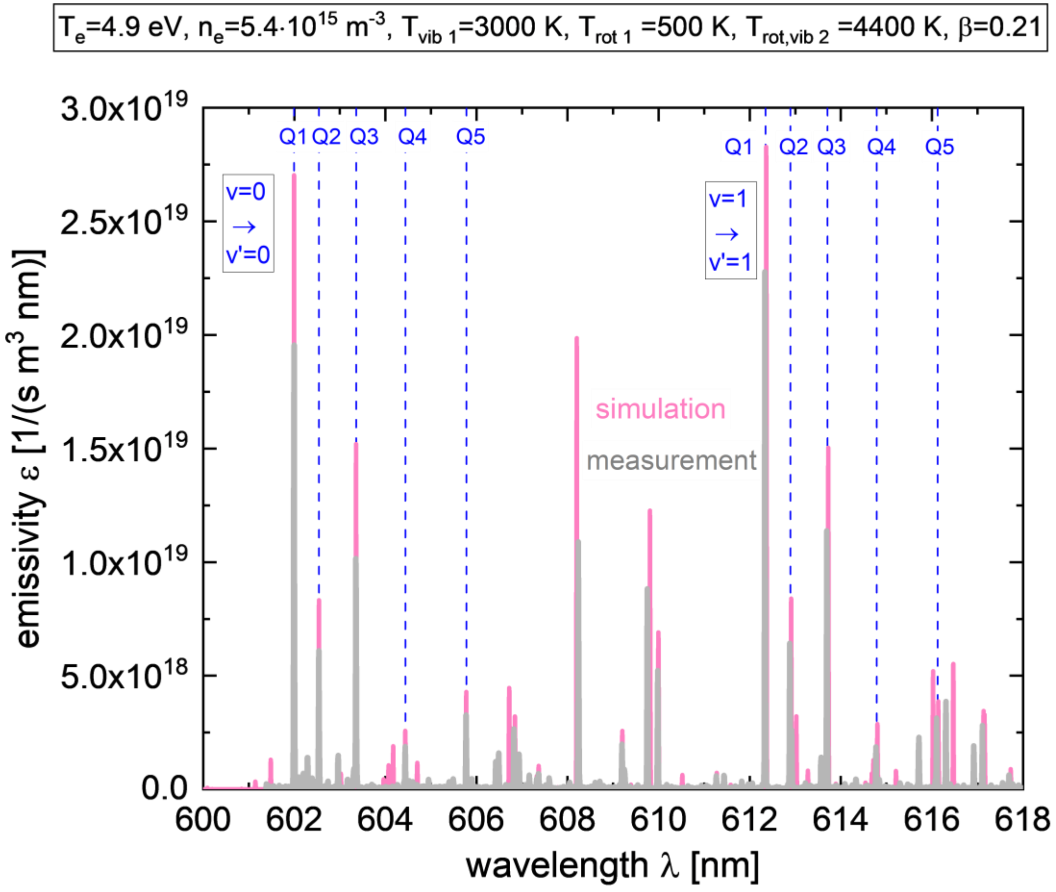
[12]: B. Xiao et al, *Plasma Phys. Control. Fusion* 64, **2004**, 653

Ro-vibrational model: results

Up to now pure Corona models (e.g. Fulcher band)…

- Several successful benchmarks to the experiment.
- Example: Low-power plasma at the negative ion source test facility BATMAN Upgrade

Simulation predicts absolute emissivity ±20-40 % (previous model: ±140-180 %).



Next aim: broaden the set of available test cases.
Extremely useful would be: Fulcher spectra for different divertor regimes (attachment/detachment) & known plasma parameters.

Data needs: a wish list, part I

Significant progress made but still a lot of data missing. Examples are:

- **Dissociative electron attachment** to (ro-)vibrationally and electronically excited states of H_2.
  Vibrationally: full set for D_2, just a few v for H_2. Electronically: only rate constants for n=2 and n>2 states.
- **Charge exchange** of ro-vibrationally and electronically excited states of H_2 with H^+.
  Vibrationally: several sets which have to be combined. Electronically: Simple estimation for rate constant and cross sections for n>3.
- **H impact excitation** of $H_2(X^1,v) \rightarrow H_2(X^1,v')$.
  Cross sections available only for v=0.
- **H impact dissociation** of $H_2(X^1,v)$.
  Only rate coefficients available.
- **Dissociative recombination** of H_3^+.
  Only total cross section and branching ratios available.
- **Further reactions involving H_2^+ and H_3^+ influencing $H_2(X^1,v)$.**
- **H_2 impact excitation** of $H_2(X^1,v) \rightarrow H_2(X^1,v')$.
  Only rate coefficients available for $\Delta v = \pm 1$.
- **H^- electron detachment** $H^- + H_2(v) \rightarrow H_2(v') + H + e^-$.
  Cross sections only for v=0 without final state resolution.
- **Lifetimes** of (ro-)vibrationally and electronically excited states of $H_2 \Rightarrow$ predissociation, autoionisation, quenching, …
- Probabilities for typical **wall processes** $H_2(X^1,v) \rightarrow H_2(X^1,v')$, under the involvement of e.g. tungsten, beryllium, …

Describing the plasma edge: input data for D_2 essential.
Ideally also T_2, but scaling may be possible,
following the data from H_2 and D_2.

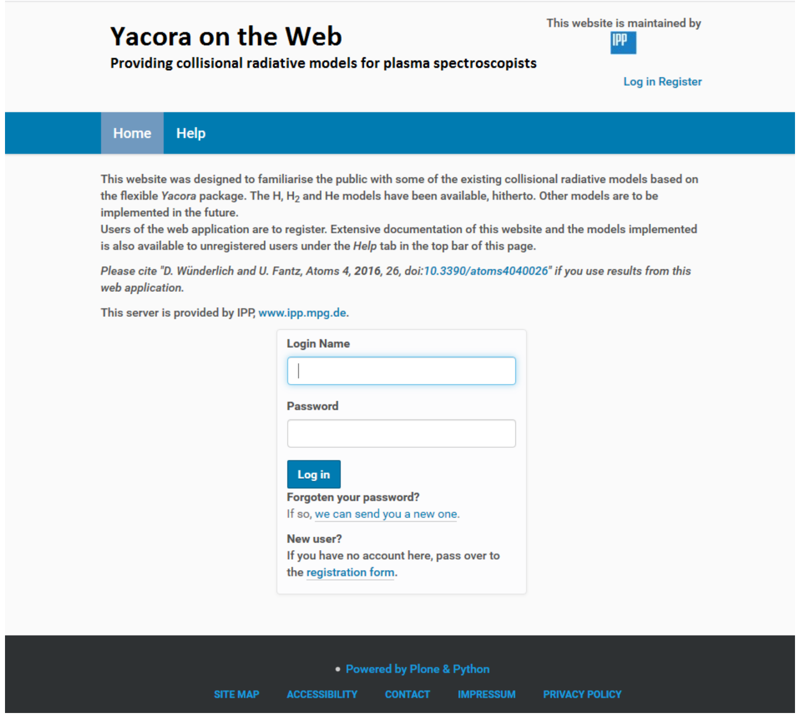
Conclusions & Yacora on the Web

CR models based on Yacora …

- Well benchmarked models for several atomic and molecular species.
- Applied within numerous collaborations worldwide.
- Steady improvement of the used input data.
  Example: MCCC cross sections for H_2, solving a very persistent issue.

www.yacora.de

- Online access to selected Yacora CR models
- Available up to now: models for H, H_2 and He.



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

Data needs: a wish list, part II

… and now to something completely different …

… related to an ECR charge breeder diagnostics. Used are cross sections for:

$e^- + K^{4+} \rightarrow 2e^- + K^{5+}$,
$e^- + K^{5+} \rightarrow 2e^- + K^{6+}$,

... all charge states in between …

$e^- + K^{12+} \rightarrow 2e^- + K^{13+}$

These cross sections are available but some of these have reported uncertainties of up to 200 % which directly impacts the sensitivity of the diagnostics.