Characterization of wall response to plasma fluctuations in tokamak

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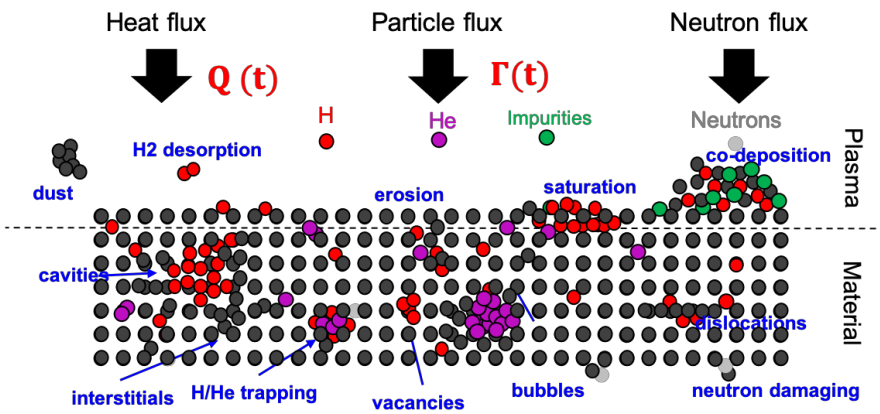
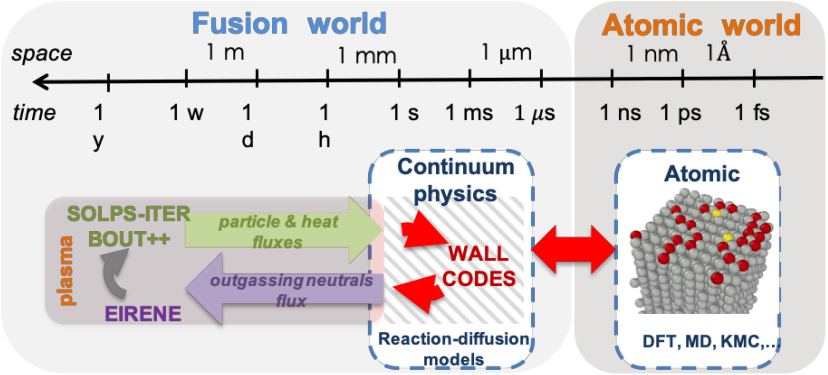
GENERAL ATOMICS

Plasma boundary conditions in SOL provided by dynamic hydrogen recycling from PFCs governed by complex and multi-faceted material processes

- Hydrogen retention and recycling in PFCs governed by **various complex and multifaceted atomic processes** in material bulk and on material surface
- Various plasma regimes in Tokamak scrape-off layer with transient features (fluctuations, ELMs,…): PFCs exposed to a **broad range of plasma conditions fluctuating in time**
- Example of synergetic effects between plasma and H recycling: Re-healing of giant ELMs determined by wall outgassing, which controls giant ELMs frequency [3]
- Integrated modeling of PMI using macroscopic plasma models and atomic wall models **virtually impossible** due to the multiscale nature of PMI



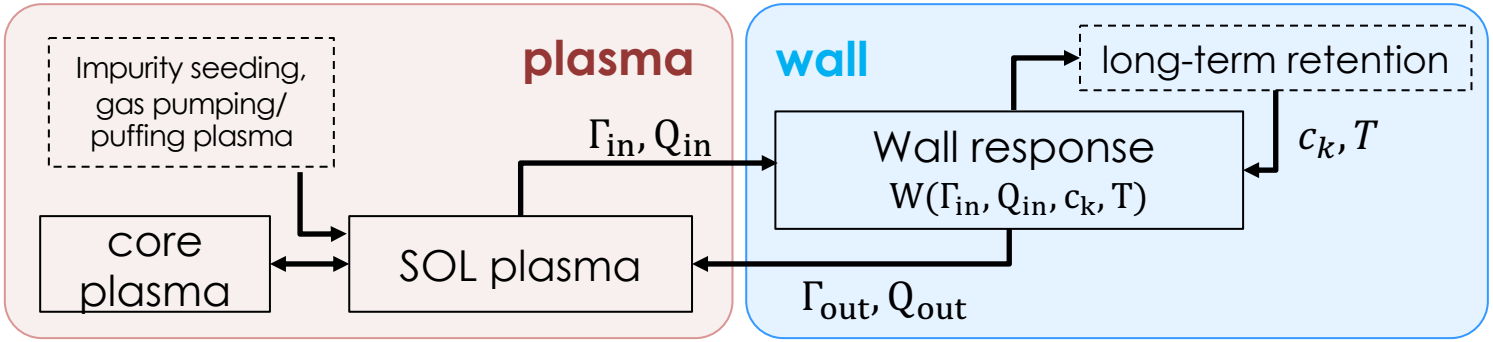


Reduced continuum models of hydrogen retention and recycling (e.g. FACE[1], Xolotl[2],…) must be developed to describe dynamic wall recycling and **provide <u>time-dependent</u> boundary conditions for plasma solvers** (SOLPS, BOUT++, …)

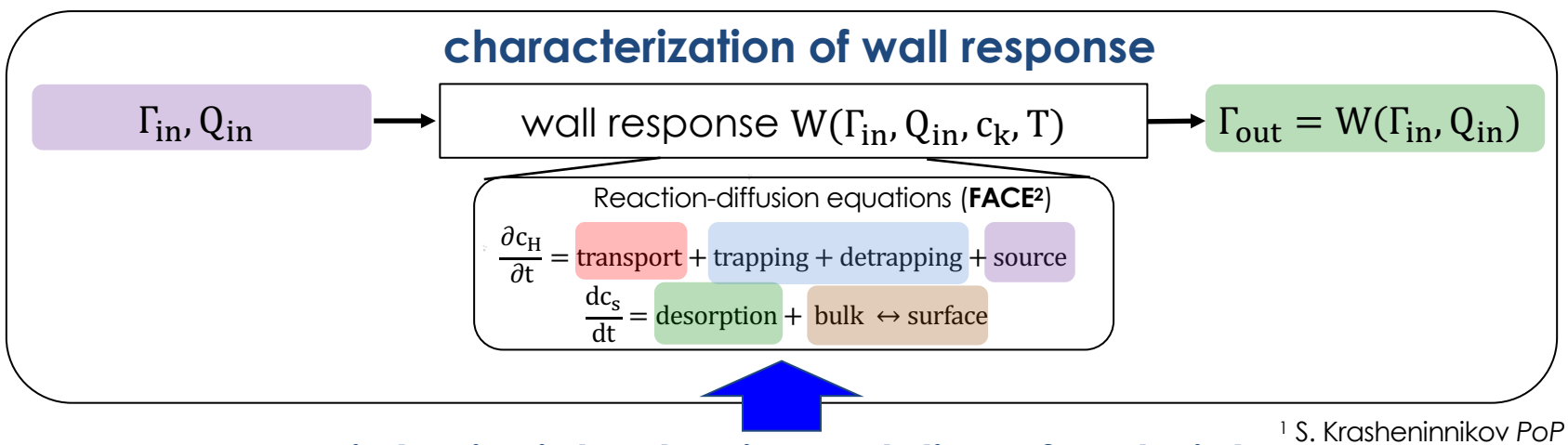
[1] R. Smirnov *Fusion Sc. Tech.* 2017 [2] S. Blondel *FTS* 2017 [3] A. Pigarov *JNM* 2014

Characterization of wall response using reaction-diffusion models in conjunction with first-principle atomic modeling

- Splitting between plasma models and wall models possible when considering linear wall & plasma responses[1]



- Fully coupling plasma and wall models however requires to develop an advanced numerical framework → **we focus solely in this work on the analysis of the wall response**
- **Atomic and macroscopic modeling of H transport, reaction and desorption processes** must be combined to describe hydrogen recycling from PFCs (wall response)



[1] S. Krasheninnikov *PoP* 2018
[2] R. Smirnov *FST* 2017

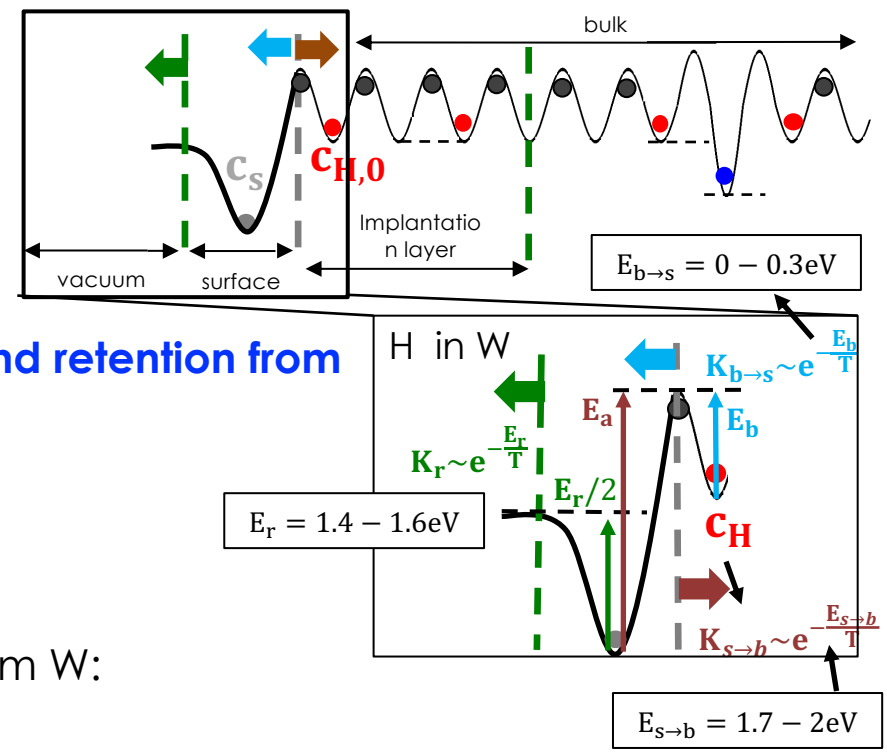
Modeling of tungsten wall response in ITER relevant conditions

- Continuum models of hydrogen retention and recycling - and thus of wall response – include **hydrogen transport, desorption and trapping in material in various regimes**

Reaction-diffusion equations

bulk:
$$\frac{\partial c_H}{\partial T} = D\frac{\partial c_H}{\partial x^2} - \frac{\partial c_{H,trap}}{\partial T} + \frac{\Gamma_{H,in}}{\lambda_{imp}}$$
$$\frac{\partial c_{c_{H,trap}}}{\partial T} = \nu_{tr}\, c_H \left(c_{trap} - c_{c_{H,trap}}\right) - \nu_{dt}\, c_{c_{H,trap}}$$

surface:
$$\frac{\partial c_s}{\partial T} = -K_r c_s^{\alpha} + K_{b\to s}\, c_{H,0} - K_{s\to b}\, c_s$$



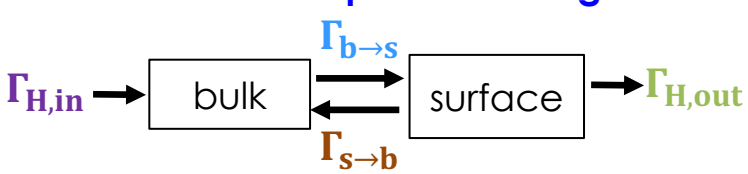
- **Focus on wall response induced H recycling and retention from tungsten in ITER relevant conditions:**
  - W divertor **temperature from 500K to 1400K**
  - W divertor exposed to DT flux $\Gamma_{H,in} \approx 10^{24}\ m^{-2}\ s^{-1}$
  - High recycling regime $\Gamma_{H,in} \approx \Gamma_{H,out}$
- H molecular recombination and desorption from W:
  - Second order kinetic: $\alpha = 2$
  - Large recombination and reabsorption barrier $E_r \sim 1.4eV$ [1], $E_{s\to b} = 2eV$ [2]
- H transport and trapping in W:
  - Diffusion $E_D \sim 0.2eV$ [3] and vacancy and defects with $E_{dt} \sim 0.85 - 2eV$ [4]
  - Implantation of He in W can strongly affect H transport and recycling in W [5]

[1] P. Tamm *Journal of Chem. Phys.* 1969
[2] D. Johnson *J. Mater. Res.* 2010
[3] N.Fernandez *Acta Materialia* 2015
[4] E. Hodille *Nuclear Fusion* 2017
[5] M.J. Baldwin *Nuclear Fusion* 2017

Influence of surface processes on hydrogen outgassing from W remains uncertain in ITER relevant conditions

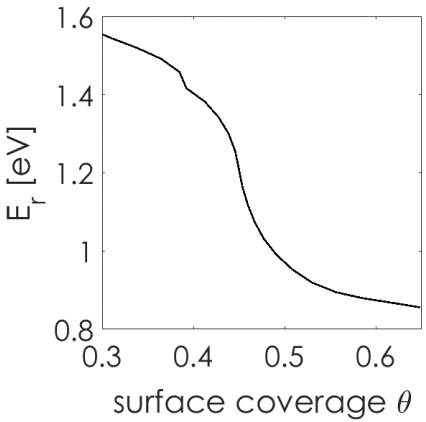
- Physical parameters of continuum models can be empirically constrained with dedicated controlled experiments (TDS, permeation,…) but large uncertainties might remain. For instance, some H surface processes on W still not well understood:
  - H_2 dissociation rate on W independent of H pressure [1]
  - Decrease of activation energy of H desorption when W surface is saturated with H [2]
- Such processes may be important in fusion relevant outgassing regimes!
- Within this framework, dynamic wall outgassing response is analyzed here using

① **R-D models (FACE) to characterization of bulk and surface responses in relevant ITER operational regime**

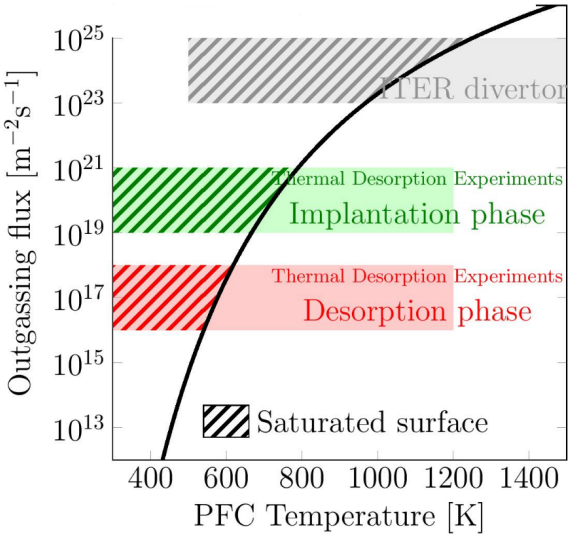


② **First-principle atomic modeling (LAMMPS) of H desorption from W material**

Activation energy of H desorption from W (experiment)[2]



Maximum H desorption flux from unsaturated W surface



[1] W. Zheng *Surface Science* 2006 [2] P. Alnot, *Surface Science*1989

Bulk outgassing response to fluctuations of plasma particle flux by diffusion of free hydrogen in the bulk is equivalent to a low-pass filter

- Outgassing response to **fluctuations of incoming particle flux $\Gamma_{H,in} = \bar{\Gamma}_{H,in} + \Delta\Gamma_{H,in} \sin \omega t$** from bulk material at fixed material temperature T <u>without traps</u> (free H only)

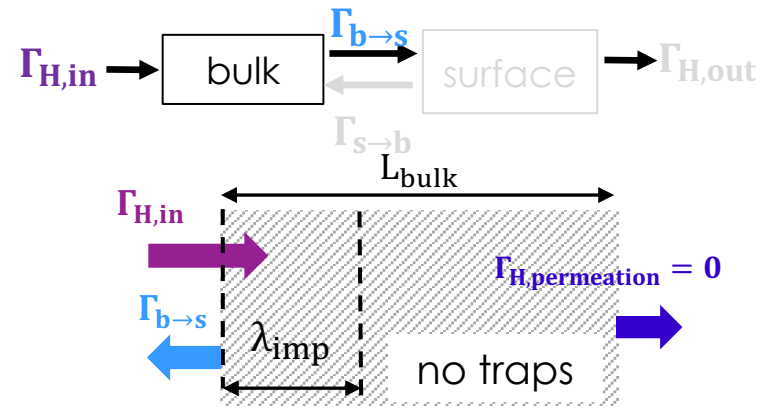
- Hydrogen release from bulk governed by three time scales: $\tau_D = \frac{\lambda_{imp}^2}{D}$; $\tau_{b\to s} = \frac{\lambda_{imp}}{K_{b\to s}}$; $\tau_{bulk} = \frac{L_{bulk}^2}{D}$

- Hydrogen release bulk onto surface assumed to be not limited by surface processes $\left(\frac{\tau_{b\to s}}{\tau_D} \ll 1\right)$

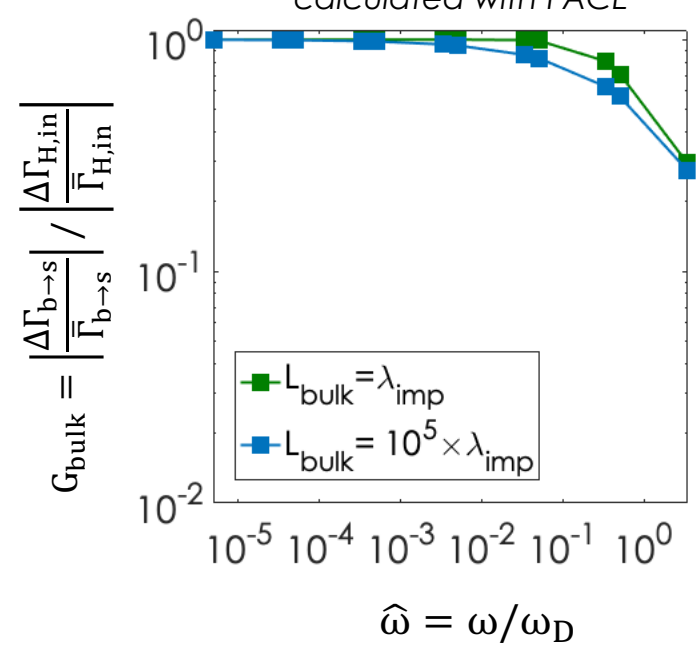
- **Bulk response to $\Gamma_{H,in}$ fluctuations ~ low-pass filter with $\omega_{cutoff} = \omega_D = \frac{D}{\lambda_{imp}^2}$**

- **$\omega_D > 1\text{MHz}$ for $400\text{K} < T$ with $\lambda_{imp} \sim 50\text{nm}$**

- Permeation barrier in material (e.g. formation of sub-surface He bubbles) may slightly affect bulk response to $\Gamma_{H,in}$ fluctuations

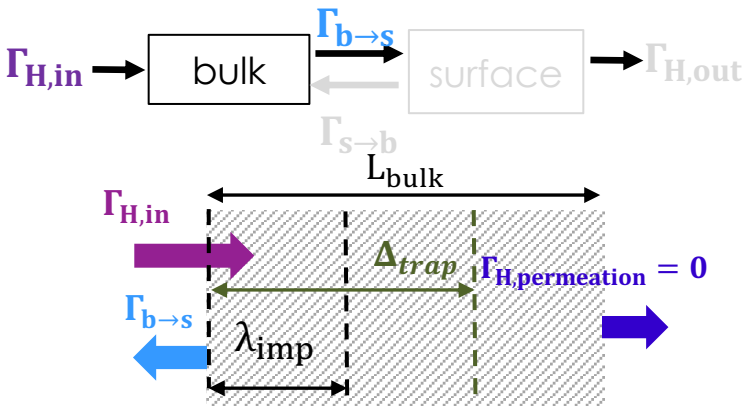


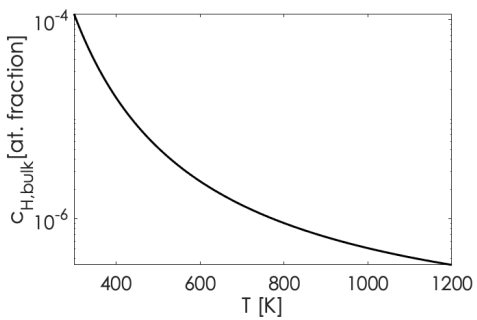
Magnitude of bulk response calculated with FACE



Bulk outgassing response to fluctuations of plasma particle flux weakly affected by the presence of traps in material

- Outgassing response to **fluctuations of incoming particle flux $\Gamma_{H,in} = \bar{\Gamma}_{H,in} + \Delta\Gamma_{H,in} \sin \omega t$** from bulk material at fixed material temperature T <u>with traps</u>
- Solute H concentration $c_{H,bulk} \sim \frac{\Gamma_{in}\lambda_{imp}}{D}$
- Effects of trapped hydrogen on dynamic outgassing only significant when trap concentration is large $c_{trap} > c_{H,bulk}$



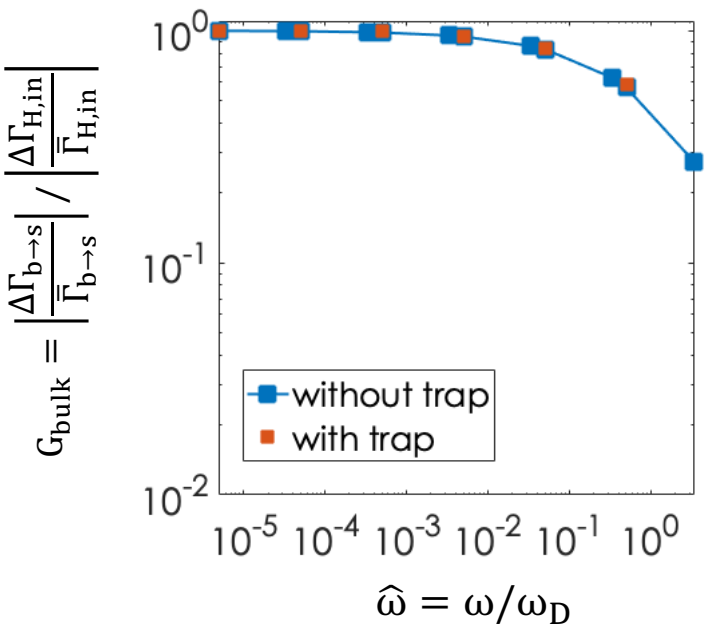


- However, large trap concentration leads to fast hydrogen trapping ($\nu_{tr} c_{trap} \gg \nu_{dt}$) at T<1500K

⇒ empty traps concentrations small compared to solute hydrogen concentration in material

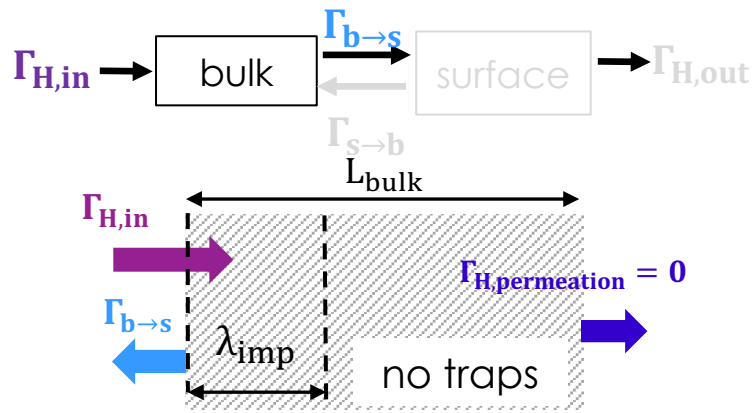
⟹ **Outgassing response to $\Gamma_{H,in}$ fluctuations from bulk weakly affected by hydrogen trapping in material bulk**

Magnitude of bulk response calculated with FACE



Bulk outgassing response to fluctuations of material temperature by diffusion of free hydrogen in the bulk is equivalent to a high-pass filter

- Outgassing response to **fluctuations of material temperature $T = \bar{T} + \Delta T \sin \omega t$** from bulk material at fixed incoming particle flux $\Gamma_{H,in}$ without traps (free H only)

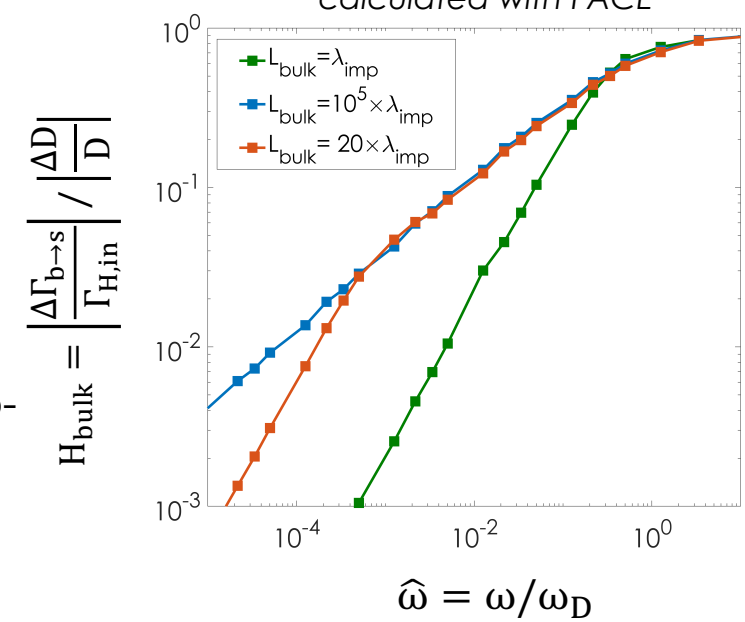


- Hydrogen release bulk onto surface assumed to be not limited by surface processes $\left(\frac{\tau_{b\to s}}{\tau_D} \ll 1\right)$

- **Bulk response to T fluctuations ~ high-pass filter with $\omega_{cutoff} = \omega_D = \frac{D}{\lambda_{imp}^2}$ and $\left|\frac{\Delta\Gamma_{b\to s}}{\Gamma_{H,in}}\right| \propto \left|\frac{\Delta D}{D}\right| \approx \frac{E_D}{T}\frac{\Delta T}{T}$**
  - **$\omega_D > 1\text{MHz}$ for $400\text{K} < T$ with $\lambda_{imp} \sim 50\text{nm}$**
  - **Bulk response → 0 at high temperature**

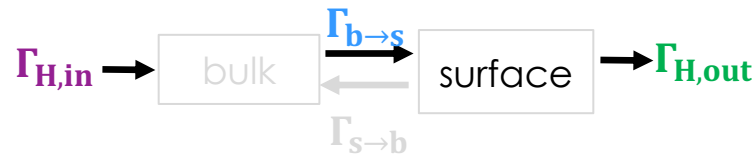
- Permeation barrier in material (e.g. formation of sub-surface He bubbles) may strongly affect bulk response to material temperature fluctuations

Magnitude of bulk response calculated with FACE



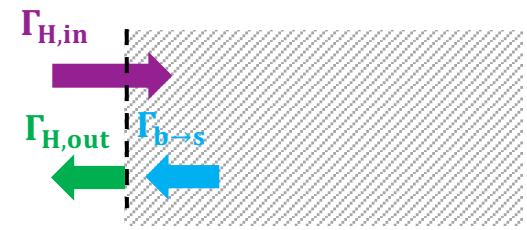
Surface outgassing response to fluctuations of plasma particle flux is equivalent to a low-pass filter and is affected by surface saturation with hydrogen

- Outgassing response to **fluctuations of incoming particle flux** $\Gamma_{H,in} = \bar{\Gamma}_{H,in} + \Delta\Gamma_{H,in} \sin \omega t$ from material surface at fixed material temperature T

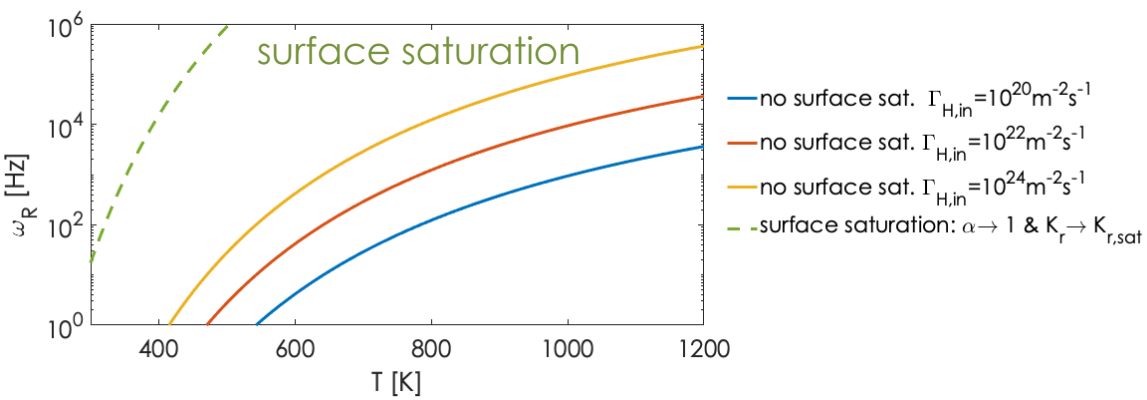


- H surface concentration described by

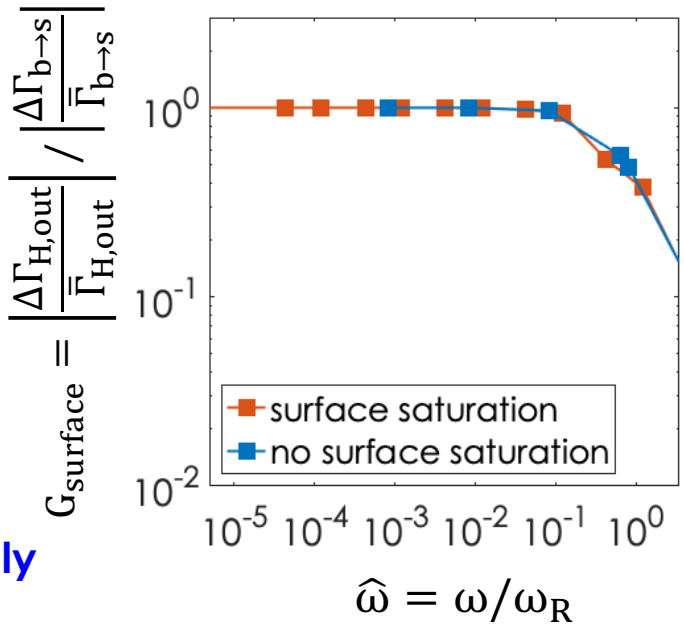
$$\frac{\partial c_s}{\partial t} = -\underbrace{K_{r,0} e^{-\frac{E_r}{T}} (c_s)^{\alpha}}_{\Gamma_{H,out}} + \Gamma_{b\to s}\,, \quad \begin{cases} \alpha = 2 & \text{(non-saturated surf.)} \\ \alpha \to 1 & \text{(saturated surf.)} \end{cases}$$



- **Surface (linear) response to $\Gamma_{H,in}$ fluctuations ~ low-pass filter with $\omega_{cutoff} = \omega_R = K_r^{\alpha^{-1}} (\Gamma_{b\to s})^{1-\alpha^{-1}}$**



Magnitude of surface response calculated with FACE



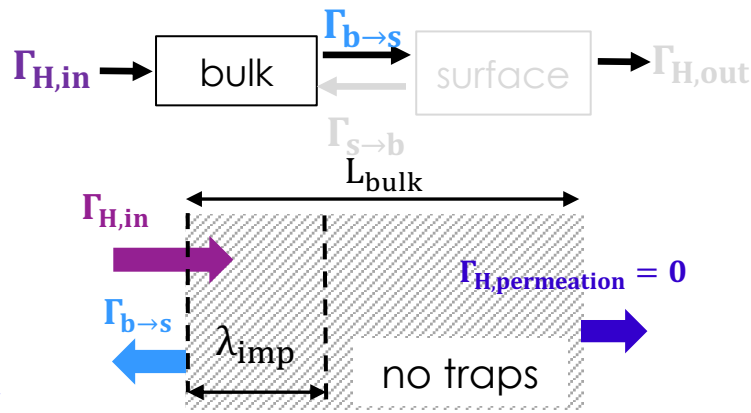
- **Saturation of surface ($E_r = 1.4\text{eV} \to 1\text{eV}$) strongly modify characteristic frequency of surface response**

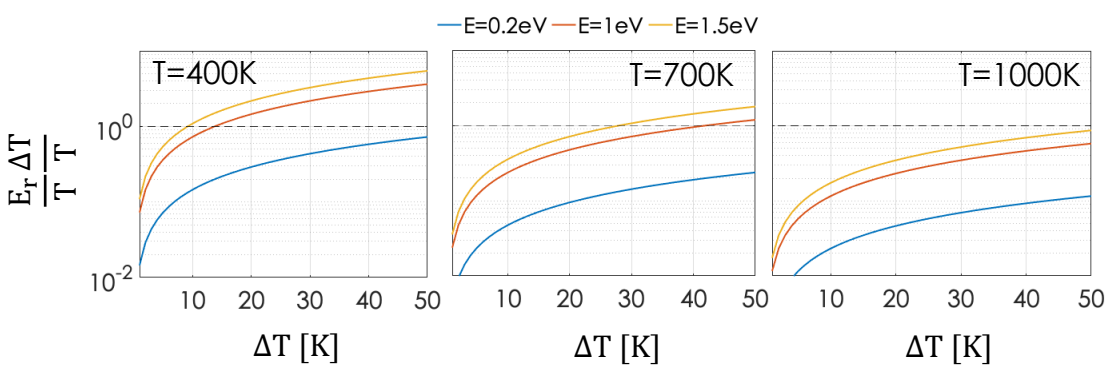
Surface outgassing response to fluctuations of material temperature is equivalent to a high-pass filter and may be large when surface is not saturated

- Outgassing response to **fluctuations of material temperature** $\mathbf{T = \overline{T} + \Delta T \sin \omega t}$ from surface material at fixed incoming particle flux $(\Gamma_{b\to s} = \Gamma_{H,in})$
- H surface concentration described by

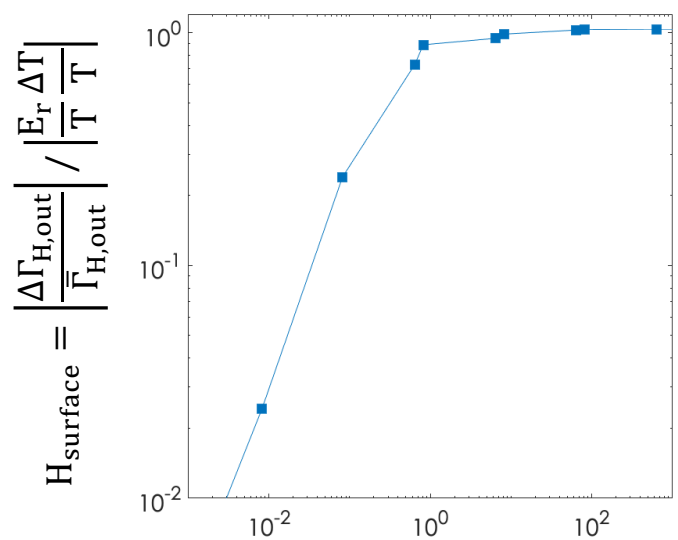
$$\frac{\partial c_s}{\partial t} = -K_{r,0} e^{-\frac{E_r}{T}} (c_s)^{\alpha} + \Gamma_{b\to s}$$

- **Surface response to T fluctuations ~ high-pass filter with** $\omega_{cutoff} = \omega_R = K_r^{\alpha^{-1}} (\Gamma_{b\to s})^{1-\alpha^{-1}}$ **and** $\left|\frac{\Delta\Gamma_{H,out}}{\overline{\Gamma}_{H,out}}\right| \propto \frac{E_r}{T}\frac{\Delta T}{T}$





Magnitude of surface response calculated with FACE



$\hat{\omega} = \omega/\omega_R$

- **Small fluctuations of the material surface temperature can induce large outgassing response at high frequency when surface is non-saturated**

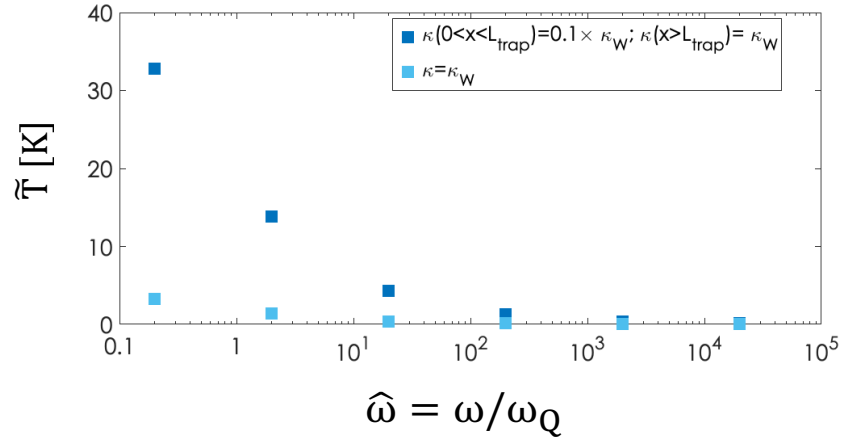
Fluctuations of temperature near material surface may be large due to reduced thermal conductivity caused by impurities and defects

- H outgassing material response to temperature fluctuations governed by activated processes:

$$\frac{\tilde{\Gamma}_{H,out}}{\bar{\Gamma}_{H,out}} \propto \frac{E}{T^2}\,\Delta T\,\frac{\omega/\omega_c}{\sqrt{(\omega/\omega_c)^2+1}}$$

- Rudimentary model of heat deposition on PFCs surface shows that

$$\frac{\tilde{T}}{\bar{T}} \propto \frac{\kappa}{\rho c_p}\frac{\tilde{Q}}{\bar{Q}}\frac{1}{\sqrt{(\omega/\omega_Q)^2+1}} \text{ with } \omega_Q \approx \frac{L_{bulk}^2}{\kappa}$$



- Modeling of realistic dynamic temperature response of PFCs is complex and must include
  - Realistic PFC geometry and cooling components
  - Reduction of thermal conductivity due to impurities and defects in material[1,2], which may lead to larger temperature near the surface
  - Macroscopic plasma dynamics such as strike point motion due to vertical plasma displacement[3] (e.g. strike point excursion in ITER ~ several cms)

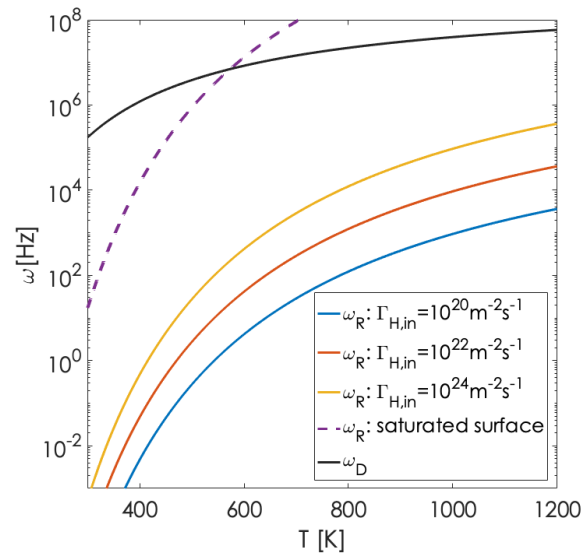
- **Realistic thermal wall model must be provided to predict outgassing material response to heat flux fluctuations … but <u>any experimental measurements</u> of wall temperature fluctuations in tokamak will be extremely valuable!**

[1] S. Cui et al, Journal of Nuclear Materials 2017 [2] L. Hu et al, Appl. Phys. Lett. 2017 [3] R. Pitts et al. Nuclear Fusion 2019

H surface processes on W may provide time scale separation between plasma fluctuations and outgassing response from W wall

- Outgassing response from wall to **fluctuations of plasma particle flux ~ low-pass filter**
- Outgassing response from wall to **fluctuations of material temperature ~ high-pass filter**
- Outgassing response from W bulk determined by free H diffusion ($\omega_{cutoff} \approx \omega_D = \frac{D}{\lambda_{imp}^2}$)
- Outgassing response from W surface determined by H recombination and desorption $\left(\omega_{cutoff} \approx \omega_R = K_r^{\alpha^{-1}} (\Gamma_{b\to s})^{1-\alpha^{-1}}\right)$
- Modeling of thermal fluctuations induced by fluctuations of heat flux difficult because of uncertainties in effects of implantation of plasma species on thermal conductivity of W
- In summary, H surface processes on W may induce **time scale separation between slow wall outgassing response and fast plasma fluctuations observed in divertor:**
  - $\omega \neq \omega_{cutoff}$: may largely simplified coupling of SOL turbulence simulations to wall dynamics
  - $\omega \approx \omega_{cutoff}$: dephasing between neutral outgassing flux and plasma flux

Cut-off frequency for H in W



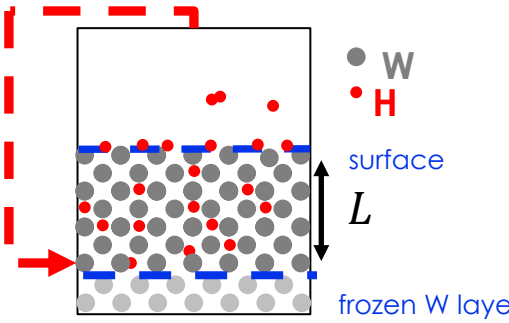
⇒ **Characterization of H processes on W surface with MD simulations**

Can H desorption from W be modeled with currently available W-H interatomic potentials?

- First-principle atomic modeling (MD simulations) may help to identify and characterize key mechanisms governing H outgassing from W:
  - Thermal desorption
  - Ion-induced desorption
  - H surface saturation effects
- Two types of W-H interatomic potentials available in literature: Tersoff [1] & EAM [2] potentials
- H molecular desorption from W cannot be reproduced with Tersoff potential [3]
- **But can EAM interatomic potential model H desorption from W?**
- Modeling of H desorption from W with MD simulations (LAMMPS):
  - Kinetic of surface desorption a priori unknown :

$$\Gamma_{desorption} = K_0\, e^{-\frac{E_{des}}{T}} c_s^{\alpha(c_H,T)}$$

  - MD simulations method to characterize **non-perturbed** H desorption at various material temperature T and various total amount of H in simulation to obtain $\Gamma_{desorption}(c_s, T)$ as a function of the H surface concentration c_s
  - Kinetic order of desorption α and activation energy of desorption E_{des} can be derived from $\Gamma_{desorption}(c_s, T)$, provided that H transport ($\tau_D = \frac{L^2}{D}$) is faster than H desorption (τ_{des}) $= K_r c_s^{\alpha-1}$ $\Rightarrow$ $L \sim 15nm$ with $T = 900K - 1400K$



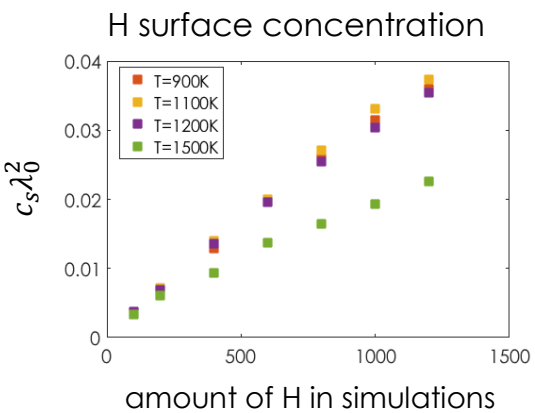
[1] N. Juslin *JAP* 2005
[2] L. Wang *JPCM* 2017
[3] J. Guterl *JNM* 2013

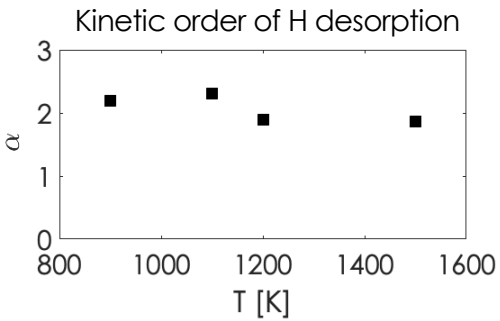
Hydrogen molecular desorption from W simulated with EAM potential at low H surface concentration in good agreement with experimental data

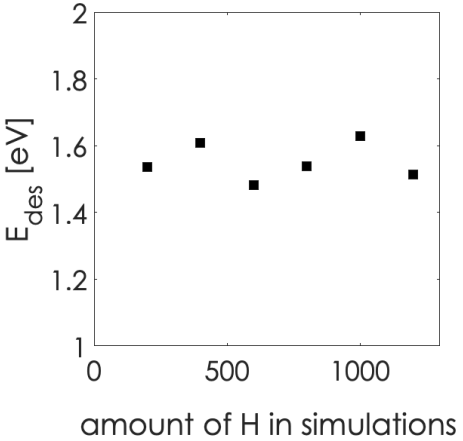
- MD simulations with EAM potential show H desorption from W as H_2 between T=900-1500K at low H surface concentration ($c_s\lambda_0^2 << 1$):

$$\Gamma_{desorption} = K_0\, e^{-\frac{E_{des}}{T}}\, c_s^{\alpha(T)} \text{ with } \alpha = 2$$

- Activation energy for molecular desorption from W <100> surface $E_{des} \sim 1.4 - 1.6\text{eV}$ **in good agreement with experimental H_2 desorption activation energies:**
  - Markelj[1] (2013): 1.6-1.7eV up to 2.2eV (polycrystalline)
  - Tamm[2] (1969): single crystal <100>: 1.4eV
- Pre-exponential factor $\mathbf{K_0 \sim \nu_0 \lambda_0^2 \sim 10^{-6} m^2 s}$ in good agreement with experimental estimations:
  - Markelj[1] (2013): $2 - 7\times10^{-7}\ m^2 s$
  - Tamm[2] (1969): single crystal <100>: $4\times10^{-6}\ m^2 s$
- **EAM potential well reproduces thermal H molecular desorption from W at low H surface concentration (unlike Tersoff potential)!**







[1] S. Markejl *Applied Surface Science* 2015 [2] P. Tamm *Journal of Chem. Phys.* 1969

Hydrogen molecular desorption simulated with EAM potential at high H surface concentration in qualitative agreement with experimental data

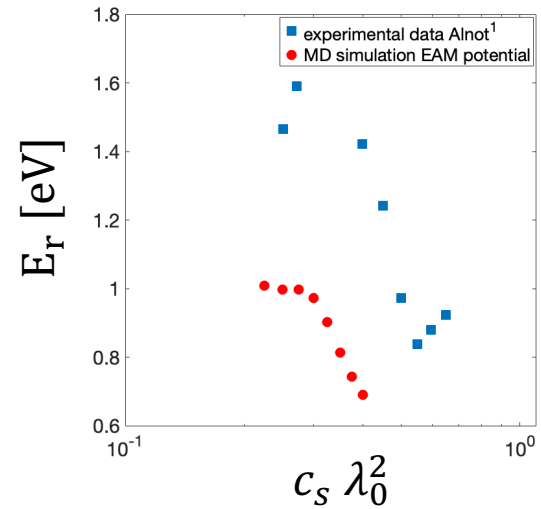
- H desorption from W saturated surface $(c_s\lambda_0^2 > 0.1)$ expected for ITER relevant conditions
- H surface concentration determined by equilibrium with H bulk concentration:

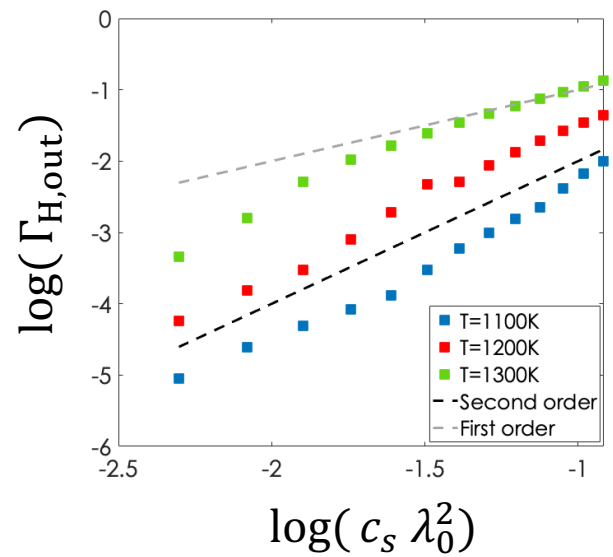
$$K_r \; c_s^{\alpha} = \frac{D}{L} c_{bulk}$$

- H bulk and surface concentrations varied through total amount of H in MD simulation:

$$N_{tot} = \int_S c_s \, dS + \int_V c_{bulk} \, dV$$

- Narrow parameter range to simulate H desorption from W saturated surface :
  - H desorption slow $E_r > 1eV \;\; => \;\; T > 900K, c_s > 0.1$
  - Formation of H platelet (self-clustering) at large bulk concentration [2] induces massive H trapping $\Rightarrow c_{bulk}\,\lambda_0^3 < 0.04 \Rightarrow c_s < 0.5$ and T<1400K
- **Evolution of E_r with c_s at large surface concentration in qualitative agreement with experimental data**
- **Transition from second to first order kinetic of desorption as surface becomes saturated**



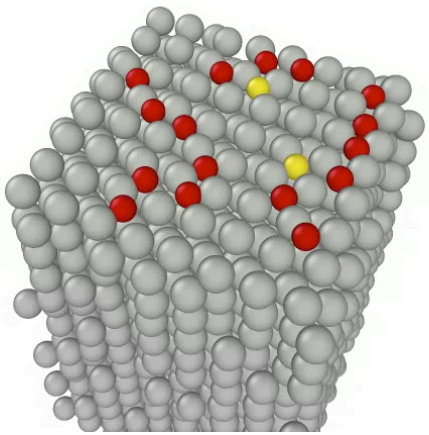


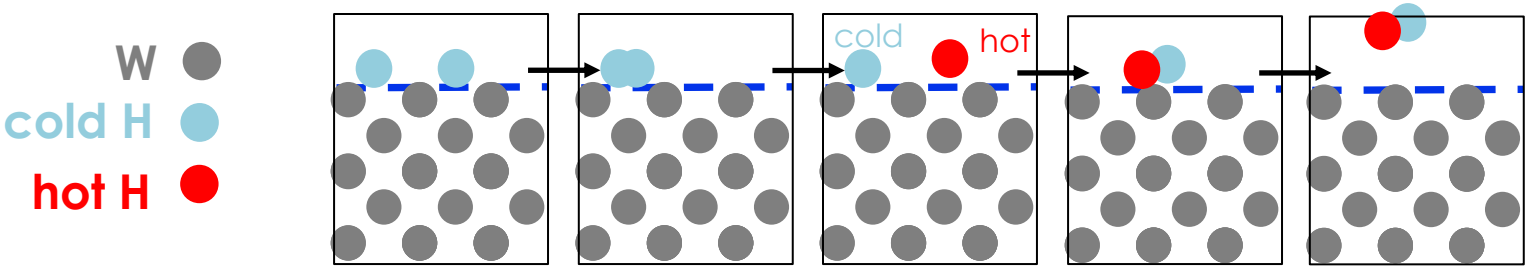
[1] P. Alnot *Surface Science* 1989
[2] R. Smirnov *Nuclear Fusion* 2018

A "mysterious" H precursor state for molecular desorption on W in simulations…

- H desorption from W usually assumed to result from the recombination of two H atoms into a molecule, which immediately desorbs
- **However, H molecular desorption on W follows an H precursor state[1] in MD simulations using the EAM potential, whether W surface is saturated with H or not:**
  1. recombination of two thermalized (cold) H into molecule
  2. dissociation of newly formed molecule into one cold and one hot H atom onto W surface
  3. molecular desorption results from the recombination of the hot atom with another cold atom
- Lifetime of molecules newly formed by two cold H is very short (t<0.1ps): no contradiction *a priori* with H_2 dissociation on W [1]

W <110> at T=1300K (EAM potential)





- W-H EAM potential fitted using bulk processes only and uncertainties remain for H surface processes on W:
  - **additional DFT simulations required to determine whether this H precursor state is an artefact from the EAM potential**

Ion-induced desorption and ion-induced detrapping may affect H recycling and retention in fusion relevant conditions

- Hydrogen recycling and retention in PFCs are usually assumed to be governed only by activated (thermal) processes (e.g. diffusion, trapping/detrapping in defects, desorption, …)
- However, **large amount of hydrogen can be present near material surface** (e.g. H super-saturation observed experimentally [1]) **and interact with large flux of impinging particles, resulting in ion-induced processes**
  - Exampe: well-known Eley-Rideal recombination of H on W surface with low-energy impinging H [2]
- Rudimentary model of ion-induced desorption and ion-induced detrapping:

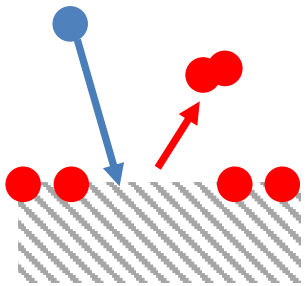
$$\frac{dc_{H,trap}}{dt} = -\sigma_{dt} c_{H,trap}\,\Gamma_{in} - \nu_{dt} c_{H,trap} + \nu_{tr}\left(c_{trap} - c_{H,trap}\right) c_H$$

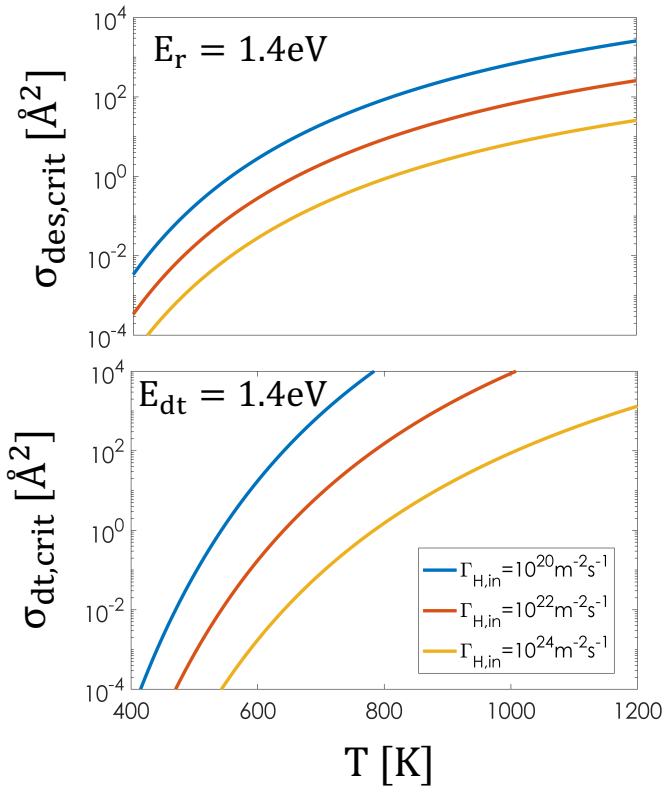
$$\frac{dc_s}{dt} = -\sigma_{des} c_s\,\Gamma_{in} - K_r\, c_s^{\alpha} + \Gamma_{b\to s}$$

- **Effects of ion-induced processes on H recycling are significant when**

$$\sigma_{dt} > \sigma_{dt,crit} = \frac{\nu_{dt}}{\Gamma_H} \text{ or } \sigma_{des} > \sigma_{des,crit} = \frac{K_r^{\alpha^{-1}}}{\Gamma_H^{\alpha^{-1}}}(1-R)^{1-\alpha^{-1}}$$

⇒ Estimations of σ_{des} and σ_{dt} with MD simulations

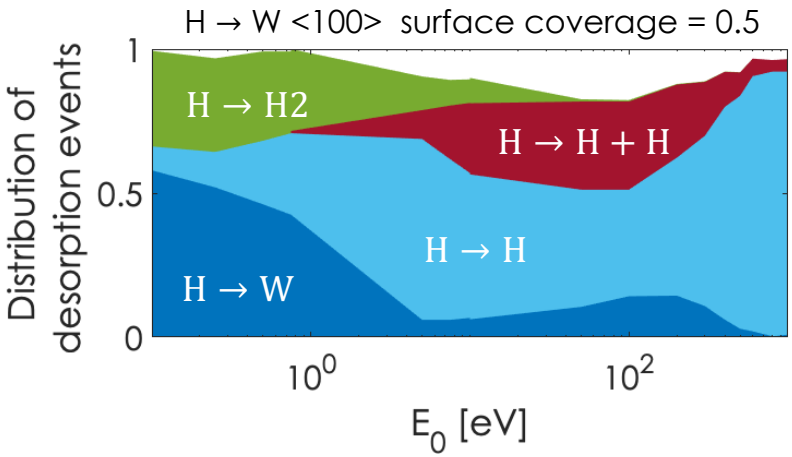


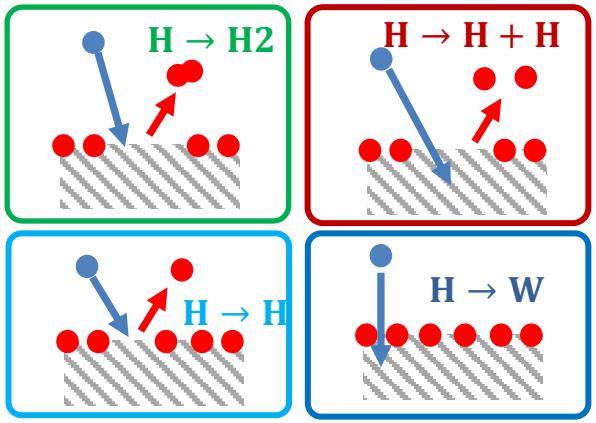


[1] L. Gao *Nuclar Fusion* 2016
[2] S.Markelj *Applied Surface Science 2013*

MD simulations suggest that ion-induced desorption may affect H recycling in various divertor plasma regimes

- MD simulations of ion–induced H desorption on W with EAM potential
- H impinging on W with 50% H coverage induces:
  - **Ion-induced H molecular desorption (Eley-Rideal)**
  - **H adsorption/implantation**
  - **H reflection**
  - **Ion-induced H atomic desorption**
- Eley-Rideal cross section: $\sigma \sim 1 - 5 \text{Å}^2$ in agreement with experimental/theoretical observations: $\sigma \sim 0.5 - 1\ \text{Å}^2$ [1]
- **Ion-induced desorption may increase:**
  - **H molecular desorption at low energy**
  - **H effective reflection at high energy**
- Some experimental observations in DIII-D suggest such ion-induced desorption during ELMs [2]
- Electronic effects may strongly affect Eley-Rideal mechanism at low energy [3]



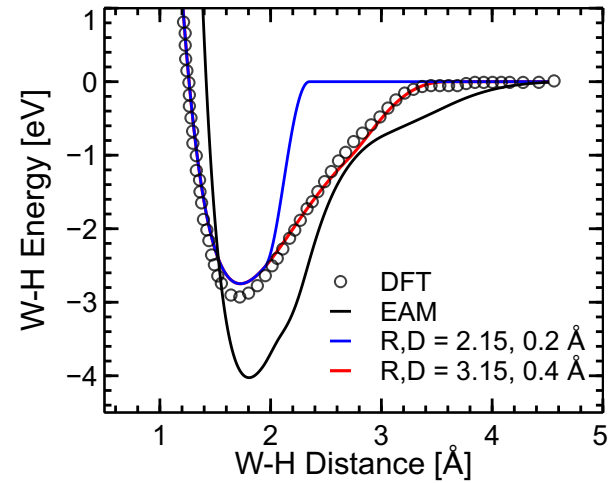


[1] B. Jackson *J. Chem. Phys.* 1992 [2] I. Bykov *PSI 2018* [3] O. Galparsoro, *Phys. Chem.* 2016

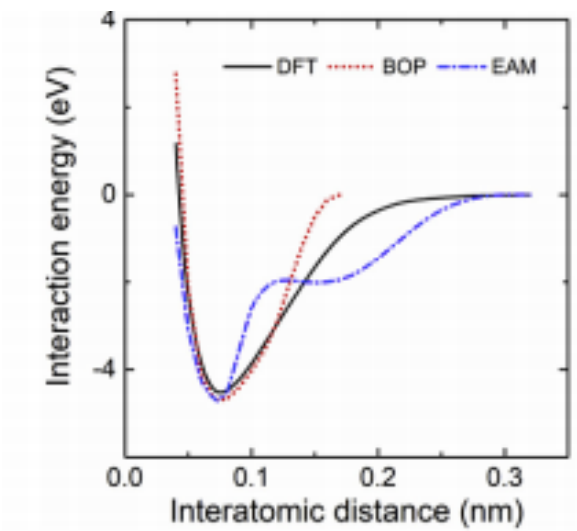
Benchmarking and improvement of EAM W-H interatomic potential against DFT simulations for H surface processes on W is crucial!

- **EAM potential seems to able to reproduce some key features of H desorption from W** (second-order thermal molecular desorption from non-saturated surface & Eley-Rideal H recombination)
- However, **EAM potentials have known intrinsic limitations** in modeling of BCC metal.
- Moreover, the **current EAM H-W potential does not well reproduce features of H_2 and H-W interactions in vacuum predicted with DFT** (in contrast with the Tersoff potential!)
- Is the plateau in H-H interaction energy at $r = 1\text{Å}$ obtained with the EAM potential responsible for the H recombination precursor state observed in MD simulations?
- **Can EAM potential actually be used to model H-W surface processes and accommodate multi-components chemistry (SiC-H-W[2], W-H-N) relevant for fusion reactor conditions?**
  - See poster PA069 (S. Bringuier)

H-W in vacuum [1,2]



H-H in vacuum [1]



[1] L. Yang JNM 2018
[2] Binguier this conference PQ069

Summary

- Outgassing response from wall to
  - fluctuations of plasma particle flux ~ low-pass filter
  - fluctuations of material temperature ~ high-pass filter
- W wall response to plasma fluctuations strongly determined by **surface processes** (desorption, saturation), which may induce **time scale separation between fast plasma fluctuations and slow outgassing response**
- Atomic characterization of H surface processes on W with molecular dynamics simulations:
  - **Unlike bond-order potential (Tersoff) potential, recently developed EAM potential can reproduce H thermal molecular desorption from W**
  - **MD simulation of H desorption from W surface saturated with H in qualitative agreement with experimental observations**
- MD simulations framework developed to characterize ion-induced desorption processes
- Uncertainties remain in the modeling of H surface processes on W with EAM potential:
  - **Validation of the EAM W-H interatomic potential with DFT simulations of surface processes is necessary!**