Novel method for studying hydrogen storage process in the nanometer length scale using count rate of neutrons scattered at a small angle and probabilistic structure generation

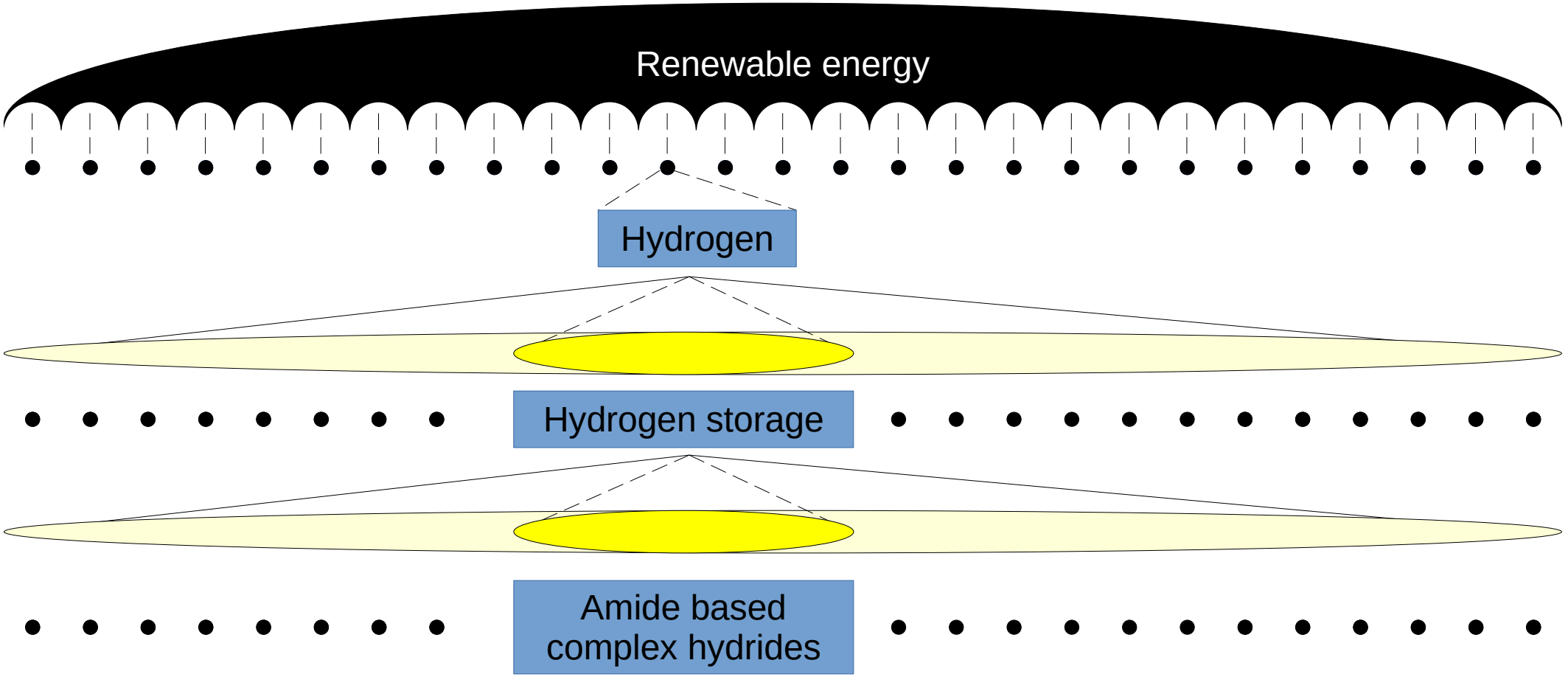
Arnab Majumdar, Martin Müller, Sebastian Busch

MLZ is a cooperation between:

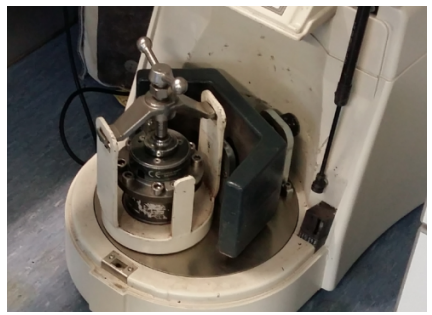
Agenda

1	Scientific problem
2	Calculation of neutron count rate from simulation
3	Structure generation using probabilistic simulation

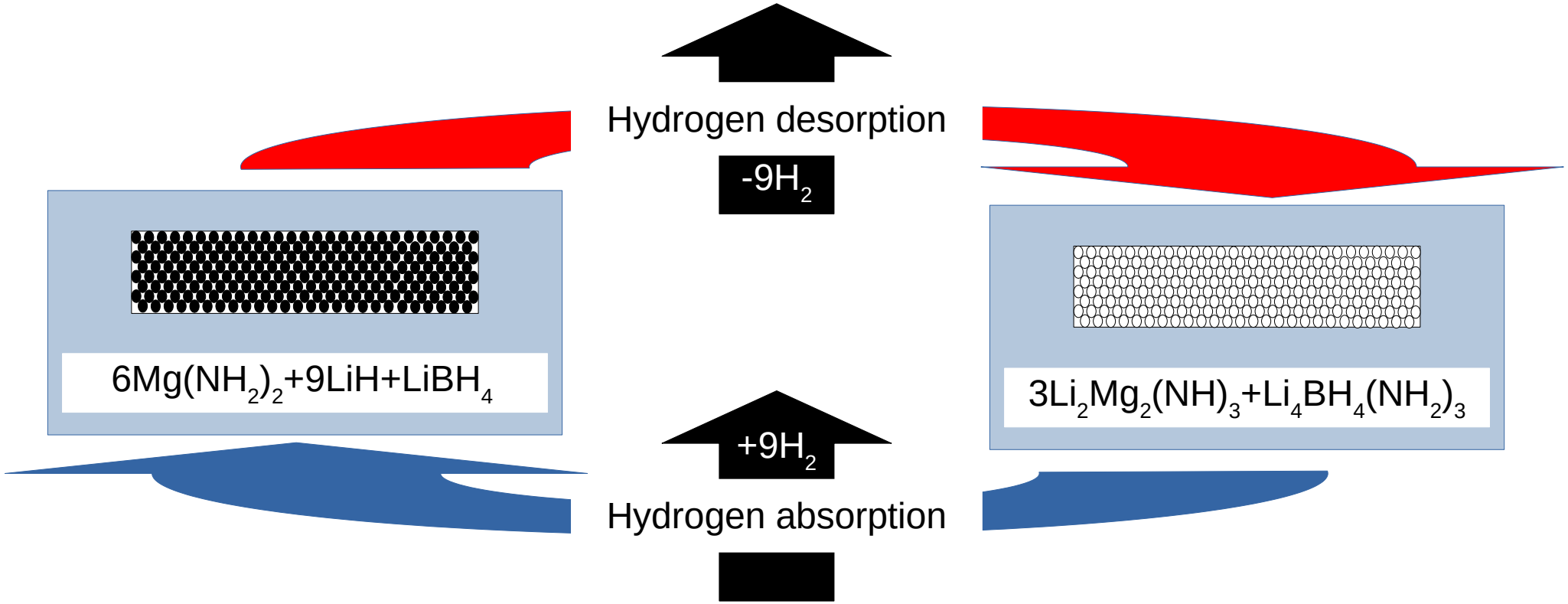
Focus


Renewable energy
Hydrogen
Hydrogen storage
Amide based
complex hydrides

Hydrogen storage using amide based complex hydrides

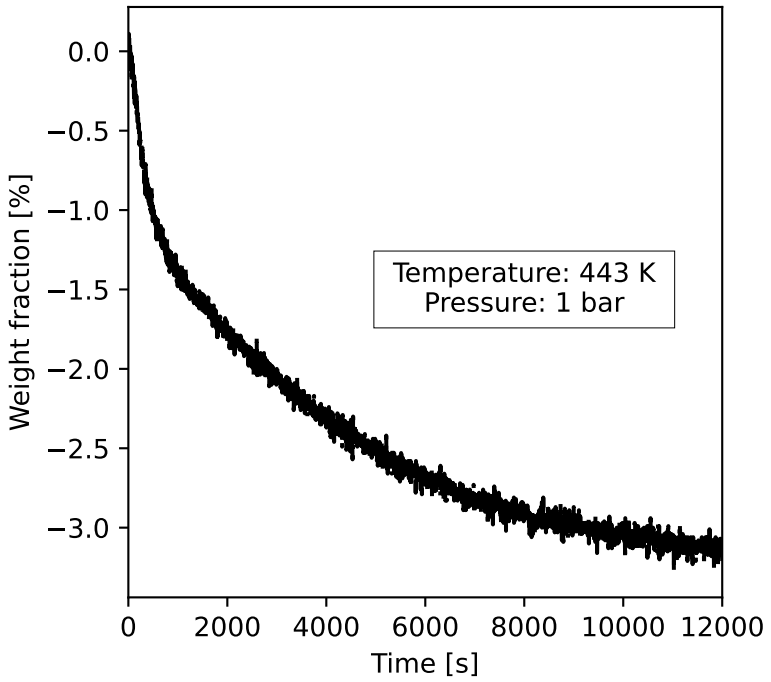


Ball milled sample

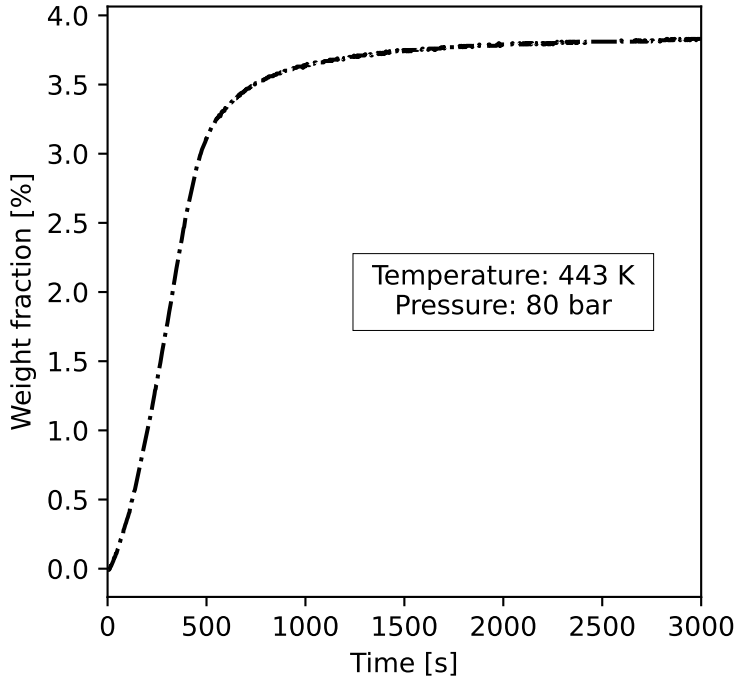


Engineering length scale: Volumetry measurement

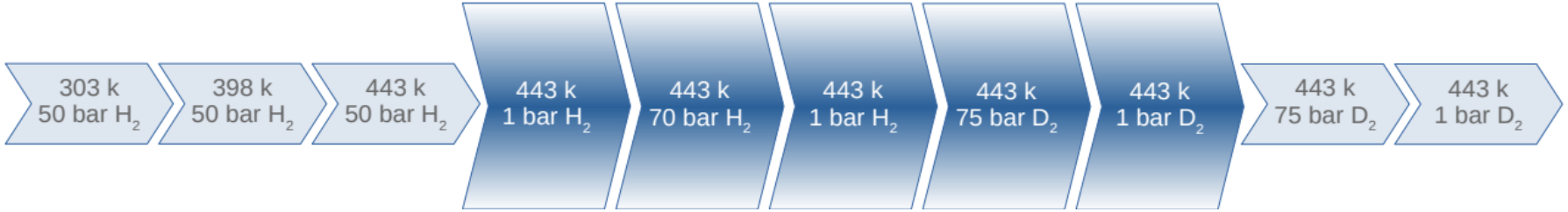
Desorption

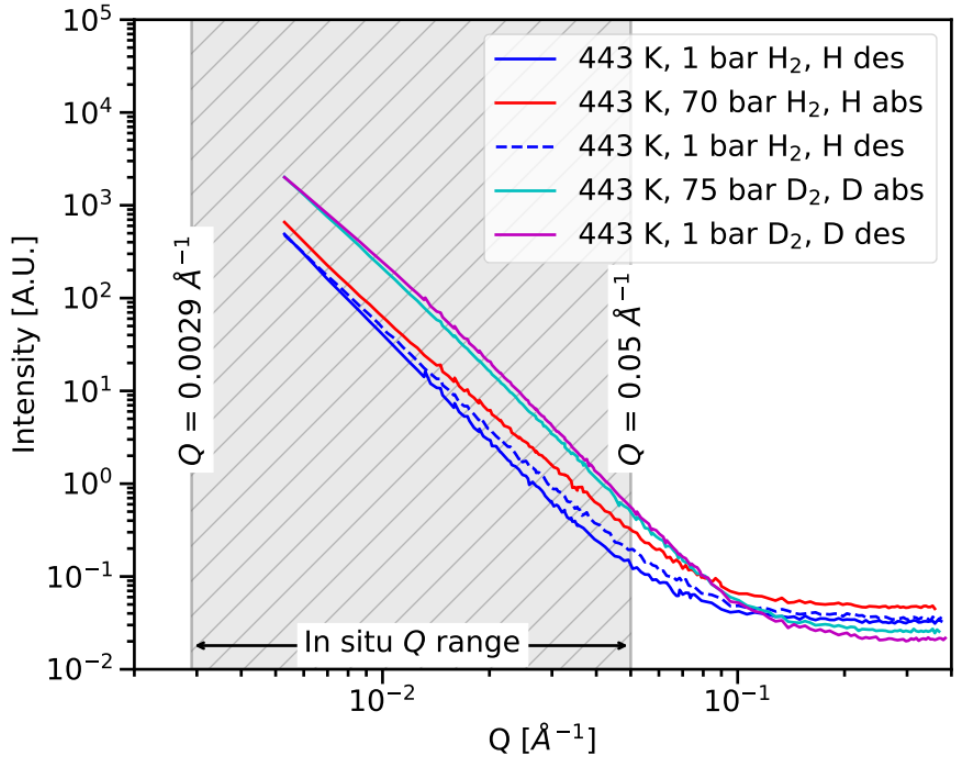


Absorption



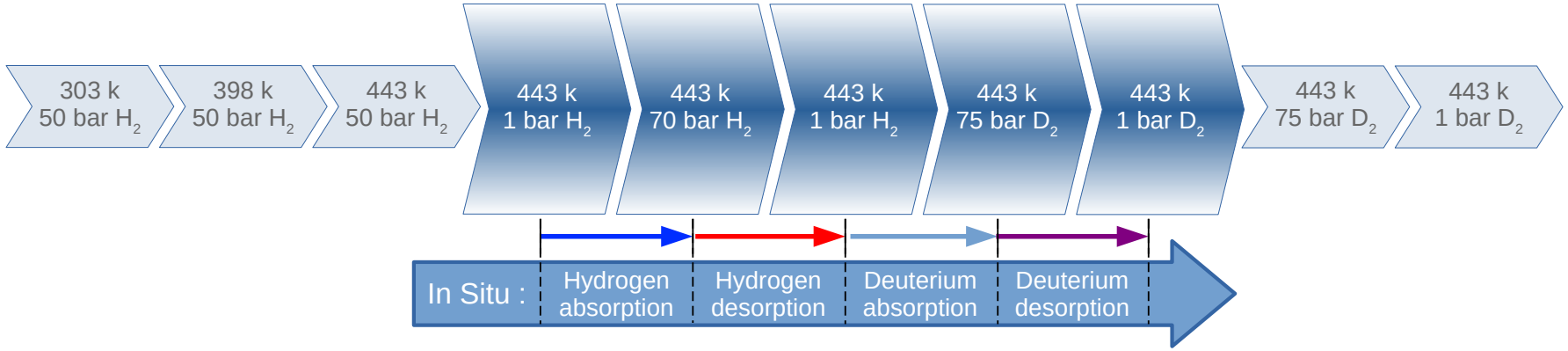
Nanometer length scale: Small Angle Neutron Scattering (SANS) measurement

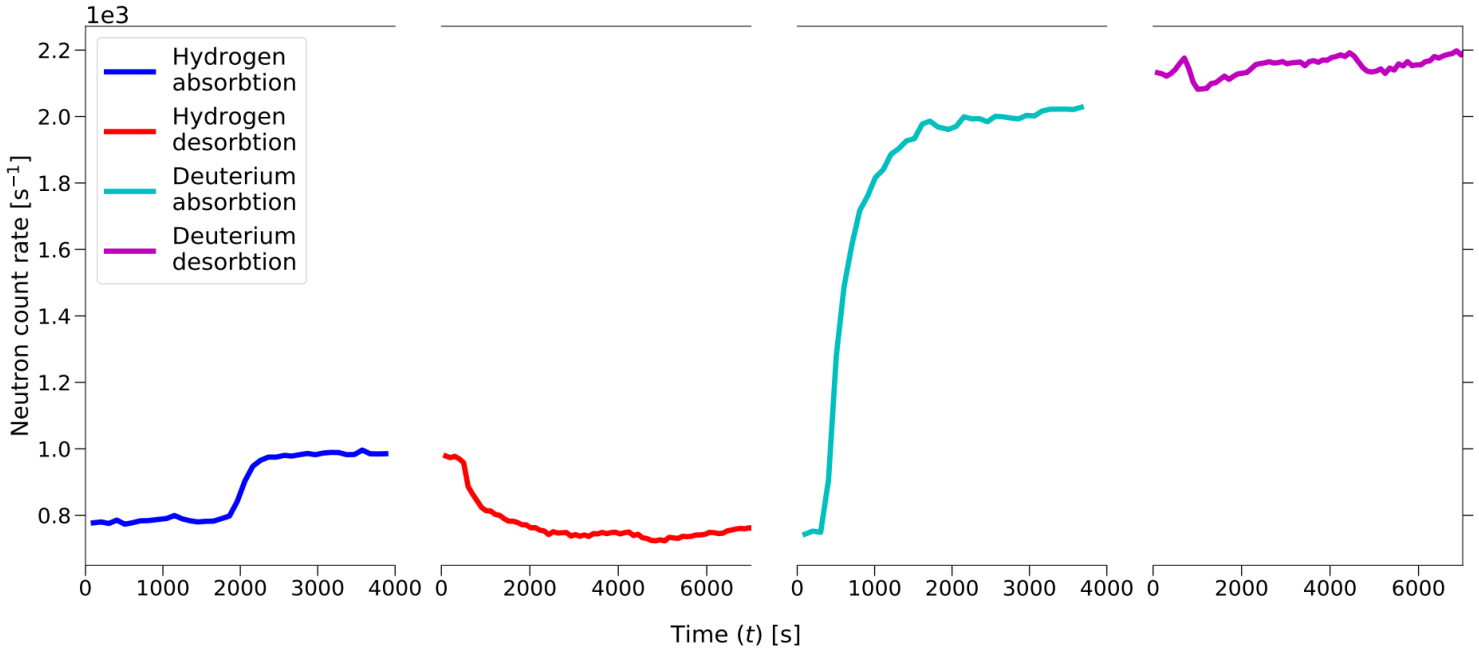




N. Aslan et al. DOI: https://doi.org/10.3233/JNR-190116

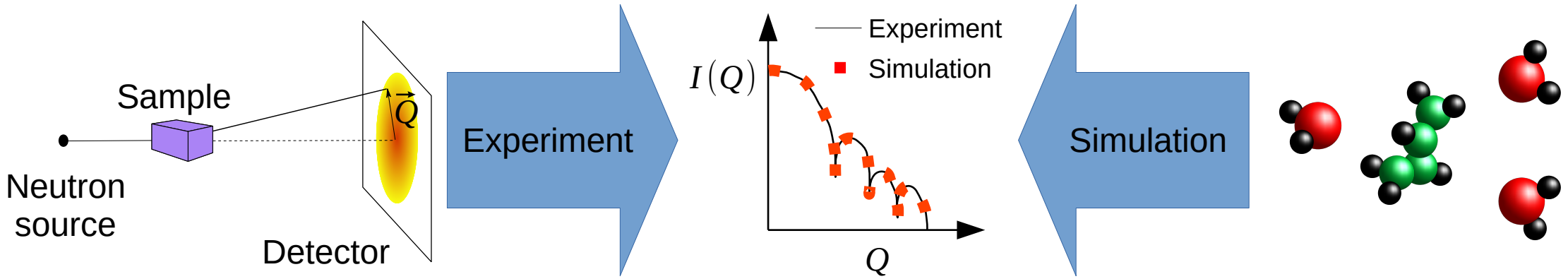
Nanometer length scale: In situ SANS measurement





N. Aslan et al. DOI: https://doi.org/10.3233/JNR-190116

Complementary use of simulations and SANS experiments



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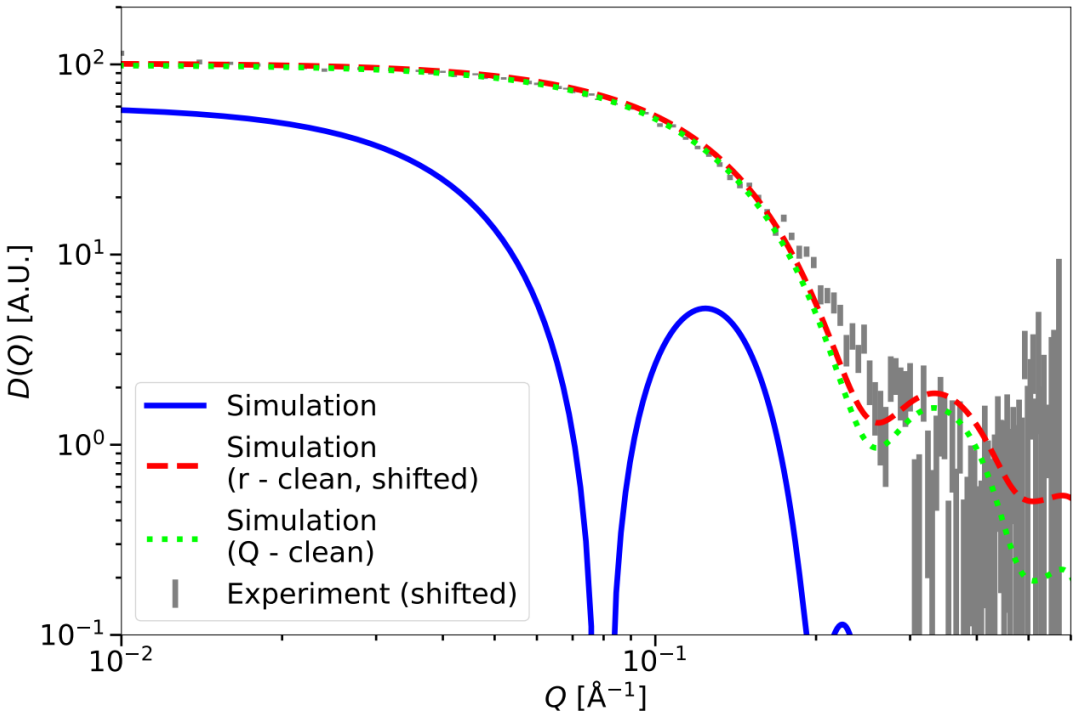
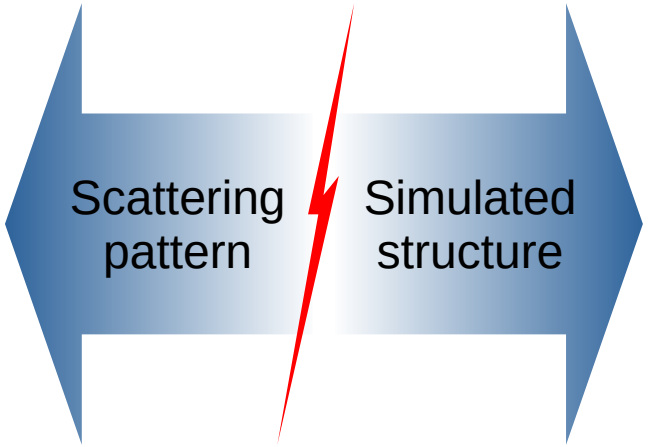
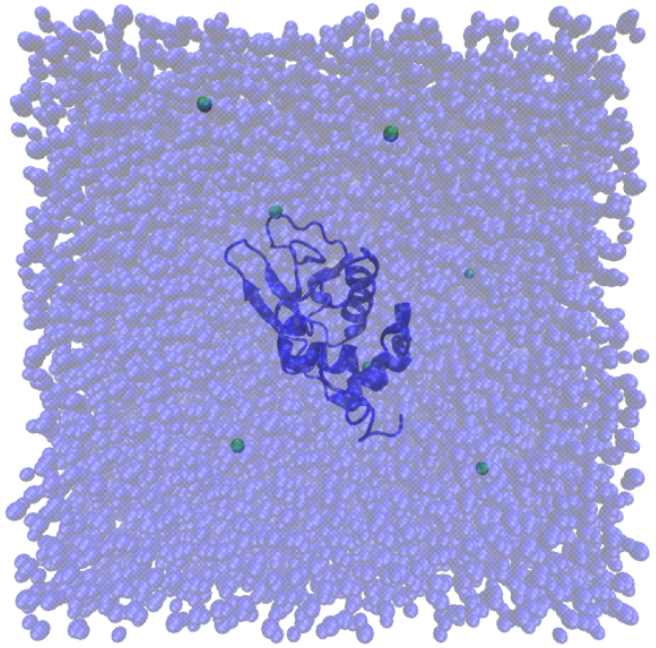
Q - clean method: Removal of finite size effect



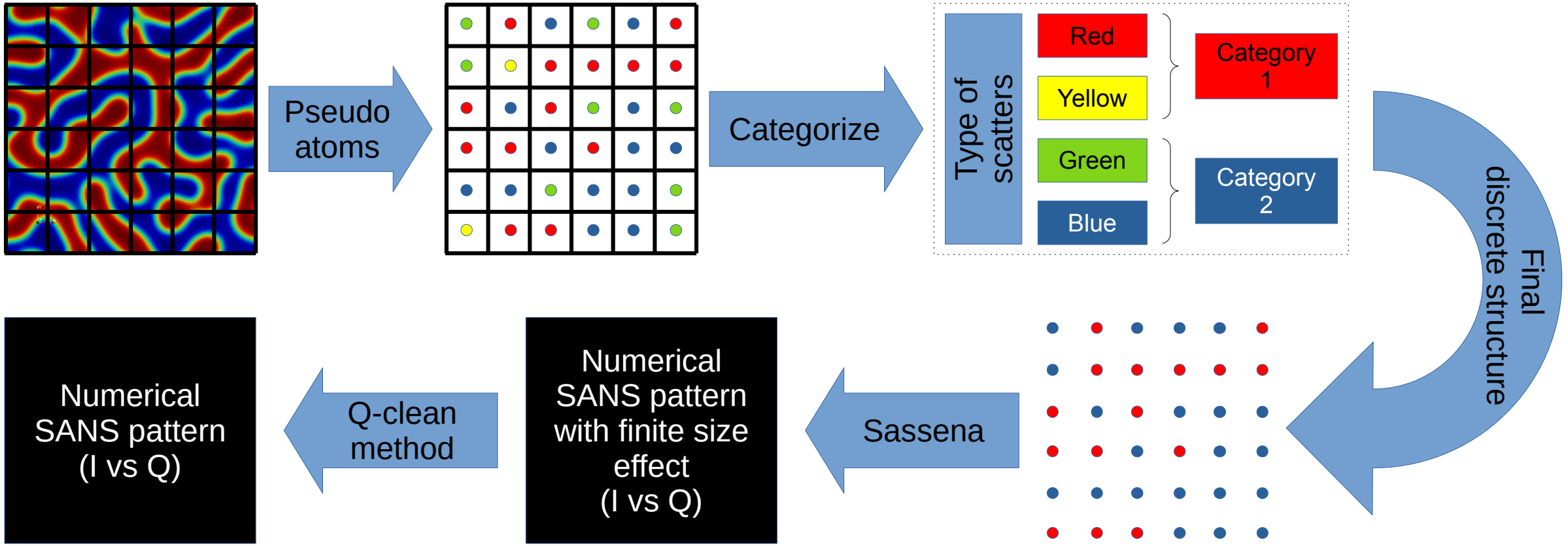




Key publications:
SANS measurement: J. Trewhella et al.*
Calculation of diffraction pattern: A. Majumdar et al.**

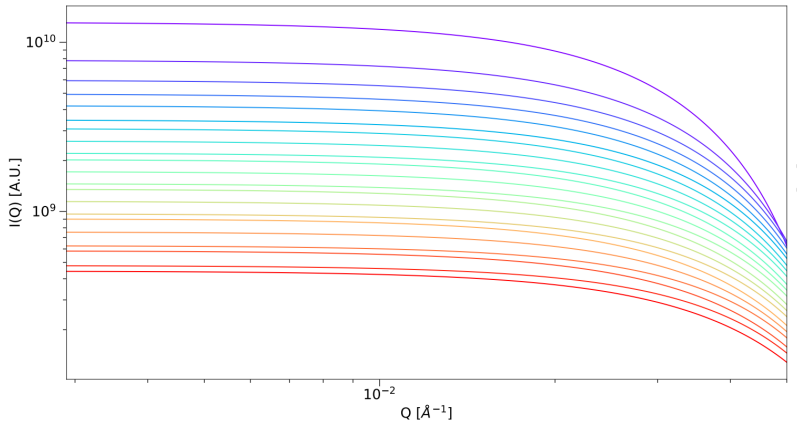
*DOI: https://doi.org/10.1107/s2059798322009184, **DOI: https://doi.org/10.3390/ijms25031547

Numerical method: Calculation of SANS pattern from SLD distribution

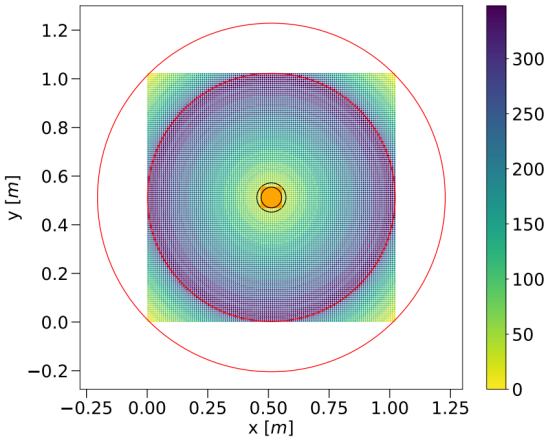


Calculation of neutron count rate per unit flux or effective cross-section

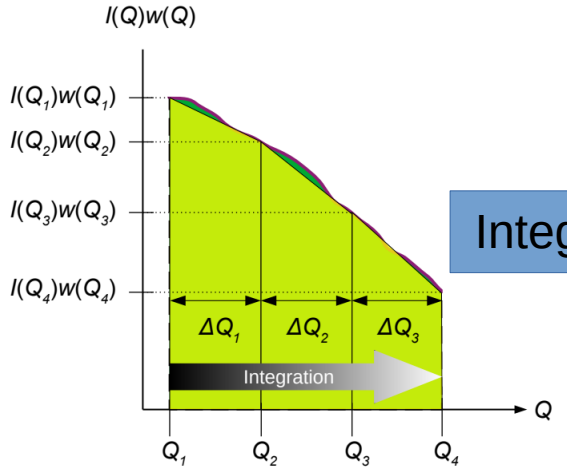
I(Q)



w(Q)



I(Q) w(Q)



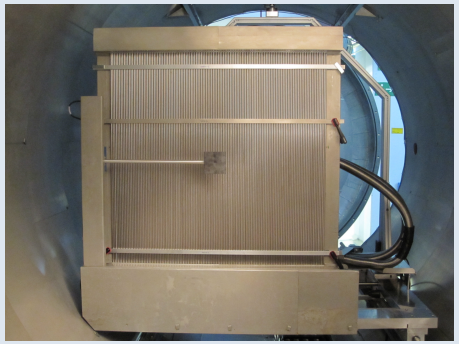
Integration

Neutron count rate per unit flux or Effective cross-section

Compare

Neutron count rate

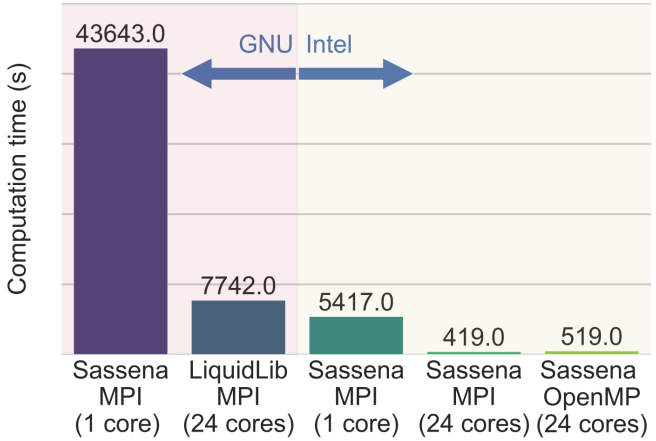
SANS1 detector



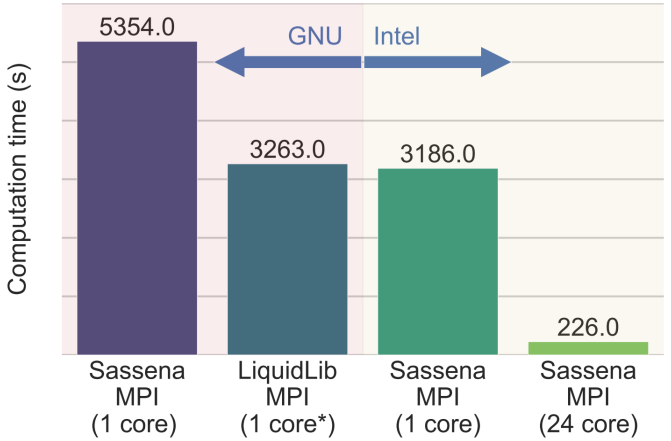
S. Mühlbauer et al. DOI: https://doi.org/10.3233/JNR-190116, L. Karge et al. DOI: https://doi.org/10.1107/S1600576717011463

Sassena - Software development

Computation time (Base configuration)

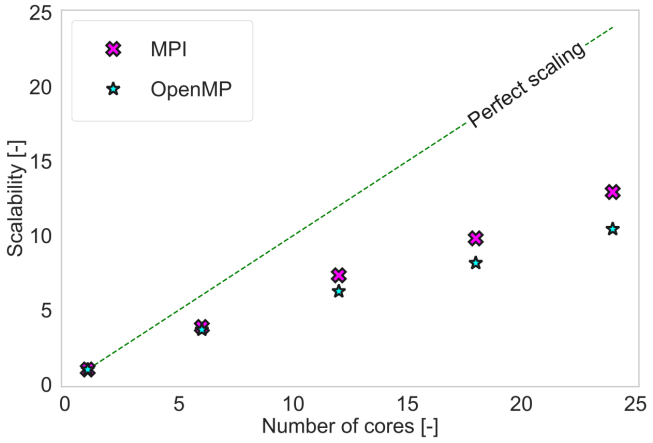


Coh Incoh

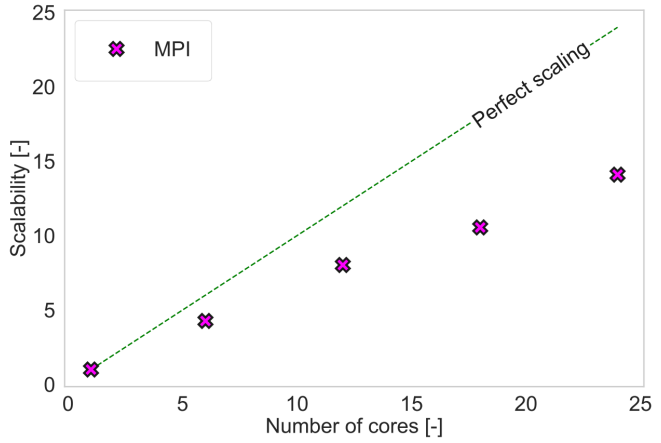


Gitlab project (Sassena)

Scalability (N cores) = Computation time (N cores) ÷ Computation time (1 core)



Coh Incoh



Sassena: B. Lindner et. al. DOI: doi:10.1016/j.cpc.2012.02.010, A. Majumdar et al. DOI: https://doi.org/10.3390/ijms25031547,
Liquidlib: N.Walter et. al. DOI: https://doi.org/10.1016/j.cpc.2018.03.005

Agenda

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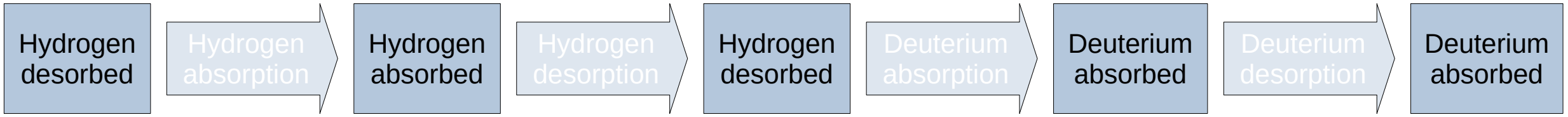
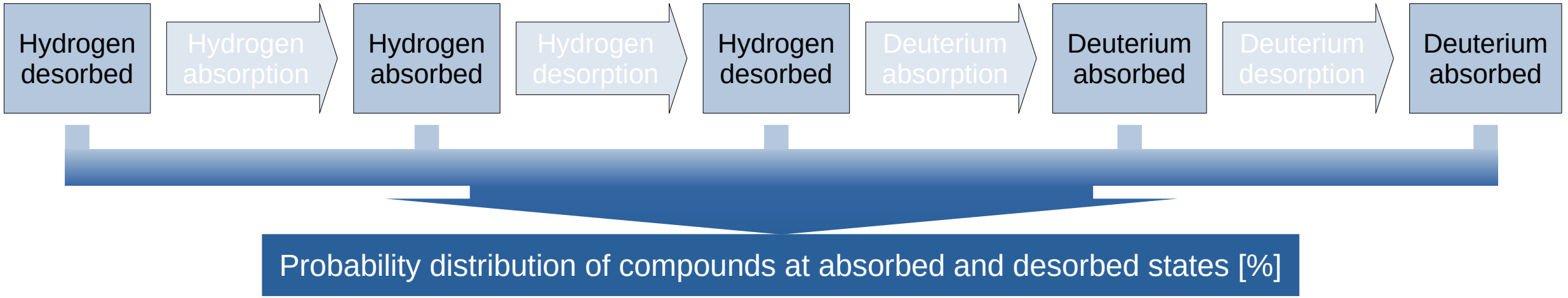
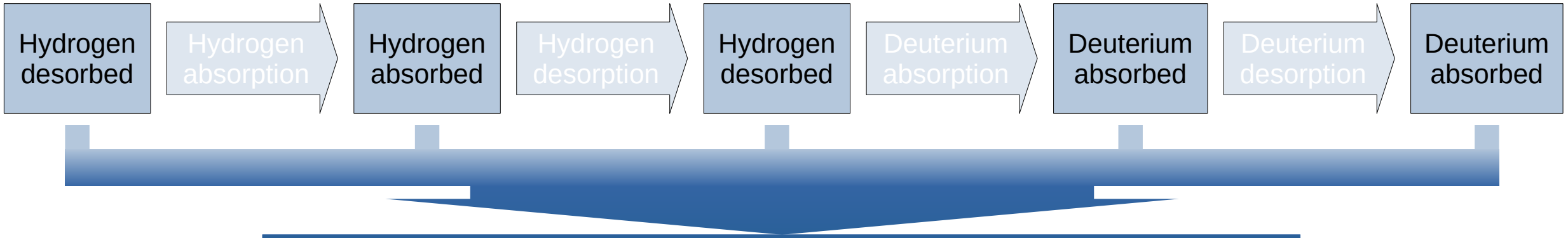
Probabilistic simulation: Generation of structures at end states of a process



Probabilistic simulation: Generation of structures at end states of a process

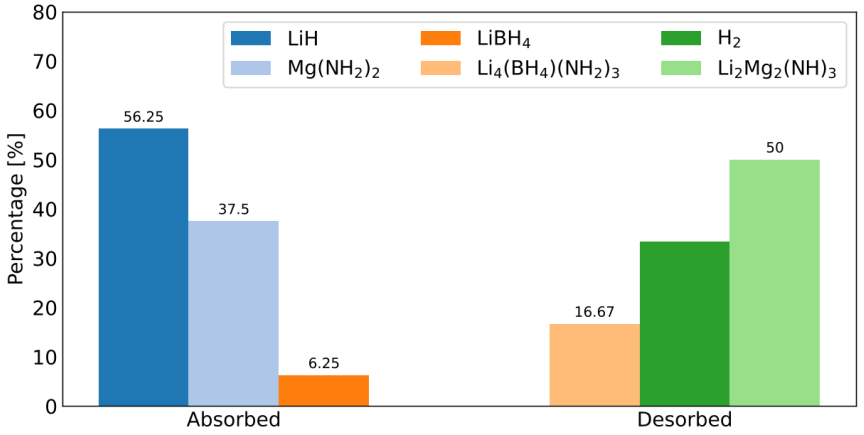


Probabilistic simulation: Generation of structures at end states of a process

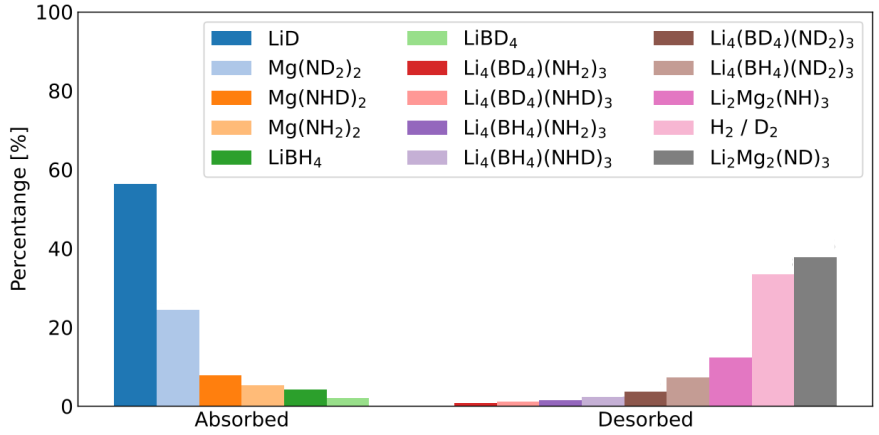


Probability distribution of compounds at absorbed and desorbed states [%]

Hydrogen absorbed and desorbed states



Deuterium absorbed and desorbed states

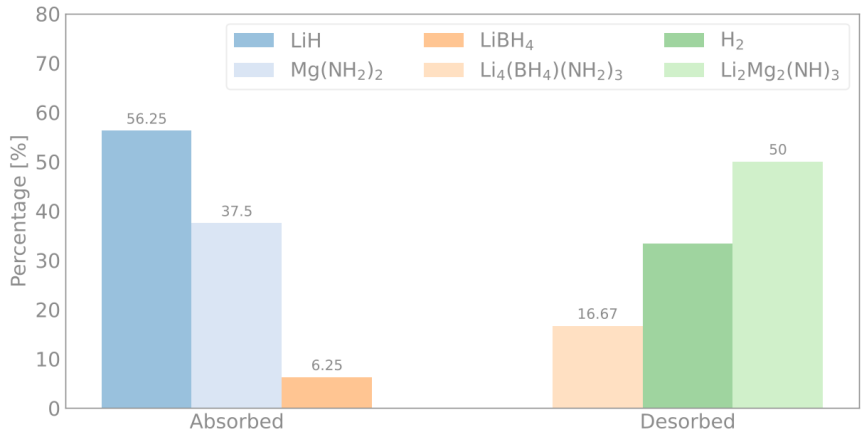


Probabilistic simulation: Unknown parameters for generation of structures

Hydrogen desorbed → Hydrogen absorption → Hydrogen absorbed → Hydrogen desorption → Hydrogen desorbed → Deuterium absorption → Deuterium absorbed → Deuterium desorption → Deuterium absorbed

Probability distribution of compounds at absorbed and desorbed states [%]

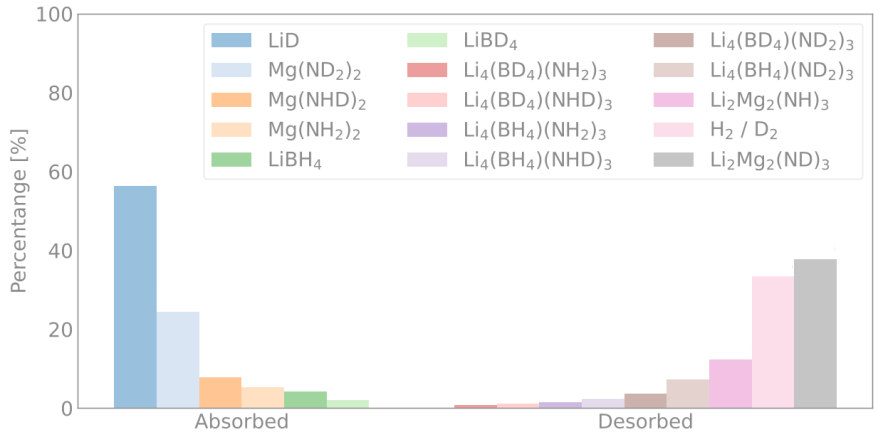
Hydrogen absorbed and desorbed states



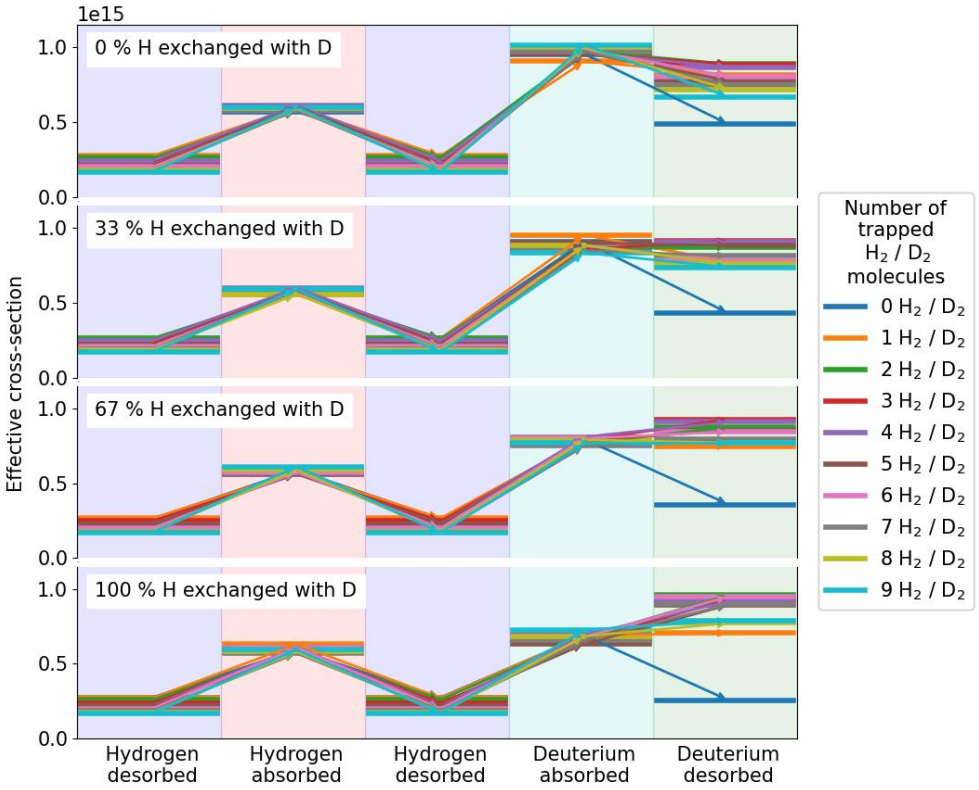
Two unknowns

- H-D exchange in BH_4^-
- Amount of trapped gas

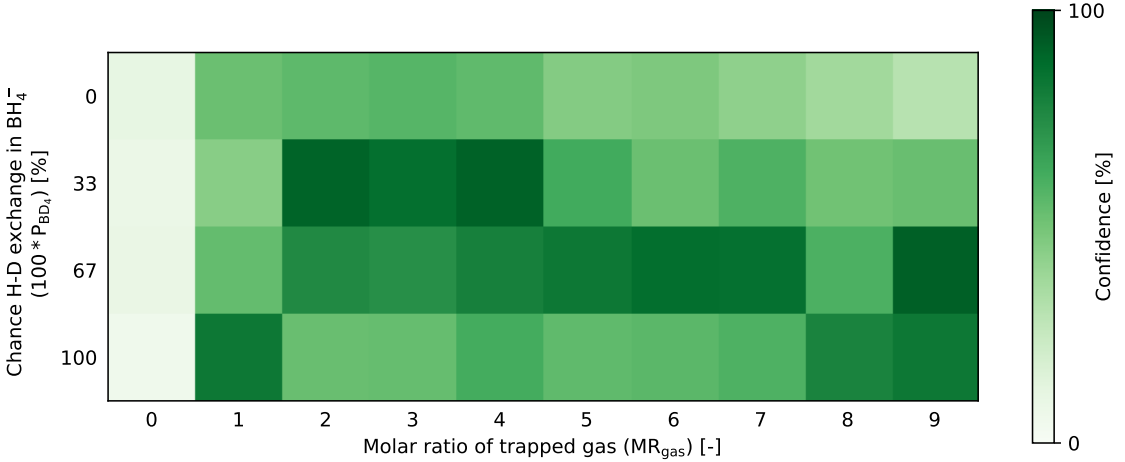
Deuterium absorbed and desorbed states



Probabilistic simulation: Optimization of unknown parameters



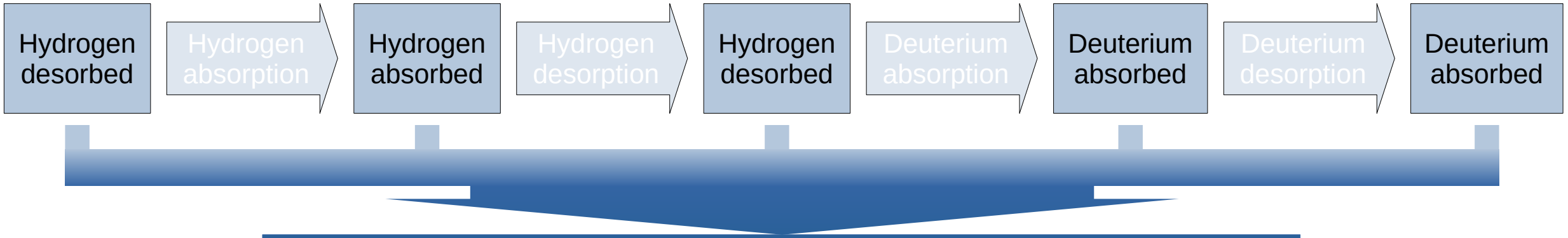
Confidence matrix



Values of unknowns after optimization

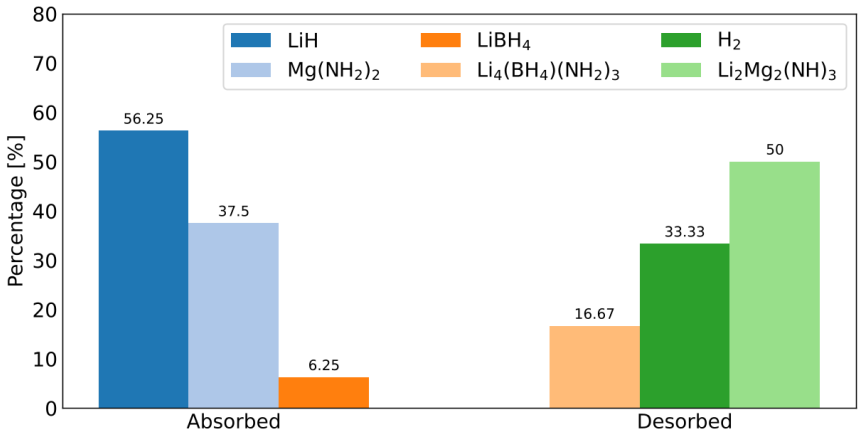
- 33% of BH_4^- subjected to H-D exchange
- 33.33% probability of having H_2 / D_2 in structures generated for desorbed states

Probabilistic simulation: Generation of structures at end states of a process

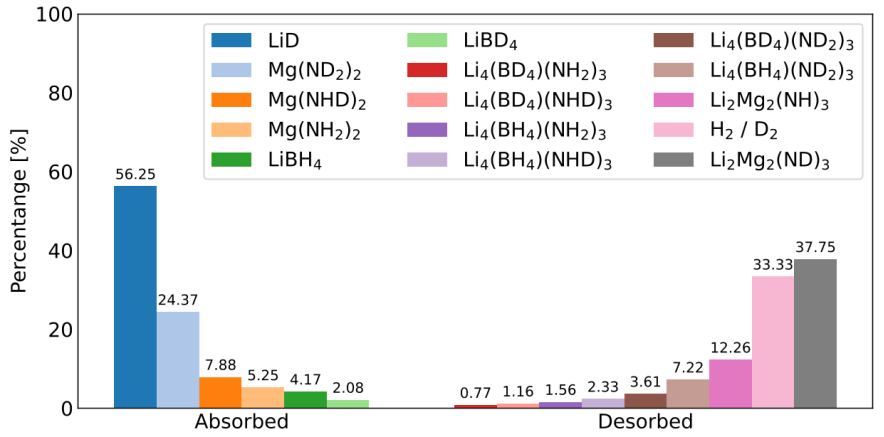


Probability distribution of compounds at absorbed and desorbed states [%]

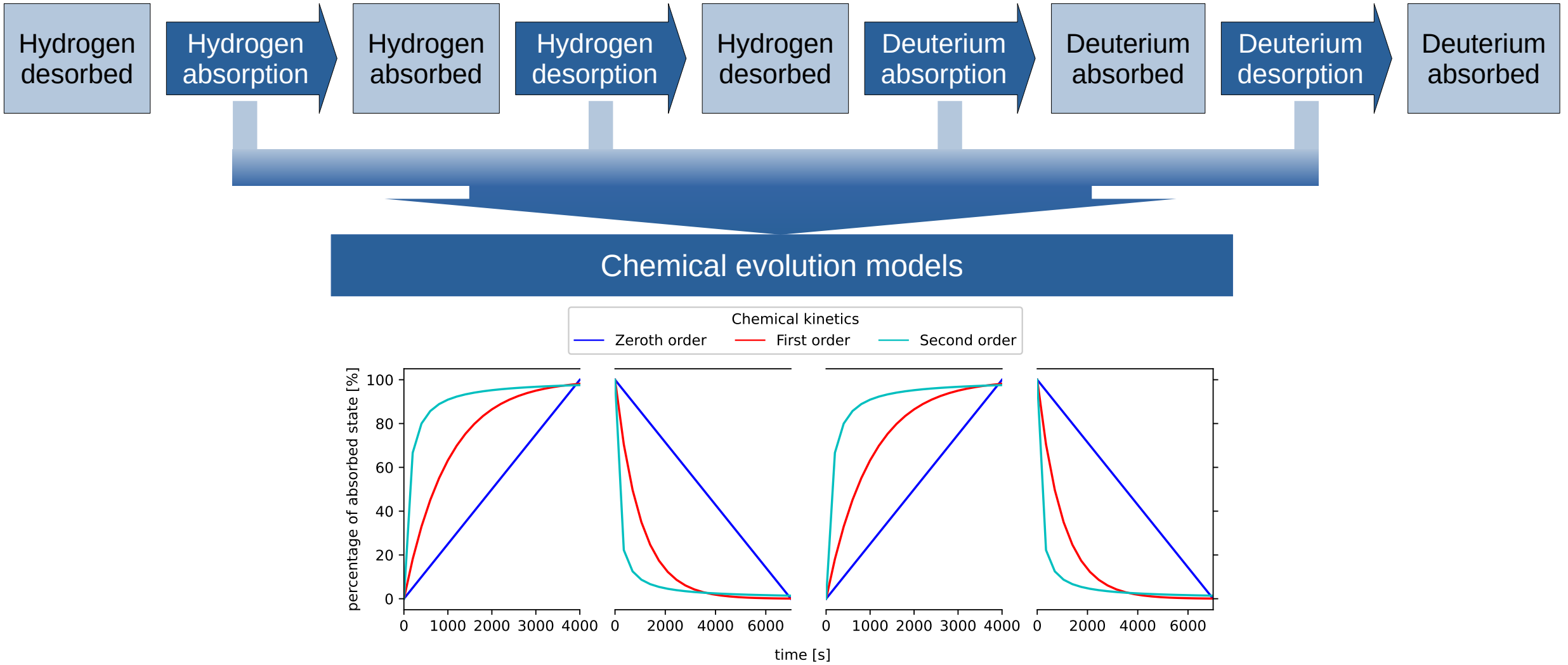
Hydrogen absorbed and desorbed states



Deuterium absorbed and desorbed states

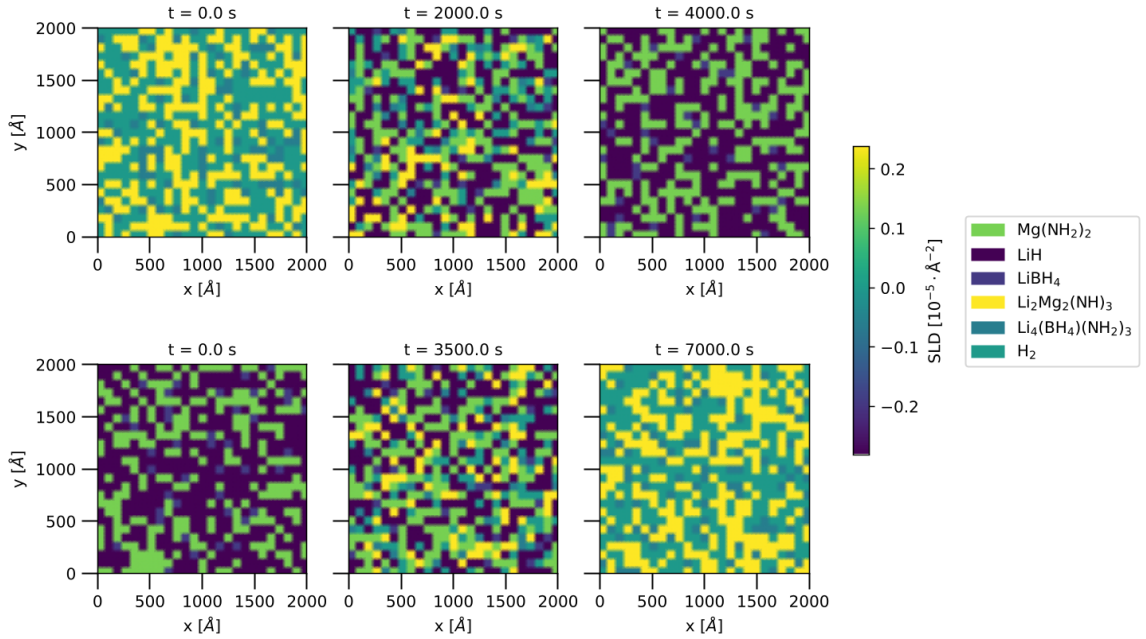


Probabilistic simulation: Chemical evolution of generated structures

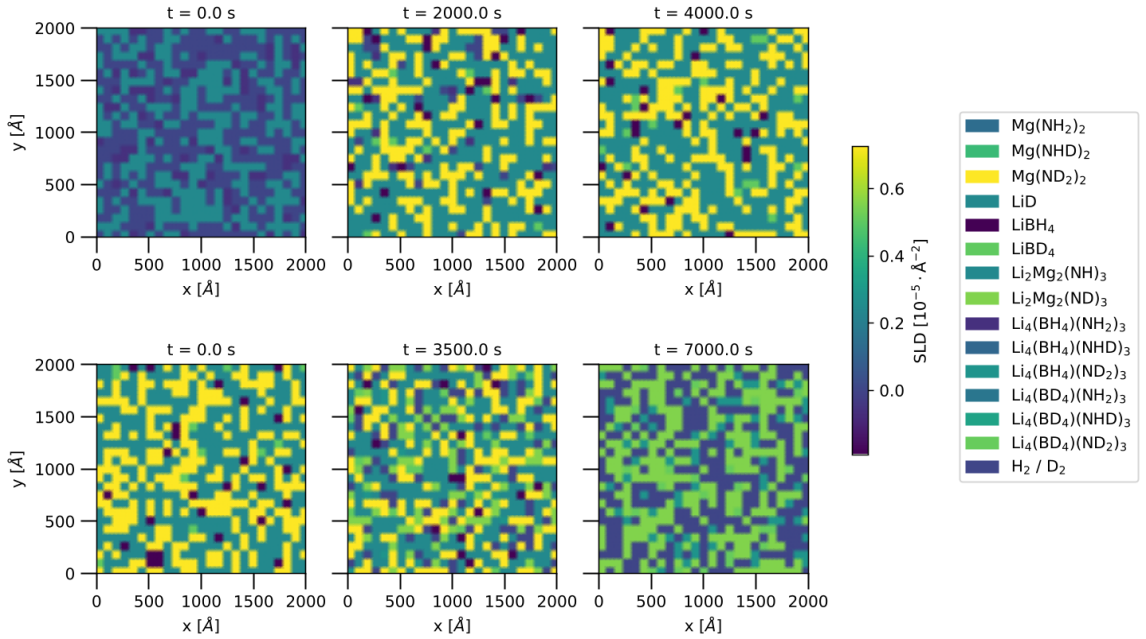


Probabilistic simulation: SLD distribution of generated structures

Hydrogen absorption



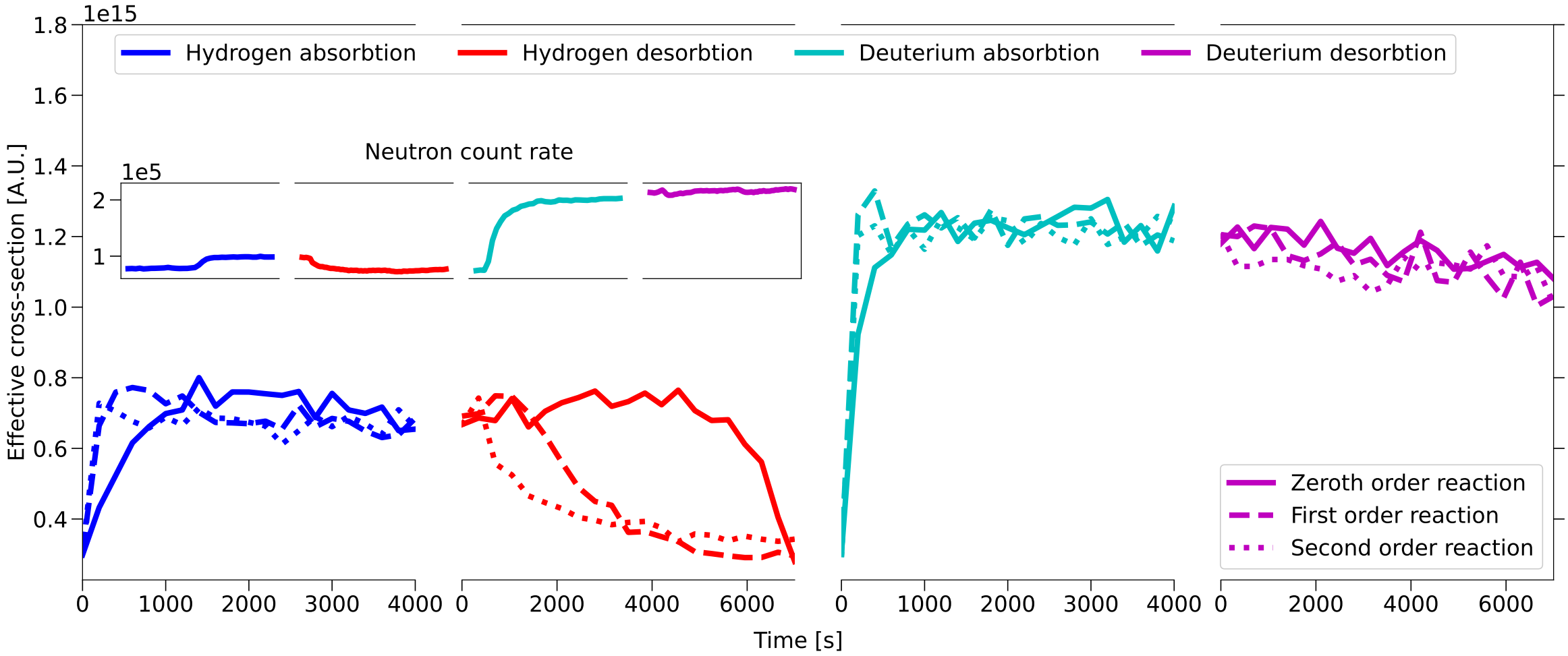
Deuterium absorption



Hydrogen desorption

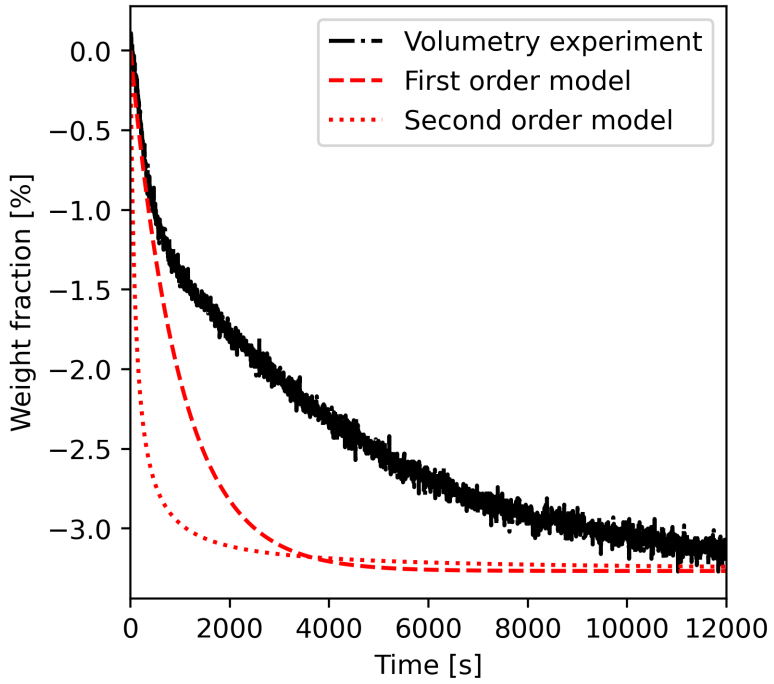
Deuterium desorption

Nanometer length scale: Neutron count rate vs effective cross-section

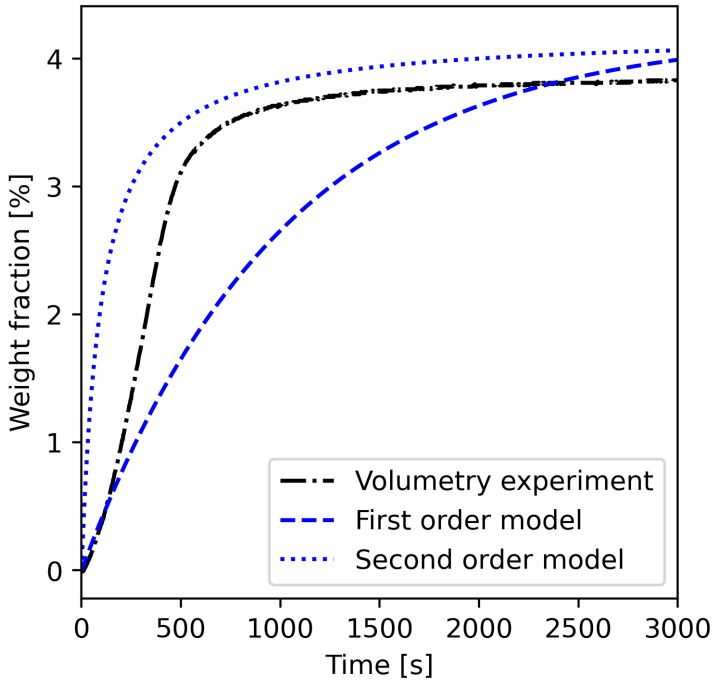


Effect of nanoscopic phenomena on engineering length scale

Hydrogen desorption



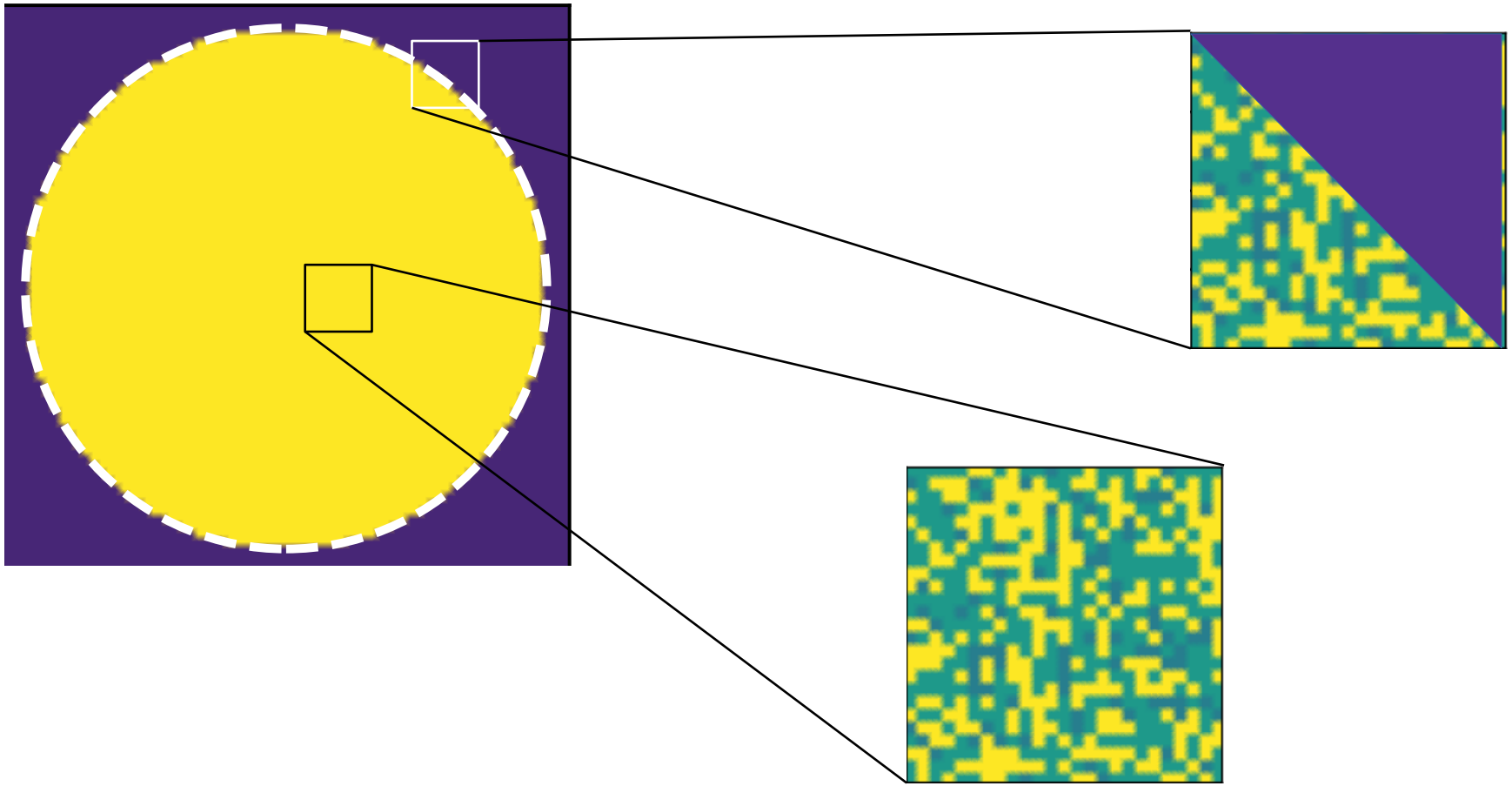
Hydrogen absorption



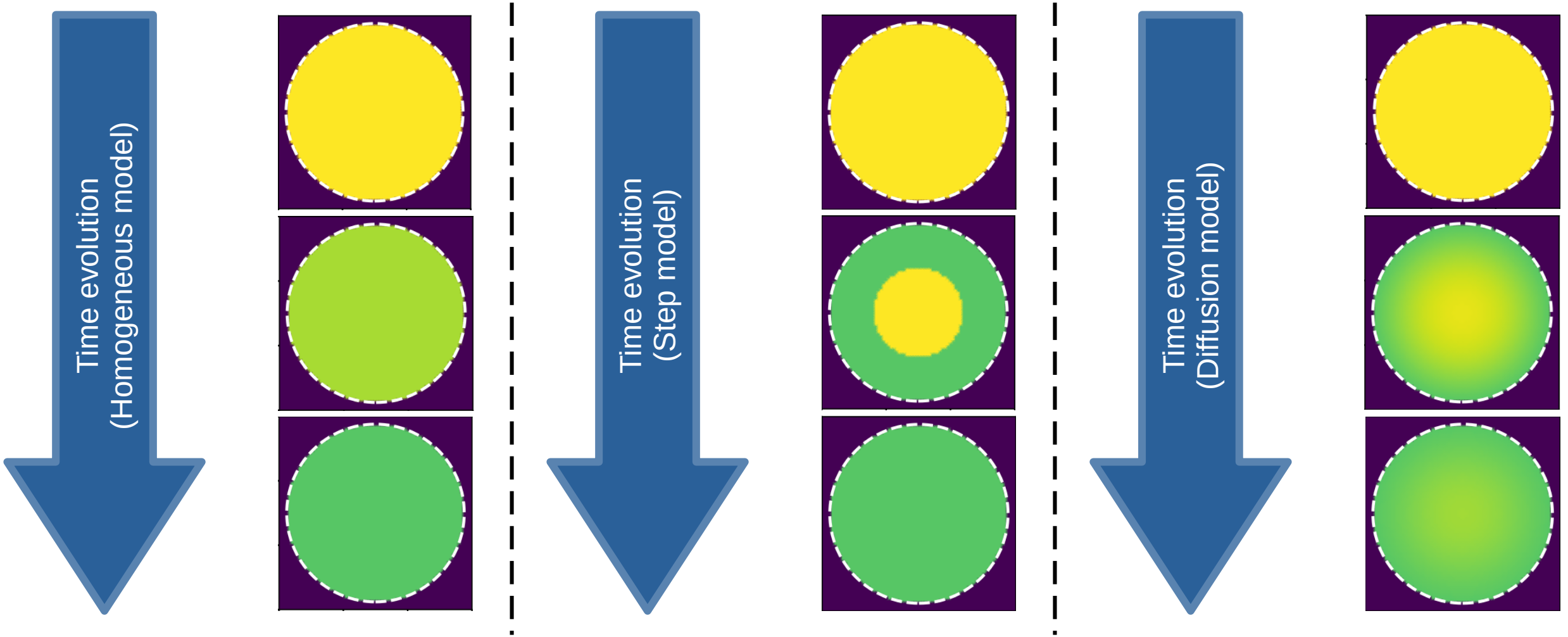
Thank you

Backup

Bigger length scale: Contrast between storage material and gas around it

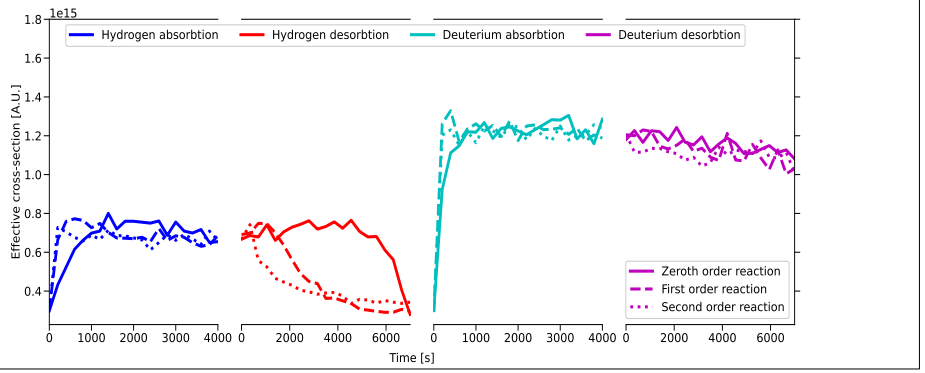


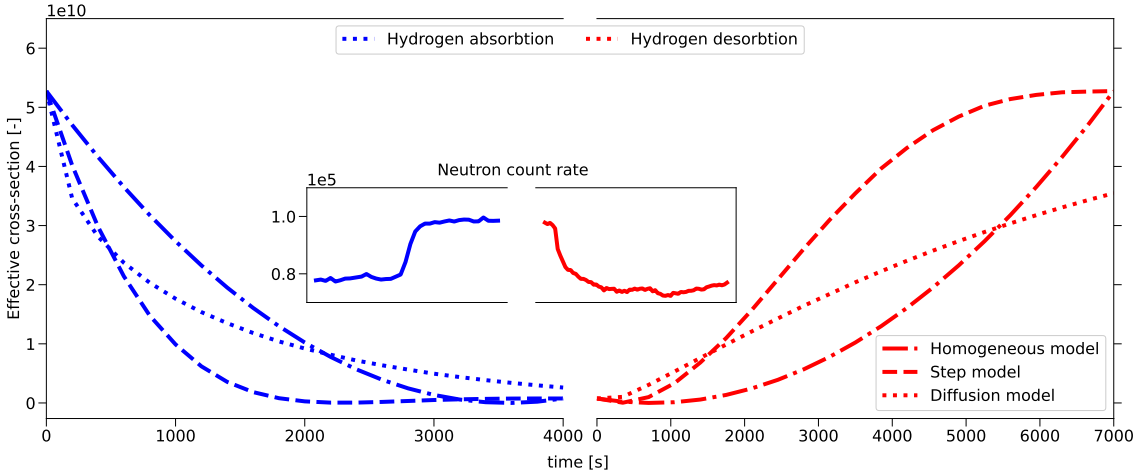
Bigger length scale: Chemical diffusion in a single grain

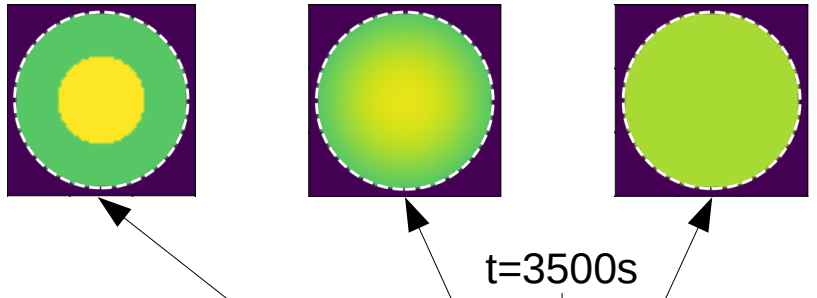


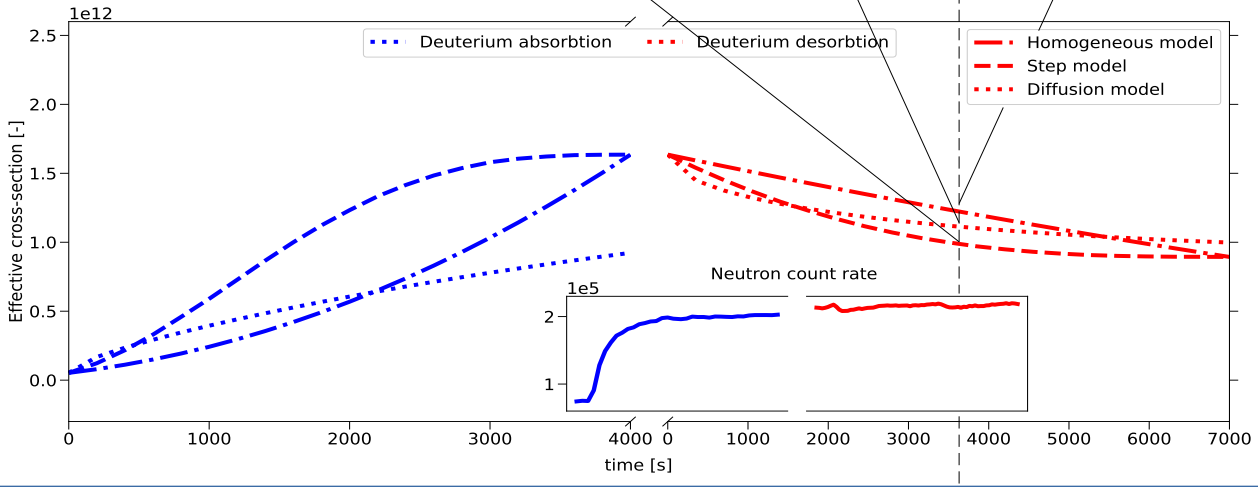
Bigger length scale: Neutron count rate vs effective cross-section

Nanometer length scale: Probabilistic model









Bigger length scale: Chemical diffusion in a single grain