Some cross-diffusion models for the evolution of chemical species in a crystalline solid

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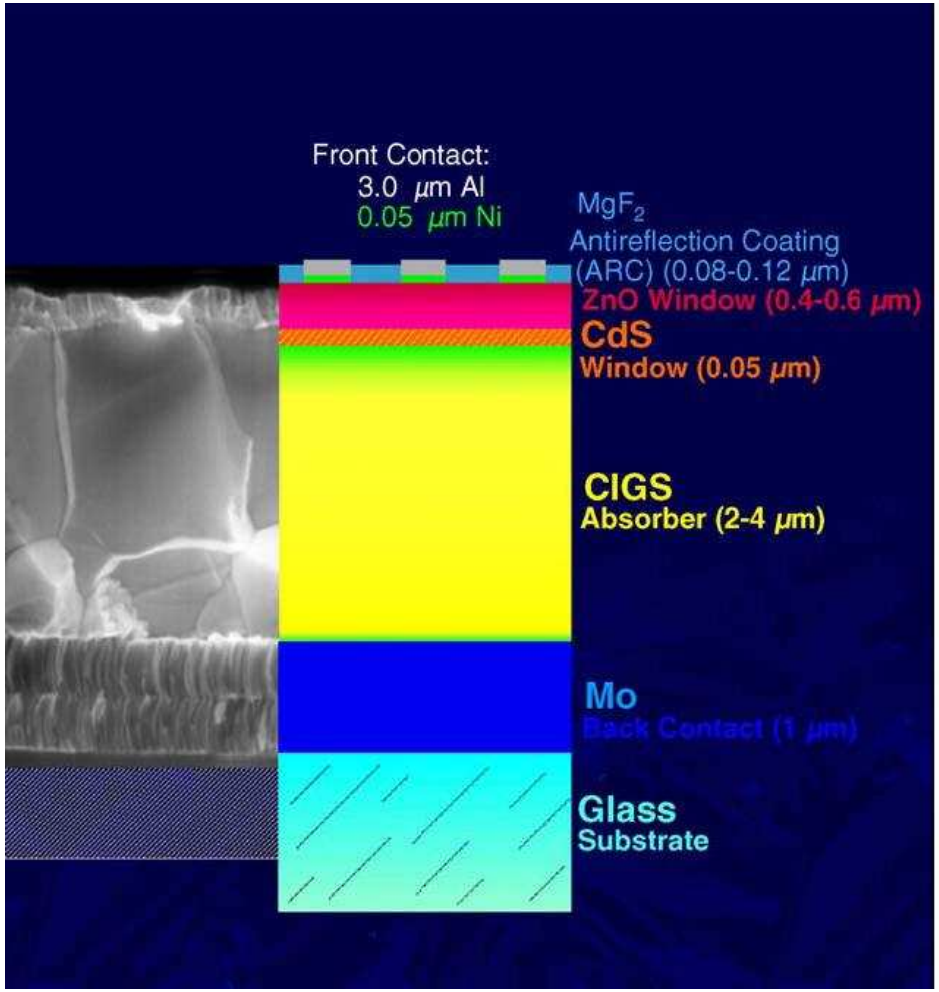
Motivation: thin film CIGS solar cell production

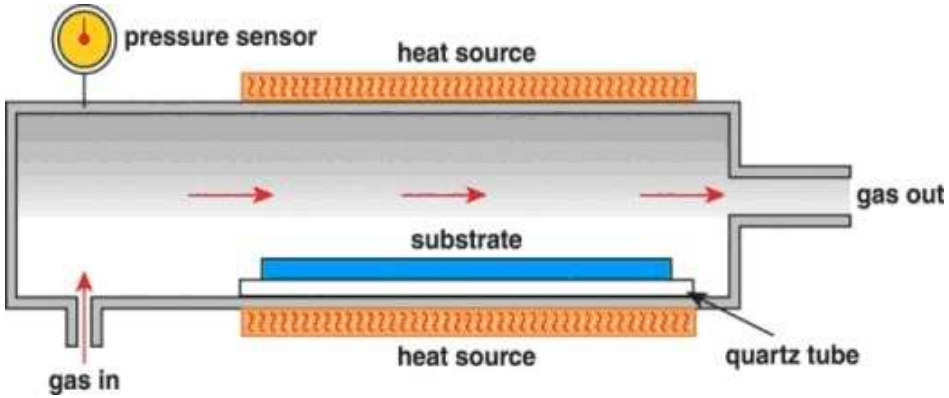
Collaboration with IPVF (Institut Photovoltaïque de France).

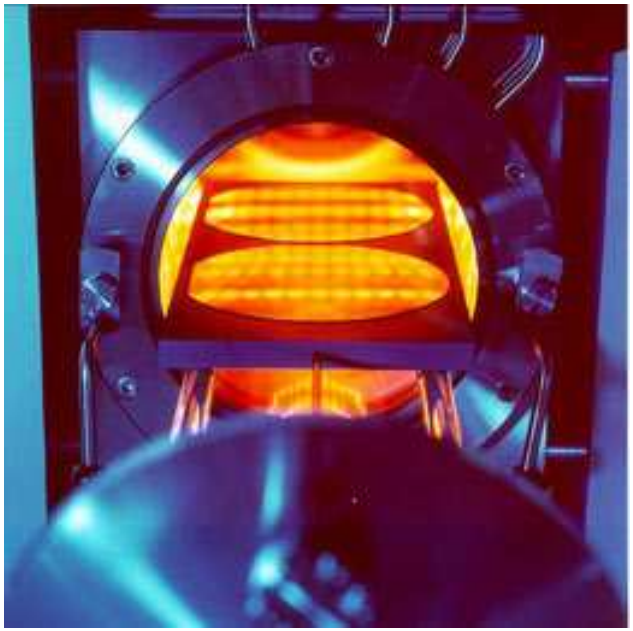
Optimal control of the production process of thin film CIGS (Copper, Indium, Gallium, Selenium) solar cell devices

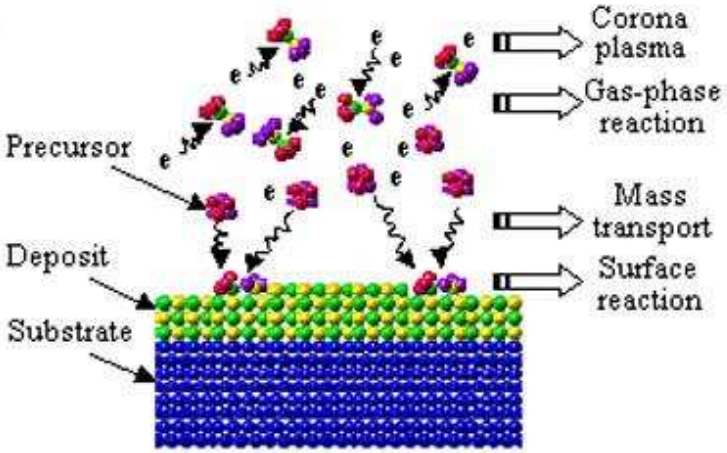
Typical composition of a CIGS solar cell



Production process: Physical Vapor Decomposition (PVD)







Modelisation

5 different chemical species: Cu, In, Ga, Se, Mo

Physical phenomena to take into account in the model:

- the evolution of the surface of the thin film surface during the fabrication process
- the diffusion of the different chemical species within the bulk of the film, due to the high temperature of the chamber

Talk of today: only the second point

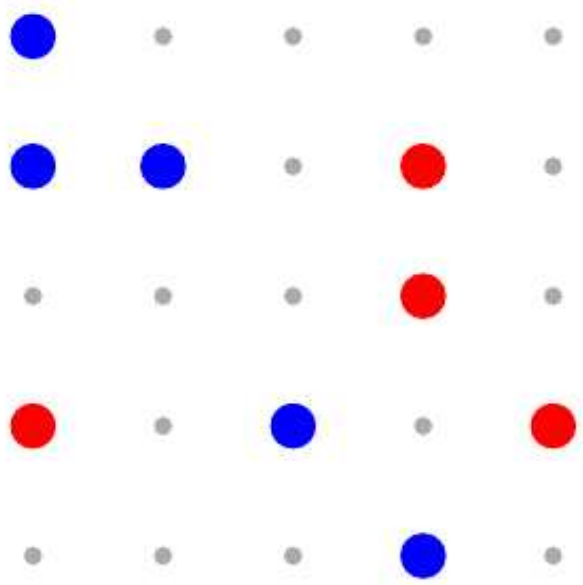
Modelisation on the atomistic scale

Multi-species symmetric exclusion process on a grid

Lattice-based stochastic hopping model

For the sake of simplicity:

- only two chemical species red and blue
- Atoms are clamped on an infinite periodic grid (sites of the crystalline solid): $\mathbb{Z}^d$



Multi-species symmetric exclusion process on a grid

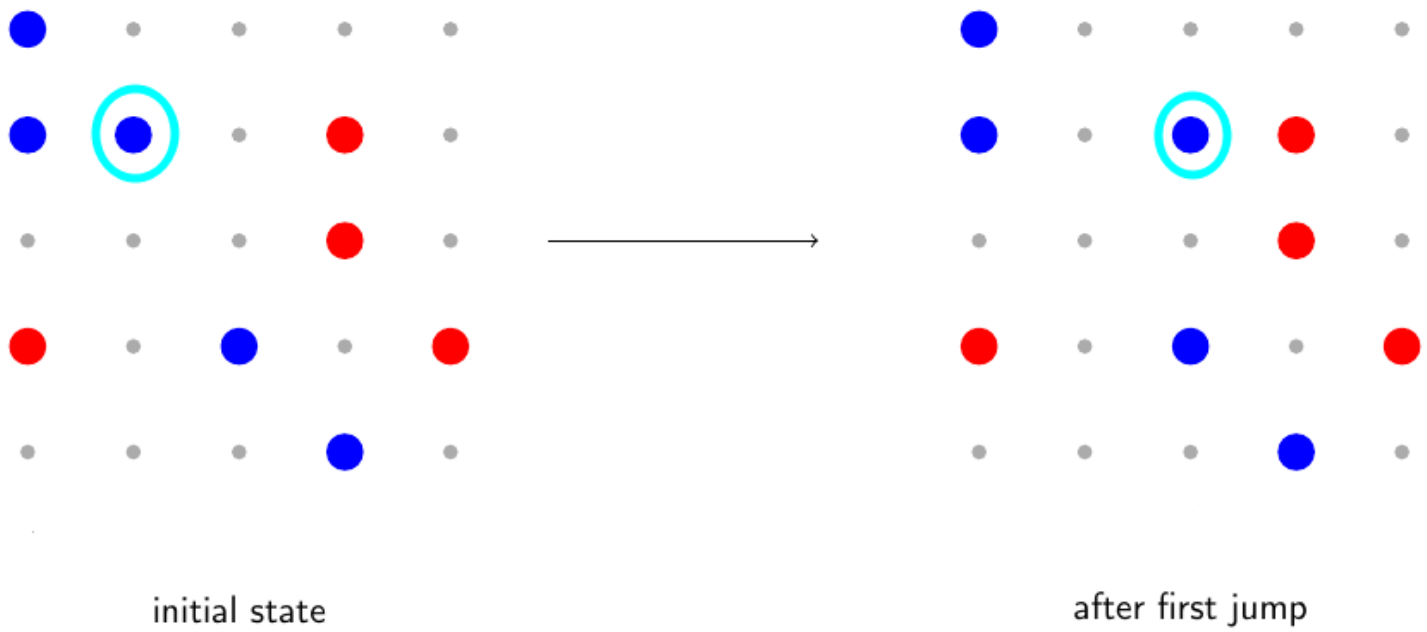
Lattice-based stochastic hopping model

- At some given random times (following an exponential law), one random particle may jump to a neighbouring site.
- Let $K \in \mathbb{N}^*$.
  Let $(\mathbf{v}_k)_{1 \leq k \leq K} \subset \mathbb{Z}^d \setminus \{0\}$ be the set of K possible jump directions of each particle.
  Let $(p_k)_{1 \leq k \leq K} \subset (0, 1)$ be the probability rate of each jump direction.
- Let us assume that the selected random particle is located at the site $\mathbf{x} \in \mathbb{Z}^d$. A jump direction $\mathbf{v}_k$ is randomly selected with probability p_k.
  - If the site located at position $\mathbf{x} + \mathbf{v}_k$ is already occupied by another particle, nothing happens
  - Otherwise, the particle jumps at the new site located at $\mathbf{x} + \mathbf{v}_k$.
- Select another random time, another random particle...

Modelisation on the atomistic scale

Multi-species symmetric exclusion process on a grid

Lattice-based stochastic hopping model



Hydrodynamic limit: deterministic cross-diffusion system

[Quastel, 1992], [Erignoux, 2016]

Limit as

- the number of particles grows to infinity
- the distance between neighbouring sites decreases to 0

For the sake of simplicity, on the torus $\mathbb{T}^d := \mathbb{R}^d/\mathbb{Z}^d$ (periodic boundary conditions)

- $\rho_{\texttt{red}}(\mathbf{x}, t)$ density of red particles at time $t > 0$ and point $\mathbf{x} \in \mathbb{T}^d$
- $\rho_{\texttt{blue}}(\mathbf{x}, t)$ density of blue particles at time $t > 0$ and point $\mathbf{x} \in \mathbb{T}^d$

$$\frac{\partial}{\partial t}\begin{pmatrix}\rho_{\texttt{red}}\\ \rho_{\texttt{blue}}\end{pmatrix} = \frac{1}{2}\nabla_{\mathbf{x}}\cdot\left[\begin{pmatrix}\frac{\rho_{\texttt{blue}}}{\rho}\mathbb{D}_s(\rho) + \frac{\rho_{\texttt{red}}}{\rho}\mathbb{D} & \frac{\rho_{\texttt{red}}}{\rho}(\mathbb{D} - \mathbb{D}_s(\rho))\\ \frac{\rho_{\texttt{blue}}}{\rho}(\mathbb{D} - \mathbb{D}_s(\rho)) & \frac{\rho_{\texttt{red}}}{\rho}\mathbb{D}_s(\rho) + \frac{\rho_{\texttt{blue}}}{\rho}\mathbb{D}\end{pmatrix}\begin{pmatrix}\nabla_{\mathbf{x}}\rho_{\texttt{red}}\\ \nabla_{\mathbf{x}}\rho_{\texttt{blue}}\end{pmatrix}\right]$$

where $\rho(\mathbf{x}, t) := \rho_{\texttt{red}}(\mathbf{x}, t) + \rho_{\texttt{blue}}(\mathbf{x}, t)$.

Definition of $\mathbb{D}$ and $\mathbb{D}_s(\rho)$

It holds that there exists a unique solution to the previous system provided that

$$\rho_{\texttt{red}}(\mathbf{x}, t=0) \geq 0,\ \rho_{\texttt{blue}}(\mathbf{x}, t=0) \geq 0,\ \rho(\mathbf{x}, t=0) = \rho_{\texttt{red}}(\mathbf{x}, t=0) + \rho_{\texttt{blue}}(\mathbf{x}, t=0) \leq$$

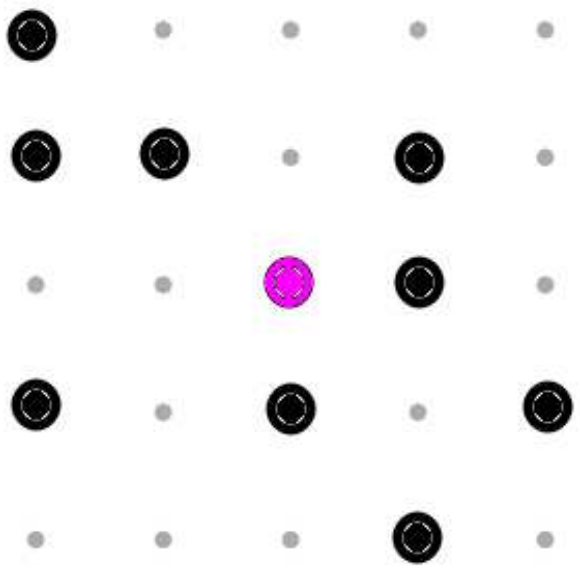
Then, it holds that for all $t > 0$,

$$\rho_{\texttt{red}}(\mathbf{x}, t) \geq 0,\ \rho_{\texttt{blue}}(\mathbf{x}, t) \geq 0,\ \rho(\mathbf{x}, t) = \rho_{\texttt{red}}(\mathbf{x}, t) + \rho_{\texttt{blue}}(\mathbf{x}, t) \leq 1.$$

- **Definition of $\mathbb{D} \in \mathbb{R}^{d \times d}$:** $\mathbb{D} := \sum_{k=1}^{K} p_k \mathbf{v}_k \mathbf{v}_k^T$
- **Definition of $\mathbb{D}_s(\rho) \in \mathbb{R}^{d \times d}$ for a given $\rho \in [0, 1]$:** The matrix $\mathbb{D}_s(\rho)$ is symmetric, non-negative and called the **auto-diffusion matrix** of the system.

Auto-diffusion matrix: probabilistic interpretation

Tagged particle process



- We consider a system of (identical) particles located on some sites of the lattice $\mathbb{Z}^d$
- At time $t = 0$:
  - there is one particle at the site 0: the tagged particle
  - all the other sites of the grid are independently occupied with a probability ρ
- the system of particles evolves following the same jumping rules as the original blue/red particle system

For all $t > 0$, let $\mathbf{w}_t \in \mathbb{Z}^d$ be the position at time t of the tagged particle.

Auto-diffusion matrix: probabilistic interpretation

For all $\mathbf{u} \in \mathbb{R}^d$,

$$\mathbf{u}^T \mathbb{D}_s(\rho)\mathbf{u} = 2 \lim_{t \to +\infty} \frac{\mathbb{E}\left[(\mathbf{u} \cdot \mathbf{w}_t)^2\right]}{t}$$

Remark: Since $\mathbb{D}_s(\rho)$ is symmetric, by polarization, it is sufficient to know the value of

$$\mathbf{u}^T \mathbb{D}_s(\rho)\mathbf{u}$$

for all $\mathbf{u} \in \mathbb{R}^d$ to know the value of the whole matrix $\mathbb{D}_s(\rho)$.

Computation in practice:

- Introduce a finite-size grid with periodic boundary conditions
  - Exponential convergence with respect to the size of the finite grid [Landim, Olla, Varadhan, 2022]
- Approximate the expectation $\mathbb{E}\left[(\mathbf{u} \cdot \mathbf{w}_t)^2\right]$ with Monte-Carlo simulations
  - Very high variance leads to **very large statistical noise**

Auto-diffusion matrix: optimization interpretation

For a given vector $\mathbf{u} \in \mathbb{R}^d$ and $\rho \in [0,1]$, the auto-diffusion coefficient $\mathbf{u}^T \mathbb{D}_s(\rho)\mathbf{u}$ can be equivalently expressed as the solution of a **high-dimensional optimization problem**.

- Let $S := \mathbb{Z}^d \setminus \{0\}$.
- An element $\boldsymbol{\eta} := (\eta_{\mathbf{s}})_{\mathbf{s}\in S} \in \{0,1\}^S$ denotes a possible environment around the tagged particle.
- Let $H := \left\{\Psi : \{0,1\}^S \to \mathbb{R}\right\}$ the set of real-valued functions defined on the set of possible environments

$$\mathbf{u}^T \mathbb{D}_s(\rho)\mathbf{u} := 2 \inf_{\Psi \in H} \mathbb{E}_\rho \left[J_\Psi(\boldsymbol{\eta})\right]$$

where

- $J_\Psi : \{0,1\}^S \to \mathbb{R}$ (see next slide)
- $\mathbb{E}_\rho$ denotes the expectation on all possible environments $\boldsymbol{\eta}$ so that each site is independently occupied with probability ρ.

Auto-diffusion matrix: optimization interpretation

Let $\boldsymbol{\eta} := (\eta_{\mathbf{s}})_{\mathbf{s}\in S} \in \{0,1\}^S$

- For all $\mathbf{y} \neq \mathbf{z} \in S$, $\boldsymbol{\eta}^{\mathbf{y},\mathbf{z}} := (\eta_{\mathbf{s}}^{\mathbf{y},\mathbf{z}})_{\mathbf{s}\in S}$ is the element of $\{0,1\}^S$ such that

$$\eta_{\mathbf{s}}^{\mathbf{y},\mathbf{z}} := \begin{cases} \eta_{\mathbf{s}} & \text{if } \mathbf{s} \neq \mathbf{y}, \mathbf{z}, \\ \eta_{\mathbf{y}} & \text{if } \mathbf{s} = \mathbf{z}, \\ \eta_{\mathbf{z}} & \text{if } \mathbf{s} = \mathbf{y}. \end{cases}$$

- For all $\mathbf{w} \in S$, $\boldsymbol{\eta}^{\mathbf{0},\mathbf{w}} := (\eta_{\mathbf{s}}^{\mathbf{0},\mathbf{w}})_{\mathbf{s}\in S}$ is the element of $\{0,1\}^S$ such that

$$\eta_{\mathbf{s}}^{\mathbf{0},\mathbf{w}} := \begin{cases} \eta_{\mathbf{s}+\mathbf{w}} & \text{if } \mathbf{s} \neq -\mathbf{w}, \\ 0 & \text{if } \mathbf{s} = -\mathbf{w}. \end{cases}$$

$$\begin{aligned} J_{\Psi}(\boldsymbol{\eta}) \quad = \quad & \sum_{k=1}^{K} p_k \Bigg[(1-\eta_{\mathbf{v}_k}) \Big(\mathbf{u}\cdot\mathbf{v}_k + \Psi(\boldsymbol{\eta}^{\mathbf{0},\mathbf{v}_k}) - \Psi(\boldsymbol{\eta}) \Big)^2 \\ & + \frac{1}{2} \sum_{\substack{\mathbf{y}\in S \\ \mathbf{y}+\mathbf{v}_k \neq \mathbf{0}}} \Big(\Psi(\boldsymbol{\eta}^{\mathbf{y}+\mathbf{v}_k,\mathbf{y}}) - \Psi(\boldsymbol{\eta}) \Big)^2 \Bigg] \end{aligned}$$

Deterministic interpretation: finite-size grid approximation

Computation in practice:

- Introduce a finite-size grid with periodic boundary conditions

Exponential convergence with respect to the size of the finite grid [Landim, Olla, Varadhan, 2022]

Let $M \in \mathbb{N}^*$ and $S_M := \{-M, \cdots, M\}^d \setminus \{0\}$ so that $|S_M| = (2M+1)^d - 1$.
Let $H_M := \left\{\Psi : \{0,1\}^{S_M} \to \mathbb{R}\right\}$

For all $\mathbf{u} \in \mathbb{R}^d$ and $\rho = \frac{\ell}{|S_M|}$ with $\ell \in \{0, 1, \cdots, |S_M|\}$,

$$\mathbf{u}^T \mathbb{D}_s(\rho)\mathbf{u} \approx \mathbf{u}^T \mathbb{D}_s^M(\ell)\mathbf{u} := 2 \inf_{\Psi \in H_M} \sum_{\substack{\boldsymbol{\eta} := (\eta_{\mathbf{s}})_{\mathbf{s} \in S_M} \in \{0,1\}^{S_M} \\ \sum_{\mathbf{s} \in S_M} \eta_{\mathbf{s}} = \ell}} J_\Psi(\boldsymbol{\eta})$$

High-dimensional optimization problem $\hookrightarrow$ Curse of dimensionality

$$H_M \approx (\mathbb{R}^2)^{S_M}$$

Numerical approach investigated here

Tensor methods + greedy algorithms

First step: reformulation of the optimization problem

Let us introduce $\boldsymbol{\Psi}^{\mathrm{opt}} \in H_M$ solution to

$$\boldsymbol{\Psi}^{\mathrm{opt}} := \underset{\boldsymbol{\Psi} \in H_M}{\mathrm{argmin}} \sum_{\boldsymbol{\eta} := (\eta_{\mathbf{s}})_{\mathbf{s} \in S_M} \in \{0,1\}^{S_M}} J_{\boldsymbol{\Psi}}(\boldsymbol{\eta})$$

Then, for all $\ell \in \{0, 1, \cdots, |S_M|\}$,

$$\mathbf{u}^T \mathbb{D}_s^M(\ell)\mathbf{u} = 2 \sum_{\substack{\boldsymbol{\eta} := (\eta_{\mathbf{s}})_{\mathbf{s} \in S_M} \in \{0,1\}^{S_M} \\ \sum_{\mathbf{s} \in S_M} \eta_{\mathbf{s}} = \ell}} J_{\boldsymbol{\Psi}^{\mathrm{opt}}}(\boldsymbol{\eta})$$

We are going to present a numerical approach in order to obtain a numerical approximation of $\boldsymbol{\Psi}^{\mathrm{opt}}$ which circumvents the curse of dimensionality

Second step: definition of a pure rank-1 tensor

For all $\mathbf{s} \in S_M$, let $R^{\mathbf{s}} : \{0,1\} \to \mathbb{R}$ **Remark:** $R^{\mathbf{s}} \approx \mathbb{R}^2$

The pure rank-1 tensor associated to the family $(R^{\mathbf{s}})_{\mathbf{s}\in S_M}$ is the function $\otimes_{\mathbf{s}\in S_M} R^{\mathbf{s}} \in H_M$ defined as follows:

for all $\boldsymbol{\eta} = (\eta_{\mathbf{s}})_{\mathbf{s}\in S_M}$,

$$\left(\otimes_{\mathbf{s}\in S_M} R^{\mathbf{s}}\right)(\boldsymbol{\eta}) = \Pi_{\mathbf{s}\in S_M} R^{\mathbf{s}}(\eta_{\mathbf{s}})$$

Note that the knowledge of $\otimes_{\mathbf{s}\in S_M} R^{\mathbf{s}}$ only requires the knowledge of $|S_M|$ vectors of $\mathbb{R}^2$.

We are going to compute a numerical approximation of Ψ^{opt} under the form

$$\Psi^{\mathrm{opt}} \approx \Psi_n := \sum_{k=1}^{n} \otimes_{\mathbf{s}\in S_M} R_k^{\mathbf{s}}$$

with n small and where for all $\mathbf{s}_{\in} S_M$ and all $1 \leq k \leq n$, $R_k^{\mathbf{s}} : \{0,1\} \to \mathbb{R}$

The number of terms n is then called the **rank** of the tensor approximation of Ψ^{opt}

Third step: greedy algorithm

Let us denote by $\mathcal{E} : H_M \to \mathbb{R}$ the (quadratic) functional defined by

$$\forall \Psi \in H_M, \quad \mathcal{E}(\Psi) := \sum_{\boldsymbol{\eta} := (\eta_{\mathbf{s}})_{\mathbf{s} \in S_M} \in \{0,1\}^{S_M}} J_\Psi(\boldsymbol{\eta})$$

so that

$$\Psi^{\mathrm{opt}} = \operatorname*{argmin}_{\Psi \in H_M} \mathcal{E}(\Psi).$$

Greedy algorithm:

- **Initialization :** Start from $\Psi_0 = 0$
- **Iteration** $n \geq 1$**:** Select $(R_n^{\mathbf{s}})_{\mathbf{s} \in S_M}$ solution to

$$(R_n^{\mathbf{s}})_{\mathbf{s} \in S_M} \in \operatorname*{argmin}_{\substack{(R^{\mathbf{s}})_{\mathbf{s} \in S_M} \\ \forall \mathbf{s} \in S_M,\ R^{\mathbf{s}} : \{0,1\} \to \mathbb{R}}} \mathcal{E}\left(\Psi_{n-1} + \otimes_{\mathbf{s} \in S_M} R^{\mathbf{s}}\right)$$

Set $\Psi_n := \Psi_{n-1} + \otimes_{\mathbf{s} \in S_M} R_n^{\mathbf{s}}$.

Theoretical results on greedy algorithm for tensor approximation

Theorem (Cancès, VE, Lelièvre, 2011)

The sequence $(\Psi_n)_{n\in\mathbb{N}}$ *converges to* Ψ^{opt} *exponentially fast with respect to* n.

In practice? Fixed-point algorithm

$$(R_n^{\mathbf{s}})_{\mathbf{s}\in S_M} \in \underset{\substack{(R^{\mathbf{s}})_{\mathbf{s}\in S_M} \\ \forall \mathbf{s}\in S_M,\ R^{\mathbf{s}}:\{0,1\}\to\mathbb{R}}}{\operatorname{argmin}} \mathcal{E}\left(\Psi_{n-1} + \otimes_{\mathbf{s}\in S_M} R^{\mathbf{s}}\right)$$

Alternating Least Square (ALS) algorithm
Define some ordering of the sites of S_M so that

$$S_M = \{\mathbf{s}_1, \cdots, \mathbf{s}_{|S_M|}\}$$

Finding a family $(R^{\mathbf{s}})_{\mathbf{s}\in S_M}$ then amounts to finding a family $(R_n^{\mathbf{s}_p})_{1\le p\le |S_M|}$

$$\forall \boldsymbol{\eta} = (\eta_{\mathbf{s}_p})_{1\le p\le |S_M|}, \qquad \otimes_{\mathbf{s}\in S_M} R^{\mathbf{s}}(\boldsymbol{\eta}) = \Pi_{p=1}^{|S_M|} R^{\mathbf{s}_p}(\eta_{\mathbf{s}_p})$$

- **Initialization:** Start from $(R_{n,0}^{\mathbf{s}_p})_{1\le p\le |S_M|}$ which are randomly chosen
- **Iteration** $m \ge 1$**:** For all $1 \le p \le |S_M|$, find

$$R_{n,m}^{\mathbf{s}_p} = \underset{R:\{0,1\}\to\mathbb{R}}{\operatorname{argmin}}\ \mathcal{E}\left(\Psi_{n-1}(\boldsymbol{\eta}) + \Pi_{p'=1}^{p-1} R_{n,m}^{\mathbf{s}_p}(\eta_{\mathbf{s}_{p'}}) R(\eta_{\mathbf{s}_p}) \Pi_{p'=p+1}^{|S_M|} R_{n,m-1}^{\mathbf{s}_p}(\eta_{\mathbf{s}_{p'}})\right)$$

In practice? Fixed-point algorithm

Since $\mathcal{E}$ is quadratic, computing

$$R_{n,m}^{\mathbf{s}_p} = \underset{R:\{0,1\}\to\mathbb{R}}{\operatorname{argmin}} \ \mathcal{E}\left(\Psi_{n-1}(\boldsymbol{\eta}) + \Pi_{p'=1}^{p-1} R_{n,m}^{\mathbf{s}_p}(\eta_{\mathbf{s}_{p'}}) R(\eta_{\mathbf{s}_p}) \Pi_{p'=p+1}^{|S_M|} R_{n,m-1}^{\mathbf{s}_p}(\eta_{\mathbf{s}_{p'}})\right)$$

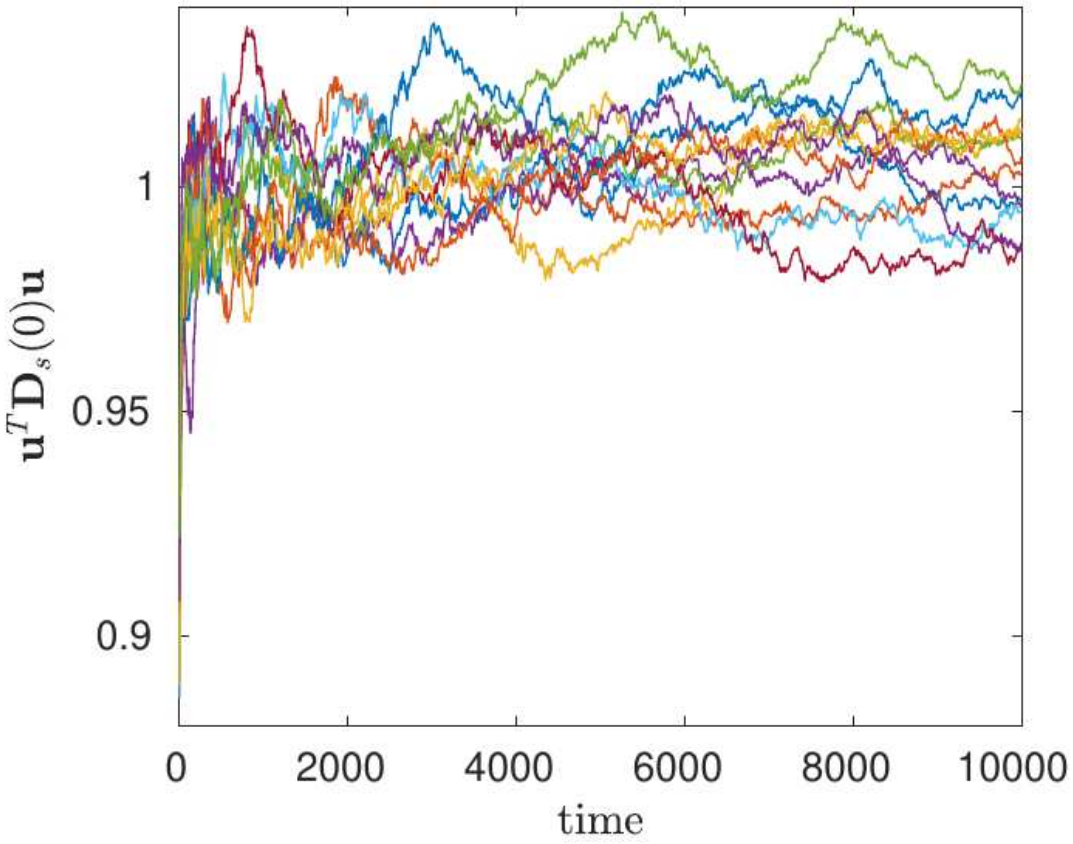
also amounts to solving a quadratic problem to find a two-dimensional vector.

Resolution of a two-dimensional linear problem

Because of the particular structure of the functional $\mathcal{E}$, the assembly of this two-dimensional linear problem can be done with complexity which evolves linearly with $n|S_M|$.

Variance of the naive Monte-Carlo method

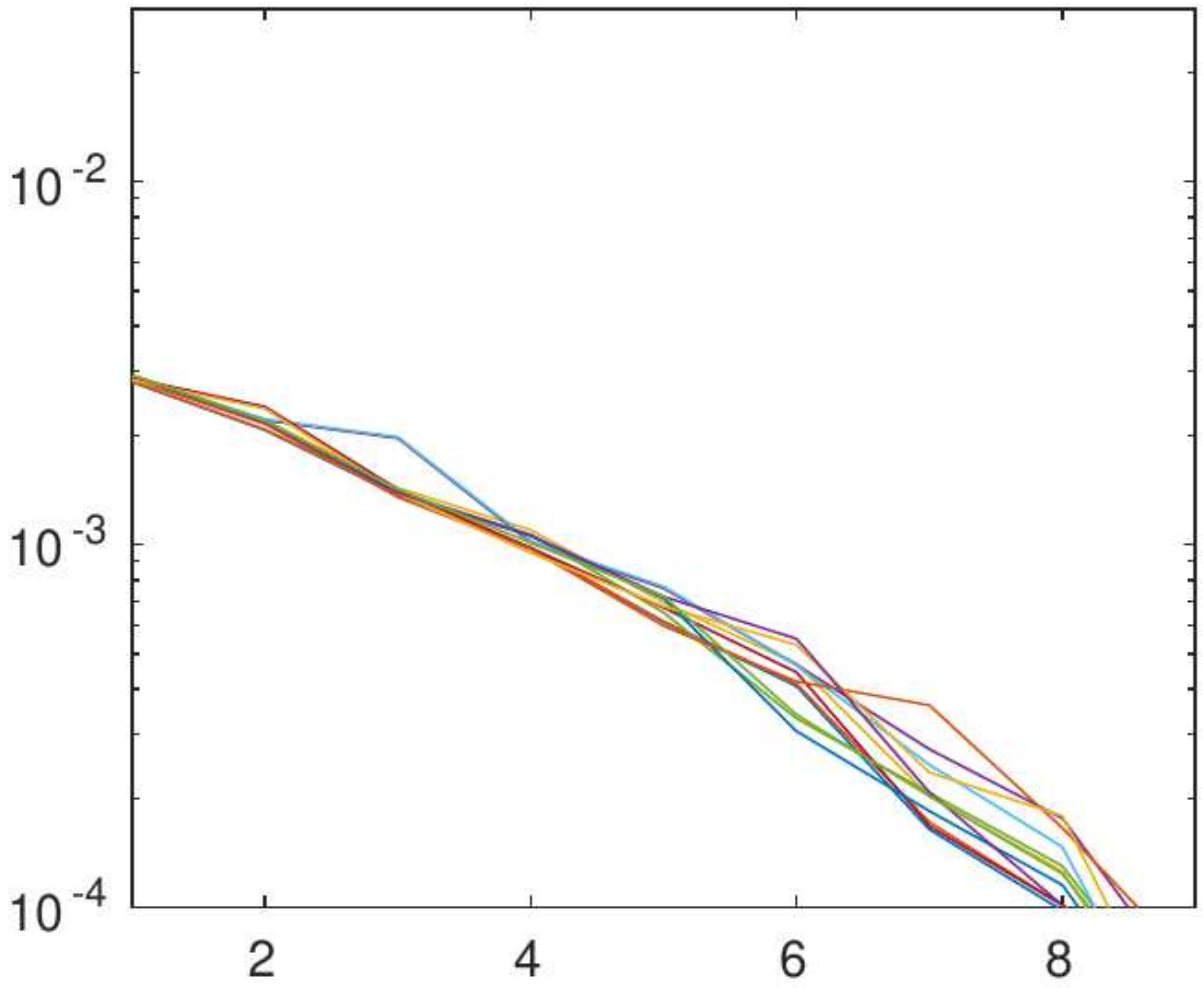
$$\mathbf{u}^T \mathbb{D}_s \left(\frac{\ell}{|S_M|} \right) \mathbf{u} = 2 \lim_{t \to +\infty} \frac{\mathbb{E}\left[(\mathbf{u} \cdot \mathbf{w}_t)^2 \right]}{t}$$



$d = 2$, $M = 2$, $\mathbf{u} = (1, 0)$, $\ell = 0$

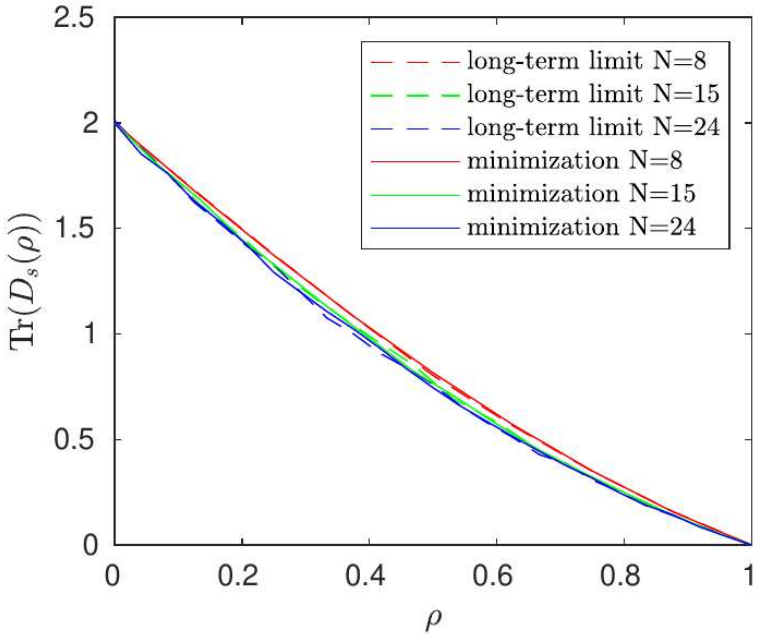
Convergence as of the greedy algorithm

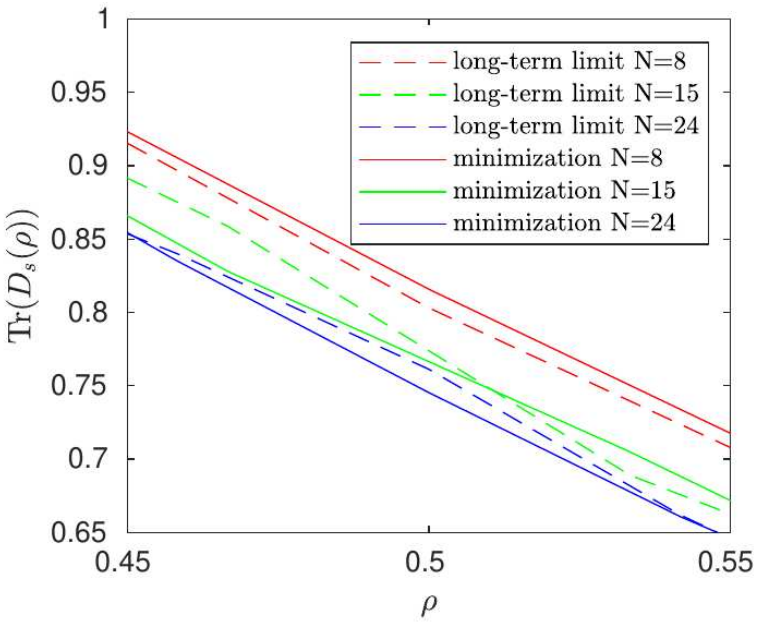
Convergence of $\mathcal{E}(\Psi_n)$ to $\mathcal{E}(\Psi^{\mathrm{opt}})$ as a function of n



Numerical results

$n = 10$





- Tensor method: 3 minutes
- Monte-Carlo method: 15 minutes

What next?

Perspectives:

- Couple this tensor/greedy approach together with domain decomposition method to solve larger problems
- Develop a finite-volume scheme in order to solve the original cross-diffusion system.

$$\frac{\partial}{\partial t}\begin{pmatrix}\rho_{\texttt{red}}\\ \rho_{\texttt{blue}}\end{pmatrix} = \frac{1}{2}\nabla_{\mathbf{x}}\cdot\left[\begin{pmatrix}\frac{\rho_{\texttt{blue}}}{\rho}\mathbb{D}_s(\rho)+\frac{\rho_{\texttt{red}}}{\rho}\mathbb{D} & \frac{\rho_{\texttt{red}}}{\rho}(\mathbb{D}-\mathbb{D}_s(\rho))\\ \frac{\rho_{\texttt{blue}}}{\rho}(\mathbb{D}-\mathbb{D}_s(\rho)) & \frac{\rho_{\texttt{red}}}{\rho}\mathbb{D}_s(\rho)+\frac{\rho_{\texttt{blue}}}{\rho}\mathbb{D}\end{pmatrix}\begin{pmatrix}\nabla_{\mathbf{x}}\rho_{\texttt{red}}\\ \nabla_{\mathbf{x}}\rho_{\texttt{blue}}\end{pmatrix}\right]$$

Jad Dabaghi, Virginie Ehrlacher, Christoph Strössner, Tensor approximation of the self-diffusion matrix of tagged particle processes, 2022, arXiv:2204.03943

CEMRACS 2022 project: tensor/greedy approach for Mean-Field Games systems

Crowd motion applications: [Achdou, Laurière, 2020]

$$\begin{cases} \partial_t u - \nu \Delta u + H(x, m, \nabla u) = 0 \\ \partial_t m - \nu \Delta m - \operatorname{div}(m H_p(x, m, \nabla u)) = 0 \\ m(0, x) = m_0(x) \\ u(T, x) = \phi(x, m(T, x)) \end{cases}$$

with Luca Nenna, Laïla Baroukh, Damien Prel and Léopold Trémant.

More on Thursday!

Thank you for your attention!