Vacancy elastodiffusion around cavities in aluminium: Fast First passage algorithms based on Krylov subspace projection techniques

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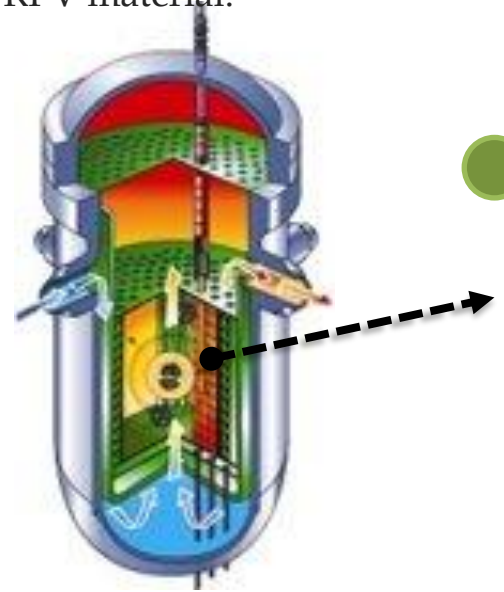
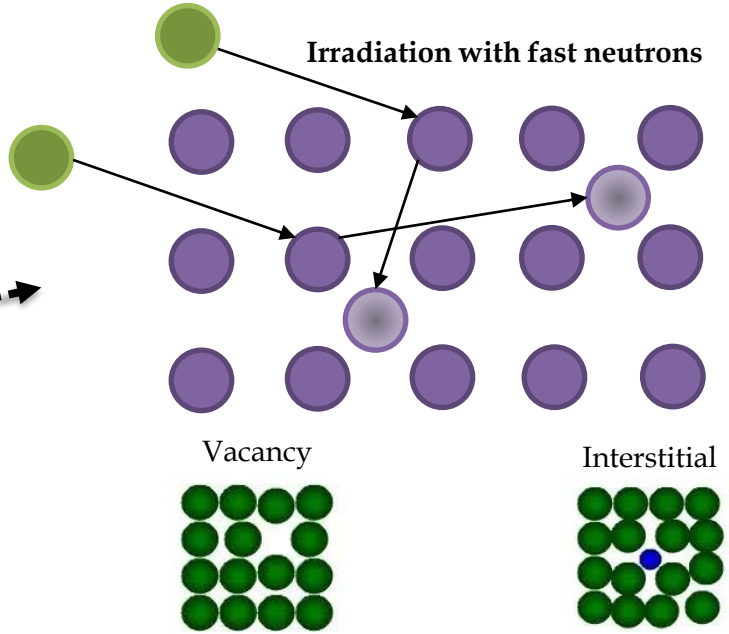
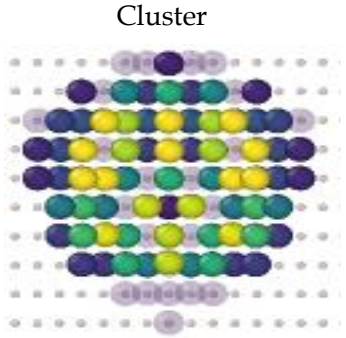
NEUTRON IRRADIATION

- **Pressurized Water Reactor(s)** are subjected to neutron irradiation which introduces vacancies and interstitials.
- Over time, these defects form vacancy clusters and interstitials loops. Affects the mechanical properties of RPV material.



Schematic view of PWR





Damage in the material's structure

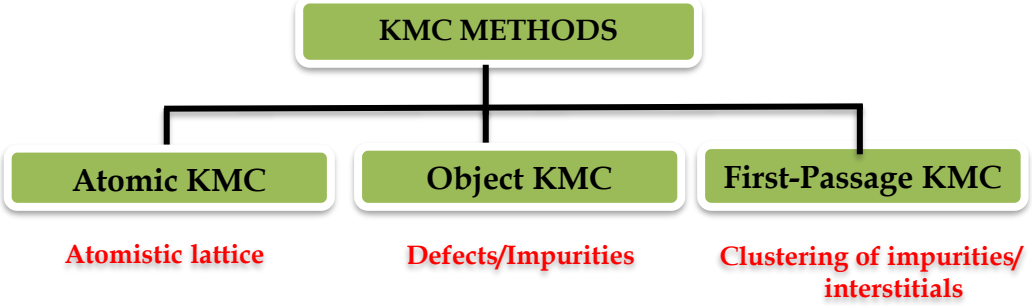
Aim: To simulate the microstructural evolution occurring in irradiated steels using KMC simulations

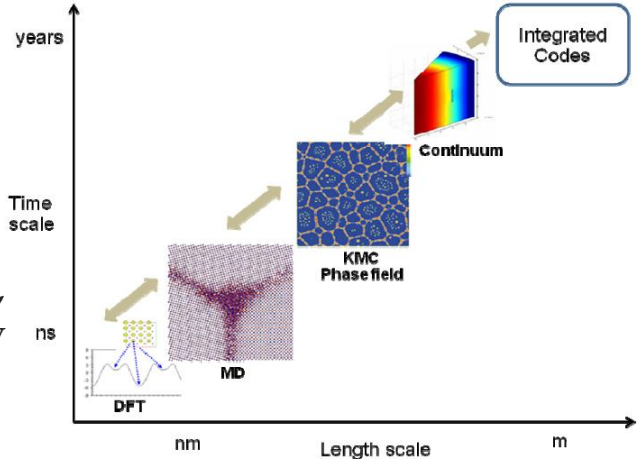
PRESENTATION OUTLINE

1. Introduction to Absorbing Markov Chains
2. Eigenvalue subspace model projection (ESMP)
3. Model Order Reduction: Krylov subspace projection methods
4. General Conclusions and Tasks ahead

PHYSICAL BACKGROUND: KINETIC MONTE CARLO SIMULATIONS

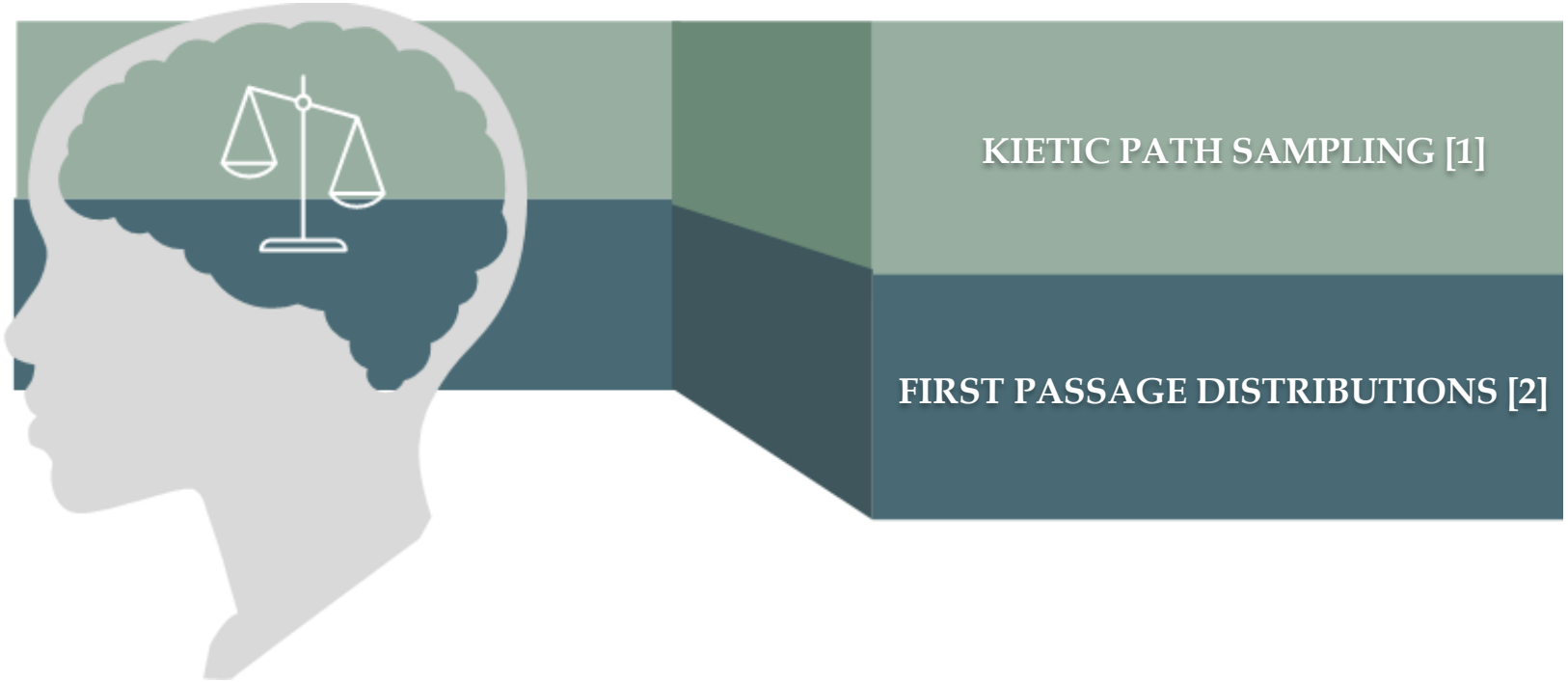
- **KMC Simulations** are used to study the early stage of micro-structural evolution at equilibrium or non equilibrium steady state, ranging from hours to days depending on temperature.





- Governed by the Master equation – time evolution of a system, probability of the states at any given time, interchanging states by transition rate matrix
- Efficiency reduces – transition rate matrix exhibits wide spectrum
- System transits huge number of times between configurations separated by small energy barriers – forms trapping basins
- Average escape time >> Characteristic time
- Recurrent issue in KMC simulations
- Acceleration techniques ways to improve KMC simulations

PHYSICAL BACKGROUND:
KINETIC MONTE CARLO SIMULATIONS



[1] Path Factorization Approach to Stochastic Simulations, M.Athènes, and V.Bulatov, *Phys. Rev. Lett*. **113, 230601 (2014**).
[2] Elastodiffusion and cluster mobilities using kinetic Monte Carlo simulations: Fast first-passage algorithms for reversible diffusion processes, Manuel Athènes, Savneet Kaur, Gilles Adjanor, Thomas Vanacker, and Thomas Jourdan, *Phys. Rev. Materials* **3, 103802 (2019)**.

PHYSICAL BACKGROUND: ABSORBING MARKOV PROCESS

- System to be in a transient state, and absorbing states are not considered yet.
- Master equation: $\dot{\boldsymbol{p}}_{\boldsymbol{t}}^{\boldsymbol{T}} = \boldsymbol{p}_{\boldsymbol{t}}^{\boldsymbol{T}} \boldsymbol{K}^{\boldsymbol{a}}$
- Evolution Operator: $\boldsymbol{P}(\boldsymbol{t_0}, \boldsymbol{t_1}) = \exp\left[\int_{t_0}^{t_1} \boldsymbol{K} dt\right] = \exp[(\boldsymbol{t_1} - \boldsymbol{t_0})\boldsymbol{K}]$
- A transition rate matrix is defined as $\boldsymbol{K}^{\boldsymbol{a}}$ associated with absorbing Markov chains.

$$\boldsymbol{K}^{\boldsymbol{a}} = \begin{pmatrix} -\boldsymbol{A} & \boldsymbol{A}\vec{\boldsymbol{1}} \\ \vec{\boldsymbol{0}}^{\boldsymbol{T}} & \boldsymbol{0} \end{pmatrix}$$

where $\boldsymbol{A}$ is N x N matrix, vector $\vec{0} = (0, \ldots, 0)^T$ and $\vec{\boldsymbol{1}}$ is column vector.

- Probability of a walker in absorbing states is conserved over time. Expressing evolution operator by using matrix –A gives the probability of being in state j ≤ N from state i ≤ N at time t is,

$$\boldsymbol{P}_{\boldsymbol{ij}}^{\boldsymbol{a}}(\boldsymbol{t}) = \boldsymbol{e}_{\boldsymbol{i}}^{\boldsymbol{t}} \exp(-\boldsymbol{A}\boldsymbol{t})\boldsymbol{e}_{\boldsymbol{j}}$$

- Mean First Passage time : $\boldsymbol{\tau}_{\boldsymbol{i}} = \int_0^\infty \boldsymbol{t} \frac{\boldsymbol{d}}{\boldsymbol{dt}} \boldsymbol{P}_{\boldsymbol{i}}^{\boldsymbol{a}}(\boldsymbol{t}) \boldsymbol{dt} = \int_0^\infty \boldsymbol{e}_{\boldsymbol{i}}^{\boldsymbol{T}} \exp(-\boldsymbol{A}\boldsymbol{t}) \vec{\boldsymbol{1}} \boldsymbol{dt} = \boldsymbol{e}_{\boldsymbol{i}}^{\boldsymbol{T}} \boldsymbol{A}^{-\boldsymbol{1}} \vec{\boldsymbol{1}}$$

PHYSICAL BACKGROUND: REVERSIBILITY OF MARKOV CHAIN

- Markov process obeys detailed balance

$$\rho_i K_{ij} = \rho_j K_{ji}$$

- Also, considering matrix to be symmetric positive definite

$$\sqrt{\rho_i} A_{ij} / \rho_j = \sqrt{\rho_j} A_{ji} / \rho_i$$
$$\Longrightarrow A^I_{ij} = \sqrt{\rho_i} A_{ij} / \rho_j = A^I_{ji}$$
$$\Longrightarrow \boldsymbol{A = R^{-1} A R = (R)^{-1} A (R)}$$

Where $\boldsymbol{B}$ is a preconditioner = 1, $\boldsymbol{R}$ are the $N \; x \; N$ diagonal matrix

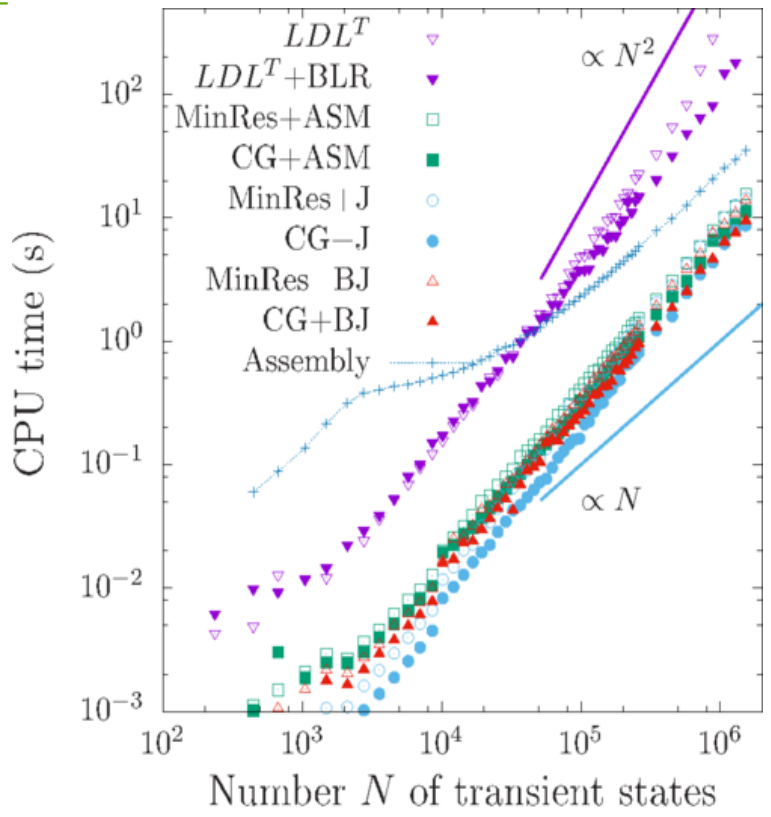
- Linear system problem, Ax=b
- This method can also be as linearized KMC, preserving the spectral properties
- Also $\boldsymbol{R}$ makes diagonal similarity transformation, results in generalized symmetric eigenvalue problem. That will result in all eigenvalues to be positive.

NUMERICAL RESULTS AND ANALYSIS: COMPARISON OF LINEAR SOLVER WITH DIRECT SOLVER

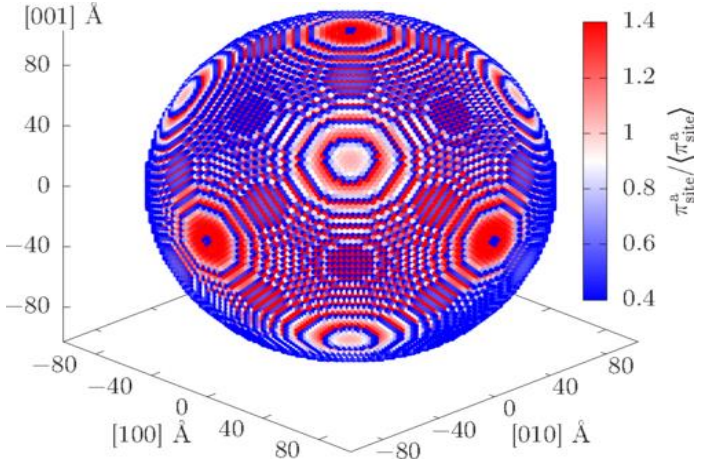
PETSC - Portable, Extensible Toolkit for Scientific Computations

Single vacancy emission from cavity	
Temperature	600K
Transient states	259320
Vacancies	2243
Radius for Cavity	20.07$\dot{A}$
Radius for protective sphere	101$\dot{A}$

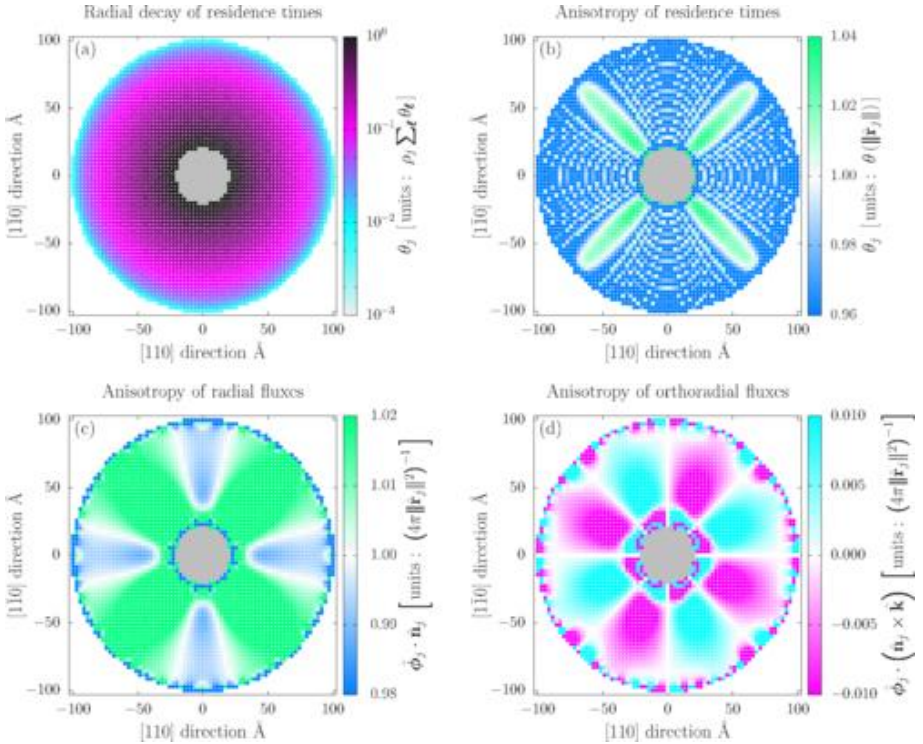
- ✓ Iterative solvers perform better than the direct solvers
- ✓ Conjugate Gradient (CG) is more efficient then LDL^T factorization.
- ✓ Sparsity decreased by a factor of 10 - 20



NUMERICAL RESULTS AND ANALYSIS: ILLUSTRATION ON VACANCY EMISSION FROM A CAVITY



Absorbing Probabilities: Anisotropy on peripheral sites



Residence time and fluxes associated with a single vacancy emission from a spherical cavity.

EIGENVALUE SUBSPACE MODEL PROJECTION (ESMP)

- Spectrum of matrix $\boldsymbol{A}$ can be evaluated using generalized symmetric eigenvalue problem (GSEP)

$$\boldsymbol{A^I \Phi = \Phi \Lambda}$$
$$\boldsymbol{\Longrightarrow A^I_{ij} = R \Phi \Lambda \Phi^T R^{-1}}$$
$$\boldsymbol{\Longrightarrow \exp[-At] = R \Phi \exp[\Lambda t] \Phi^T R^{-1}}$$

Where $\boldsymbol{\Lambda = \Lambda_{ii} = \lambda_i}$ eigenvalues and $\boldsymbol{\Phi = (\varphi_1, \varphi_2, \dots, \varphi_N)}$ orthonormal basis of eigenvectors.

- Introducing scaling and rescaling vectors, $\boldsymbol{r = R\vec{1}}$

$$\boldsymbol{\Longrightarrow g_i = e_i \odot r \;\&\; d_i = e_i \oslash r}$$
$$\boldsymbol{\Longrightarrow g_i^h = \varphi_h^T g_i \;\&\; d_j^h = \varphi_h^T d_j}$$

- The evolution operator is given by , $\boldsymbol{P_{ij}^a(t) = \sum_{h=1}^{N} g_i^h d_j^h \exp(-\lambda_h t)}$

FIRST PASSAGE PROBABILITIES AND DISTRIBUTIONS

- The survival probability after duration time t for the given system at state j started from state i is = probability of not having been absorbed/ probability in being N transient states,

$$\boldsymbol{S(t) \triangleq \sum_{j=1}^{N} P_{ij}^{a}(t) = \left(e_i^T, 0\right) exp[K^a t] \begin{pmatrix} \vec{1} \\ 0 \end{pmatrix} = e_i^T exp[-At]\vec{1}}$$

$$\boldsymbol{S_N(t) = S(t)}$$

$$\boldsymbol{\Rightarrow S_k\,(t) = \sum_{h=1}^{k} \alpha_h\, exp(-\lambda_h\, t)}$$

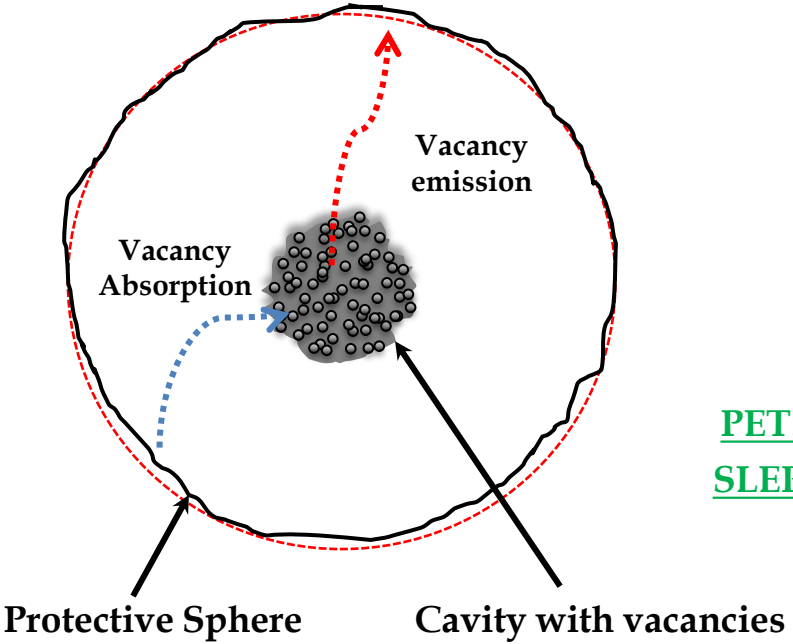
Where $\boldsymbol{\alpha_h = \left[\pi_0^T \varphi_h\right]\left[\vec{1}^T \varphi_h\right] = g_i^h \sum_{j=1}^{N} d_j^h}$ are the weighing coefficients.

- KMC simulations draw first passage times with appropriate statistics
- ***No Passage Distributions*** – The probability of system being in state j started from state i but the process is not absorbed
- ***Quasi Stationary Distribution*** – Limit of no passage distribution when time goes to infinity .

- And, if kinetics is governed by QSD, then $\boldsymbol{\langle t \rangle_{FP} = \sum_k \alpha_k/\lambda_k \approx 1/\lambda_1}$

NUMERICAL RESULTS AND ANALYSIS: SYSTEM PROPERTIES

- 1 to *N* transient states
- *N*+1 state in absorbing state, which acts like a sink trapping system
- Transient state ⇒ Absorbing state
- Transient state ⇍ Absorbing state



Cases	Single Vacancy Emission		Single Vacancy Absorption
Transient States	236	259320	34801
Cavity, R($\dot{A}$)	4.04	20.7	20.87
Protective, R($\dot{A}$)	10.1	101	-

PETSC - Portable, Extensible Toolkit for Scientific Computations

SLEPC - Scalable Library for Eigenvalues Problem Computations

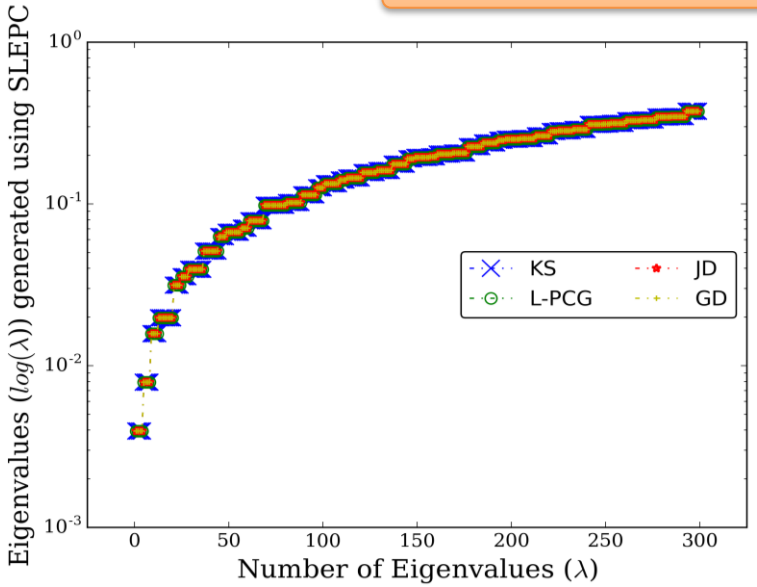
Eigenvalue problem , $\boldsymbol{Ax = \lambda x \Rightarrow A^{-1}x = \lambda^{-1}x}$

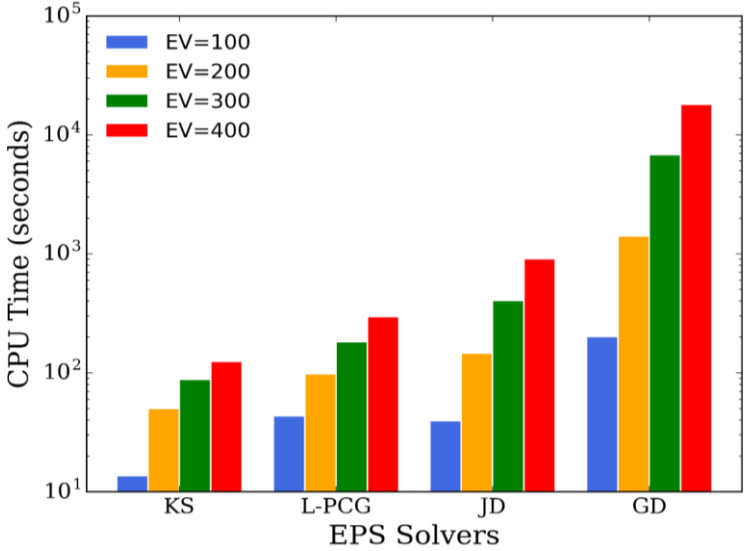
NUMERICAL RESULTS AND ANALYSIS: ITERATIVE SOLVERS

- ✓ Aim: To check with the speed up and efficiency of iterative solvers
- ✓ 2D Laplacian toy model with absorbing sink at the periphery, with periodic boundary conditions.
- ✓ Assumed value for K = 0.001 for absorption rate.

The evolution of parameters that are analyzed are:

- ➢ Eigenvalues & CPU time for the simulations
- ➢ Efficiency & Speed up

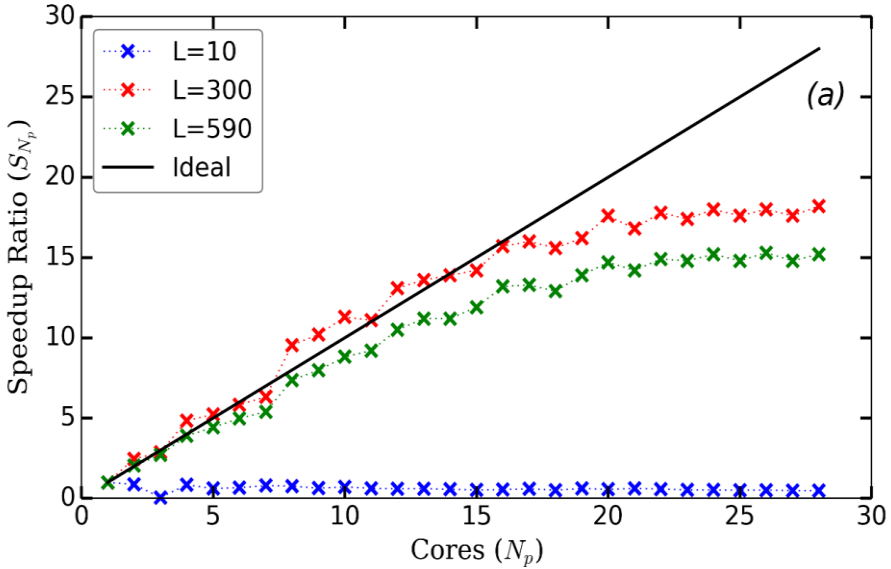




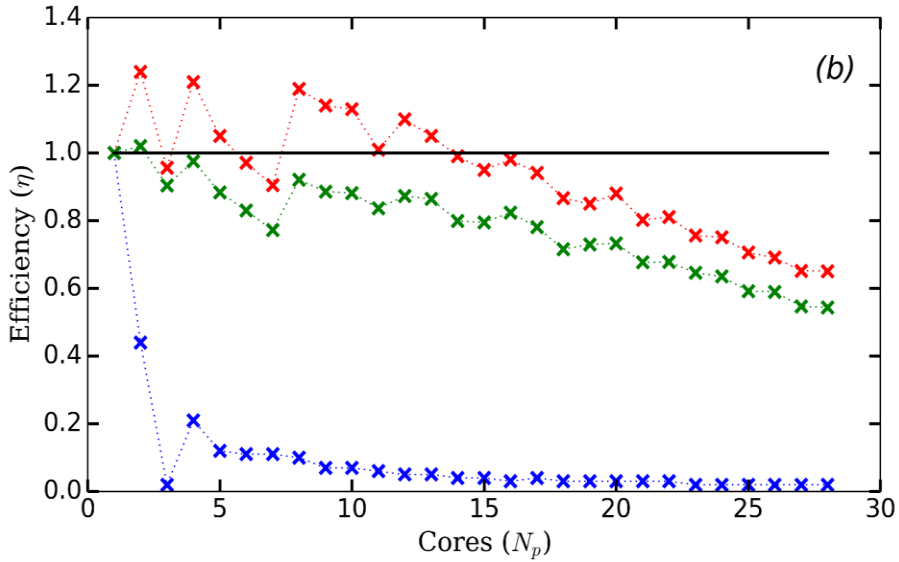
NUMERICAL RESULTS AND ANALYSIS: EFFICIENCY AND SPEEDUP

- ✓ Scalability of the solvers: simulations with MPI (upto 28 processors)

$$S_{N_p} = \frac{T(1)}{T(N_p)}$$



$$\eta = \frac{S_{N_p}}{N_p}$$

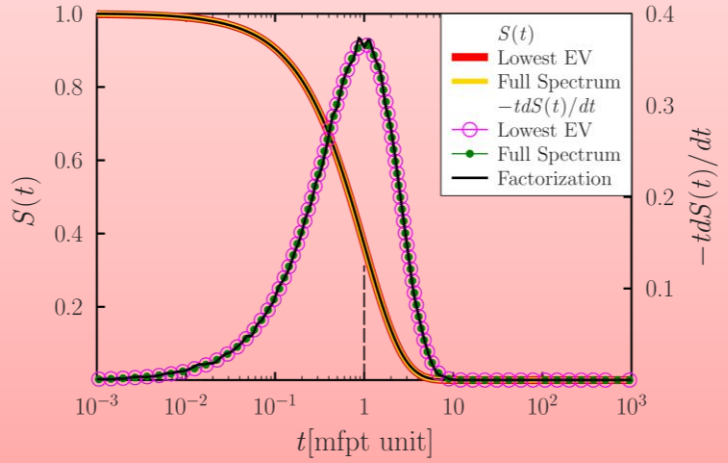


NUMERICAL RESULTS AND ANALYSIS: SINGLE VACANCY EMISSION

- ✓ Eigen solver scheme used : **KRYLOVSCHUR**
- ✓ Accounts for short time kinetics

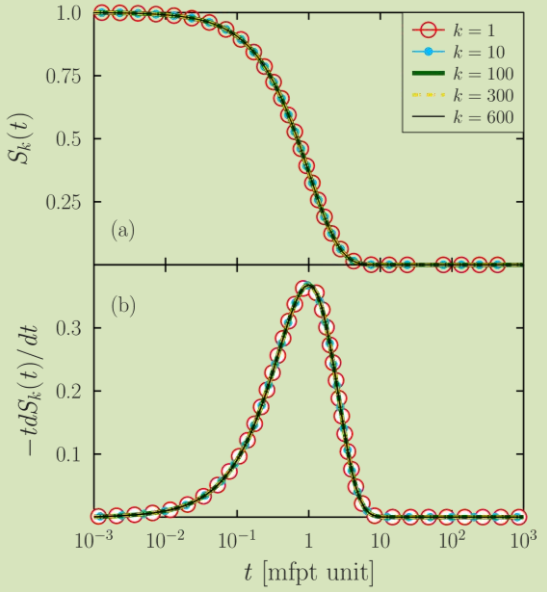
Case: Small system with 236 transient states

- ✓ CPU costs = 0.01689 seconds
- ✓ Early kinetics governed by quasi stationary distribution
- ✓ Comparison of first passage time with randomization approach (kinetic path sampling)



Case: Large system with 259320 transient states

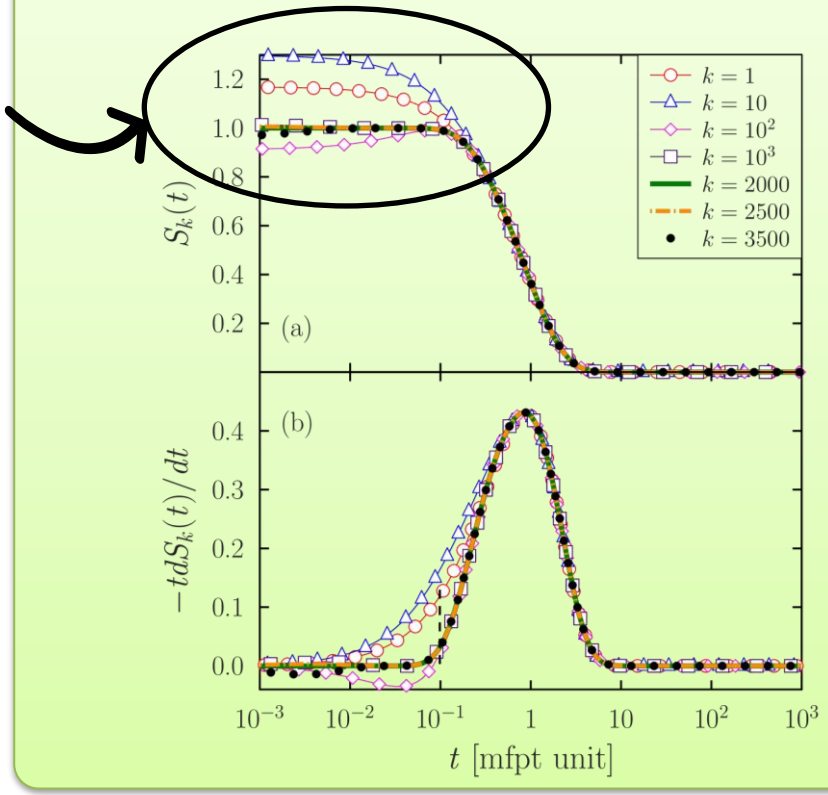
- ✓ CPU costs = 26554 seconds
- ✓ Early kinetics governed by quasi stationary distribution
- ✓ 0,25% eigen spectrum – accounts for QSD



NUMERICAL RESULTS AND ANALYSIS: SINGLE VACANCY ABSORPTION

- Trapping kinetics are governed by times greater than mean first passage times
- Predominance of substantial amount of eigenmodes - extracted 3500 eigenvalues
- CPU costs: 5002 seconds (28 Processors – parallel computation)
- 10% of the decaying exponentials need to be included for the early stage absorption kinetics!
- Error quantification: Truncated and reduced quantities, $\boldsymbol{\mathcal{T}_k}$ and $\boldsymbol{S_k(0)}$

Case: Artificial absorbing sink with 34801 transient states

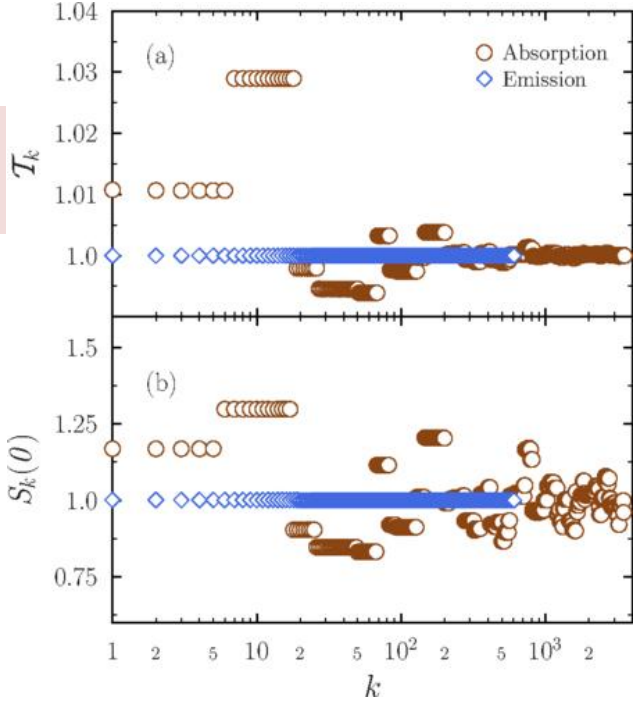


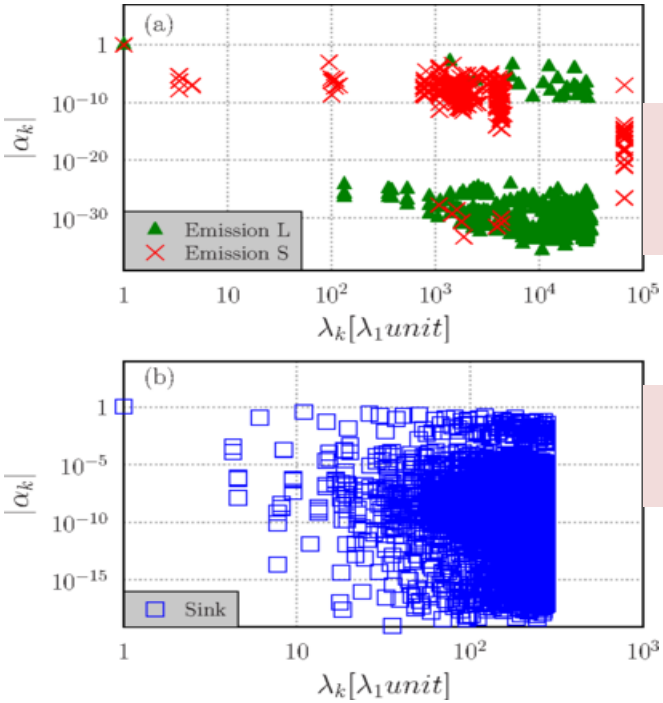
NUMERICAL RESULTS AND ANALYSIS: ERROR QUANTIFICATION

- Non monotonous convergence proceeds with a plateau
- Scatter plot of computed eigenvalues associated to its weighing coefficient
- Spectral gaps between λ_1 and λ_2
- Memory constraint for huge sparse matrices

$$\mathcal{T}_k = \frac{\sum_{h=1}^{k} \alpha_h/\lambda_h}{\sum_{h=1}^{N} \alpha_h/\lambda_h}$$

$$S_k(0) = \sum_{h=1}^{k} \alpha_h$$





$$\alpha_h = \sum_{h=1}^{k} \alpha_h$$

$$\lambda_k = \frac{\sum_{h=1}^{k} \lambda_h}{\lambda_1}$$

MODEL ORDER REDUCTION: RESTARTED KRYLOV SUBSPACE (R-KSP)

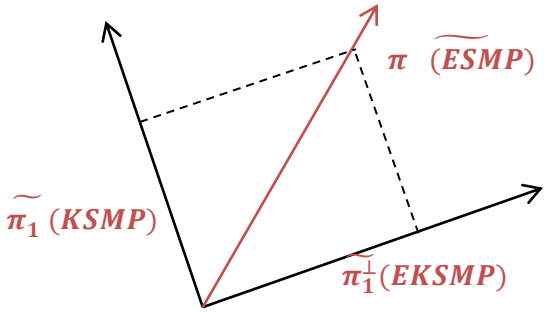
- ESMP is limited to high storage and computational requirements
- Method based on Krylov Subspaces:

$$\mathcal{K}_l(A, b) = \mathbf{span}(\mathbf{b}, \mathbf{Ab}, A^2 b, \dots, A^{l-1} b)$$

- Restarted Arnoldi method – Apply initial vector to the sparse matrix

$$y = f(Ab)$$

Where f is either inverse or exponential scaled function.



$$V_l^T A V_l = T_l \Rightarrow f_l = \beta V_l f(T_l)\, e_1$$

Krylov Subspace Model Projection (KSMP)

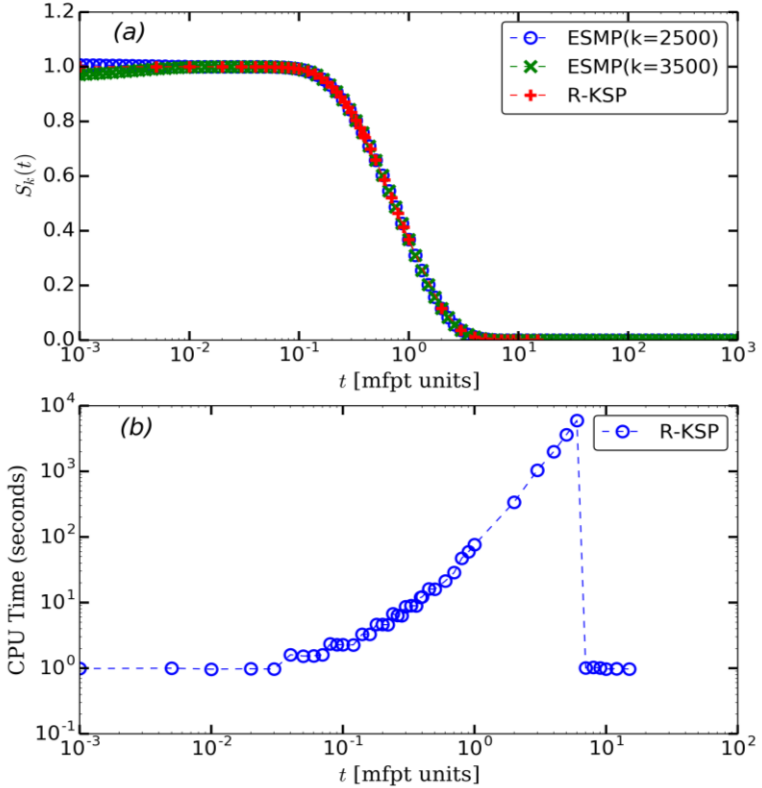
- ✓ Solves for $f_l = \beta V_l f(T_l)\, e_1$
  where T_l is the reduced tridiagonal matrix, constructed using krylov basis
  $$S_{k,l}(A, b) = \mathcal{K}_l(A, b - Pb)$$
- ✓ No contribution to the QSD
  $$S_{k,l}(A, b) = \widetilde{\alpha_1}\, e^{-\widetilde{\lambda_1} t}$$

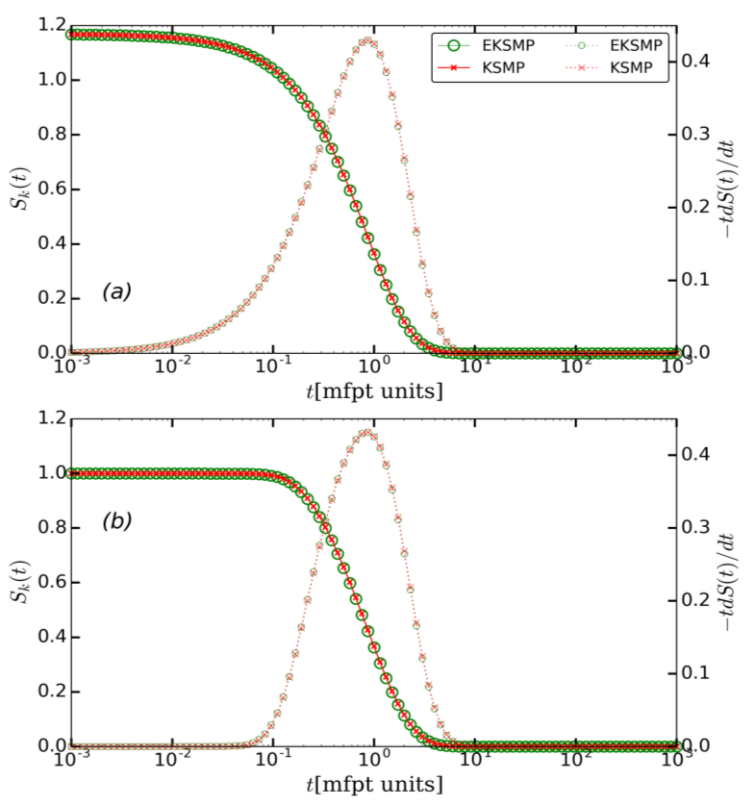
Eigenvector and Krylov Subspace Model Projection (EKSMP)

- ✓ Solves for
  $$S_{k,l}(A, b) = \mathcal{E}_k(A) \oplus \mathcal{K}_l(A, b - Pb)$$
  $\mathcal{E}_k(A)$ – Eigen subspace
  $\mathcal{K}_l(A, b - Pb)$– Orthogonal Krylov Space
- ✓ $S_{k,l}(A, b) = \underbrace{\alpha_1 e^{-\lambda_1 t}}_{\text{QSD}} \oplus \underbrace{\widetilde{\alpha_1}\, e^{-\widetilde{\lambda_1} t}}_{\text{Long time kinetics}}$

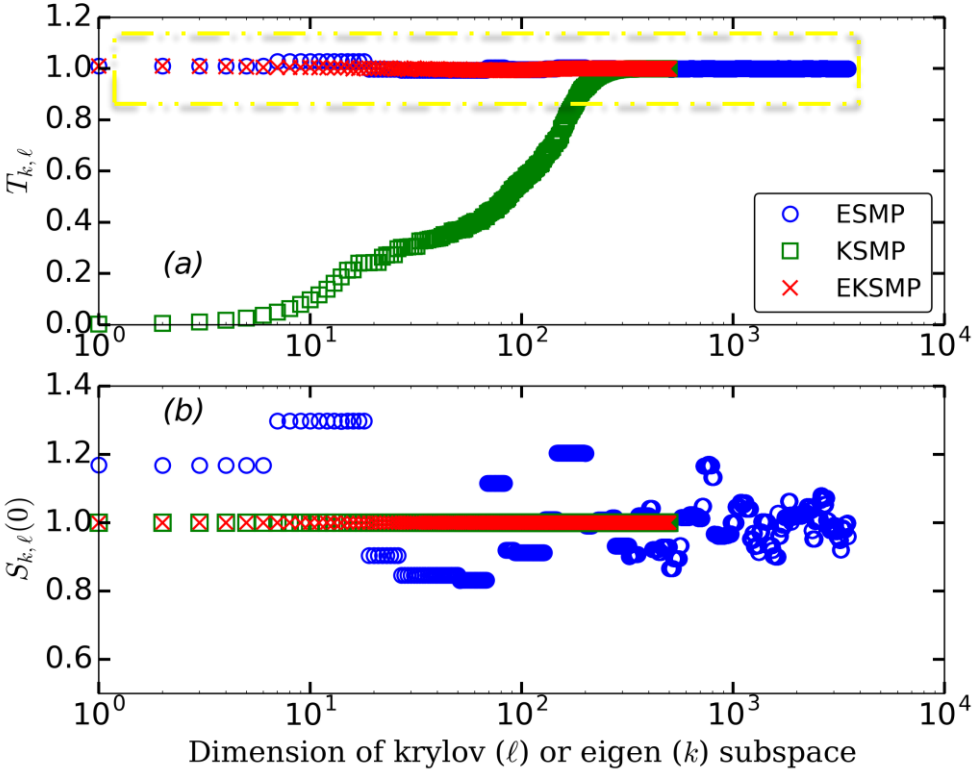
NUMERICAL RESULTS AND ANALYSIS: KRYLOV PROJECTION TECHNIQUES

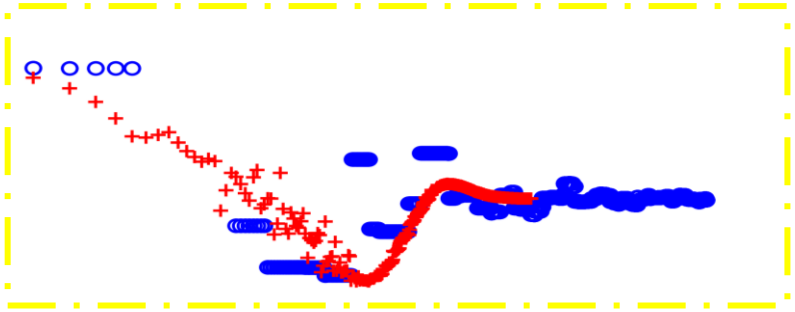
- ✓ Evaluated a Survival Probability distribution using R-KSP, ESMP, KSMP and EKSMP





NUMERICAL RESULTS AND ANALYSIS: COMPARISON BETWEEN THE METHODS





Method	Time (in seconds)
ESMP	5000 (28 procs)
KSMP	88.77
EKSMP	241.8

GENERAL CONCLUSIONS AND TASKS AHEAD

- Understanding of the diffusion process and absorbing Markov chains with reversibility
- Accelerating the KMC simulations by solving first-passage problems through partial factorization of transition matrix.
- Factorization is direct and invertible. It allows to generate first-passage times to escape trapping basins.
- Iterative Krylov subspace projection methods make it possible to extract the quasistationary distribution (QSD) and correctly approximate the untrapping events at short times.

? To calculate sink strengths and fully characterize recombination laws. Crucial information for simulations at larger scale (FPKMC, EKMC, RECD)

? To deal with larger trap sizes than with direct factorization

Thank You for your attention.

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