Kinetic Monte Carlo method for the acceleration of defect evolution simulation

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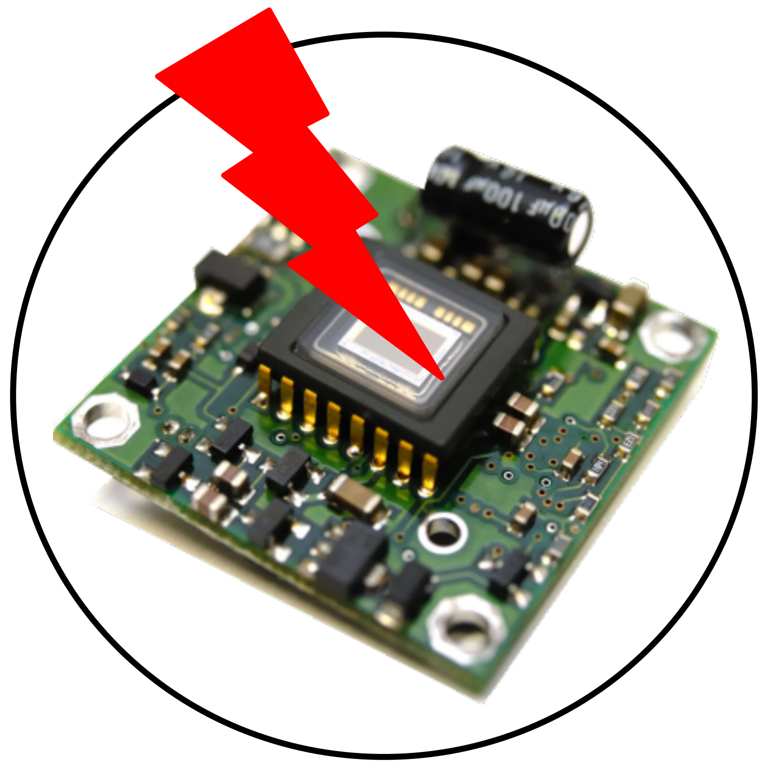
Kinetic Monte Carlo method for the acceleration of defect evolution simulation

Target:

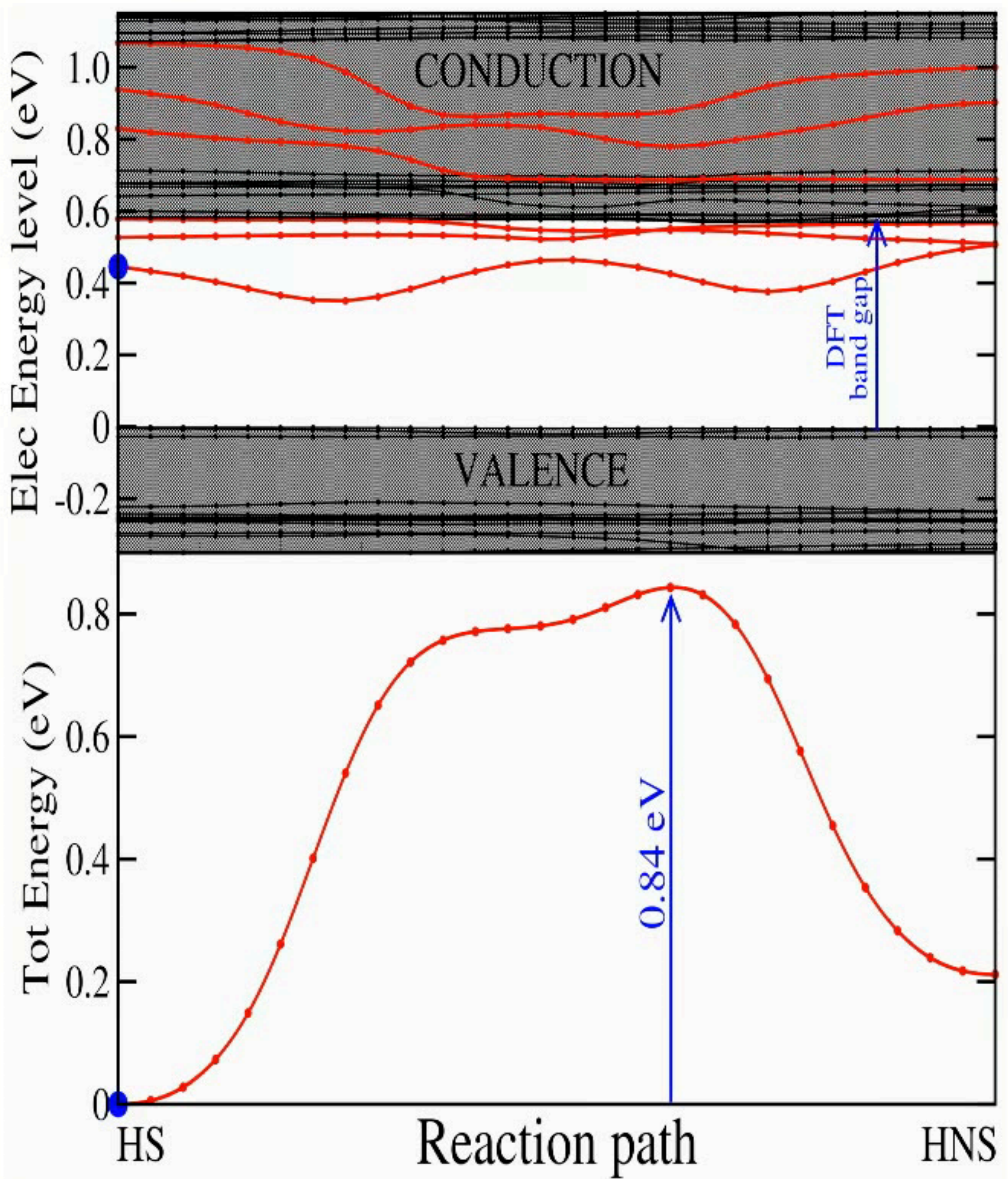
- Theoretical basis of kinetic Monte Carlo
- Importance of the events catalog
- Challenge of Off-Lattice kMC: Complexity increase

Interest of kinetic Monte Carlo

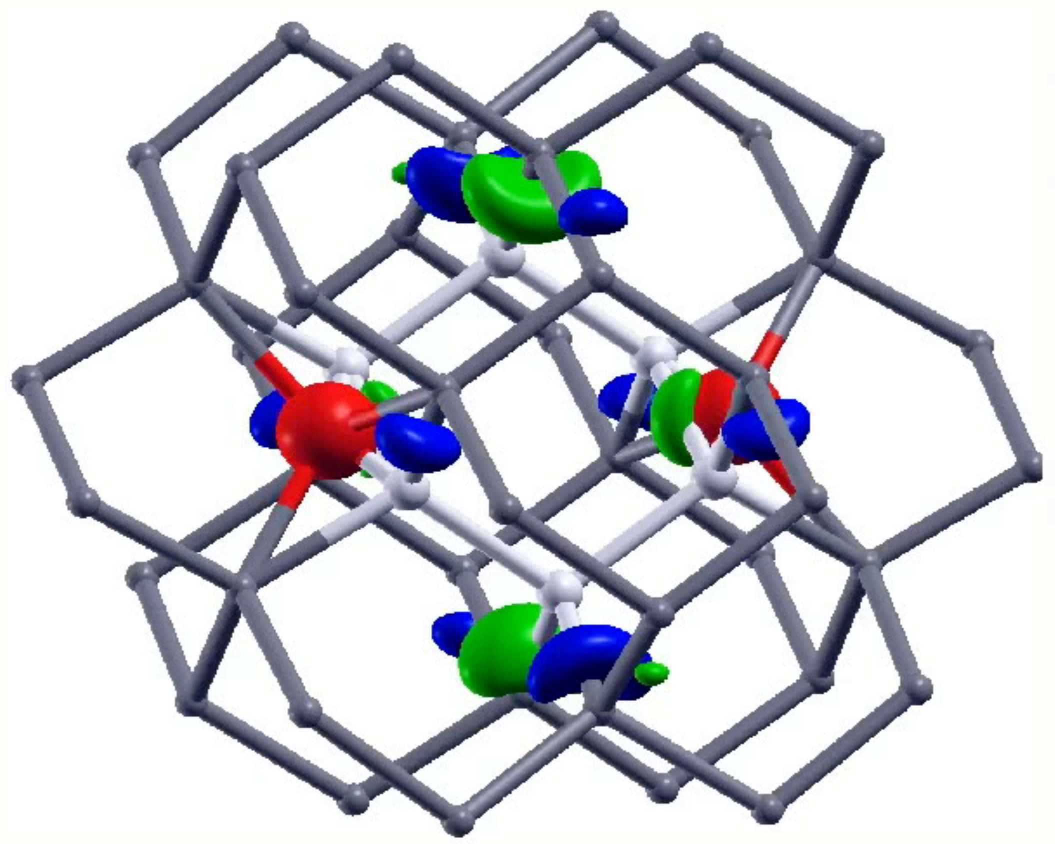
protons, ions

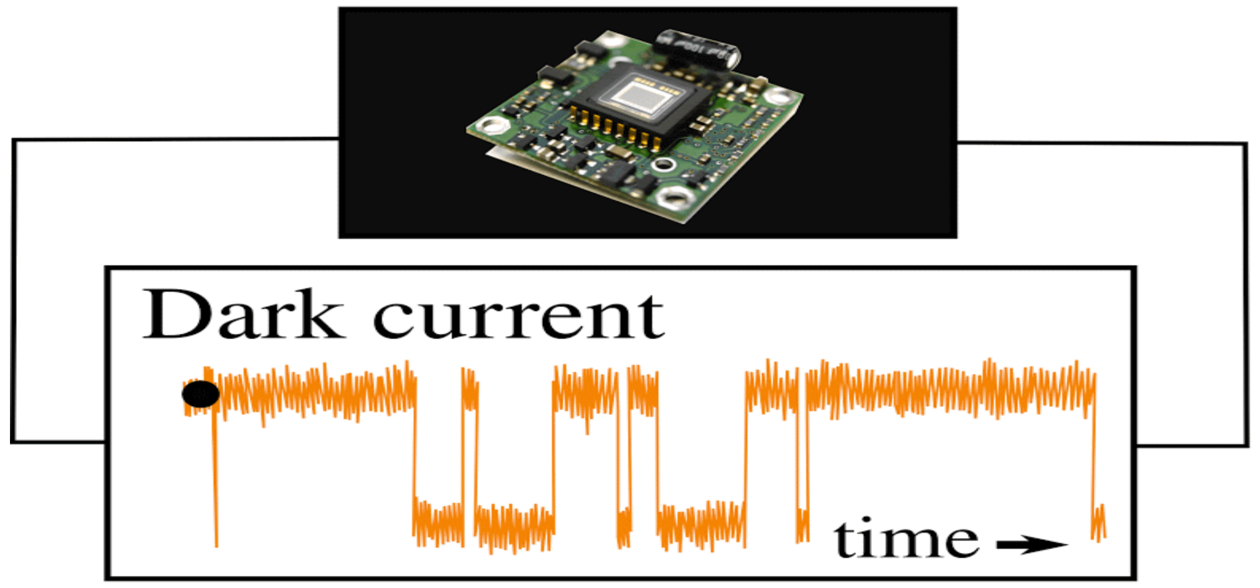


Silicon-based image sensor

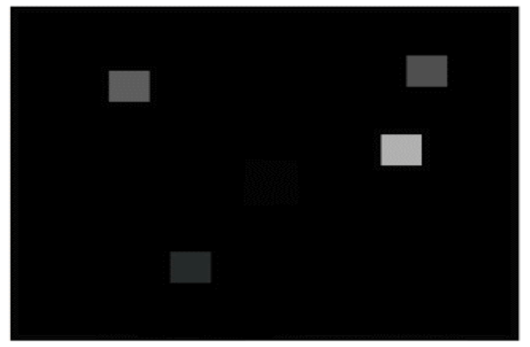


Si-Bivacancies

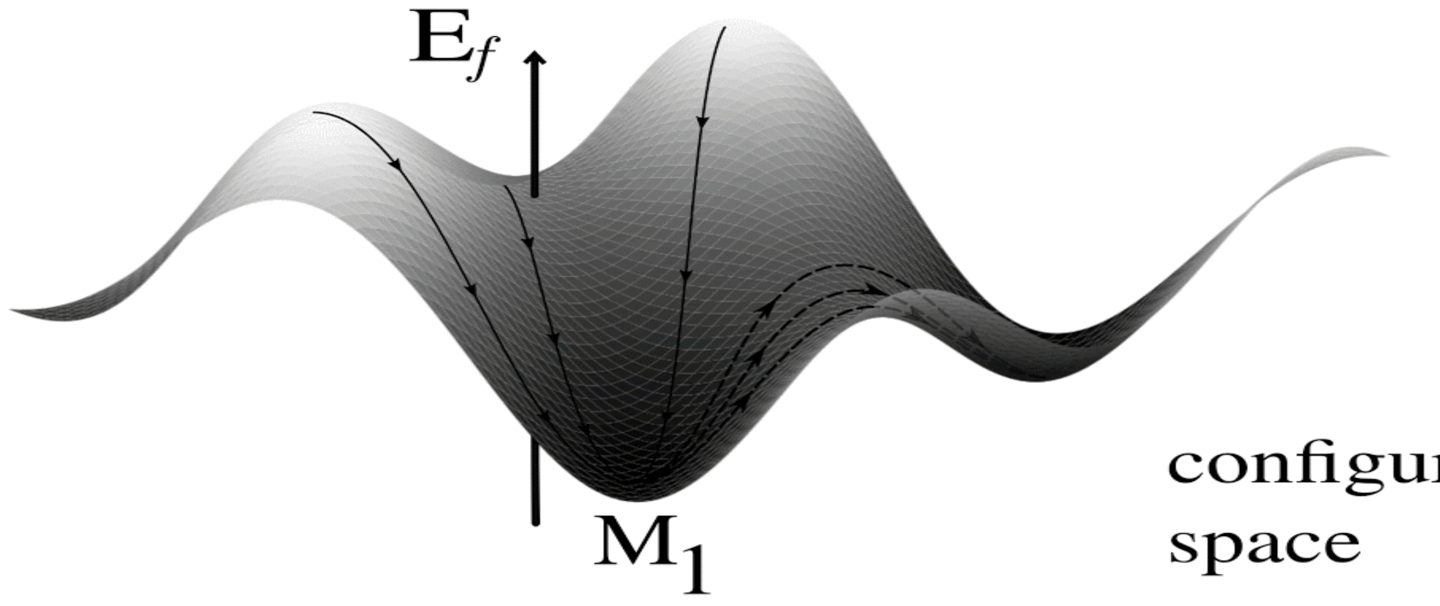




Random Telegraph Signal



pixels

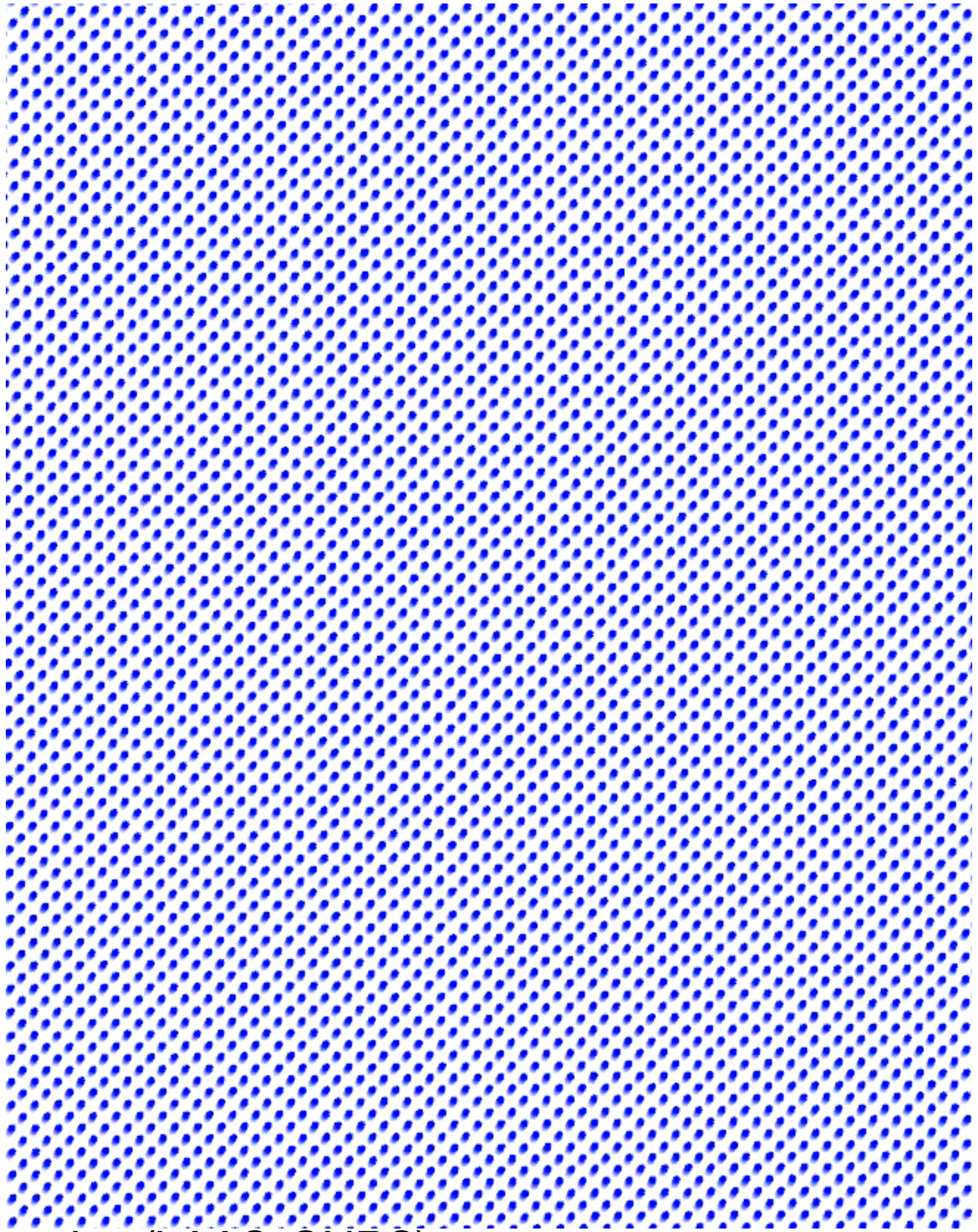


configuration space

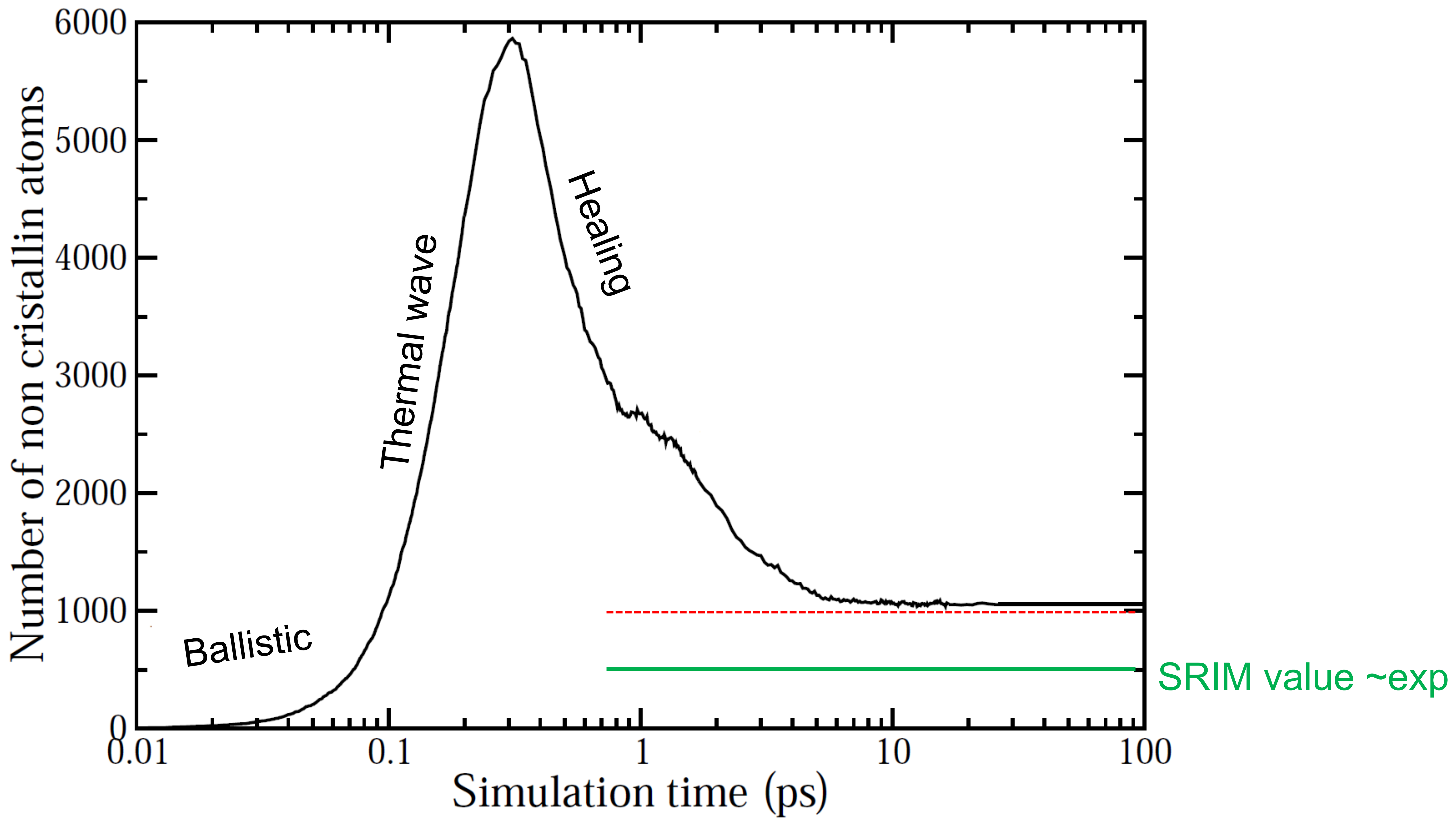
credit to G.Herrero-Saboya

Interest of kinetic Monte Carlo

> Molecular Dynamics: LAMMPS with Stilinger-Weber (SW) Potential

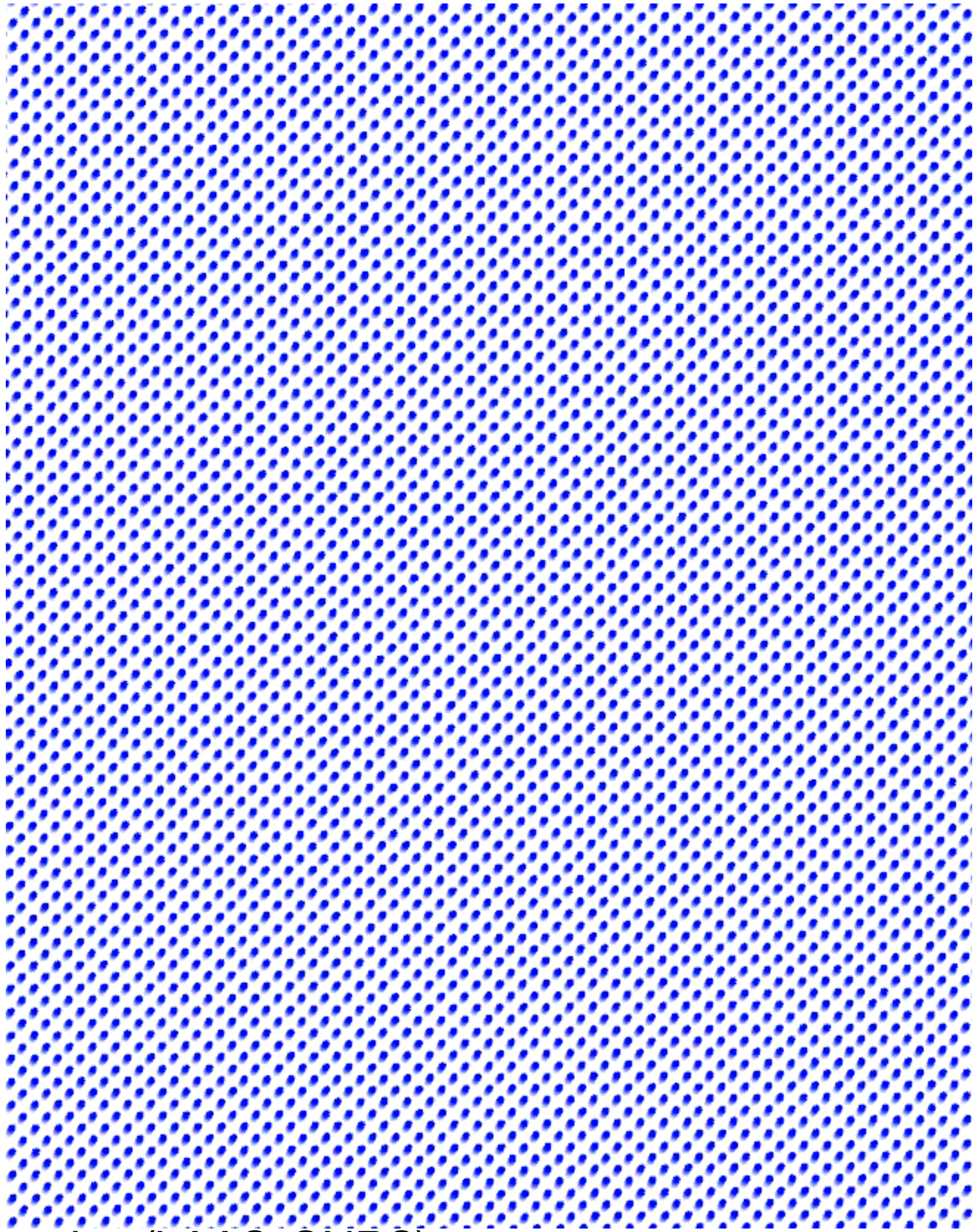


> Evolution of the number of defects   pka = 10keV

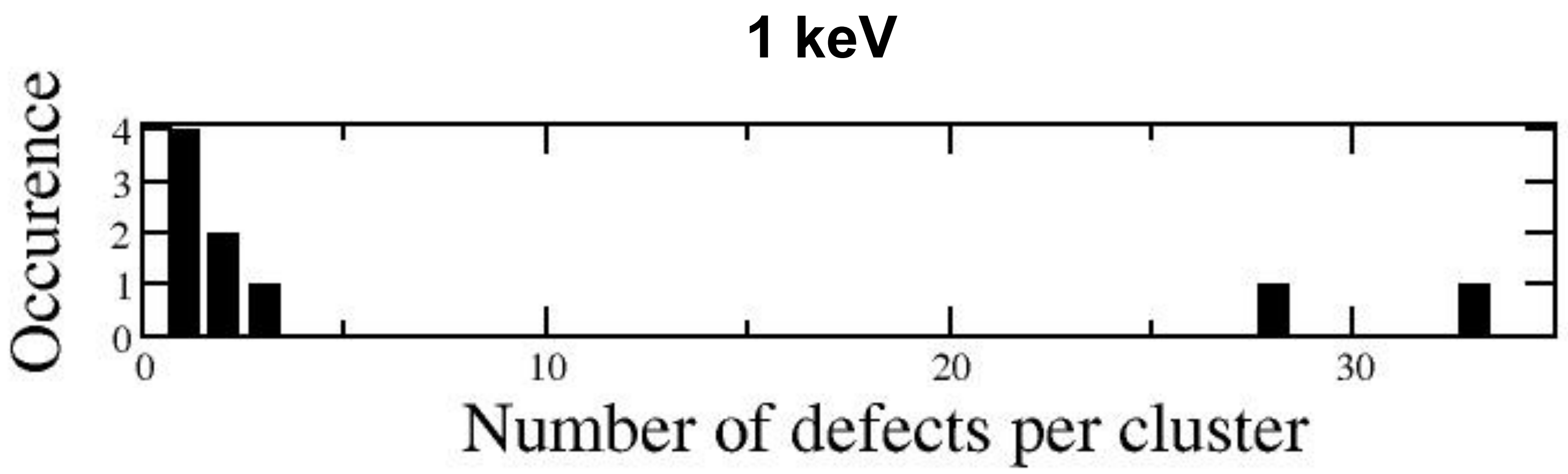


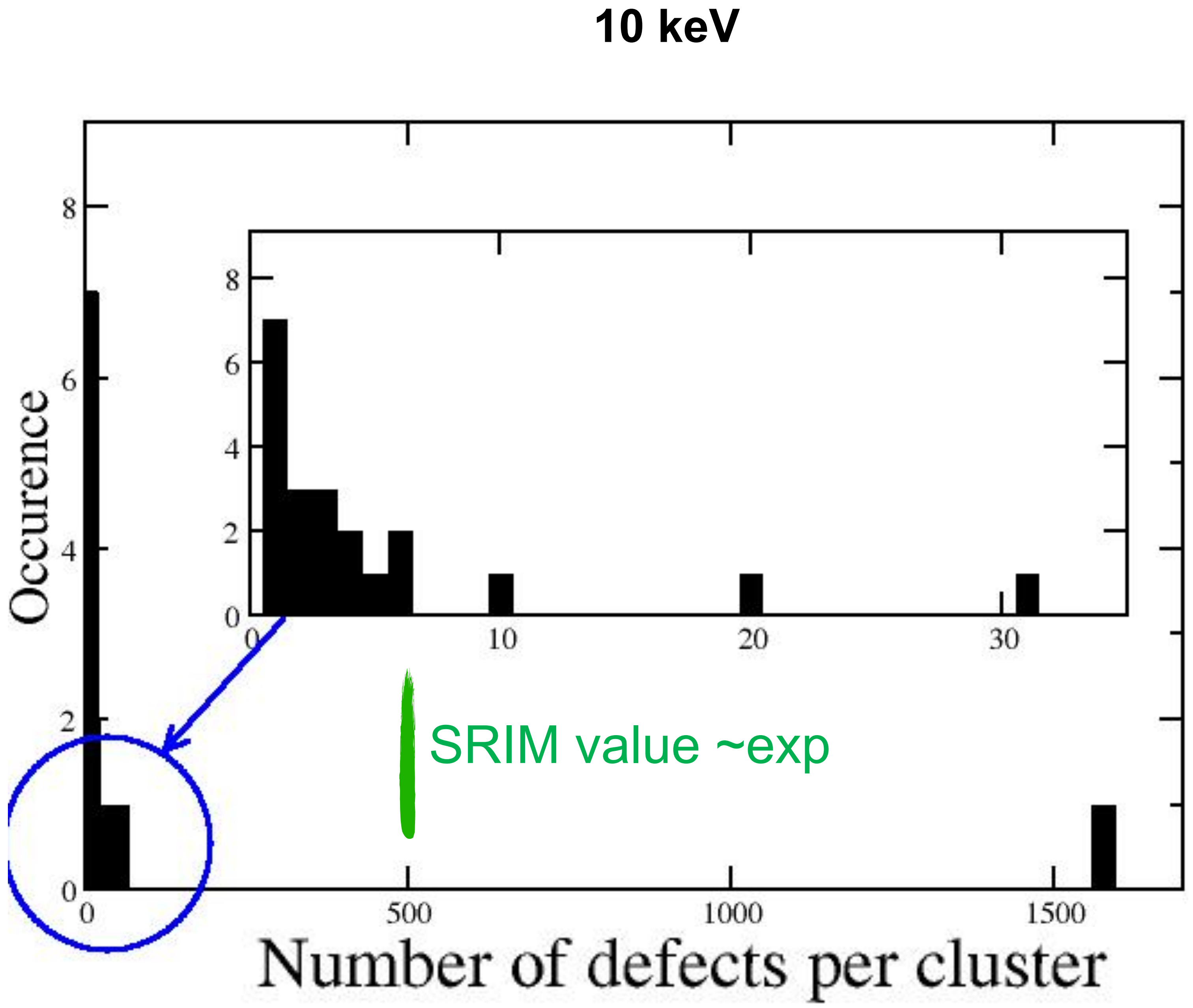
Interest of kinetic Monte Carlo

> Molecular Dynamics: LAMMPS with SW Potential



At the end of the MD simulation (1 ns)

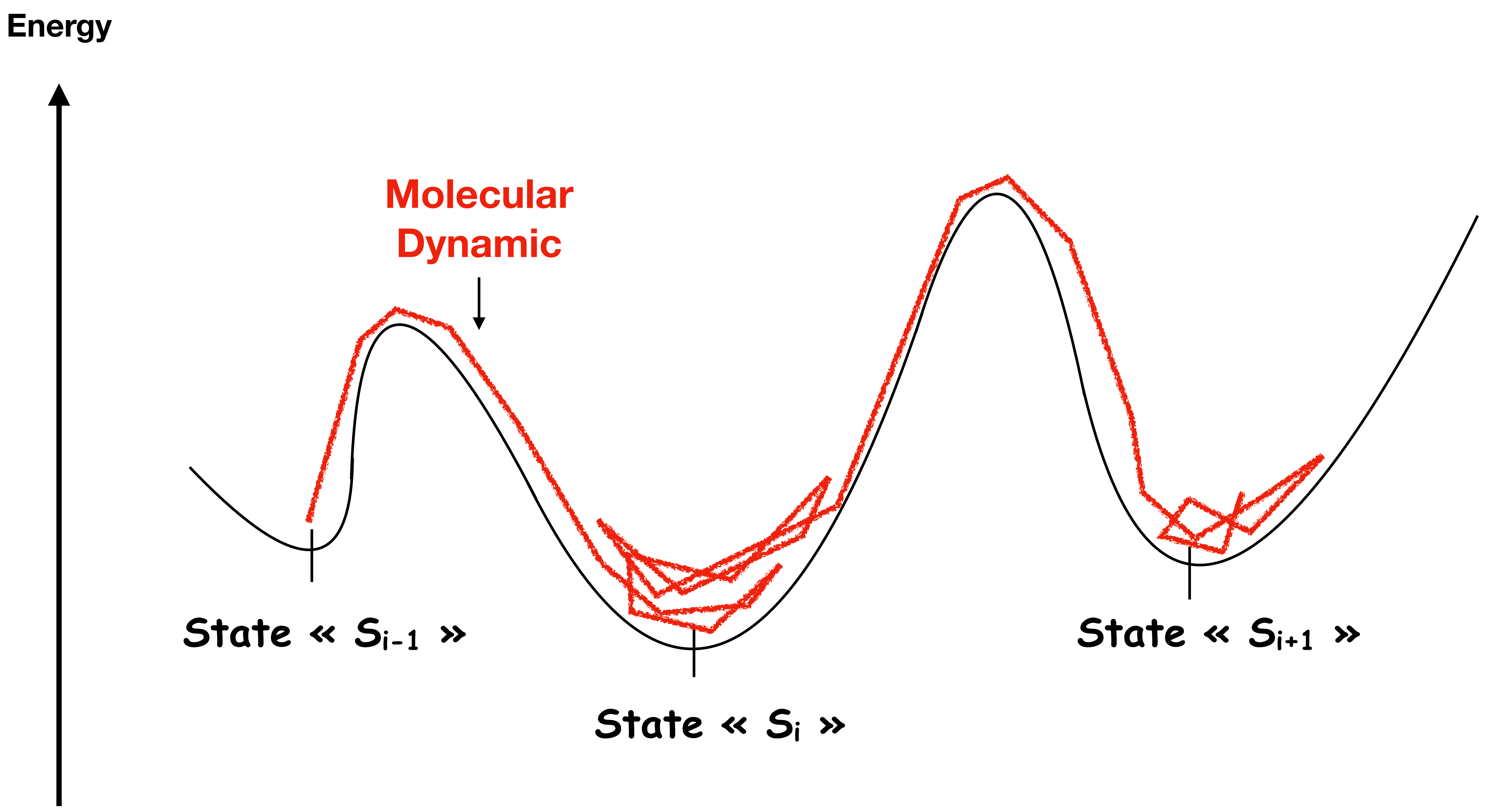




Limitation of the Molecular Dynamics

➢ Need to access to longer times (seconds or more)

Molecular Dynamics VS kinetic Monte Carlo



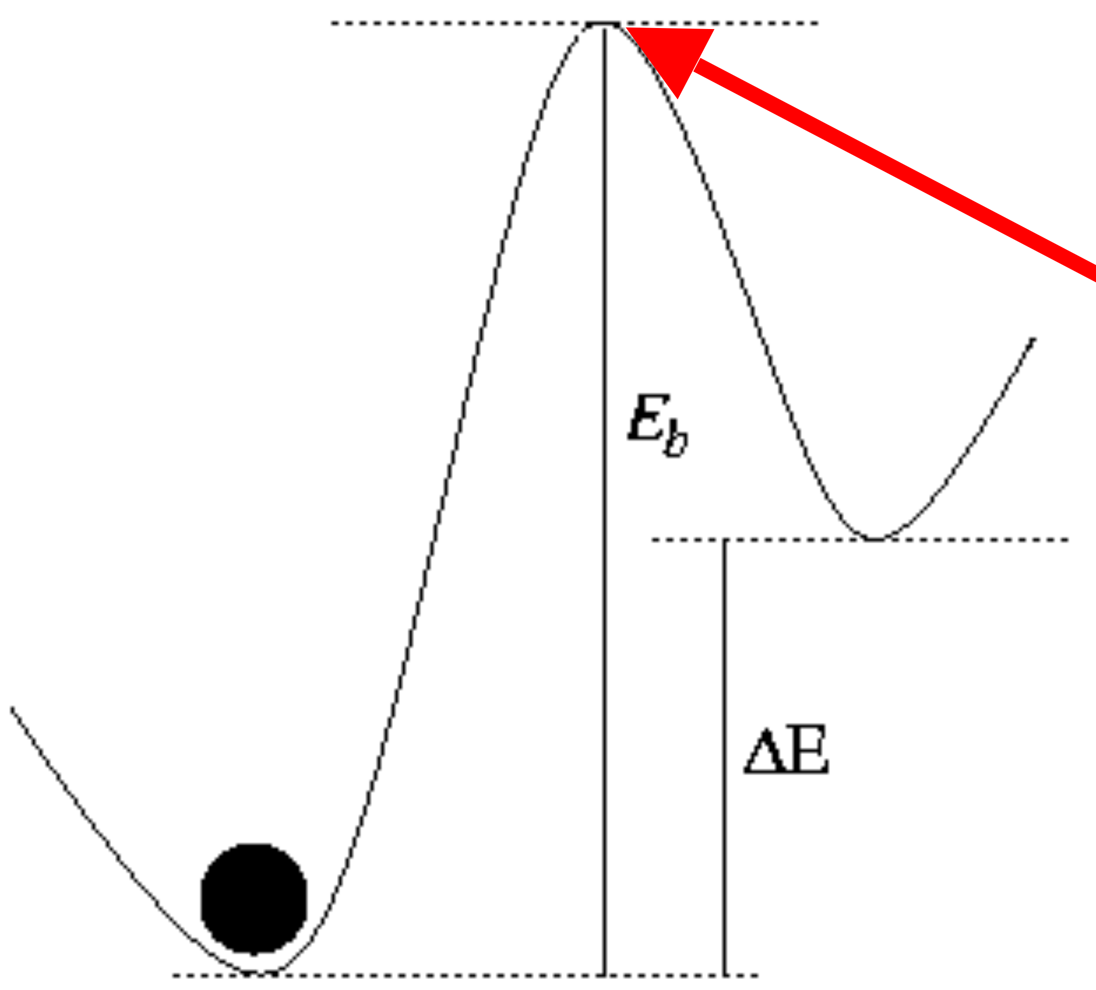
kinetic Monte Carlo

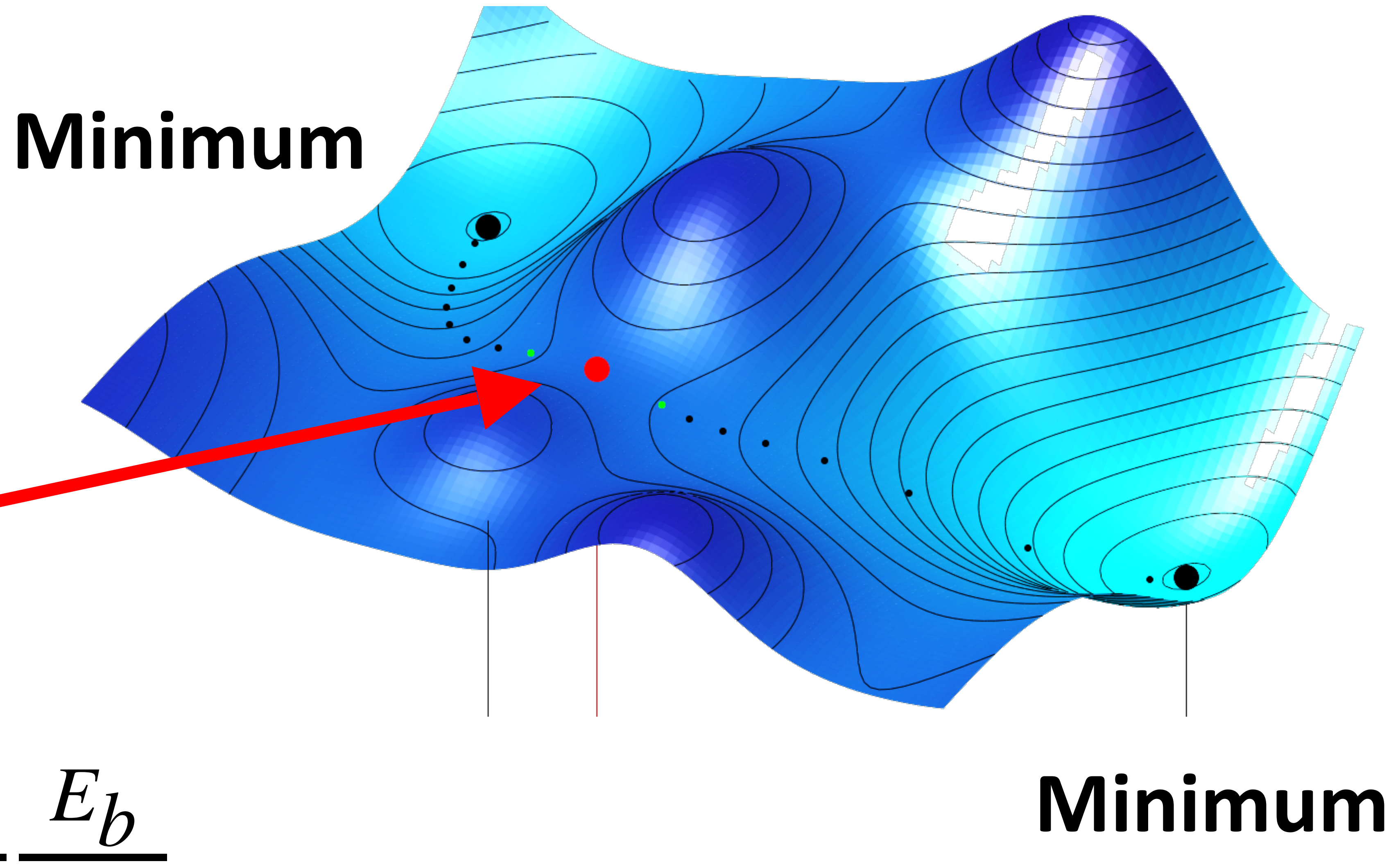
Markovian chain of state $\{S_{i-1}, S_i, S_{i+1}\}$

Interest of kinetic Monte Carlo

➢**Transition State Theory** is an approach for modeling the rate of chemical reactions based on the idea of a transition state or activated complex.

➔ The chemical reaction = a process where the reactants cross an energy barrier E_b to form products. This barrier is associated with a transition state where the reactants are transformed into products.





Reaction rate

$$r_{i\rightarrow j} = \nu \times e^{-\frac{E_b}{k_B T}}$$

Lifetime of event as function T and activation barrier

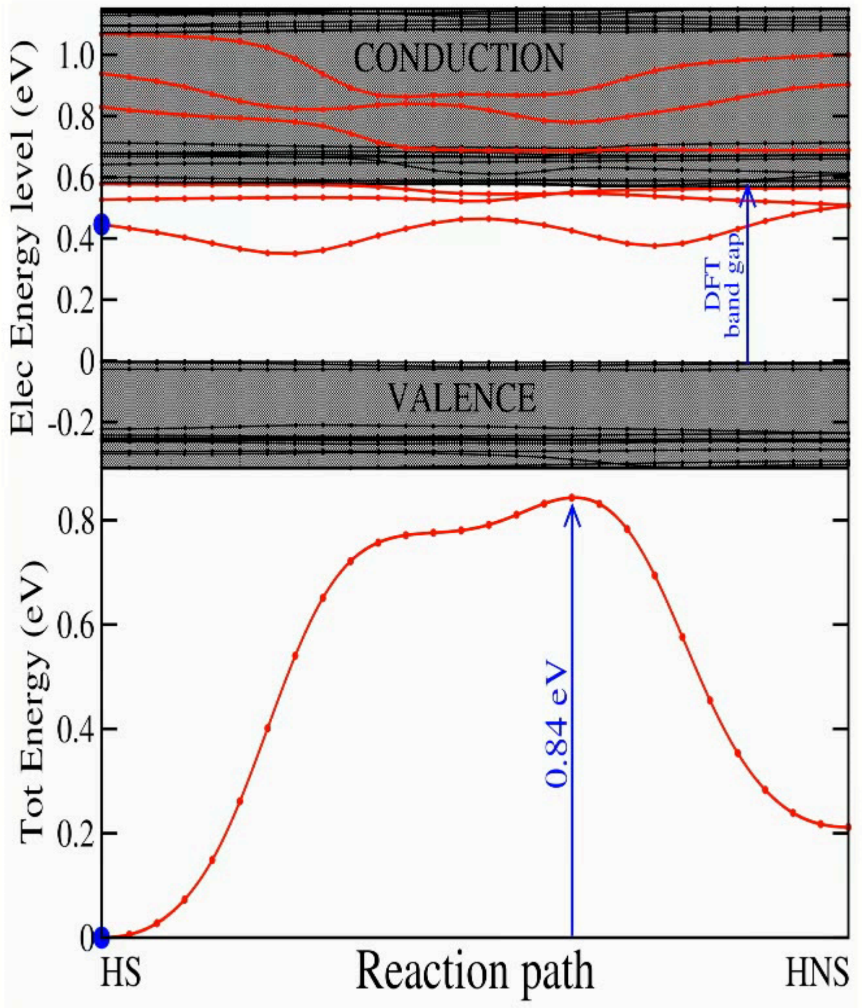
$$t = \frac{1}{\nu} e^{\frac{E_{ac}}{k_B T}}$$

$t > 10^{-4}\, s$

Temp (K)	50	100	200	300	400	500	600	700	800	900	1000
Energy (eV)											
0.1	1.20E-003	1.10E-008	3.31E-011	4.79E-012	1.82E-012	1.02E-012	6.92E-013	5.25E-013	4.27E-013	3.63E-013	3.19E-013
0.2	1.44E+007	1.20E-003	1.10E-008	2.29E-010	3.31E-011	1.04E-011	4.79E-012	2.75E-012	1.82E-012	1.32E-012	1.02E-012
0.3	1.73E+017	1.32E+002	3.63E-006	1.10E-008	6.02E-010	1.06E-010	3.31E-011	1.45E-011	7.76E-012	4.79E-012	3.25E-012
0.4	2.08E+027	1.44E+007	1.20E-003	5.24E-007	1.10E-008	1.08E-009	2.29E-010	7.58E-011	3.31E-011	1.74E-011	1.04E-011
0.5	2.50E+037	1.58E+012	3.98E-001	2.51E-005	1.99E-007	1.10E-008	1.58E-009	3.98E-010	1.41E-010	6.31E-011	3.31E-011
0.6	3.00E+047	1.73E+017	1.32E+002	1.20E-003	3.63E-006	1.12E-007	1.10E-008	2.09E-009	6.02E-010	2.29E-010	1.06E-010
0.7	3.60E+057	1.90E+022	4.36E+004	5.75E-002	6.60E-005	1.14E-006	7.58E-008	1.10E-008	2.57E-009	8.31E-010	3.37E-010
0.8	4.33E+067	2.08E+027	1.44E+007	2.75E+000	1.20E-003	1.16E-005	5.24E-007	5.75E-008	1.10E-008	3.02E-009	1.08E-009
0.9	5.20E+077	2.28E+032	4.78E+009	1.32E+002	2.19E-002	1.18E-004	3.63E-006	3.02E-007	4.67E-008	1.10E-008	3.43E-009
1	6.25E+087	2.50E+037	1.58E+012	6.30E+003	3.98E-001	1.20E-003	2.51E-005	1.58E-006	1.99E-007	3.98E-008	1.10E-008
1.1	7.50E+097	2.74E+042	5.23E+014	3.01E+005	7.23E+000	1.22E-002	1.74E-004	8.31E-006	8.51E-007	1.44E-007	3.50E-008
1.2	9.01E+107	3.00E+047	1.73E+017	1.44E+007	1.32E+002	1.25E-001	1.20E-003	4.36E-005	3.63E-006	5.24E-007	1.12E-007
1.3	1.08E+118	3.29E+052	5.74E+019	6.90E+008	2.39E+003	1.27E+000	8.31E-003	2.29E-004	1.55E-005	1.90E-006	3.56E-007
1.4	1.30E+128	3.60E+057	1.90E+022	3.30E+010	4.36E+004	1.29E+001	5.75E-002	1.20E-003	6.60E-005	6.91E-006	1.14E-006
1.5	1.56E+138	3.95E+062	6.29E+024	1.58E+012	7.93E+005	1.32E+002	3.98E-001	6.30E-003	2.82E-004	2.51E-005	3.63E-006
1.6	1.87E+148	4.33E+067	2.08E+027	7.57E+013	1.44E+007	1.34E+003	2.75E+000	3.31E-002	1.20E-003	9.11E-005	1.16E-005
1.7	2.25E+158	4.74E+072	6.89E+029	3.62E+015	2.62E+008	1.37E+004	1.90E+001	1.74E-001	5.12E-003	3.31E-004	3.70E-005
1.8	2.70E+168	5.20E+077	2.28E+032	1.73E+017	4.78E+009	1.39E+005	1.32E+002	9.11E-001	2.19E-002	1.20E-003	1.18E-004
1.9	3.25E+178	5.70E+082	7.55E+034	8.29E+018	8.69E+010	1.42E+006	9.11E+002	4.78E+000	9.32E-002	4.36E-003	3.76E-004
2	3.90E+188	6.25E+087	2.50E+037	3.97E+020	1.58E+012	1.44E+007	6.30E+003	2.51E+001	3.98E-001	1.58E-002	1.20E-003

$t > 10^{4}\, s$

bivacancies



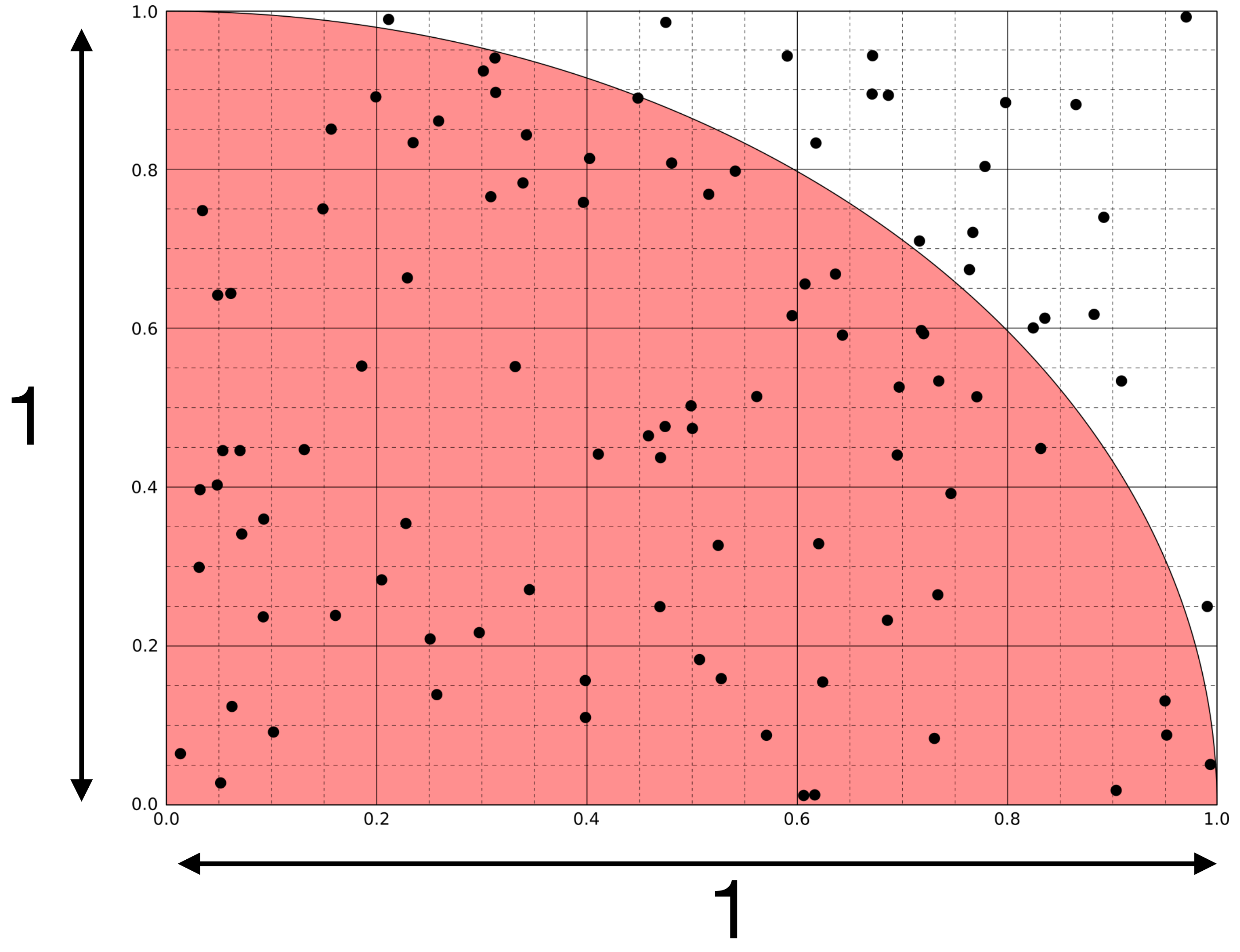
$E_b = 0.84\ eV$

kinetic Monte Carlo (kMC)

Monte Carlo (MC) method:

Random number to compute quantities

> Statistical method



$$\frac{S_{\bigcirc}}{4}\frac{1}{S_{\square}} = \frac{\pi R^2}{4}\frac{1}{R^2} = \frac{\pi}{4}$$

$$\frac{N_{\bigcirc}}{N_{\square}} \sim \frac{\pi}{4}$$

kinetic Monte Carlo (kMC) method:

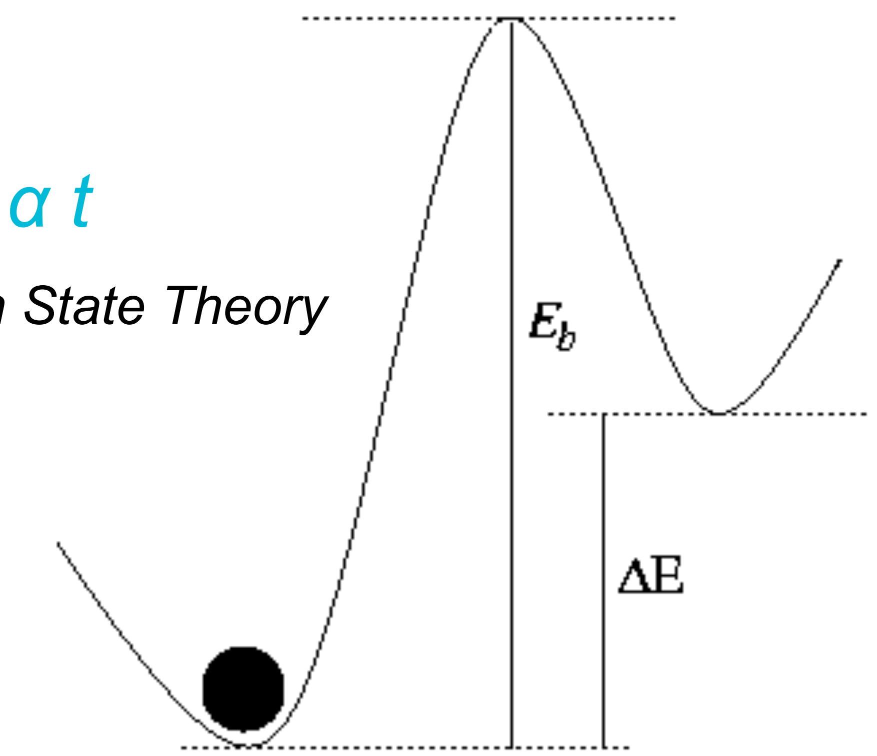
Random number to change system following a probability

> Probabilistic method

Transition State between discrete states over the time

E_b *α t*

Transition State Theory



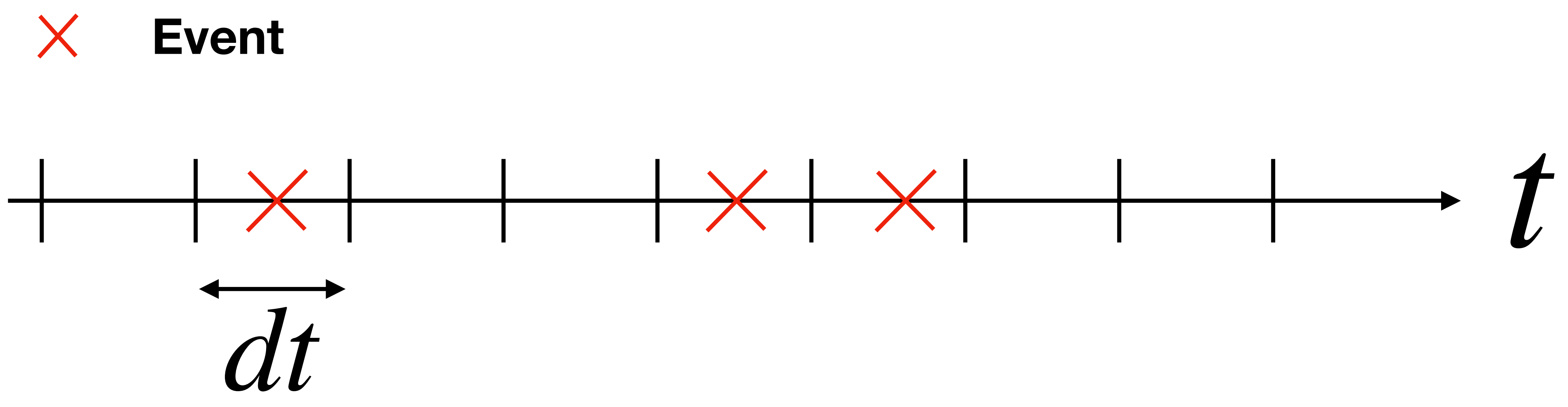
kinetic Monte Carlo (kMC)

> Frequentist representation

The time is divided in interval dt

$$t = n \,.\, dt$$

➔ some event happens during this time t



If we take dt small enough we can have, at most, one event by interval

➔ we define the average rate:

$$r = \lim_{dt \to 0, t \to \infty} \sum^{n} \frac{N_{evt}}{t}$$

➔ and the probability to have an event in time dt

$$p = r \,.\, dt$$

kinetic Monte Carlo (kMC)

> *Binomial representation*

Binomial law b(n,p):
- n the number of experiment
- p (=r.dt) the probability to have a success

The probability to have x event in n trial: $P(X=x) = \binom{n}{x}(r\,.\,dt)^x(1-r\,.\,dt)^{n-x}$ $\qquad \binom{n}{x} = \frac{n!}{x!(n-x)!}$

> *Convergence to the Poisson law*

We have $n\,.\,p = n\,.\,r\,.\,dt = r\,.\,t > 0$

In the limit of small *dt*: $p \to 0$, $n \to \infty$

➔ The binomial law **`b(n,r.dt)`** converge to Poisson law of parameter $\lambda = r\,.\,t$

$$P(X=x) = \frac{(rt)^x}{x!}e^{-rt}$$

The probability of having x events in n attempt

become -> the probability of having x event in time t

kinetic Monte Carlo (kMC)

Theorem:
- If $\mathbf{N_t}$ is number of event in time t and
- $\mathbf{T_n}$ is time between (n-1) and n-th event

Then $\mathbf{N_t}$ is Poisson process of parameter **K.t** if and only if $\mathbf{T_n}$ **are independent** and **follow the exponential law** with same parameter **K.t.**

Poisson law of parameter λ

$$P(X = x) = \frac{(\lambda t)^x}{x!} e^{-\lambda t}$$

Probability of having x event in time t

Exponential law of parameter λ

$$P(X > t) = e^{-\lambda t}$$

Probability of having an event after a time t

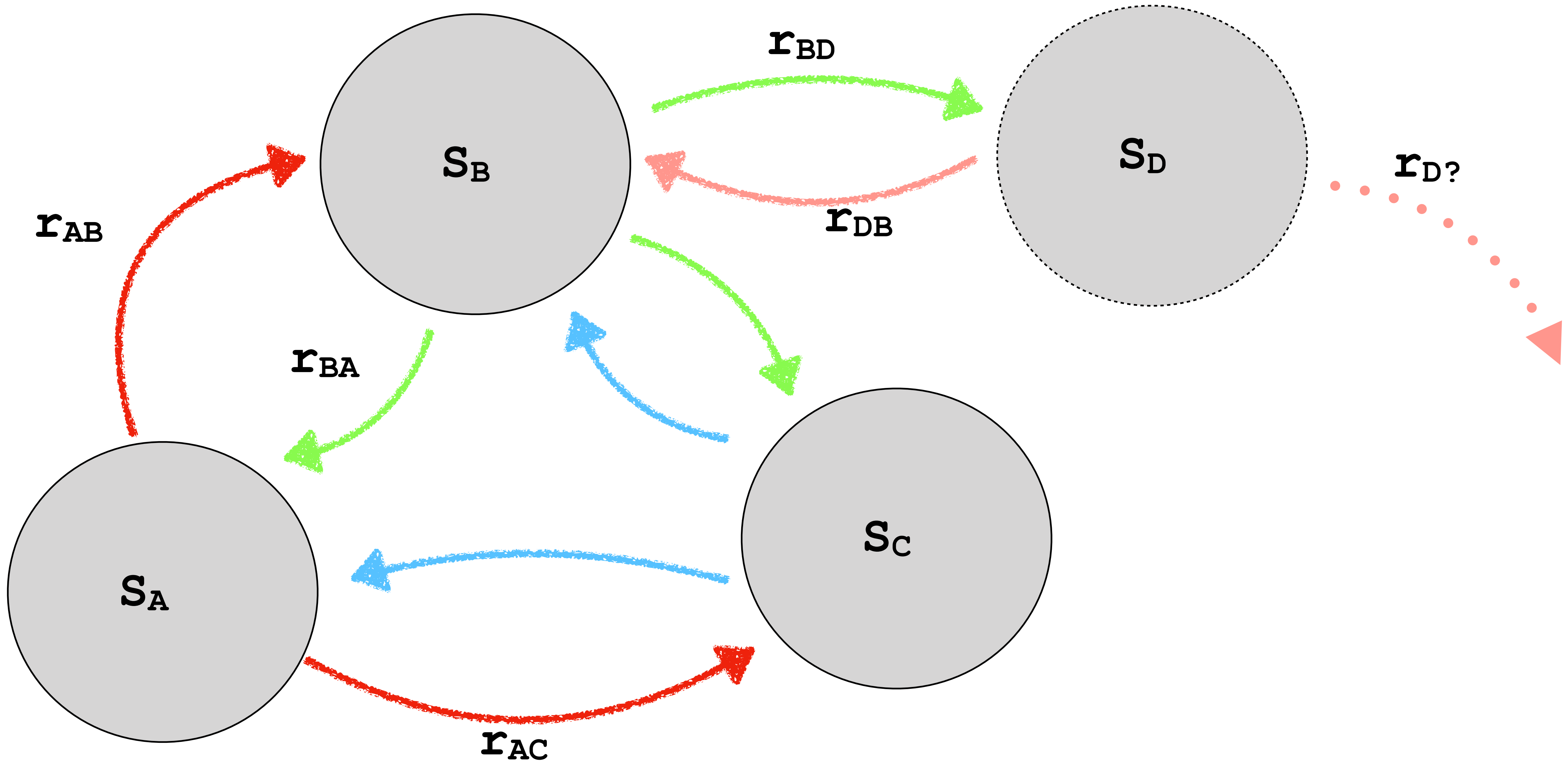
The time before the event happen:

$$t = -\frac{\ln u}{\lambda}$$

kinetic Monte Carlo (kMC)

T_n independent means that the event happens at time t is independent of event happened in previous time

➔ **Markov Chain hypothesis**



$$r_A = \sum_I r_{AI} \qquad T_1 = -\frac{\ln u}{r_A}$$

$$r_B = \sum_I r_{BI} \qquad T_2 = -\frac{\ln u}{r_B}$$

$$\ldots$$

Chain: S_A -> S_B -> S_A -> S_C -> S_B -> S_D -> ...

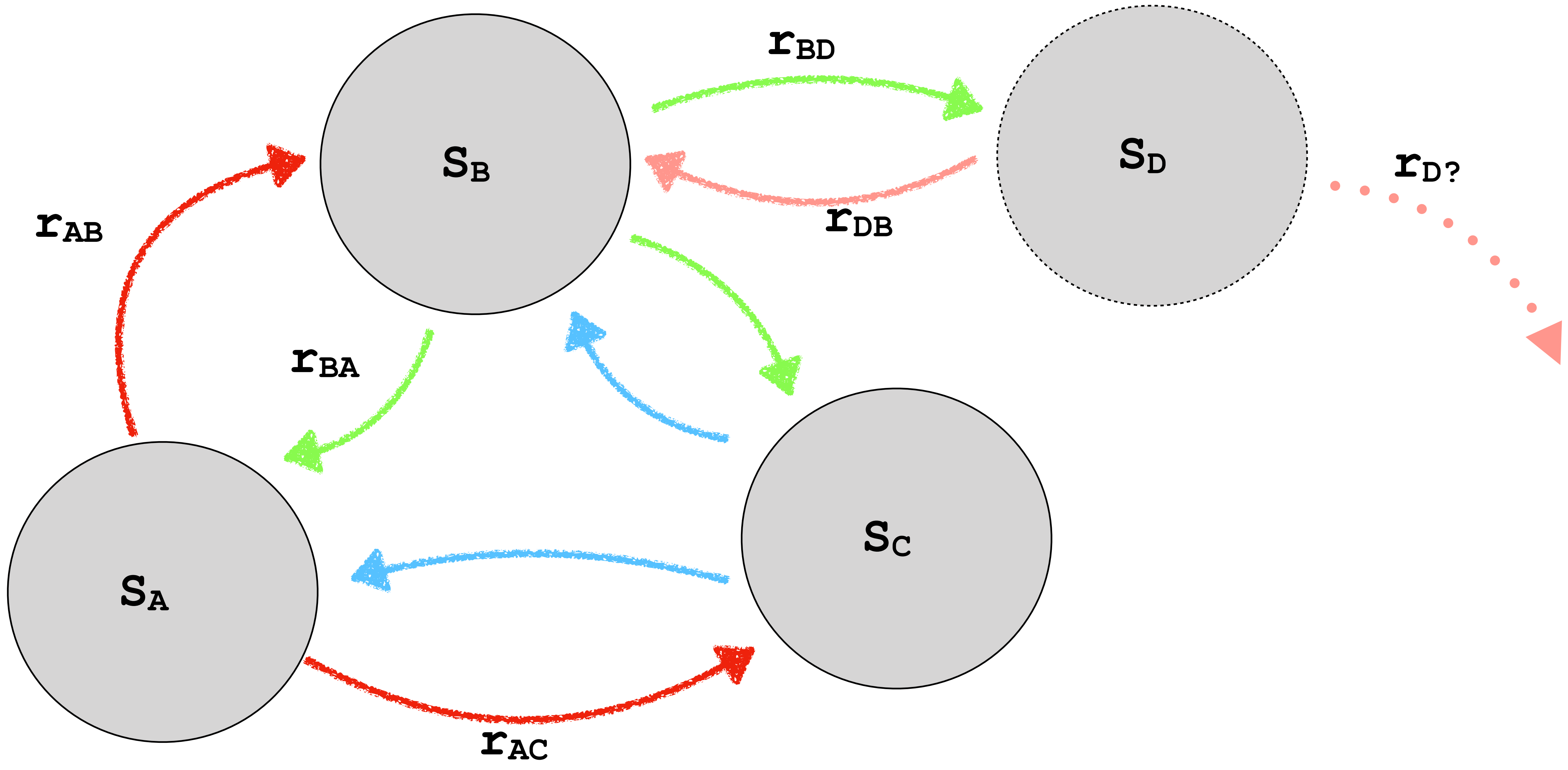
T_1 T_2 T_3 T_4 T_5

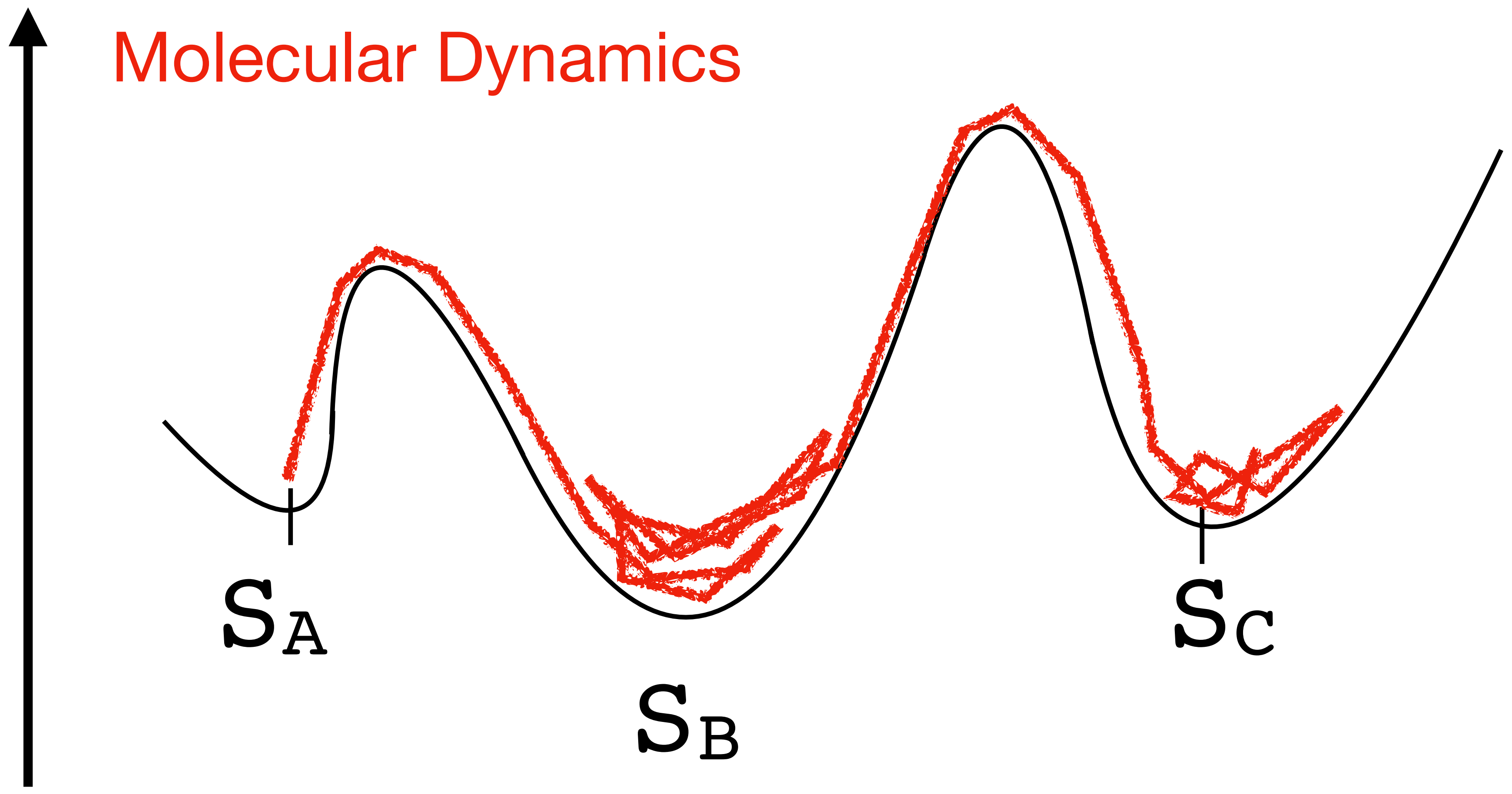
$$t = \sum T_i$$

kinetic Monte Carlo (kMC)

T_n independent means that the event happens at time t is independent of event happened in previous time

➔ **Markov Chain hypothesis**





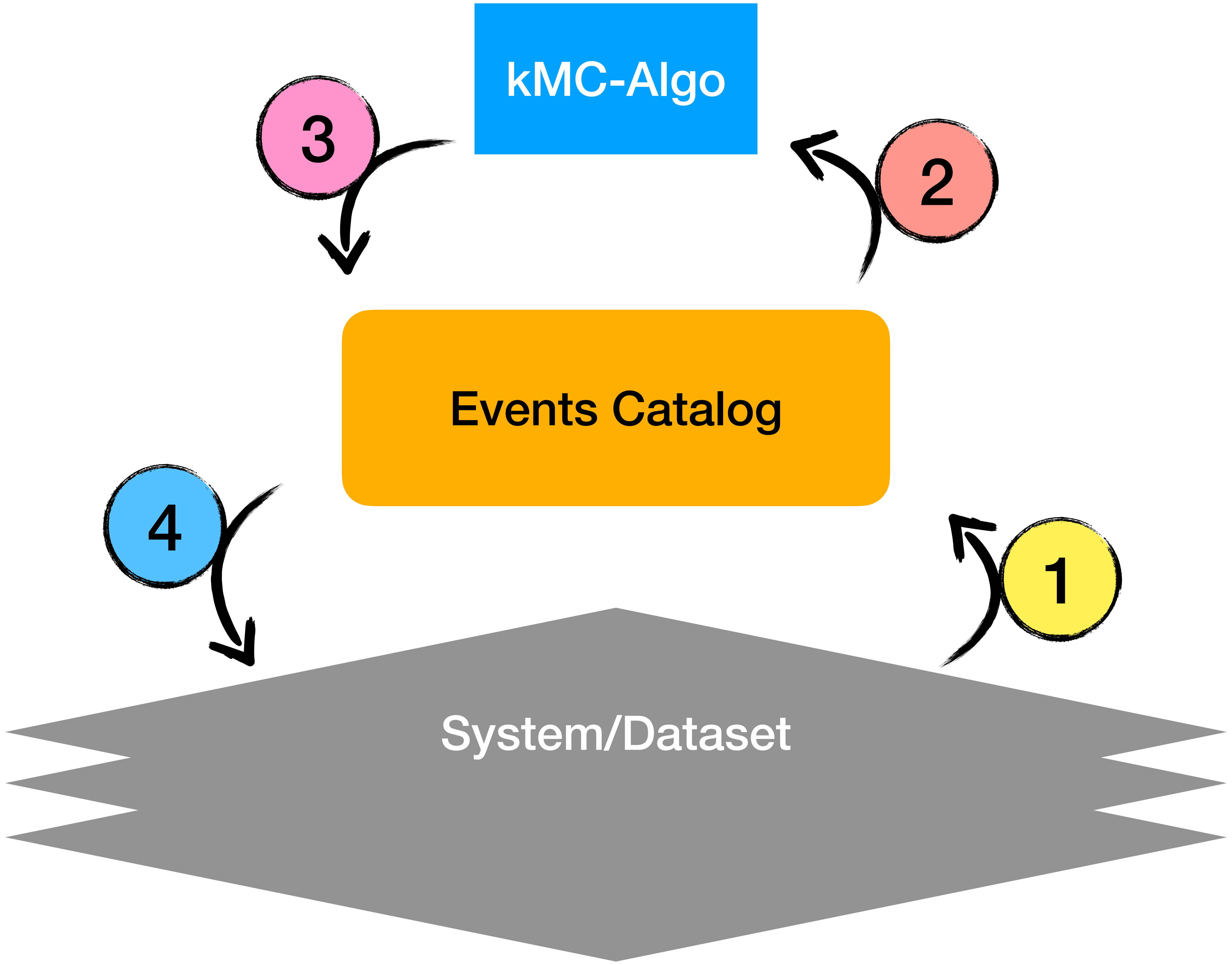
Chain: S_A -> S_B -> S_A -> S_C -> S_B -> S_D -> …

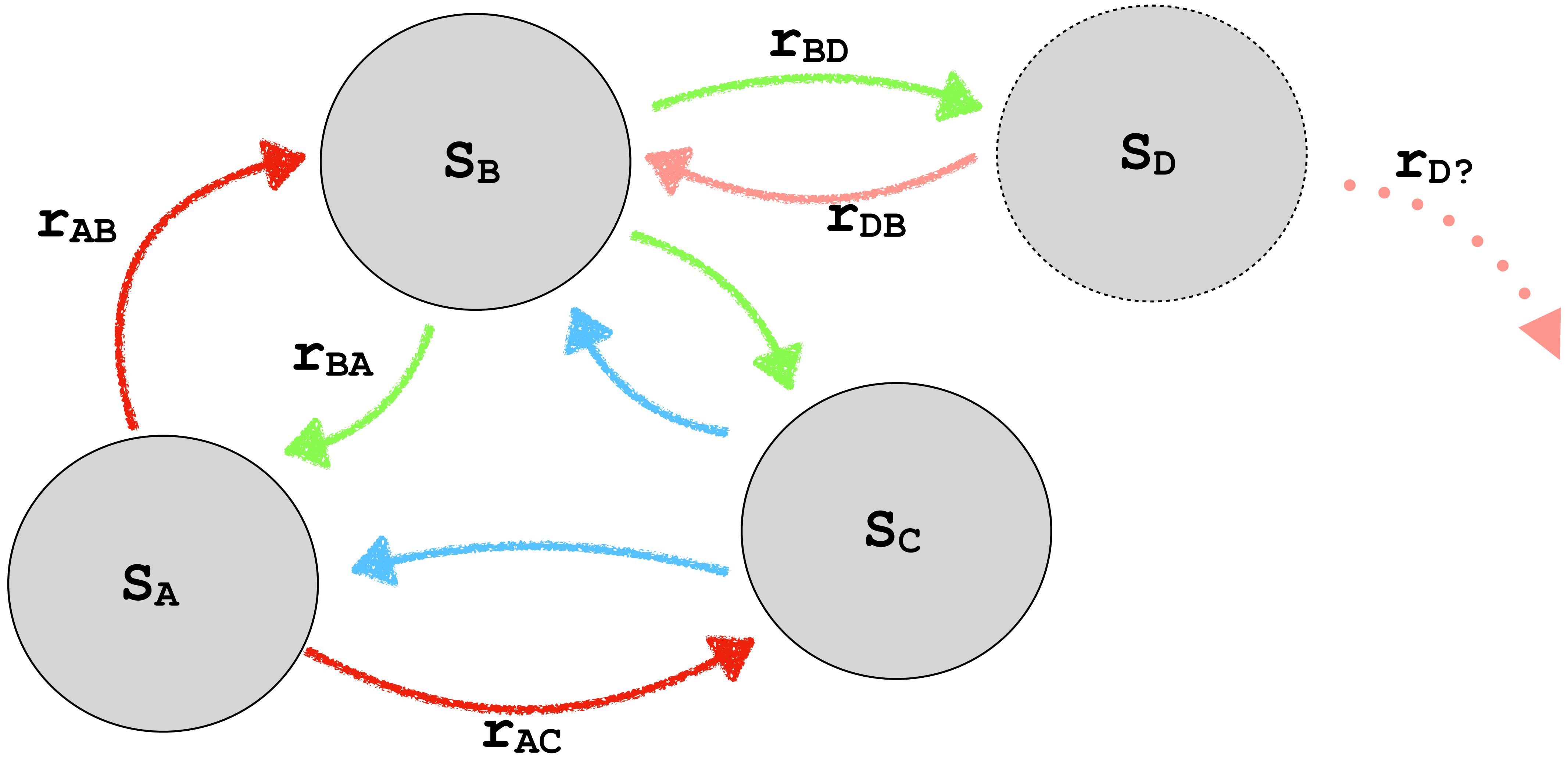
$t = \sum T_i$

Master equation

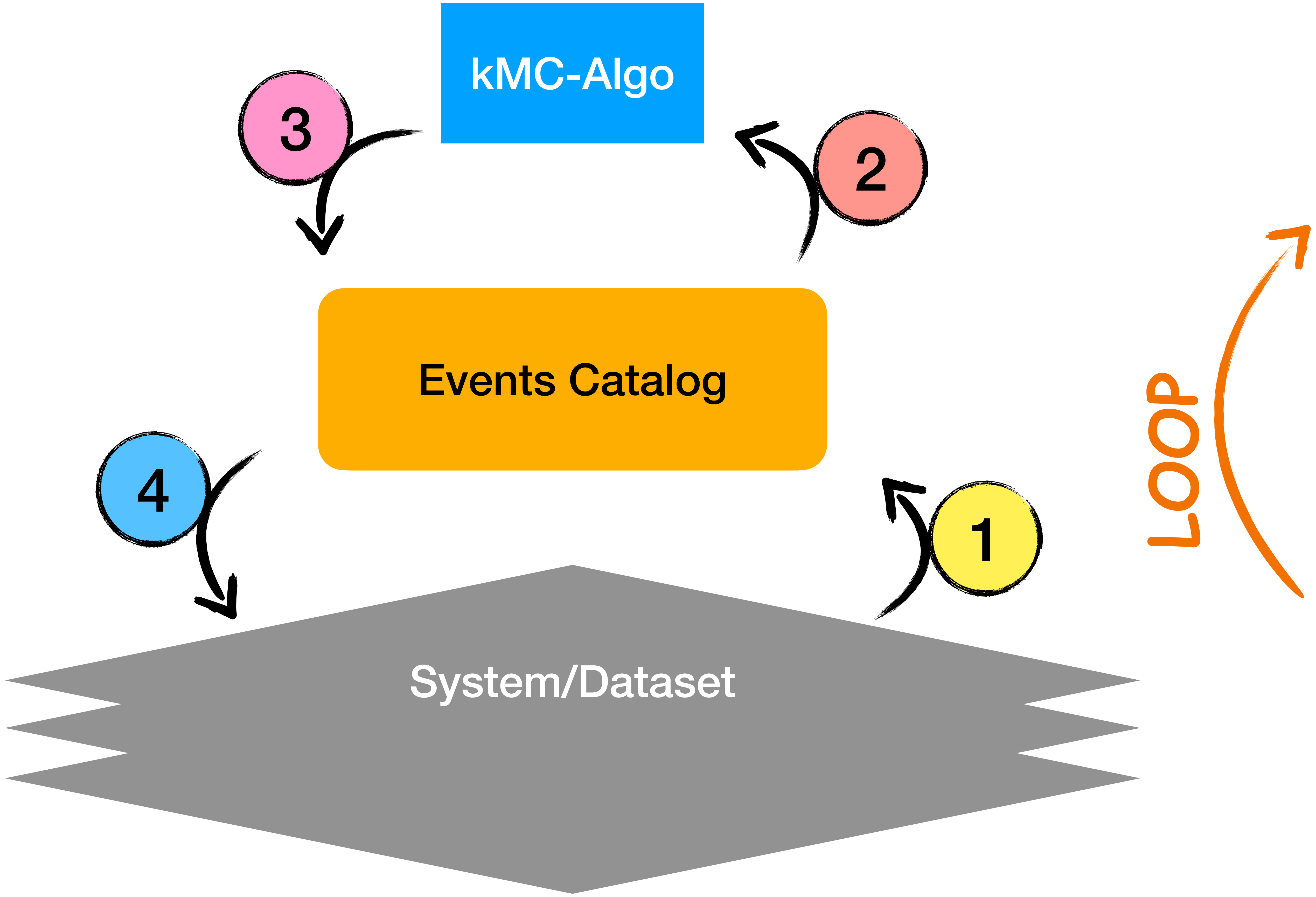
$$\frac{\partial P(\sigma, t)}{\partial t} = \sum_{\sigma'} r_{\sigma' \to \sigma} \, . \, P(\sigma', t) - r_{\sigma \to \sigma'} \, . \, P(\sigma, t)$$

kinetic Monte Carlo (kMC): Resume





kinetic Monte Carlo (kMC): Resume



1. Compare the system to the catalog of event
2. Give the list of possible event to the kMC algorithm
3. Return the site and the event selected
4. Apply the event on the selected site

Kinetic Monte Carlo algorithm

Residence time algorithm (Bortz, Kalos and Lebowitz)

Step 0 - Set the time t = 0, initial configuration

Step 1 - Form a list of all the rates r_{ij} of all possible transitions P_i in the system

Step 2 - Calculate the partition function $Z = \sum_j r_{ij}$ for i = 1,…, N where N is the total number of transitions.

Step 3 - Get a uniform random number n ∈ [0, 1]

Step 4 - Find the event to carry out *i* by finding the i for which

$$P(ik) < n \,.\, Z < P(im)$$

Step 5 - Apply event *i*, change the local atomic configuration

Step 6 - Find all P_i and recalculate all r_{ij} which may have changed due to the transition

Step 7 - Get a new uniform random number n ∈ [0, 1]

Step 8 - Update the time with t = t + dt where:

Step 9 - Return to step 1

Rate of the event Probability

Stochastic choice of event

Temporal evolution

First reaction algorithm (Gillepsie)

Step 0 - Set the time t = 0, initial configuration

Step 1 - Form a list of all the rates r_{ij} of all possible transitions Pi in the system

Step 3 - For each possible event S_i → S_j, get a random number u ∈ [0, 1]

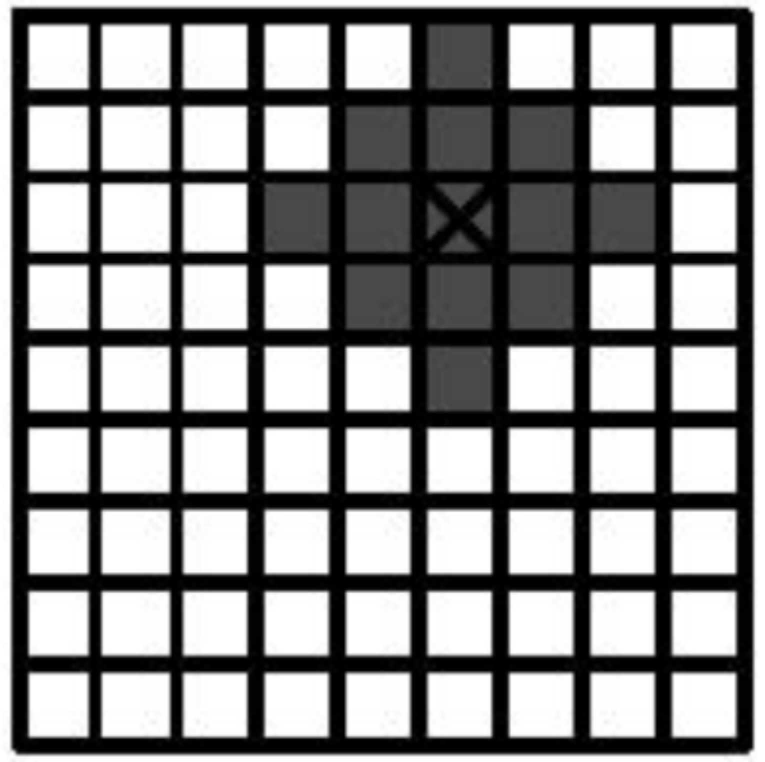
and compute associated time $\tau = -\dfrac{\ln u}{r_{ij}}$

Step 4 - Select the event S_i → S_j with the lowest τ

Step 5 - Apply selected transition, and increase time by τ .

Step 6 - Return to step 1

(advantage: previous computations are stored, only nearest neighbors and associated events are updated)



Rate of the event Probability

Stochastic choice of event

Temporal evolution

Standard algorithm / Rejection

Step 0 - Set the time t = 0, initial configuration

Step 1 - Form a list of all the rates r_{ij} of all possible transitions P_i in the system

Step 2 - Calculate the partition function $Z = \sum_{i,j} r_{ij}$ for i = 1,…, N where N is the total number of transitions.

Step 3 - Choose a transition at random $S_i \rightarrow S_j$

Step 4 - Get a random number $u_1 \in [0,1]$

Apply event if $u_1 < \frac{r_{ij}}{Z}$

Step 5 - If transition accepted, pick a random number $u_2 \in [0,1]$, and increase time

$$\tau = -\frac{\ln u_2}{Z}$$

Otherwise the event is rejected

$$\epsilon = \frac{N_{rejected}}{N_{tot}}$$

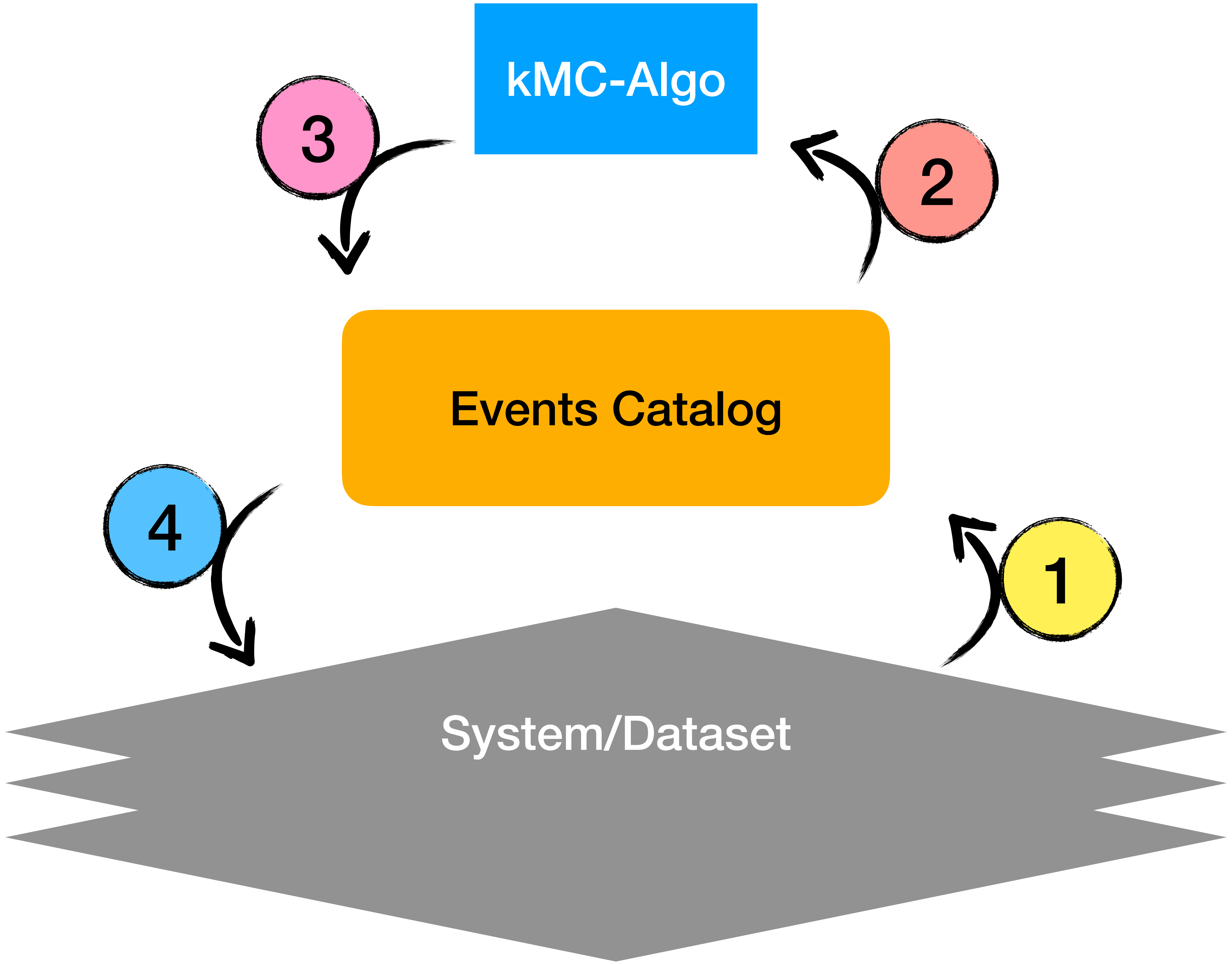
Step 6 - Return to step 1

Rate of the event Probability

Stochastic choice of event

Temporal evolution

kinetic Monte Carlo (kMC): Resume



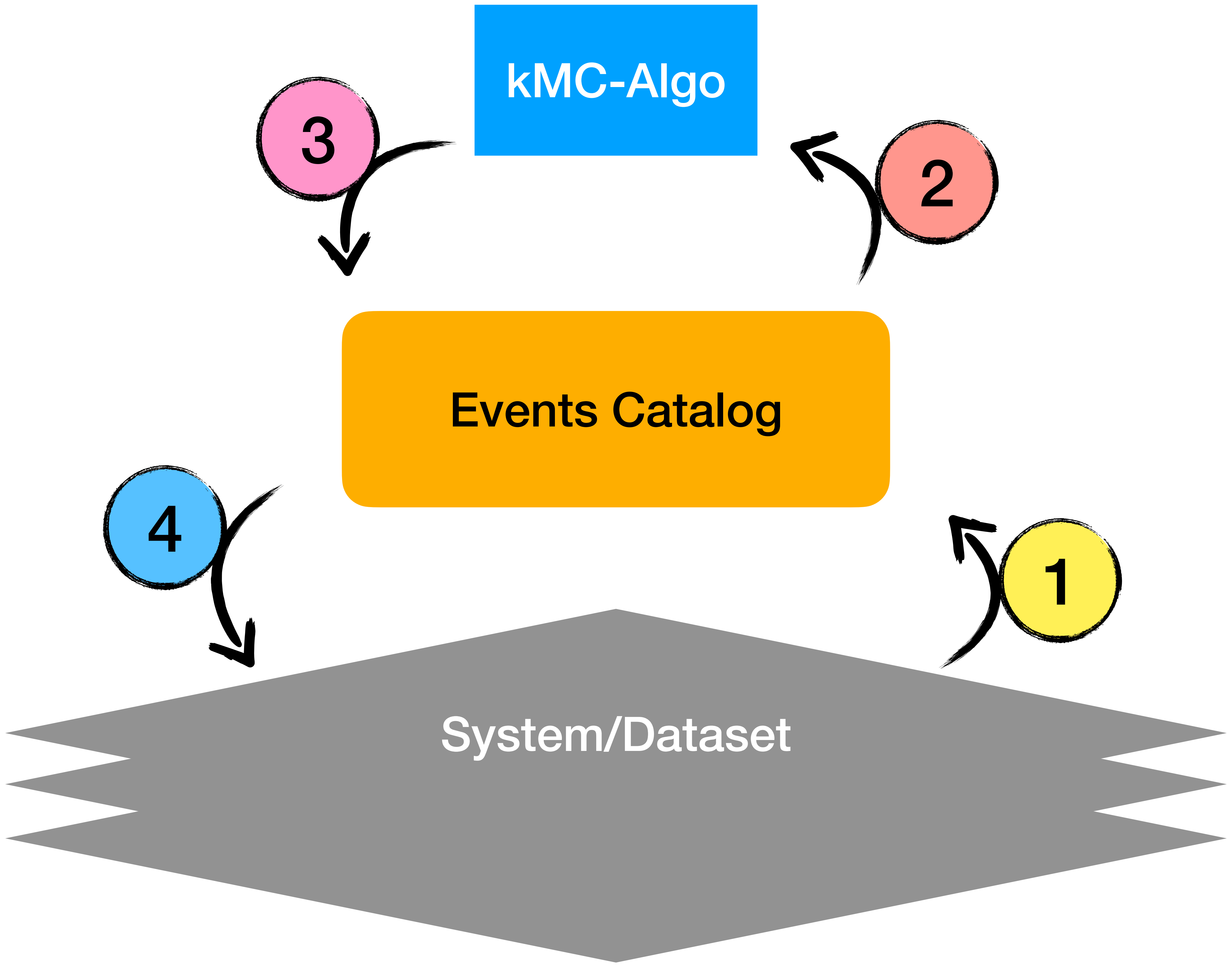
kinetic Monte Carlo:

- Versatile method, large kind of system can be describes
- Large system dimension & long time evolution

Difficulties:

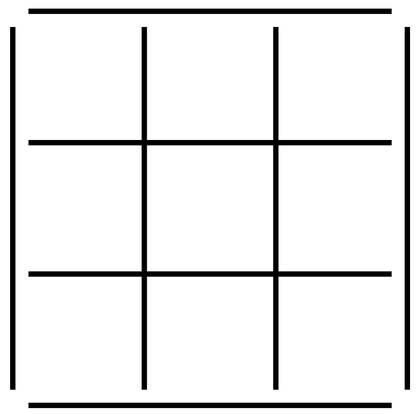
- Description depend on the list of event
- Not fully Parallelizable but some optimization exist

kinetic Monte Carlo (kMC): Resume



→ **On-Lattice kMC**

Approximation of the system by a grid of point



+ events catalog

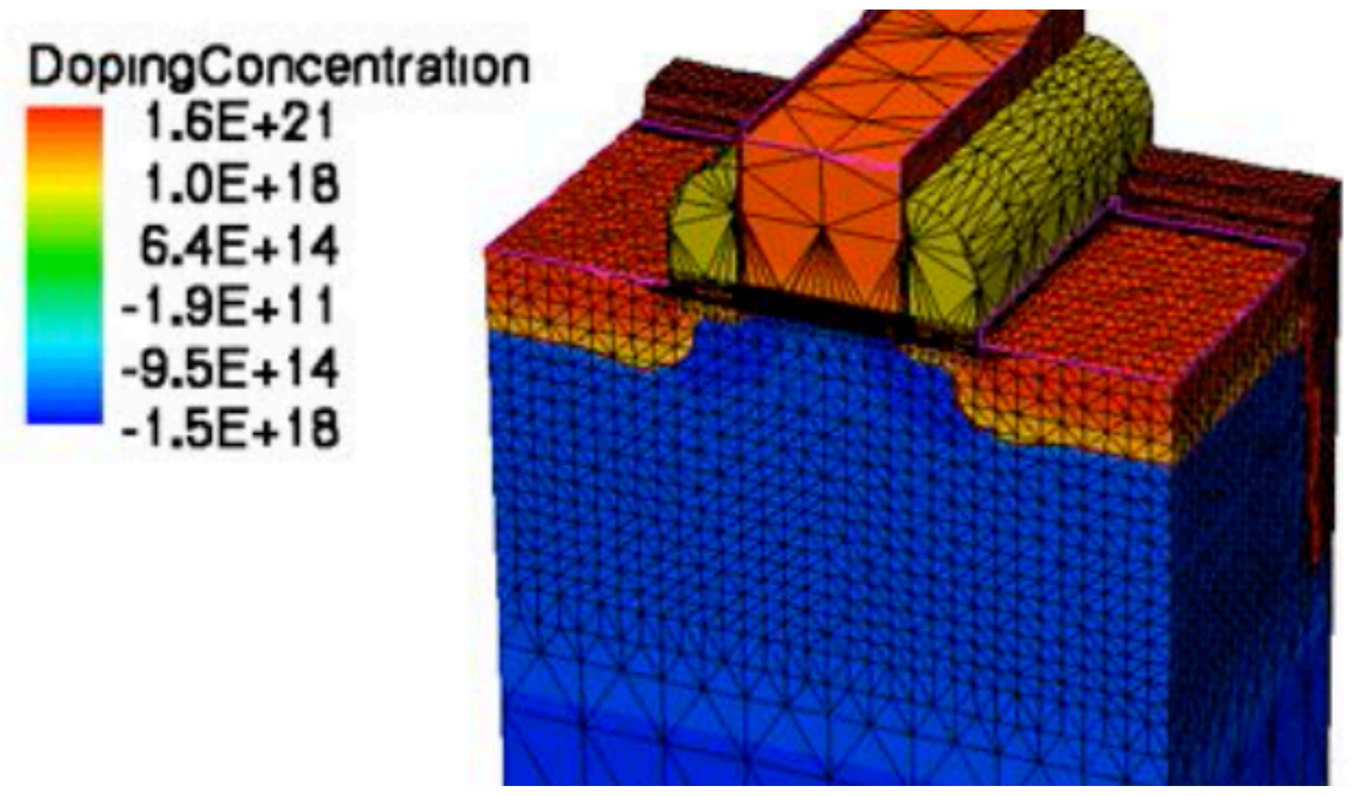
OnLattice kMC application

➢ Methodology of industrial interest

- to optimize manufacturing processes and reduce costs
- to simulate processes and minimize defects

SYNOPSYS®

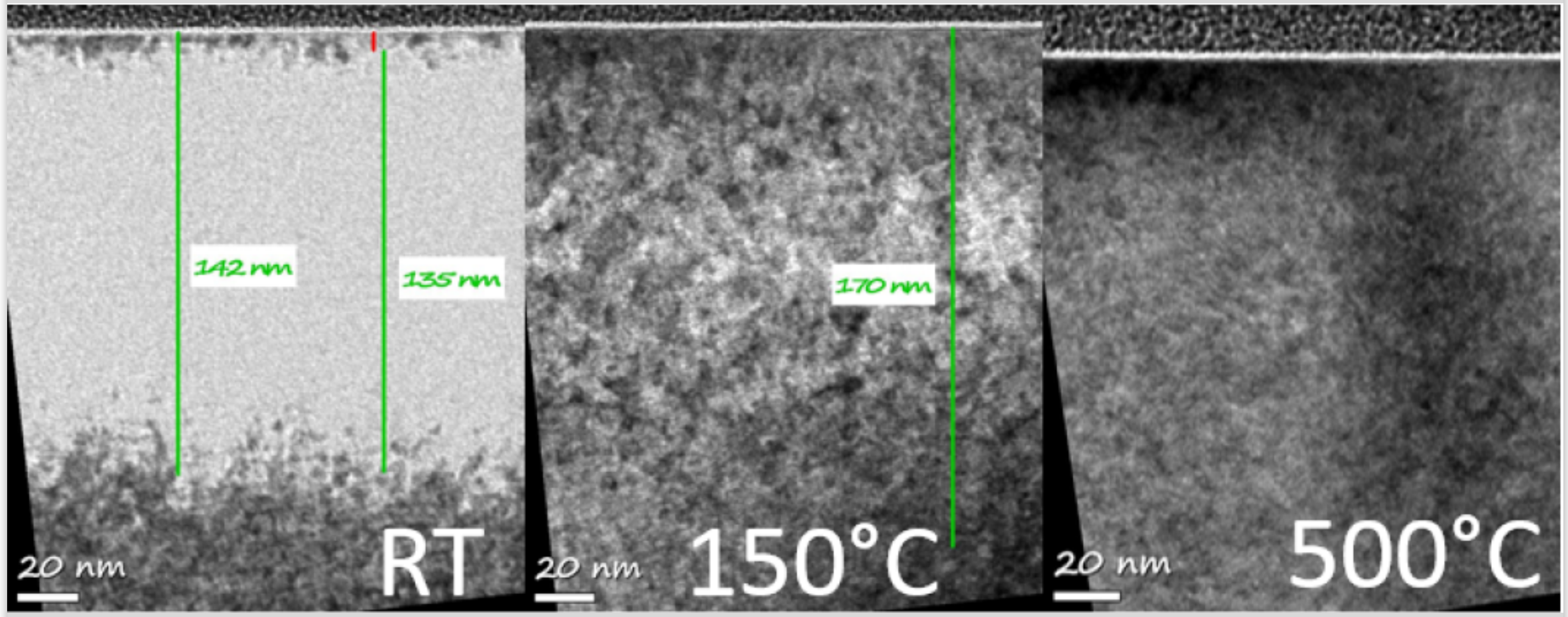
TCAD



OnLattice kMC application

Implantation of As in Silicon

EXPERIMENTAL EVIDENCES

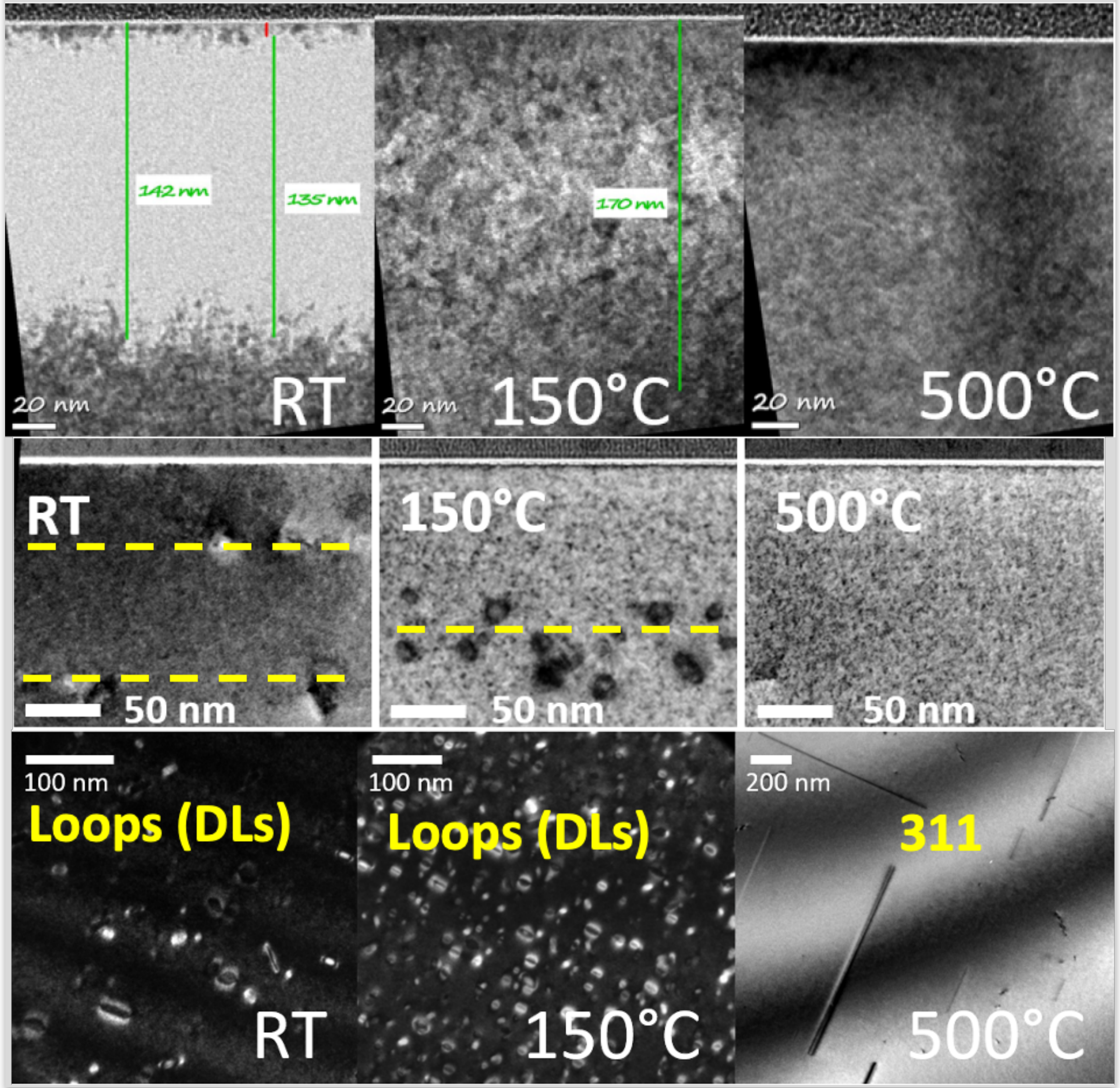


TEM as-implanted

- Amorphous layer in RT implant
- No amorphous layer for 150°C and 500°C implants
- Damaged area in 150 °C case

OnLattice kMC application
Implantation of As in Silicon

EXPERIMENTAL EVIDENCES



TEM as-implanted

- Amorphous layer in RT implant
- No amorphous layer for 150°C and 500°C implants
- Damaged area in 150 °C case

TEM post-annealing

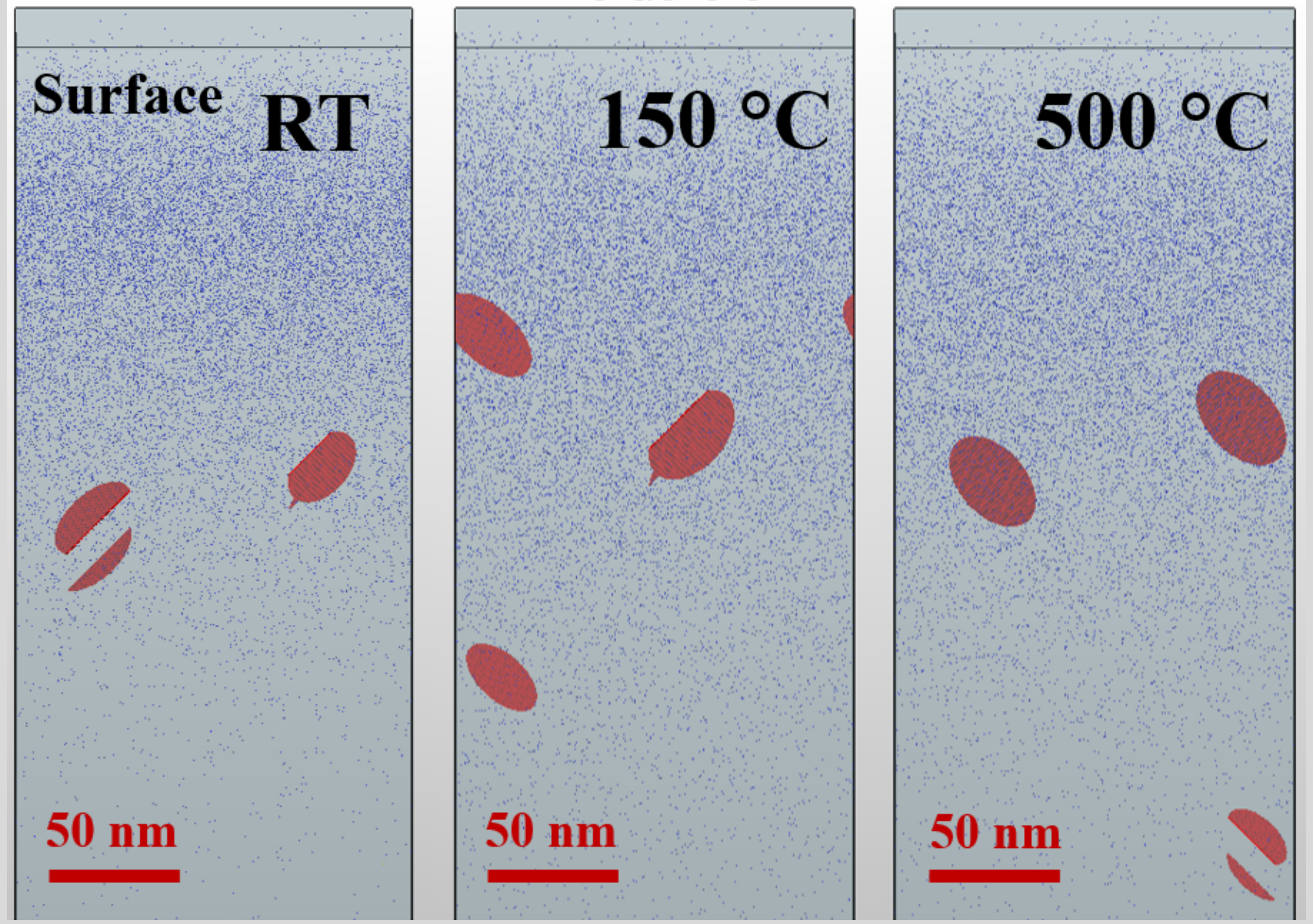
- **Defect location**: Amorphous layer has an impact on the depth of the defects
- **Defect type**: Dislocation loops (DLs) in RT and 150°C / {311} defects in 500°C case
- **Defect density**: More interstitials trapped in RT and 150°C than in the 500°C

I in DLs		I in {311}
RT	150°C	500 °C
1.4 10^{14} cm^{-2}	1.6 10^{14} cm^{-2}	5.4 10^{12} cm^{-2}

OnLattice kMC application

Implantation of As in Silicon

Modeling – KMC default calibration



KMC post-annealing

Defect type:

- **RT**: DLs simulated **agree with TEM**
- **150°C**: DLs simulated **agree with TEM**
- **500°C**: DLs simulated **different from {311} in TEM**

Defect density

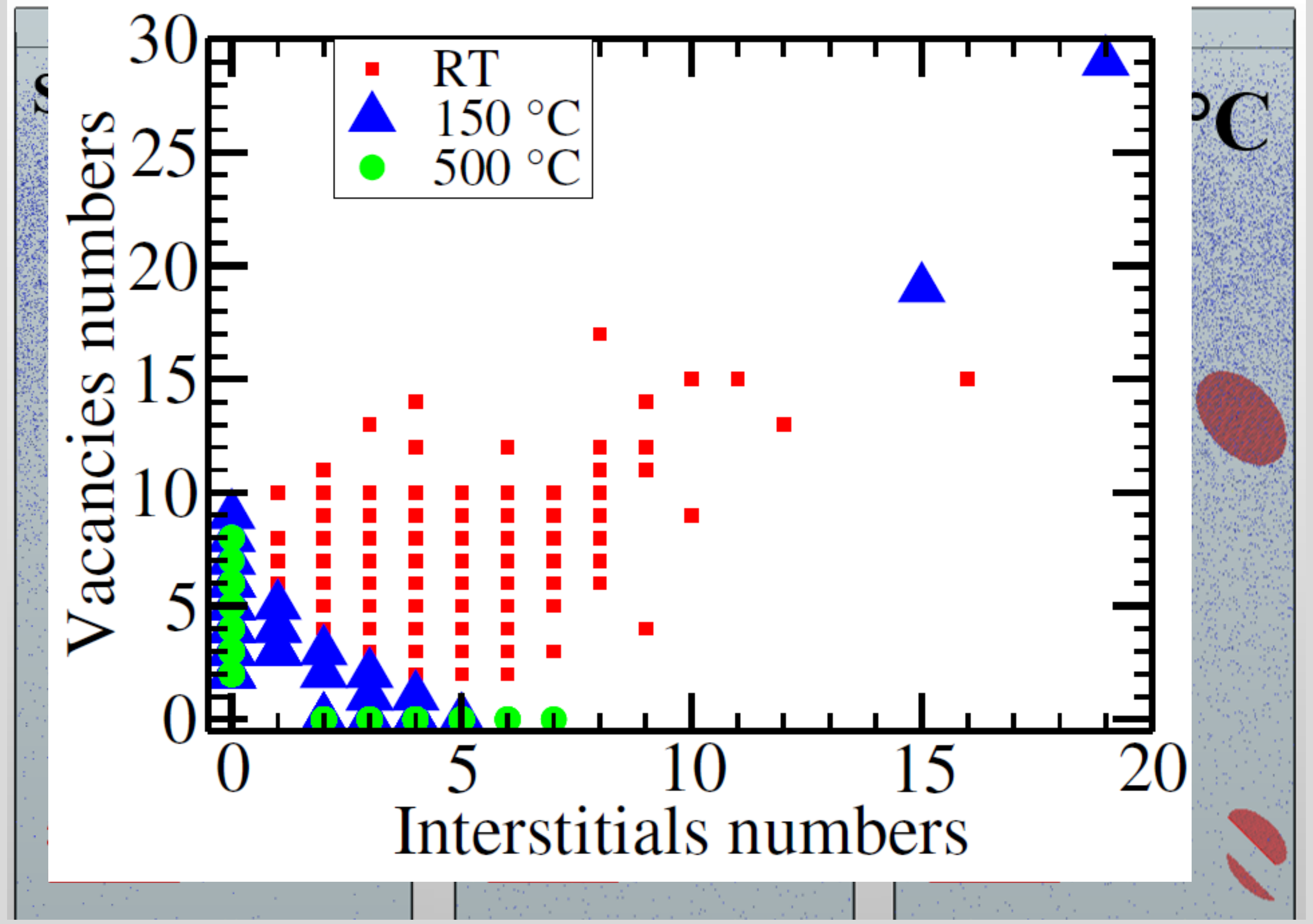
	RT	150°C	500°C
TEM	$1.4\ 10^{14}\ cm^{-2}$	$1.6\ 10^{14}\ cm^{-2}$	$5.1\ 10^{12}\ cm^{-2}$
KMC	$1.4\ 10^{14}\ cm^{-2}$ ✓	$2.4\ 10^{14}\ cm^{-2}$ ✓	$1.9\ 10^{14}\ cm^{-2}$ X

KMC simulations are accurate for RT and 150°C cases but not for 500°C implants

OnLattice kMC application

Implantation of As in Silicon

Modeling – KMC default calibration



What parameter to change?

- Same annealing sequence after the implants
  → **The difference should be observed in the as-implanted state of the material**

- What are the defects produced by a single As atom implanted?

  In 500°C SMIC with size < 7

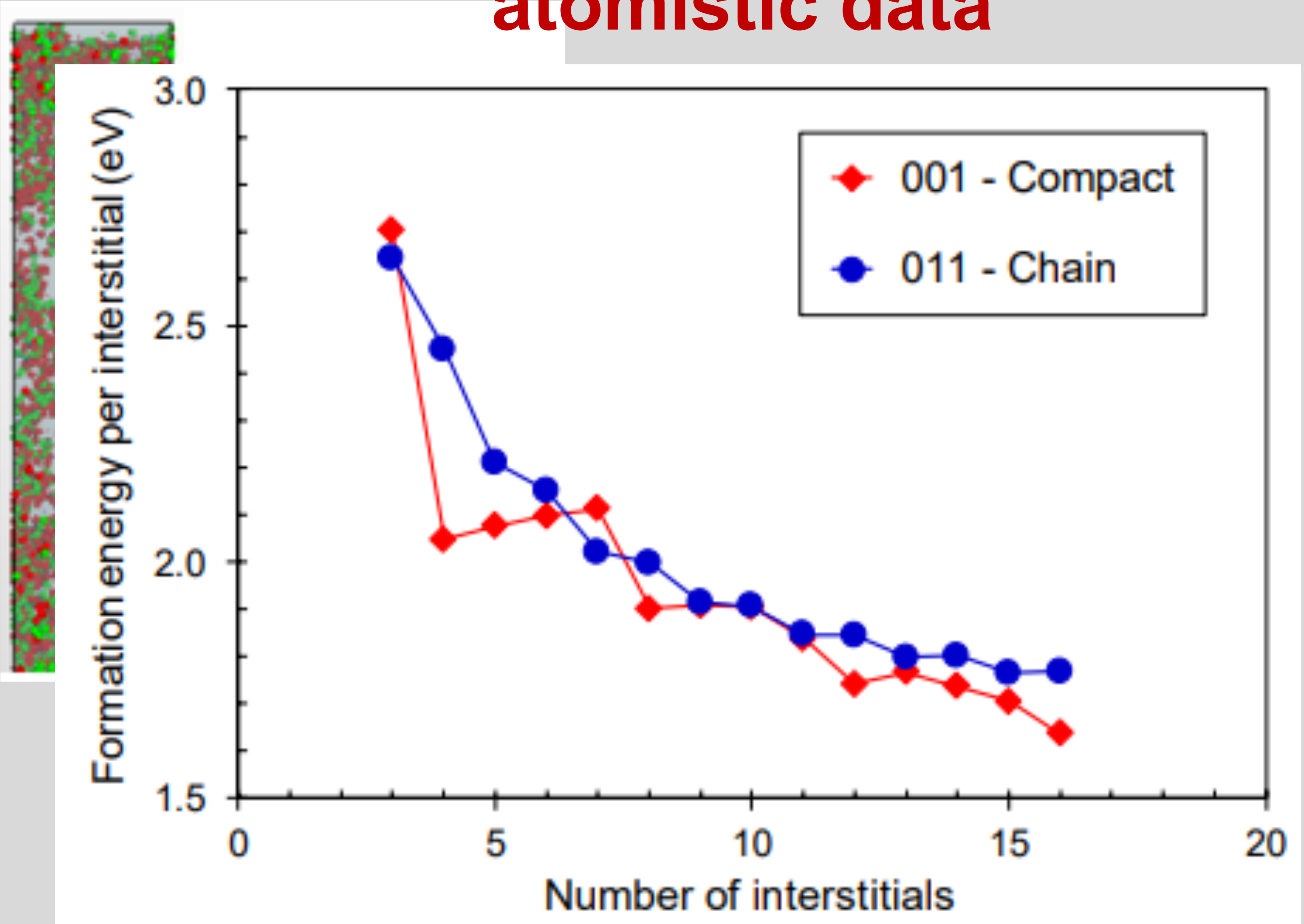
Calibration of small interstitial clusters (SMICs) is required

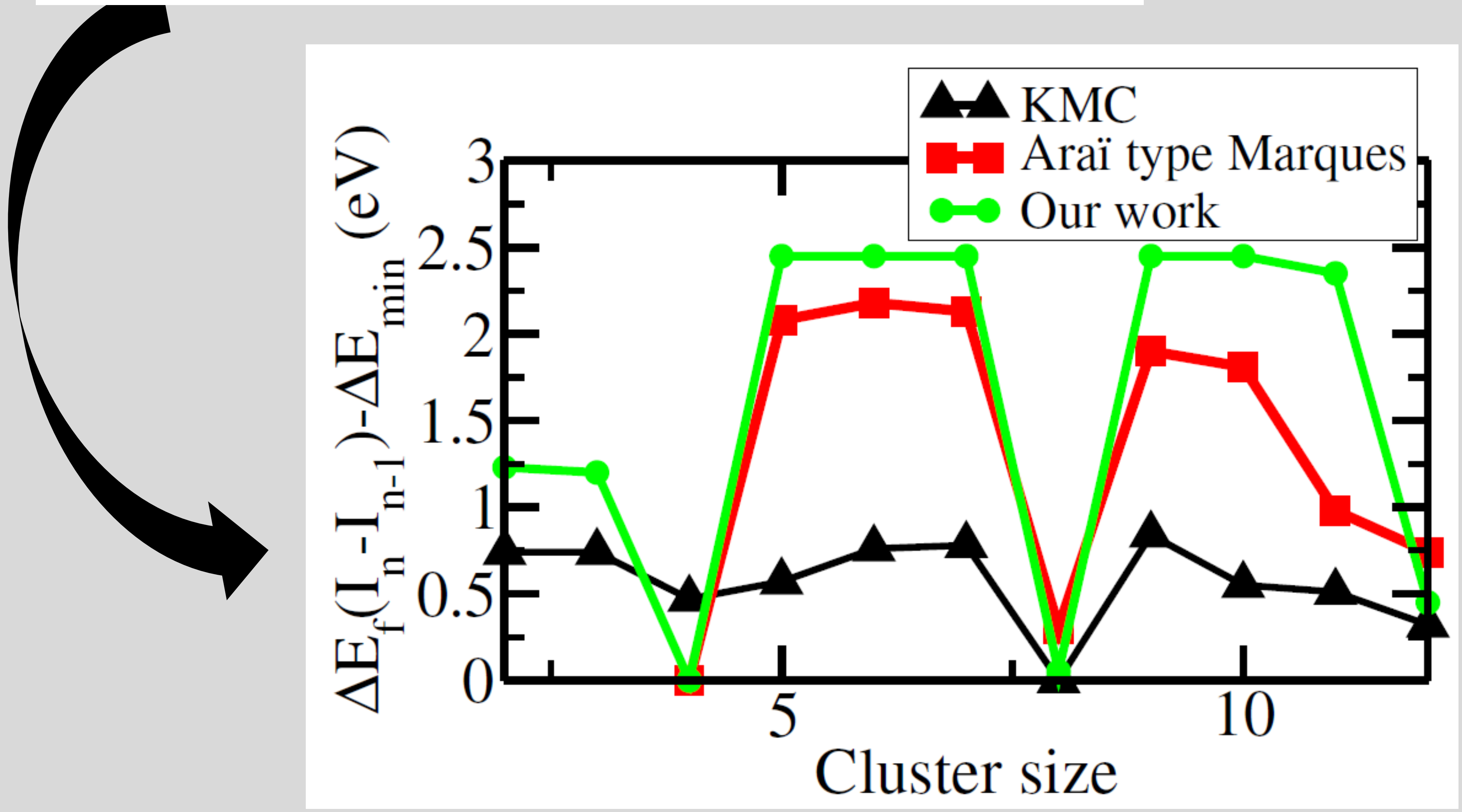
Histogram of interstitials-vacancies defects for an As atom implanted at RT, 150°C and 500°C

OnLattice kMC application

Implantation of As in Silicon

Modeling – KMC – New calibration from atomistic data



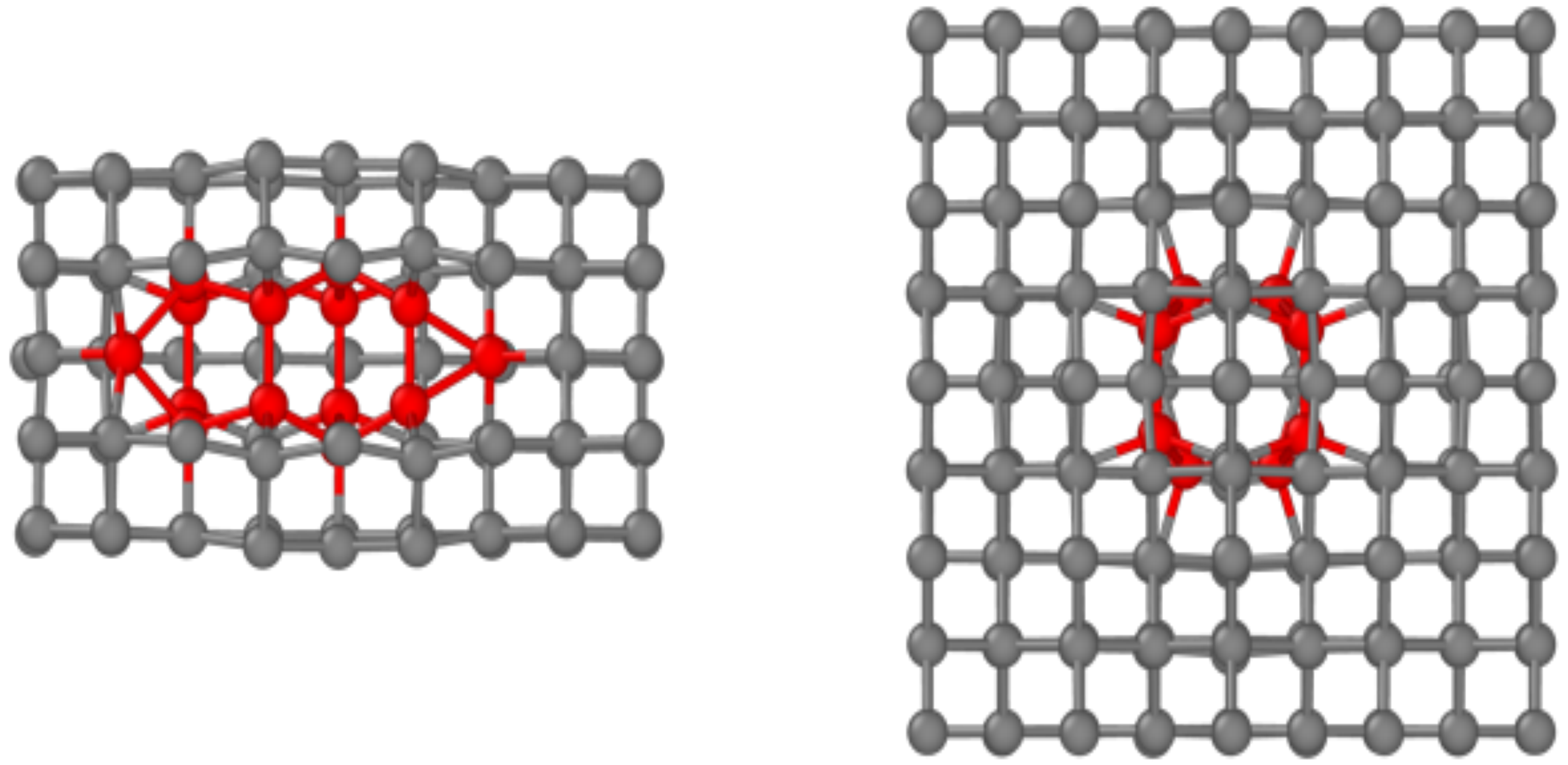


What are the SMIC energies in literature ?

- Two SMIC types in literature :

Chain-like and Araï like (001-Compact)

Ref. MD simulation - Marques (2019). Acta Mater. 166, 192-201

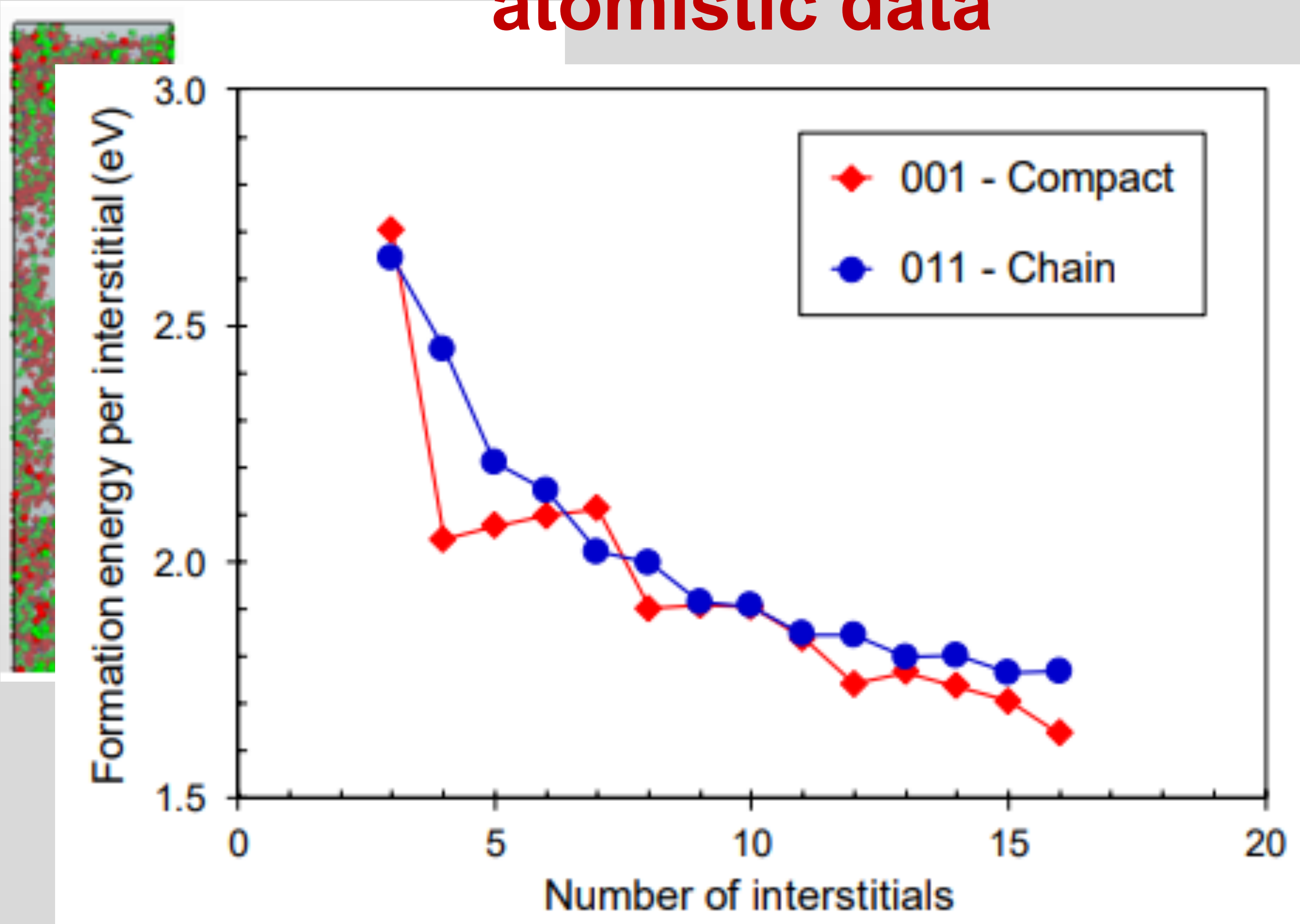


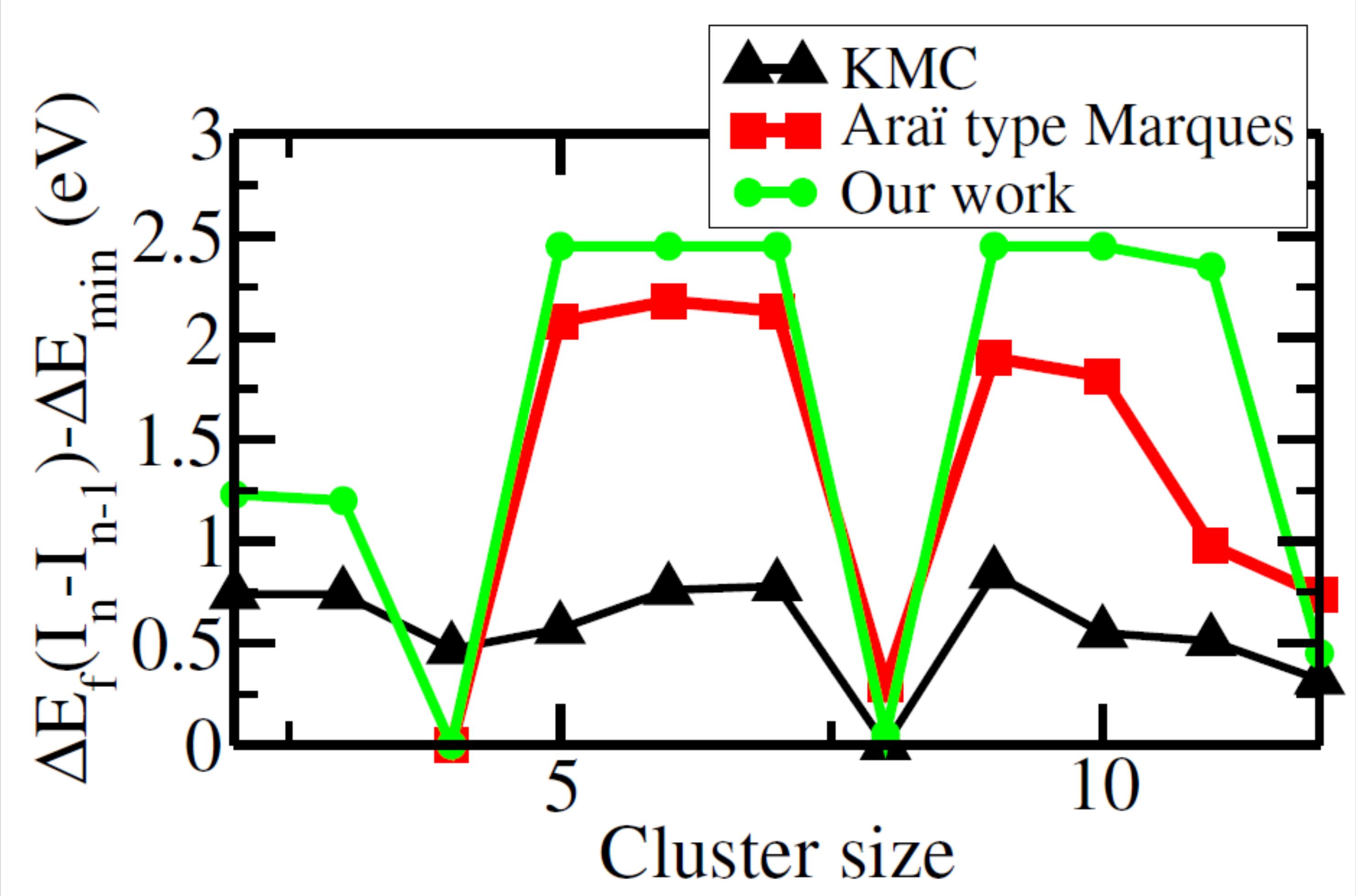
SMIC of Araï type should be considered in detail

OnLattice kMC application

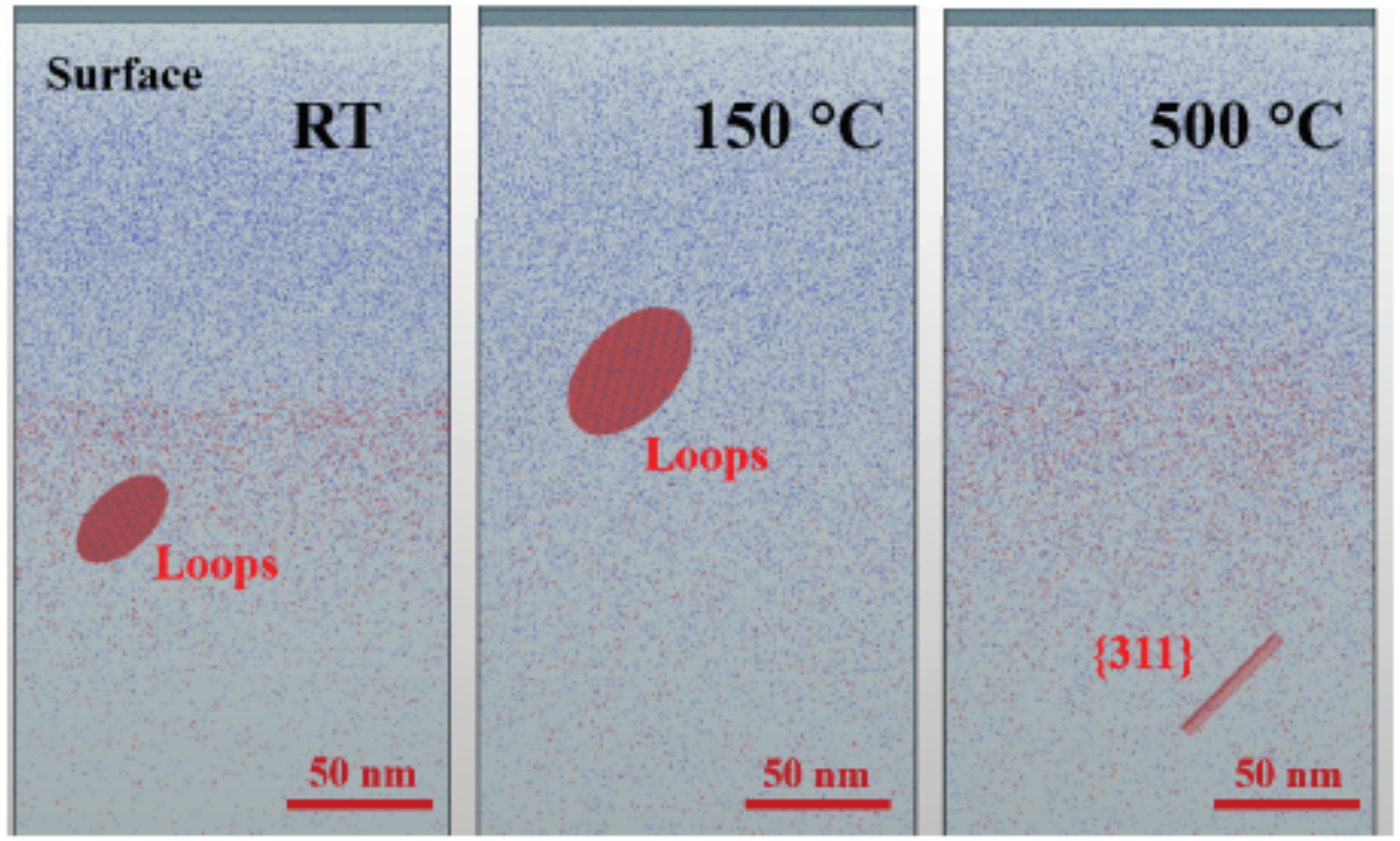
Implantation of As in Silicon

Modeling – KMC – New calibration from atomistic data





What if the MD trend for Araï SMICs energies is used to fit experiments?



✔ **Defect type accurate**

n 311

0 °C

TEI

0^{12} cm^{-2}

KM

0^{12} cm^{-2}

Reproduce the extended defects trends for the 3 implantations

➢ **Need for fine atomistic ingredient in TCAD**

OnLattice kMC: Resume

➢ List of events

Known in advance: KMC only uses events
Assumption that **events are well defined and that their transition rates** are constant or can be calculated

However, in complex systems, events may be dependent on the environment and local conditions, making them difficult to describe. Same for long range interaction.

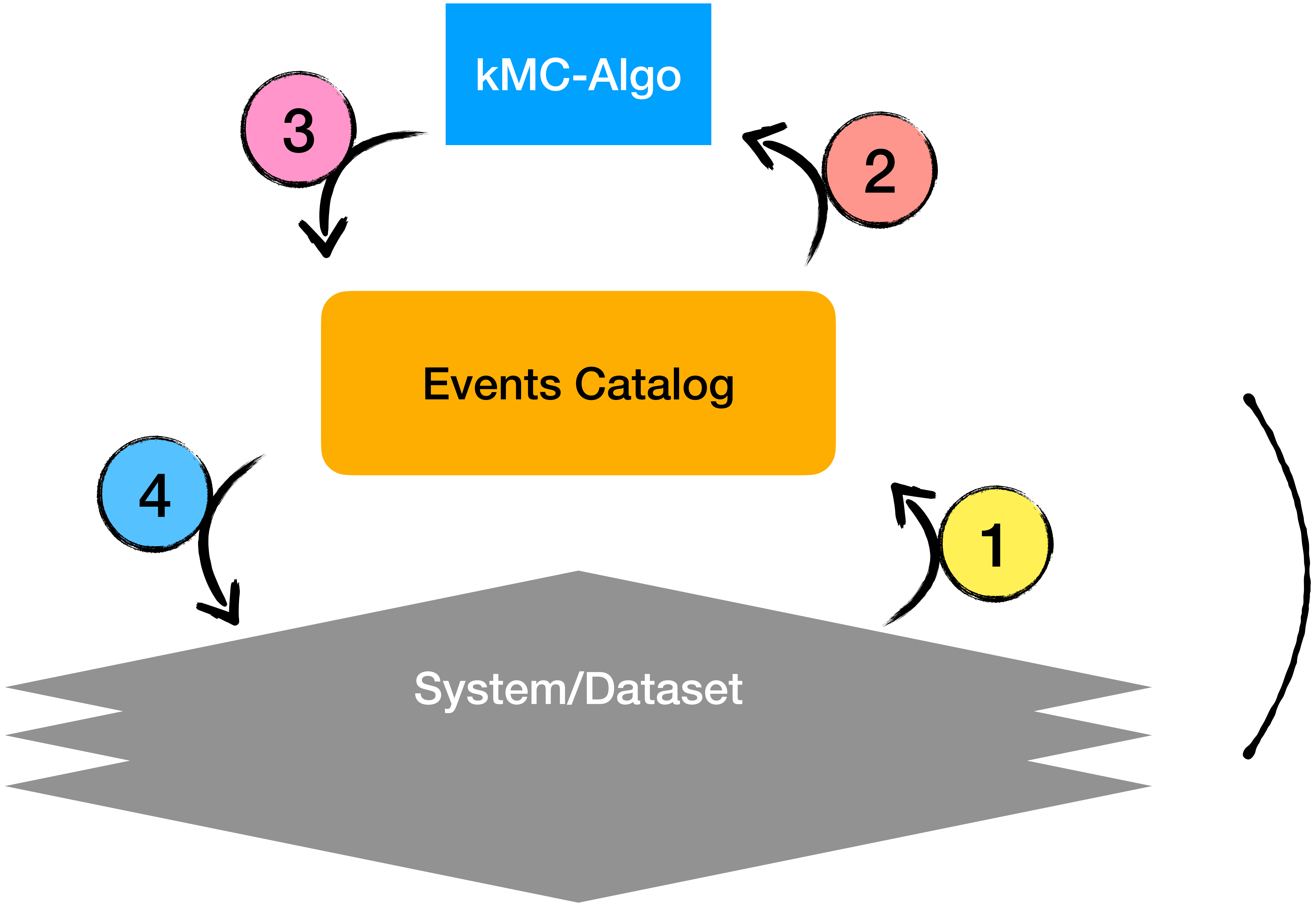
Limits:
Calculation of Rates: The accuracy of KMC is highly dependent on the *accuracy of transition rates*. Calculating these rates accurately can be difficult, especially for complex systems.

Modelling error: *Incorrect transition rates* can lead to bad results, reducing the reliability of simulations.

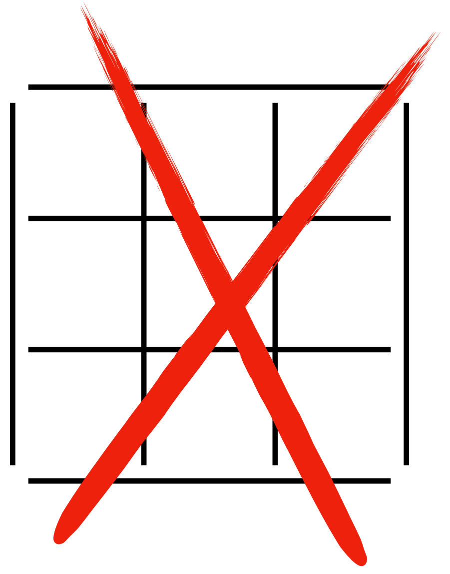
Model accuracy: The importance of *accuracy in transition rate models* for reliable results.

Beyond OnLattice kMC

➢ On the fly kinetic Monte Carlo / Adaptive KMC (AKMC)



Approximation of the system by E/F Engine

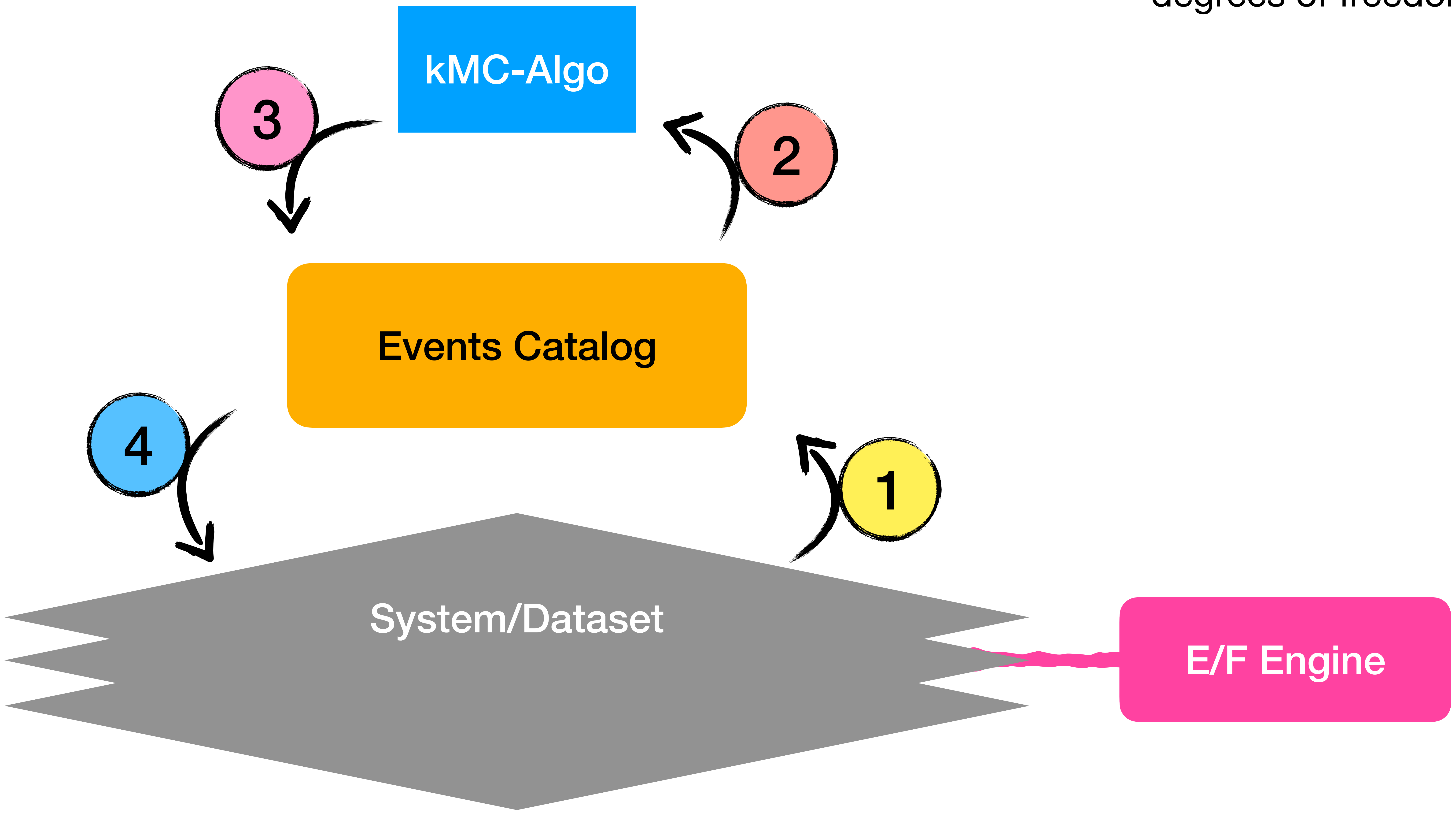


+ events catalog

Beyond OnLattice kMC

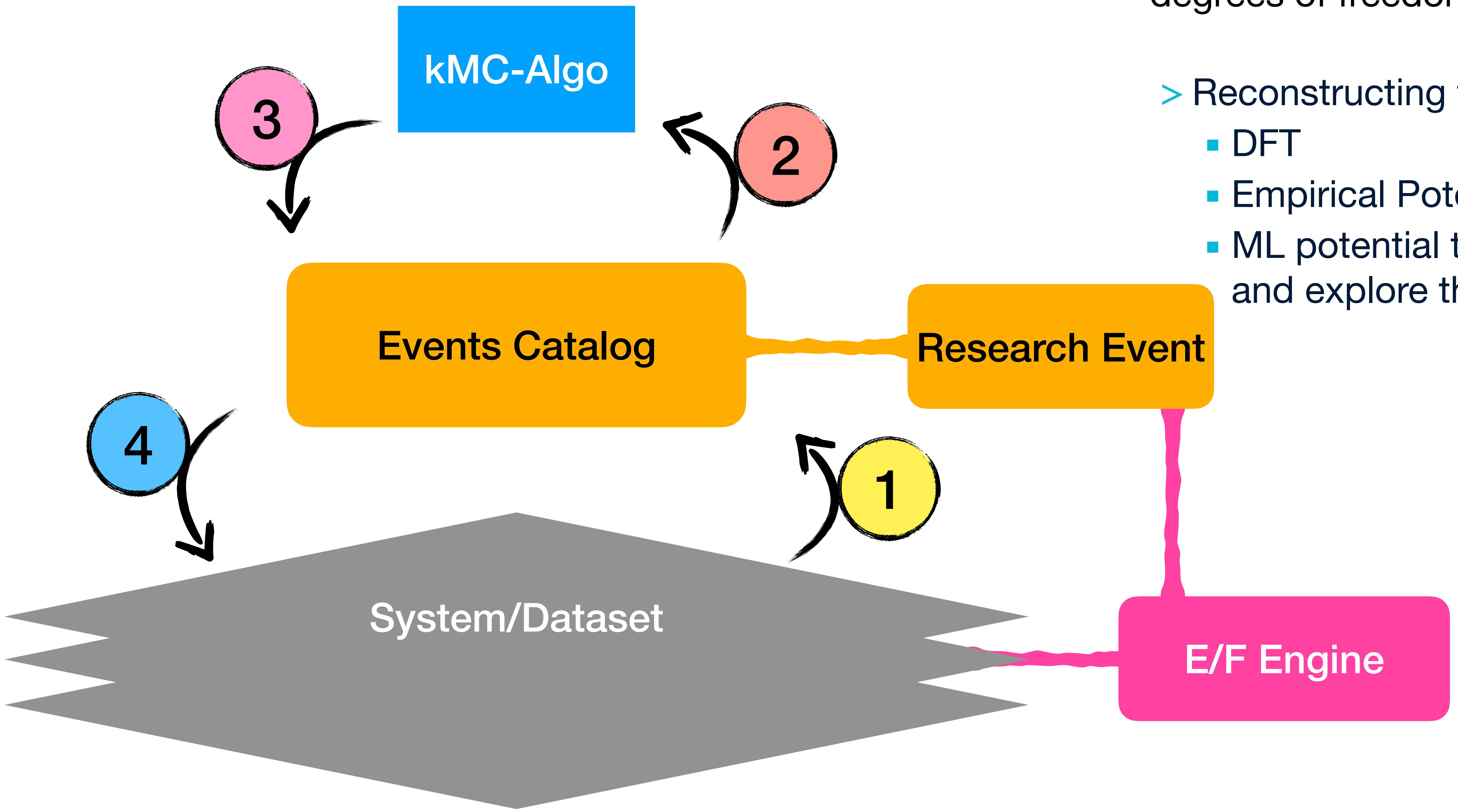
- On the fly kinetic Monte Carlo / Adaptive KMC (AKMC)

The **representation of the event** has to take into account the degrees of freedom of the E/F Engine



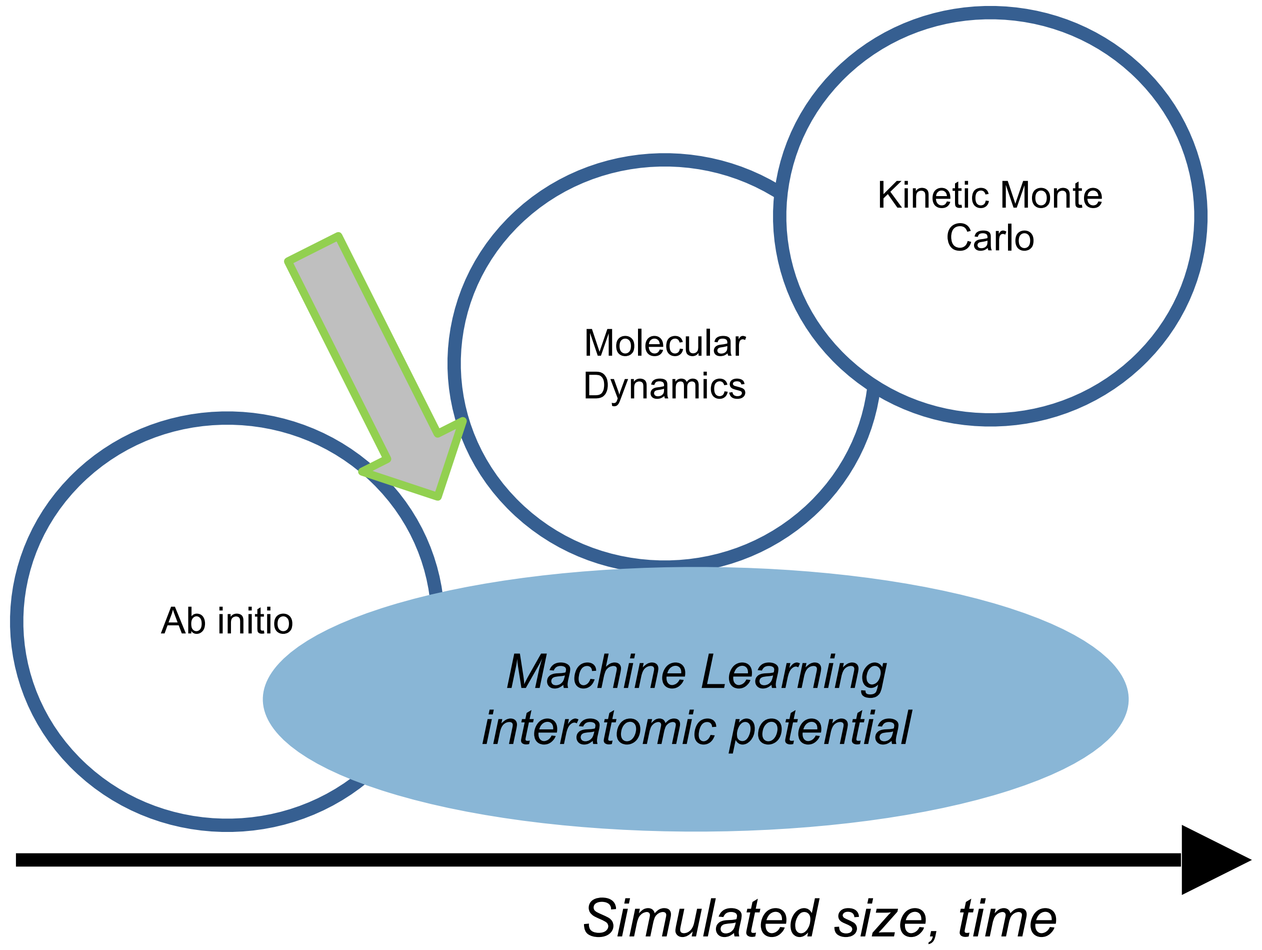
Beyond OnLattice kMC

➢ On the fly kinetic Monte Carlo / Adaptive KMC (AKMC)



The **representation of the event** has to take into account the degrees of freedom of the E/F Engine

> Reconstructing the catalogue on the fly
- DFT
- Empirical Potential (MD)
- ML potential to improve the estimation of transition rates and explore the state space more efficiently



Beyond OnLattice kMC

- On the fly kinetic Monte Carlo / Adaptive KMC (AKMC)

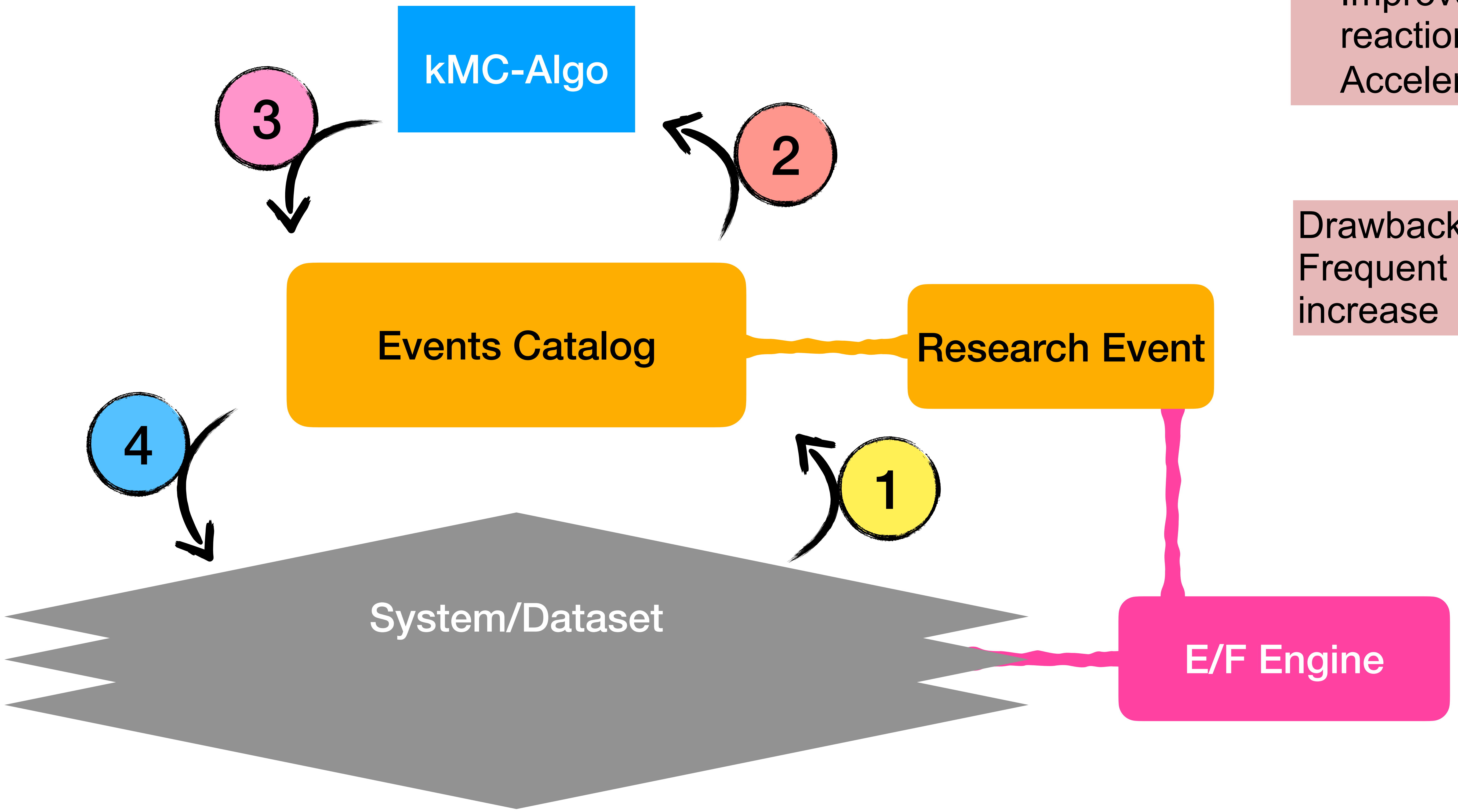
Benefits:

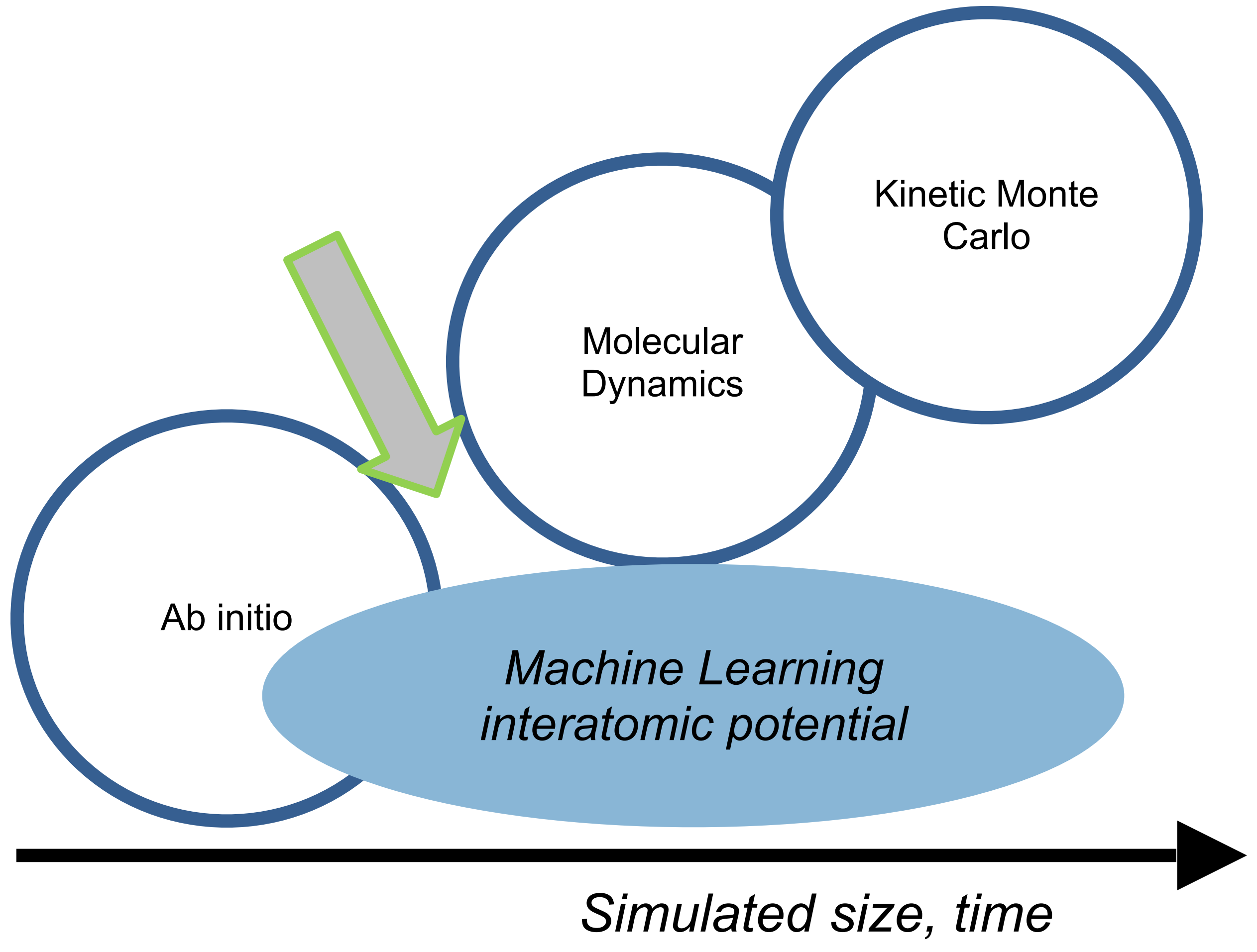
Improves the accuracy of transition rates and reaction paths.

Accelerates state space exploration.

Drawbacks:

Frequent update of the catalog, computational cost increase





OpenSource OffLattice kMC

Adaptative events catalog needs an automatic way to search new events

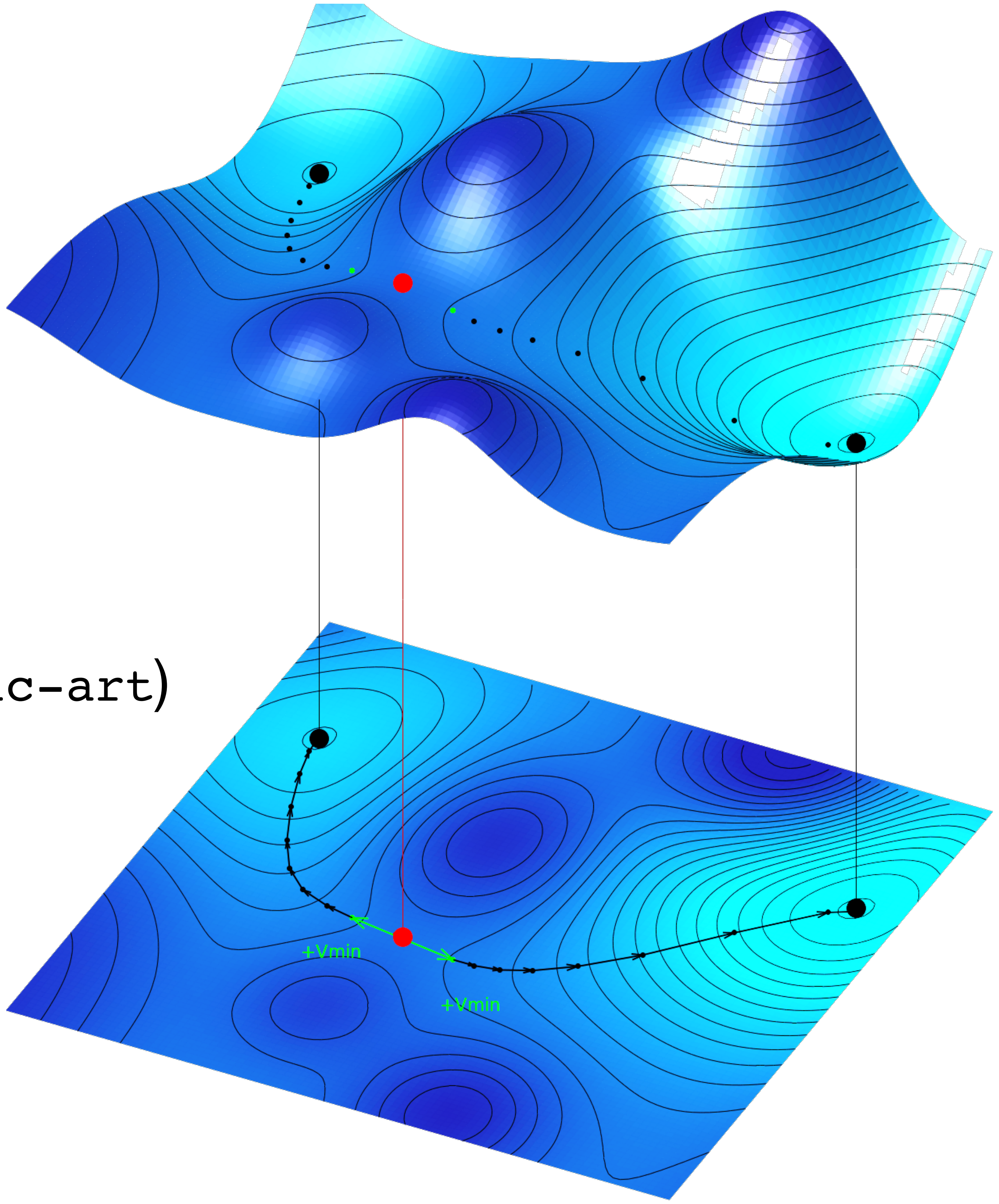
ARTn (Activation Relaxation Technique nouveau) Normand Mousseau

plugin-ARTn (https://gitlab.com/mammasmias/artn-plugin)

Software: **kART** (https://normandmousseau.com/fr/research/kinetic-art)

Dimere method (Graeme Henkelman and Hannes Jonsón)

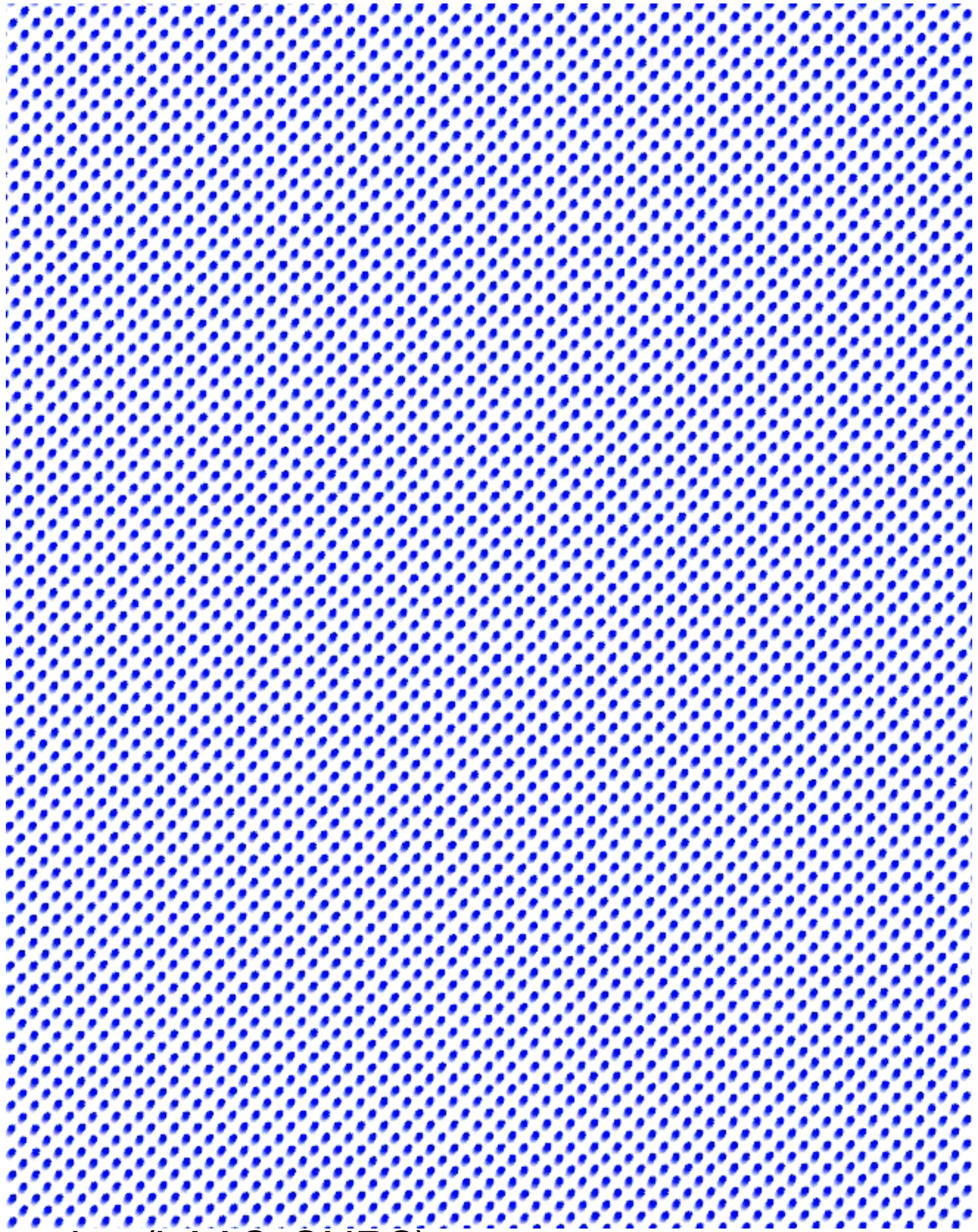
Software: **eON** (https://github.com/TheochemUI/eOn)



Accurate Transition states
Exhaustive list of events

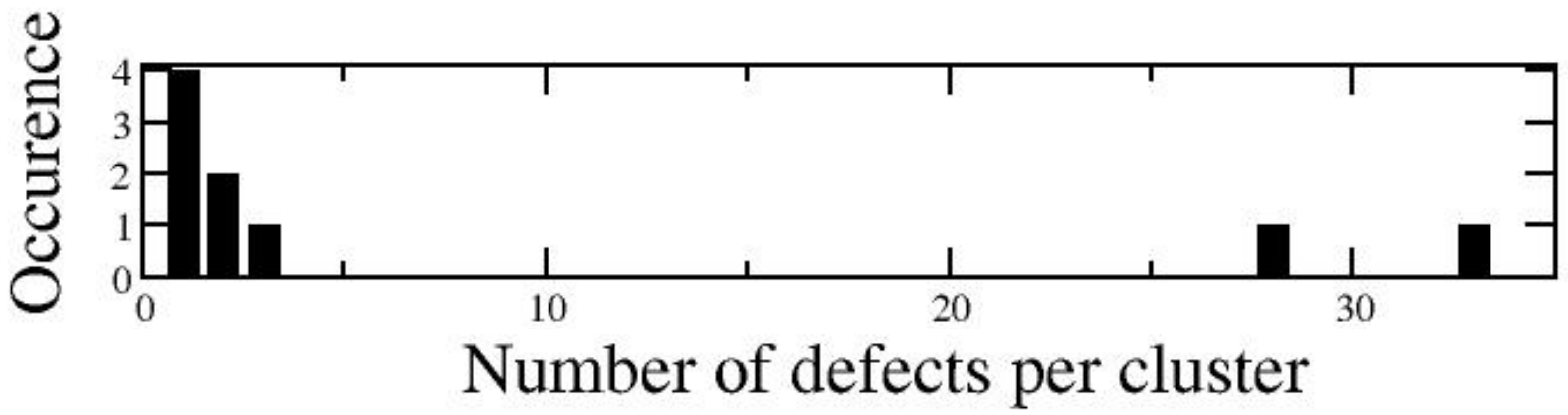
Long time simulation

> Molecular Dynamics: LAMMPS with SW Potential

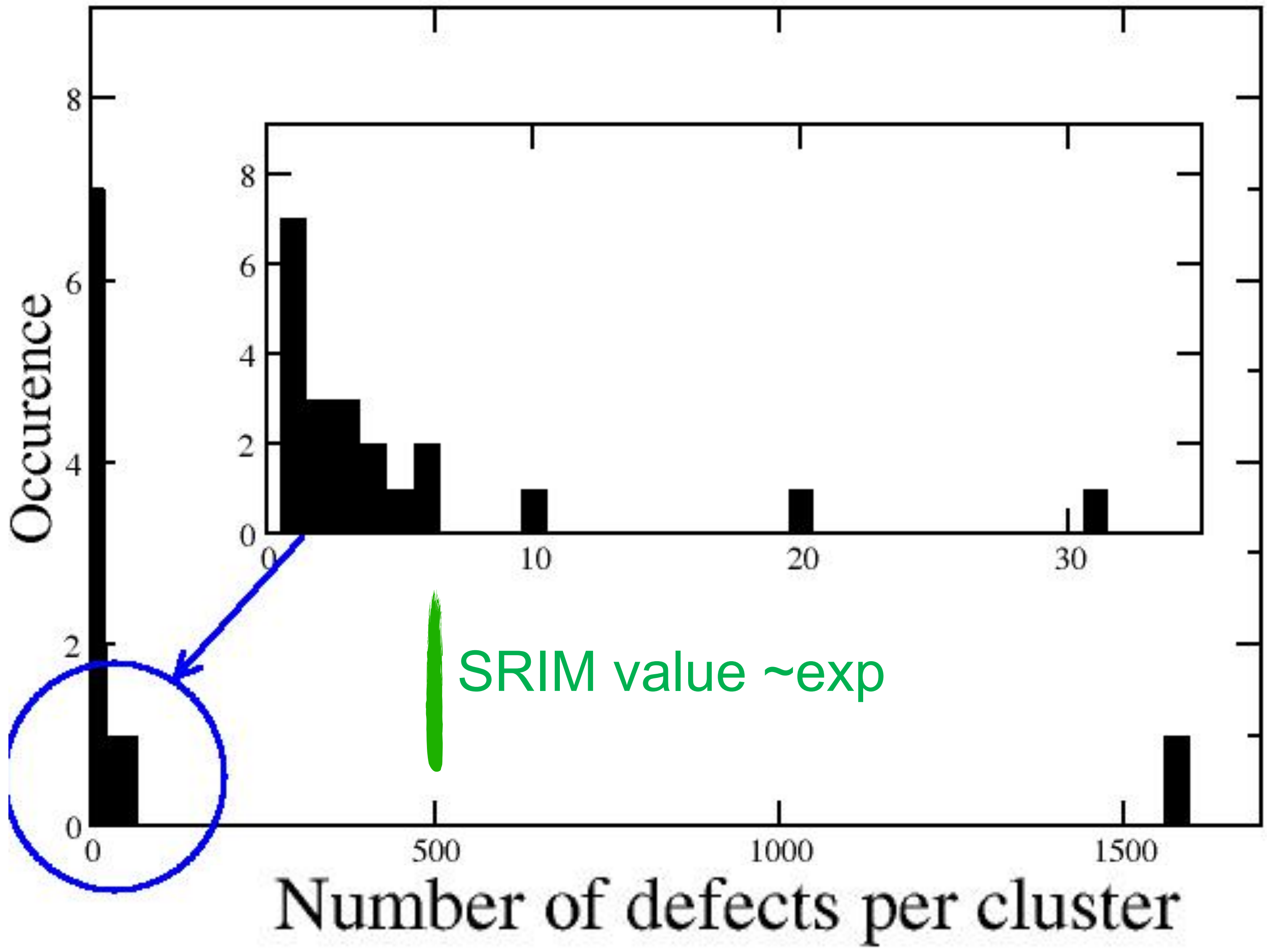


At the end of the MD simulation (1 ns)

1 keV

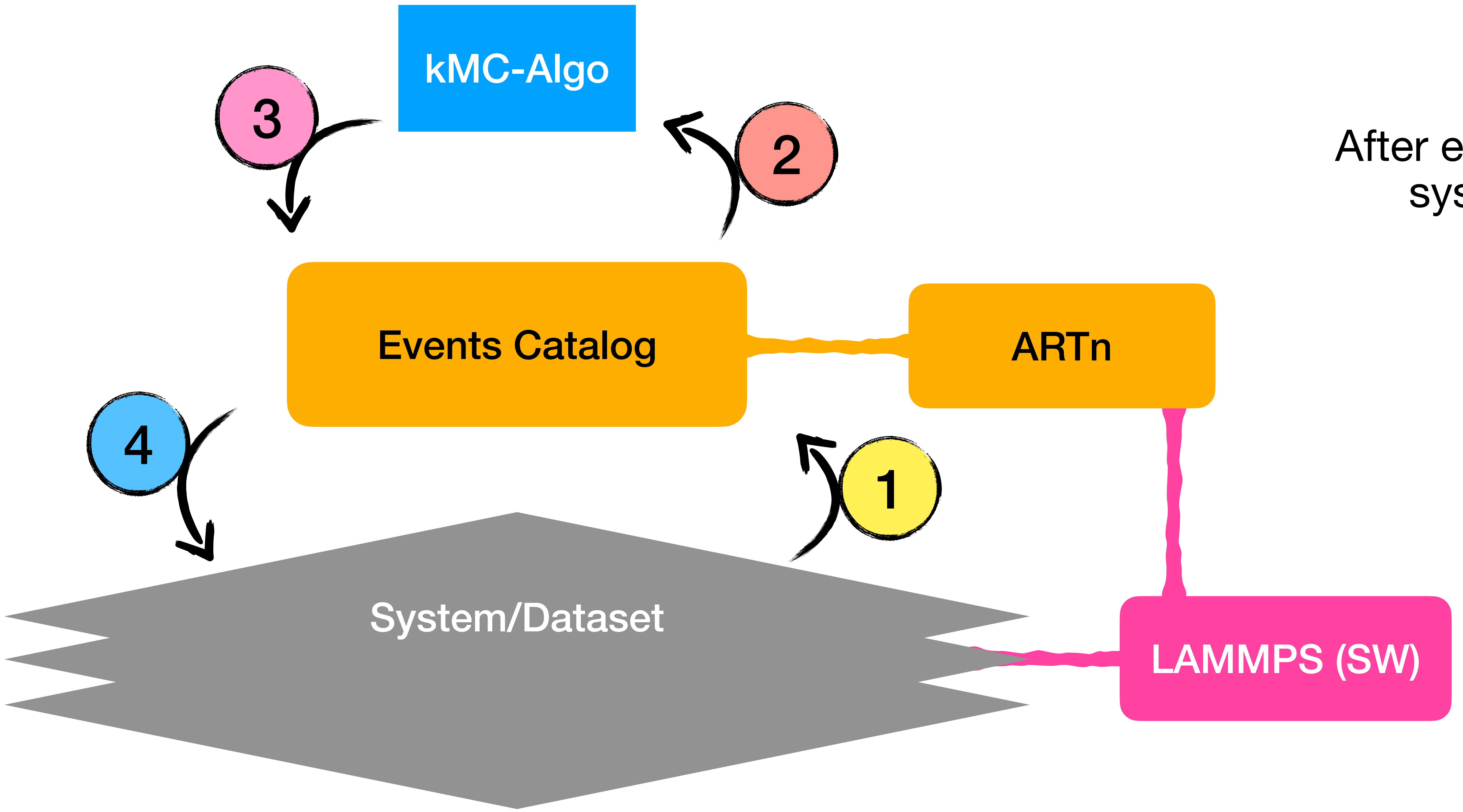


10 keV



Long time simulation

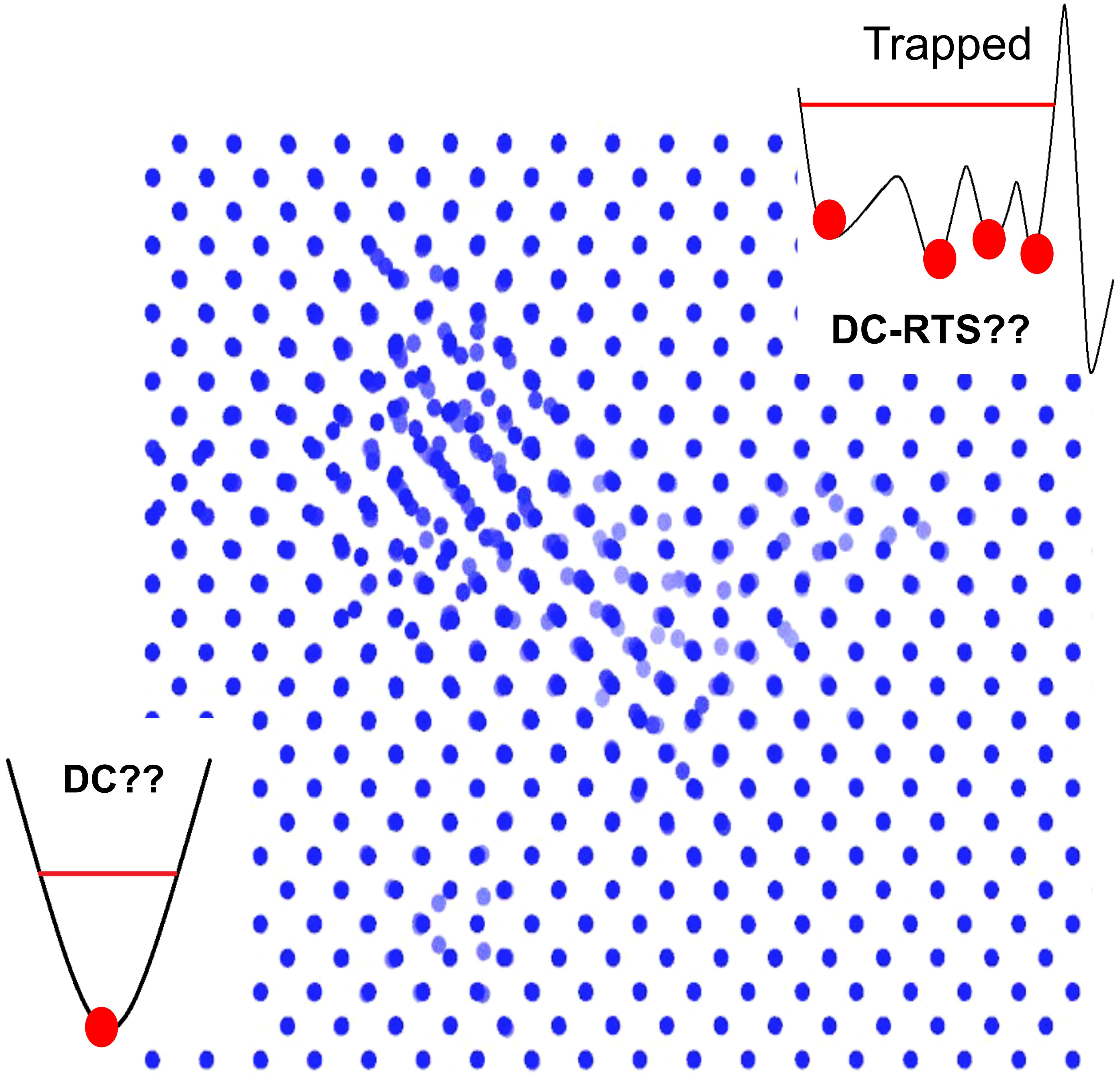
> Output of Molecular Dynamics ➜ kinetic ART (Code kMC)

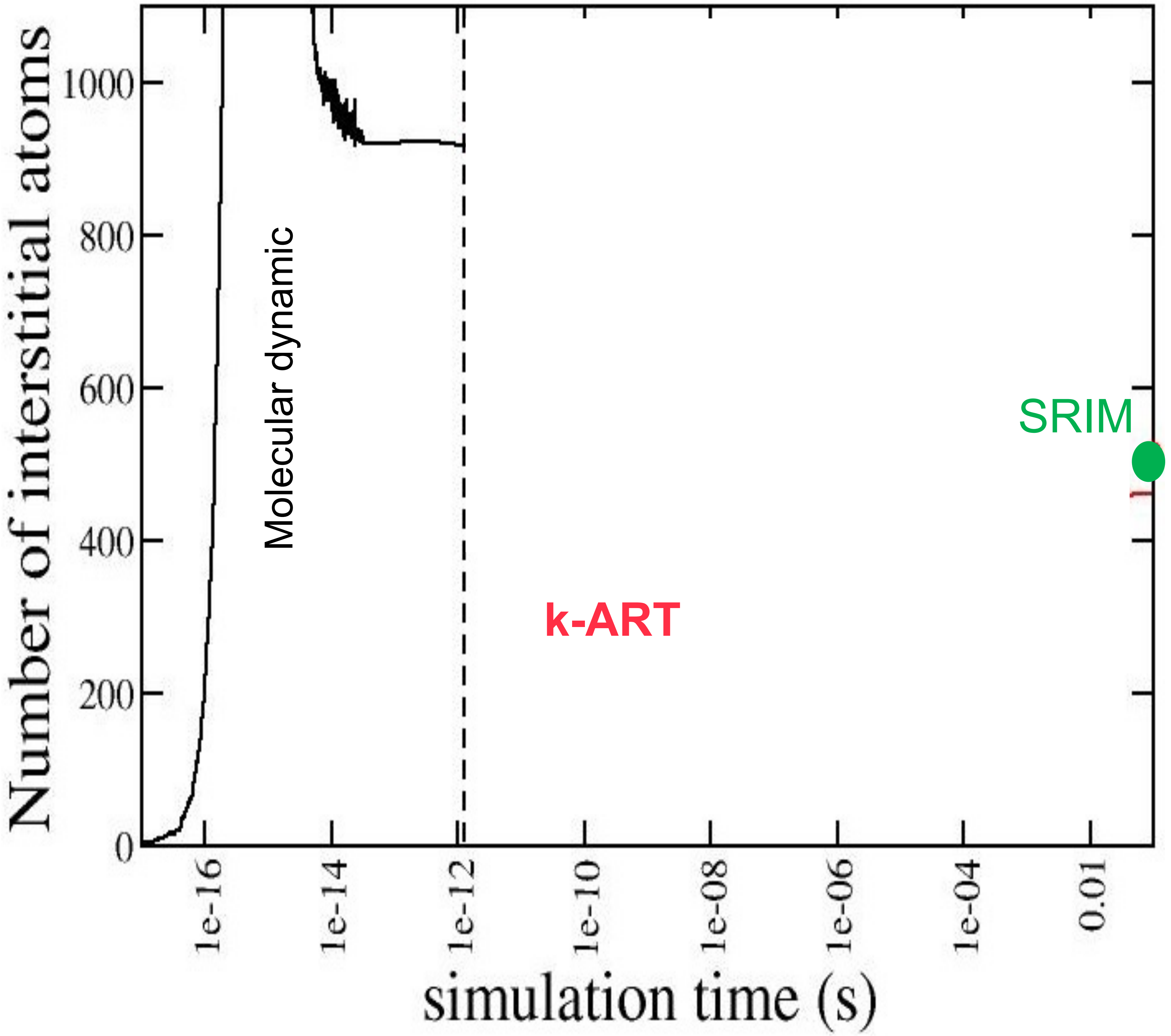


After each kMC step (event application) the system is relaxed by the E/F Engine

Long time simulation

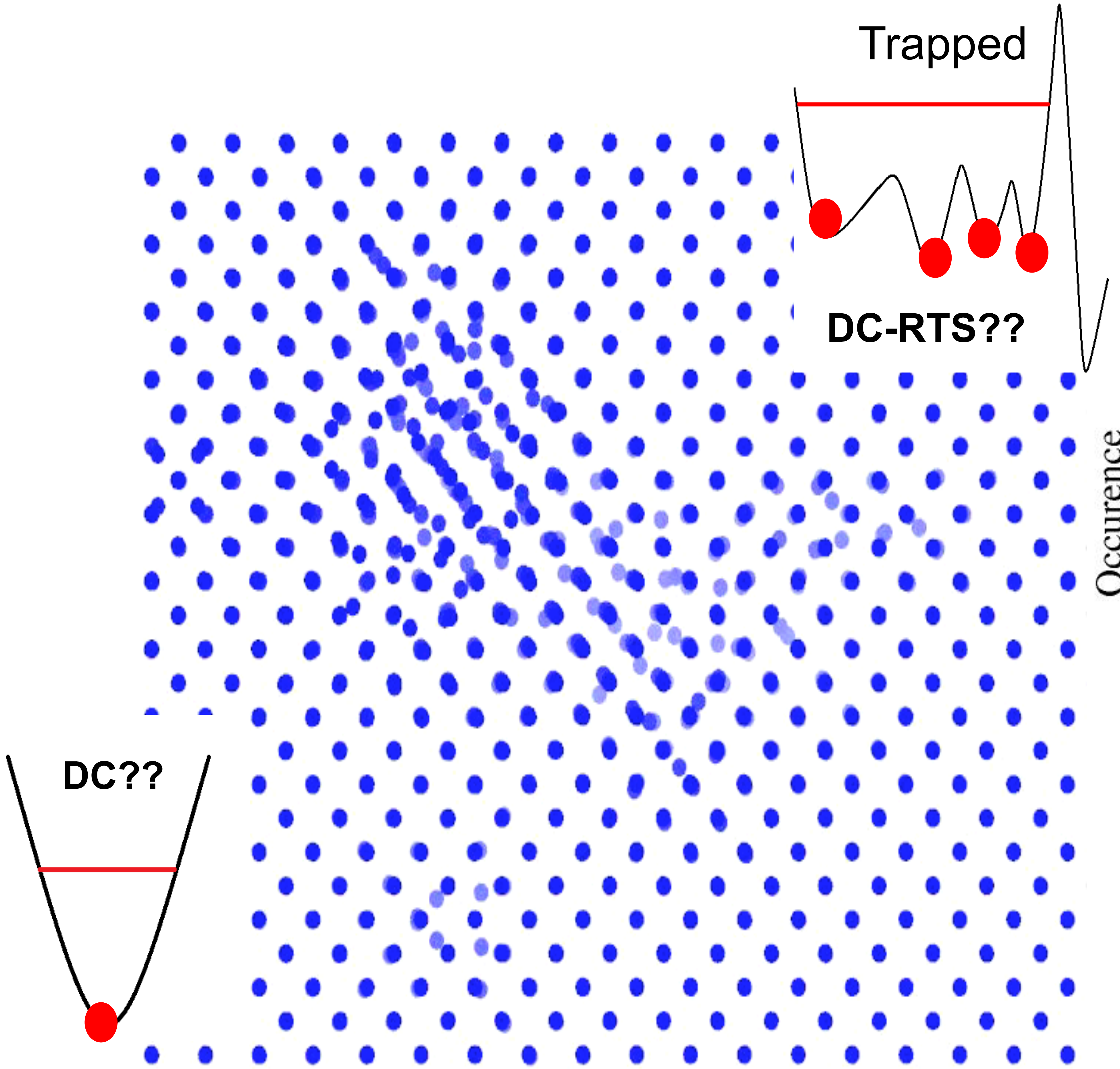
> Output of Molecular Dynamics → kinetic ART (Code kMC)





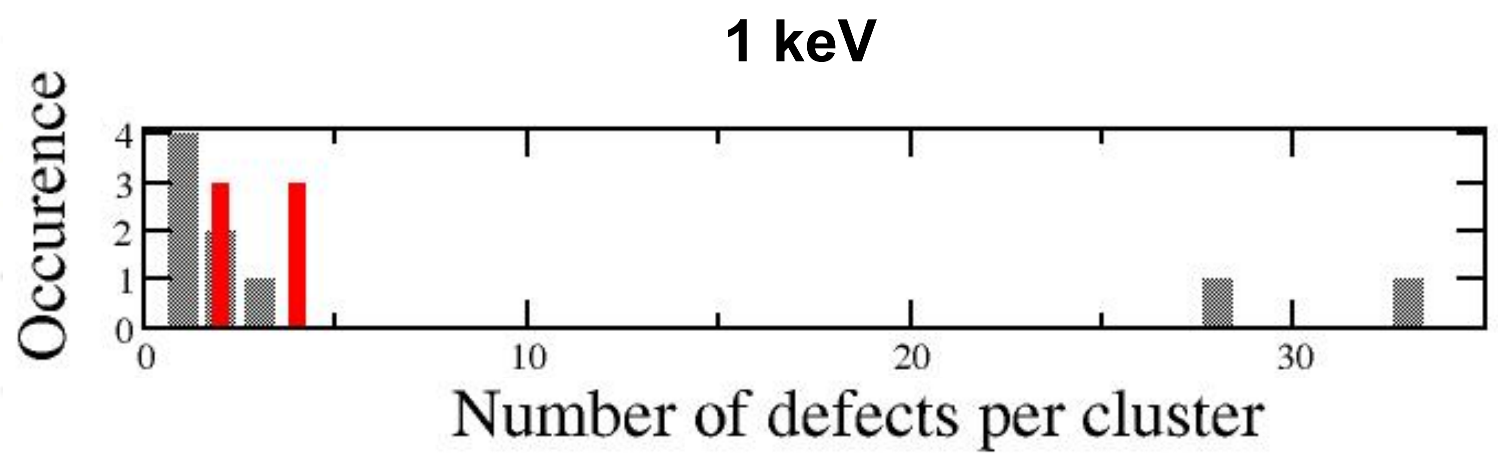
Long time simulation

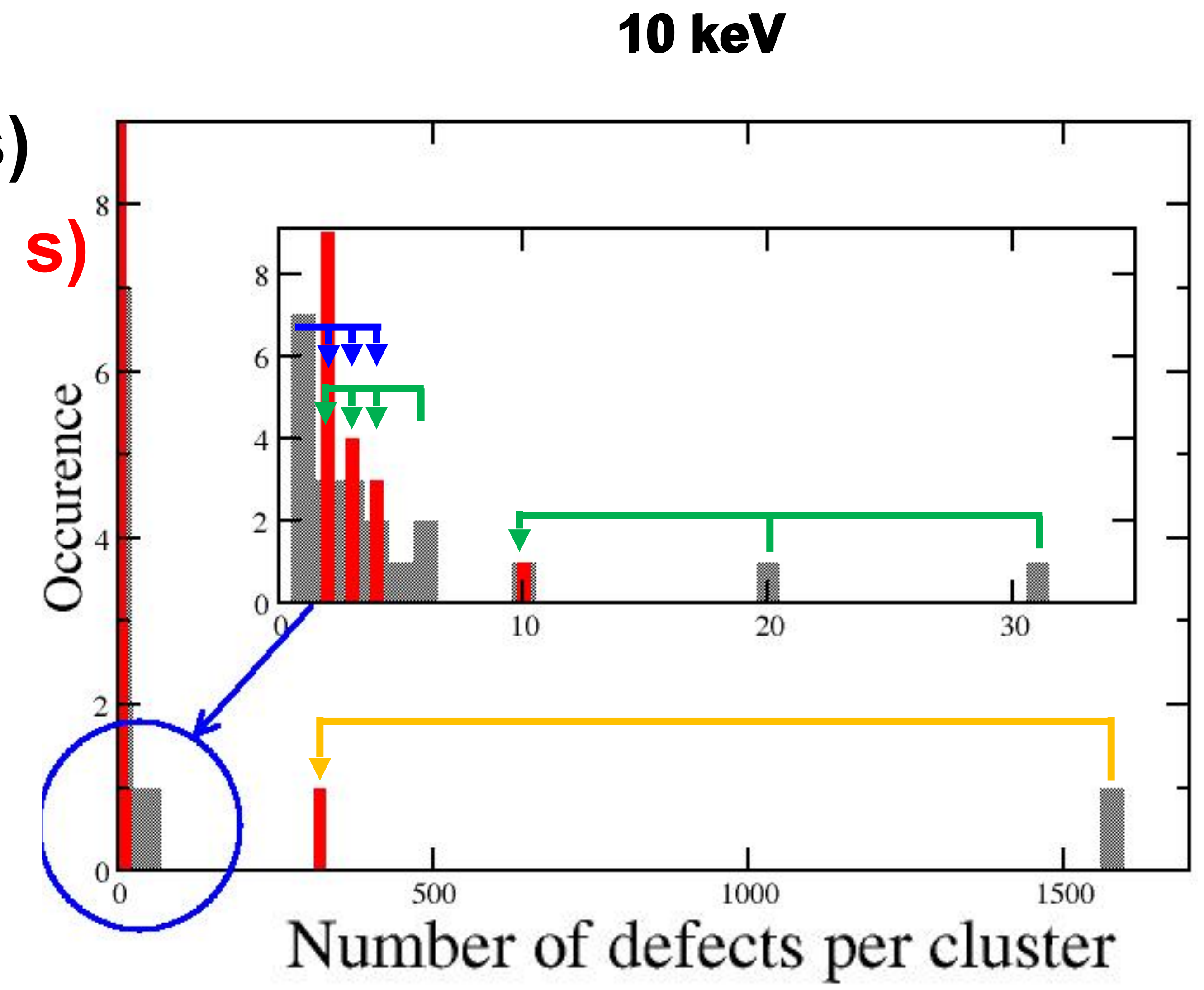
> Output of Molecular Dynamics ➜ kinetic ART (Code kMC)



At the end of the MD simulation (1 ns)

At the end of the k-ART simulation (1 s)





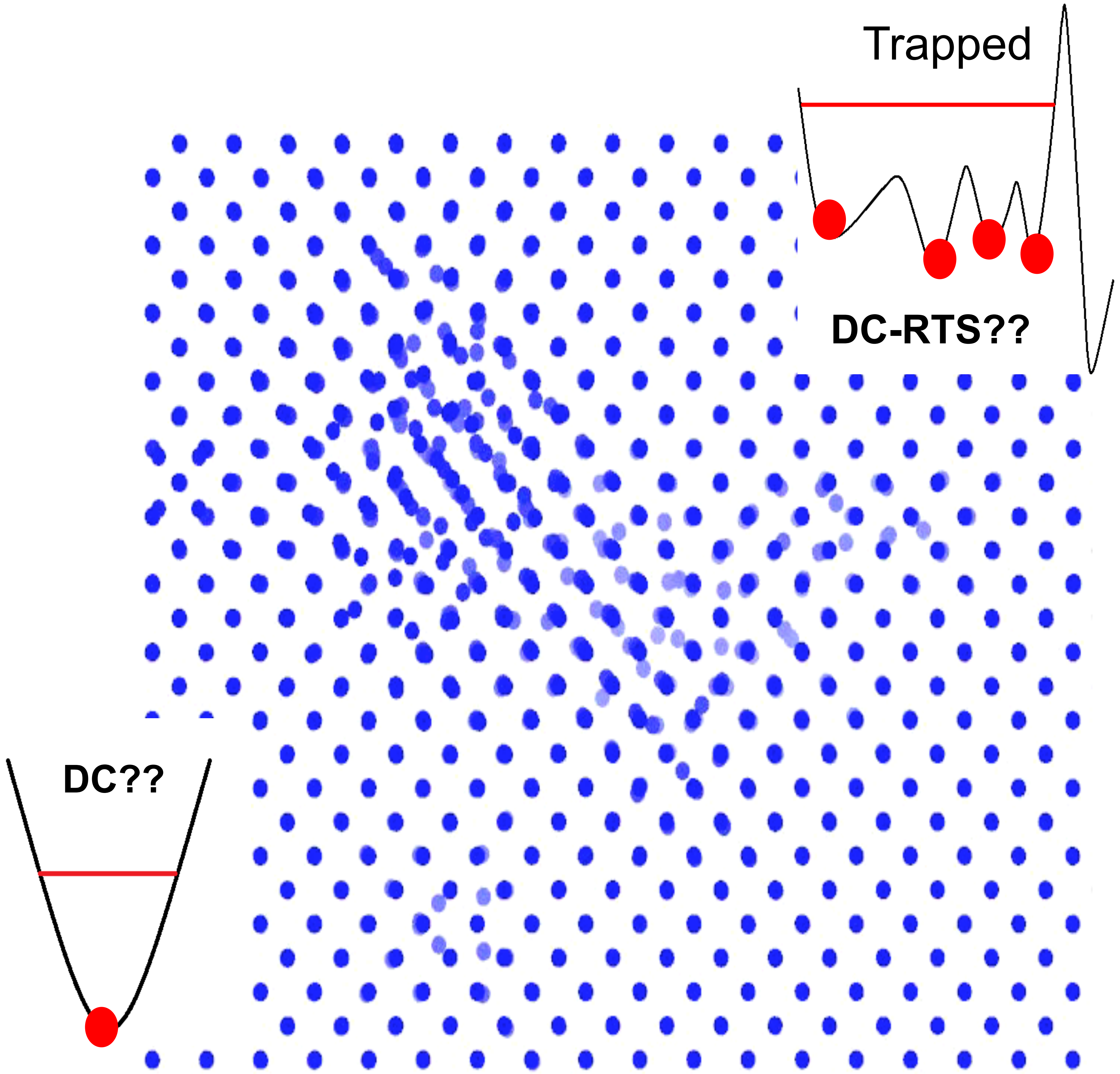
Large Sized Clusters (100 keV):
Partial healing

Medium Sized Clusters:
Healing

Point defects:

Diffusion + Agglomeration into small clusters or Recrystallisation

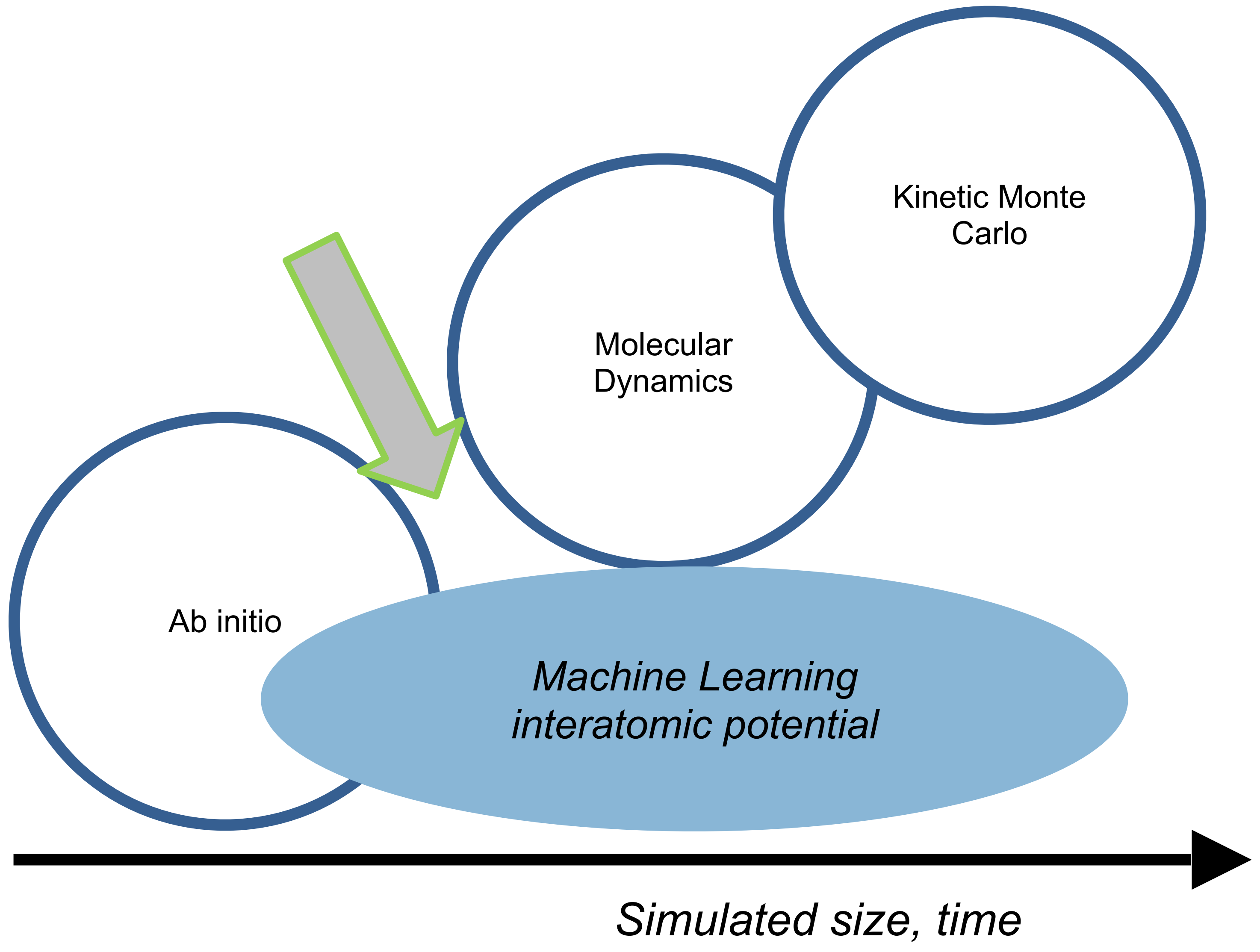
Long time simulation: Feedback



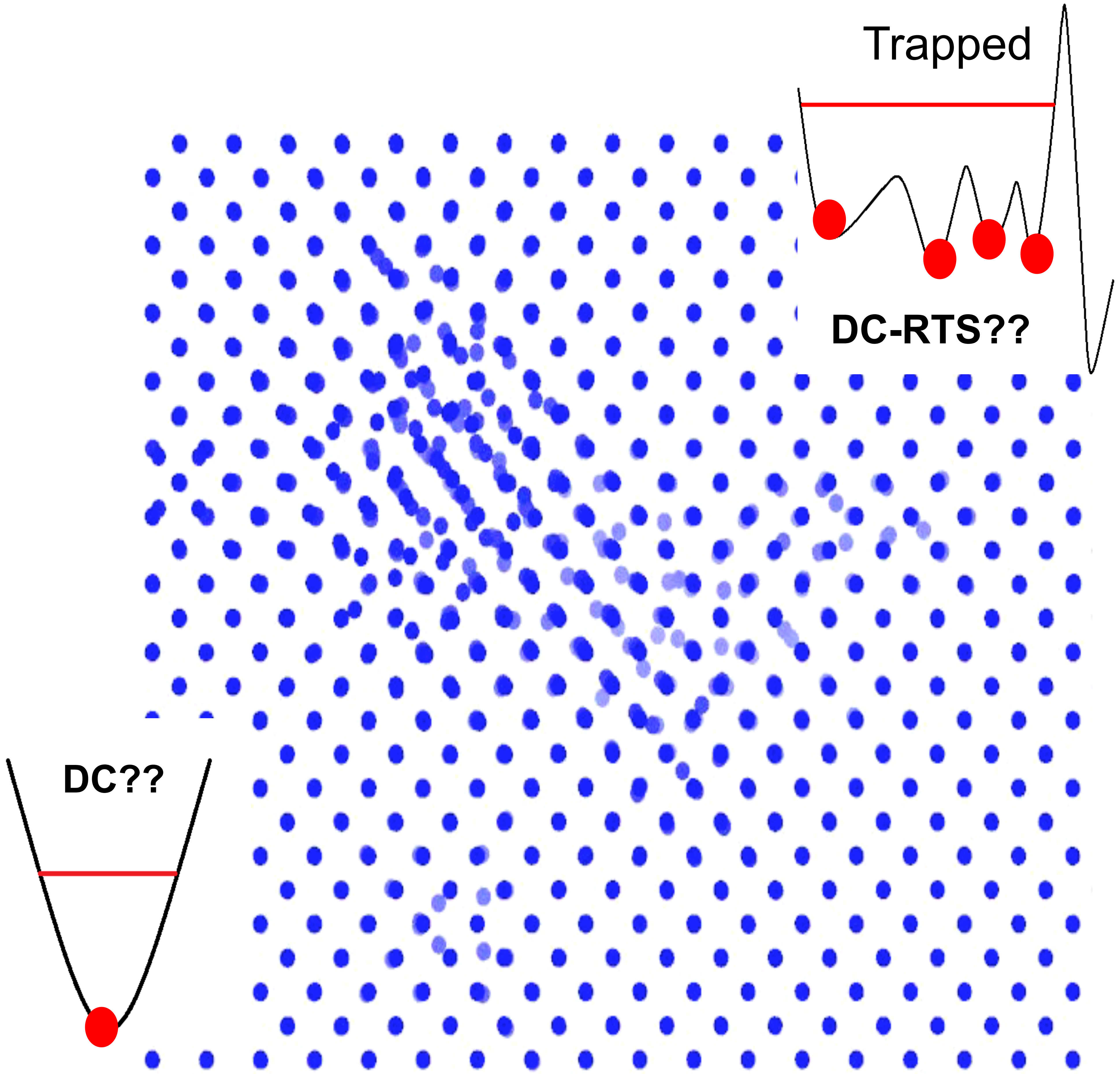
> Output of kART ➜ ab initio calculation

The defect structure of complex defect are really different than those expected ab initio words

➜ Gabriela Herrero-Saboya: Talk tomorrow



Long time simulation: Feedback



> Output of kART ➜ ab initio calculation

The defect structure of complex defect are really different than those expected ab initio words

➜ Gabriela Herrero-Saboya: Talk tomorrow

OffLattice kMC (kMC+ARTn+E/F Engine)

➜ Deal with complex system on long time

➜ Computation cost of relaxation/research event

How to keep/reuse the information already computed?

➜ Event configuration in continuous configuration space

Thank You for your attention