Kinetic Monte Carlo Simulation

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Predicting long-time dynamics
Rare Events

- Infrequent transitions from a local minimum to another local minimum

- Transition state theory to understand chemical reaction rates: Michael Polanyi & Henry Eyring in 1920’s & 1930’s

- Renewed interests in understanding self-organization (protein folding, life, etc.)
Energy Landscape

- Discrete abstraction: Partitioned configuration space

\[ \mathbb{R}^{3N} = \bigcup_{\alpha} \mathbb{R}_\alpha; \mathbb{R}_\alpha \cap \mathbb{R}_\beta = \emptyset \]

where a $3N$-dim. configuration $q \in \mathbb{R}_\alpha$ converges to the $\alpha$-th local minimum upon local minimization ($N$ is the number of atoms)
Phase Space Distribution

- Phase-space distribution \( f(q, p, t) \): probability to find the system at \( 3N \)-dim. position \( q = (q_1, ..., q_N) \) & \( 3N \)-dim. momentum \( p = (p_1, ..., p_N) \) at time \( t \)
- Probability to find the system in \( \mathcal{R}_\alpha \) at time \( t \) (\( h \) is the Planck constant)
  \[
  P_\alpha(t) = \frac{1}{h^{3N}} \int_{\mathcal{R}_\alpha} dq \int dp f(q, p, t)
  \]
Outline

• Master equation

\[
\frac{dP_\alpha}{dt} = -\sum_\beta W_{\beta\alpha} P_\alpha (t) + \sum_\beta W_{\alpha\beta} P_\beta (t)
\]

\( W_{\beta\alpha} \): Transition rate from state \( \alpha \) to state \( \beta \)

• How to compute \( W_{\alpha\beta} \)? Transition state theory

\[
W_{\beta\alpha} \equiv v_\alpha \exp \left( -\frac{V_s - V_\alpha}{k_B T} \right)
\]

Vibration frequency at the \( \alpha \)-th local minimum

Minimum energy path along a saddle point (transition state)

• How to simulate the master equation: Kinetic Monte Carlo simulation

Let \( \{r_1, r_2, \ldots\} \) be a set of possible escape events, \( r = \sum_i r_i \), and \( u_1 \) & \( u_2 \) are uniform random numbers in \([0,1]\):

1. Pick the next event \( i \) as

\[
i = \min_j \left\{ \sum_{k=1}^j \frac{r_k}{r} > u_1 \right\}
\]

2. Advance the time by

\[
t = -\ln(u_2)/r
\]
Dynamics of Phase Space Distribution

- Phase-space distribution $f(q, p, t)$: probability to find the system at $3N$-dim. position $q = (q_1, ..., q_N)$ & $3N$-dim. momentum $p = (p_1, ..., p_N)$ at time $t$

- Probability to find the system in $\mathcal{R}_\alpha$ at time $t$ ($h$ is the Planck constant)

$$P_\alpha(t) = \frac{1}{h^{3N}} \int_{\mathcal{R}_\alpha} dq \int dp f(q, p, t)$$

- Time derivative ($L = $ Liouville operator; $H(q, p) = $ Hamiltonian)

$$\frac{dP_\alpha}{dt} = \frac{1}{h^{3N}} \int_{\mathcal{R}_\alpha} dq \int dp \frac{\partial}{\partial t} f(q, p, t)$$

$$= \frac{1}{h^{3N}} \int_{\mathcal{R}_\alpha} dq \int dp [-Lf(q, p, t)]$$

$$= \frac{1}{h^{3N}} \int_{\mathcal{R}_\alpha} dq \int dp \left[ -\left( \frac{\partial H}{\partial p} \cdot \frac{\partial}{\partial q} - \frac{\partial H}{\partial q} \cdot \frac{\partial}{\partial p} \right) f(q, p, t) \right]$$

See supplementary note 1: “Liouville equation”
Population Dynamics

- Let

\[ H(q, p) = \sum_{i=1}^{3N} \frac{p_i^2}{2m_i} + V(q) \]

then

\[ \frac{dP_\alpha}{dt} = -\frac{1}{h^{3N}} \int_{\partial R_\alpha} dq \int d\mathbf{p} \sum_{i=1}^{3N} \frac{p_i}{m_i} \frac{\partial}{\partial q_i} f(q, p, t) + \frac{1}{h^{3N}} \int_{\partial R_\alpha} dq \int d\mathbf{p} \sum_{i=1}^{3N} \frac{\partial V}{\partial q_i} \frac{\partial}{\partial p_i} f(q, p, t) \]

\[ = -\frac{1}{h^{3N}} \int_{\partial R_\alpha} dq \int d\mathbf{p} \sum_{i=1}^{3N} \frac{\partial}{\partial q_i} \left( \frac{p_i}{m_i} f(q, p, t) \right) \]

Gauss’ theorem

~ telescoping

where \( dS \) is the surface element pointing outward normal to the surface \( \partial R_\alpha \) that outlines \( R_\alpha \)

- \( dP_\alpha /dt \) is negative of the outward flux through \( \partial R_\alpha \)
Population Flux

- Partition the surface $\partial R_\alpha$ into
  $$\partial R_\alpha = \sum_\beta S_{\beta \alpha}$$

where $S_{\beta \alpha}$ is the surface splitting $R_\alpha \& R_\beta$ (normal pointing from $\alpha$ to $\beta$)

\[
\frac{dP_\alpha}{dt} = -\sum_\beta \frac{1}{h^{3N}} \int \sum_{i=1}^{3N} dS_i \int dP \sum_{i=1}^{3N} \frac{p_i}{m_i} \Theta \left( \sum_{i} dS_i \frac{p_i}{m_i} \right) f(q,p,t) \text{ outgoing}
\]

\[
+ \sum_\beta \frac{1}{h^{3N}} \int \sum_{i=1}^{3N} dS_i \int dP \sum_{i=1}^{3N} \frac{p_i}{m_i} \Theta \left( \sum_{i} dS_i \frac{p_i}{m_i} \right) f(q,p,t) \text{ incoming}
\]

where $\Theta(x) = 1 (x \geq 0) \& 0 (x < 0)$ is the step function
Local Equilibration Approximation

• Assume that within each $\mathcal{R}_\alpha$, the phase space distribution is locally in thermal equilibrium, weighted to reproduce the current population (time scale of inter-state population transfer >> intra-state thermal equilibration time)

$$f(q,p,t) \simeq \frac{P_\alpha(t)}{P_\alpha(eq)} f_{eq}(q,p)$$

where

$$f_{eq}(q,p) = \frac{1}{\mathcal{Q}} \exp\left(-\frac{H(q,p)}{k_B T}\right)$$

and the partition function is split into

$$Q = \int \int \frac{dq dp}{h^{3N}} \exp\left(-\frac{H(q,p)}{k_B T}\right) = \sum_\alpha \int \int \frac{dq dp}{h^{3N}} \exp\left(-\frac{H(q,p)}{k_B T}\right) = \sum_\alpha Q_\alpha$$

$$\therefore P_\alpha(eq) = \frac{Q_\alpha}{Q} = \frac{1}{Q} \int \int \frac{dq dp}{h^{3N}} \exp\left(-\frac{H(q,p)}{k_B T}\right)$$
Master Equation

- Substituting the local-equilibration approximation into the population-flux equation, we obtain

\[
\frac{dP_{\alpha}}{dt} = -\sum_{\beta} \frac{1}{h^{3N}} \int_{S_{\beta \alpha}} \sum_{i=1}^{3N} dS_i \int d\mathbf{p} \sum_{i=1}^{3N} p_i \left( \sum_i dS_i \frac{p_i}{m_i} \right) f_{eq}(\mathbf{q}, \mathbf{p}) \frac{P_{\alpha}(t)}{P_{\alpha}(eq)} \\
+ \sum_{\beta} \frac{1}{h^{3N}} \int_{S_{\alpha \beta}} \sum_{i=1}^{3N} dS_i \int d\mathbf{p} \sum_{i=1}^{3N} p_i \left( \sum_i dS_i \frac{p_i}{m_i} \right) f_{eq}(\mathbf{q}, \mathbf{p}) \frac{P_{\beta}(t)}{P_{\beta}(eq)}
\]

\[
\therefore \frac{dP_{\alpha}}{dt} = -\sum_{\beta} W_{\beta \alpha} P_{\alpha}(t) + \sum_{\beta} W_{\alpha \beta} P_{\beta}(t)
\]

\[
W_{\alpha \beta} = \frac{1}{h^{3N}} \int_{S_{\alpha \beta}} \sum_{i=1}^{3N} dS_i \int d\mathbf{p} \sum_{i=1}^{3N} p_i \left( \sum_i dS_i \frac{p_i}{m_i} \right) f_{eq}(\mathbf{q}, \mathbf{p}) \frac{1}{P_{\beta}(eq)}
\]

\[
= \frac{1}{h^{3N}} \int_{S_{\alpha \beta}} \sum_{i=1}^{3N} dS_i \int d\mathbf{p} \sum_{i=1}^{3N} p_i \left( \sum_i dS_i \frac{p_i}{m_i} \right) e^{-H(\mathbf{q}, \mathbf{p})/k_B T} \left/ \frac{1}{h^{3N}} \int d\mathbf{q} d\mathbf{p} e^{-H(\mathbf{q}, \mathbf{p})/k_B T} \right.
\]

See supplementary note 2: “Master equation”

A. P. J. Jansen, *Introduction to KMC Simulations of Surface Reactions* (Springer, ’12)
**Transition State Theory**

- Reaction coordinate $q_1$ along a minimum-energy path separates the phase space into 2 regions—$A$ ($q_1 < 0$) & $B$ ($q_1 > 0$); all the other coordinates & momenta are collectively denoted as $X = (q, p) = (q_2, ..., q_{3N}, p_2, ..., p_{3N})$

- Probability to find the system being in $B$

  $$P_B(t) = \iiint \frac{dq_1 dp_1 dX}{h^{3N}} \Theta(q_1) f(q_1, p_1, X, t)$$

- Time derivative ($L = $ Liouville operator)

  $$\frac{dP_B}{dt} = \iiint \frac{dq_1 dp_1 dX}{h^{3N}} \Theta(q_1) \frac{\partial}{\partial t} f(q_1, p_1, X, t)$$

  $$= \iiint \frac{dq_1 dp_1 dX}{h^{3N}} \Theta(q_1) (-Lf(q_1, p_1, X, t))$$

  $$= \iiint \frac{dq_1 dp_1 dX}{h^{3N}} (L\Theta(q_1)) f(q_1, p_1, X, t)$$

  $$= \iiint \frac{dq_1 dp_1 dX}{h^{3N}} \left( \frac{p_1}{m_1} \delta(q_1) \right) f(q_1, p_1, X, t)$$

  $$= \iint \frac{dp_1 dX}{h^{3N}} \frac{p_1}{m_1} f(0, p_1, X, t)$$
Local Equilibration Approximation

- Split the integral into the gain (A → B) & loss (B → A) terms

\[
\frac{dP_B}{dt} = \left( \frac{dP_B}{dt} \right)_{A \rightarrow B} + \left( \frac{dP_B}{dt} \right)_{B \rightarrow A}
\]

\[
= \int_0^\infty \frac{dp_1}{h} \int \frac{dX}{h^{3N-1}} \frac{p_1}{m_1} f(0, p_1, X, t) + \int_{-\infty}^0 \frac{dp_1}{h} \int \frac{dX}{h^{3N-1}} \frac{p_1}{m_1} f(0, p_1, X, t)
\]

- Regions A & B locally (i.e., within the region) maintain the equilibrium distribution weighted to reproduce the current population

\[
f_{\alpha, \text{local}} = \frac{P_{\alpha}(t)}{P_{\alpha}(\text{eq})} f_{\text{eq}} \quad (\alpha = A, B)
\]

\[
f_{\text{eq}} = \frac{1}{Q} \exp\left(-\frac{H}{k_B T}\right)
\]

\[
Q = \iiint \frac{dq_1 dp_1 dX}{h^{3N}} \exp\left(-\frac{H}{k_B T}\right) = Q_A + Q_B
\]

\[
Q_A = \iiint_{q_1 < 0} \frac{dq_1 dp_1 dX}{h^{3N}} \exp\left(-\frac{H}{k_B T}\right) \quad Q_B = \iiint_{q_1 > 0} \frac{dq_1 dp_1 dX}{h^{3N}} \exp\left(-\frac{H}{k_B T}\right)
\]

\[
P_{\alpha}(\text{eq}) = \frac{Q_{\alpha}}{Q} \quad (\alpha = A, B)
\]
Transition State Theory

- Substituting the local equilibration approximation to the flux equation

\[
\left( \frac{dP_B}{dt} \right)_{A \to B} = \int_0^\infty dp_1 \int \frac{dX}{h^{3N-1}} \frac{p_1}{m_1} f_{eq}(0, p_1, X) \frac{P_A(t)}{P_A(\text{eq})}
\]

\[
\left( \frac{dP_B}{dt} \right)_{B \to A} = -\int_0^\infty dp_1 \int \frac{dX}{h^{3N-1}} \frac{p_1}{m_1} f_{eq}(0, p_1, X) \frac{P_B(t)}{P_B(\text{eq})}
\]

\[
\therefore \frac{dP_A(t)}{dt} = k_{BA} P_A(t) - k_{AB} P_B(t)
\]

\[
k_{BA} = \int_0^\infty dp_1 \int \frac{dX}{h^{3N-1}} \frac{p_1}{m_1} f_{eq}(0, p_1, X) \frac{1}{P_A(\text{eq})}
\]

\[
k_{AB} = \int_0^\infty dp_1 \int \frac{dX}{h^{3N-1}} \frac{p_1}{m_1} f_{eq}(0, p_1, X) \frac{1}{P_B(\text{eq})}
\]

- Analytical integration over \( p_1 \)

\[
k_{BA} = \int_0^\infty dp_1 \frac{p_1}{m_1} \exp(-p_1^2 / 2m_1k_B T) \int \frac{dX}{h^{3N-1}} \frac{\exp(-H / k_B T)_{q_1=p_1=0}}{Q} \]

\[
Q^* = \int \frac{dX}{h^{3N-1}} \exp(-H / k_B T)_{q_1=p_1=0}
\]

\[
Q^* = \int \frac{dX}{h^{3N-1}} \exp(-H / k_B T)_{q_1=p_1=0}
\]

\[
\frac{Q^*}{Q_A} = \frac{k_B T}{h} \frac{Q^*}{Q_A}
\]
Harmonic Transition State Theory

- In region A, we assume
  \[ V(q_1, \ldots, q_{3N}) = V_A + \frac{1}{2} \sum_j m_j \left( \omega_j^A \right)^2 (q_j - b_j)^2 \]
  \[ \therefore Q_A = \left( \frac{2\pi k_B T}{\hbar} \right)^{3N} \exp \left( -\frac{V_A}{k_B T} \right) \prod_{j=1}^{3N} \omega_j^A \]

- At the dividing surface, we assume
  \[ V(q_1, \ldots, q_{3N}) = V_S - \frac{1}{2} a_{11} q_1^2 + \frac{1}{2} \sum_{j=2}^{3N} m_j \left( \omega_j^* \right)^2 q_j^2 \]
  \[ Q^* = \int \frac{dX}{\hbar^{3N-1}} \exp \left( -\frac{H}{k_B T} \right) _{q_1=p_1=0} = \left( \frac{2\pi k_B T}{\hbar} \right)^{3N-1} \exp \left( -\frac{V_S}{k_B T} \right) \prod_{j=2}^{3N} \omega_j^* \]

\[ \therefore k_{BA} = \frac{1}{2\pi} \exp \left( -\frac{V_S - V_A}{k_B T} \right) \frac{\prod_{j=1}^{3N} \omega_j^A}{\prod_{j=2}^{3N} \omega_j^*} \exp \left( -\frac{V_S - V_A}{k_B T} \right) \]

See supplementary note 3: “Transition state theory”
Digression: Save the World?

• Solar land-area requirement (with 10% energy conversion efficiency) to supply the global energy [Nathan Lewis, *Caltech*]

• Need better catalyst for splitting water [Lewis & Nocera, *PNAS* 103, 15729 ('06)]

\[ 2H_2O \xrightarrow{hv} 2H_2 + O_2 \]

**COMPUTATIONAL METHODS**

A search engine for catalysts

Trial and error has been the traditional method of finding the best catalyst for a reaction. A computational approach can reduce the lab work required.

*Nature Mater.* 5, 847 ('06)
A Job for Superatom

Molecular Dynamics Simulations of Rapid Hydrogen Production from Water Using Aluminum Clusters as Catalyzers

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\[
k_{H_2} = \frac{k_B T}{h} \exp\left(-\frac{\Delta}{k_B T_{room}}\right) = 10^{11} \text{s}^{-1}
\]
H₂ Production from Water Using LiAl Particles

16,661-atom QMD simulation of Li₄₄₁Al₄₄₁ in water on 786,432 IBM Blue Gene/Q cores

• Scalable to industrially relevant particle sizes

K. Shimamura et al., Nano Lett. 14, 4090 (’14)
Poisson Process

- **Poisson process** = sequence of events, in which the probability of an event to occur in time \([t,t+\delta]\) is \(r\delta\) (\(r\) is the rate) independent of history as \(\delta \to 0\).

- Probability \(P(n,t)\) that \(n\) events occur in time interval \(t=N\delta\):

\[
P(n,t) = C(N,n)(r\delta)^n (1 - r\delta)^{N-n} = \frac{N!}{n!(N-n)!} (r\delta)^n (1 - r\delta)^{N-n}
\]

\[
\xrightarrow{\frac{rt}{N} \to \infty} \frac{(rt)^n}{n!} e^{-rt}
\]

\[
\xrightarrow{N \to \infty} 1
\]

- **Sum rule:**

\[
\sum_{n=0}^{\infty} P(n,t) = \sum_{n=0}^{\infty} \frac{(rt)^n}{n!} e^{-rt} = 1
\]
Kinetic Monte Carlo Simulation

- **Probability density** $P(t)$ of time $t$ between successive events
  
  $P(t)dt = \text{probability(\text{n} - \text{e} - \text{v} - \text{in} - [0, t] \wedge 1 - \text{n} - \text{e} - \text{v} - \text{in} - [t, t+dt])}$

  $= P(0, t) \times rdt = e^{-rt} \times rdt$

  $\therefore P(t) = re^{-rt}$

- **Random time-interval generation**: Let $u$ be a uniform random number in $[0,1]$ & generate $t = -\ln(u)/r \in [0,\infty]$

  $\therefore P(t) = P(u) \left| \frac{du}{dt} \right| = 1 \times re^{-rt} = re^{-rt}$

- **Kinetic MC algorithm**: Let $\{r_1, r_2, \ldots\}$ be a set of possible events, $r = \Sigma_i r_i$, and $u_1 \& u_2$ are uniform random numbers in $[0,1]$:

  1. **Pick the next event** $i$ as $i = \min_j \left\{ \frac{j \sum_k r_k}{r} > u_1 \right\}$

  2. **Advance the time by** $t = -\ln(u_2)/r$


  See supplementary note 4: “Kinetic Monte Carlo simulation”
Divide-\&-Conquer KMC Algorithm

- Domain decomposition: Concurrent events among multiple domains, $d$
  \[ \Delta t = -\ln(rnd) / \sum_{d} r_d = O(N^{-1}) \implies -\ln(rnd) / \max_{d} (r_d) = O(1) \]

- Colored domain blocks: Avoids conflicting events by allowing concurrent events only with domains of the same color, which are well-separated

- Neighbor-domain caching for spatial decomposition via message-passing

- Dual linked-list cell method: (1) small cells for constructing neighbor lists for nearest-neighbor hopping events; (2) large cells for domain-block coloring

E. Martinez et al., J. Comp. Phys. 230, 1359 (’11)
Scalable Parallel KMC

- Benchmark tests on electron transfer in heme aggregates
- Better weak-scaling for coarser granularity ($N$ hemes on $P$ processors)

- Weak-scaling parallel efficiency 0.935 for a 4.2 billion-heme system on 1,024 Intel Xeon processors
Temporal Locality in Long-Time Dynamics

- **Temporal locality**: Rare transitions between local minimum-energy states
- **Transition state theory**: Reformulate *sequential* long-time dynamics as *parallel* search for low activation-barrier transition events
- **Discrete graph abstraction**: Linear combinations of atomistic events (LCAE)
  

- **Directionally heated nudged elastic band (NEB) method**: Search for thermally activated events without the knowledge of final states

\[
\mathbf{M} \dot{\mathbf{R}}_s = \mathbf{F}_s - \mathbf{M} \gamma_s \dot{\mathbf{R}}_s \quad (s = 0, \ldots, S - 1)
\]

\[
\mathbf{F}_s = \begin{cases}
\frac{\partial V}{\partial \mathbf{R}_s} + \mathbf{F}_s^{spr} \bigg|_\parallel & (1 \leq s \leq S - 2) \\
\frac{\partial V}{\partial \mathbf{R}_s} & (s = 0, S - 1)
\end{cases}
\]
Space-Time-Ensemble Parallel (STEP) NEB

- Path ensemble method (PEM): Long-time simulation in the framework of kinetic Monte Carlo—molecular kinetics simulation

\[ r_b = \left( t_{\text{therm}} + t_{\text{heat}} \exp \left( \frac{\Delta_b}{k_B} \left( \frac{1}{T} - \frac{1}{T_{\text{heat}}} \right) \right) \right)^{-1} \]

\[ P_b = \frac{r_b}{r} = \frac{r_b}{\sum_{b=0}^{B-1} r_b} \]

- Space-time-ensemble parallelism (STEP) = spatial decomposition within each state (\(D\) domains)
  - + temporal parallelism across the states within each band (\(S\) states)
  - + band ensemble (\(B\) bands)

- Hierarchical concurrency
  \[ P = BSD \]

Divide-&-Conquer Protein Folding

- Levinthal paradox (1968): How the Nature folds an amino-acid sequence into a global energy minimum 3D structure (which is known to be NP complete) within microseconds (~ billion molecular-dynamics steps).

- Sequential KMC not good enough.

Zip-&-assembly algorithm (Ken Dill at UCSF)
1. (Divide) Chop the amino-acid sequence into ~10 residue fragments.
2. (Conquer) For each fragment, perform replica-exchange (~ temperature accelerated) molecular dynamics simulation & detect the formation of any stable hydrophobic contacts.
3. (Combine) Grow the stable fragments by adding surrounding residues while freezing (~ constraint) the found stable contacts.

S.B. Ozkan et al., *PNAS* **104**, 11987 ('07)
Parsing Protein-Folding Routes

Computational linguistics

(1) Formal grammar to describe protein-folding routes
(2) Dynamic programming for an efficient algorithm for the folding routes

K. A. Dill et al., *Polymer* 48, 4289 ('07)
W. Dyrka et al., *Alg. Mol. Biol.* 8, 31 ('13)
Singlet Fission in Amorphous DPT

- Photo-current doubling by splitting a singlet exciton into 2 triplet excitons
- Singlet fission (SF) in mass-produced disordered organic solid → efficient low-cost solar cells
- Exp’l breakthrough: SF found in amorphous diphenyl tetracene (DPT)
- Ultrafast transient absorption measurements identified *two time scales* (1 & 100 ps) for exciton population dynamics
- Hypothesis: Existence of *SF hot spots* [S. T. Roberts *et al.*, *JACS* 134, 6388 (*'12)]

**Problem:** *Molecular origin of SF hot spots?*
Divide-Conquer-Recombine KMC

- Move up from molecules to microstructures
- Challenge: Unprecedented $10^4$-atom NAQMD simulation
- Computational approach: Divide-conquer-recombine (DCR) NAQMD

DCR-NAQMD (phonon-assisted exciton dynamics) + time-dependent perturbation theory (singlet-fission rate) + kinetic Monte Carlo calculations of exciton population dynamics in 6,400-atom amorphous DPT

NAQMD-informed Kinetic Monte Carlo

- NAQMD-KMC exciton population dynamics reproduces the experimentally observed two time scales (~1 & 100 ps) in amorphous DPT

![Graphs showing NAQMD-KMC and Experiment results](image)

W. Mou et al., *APL* **102**, 173301 ('13)

S. T. Roberts *et al.*, *JACS* **134**, 6388 ('12)

Cover image: *Appl. Phys. Lett.* (Apr. 29, '13)