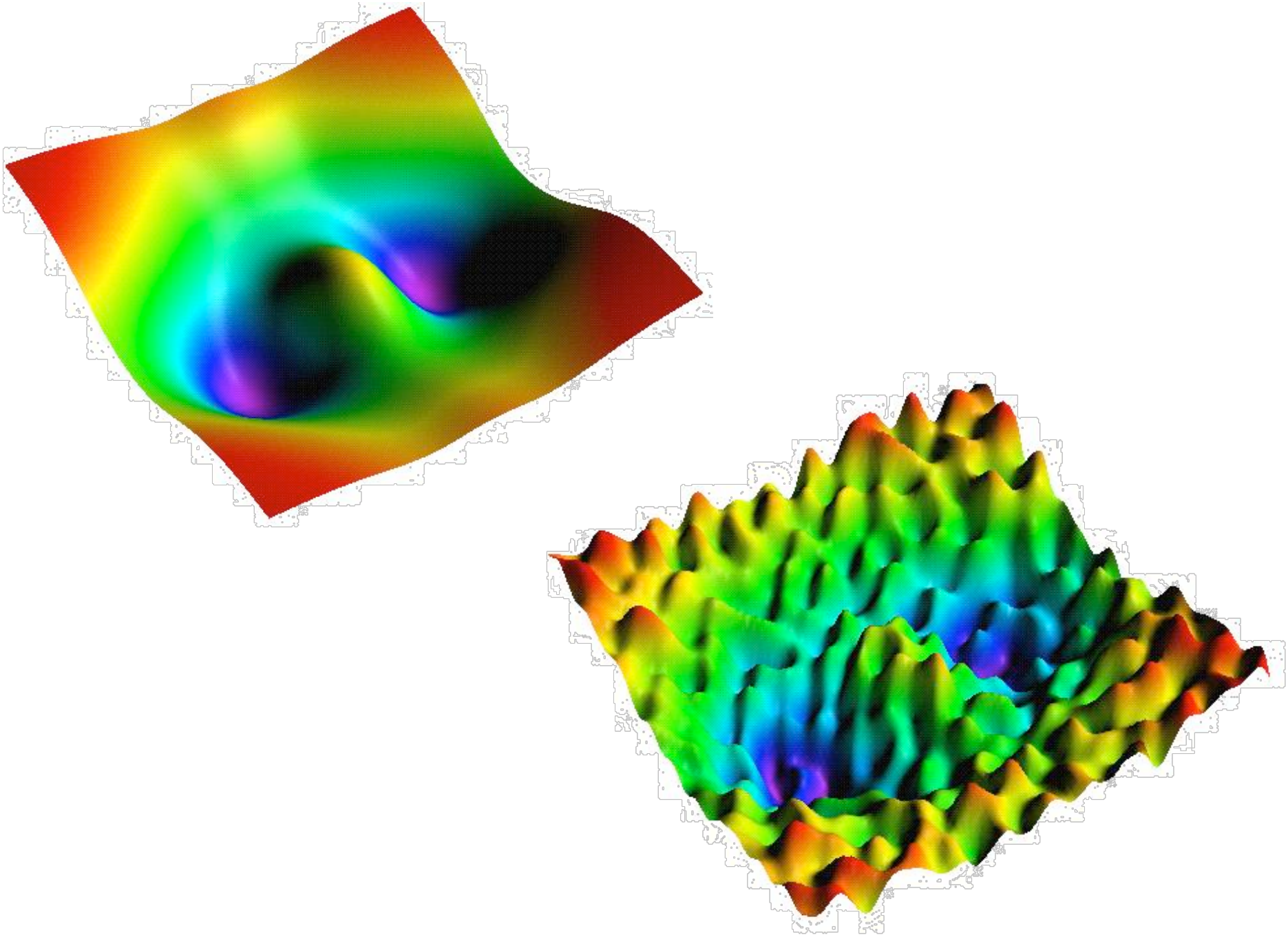
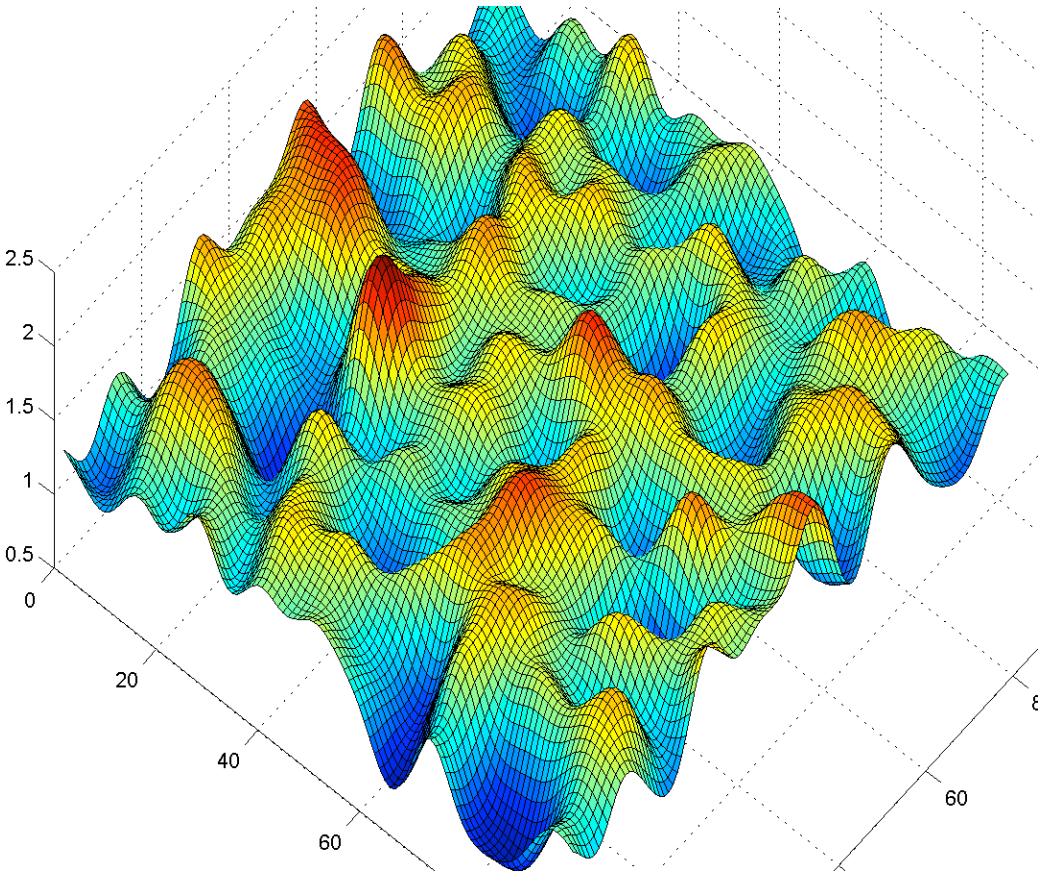
Lecture III

Path Sampling Methods

Outline

1. Introduction: Simple vs. complex
2. Transition path ensemble
3. Sampling the transition path ensemble: Monte Carlo simulation in trajectory space
4. Shooting and Shifting
5. Kinetics from the transition path ensemble
6. Transition interface sampling (& PPTIS)
7. Forward flux sampling
8. Applying TPS: freezing of LJ and GCM



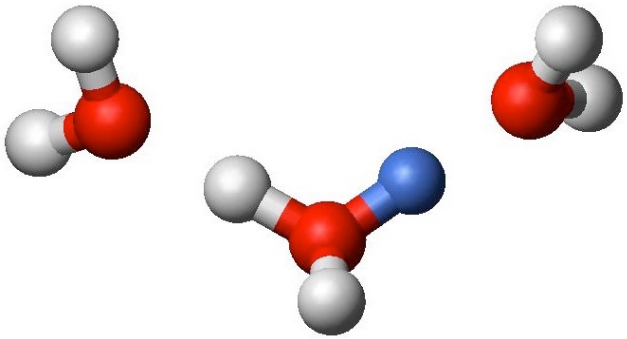


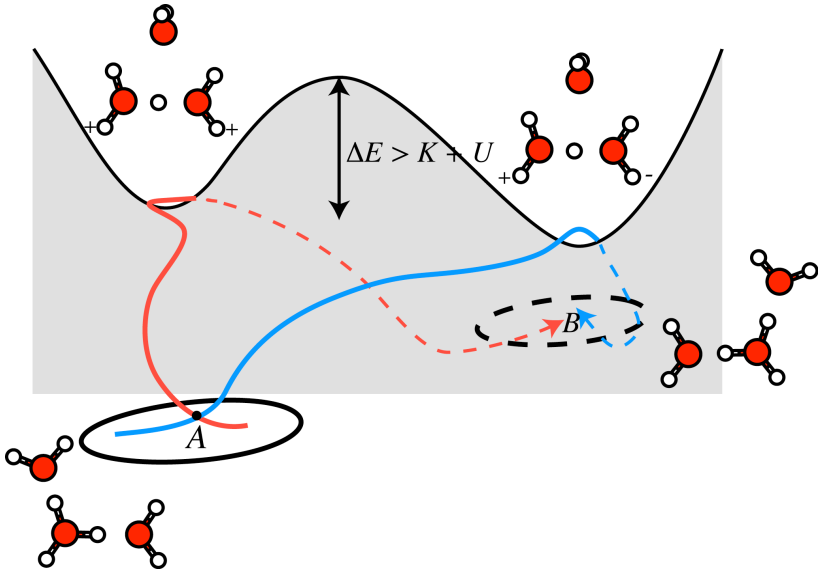
Gaussian Core Model

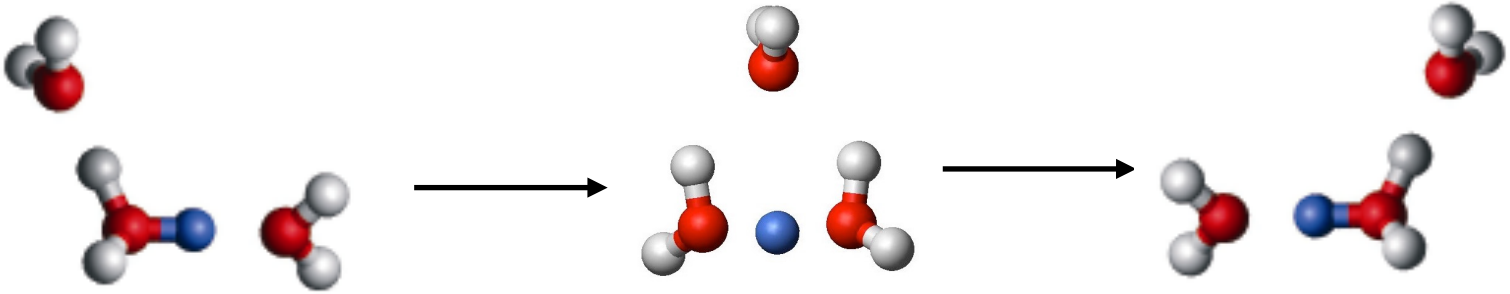
$$\phi(r) = \varepsilon e^{-(r/\sigma)^2}$$

Protonated water trimer

$(H_2O)_3H^+$

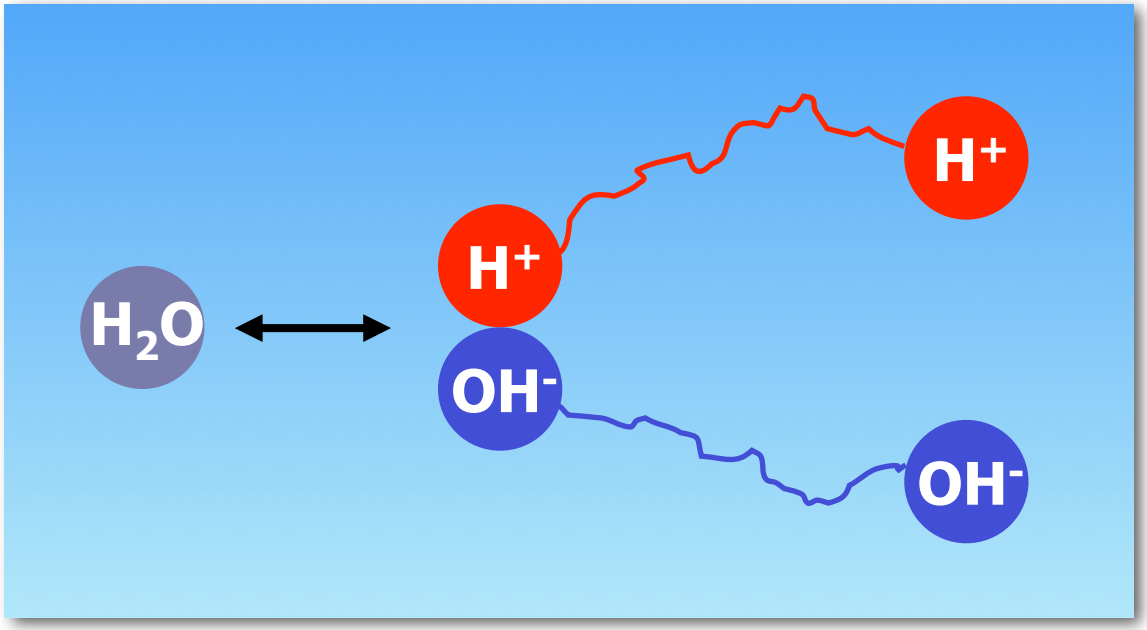




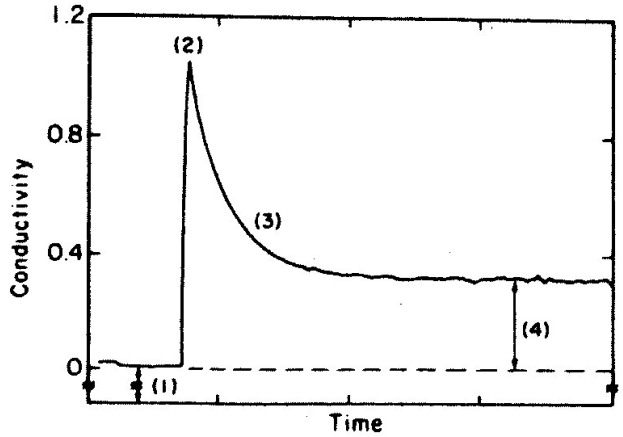


P. L. Geissler, T. v. Voorhis, C. Dellago, CPL 324, 149 (2000)

Autoionization in liquid water



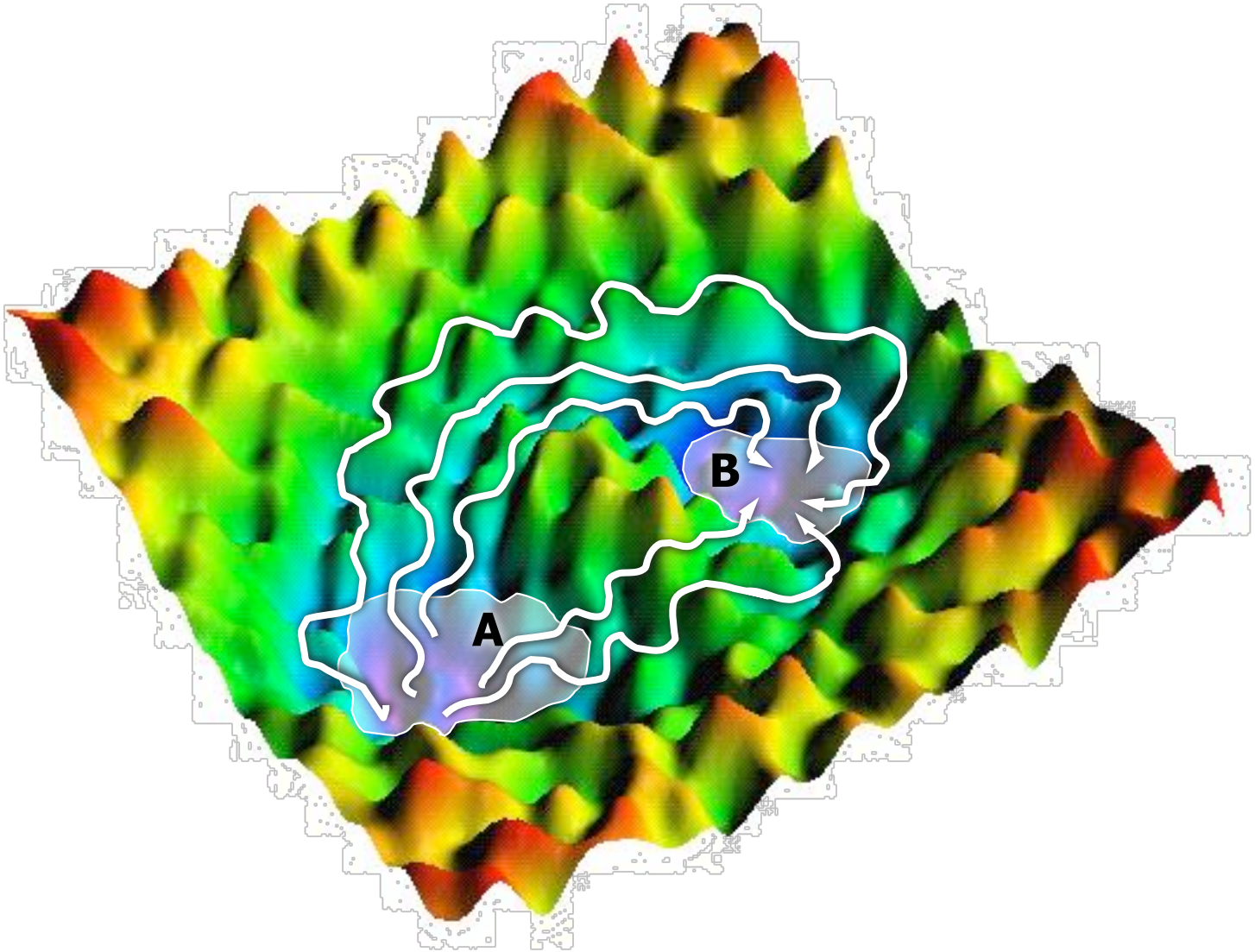
$$H_2O \rightleftharpoons OH^- + H^+$$



Average life time > 10 h

M. Eigen and L. De Maeyer, Z. Elektrochemie 59, 987 (1955)

Transition path sampling

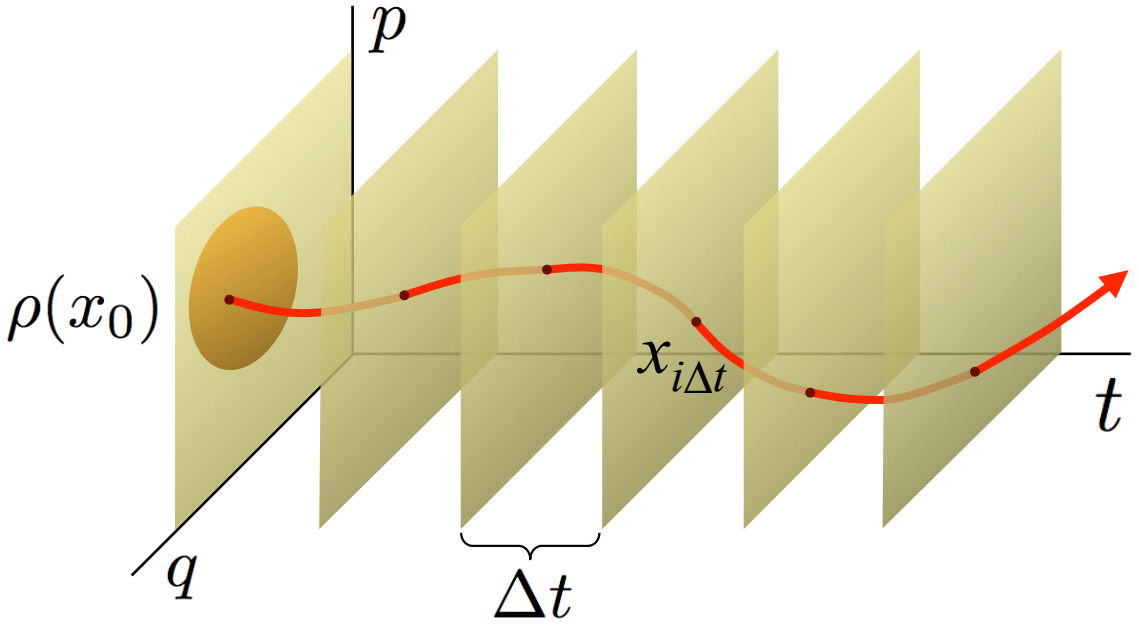


C. Dellago, P. G. Bolhuis, F. S. Csajka, D. Chandler, JCP 108, 1964 (1998)
C. Dellago, P. L. Geissler, P. G. Bolhuis, Adv. Chem. Phys. 123, 1 (2002)

Path probability density

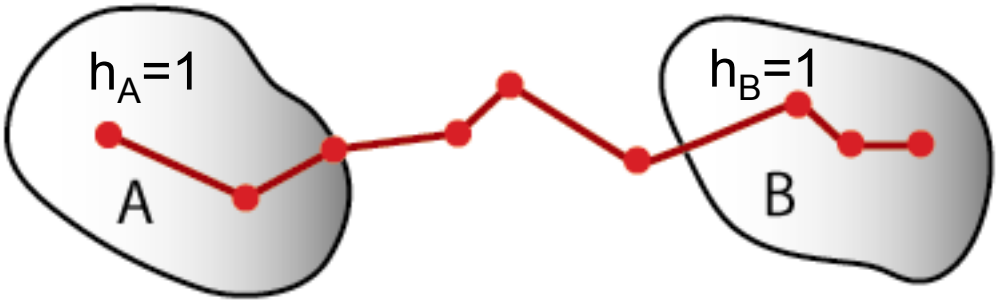
$$x(\mathcal{T}) \equiv \{x_0, x_{\Delta t}, x_{2\Delta t}, \ldots, x_{\mathcal{T}}\}$$

Path = Sequence of states



$$\mathcal{P}[x(\mathcal{T})] = \rho(x_0) \prod_{i=0}^{\mathcal{T}/\Delta t - 1} p(x_{i\Delta t} \to x_{(i+1)\Delta t})$$

Transition path ensemble



$$\mathcal{P}_{AB}[x(\mathcal{T})] \equiv h_A(x_0)\mathcal{P}[x(\mathcal{T})]h_B(x_\mathcal{T})/Z_{AB}(\mathcal{T})$$

$$Z_{AB}(\mathcal{T}) \equiv \int \mathcal{D}x(\mathcal{T})\, h_A(x_0)\mathcal{P}[x(\mathcal{T})]h_B(x_\mathcal{T})$$

$$\int \mathcal{D}x(\mathcal{T}) \equiv \int \cdots \int dx_0 dx_{\Delta t} dx_{2\Delta t} \cdots dx_\mathcal{T}$$

Transition proabilities

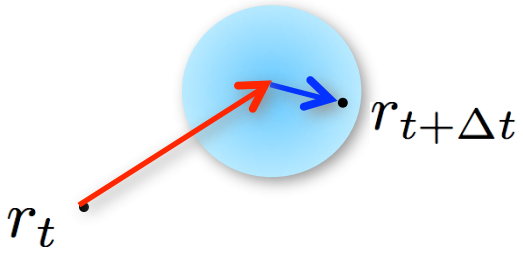
Initial conditions

Canonical: $\rho(x) = \exp\{-\beta\mathcal{H}(x)\}/Q$ $\quad Q(\beta) = \int dx \exp\{-\beta\mathcal{H}(x)\}$

Microcanonical: $\rho(x) = \delta[E - \mathcal{H}(x)]/g(E)$ $\quad g(E) = \int dx \delta[E - \mathcal{H}(x)]$

Brownian dynamics

$$m\gamma\dot{r} = -\frac{\partial V(r)}{\partial r} + \mathcal{F}$$



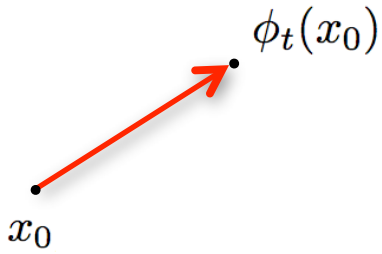
$$p(r_t \to r_{t+\Delta t}) = \frac{1}{\sqrt{2\pi\sigma^2}} \exp\left\{-\frac{(r_{t+\Delta t} - r_t + \frac{\Delta t}{\gamma m}\frac{\partial V}{\partial r})^2}{2\sigma^2}\right\}$$

$$\sigma^2 = \frac{2k_B T}{m\gamma}\Delta t$$

Newtonian dynamics

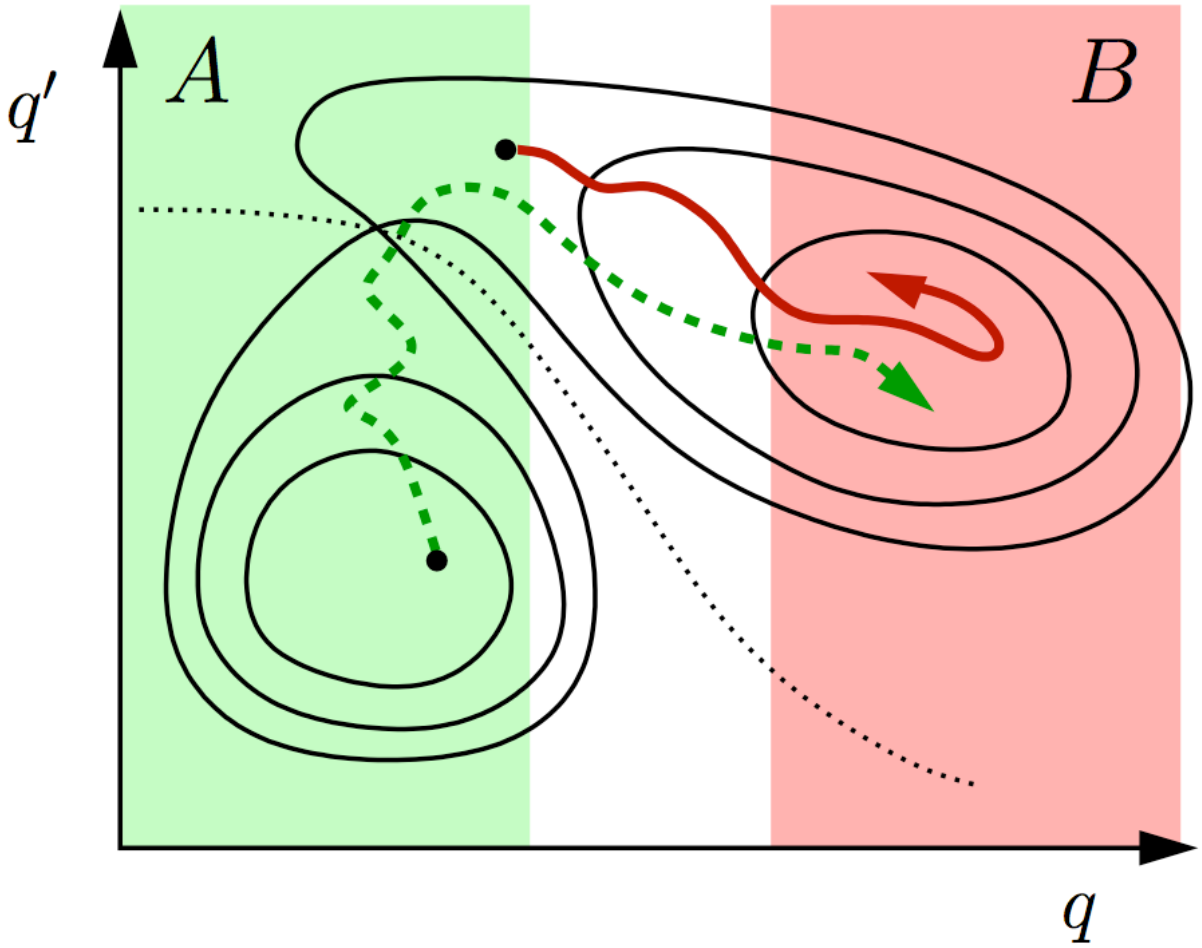
$$\dot{r} = \frac{\partial\mathcal{H}(r,p)}{\partial p}$$

$$\dot{p} = -\frac{\partial\mathcal{H}(r,p)}{\partial r}$$

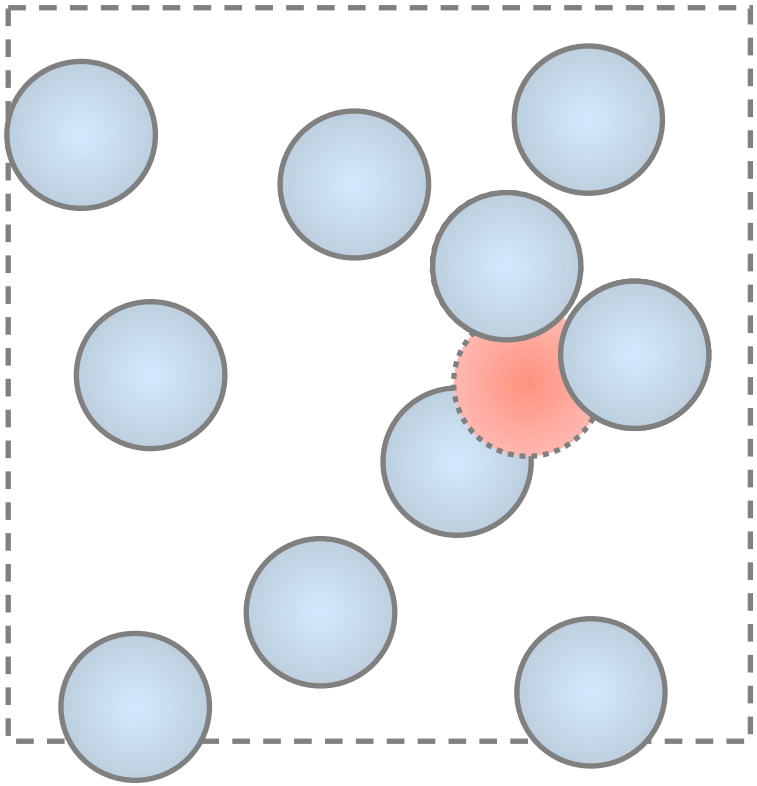


$$p(x_t \to x_{t+\Delta t}) = \delta[x_{t+\Delta t} - \phi_{\Delta t}(x_t)]$$

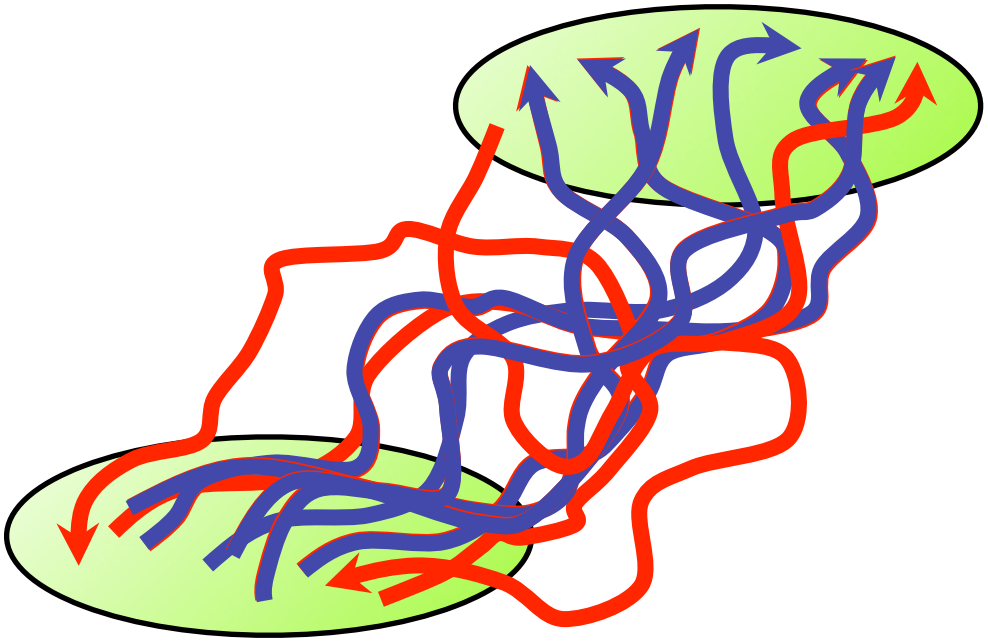
Defining the stable states



Sampling the path ensemble



Monte Carlo
simulation

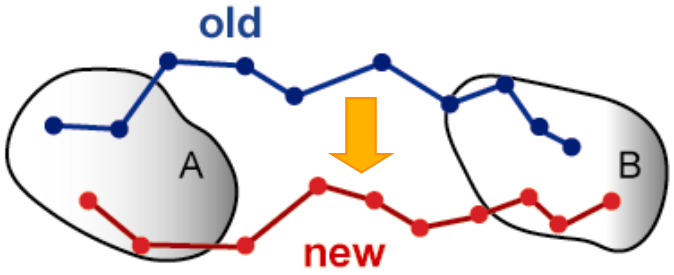


transition path
sampling

Metropolis MC of pathways

1. Generate **new** path from **old** one

$$x^{(o)}(\mathcal{T}) \longrightarrow x^{(n)}(\mathcal{T})$$



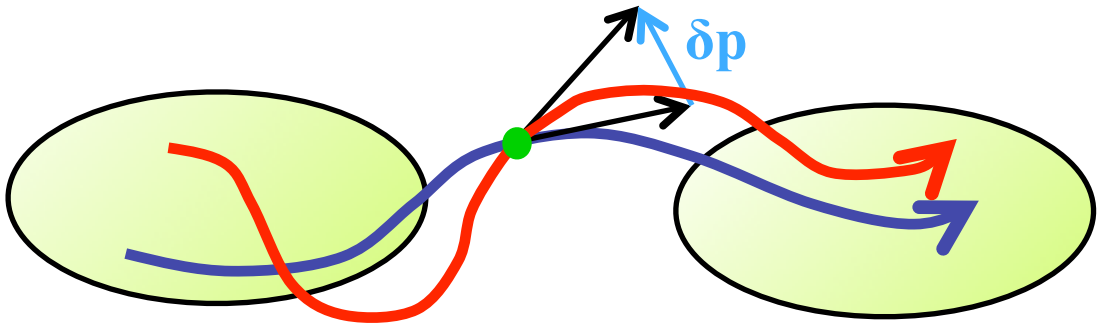
2. Accept **new** path according to **detailed balance**:

$$\mathcal{P}_{AB}[x^{(o)}(\mathcal{T})]\pi[x^{(o)}(\mathcal{T}) \to x^{(n)}(\mathcal{T})] = \mathcal{P}_{AB}[x^{(n)}(\mathcal{T})]\pi[x^{(n)}(\mathcal{T}) \to x^{(o)}(\mathcal{T})]$$

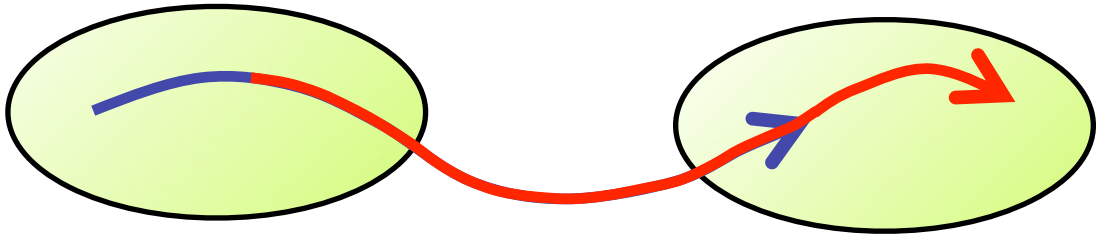
$$\pi[x^{(o)}(\mathcal{T}) \to x^{(n)}(\mathcal{T})] = P_{\text{gen}}[x^{(o)}(\mathcal{T}) \to x^{(n)}(\mathcal{T})] \times P_{\text{acc}}[x^{(o)}(\mathcal{T}) \to x^{(n)}(\mathcal{T})]$$

3. Satisfy detailed balance with the **Metropolis rule**:

$$P_{\text{acc}}[x^{(o)}(\mathcal{T}) \to x^{(n)}(\mathcal{T})] = h_A[x_0^{(n)}]h_B[x_{\mathcal{T}}^{(n)}] \min\left\{1, \frac{\mathcal{P}[x^{(n)}(\mathcal{T})]P_{\text{gen}}[x^{(n)}(\mathcal{T}) \to x^{(o)}(\mathcal{T})]}{\mathcal{P}[x^{(o)}(\mathcal{T})]P_{\text{gen}}[x^{(o)}(\mathcal{T}) \to x^{(n)}(\mathcal{T})]}\right\}$$



Shooting



Shifting

Shooting algorithm

$$P^{\mathrm{f}}_{\mathrm{gen}}[x^{\mathrm{o}}(\mathcal{T}) \to x^{\mathrm{n}}(\mathcal{T})] = \prod_{i=t'/\Delta t}^{\mathcal{T}/\Delta t-1} p\left(x^{(\mathrm{n})}_{i\Delta t} \to x^{(\mathrm{n})}_{(i+1)\Delta t}\right)$$

$$P^{\mathrm{b}}_{\mathrm{gen}}[x^{\mathrm{o}}(\mathcal{T}) \to x^{\mathrm{n}}(\mathcal{T})] = \prod_{i=1}^{t'/\Delta t} \bar{p}\left(x^{(\mathrm{n})}_{i\Delta t} \to x^{(\mathrm{n})}_{(i-1)\Delta t}\right)$$

$$P_{\mathrm{gen}}[x^{(\mathrm{o})}(\mathcal{T}) \to x^{(\mathrm{n})}(\mathcal{T})] = p_{\mathrm{gen}}[x^{(\mathrm{o})}_{t'} \to x^{(\mathrm{n})}_{t'}] \prod_{i=t'/\Delta t}^{\mathcal{T}/\Delta t-1} p\left(x^{(\mathrm{n})}_{i\Delta t} \to x^{(\mathrm{n})}_{(i+1)\Delta t}\right) \times \prod_{i=1}^{t'/\Delta t} \bar{p}\left(x^{(\mathrm{n})}_{i\Delta t} \to x^{(\mathrm{n})}_{(i-1)\Delta t}\right)$$

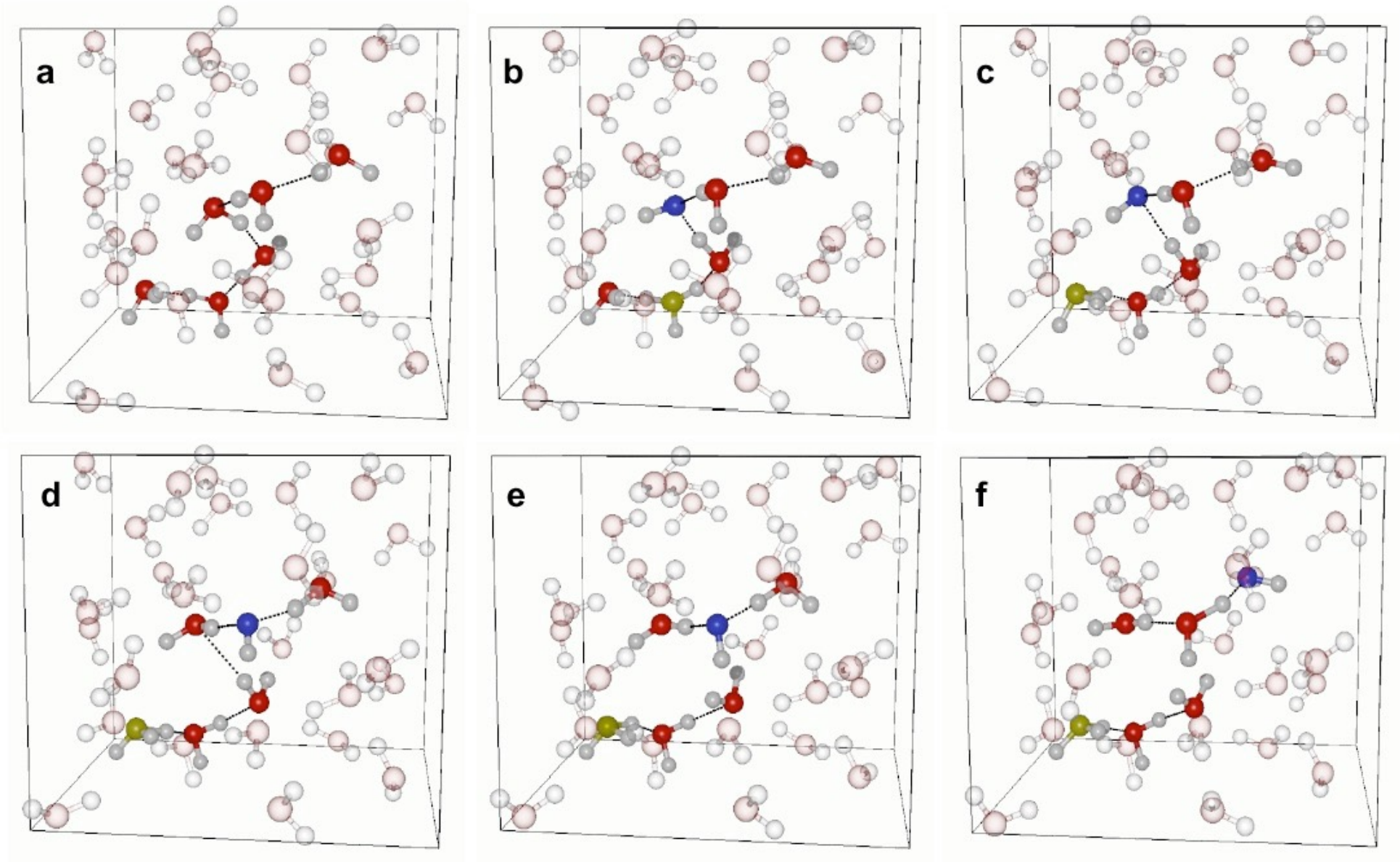
$$P_{\mathrm{acc}}[x^{(\mathrm{o})}(\mathcal{T}) \to x^{(\mathrm{n})}(\mathcal{T})] = h_A[x^{(\mathrm{n})}_0] h_B[x^{(\mathrm{n})}_{\mathcal{T}}] \min\left[1, \frac{\rho\left(x^{(\mathrm{n})}_0\right)}{\rho\left(x^{(\mathrm{o})}_0\right)} \prod_{i=0}^{t'/\Delta t-1} \frac{p\left(x^{(\mathrm{n})}_{i\Delta t} \to x^{(\mathrm{n})}_{(i+1)\Delta t}\right)}{\bar{p}\left(x^{(\mathrm{n})}_{(i+1)\Delta t} \to x^{(\mathrm{n})}_{i\Delta t}\right)} \times \frac{\bar{p}\left(x^{(\mathrm{o})}_{(i+1)\Delta t} \to x^{(\mathrm{o})}_{i\Delta t}\right)}{p\left(x^{(\mathrm{o})}_{i\Delta t} \to x^{(\mathrm{o})}_{(i+1)\Delta t}\right)}\right]$$

Detailed balance: $\dfrac{p(x \to y)}{\bar{p}(y \to x)} = \dfrac{\rho_0(y)}{\rho_0(x)}$

$$P_{\mathrm{acc}}[x^{(\mathrm{o})}(\mathcal{T}) \to x^{(\mathrm{n})}(\mathcal{T})] = h_A[x^{(\mathrm{n})}_0] h_B[x^{(\mathrm{n})}_{\mathcal{T}}] \min\left[1, \frac{\rho(x^{(\mathrm{n})}_{t'})}{\rho(x^{(\mathrm{o})}_{t'})}\right]$$

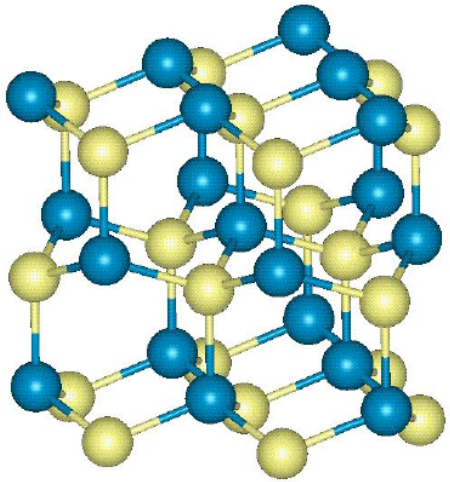
$$P_{\mathrm{acc}}[x^{(\mathrm{o})}(\mathcal{T}) \to x^{(\mathrm{n})}(\mathcal{T})] = h_A[x^{(\mathrm{n})}_0] h_B[x^{(\mathrm{n})}_{\mathcal{T}}]$$

Autoionization in liquid water



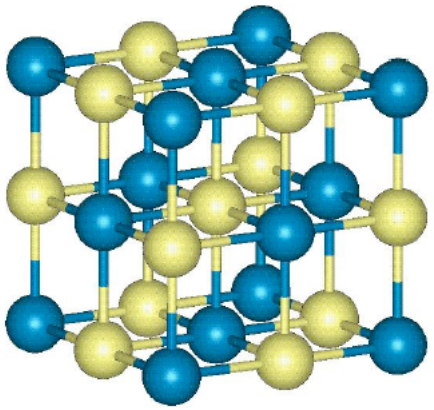
P. L. Geissler, C. Dellago, D. Chandler, J. Hutter, M. Parrinello, Science 291, 2121 (2001)

Structural transformation in CdSe

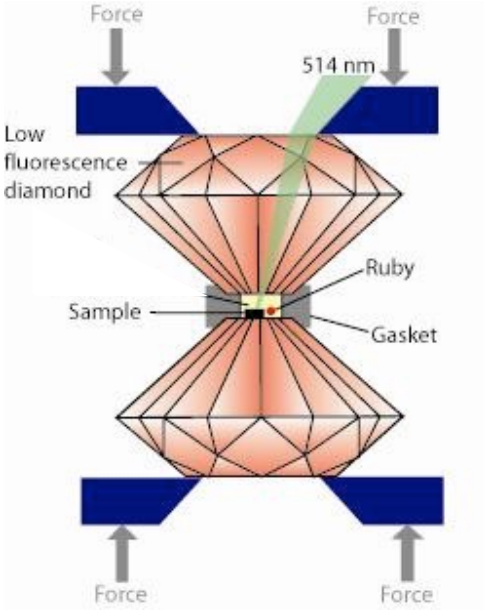


Wurtzite

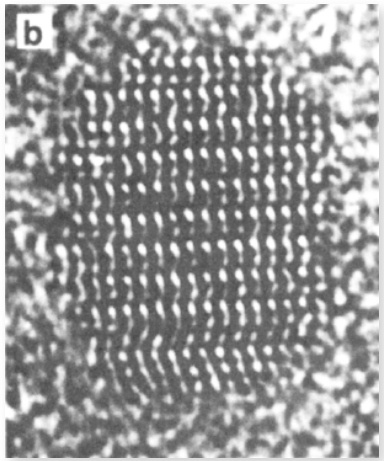
2.5 GPa

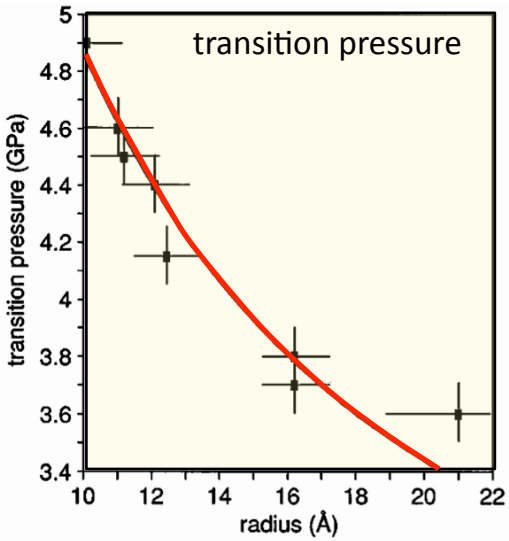


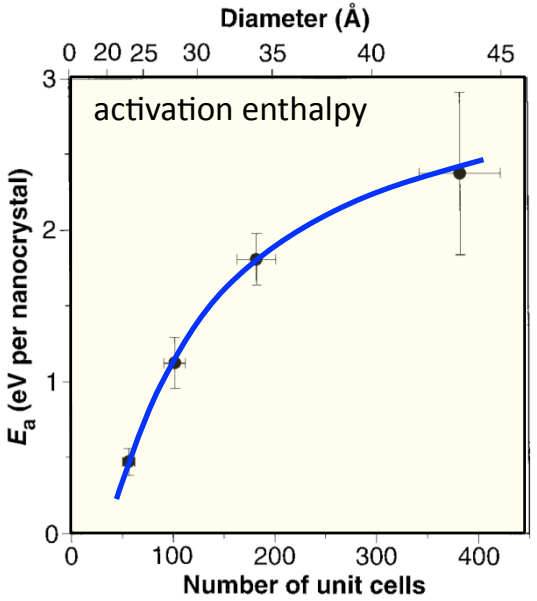
Rocksalt



10-30 Å

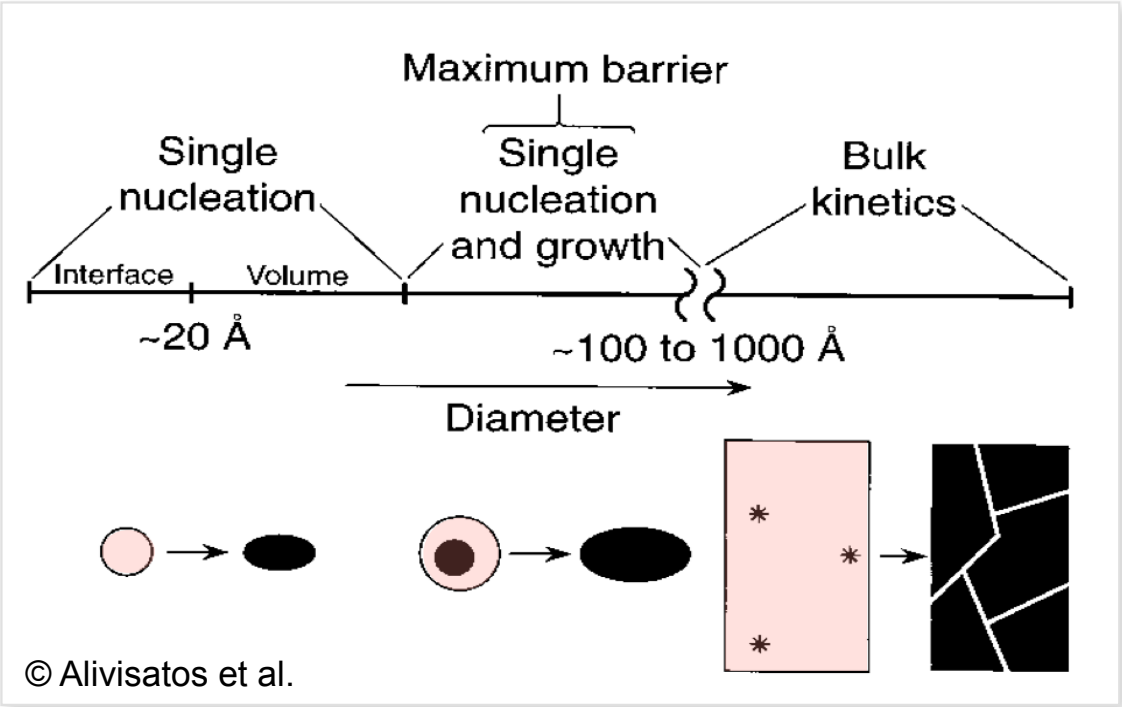


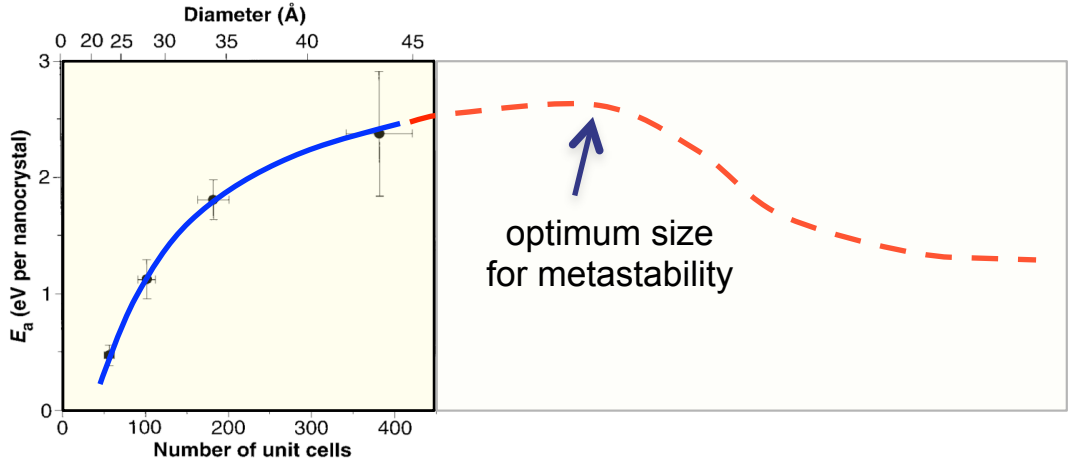




S. H. Tolbert and A. P. Alivisatos, JCP 102, 4642 (1995); Science 265, 373 (1994)

What is the mechanism?

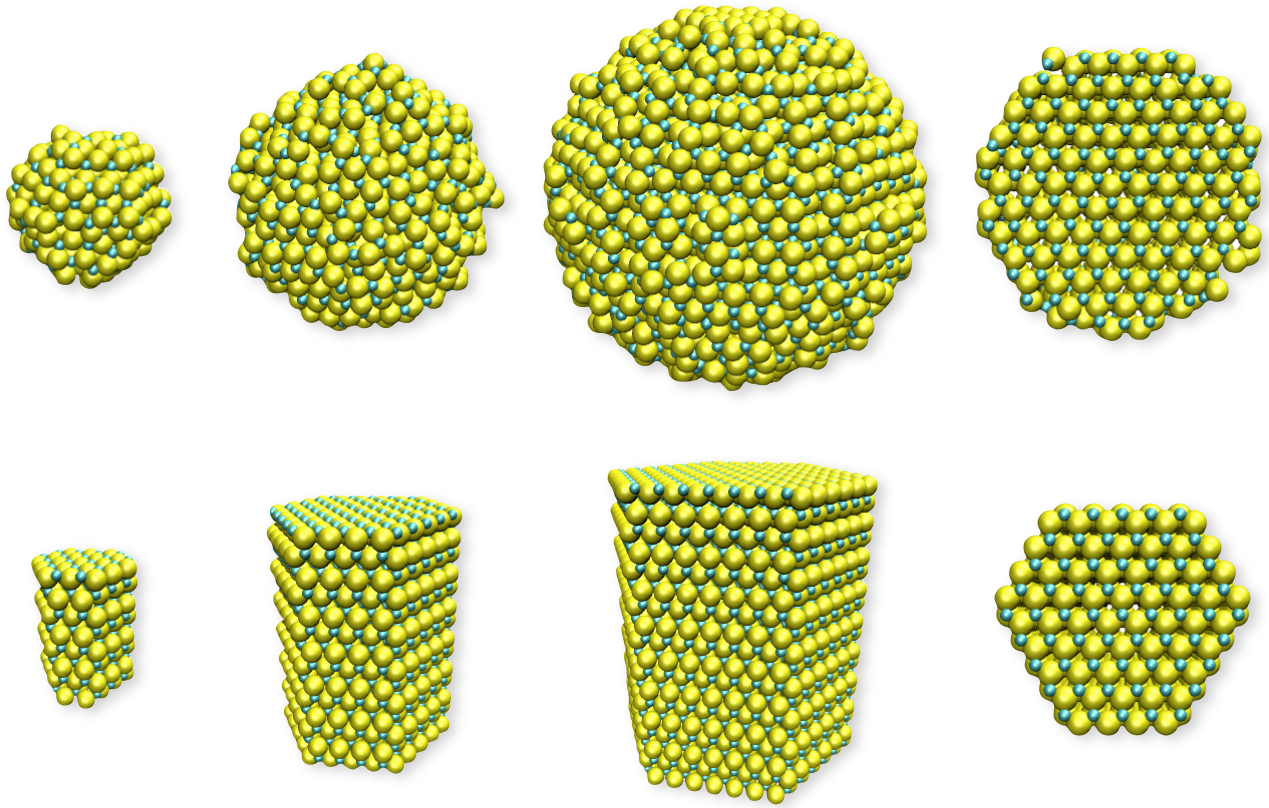


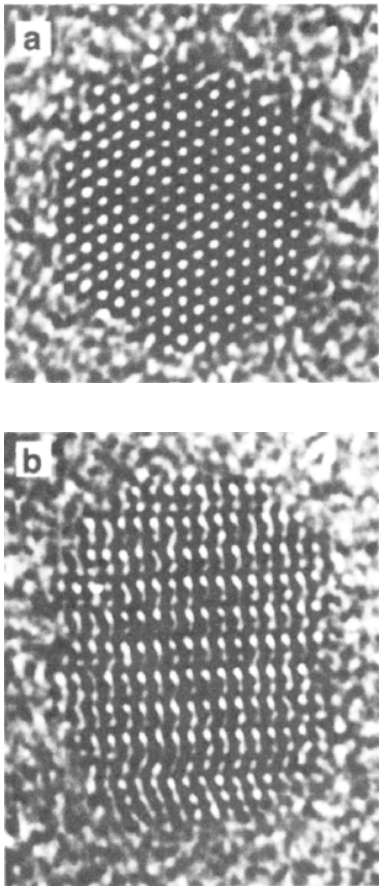


C. C. Chen, A. B. Herhold, C. S. Johnson, A. P. Alivisatos, Science 276, 398 (1997)

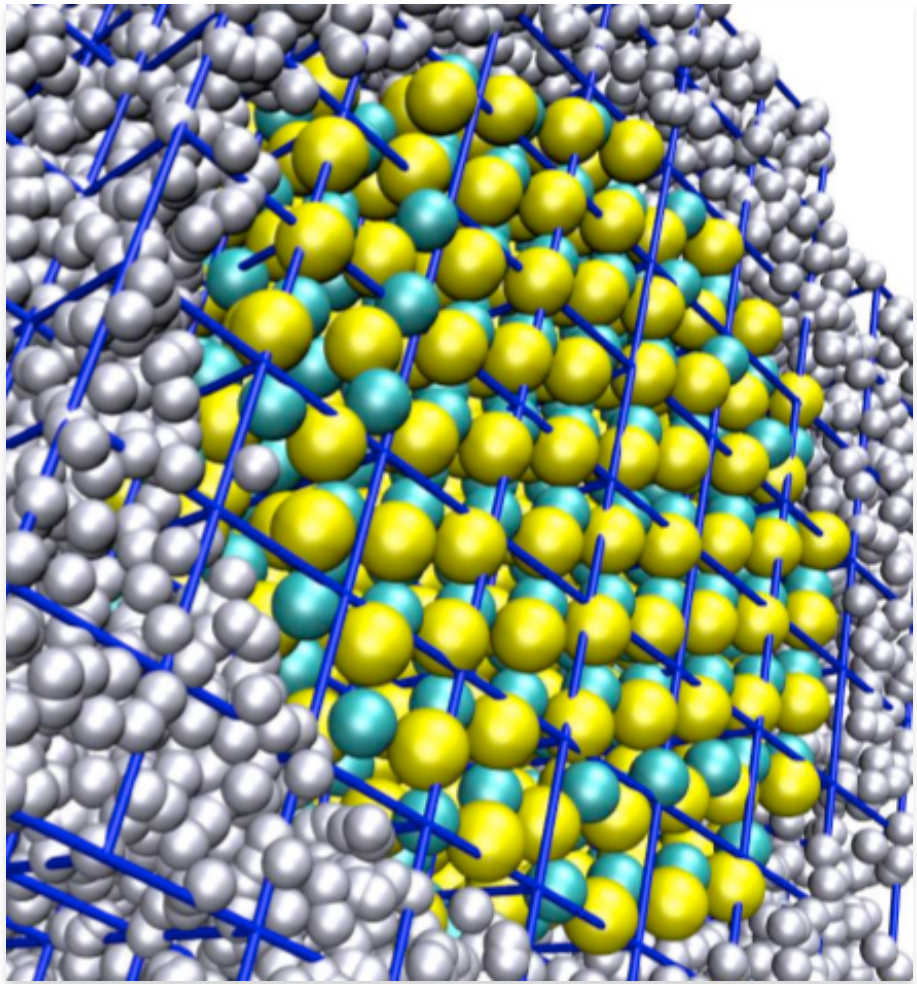
Molecular dynamics simulation

- Empirical pair potential [E. Rabani, JCP 116, 258 (2002)]
- Faceted and spherical crystals (N=120-5000, R=10-30A)
- T=300K, pressure increased in steps of 0.25 GPa / 10 ps





Ideal gas pressure bath

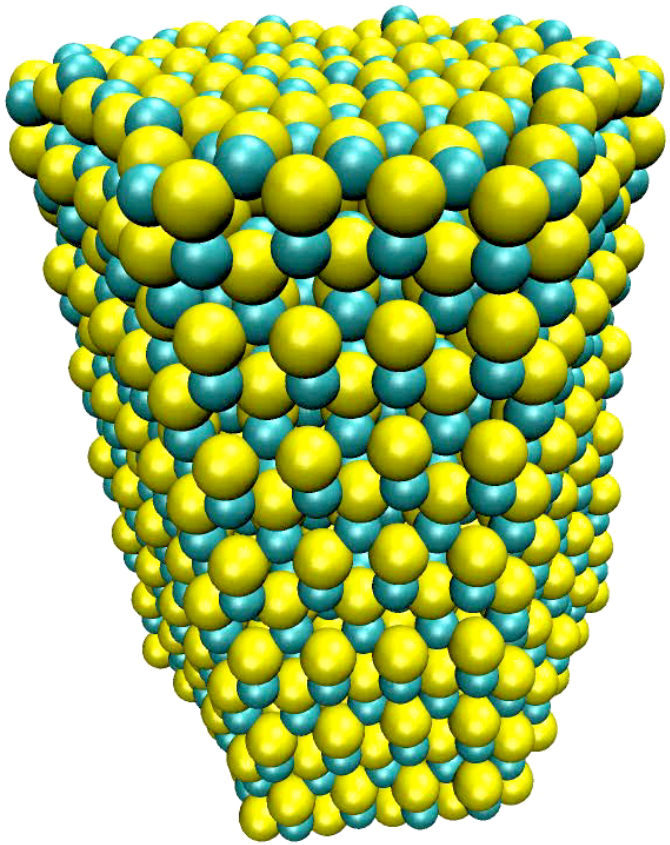


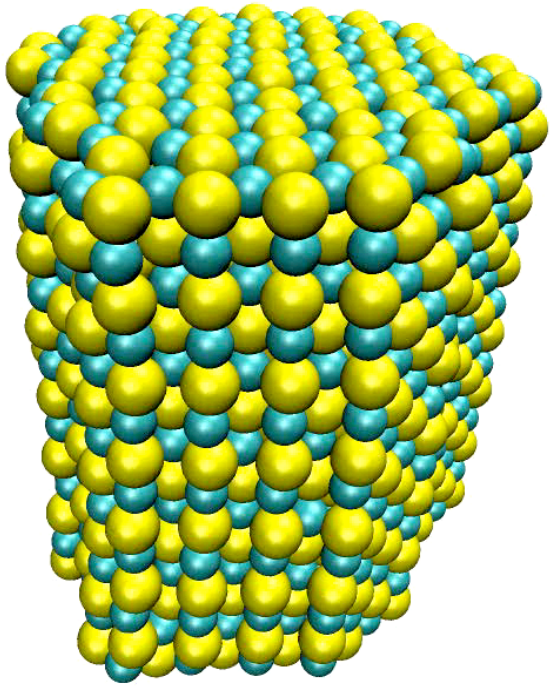
- Atmosphere of ideal gas
- Gas particles stream in through surface
- N_{id}=350.000 (at p=11GPa)
- Thermostat & barostat
- Pressure hydrostatic
- E, V, N fluctuate
- thermodynamically consistent volume

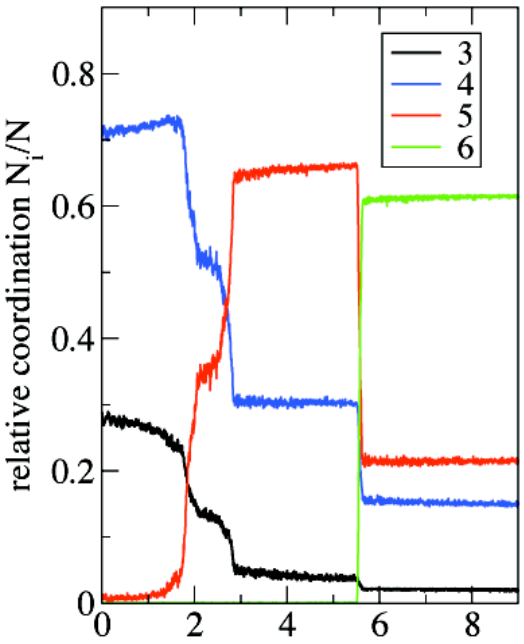
$$V_{\mathrm{crys}} = V - N_{\mathrm{gas}} k_{\mathrm{B}} T / p$$

M. Grünwald and C. Dellago, Mol. Phys. 104, 3709 (2006)
M. Grünwald, P. L. Geissler and C. Dellago, J. Chem. Phys. 127, 154718 (2007)

Transition mechanism - hysteresis



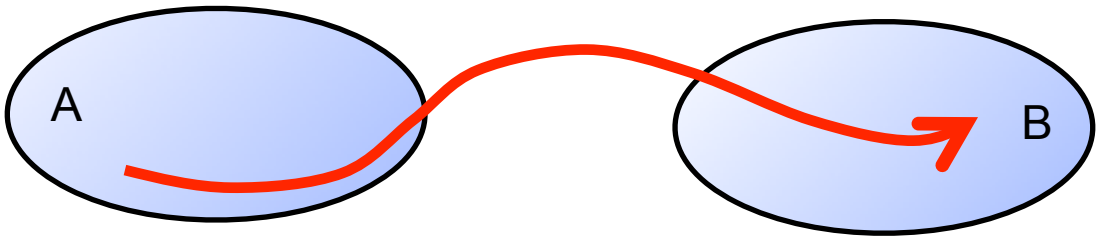


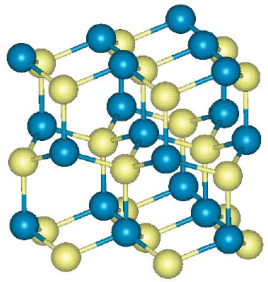


p = 6 GPa

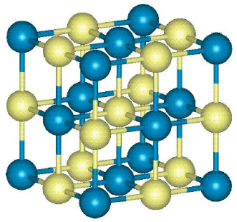
p = 0.5 GPa

TPS for the CdSe transition

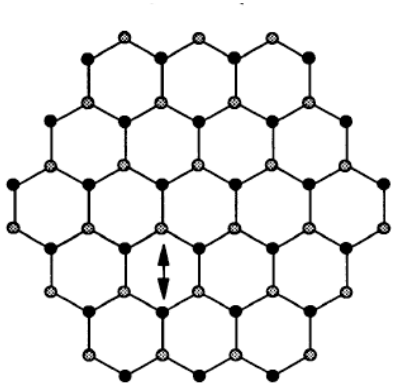




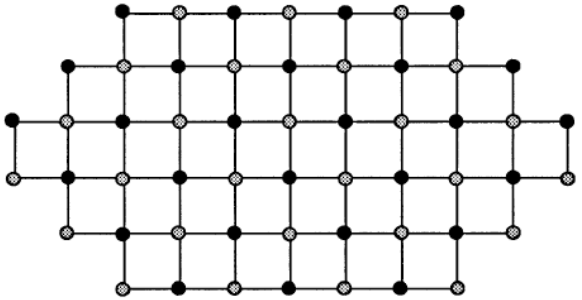
Wurtzite



Rocksalt



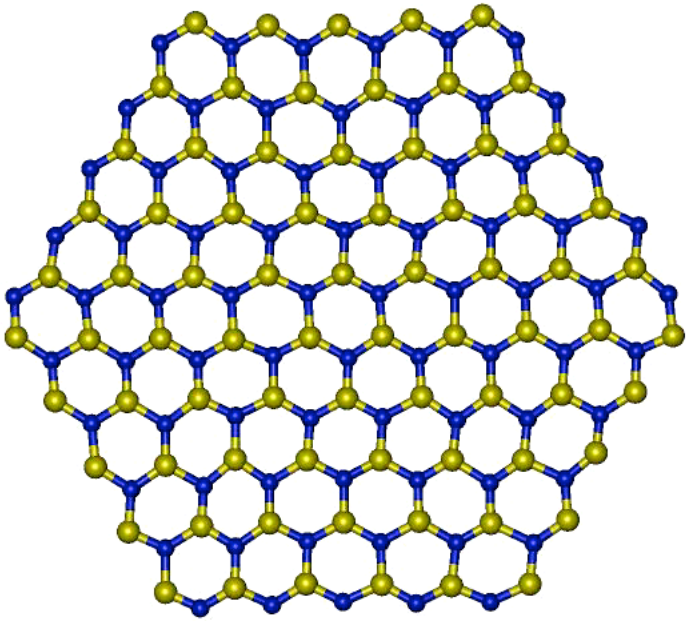
initial pathway



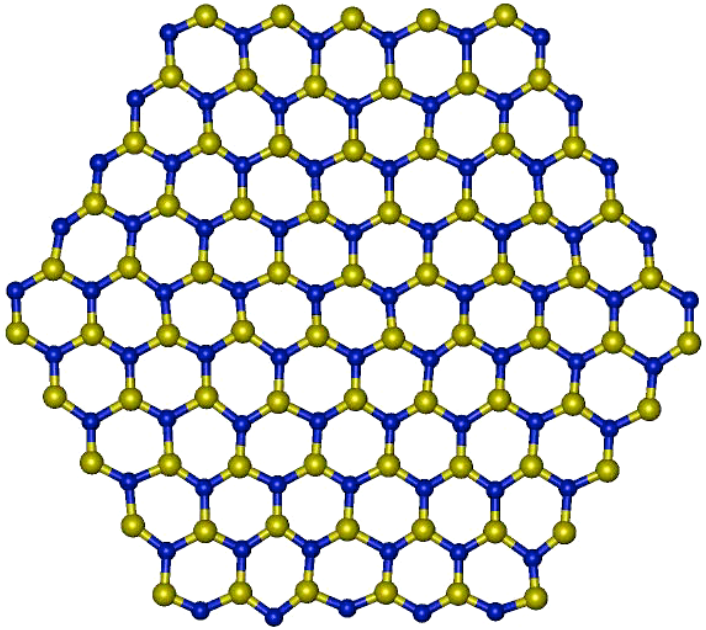
M. Grünwald, P. L. Geissler, and C. Dellago, JCP 127, 154718 (2007)
D. Zahn, Y Grin, and S. Leoni, PRB 72, 064110 (2005)
S. H. Tolbert and A. P. Alivisatos, JCP 102, 4642 (1995)

Transition Mechanism - TPS

p = 3GPa



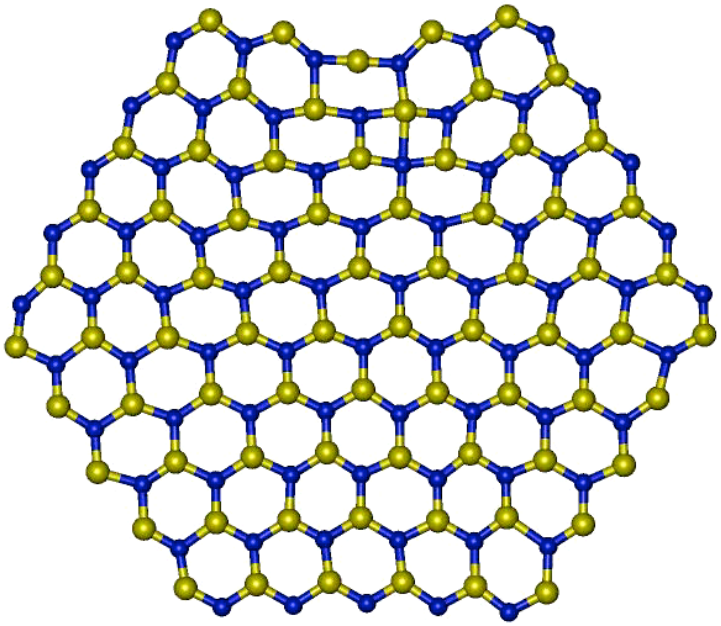
Trajectory 1



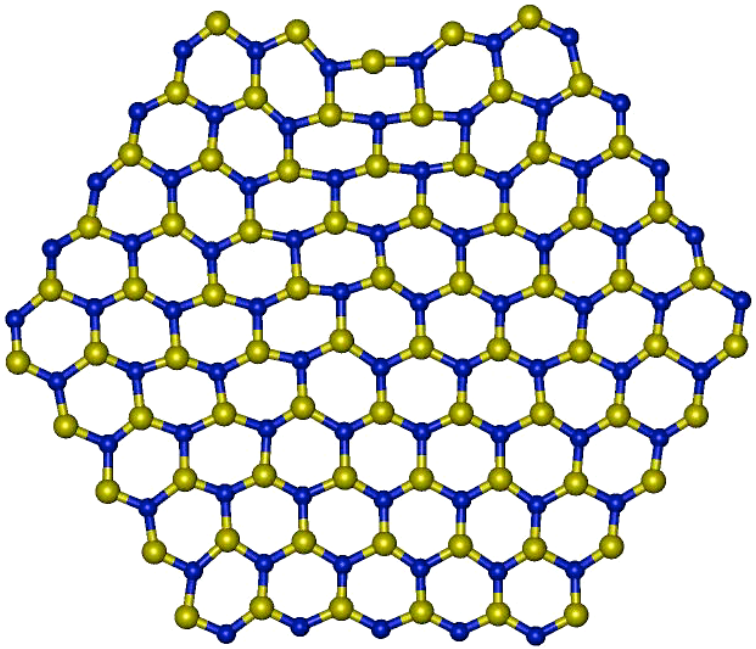
Trajectory 101

Transition Mechanism - TPS

p = 3GPa

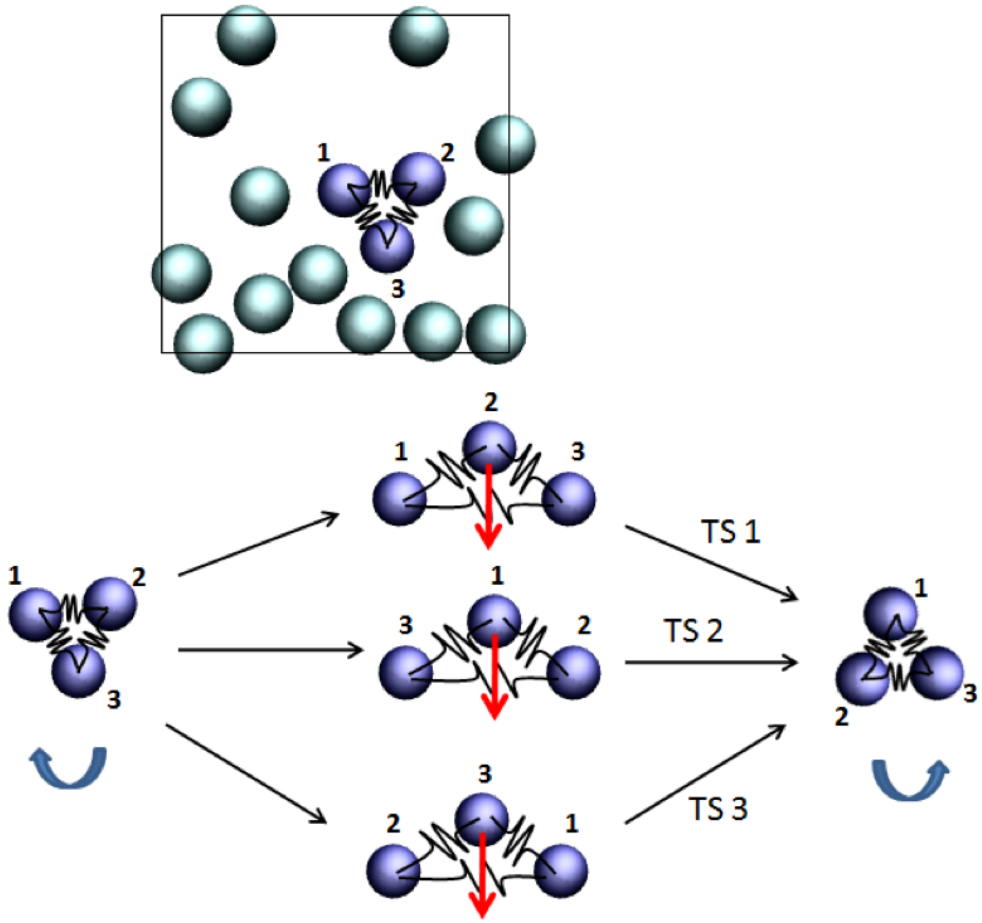


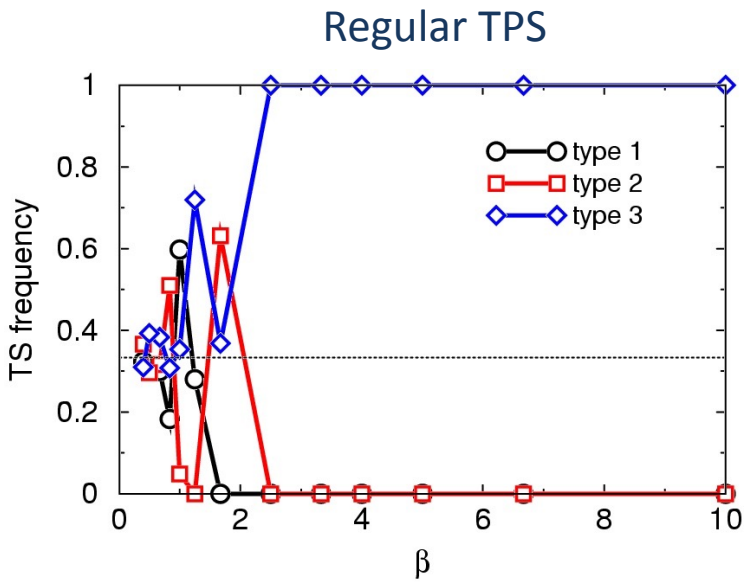
Trajectory 217



Trajectory 423

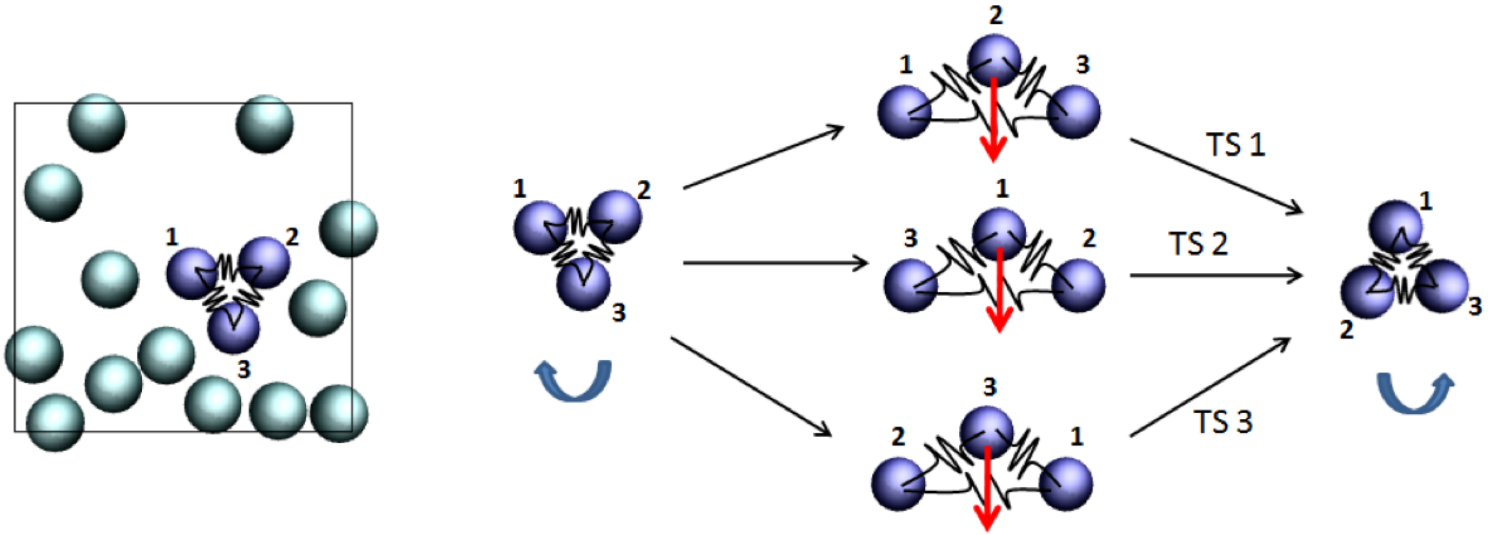
Ergodic Sampling of Trajectory Space

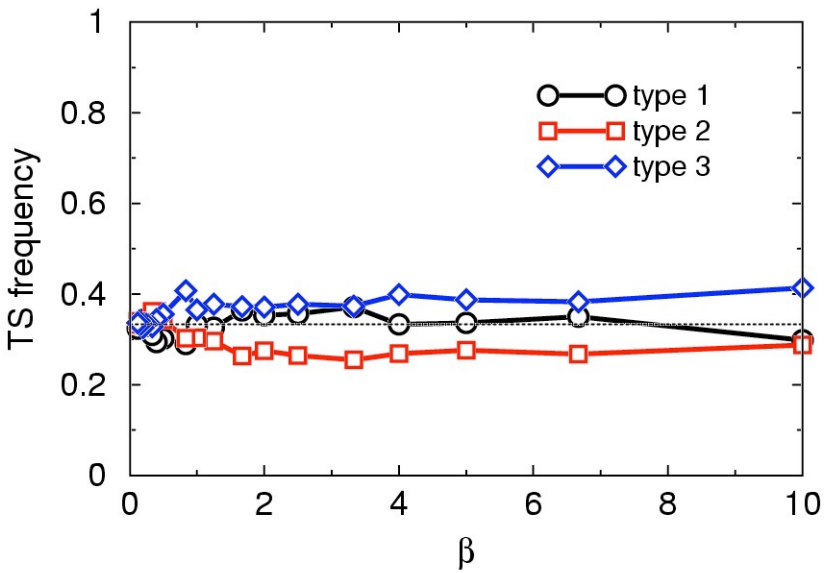




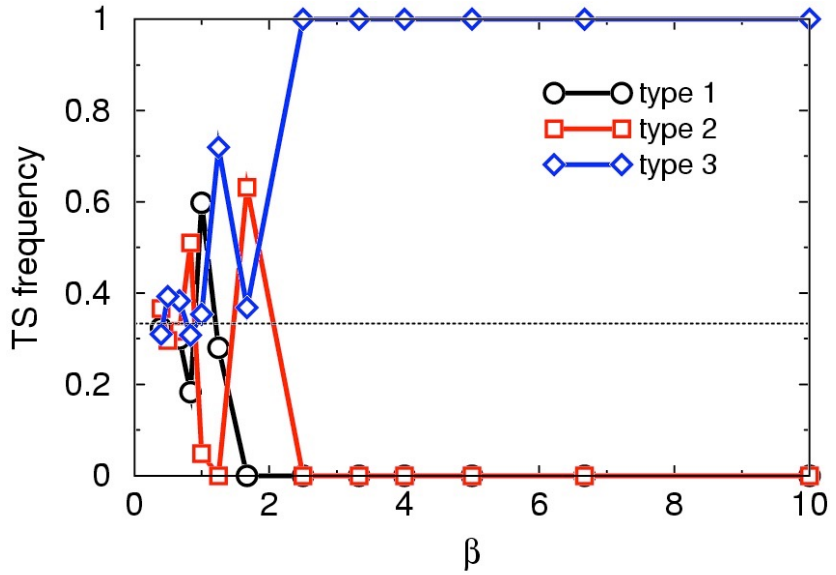
E. Borrero and C. Dellago, JCP 133, 134112 (2010)

Parallel tempering



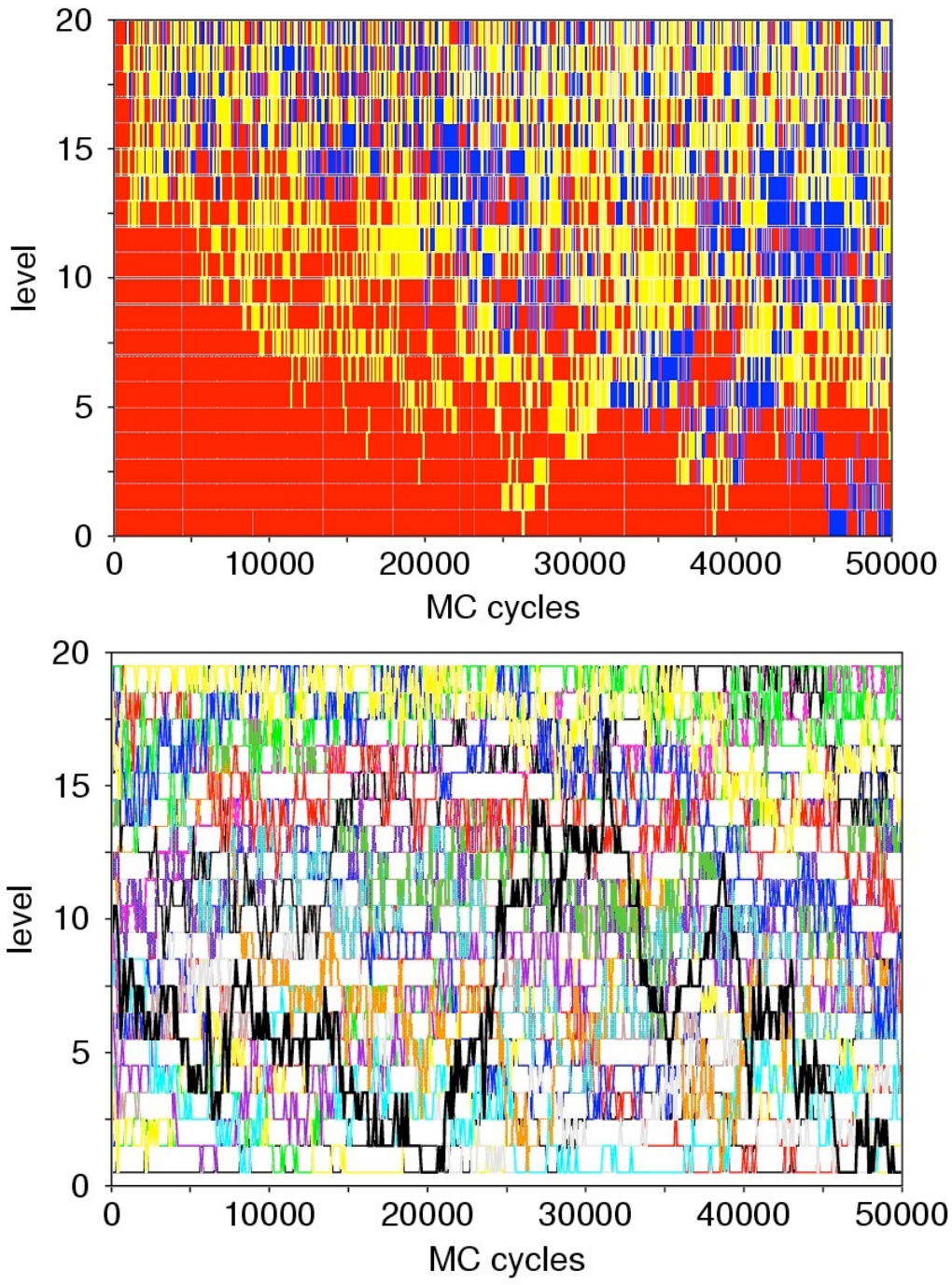


with parallel tempering

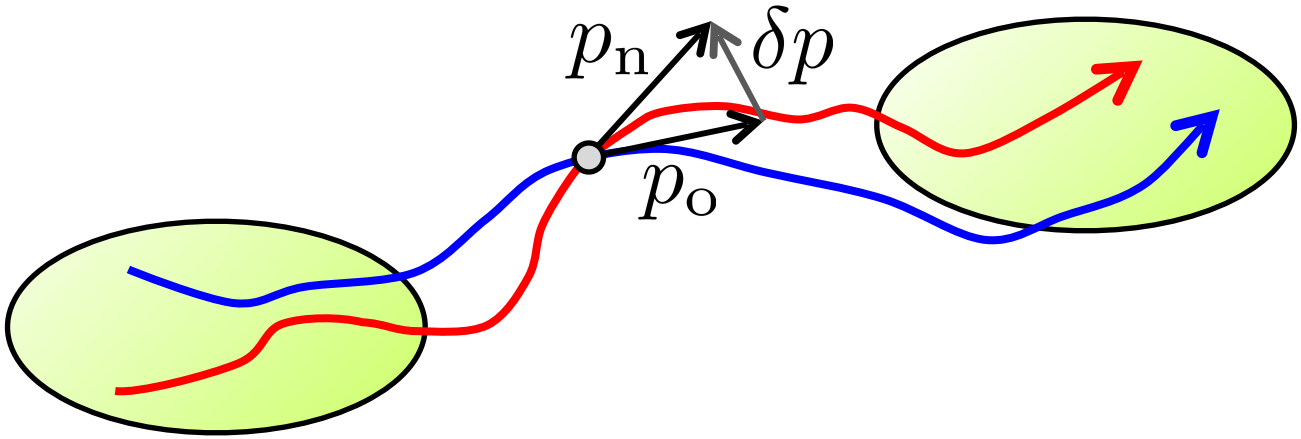


without parallel tempering

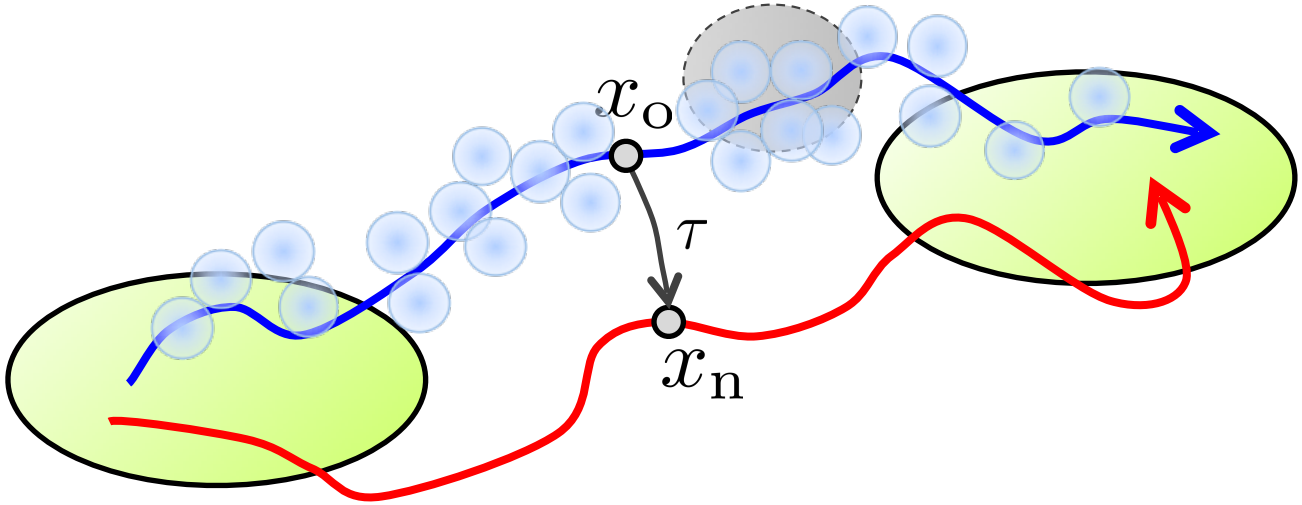
T. J. H. Vlugt and B. Smit, Phys. Chem. Comm. 2, 1 (2001)


level
MC cycles
0
5
10
15
20
0
10000
20000
30000
40000
50000

Shooting moves + metadynamics

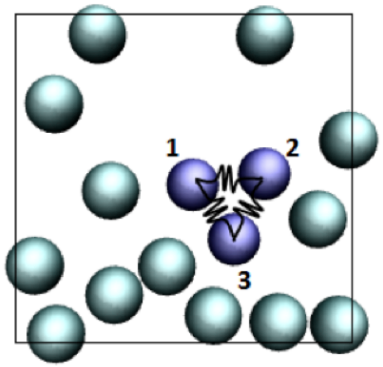


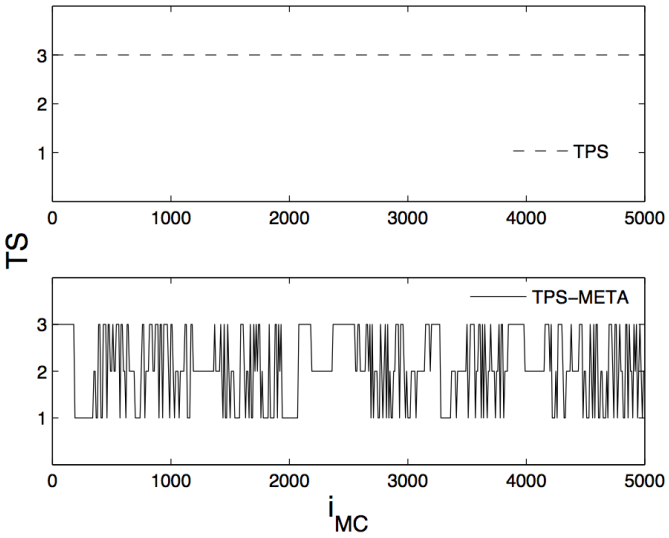
regular
shooting

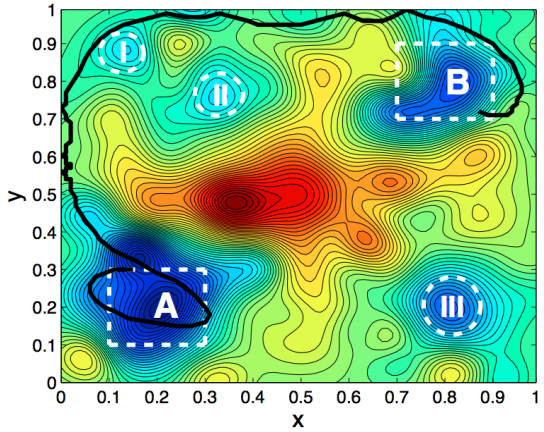


Meta-shooting

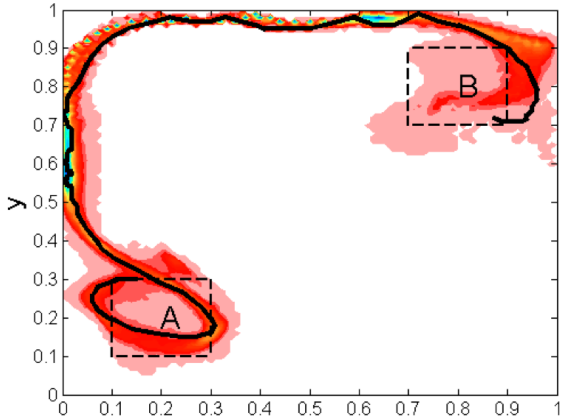
TPS + metadynamics



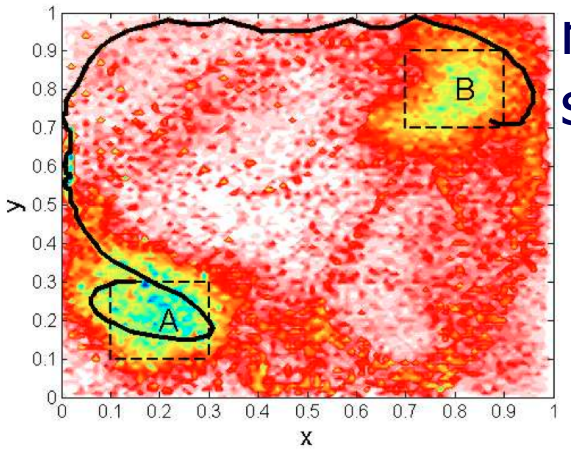




2d PES

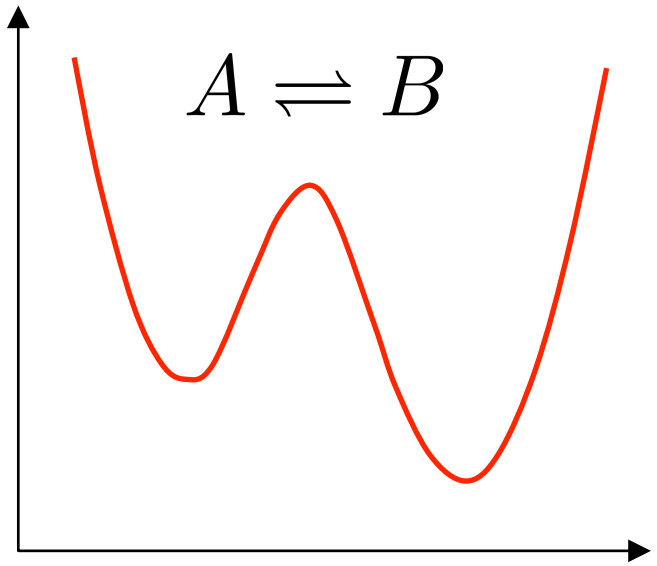


regular TPS



meta shooting

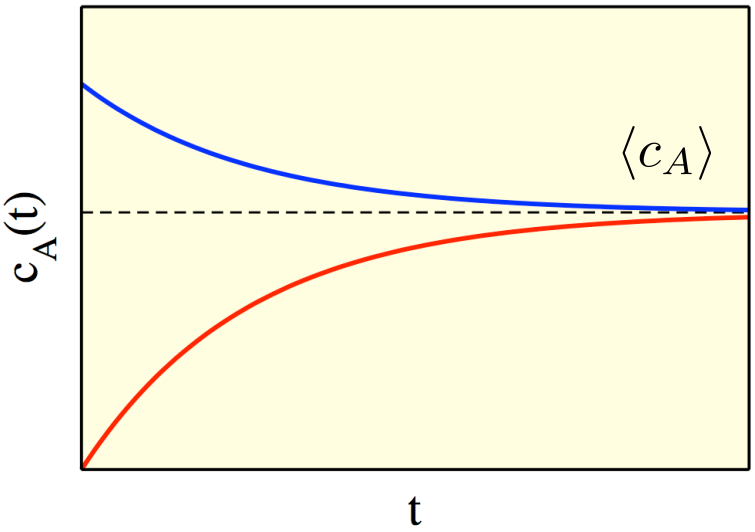
Kinetics - macroscopic picture



Populations $c_A(t),\ c_B(t)$

$$\begin{aligned} \dot{c}_A(t) &= -k_{AB}\, c_A(t) + k_{BA}\, c_B(t) \\ \dot{c}_B(t) &= k_{AB}\, c_A(t) - k_{BA}\, c_B(t) \end{aligned}$$

$$\Delta c_A(t) = c_A(t) - \langle c_A \rangle$$

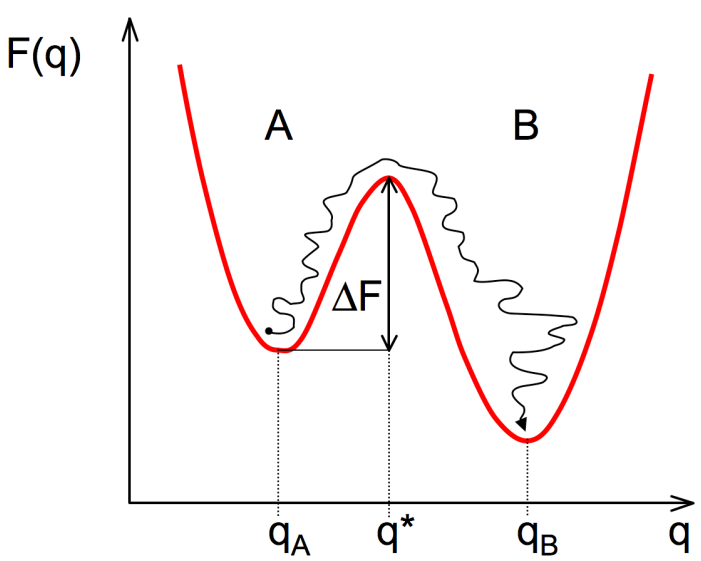


$$\Delta c_A(t) = \Delta c_A(0) \exp(-t/\tau_{\mathrm{rxn}})$$

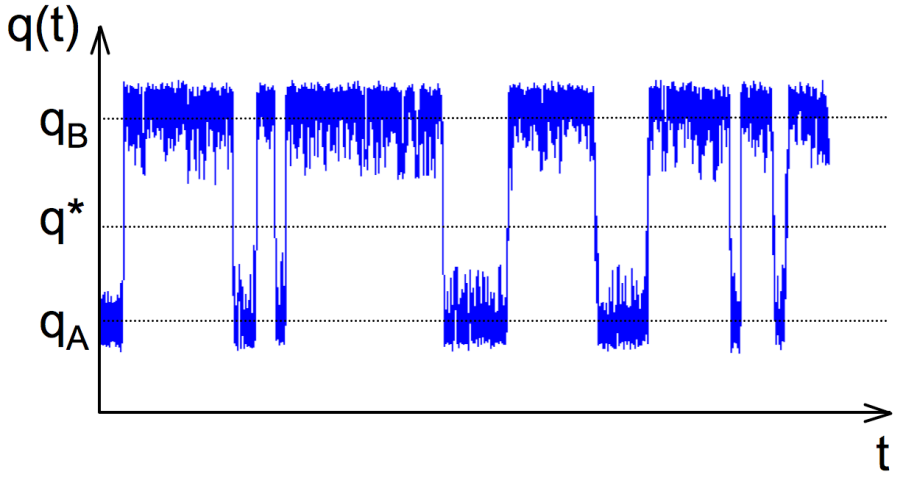
$$\tau_{\mathrm{rxn}}^{-1} = k_{AB} + k_{BA}$$

Reaction time τ_{rxn}

Microscopic picture



collective variable q(x)

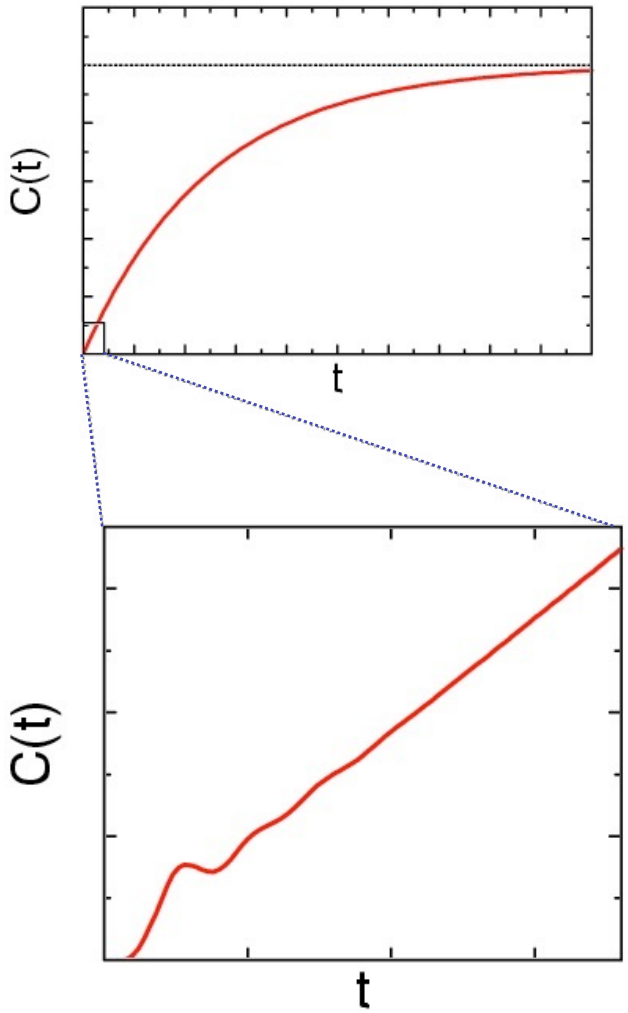


definition of stables states A and B

$$h_A\left[q(x)\right] = \theta\left[q^* - q(x)\right] = \begin{cases} 1 & \text{if } q(x) < q^* \\ 0 & \text{else} \end{cases}$$

$$h_B\left[q(x)\right] = \theta\left[q(x) - q^*\right] = \begin{cases} 1 & \text{if } q(x) > q^* \\ 0 & \text{else} \end{cases}$$

Reaction rate constants from TPS



$$P(B,t|A,0) = \frac{\langle h_A(x_0) h_B(x_t) \rangle}{\langle h_A \rangle}$$

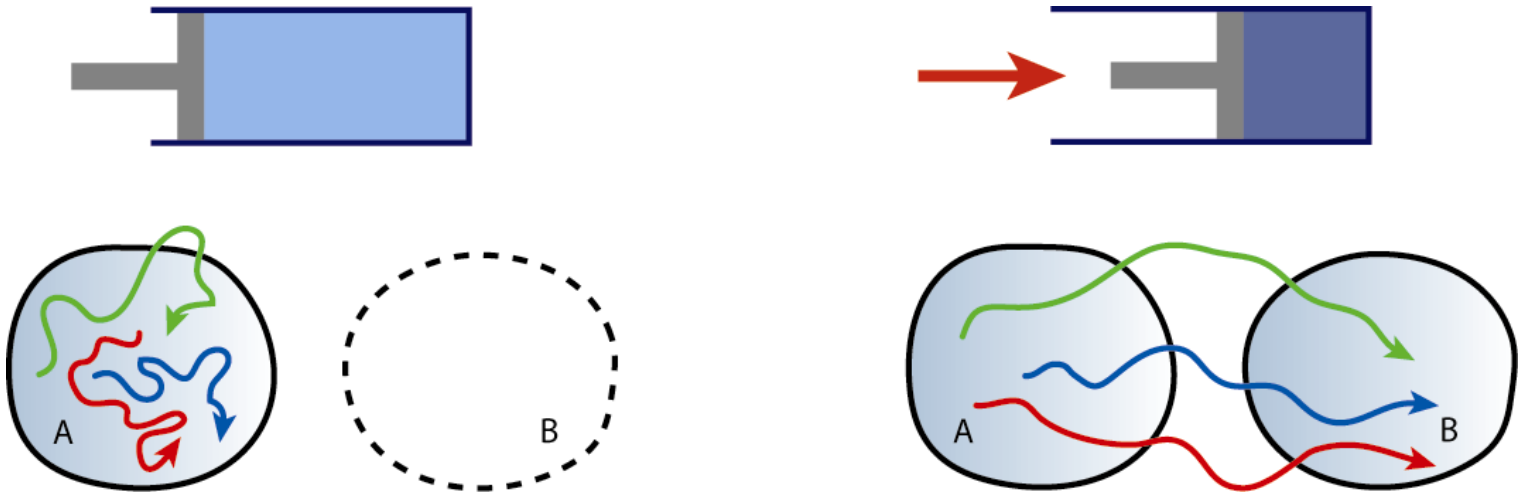
$$\frac{\langle h_A(x_0) h_B(x_t) \rangle}{\langle h_A \rangle} = \langle h_B \rangle \left(1 - e^{-t/\tau_{\mathrm{rxn}}}\right)$$

for $\tau_{\mathrm{mol}} < t \ll \tau_{\mathrm{rxn}}$

$$C(t) \equiv \frac{\langle h_A(x_0) h_B(x_t) \rangle}{\langle h_A \rangle} = k_{AB} t$$

Free energy of path ensembles

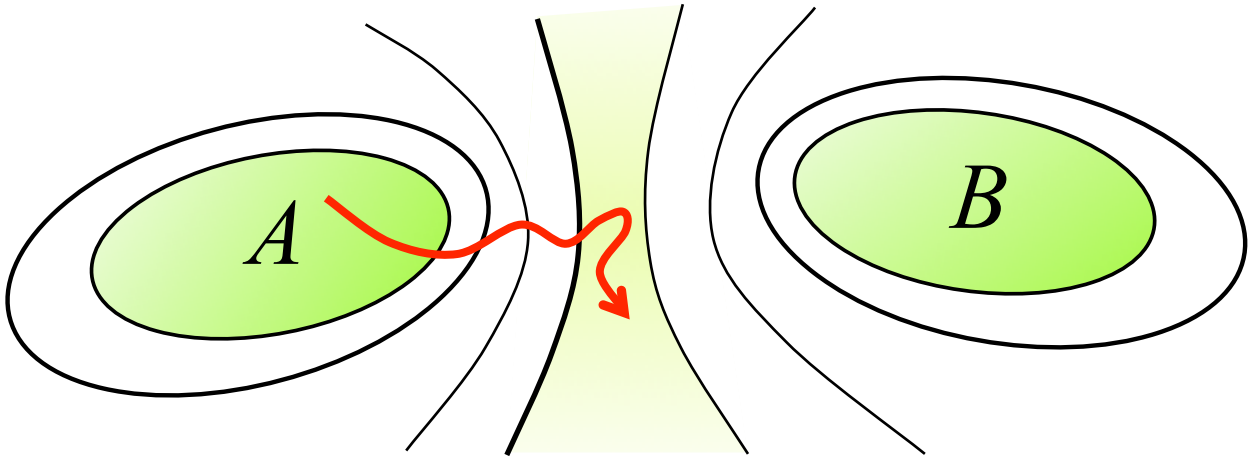
$$C(t) = \frac{\int \mathcal{D}x(t)\, h_A(x_0)\mathcal{P}[x(t)]h_B(x_t)}{\int \mathcal{D}x(t)\, h_A(x_0)\mathcal{P}[x(t)]} = \frac{Z_{AB}(t)}{Z_A}$$



$$W_{AB}(t) \equiv -\ln \frac{Z_{AB}(t)}{Z_A}$$

C. Dellago and P. L. Geissler, AIP Conf. Proc. 690, (2003)
P. L. Geissler and C. Dellago, JPC B 108, 6667 (2004)

Umbrella sampling

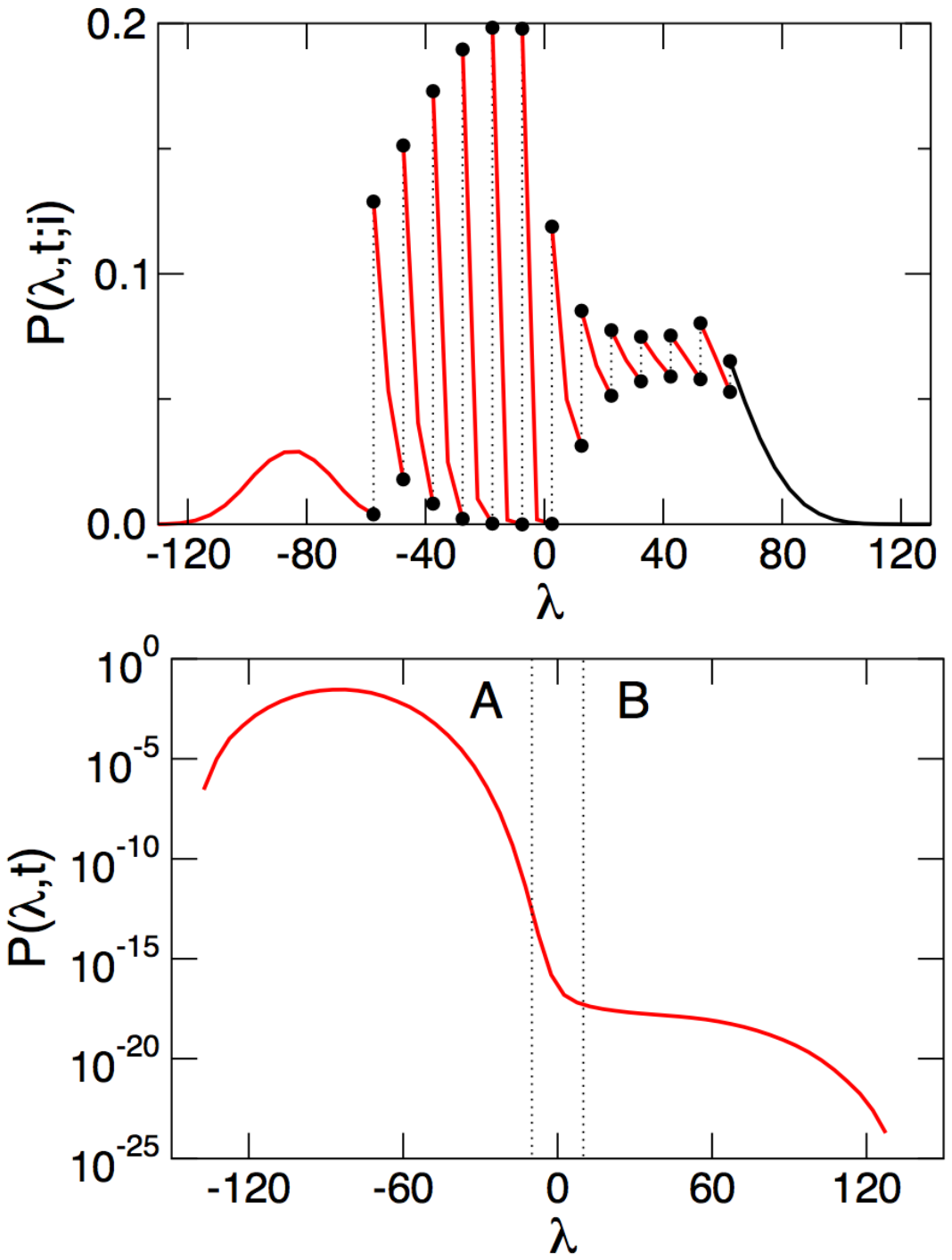


$\mathcal{W}[i]$

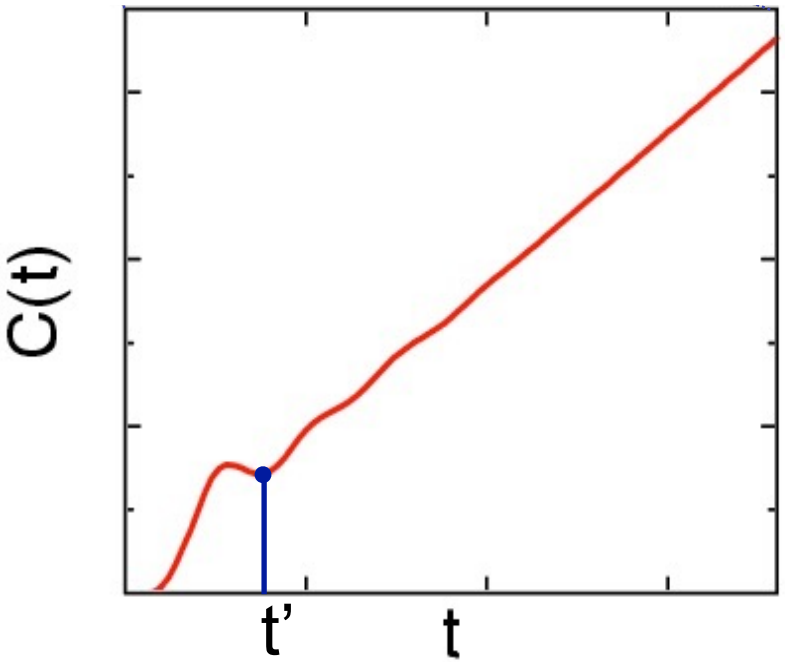
$$P_A(\tilde{\lambda},t)=\frac{\int\mathcal{D}x(t)\,h_A(x_0)\mathcal{P}[x(t)]\delta[\tilde{\lambda}-\lambda(x_t)]}{Z_A}=\langle\delta[\tilde{\lambda}-\lambda(x_t)]\rangle_A$$

$$C(t)=\exp[-W_{AB}(t)]=\int_{\lambda^B_{\min}}^{\lambda^B_{\max}}d\lambda\,P_A(\lambda,t)$$

$$P_{A\mathcal{W}[i]}(\tilde{\lambda},t)=\frac{\int\mathcal{D}x(t)\,h_A(x_0)\mathcal{P}[x(t)]h_{\mathcal{W}[i]}(x_t)\delta[\tilde{\lambda}-\lambda(x_t)]}{\int\mathcal{D}x(t)\,h_A(x_0)h_{\mathcal{W}[i]}(x_t)}$$
$$=\langle\delta[\tilde{\lambda}-\lambda(x_t)]\rangle_{A\mathcal{W}[i]}.$$


0.2
0.1
0.0
P(λ,t;i)
-120
-80
-40
0
40
80
120
λ
10^0
10^-5
10^-10
10^-15
10^-20
10^-25
P(λ,t)
A
B
-120
-60
0
60
120
λ

Reversible work to change t



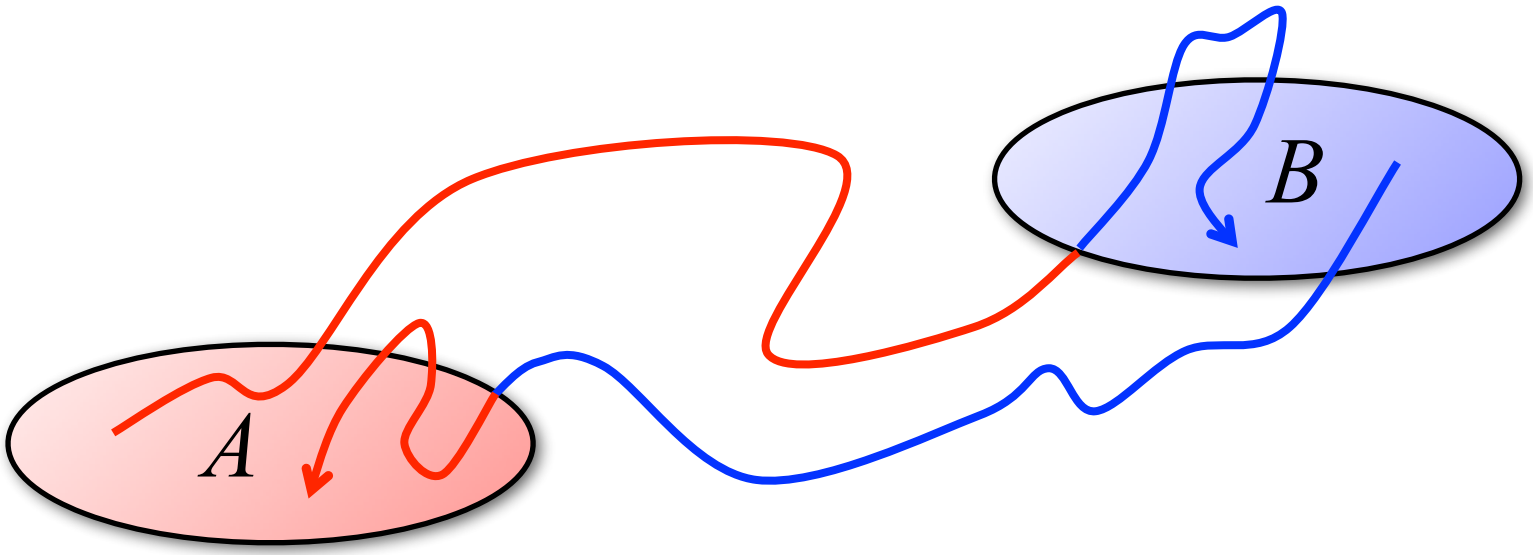
$$C(t) = \frac{\langle h_A(x_0) h_B(x_t) \rangle}{\langle h_A(x_0) h_B(x_{t'}) \rangle} \times C(t')$$

$$\exp(-\Delta W_{AB}(t,t')) = \frac{C(t)}{C(t')}$$

$$W_{AB}(t) = W_{AB}(t') + \Delta W_{AB}(t;t')$$

$$\exp(-\Delta W_{AB}(t,t')) = \frac{\langle h_A(x_0) h_B(x_t) H_B[x(\mathcal{T})] \rangle}{\langle h_A(x_0) H_B[x(\mathcal{T})] \rangle} \times \frac{\langle h_A(x_0) H_B[x(\mathcal{T})] \rangle}{\langle h_A(x_0) h_B(x_{t'}) H_B[x(\mathcal{T})] \rangle}$$

Transition interface sampling



Overall states in phase space:

$\mathcal{A}$ going *back in time* **A** reached first

$\mathcal{B}$ going *back in time* **B** reached first

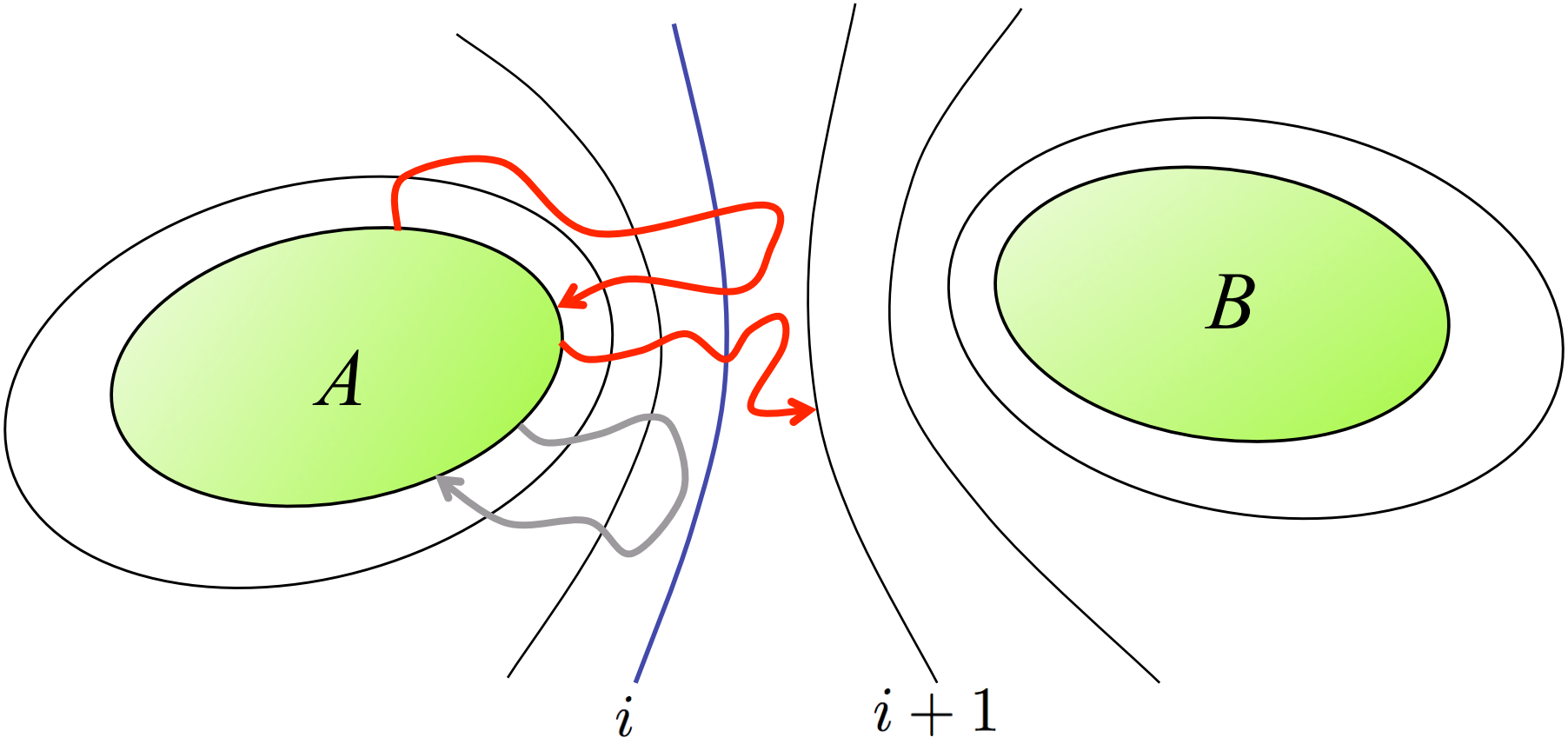
$$C(t) \equiv \frac{\langle h_{\mathcal{A}}(x_0) h_{\mathcal{B}}(x_t) \rangle}{\langle h_{\mathcal{A}} \rangle}$$

$$k_{AB} = \frac{\langle h_{\mathcal{A}}(x_0) \dot{h}_{\mathcal{B}}(x_0) \rangle}{\langle h_{\mathcal{A}} \rangle} = \frac{\langle \phi_{AB} \rangle}{\langle h_{\mathcal{A}} \rangle}$$

Effective positive flux $\langle \phi_{AB} \rangle$

T. S. van Erp, D. Moroni and P. G. Bolhuis, J. Chem. Phys. 118 , 7762 (2003)
T. S. van Erp and P. G. Bolhuis, J. Comp. Phys. 205, 157 (2005)

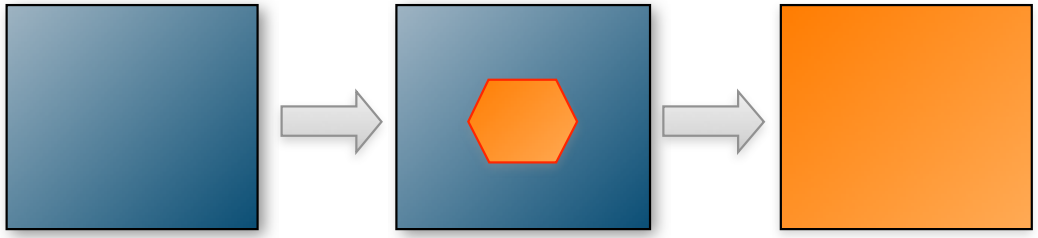
Transition interface sampling



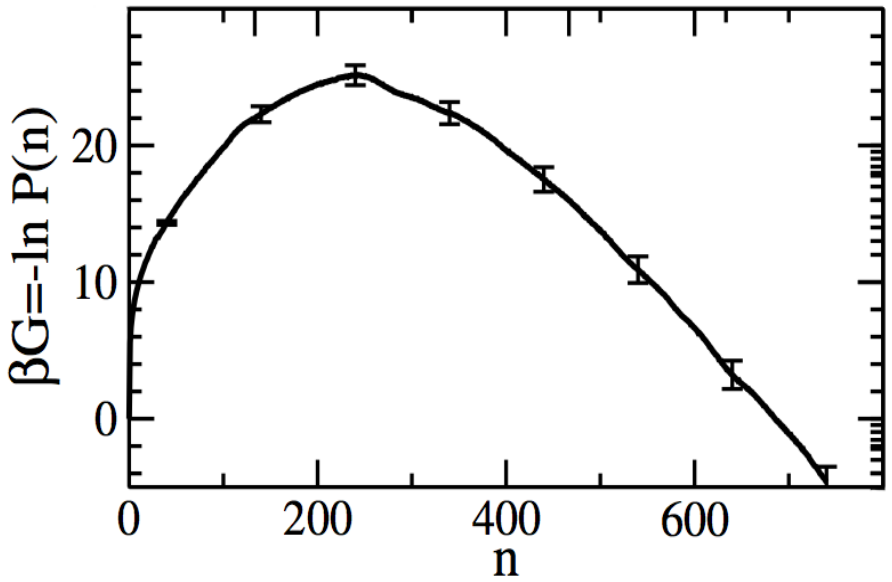
$\mathcal{P}_A(i+1|i)$ = probability that path crossing i for first time after leaving A reaches i+1 before A

$$k_{AB} = \frac{\langle \phi_{AB} \rangle}{\langle h_A \rangle} = \frac{\langle \phi_{A1} \rangle}{\langle h_A \rangle} \prod_{i=1}^{n-1} \mathcal{P}_A(i+1|i)$$

Crystallization in Lennard-Jonesium



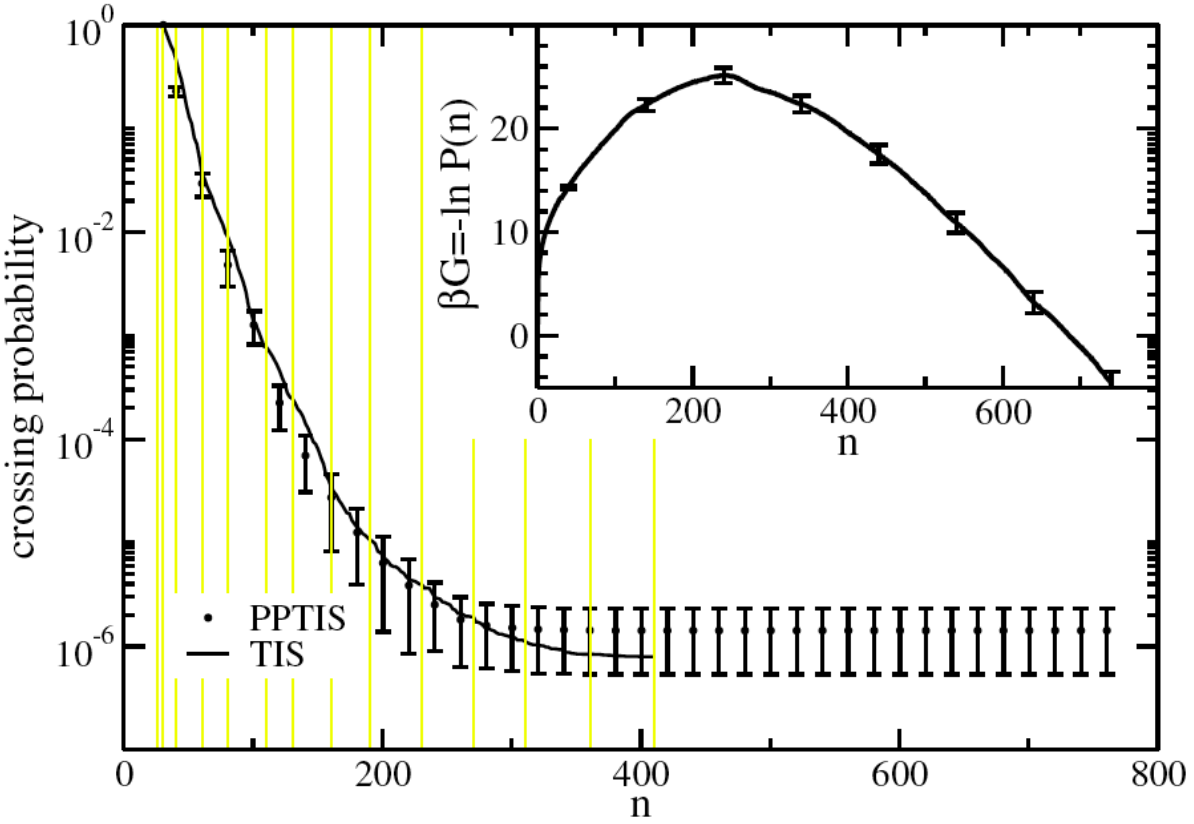
Lennard-Jones fluid
25% undercooling



n = size of the largest crystalline cluster

D. Moroni, P. R. ten Wolde, and P. G. Bolhuis, Phys. Rev. Lett. 94, 235703 (2005)

TIS for crystallization of LJ

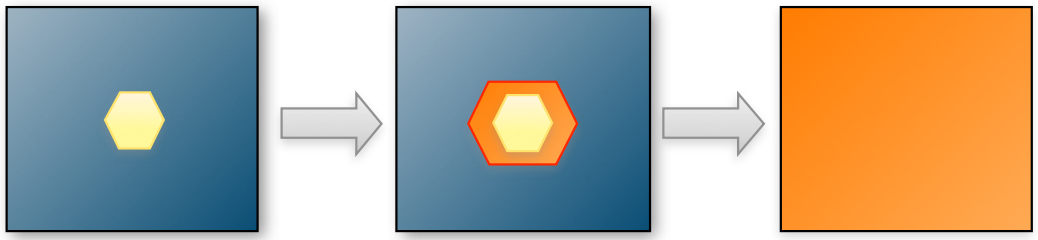


$$\frac{\langle \phi_{A1} \rangle}{\langle h_{\mathcal{A}} \rangle} = 1.29 \qquad \mathcal{P}_A(B|1) = \prod_{i=1}^{n-1} \mathcal{P}_A(i+1|i) = 8 \times 10^{-7}$$

$$k_{AB} = (1.0 \pm 0.8) \times 10^{-6}$$

D. Moroni, P. R. ten Wolde, and P. G. Bolhuis, Phys. Rev. Lett. 94, 235703 (2005)

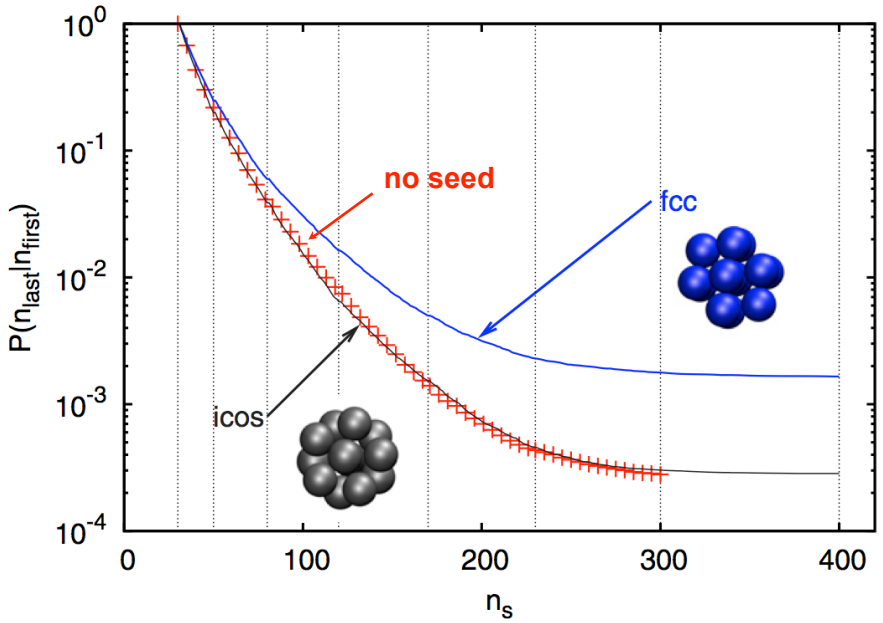
Crystallization on tiny templates



liquid+template solid

crossing probability $$P_A(\lambda_j|\lambda_1) = \prod_{i=1}^{j-1} P_A(\lambda_{i+1}|\lambda_i)$$

- **TIS**
- 28% undercooling
- Lennard-Jones particles
- N=6600 particles
- Seed size n=13
- NpH-ensemble



$J_{fcc}=1.7\times10^{-7}$

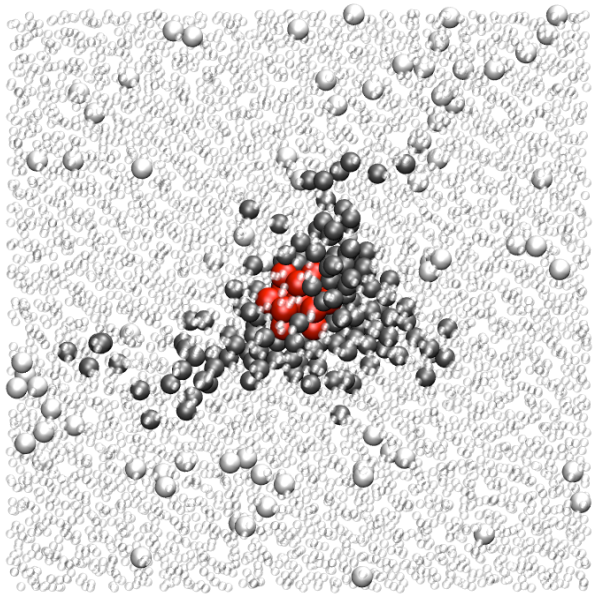
$J_{ico}=3.1\times10^{-8}$

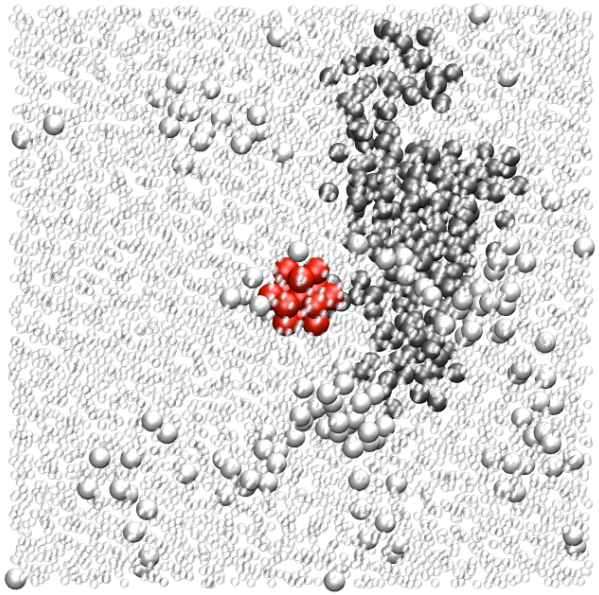
$J_{hom}=3.0\times10^{-8}$

S. Jungblut and C. Dellago, Europhys. Lett. 96, 56006 (2011)

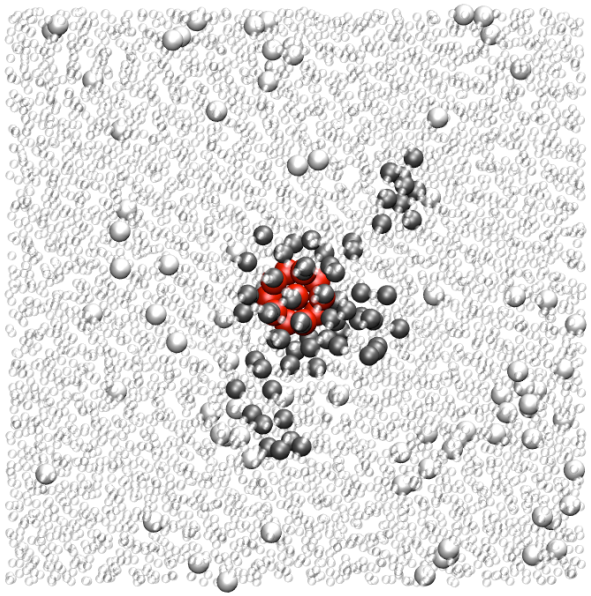
Crystallization Pathways

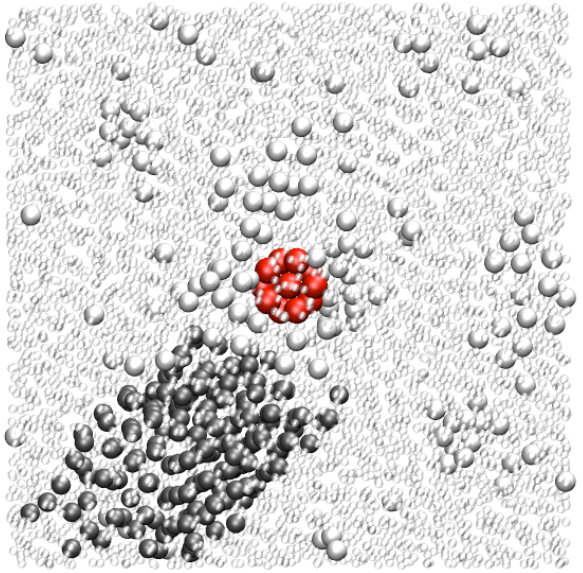
fcc template



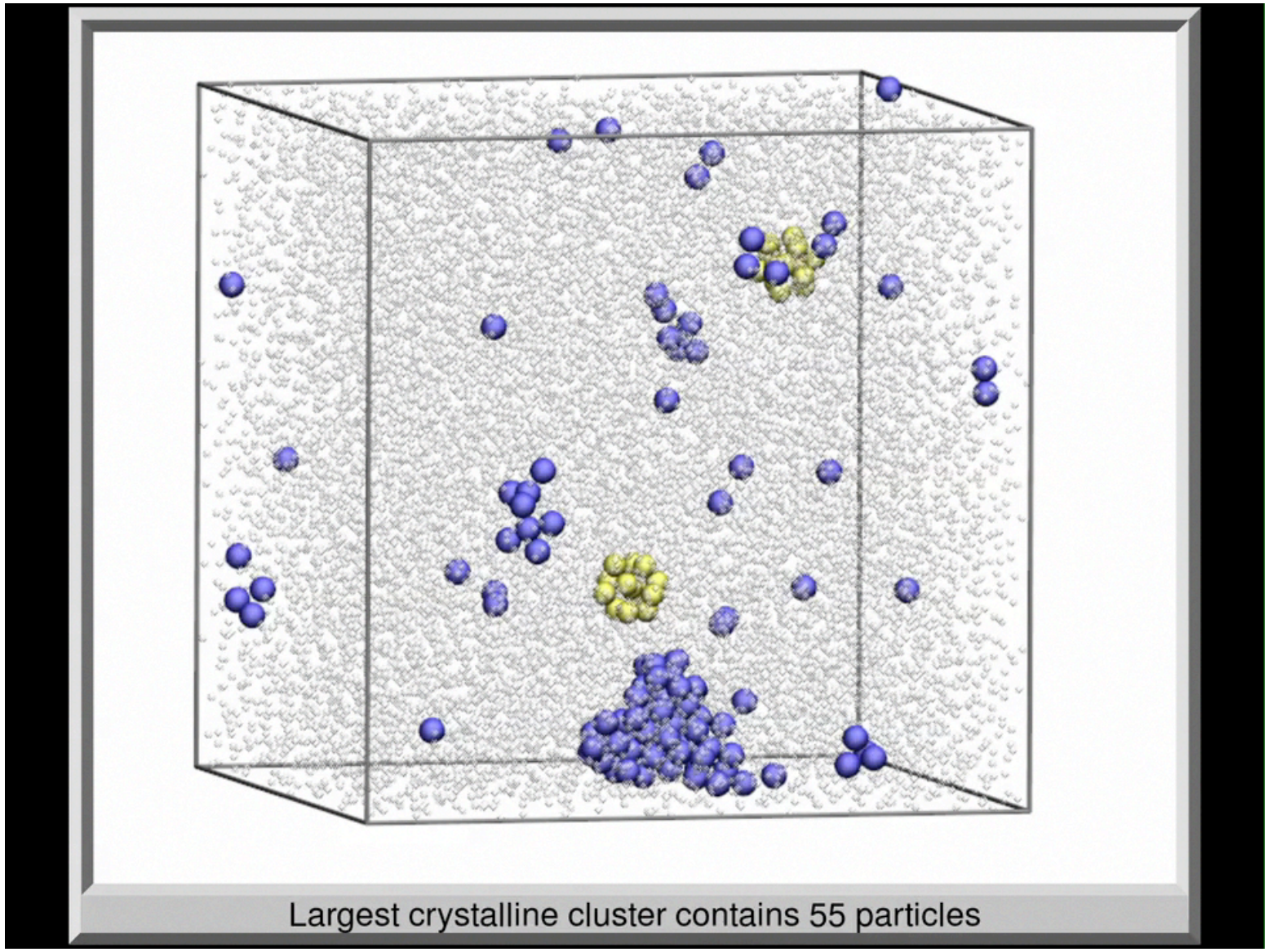


icosahedral template



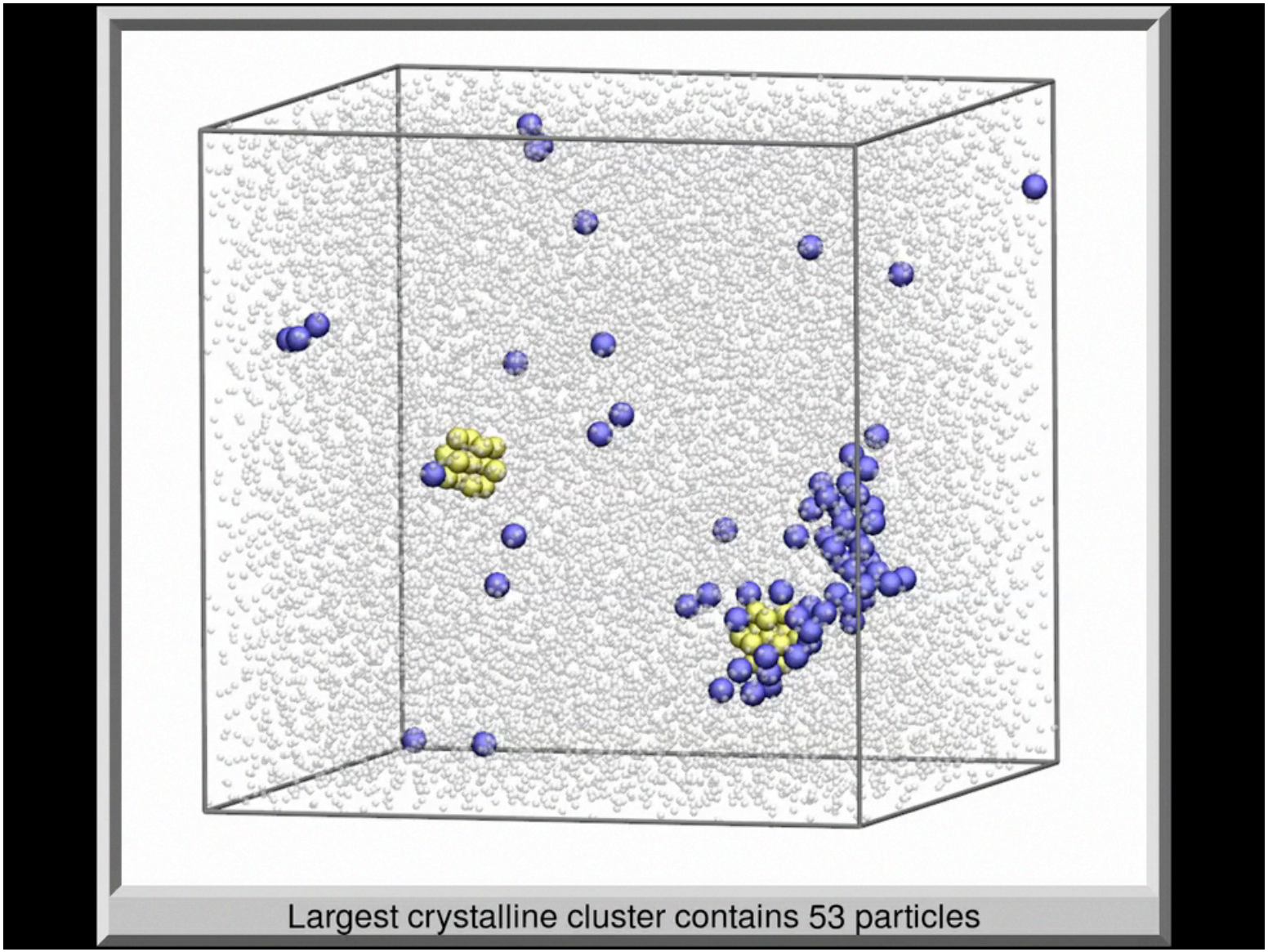


Seeded crystallization in LJ

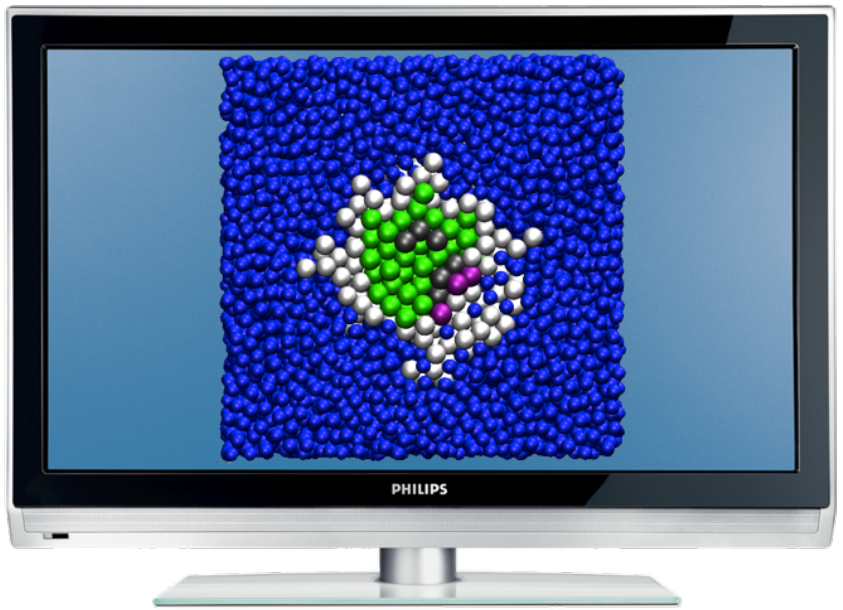


Two **icosahedral** seeds

Seeded crystallization in LJ

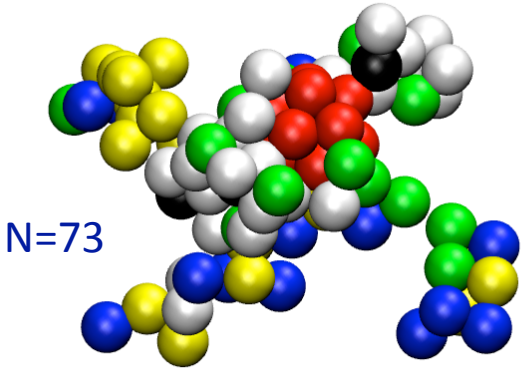


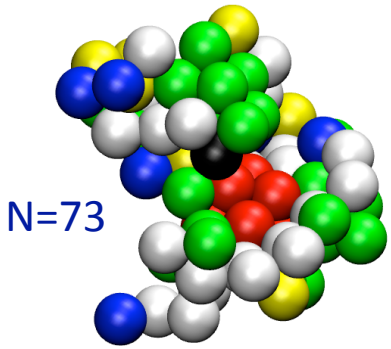
Two **fcc** seeds

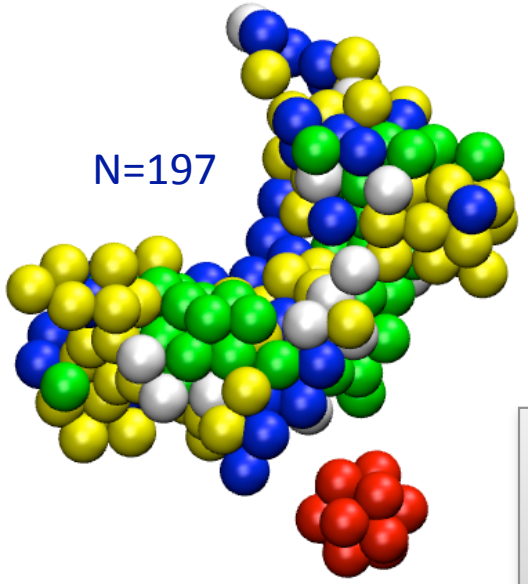


Structure of Critical Clusters

icosahedral template

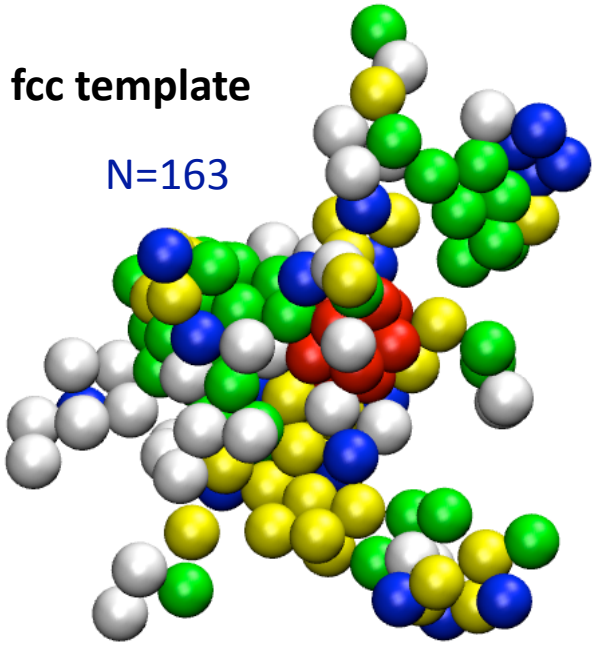


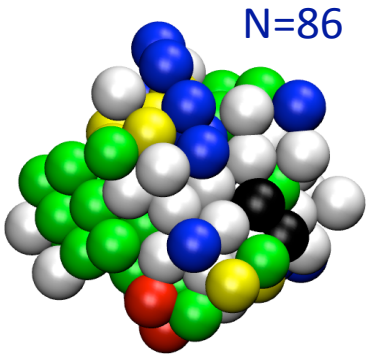


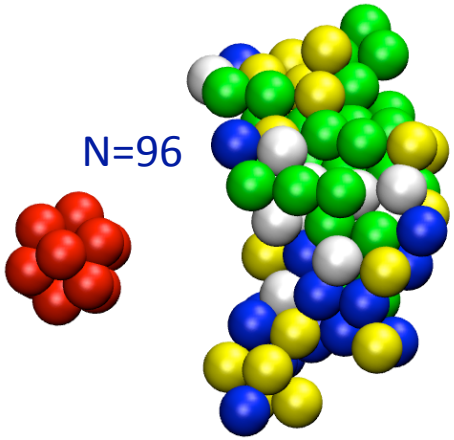


template
icosahedral
fcc
bcc
x-bcc
hcp

fcc template

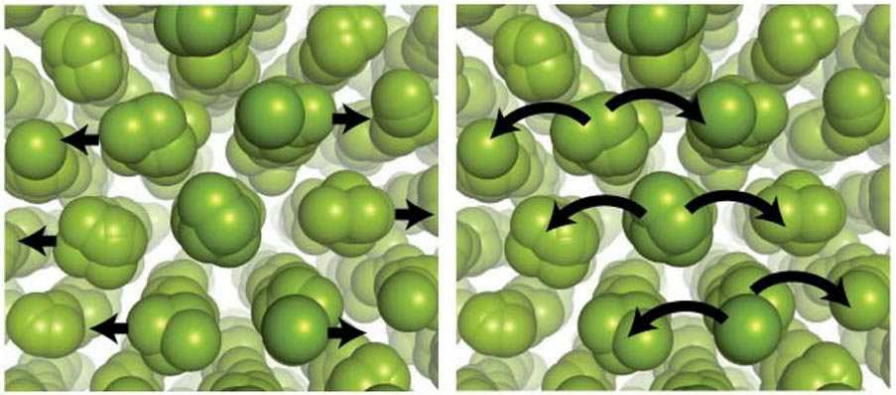


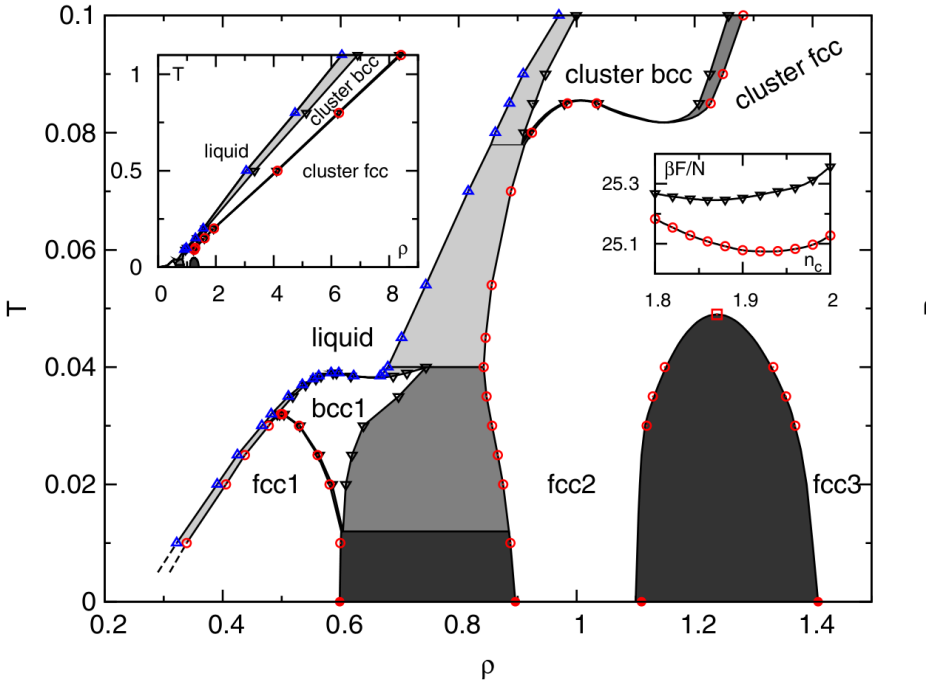




Generalized exponential model GEM4

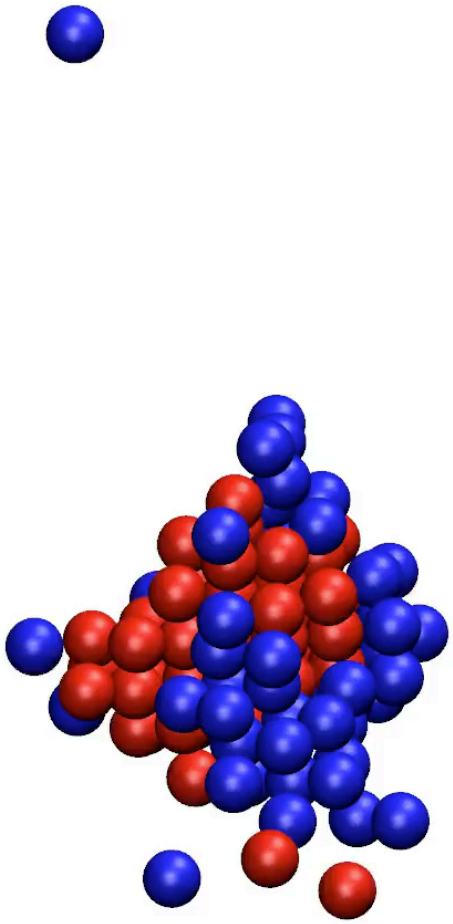
$$\phi(r) = \varepsilon \exp\{-(r/\sigma)^4\}$$





B. M. Mladek, P. Charbonneau and D. Frenkel, PRL 99, 235702 (2007)
Kai Zhang, P. Charbonneau and B. M. Mladek, PRL 105, 245701 (2010)

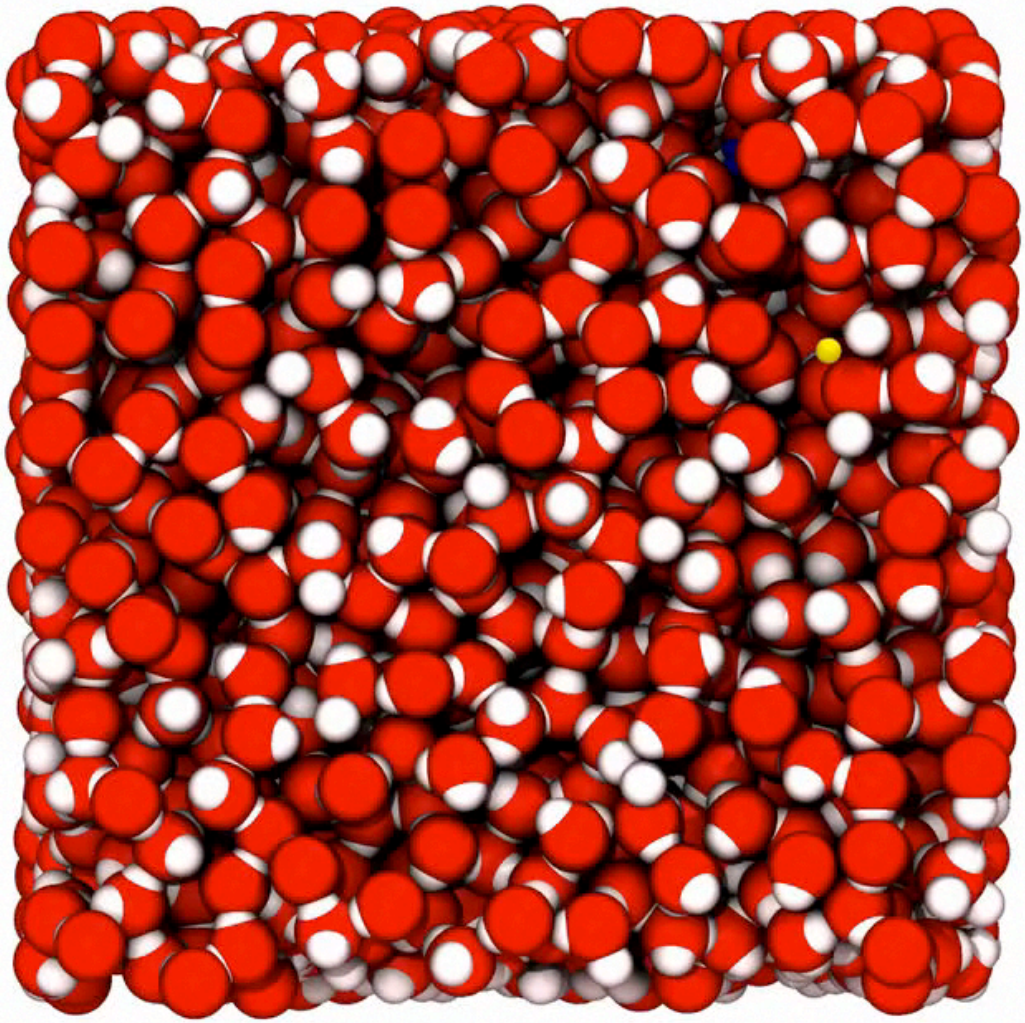
Generalized exponential model GEM4



fcc

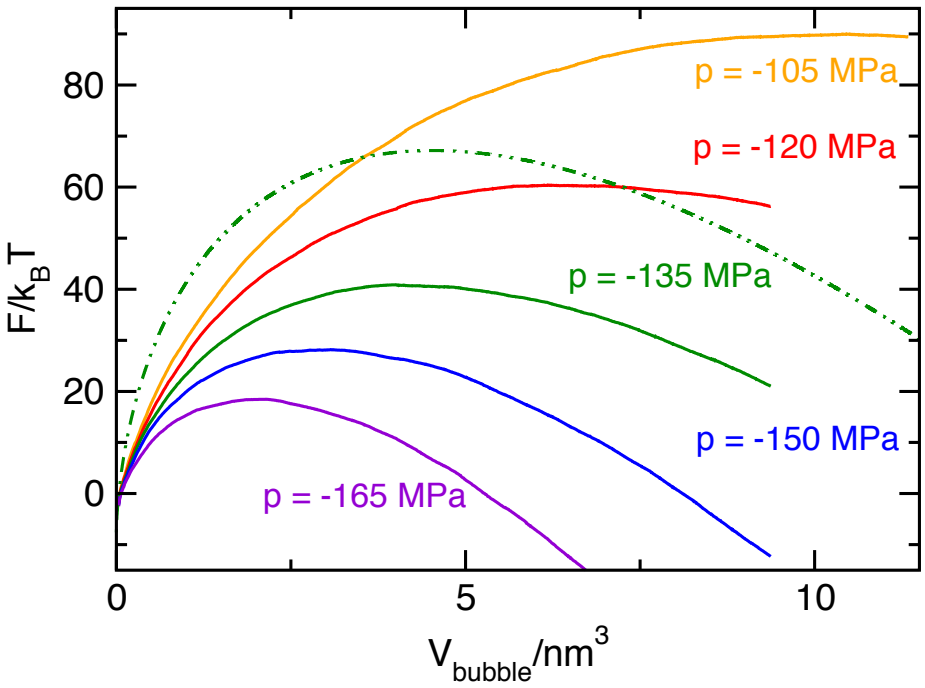
bcc

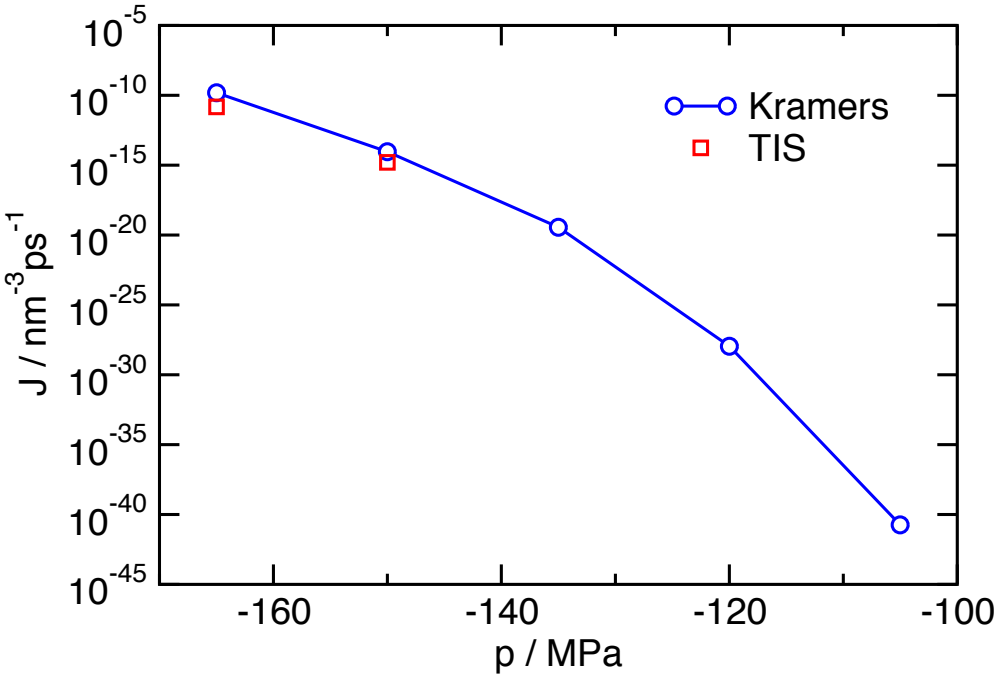
Cavitation in water at negative pressure



Free energetics and kinetics of bubble formation

$$\beta G(V_{\mathrm{bubble}}) = -\ln \rho(V_{\mathrm{bubble}})$$

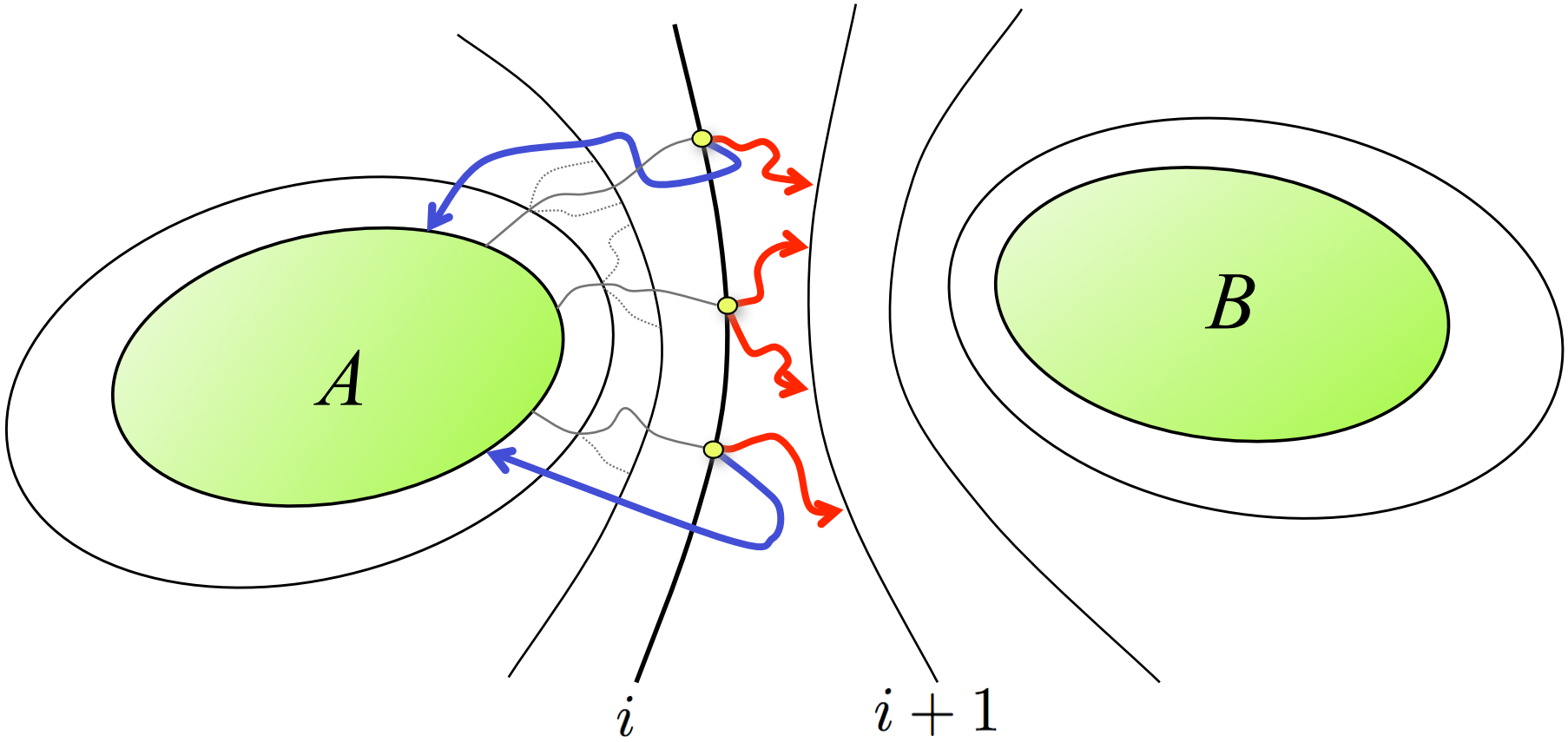




Umbrella sampling with hybrid MC moves

TIP4P/2005
NpT
N=2000
T=296.4 K

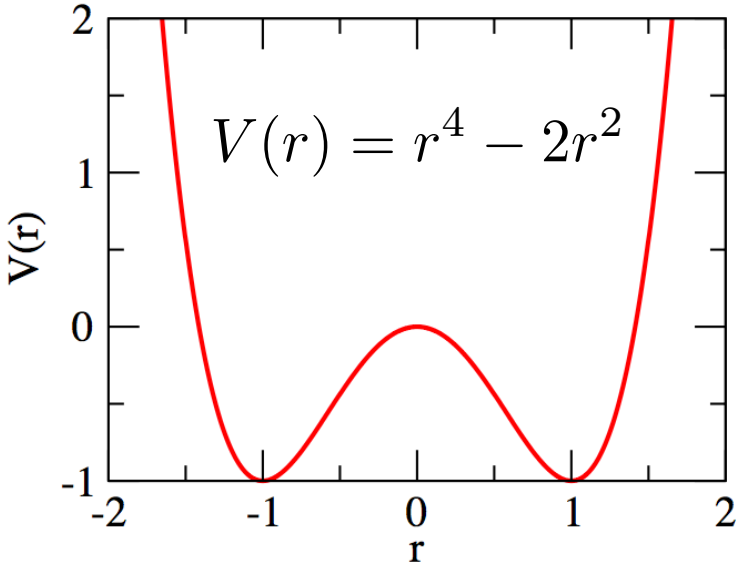
Forward flux sampling



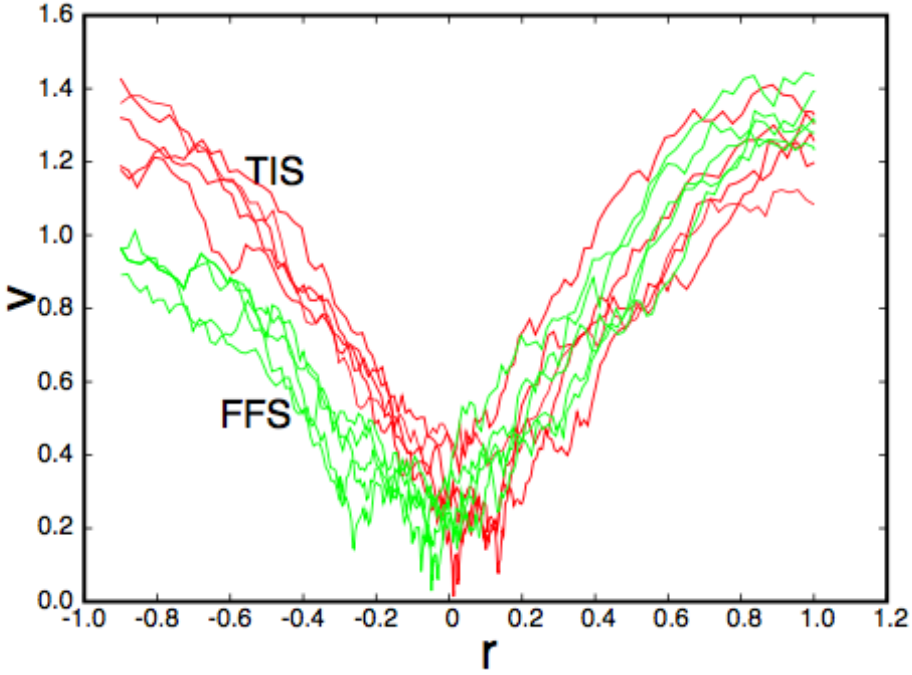
$$k_{AB} = \frac{\langle \phi_{AB} \rangle}{\langle h_{\mathcal{A}} \rangle} = \frac{\langle \phi_{A1} \rangle}{\langle h_{\mathcal{A}} \rangle} \prod_{i=1}^{n-1} \mathcal{P}_A(i+1|i)$$

R. J. Allen, D. Frenkel, P. R. ten Wolde, J. Chem. Phys. 124, 024102 (2006)
R. J. Allen, P. B. Warren, P. R. ten Wolde, Phys. Rev. Lett. 124, 018104 (2005)

Forward flux sampling



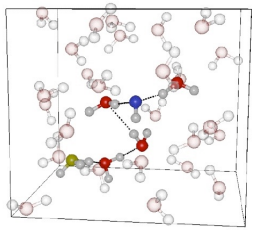
Langevin dynamics



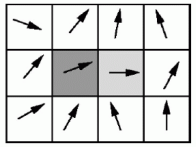
path sampling	f_A	$\mathcal{P}_A(\lambda_n\|\lambda_0)$	k= $f_A \times \mathcal{P}_A(\lambda_n\|\lambda_0)$
TIS	$0.263 \pm 1\%$	$1.52 \cdot 10^{-6} \pm 20\%$	$4.02 \cdot 10^{-7} \pm 20\%$
PPTIS	$0.263 \pm 1\%$	$1.04 \cdot 10^{-6} \pm 19\%$	$2.73 \cdot 10^{-7} \pm 19\%$
RETIS	$0.265 \pm 1\%^*$	$1.05 \cdot 10^{-6} \pm 25\%^*$	$2.79 \cdot 10^{-7} \pm 25\%^*$
FFS (long MD run)	$0.263 \pm 1\%$	$4.69 \cdot 10^{-8} \pm 6\%^*$	$1.23 \cdot 10^{-8} \pm 6\%^*$
FFS (short MD run)	$0.259 \pm 2\%$	$8.45 \cdot 10^{-9} \pm 9\%^*$	$2.18 \cdot 10^{-9} \pm 9\%^*$

T. van Erp, Adv. Chem. Phys., 151, 27 (2012)

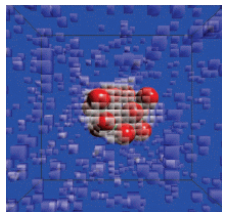
TPS applications



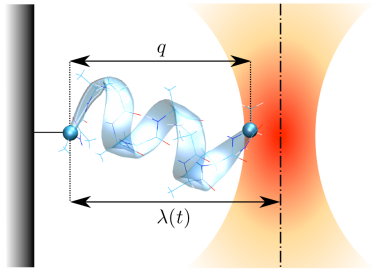
autoionization in water



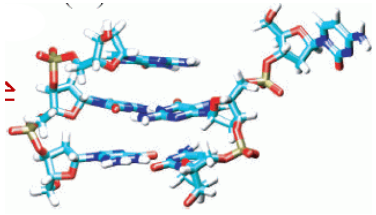
solvation dynamics



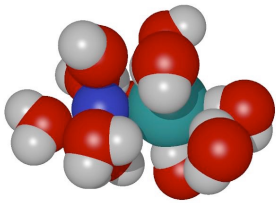
hydrophobic collapse



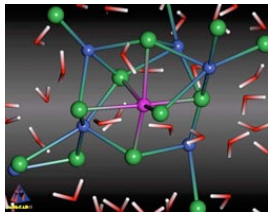
Jarzynski theorem



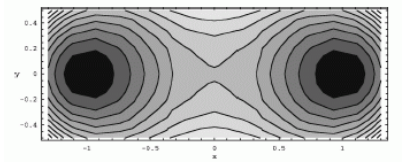
DNA binding



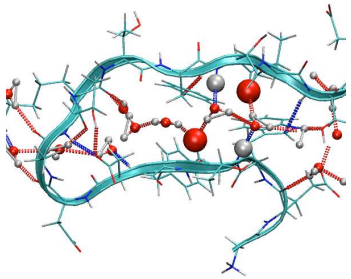
ionic dissociation



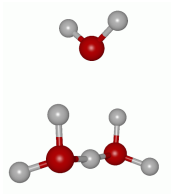
crystallization



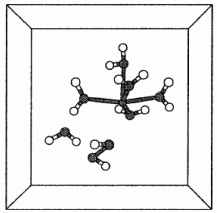
nonequilibrium dynamics



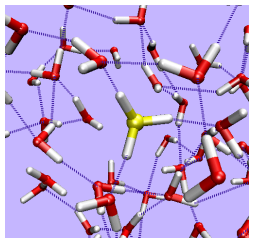
protein folding



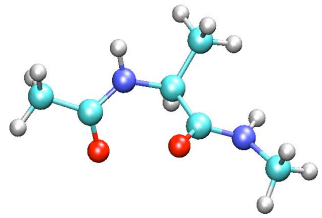
waterclusters



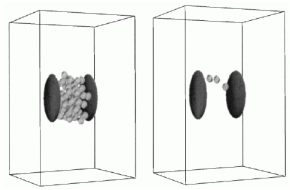
chemical reactions



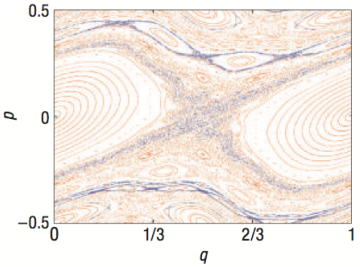
proton transfer



biomolecular isomerization



hydrophobic effect



chaotic dynamics