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Workshop: Numerical methods in molecular simulation

From mesoscopic to microscopic: Reconstruction schemes for stochastic lattice systems

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Stochastic modeling and simulations for many particle (N) systems.

Relevant scales:

- Microscale: stochastic process (spin-flip dynamics, MC algorithm)

- Coarse-grained: CGMC

- Mesoscale: Free energy and evolution for "order parameter" (LLN)

- Macroscale: Free energy and evolution for phase boundary

Question: Good numerical tool to capture effects of stochasticity.

Motivation from polymer science literature, e.g.

1. Tschöp, Kremer, Hahn, Batoulis, Bürger, 1998

2. K. Kremer, F. Müller-Plathe, 2001

3. Reinier, Akkermans, Briels, 2001

4. Fukunaga, Takimo, Doi, 2002

5. Harmandaris, Adhikari, van der Vegt, Kremer, 2006

Instead of running directly microscopic simulations, we follow the scheme: (Müller-Plathe, *Chem. Phys. Chem*, 2002)

1. Derivation of a CG model from the original microscopic model.

2. CG simulation.

3. *Reconstruction:* Reverse mapping of the CG model into a *reconstructed microscopic model*.

4. Simulation of the reconstructed microscopic model.

The model, equilibrium theory

Hamiltonian:

$$H_N(\sigma) = -\frac{1}{2}\sum_{x\in\Lambda_N}\sum_{y\neq x} J(x,y)\sigma(x)\sigma(y) + h\sum_{x\in\Lambda_N}\sigma(x)$$

where

$$J(x,y) = \frac{1}{L^d} V\left(\frac{n}{L}|x-y|\right), \quad x,y\in\Lambda_N, \quad N = n^d$$

Gibbs measure:

$$\mu_{N,\beta}(d\sigma) = \frac{1}{Z_N} e^{-\beta H_N(\sigma)} P_N(d\sigma), \quad P_N(d\sigma) = \prod_{x\in\Lambda_N} \rho(d\sigma(x))$$

(i) CG of the configuration space

Partition $\mathbf{T}^d$ into m^d boxes C_k

Block Average Transformation $\mathbf{F} : \sigma \mapsto \{\sum_{x \in C_k} \sigma(x)\}_k$

C-G configuration: $\eta \in \bar{\mathcal{S}}_M \equiv \{-q^d, ..., q^d\}^{\bar{\Lambda}_M}$.

(ii) CG of the a-priori measure

$$\rho(\eta) := P_N(\{\sigma : \mathbf{F}(\sigma) = \eta\}) = \prod_k \binom{q}{\frac{\eta(k)+q}{2}} (\frac{1}{2})^q$$

(iii) CG of the Hamiltonian Effective coarse-grained Hamiltonian:

$$e^{-\beta \bar{H}(\eta)} = \frac{1}{\rho(\eta)} \int_{\{\sigma : \mathbf{F}(\sigma)=\eta\}} e^{-\beta H_N} P_N(d\sigma)$$

Define:

$$\bar{\mu}(\eta) = \frac{1}{\bar{\bar{Z}}} e^{-\beta \bar{H}(\eta)} \rho(\eta)$$

Notice that $\mathcal{R}(\bar{\mu}|\mu_N \circ F^{-1}) = \sum_\eta \bar{\mu}(\eta) \log \frac{\bar{\mu}(\eta)}{\mu_N \circ F^{-1}} = 0$

From [Katsoulakis, Plechac, Rey-Bellet, T., M2AN 2007] we have:

$$\bar{H}(\eta) = \bar{H}^{(0)}(\eta) + \bar{H}^{(1)}(\eta) + \bar{H}^{(2)}(\eta) + m\mathcal{O}(\epsilon^3) \tag{1}$$

where

$$\begin{aligned}
\bar{H}^{(0)}(\eta) \quad &:= \quad \mathbf{E}[H_N|\eta] \equiv \int H_N(\sigma) P_N(d\sigma|\,\{\sigma : \mathbf{F}(\sigma) = \eta\}) \\
&= \quad -\frac{1}{2}\sum_{k,l\neq k} \bar{J}(k,l)\eta(k)\eta(l) - \frac{1}{2}\sum_{k} \bar{J}(k,k)\eta(k)(\eta(k)-1)
\end{aligned}$$

and

$$\bar{J}(k,l) = \frac{1}{q^2}\sum_{x\in C_k, y\in C_l} J(x-y)$$

Obtain:

$$\bar{H}(\eta) = \bar{H}^{(0)}(\eta) - \frac{1}{\beta}\log \mathbf{E}[e^{-\beta(H_N(\sigma)-\bar{H}^{(0)}(\eta))}|\eta]$$

where

$$H_N(\sigma) - \bar{H}^{(0)}(\eta) = -\frac{1}{2} \sum_{k \leq l} \sum_{\substack{x \in C_k \\ y \in C_l, y \neq x}} (J(x-y) - \bar{J}(k,l)) \sigma(x) \sigma(y) (2 - \delta_{kl})$$

Note that

$$J(x-y) - \bar{J}(k,l) \sim \mathcal{O}(\frac{q}{L^{d+1}} \|\nabla V\|_\infty)$$

which implies

$$H_N - \bar{H}^{(0)} \sim N \frac{q}{L} \|\nabla V\|_\infty$$

Thus we need to compute:

$$\bar{H}(\eta) = \bar{H}^{(0)}(\eta) - \frac{1}{\beta} \log \int \prod_{k \leq l} (1 + f_{kl}^{\Delta J}(\sigma)) \prod_{k=0}^{m-1} \tilde{\rho}_k(d\sigma)$$

where

$$f_{kl}^{\Delta J}(\sigma) := \exp\{-\beta \Delta_{kl} J(\sigma)\} - 1$$

Using cluster expansion techniques with small parameter

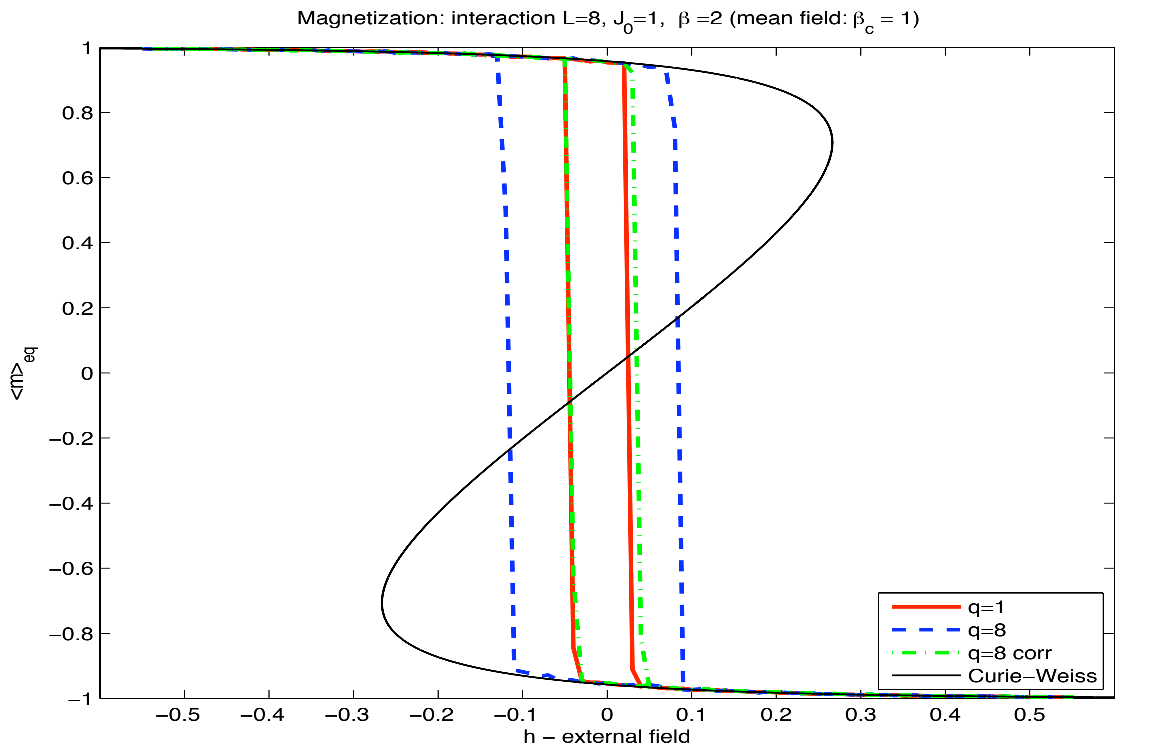
$$\epsilon \sim \beta \frac{q}{L} \|\nabla V\|_\infty$$

we obtain (1).

Then construct new higher order CG Gibbs measures:

$$\bar{\mu}^{(p)}_{m,q,\beta}(\eta) = \frac{1}{\bar{Z}^{(p)}} e^{-\beta(\bar{H}^{(0)}+\ldots+\bar{H}^{(p)})}\rho(\eta)$$

Note that $\mathcal{R}(\bar{\mu}|\bar{\mu}^{(p)}_{m,q,\beta}) \sim O(\epsilon^{p+1})$



Comparison of fully resolved $q = 1$ and coarse-grained $q = 8$ simulations. The interaction range is $L = 8$ and the inverse temperature is fixed at $\beta = 2$.

Reconstruction

Assuming a CG simulation (i.e. for some η), consider the decomposition

$$\mu_N(d\sigma) := \mu_N(d\sigma|\eta)\bar{\mu}_{\mathbf{M}}(\eta)$$

with $\mu_N(\sigma) = 0$ if $F(\sigma) \neq \eta$.

Compare with

$$\nu(d\sigma) := \nu(d\sigma|\eta)\bar{\mu}_{\mathbf{M}}^{(\mathrm{p})}(\eta)$$

Decomposition of the error in two scales:

$$\mathcal{R}(\nu|\mu_N) = \mathcal{R}(\bar{\mu}_{\mathbf{M}}^{(\mathrm{p})}|\bar{\mu}_{\mathbf{M}}) + \sum_{\eta} \bar{\mu}_M^{(p)}(\eta)\, \mathcal{R}(\nu(\cdot|\eta)|\mu_N(\cdot|\eta))$$

Goal: Construct approximations $\nu(\cdot|\eta)$ of $\mu_N(\cdot|\eta)$ s.t.

1. For every $\eta \in \bar{S}_M$ the probability measure $\nu(\cdot,\eta)$ lies within a controled distance from $\mu_N(\cdot|\eta)$.

2. For every $\eta \in \bar{S}_M$ sampling from $\nu(\cdot,\eta)$ is computationally less demanding than any "direct" sampling from $\mu_N(\cdot|\eta)$.

We decompose the domain into odd- and even-indexed reconstruction boxes D and we write:

$$\mu_N(\sigma|\eta) = \mu_{\mathbf{N},\mathcal{E}}(\sigma^{\mathcal{E}}|\eta)\mu_{N,\mathcal{O}}(\sigma^{\mathcal{O}}|\sigma^{\mathcal{E}},\eta).$$

and similarly for $\nu(\cdot|\eta)$.

Error:

$$\begin{aligned}\mathcal{R}(\nu(\cdot|\eta)|\,\mu_N(\cdot|\eta)) \quad &= \quad \sum_{\sigma^{\mathcal{O}}}\nu(\sigma^{\mathcal{O}}|\eta)\sum_{\sigma^{\mathcal{E}}}\nu(\sigma^{\mathcal{O}}|\sigma^{\mathcal{E}},\eta)\ln\frac{\nu(\sigma^{\mathcal{O}}|\sigma^{\mathcal{E}},\eta)}{\mu_{N,\mathcal{O}}(\sigma^{\mathcal{O}}|\sigma^{\mathcal{E}},\eta)} + \\ &\quad \sum_{\sigma^{\mathcal{E}}}\nu(\sigma^{\mathcal{E}}|\eta)\ln\frac{\nu(\sigma^{\mathcal{E}}|\eta)}{\mu_{\mathbf{N},\mathcal{E}}(\sigma^{\mathcal{E}}|\eta)}\sum_{\sigma\mathcal{O}}\nu(\sigma^{\mathcal{O}}|\sigma^{\mathcal{E}},\eta)\end{aligned}$$

Fix $\sigma^{\mathcal{E}} \in S_{N,\mathcal{E}}$. Take $|D| > L$

$\mu_{N,\mathcal{O}}(\cdot|\sigma^{\mathcal{E}},\eta)$ defined on $S_{N,\mathcal{O}}$ factorizes $\Rightarrow$ parallel computations.

Thus, it is not very expensive computationally to consider (with zero error)

$$\nu(\cdot|\sigma^{\mathcal{E}},\eta) := \mu_{N,\mathcal{O}}(\cdot|\sigma^{\mathcal{E}},\eta)$$

So it suffices to focus on the approximation of $\mu_{N,\mathcal{E}}(\alpha|\eta)$ for fixed $\alpha \in S_{N,\mathcal{E}}$. We have:

$$\mu_{N,\mathcal{E}}(\alpha|\eta) = \frac{e^{-\beta \bar{W}_{N,\mathcal{E}}(\alpha,\eta)}}{e^{-\beta \bar{H}_M(\eta)}} \bigotimes_{k \in \mathcal{E} \cap \bar{\Lambda}_M} \tilde{\rho}_k(\alpha^{C_k}),$$

where

$$e^{-\beta \bar{W}_{N,\mathcal{E}}(\alpha,\eta)} = E_N[e^{-\beta H_N(\sigma)}|\alpha,\eta]$$

For the computation of $\bar{W}_{N,\mathcal{E}}(\alpha, \eta)$ we treat the following cases separately:

1. If $Q < L$: mean field type approximation with error of the order $Q/L < 1$.

   Small parameter: $\epsilon \sim \beta \frac{Q}{L} \|\nabla V\|_\infty$

2. If $Q > L$ a mean field approach is not a good approximation any more and we exploit the mixing condition

   Small parameter: $\delta = \beta \frac{L}{|D|}$

$$\sup_l \sup_\eta \sup_\alpha \left| \frac{Z_l(\alpha^{D_{l-1}}, \alpha^{D_{l+1}}; \eta) Z_l(0^{D_{l-1}}, 0^{D_{l+1}}; \eta)}{Z_l(0^{D_{l-1}}, \alpha^{D_{l+1}}; \eta) Z_l(\alpha^{D_{l-1}}, 0^{D_{l+1}}; \eta)} - 1 \right| \leq C \frac{L}{|D|}$$

Case 1 If $Q < L$:

- First approximation:

$$\bar{W}^{(0)}_{N,\eta}(\alpha;\eta) = E_N[H_N(\sigma)|\alpha,\eta].$$

  Error: $\frac{1}{N} H\left(\nu_N^{(0)}(\cdot;\eta)|\mu_N(\cdot|\eta)\right) = O(\varepsilon^2)$

- Higher order corrections:

$$\bar{W}_{N,\mathcal{E}}(\alpha,\eta) - \bar{W}^{(0)}_{N,\mathcal{E}}(\alpha,\eta) = -\frac{1}{\beta} \log E_N[e^{-\beta(H_N(\sigma)-\bar{W}^{(0)}_{N,\mathcal{E}}(\alpha,\eta))}|\alpha,\eta].$$

  Error: $\frac{\beta}{N}\left(\bar{W}_{N,\mathcal{E}}(\alpha,\eta) - \sum_{l=0}^{p} \bar{W}^{(l)}_{N,\mathcal{E}}(\alpha,\eta)\right) = O(\varepsilon^{p+1})$.

  But not product!

- Higher order and product: increase the reconstruction domain

Case 2 If $Q > L$:

$$e^{-\beta \bar{W}_{N,\mathcal{E}}(\alpha,\eta)} = \int_{\{-1,1\}^{\Lambda\cap\mathcal{O}}} e^{-\beta H_N([\sigma^{\mathcal{O}},\alpha])} \prod_{k\in\bar{\Lambda}\cap\mathcal{O}} \tilde{\rho}_k(\sigma^{C_k})$$
$$= e^{-\beta \sum_{l\in\mathcal{E}} H_{l,l}(\alpha^{D_l},\alpha^{D_l})} \times$$

$$\times \prod_{l\in\mathcal{O}} \left[\int_{\{-1,1\}^{\Lambda\cap\mathcal{O}}} e^{-\beta H_{l,l}(\sigma^{D_l},\sigma^{D_l})} e^{-\beta H_{l-1,l}(\alpha^{D_{l-1}},\sigma^{D_l})} e^{-\beta H_{l,l+1}(\sigma^{D_l},\alpha^{D_{l+1}})} \bar{\rho}_l(\sigma^{D_l}) \right]$$

Let

$$[\ldots] = \frac{Z_l(0,\alpha^{D_{l+1}};\eta)\, Z_l(\alpha^{D_{l-1}},0;\eta)}{Z_l(0,0;\eta)} (f_{l-1,l+1}(\alpha)+1).$$

where

$$f_{l-1,l+1}(\alpha) = \frac{[\ldots]\, Z_l(0,0;\eta)}{Z_l(0,\alpha^{D_{l+1}};\eta)\, Z_l(\alpha^{D_{l-1}},0;\eta)} - 1$$

We obtain:

$$\prod_{l:\,\mathrm{even}} e^{-H_{l,l}(\alpha)} \prod_{l:\,\mathrm{even}} A_{l-1,l,l+1}(\eta) \prod_{l:\,\mathrm{odd}} B_l^{-1}(\eta)\times$$

$$\prod_{l:\,\mathrm{odd}} (1 + f_{l-1,l+1}(\alpha;\eta)) \prod_{l:\,\mathrm{even}} f_l^*(\alpha|_{D_l},\eta)$$

where

$$f_l^*(\alpha^{D_l},\eta) = \frac{1}{A_{l-1,l,l+1}(\eta)} Z_{l+1}(\alpha^{D_l},0;\eta)\, Z_{l-1}(0,\alpha^{D_l};\eta),$$

- First approximation:

$$\bar{W}^{(0)}_{N,\mathcal{E}}(\alpha,\eta) = \sum_{\substack{0\le l\le 2U-1\\ l \text{ even}}} H_{l,l}(\alpha^{D_l},\alpha^{D_l}) - \frac{1}{\beta}\sum_{\substack{0\le l\le 2U-1\\ l \text{ even}}} \log f^*_l(\alpha^{D_l}).$$

Error: $\frac{1}{N}H(\nu^{(0)}_N(\cdot;\eta)|\mu_N(\cdot|\eta)) = O(\frac{\delta}{N})$

- Higher order:

$$\beta\bar{W}_{N,\mathcal{E}}(\alpha,\eta) = \beta\bar{W}^{(0)}_{N,\mathcal{E}}(\alpha,\eta) - \sum_{\substack{0\le l\le 2U-1\\ l \text{ odd}}} \log(1+f_{l-1,l+1}(\alpha^{D_{l-1}},\alpha^{D_{l+1}})).$$