SAND2011-5102C

A Multiple-Timescale Simulation Method for the Morphological Evolution of Metal Interfaces

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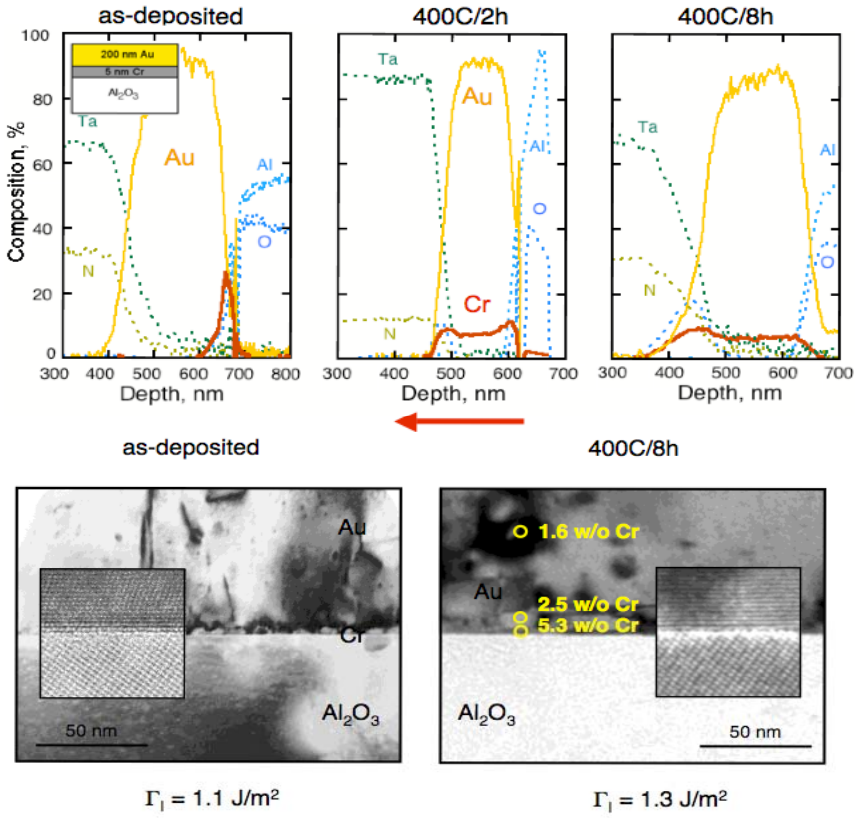
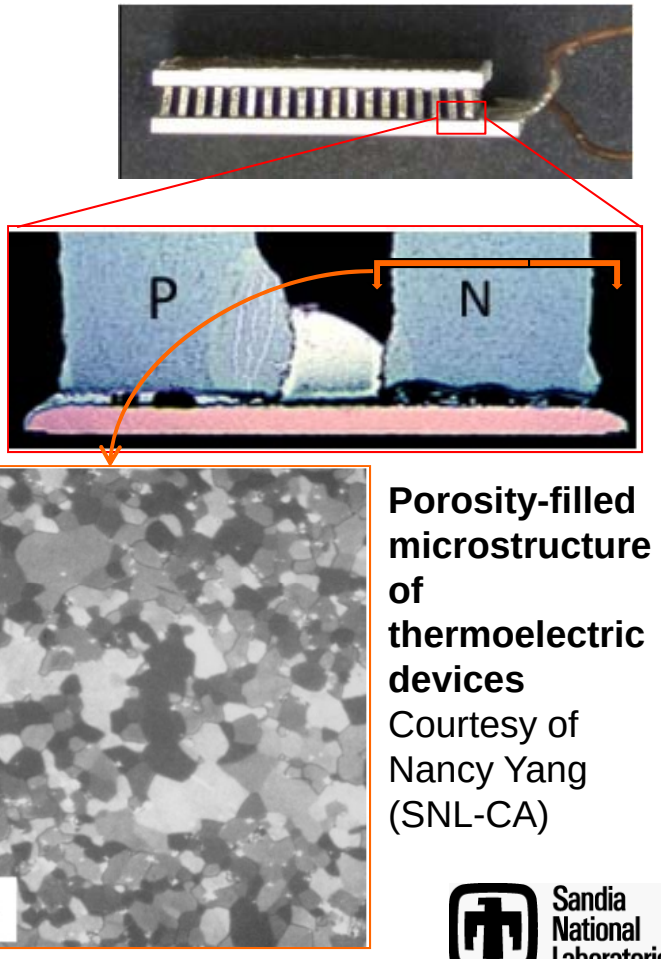
Long-Timescale Processes in Sandia Materials Applications

- **Synthesis and aging of materials driven by diffusion processes over long timescales**
  - **Neutron tubes, Semi-conductor electronics**
  - **Thermoelectrics, Fusion power systems**
  - **Radiation detectors**
  - **Gas and energy storage materials**



Aging of material interface in semi-conductor electronics
Courtesy of Neville Moody (SNL-CA)



Porosity-filled microstructure of thermoelectric devices
Courtesy of Nancy Yang (SNL-CA)

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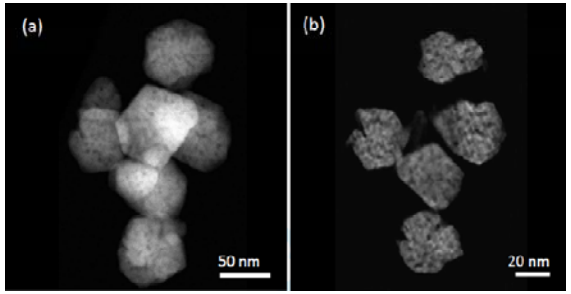
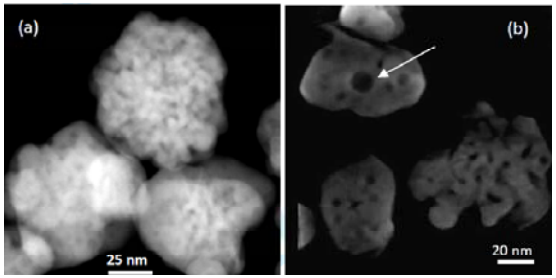
Long-Timescale Processes in Sandia Materials Applications

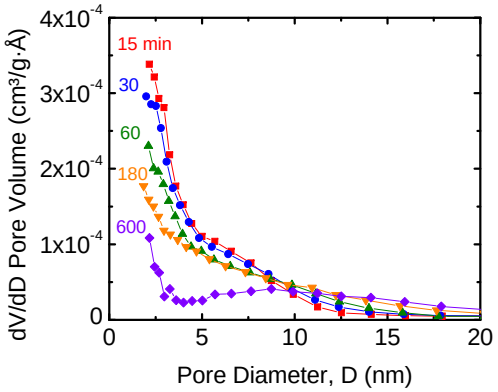
Unheated particles



After heating to 600 ° C



Aging of nanoporous Pd particles
Courtesy of Ilke Arslan (UC Davis), Markus Ong (Whitworth U) (both formerly SNL-CA)



- Current continuum models inadequate to describe many of these processes
  - Complex interactions, poorly understood physics, unknown parameters
- Timescales are too long to be reached by most atomistic simulation techniques
  - Molecular dynamics limited by atomic vibration timescale (picoseconds)
  - Diffusion processes may be taking place over hours, or even years

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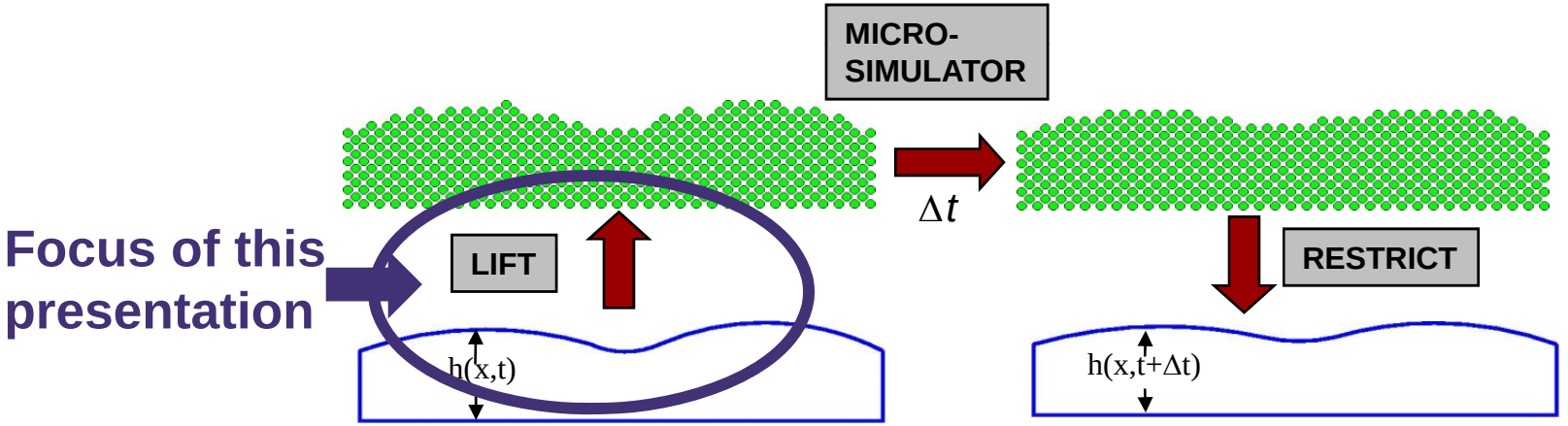
Extending to Longer Timescales: Equation-Free Projective Integration Method

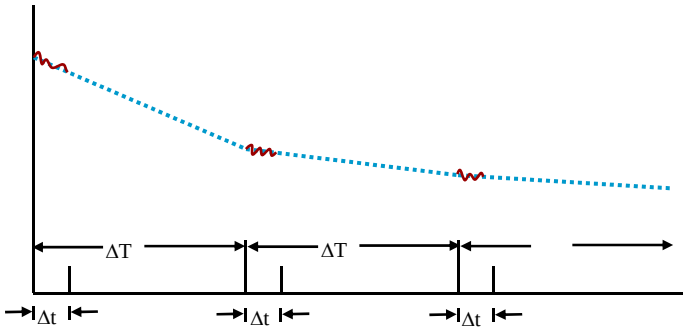
- Approach:
  - Use fine-scale simulations as “computational experiments” to get approximate coarse-scale time derivatives
  - Project coarse scales in time over longer timestep, and reinitialize fine scales
  - I. Kevrekidis et al., *Comm. Math Sci*. 1 (2003) and related papers



Focus of this presentation

Approximate: $\frac{\partial h}{\partial t} \approx \frac{1}{\Delta t}\left(h\left(t+\Delta t\right)-h\left(t\right)\right)$

Project: $h\left(t+\Delta T\right)=h\left(t\right)+\Delta T\,\frac{\partial h}{\partial t}$



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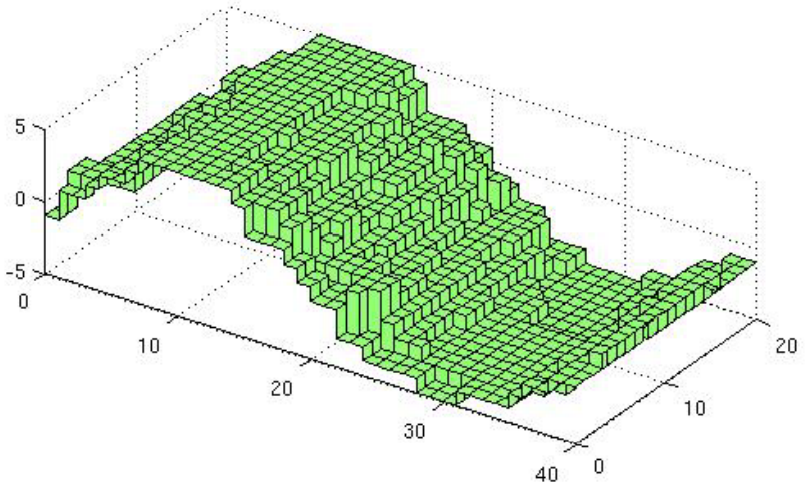
Test Application: Solid-on-Solid Model

- The Solid-on-Solid (SOS) model is a simple surface representation that mimics the physics of a real crystal surface
  - Surface represented as array of colum with integer heights
  - Energy based on nearest neighbor interactions:

$$E(\mathbf{h}) = \frac{1}{2} J \sum_{i=1}^{N} \sum_{j=1}^{neigh(i)} \left| h_i - h_j \right|$$

    - Flat surface has minimum energy



- Kinetic Monte Carlo simulations used to obtain fine-scale dynamics
  - Each “move” is a height exchange between neighboring sites
  - Implemented in SPPARKS KMC code (www.cs.sandia.gov/~sjplimp/spparks.html)

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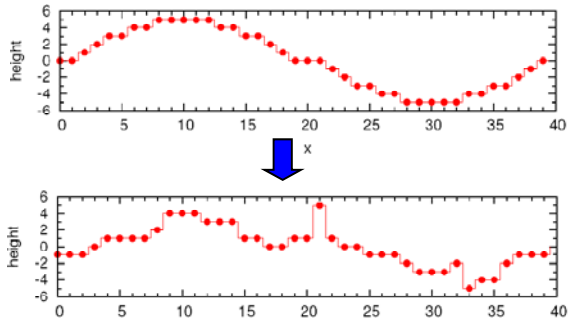
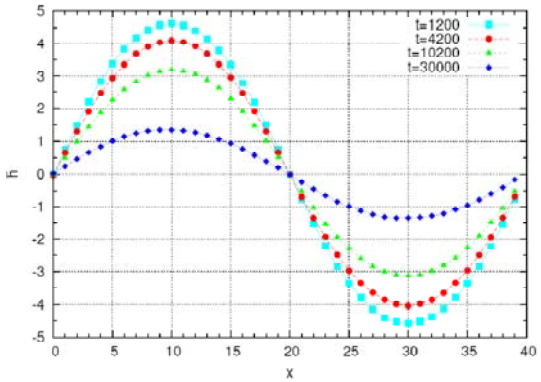
Early Results: 2D Test Problem

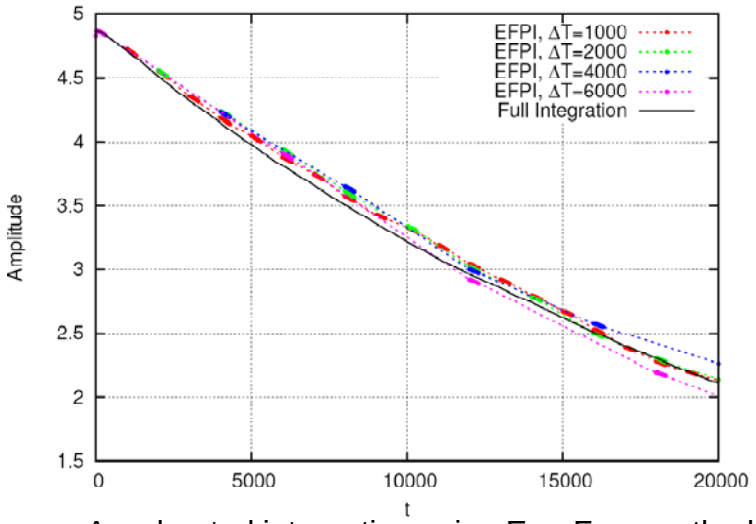
- Demonstrated acceleration factor of 20x on 2D problem
  - Goal is to predict height decay of ensemble average over many realizations (order 10^3)



A single realization of the SOS surface model



Evolution of ensemble-averaged height profile



Accelerated integration using Eqn-Free method

Findings:

- **Very important to choose correct coarse scale parameterization**
  - **Need to preserve higher order statistics (2-point spatial correlations), not just average heights, to get correct dynamics**
- **Fast variables must be identified and slaved to slow variables to preserve stability**
- **Lift operators used here are difficult to extend to the full 3D case**
- **Wagner et al., *Int. J. for Multiscale Computational Eng.* 8:423 (2010)**

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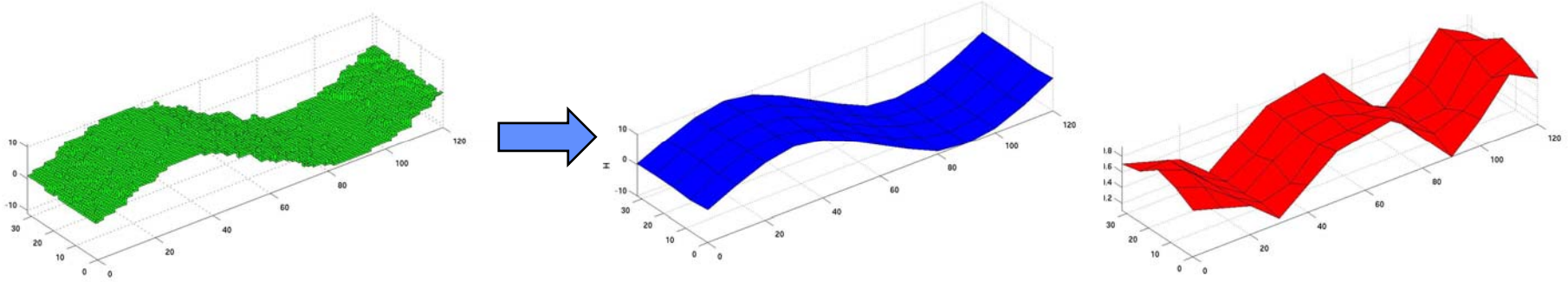
Simulated Annealing

- **Simulated Annealing**: a stochastic method (similar to Metropolis Monte Carlo) to drive a prescribed error function to zero
  - Our S.A. procedure for solid surfaces:
    - Choose a set of coarse scale functions whose ensemble-averaged values are known
      - Averages are smooth, spatially varying fields defined at nodal positions
    - For a given fine-scale configuration (surface heights at all sites), project onto smooth field spanned by cubic splines at nodes



Fine Scale Heights

Projected Heights

Projected Energy

- Run simulated annealing to minimize error defined by:

$$\text{Error} = \alpha_H \underbrace{\left| H_{actual} - H_{goal} \right|^2}_{\text{height error}} + \alpha_E \underbrace{\left| E_{actual} - E_{goal} \right|^2}_{\text{energy error}} + \alpha_G \underbrace{\left| G_{actual} - G_{goal} \right|^2}_{\text{variance error}} + \cdots$$

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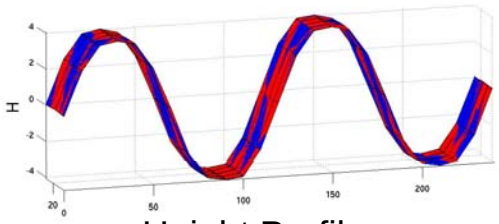
Difficulties with Simulated Annealing

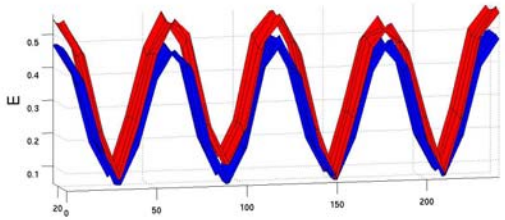
- Simulated Annealing has some problems for our application:

1. Penalty parameters must be chosen carefully to simultaneously fit all goal functions (not always possible)

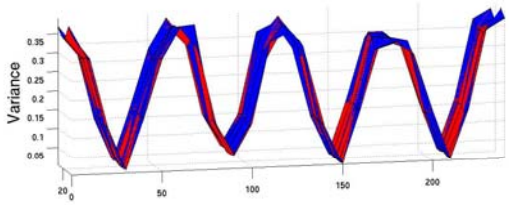
Coarse Variables: Goal vs. Actual



Height Profile

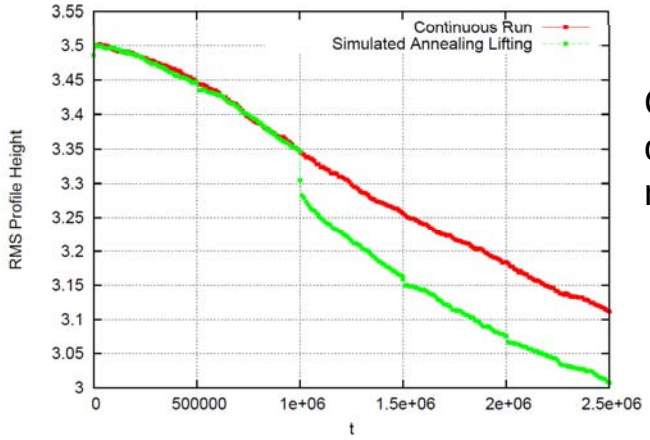


Energy Profile



Variance Profile

2. SA enforces goals on ***every*** realization, while really we want to enforce only the ensemble average
3. Most important: SA leads to realizations that do not preserve the dynamics of the original system



Comparison of “full” KMC (no restrict/lift operation) vs. run with simulated annealing restrict/lift applied every 200K timesteps

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Maximum Entropy Method

- Q: Instead of trying to specify higher order statistics (like correlation functions), can we find a way to constrain only the coarse scale values we care about, and allow other modes to come into equilibrium with them?
  - To run to equilbrium (e.g. in a Monte Carlo simulation), we need to know the probability distribution function (PDF) for the system
  - The theory of ***maximum entropy*** states that the relevant PDF is the one that maximizes the entropy of the system while still satisfying the known statistics (E.T. Jaynes, *Phys Rev.* **106**:620**,** 1957)
    - Entropy is given by:

$$H\left(P\left(\mathbf{h}_1\right), P\left(\mathbf{h}_2\right), \ldots\right) = -k \sum_i P\left(\mathbf{h}_i\right) \ln P\left(\mathbf{h}_i\right)$$

where sum is over all possible height configurations **h**

  - For example, if only the average energy of the system is known, the maximum entropy theory leads to the standard canonical ensemble distribution:

$$P\left(\mathbf{h}\right) = C \exp\left\{-\beta E\left(\mathbf{h}\right)\right\}$$

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Maximum Entropy for Solid Surfaces

- Goal: constrain the ensemble averages of some set of profile parameters A_i (e.g. Fourier modes), along with their variances σ^2_{Ai}, to prescribed values
  - Maximum entropy theory leads to PDF of the form:

$$P(\mathbf{h}) = C \exp\left\{-\beta E(\mathbf{h})\right\} \exp\left\{-\sum_i \beta_i A_i(\mathbf{h})\right\} \exp\left\{-\sum_i \alpha_i \left(A_i(\mathbf{h}) - A_i^{goal}\right)^2\right\}$$

    where α_i and β_i are Lagrange multipliers that must be determined by enforcing desired ensemble averages:

$$A_i^{goal} = \sum_{\mathbf{h}} A_i(\mathbf{h}) P(\mathbf{h}) = \langle A_i \rangle$$

$$\sigma_i^{goal} = \left(\sum_{\mathbf{h}} \left(A_i - \langle A_i \rangle\right)^2 \right)^{1/2}$$

  - Lagrange multipliers can be approximated or solved iteratively
  - Note that unlike Simulated Annealing, the maximum entropy method enforces averages over the ensemble of realizations, not individual realizations

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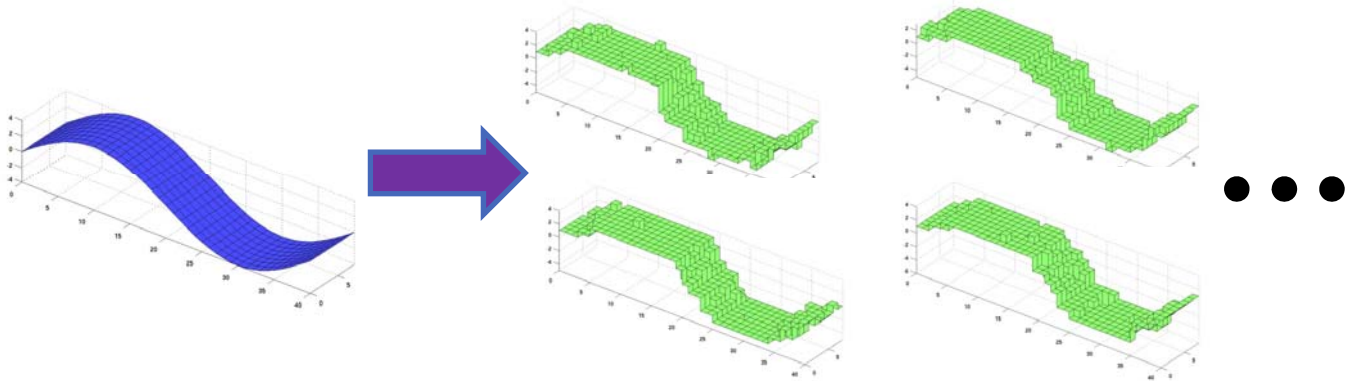
Lift Operator using Maximum Entropy

- Algorithm for lifting with Maximum Entropy method:
  - **Given:** set of coarse scale profile shape parameters with known ensemble average and std. dev.
    - E.g. set of leading order Fourier coefficients A_i
  - **Initialize** a set of heights based on the mean values, $\langle A_i \rangle$
  - **Sample**, using a Metropolis Monte Carlo sampling method, a set of realizations with the probability distribution:

$$P(\mathbf{h}) = C \exp\left\{-E(\mathbf{h})/k_B T\right\} \exp\left\{-\sum_i \beta_i A_i(\mathbf{h})\right\} \exp\left\{-\sum_i \alpha_i \left(A_i(\mathbf{h}) - A_i^{goal}\right)^2\right\}$$

    - Compute the resulting mean and standard deviation for each A_i
  - **Iterate**, if necessary, to adjust the Lagrange multipliers α_i and β_i until the desired statistics are obtained
- The resulting set of realizations obeys the desired statistics, but is otherwise in equilibrium according to the maximum entropy principle



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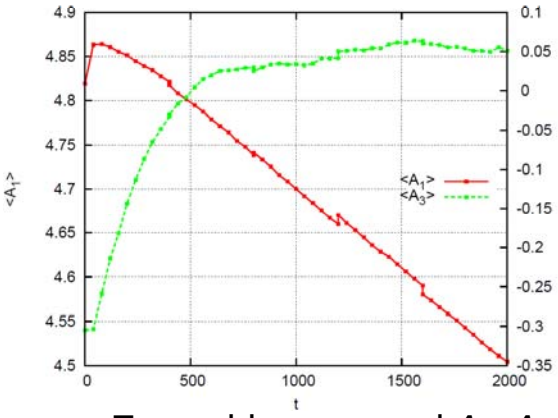
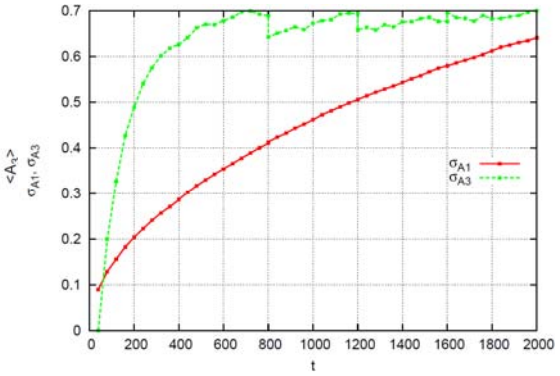
Maximum Entropy Results: 2D

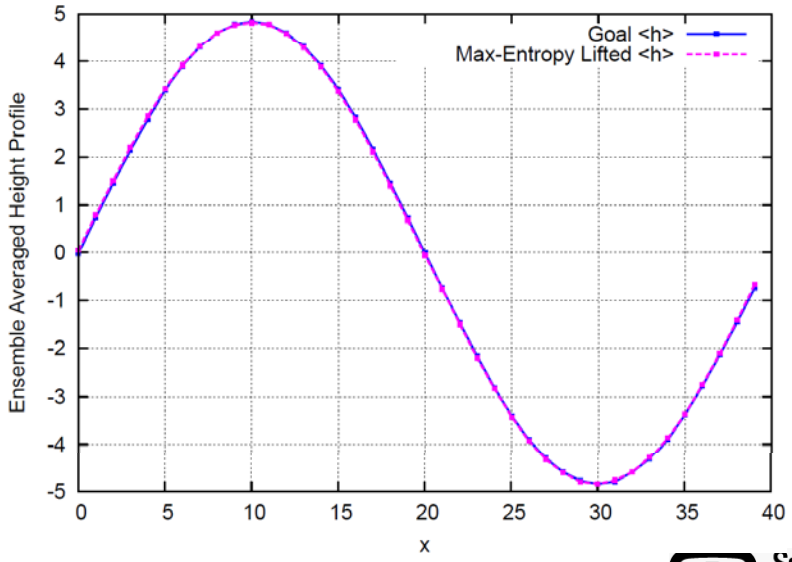
- Test our lift operator by reapplying the restrict/lift at fixed intervals during a KMC simulation
  - Operator should disturb the profile and dynamics as little as possible
  - Constrain first and third Fourier sine coefficients, A_1 and A_3, and their std. devs.
  - Use system size L=40, apply lift every 400 time units



Ensemble-averaged A_1, A_3 vs. time



Std. dev.'s of A_1, A_3 vs. time



Average height profile, before and after lift, at t=400

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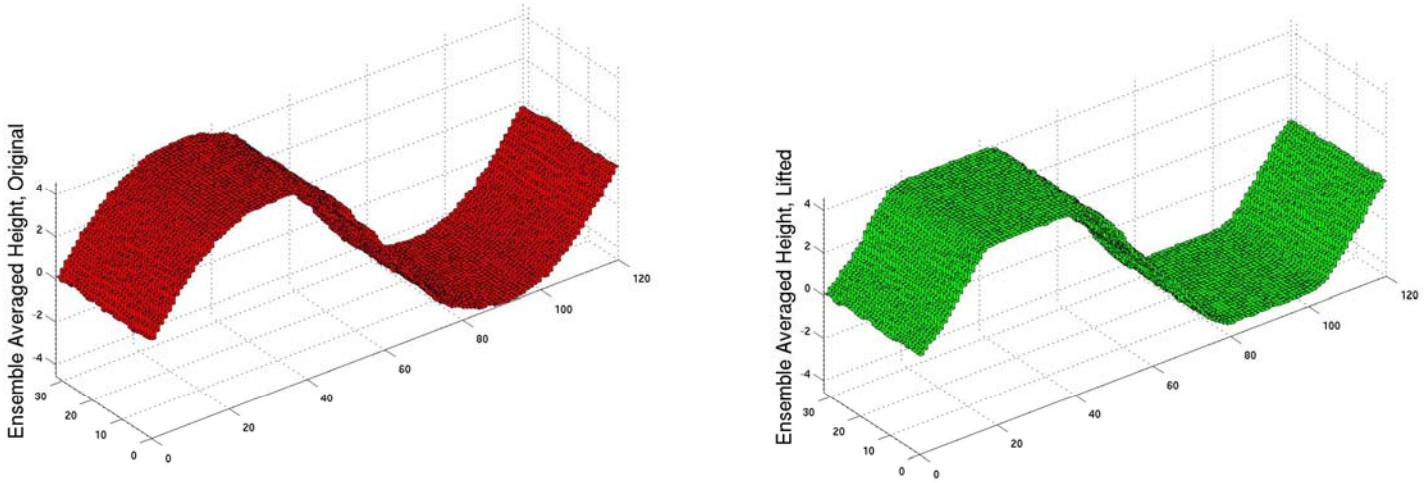
Maximum Entropy Results: 3D

- **Test**: Begin with an ensemble-averaged profile from a “real” surface simulation (full single-scale KMC) and try to reproduce it’s shape and dynamics using Maximum Entropy method
  - Constrain first 2 odd-numbered Fourier sine coefficients (A_1 and A_3)



- Shape and dynamics are not well preserved for this case
  - Better solution may be needed for Lagrange multipliers
  - But the technique succeeded in generating a low-energy average configuration!
    - Constraining more coefficients (higher order modes) may help
  - Computational expense in 3D becomes a limitation (large systems, slow equilibration times)

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Conclusions

- Two methods have been explored for a “lift” operation in the context of a multiple timescale, equation-free projection method
  - Both methods attempt to take given coarse-scale descriptions and use them to initialize an ensemble of fine scale system realizations
  - **Simulated Annealing**:
    - + Reasonable efficiency
    - + Very flexible in choice of constraint variables
    - ✓ Constraint of multiple variables must be balanced through penalty parameters
    - ✓ Constrains each realization individually rather than the entire ensemble
    - ✓ System dynamics is not well reproduced
  - **Maximum entropy method**:
    - + Constrains ensemble, not individual realizations
    - + Requires smallest amount of coarse scale information compared with similar methods
    - + Guarantees equilibrium in the max-entropy sense
    - + Reproduces dynamics well (when initialization to constraint is successful)
    - ✓ Computationally expensive
    - ✓ Requires specification of Lagrange multipliers in PDF – a difficult problem in general
    - ✓ Can lead to undesired, lower-energy configurations if under-constrained
- The Maximum Entropy method can be successful **if**:
  - Computational time to iteratively solve for Lagrange multipliers can be reduced
  - A sufficient set of coarse variables can be identified (e.g. through dimensionality reduction techniques)