SAND2020-4188C

MESOSCALE MULTIPHYSICS PHASE FIELD SIMULATOR (MEMPHIS)

RECENT PROGRESSES IN PHASE-FIELD MODELING AT SANDIA NATIONAL LABORATORIES

PRESENTED BY

RÉMI DINGREVILLE

CHIMAD PHASE-FIELD WORKSHOP X: VIRTUAL

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A Team Effort

- **Graduate students**
  M. Powers (PhD. MSE, U Mich.); E. Herman (Ph.D. Applied Math, NC State); D. Visozo (ME, GT)

- **Postdocs**
  J. Stewart, E.Y. Chen, D. Montes de Oca Zapiain, T. O'Connor

- **External collaborators**
  A. Misra (U. Mich.), K. Baldwin (LANL), N. Li (LANL), J. Singer (Rutgers)

- **Funding**

WHY MEMPHIS?

PHYLOSOPHY

1. **Not a Computer Science Project…**
   A Materials Science Project
   No need to worry about numerical methods, parallelization, etc…
2. **Modularity**
   Being able to re-utilize/combine models
   Rapid prototyping of models
3. **Easy to code**
   Accessible to users with various backgrounds
4. **Open source**

CURRENT FOCUS

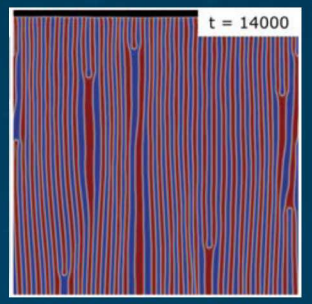
1. **Synthesis of nanostructured materials**
   Physical Vapor Deposition of thin films
   Molecular Beam Epitaxy for quantum dots
2. **Aging and degradation of materials**
   Irradiation-induced segregation
   Localized corrosion

Roadmap For Today's Talk

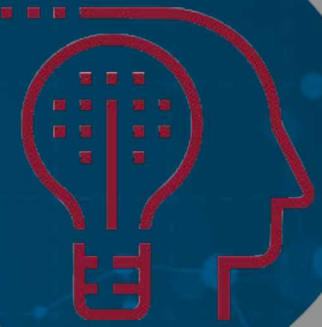
1. Code architecture

2. Verification & Validation

3. Application: PVD

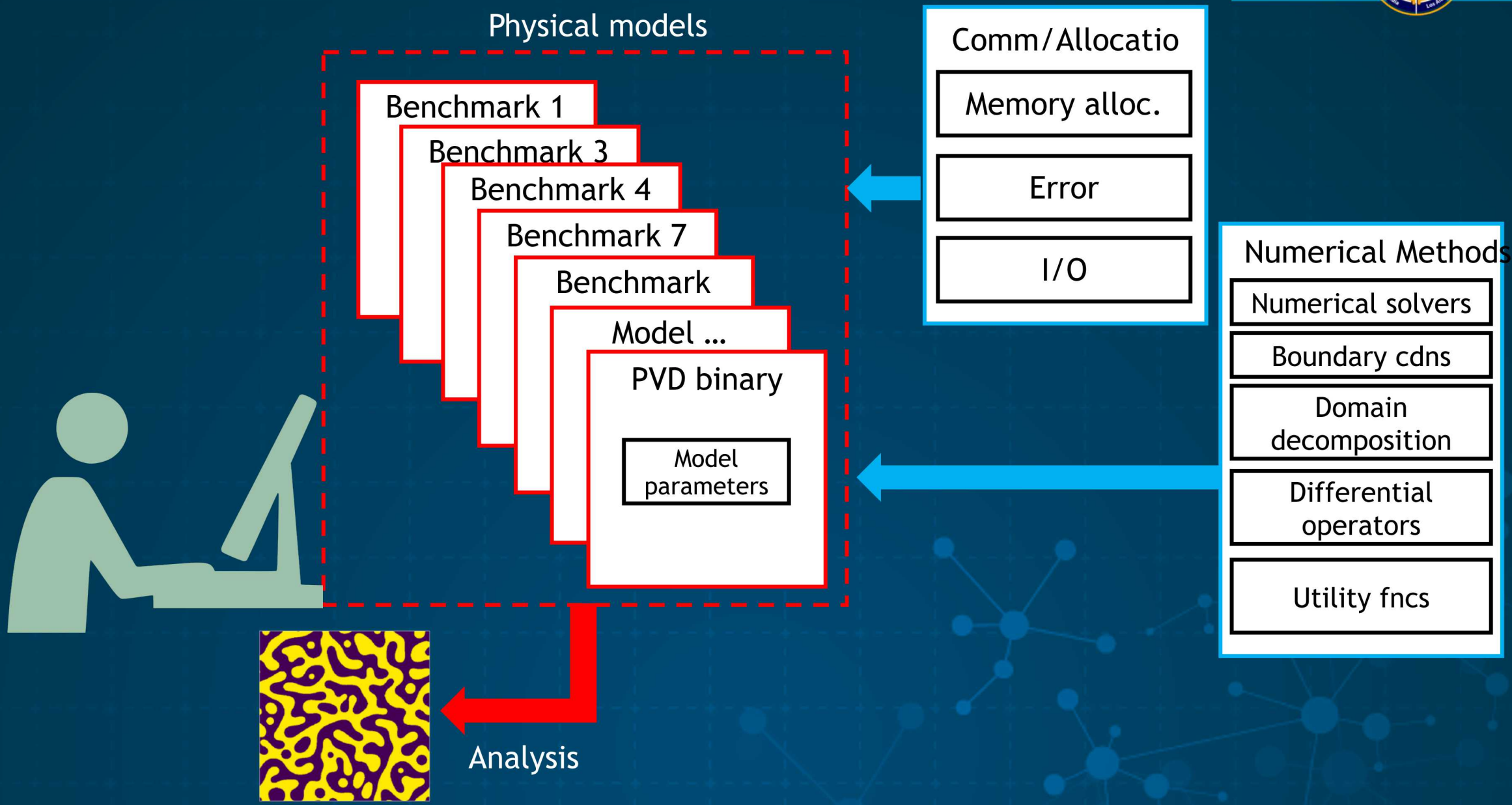


4. Integrating ML

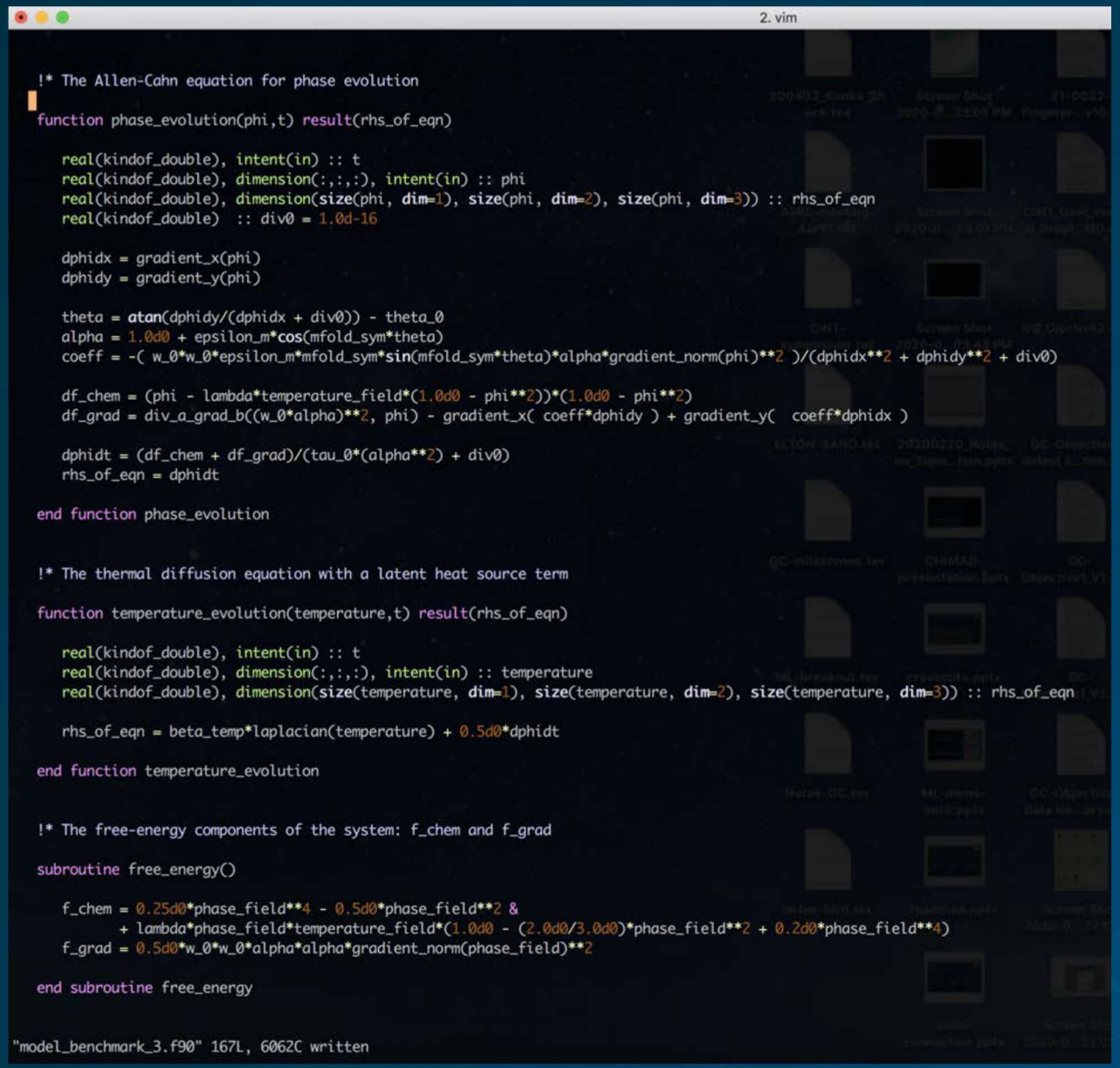


5. Moving forward

Code Architecture



Code Architecture



1. MEMPHIS is written in Fortran 90
2. Serial or parallel (using message-passing interface and spatial decomposition of comput. domain)
3. Writing a model is straight forward
4. Set boundary cdns in a variable way (Neuman /Dirichlet)
5. Various numerical integration schemes are available:
   explicit Euler, midpoint, Heun's, Gauss-Seidel
6. Output results in various formats
7. Can read an input microstructure

Verification & Validation

1. Spinodal Decomposition

Diffusion of a solute in a matrix

Total Uploads: 54

Specification

Results: 1a | 1b | 1c | 1d

2. Ostwald Ripening

Coupling of conserved and non-conserved dynamics

Total Uploads: 25

Specification

Results: 2a | 2b | 2c | 2d

3. Dendritic Growth

Solidification and dendritic growth in a single-component

Total Uploads: 14

Specification

Results: 3a

4. Elastic Precipitate

Linear elasticity via evolution of a constrained precipitate

Total Uploads: 12

Specification

Results: 4a | 4b | 4c | 4d | 4e | 4f | 4g | 4h

5. Stokes Flow

Incompressible Stokes flow in 2D

Total Uploads: 0

Specification

Results: 5a | 5b

6. Electrostatics

Coupled electrostatics and Cahn-Hilliard dynamics

Total Uploads: 8

Specification

Results: 6a | 6b

7. MMS Allen-Cahn

Use the Method of Manufactured Solutions to verify codes

Total Uploads: 9

Specification

Results: 7a | 7b | 7c

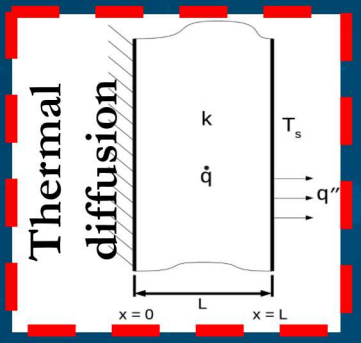
8. Nucleation

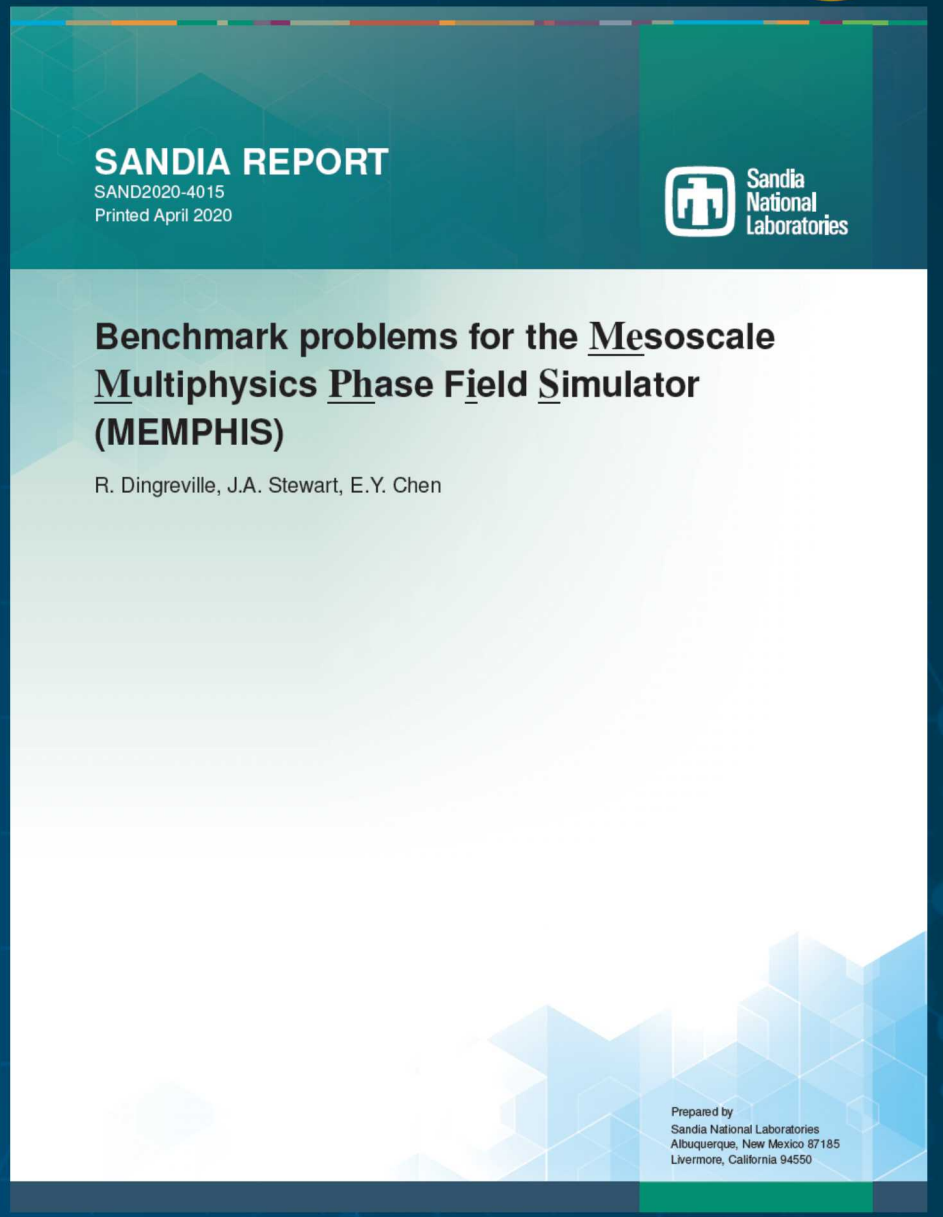
Nucleation benchmark

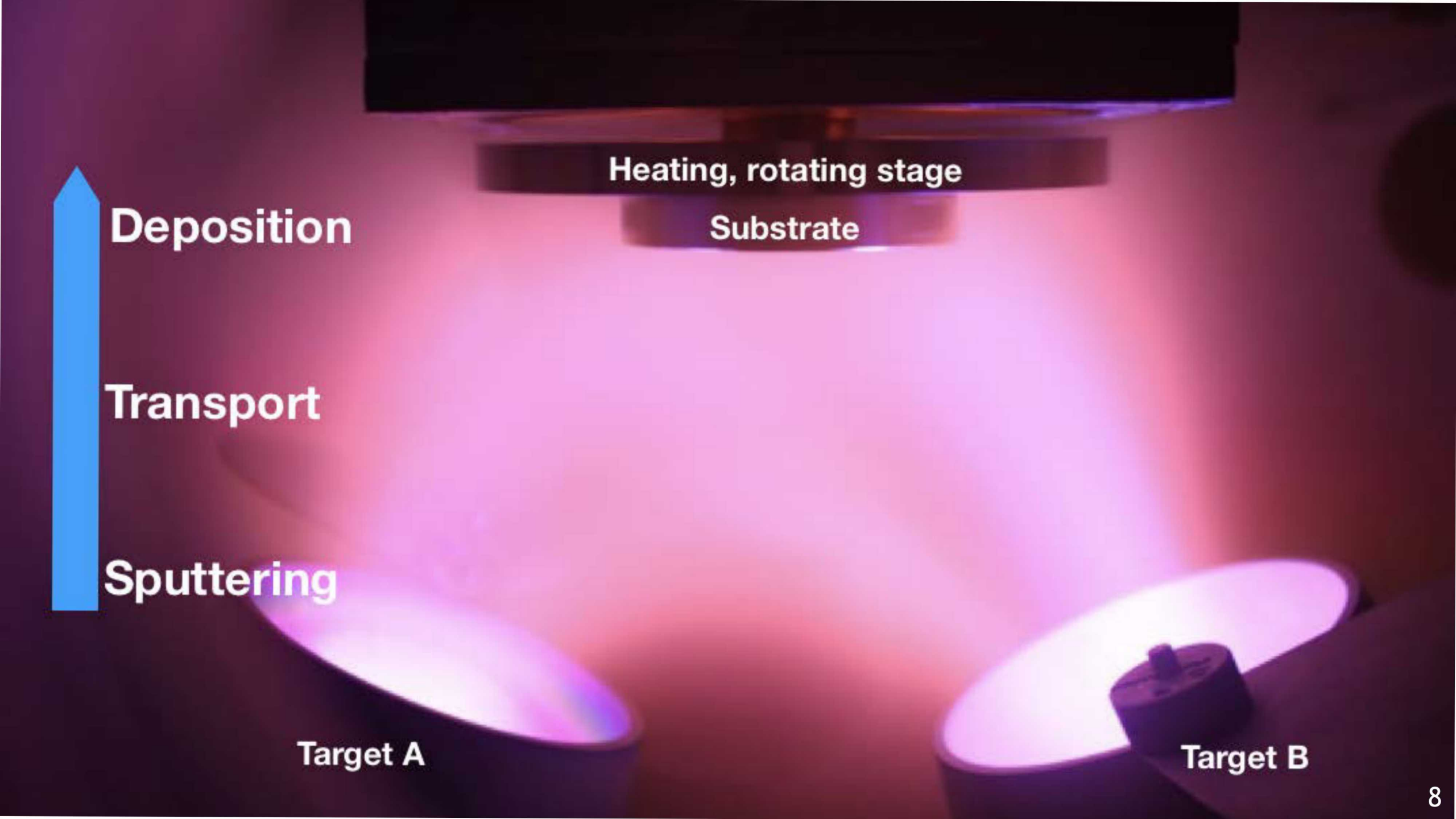
Total Uploads: 4

Specification

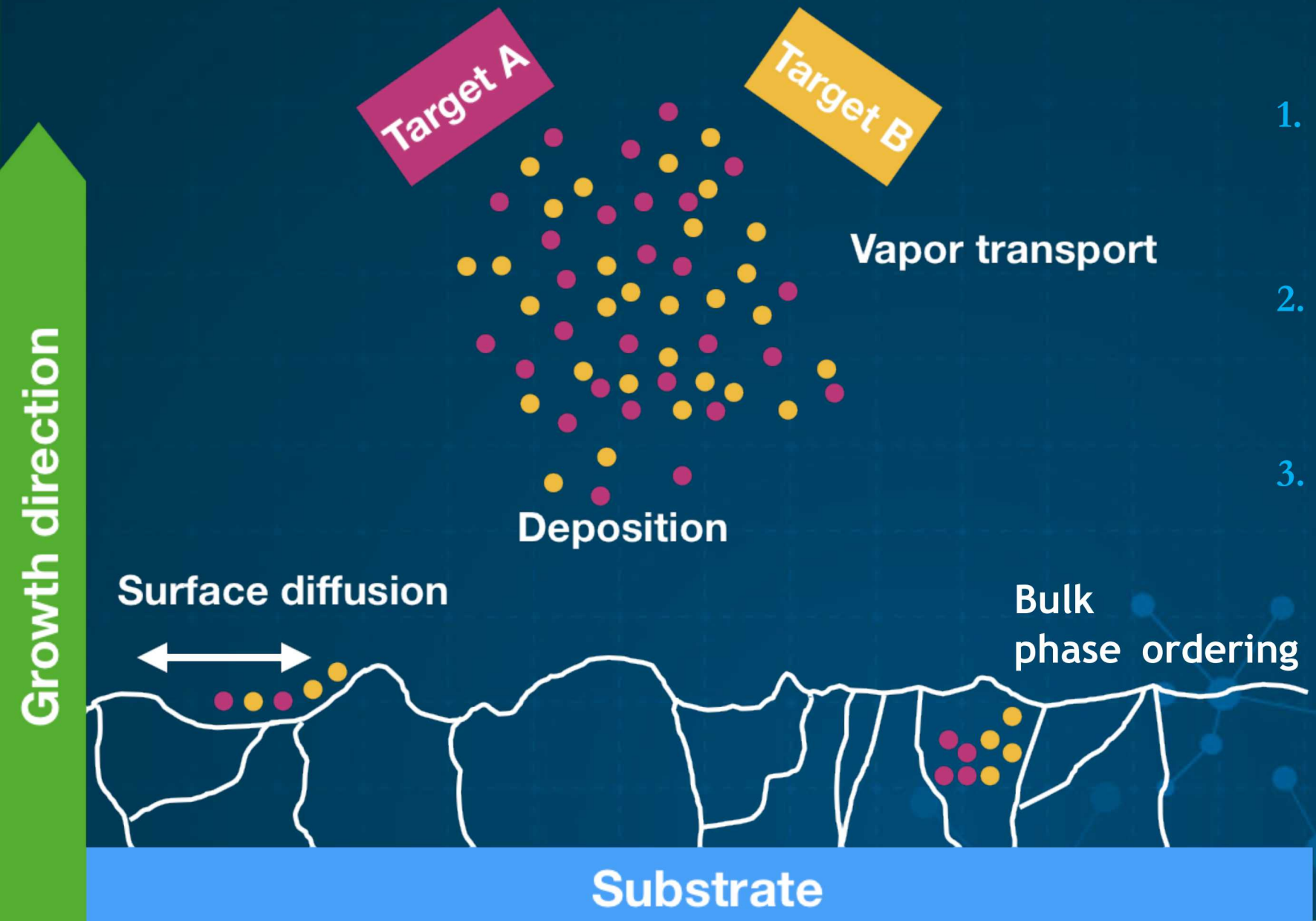
Results: 8a | 8b | 8c | 8d






Heating, rotating stage
Substrate
Deposition
Transport
Sputtering
Target A
Target B

Modeling PVD Using Phase-Field



1. **Explicitly model vapor transport**
   …so we can model multiple PVD processes
2. **No artificial deposition of vapor**
   …so we can capture hillock formation
3. **Account for surface and bulk diffusion into one model**
   …so we can account for the composition of surface vs. bulk phenomena

Modeling PVD Using Phase-Field

$$F = \int \left[\left[f_\phi + \frac{\kappa_\phi^2}{2} (\nabla \phi)^2 \right] + s(\phi) \left(f_{el} + f_c + \frac{\kappa_c^2}{2} (\nabla c)^2 \right) \right] d\Omega$$

Describing vapor/solid field

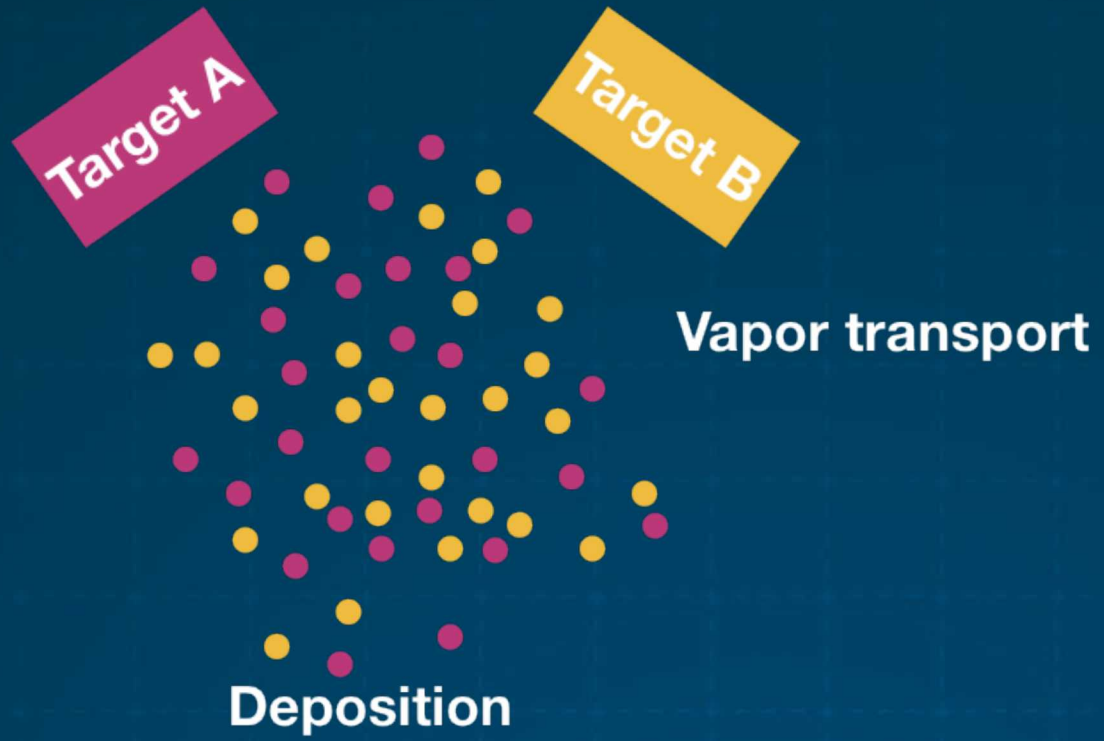
Only considered within the solid thin film

Describes concentration field

Describes two-way coupling between elastic and concentration fields (Vegard)

Substrate

Transport & Evolution of Incident Vapor



$$\frac{\partial \rho}{\partial t} = \nabla \cdot (D(\rho)\nabla\rho) - \nabla \cdot (\rho\vec{u}) - S(\vec{n})$$

Convection-diffusion

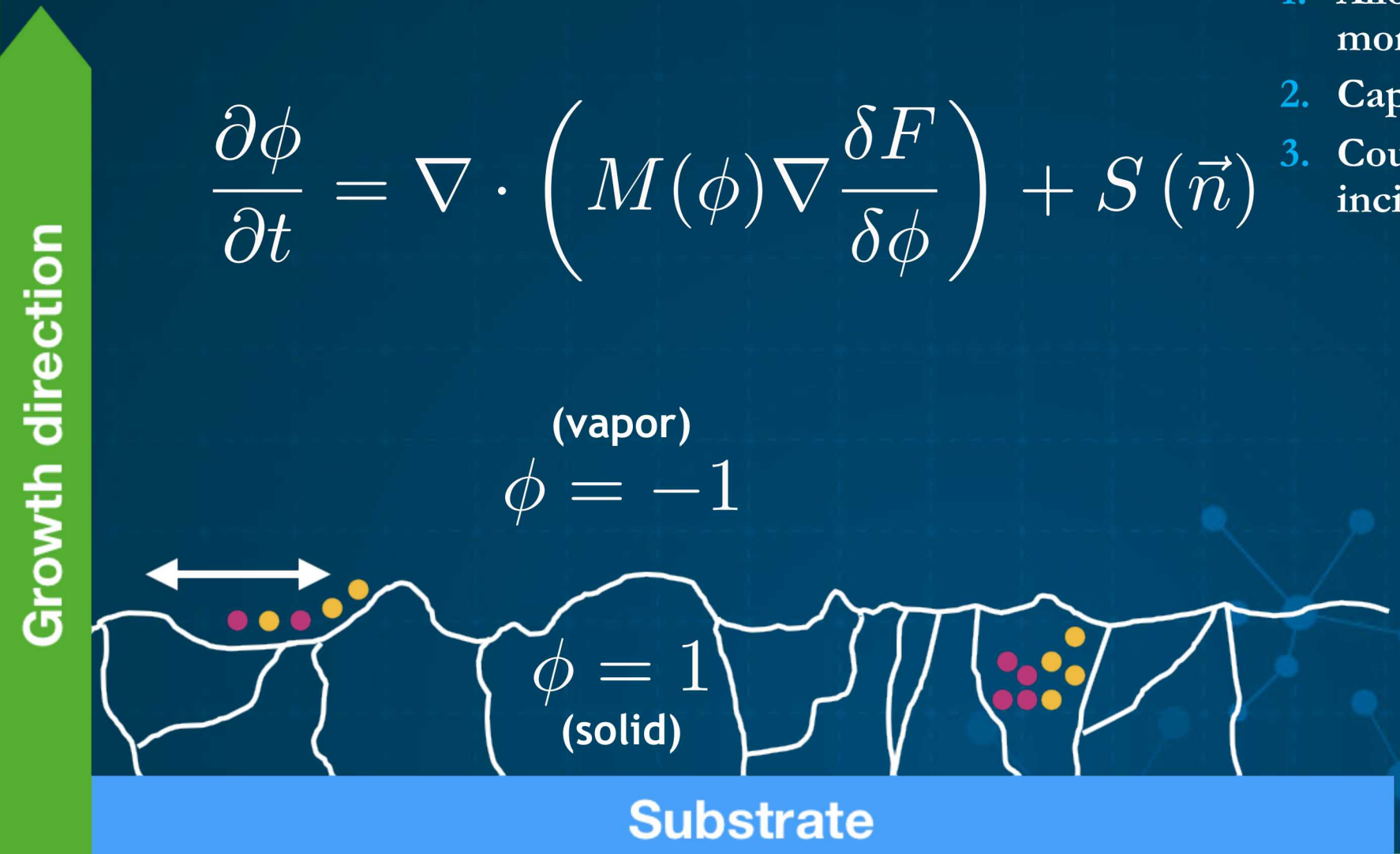
1. Local vapor pressure
2. Sputtering power (velocity)
3. Vapor density and velocity can be adapted to simulated various PVD processes
4. Random distribution of vapor phase initially
5. Source term to allow for removal of vapor that is being converted to solid at thin-film interface
6. Inclusion of LB capability in the future to include hydrodynamic effects (ALD, CVD)

Substrate

Kinetics of Thin-Film Growth

$$\frac{\partial \phi}{\partial t} = \nabla \cdot \left(M(\phi) \nabla \frac{\delta F}{\delta \phi} \right) + S(\vec{n})$$

1. Allows for arbitrary surface morphology formation
2. Captures surface-diffusion effects
3. Coupling of thin-film evolution to incident-vapor flux

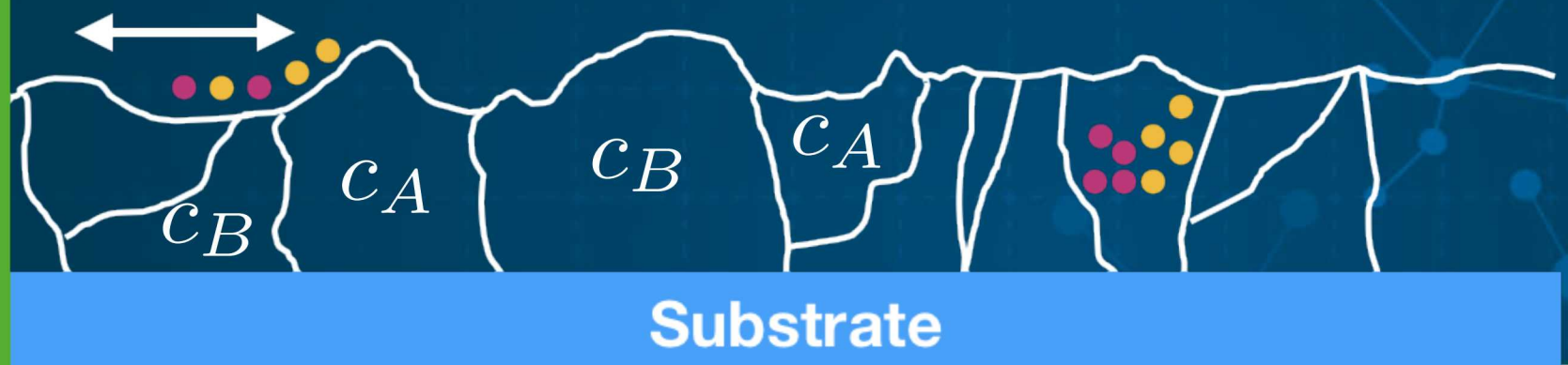


Phase Ordering in Thin-Film

$$\frac{\partial c}{\partial t} = \nabla \cdot \left(M(\phi, c) \nabla \frac{\delta F}{\delta c} \right)$$

$$M_c(\phi, c) = M^{\text{Bulk}} + M^{\text{Surf}}$$

1. Bulk or surface mobilities are selected through a **ϕ**-dependent switching function
2. Ensure that surface mobility is localized to vapor-solid interface
3. Explicitly capture both surface and bulk diffusion



Elastic Fields in the Film

$$f_{el} = (\epsilon - \epsilon^c) : \mathbb{C} : (\epsilon - \epsilon^c)$$

$$\nabla(\mathbb{C} : (\epsilon - \epsilon^c)) = 0$$

$$\epsilon^c = (c - c^*)\eta$$

1. Elastic stiffness tensor is compositionally dependent
2. Formation and evolution of phase domains give rise to compositional misfit: Thank you Vegard!
3. Two-way chemo-mechanical coupling

Modeling PVD Using Phase-Field

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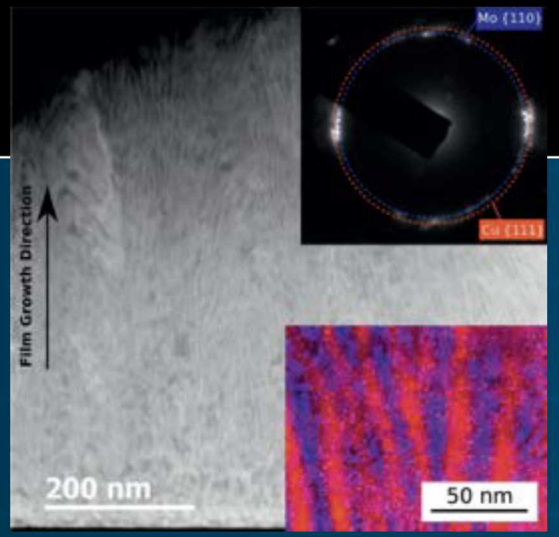
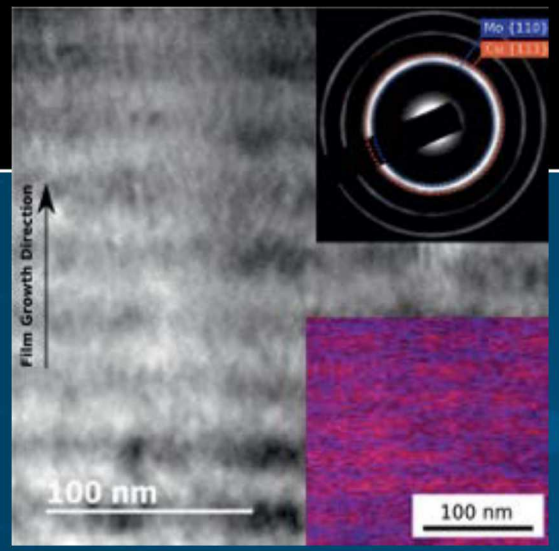
Target A | Target B

Lateral



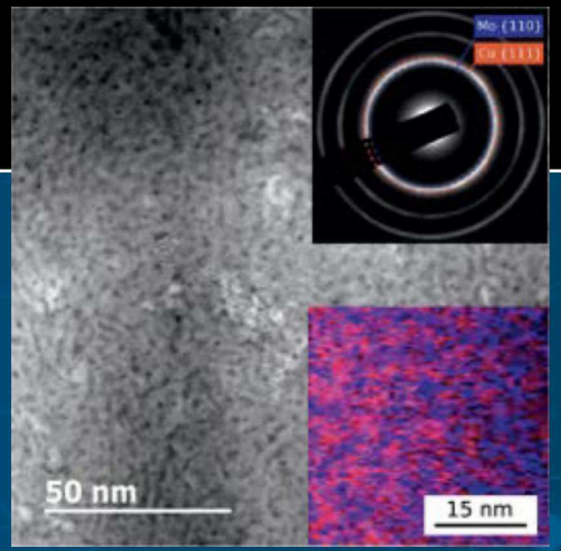
Target A | Target B

Vertical



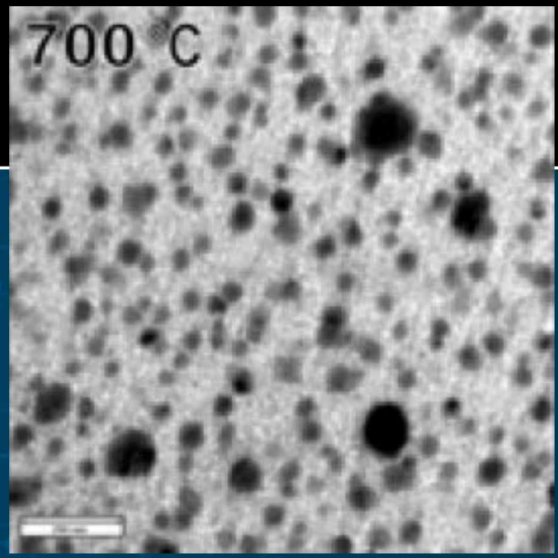
Target A | Target B

Random

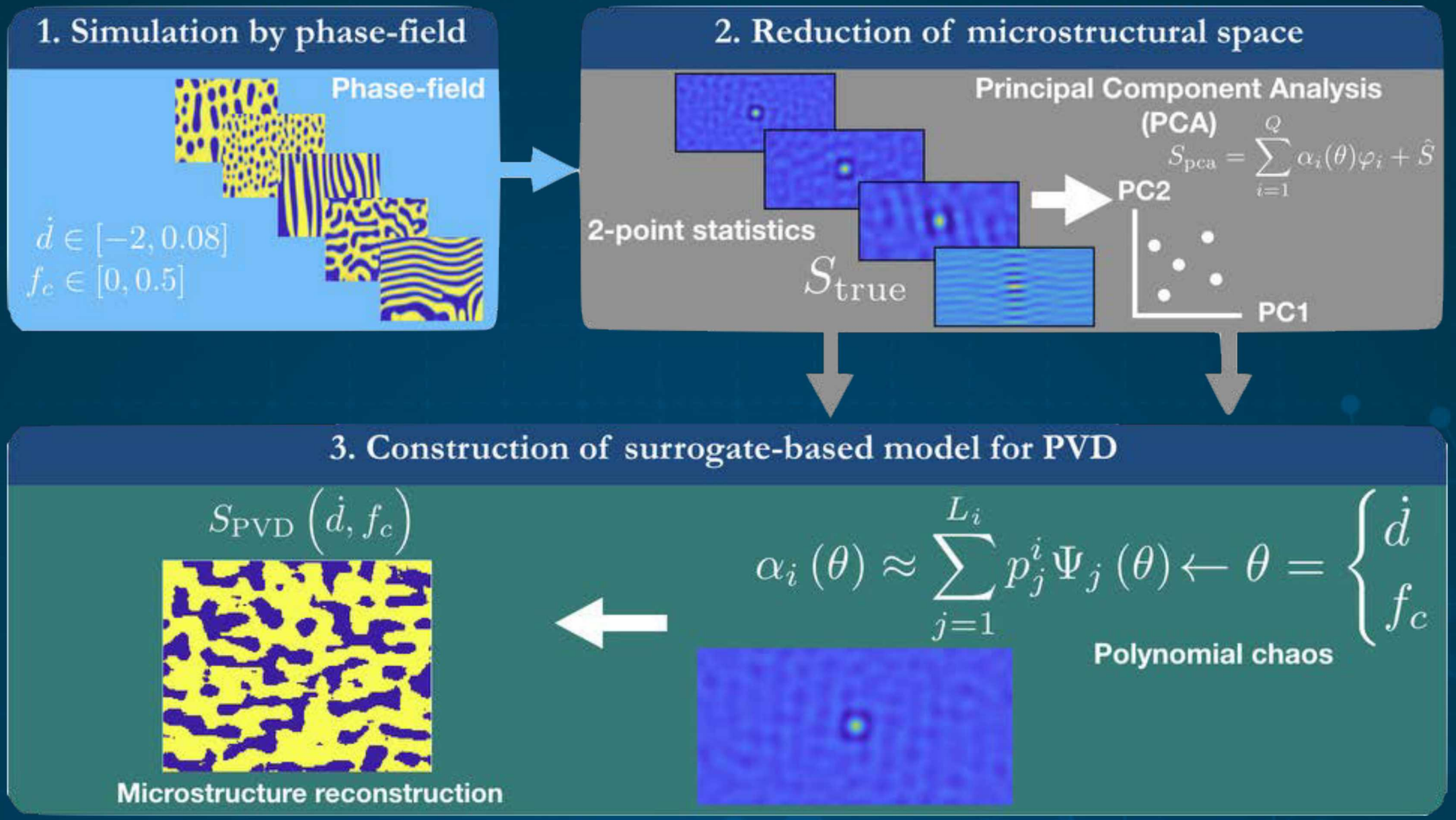


Target A | Target B

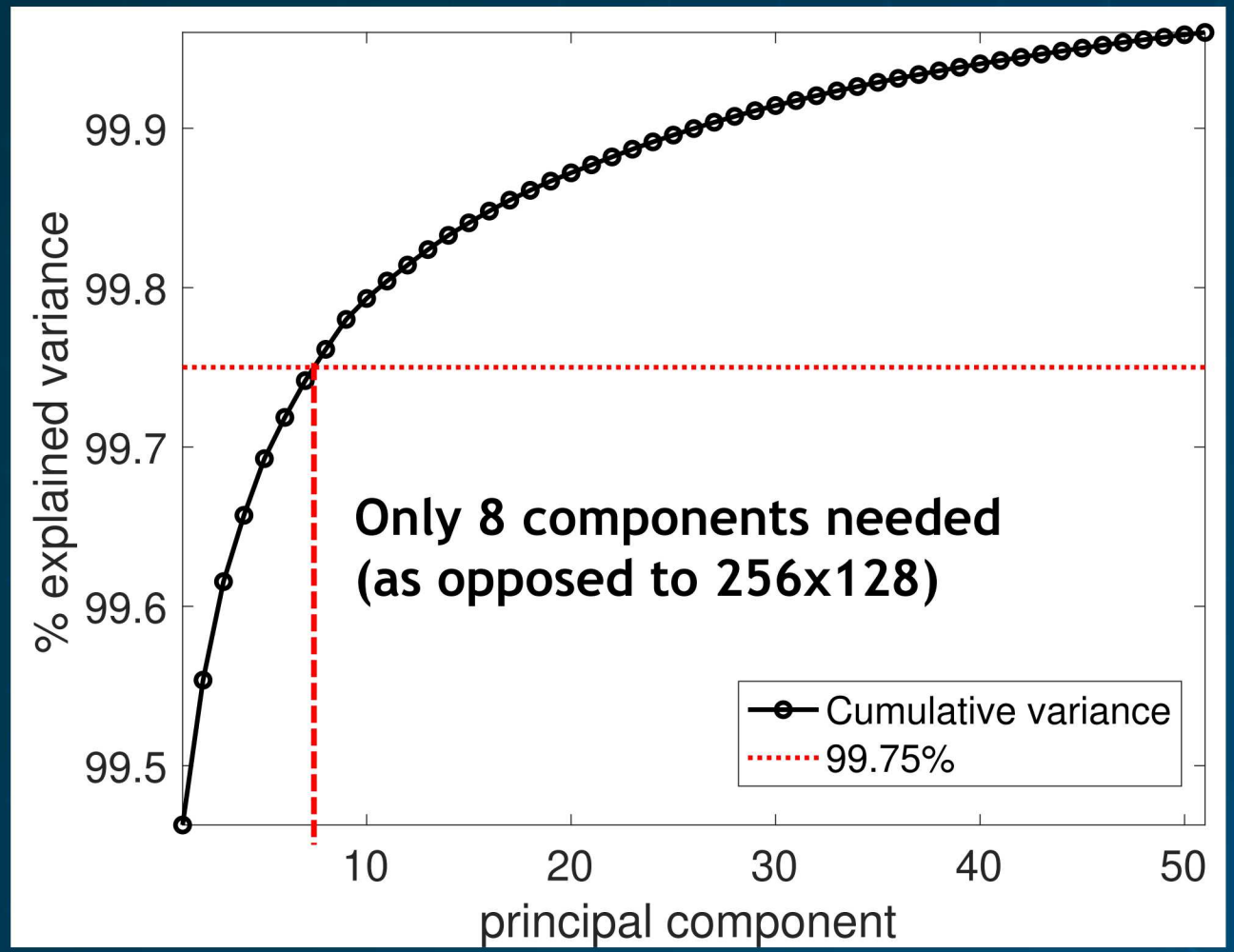
Nanoprecipitate

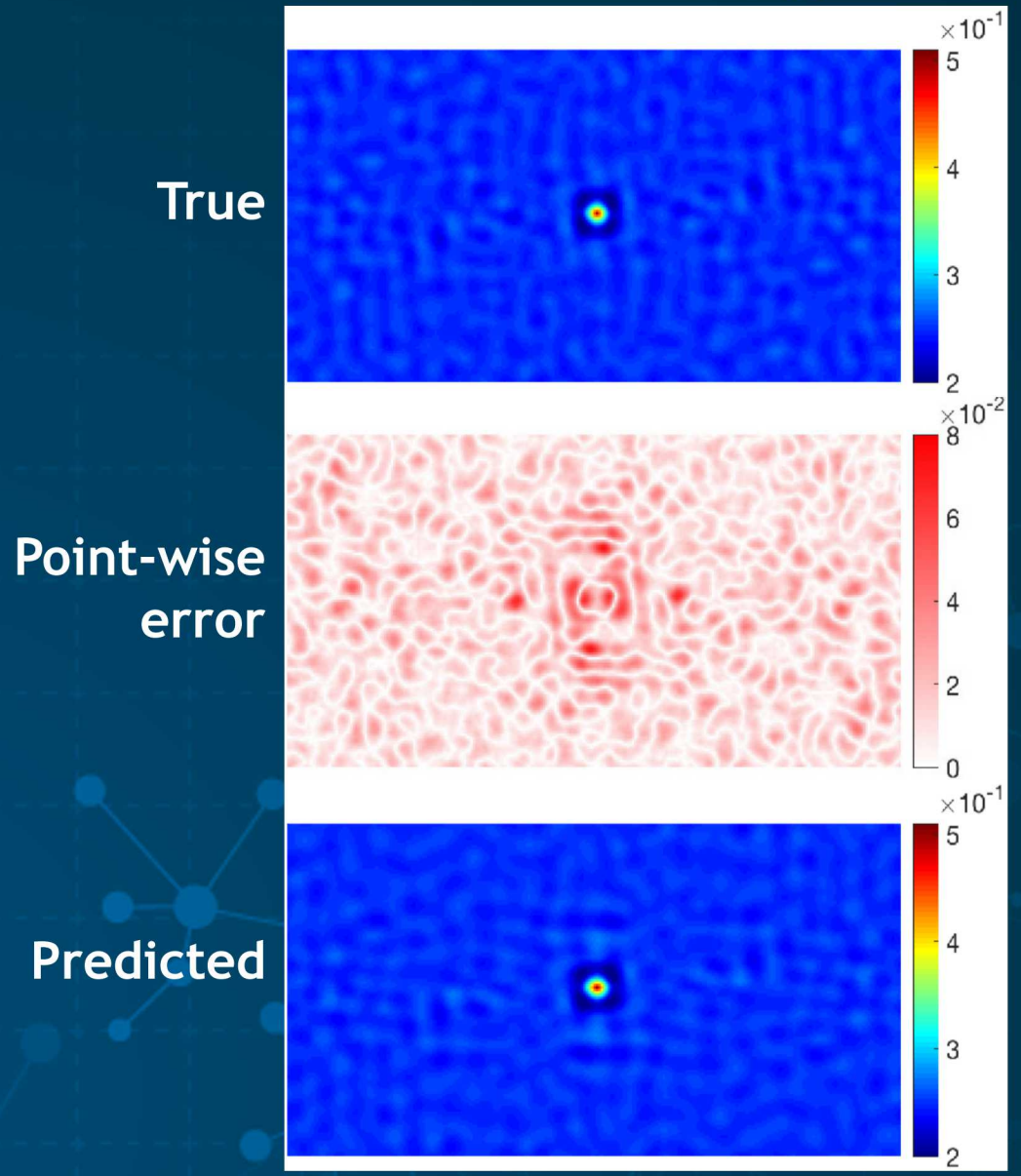


Rapidly Predicting Morphology?



Robust & Efficient Protocol





Sources of Errors

Error on 2pt-stats

$$\mathrm{err}_1(\theta) = \frac{||\mathrm{S}_{\mathrm{true}}(\theta) - \mathrm{S}_{\mathrm{pca}}(\theta)||_2}{||\mathrm{S}_{\mathrm{true}}(\theta)||_2}$$

Error on mapping process to structure

$$\mathrm{err}_3(\theta) = \frac{||\mathrm{S}_{\mathrm{true}}(\theta) - \mathrm{S}_{\mathrm{predicted}}(\theta)||_2}{||\mathrm{S}_{\mathrm{true}}(\theta)||_2}$$

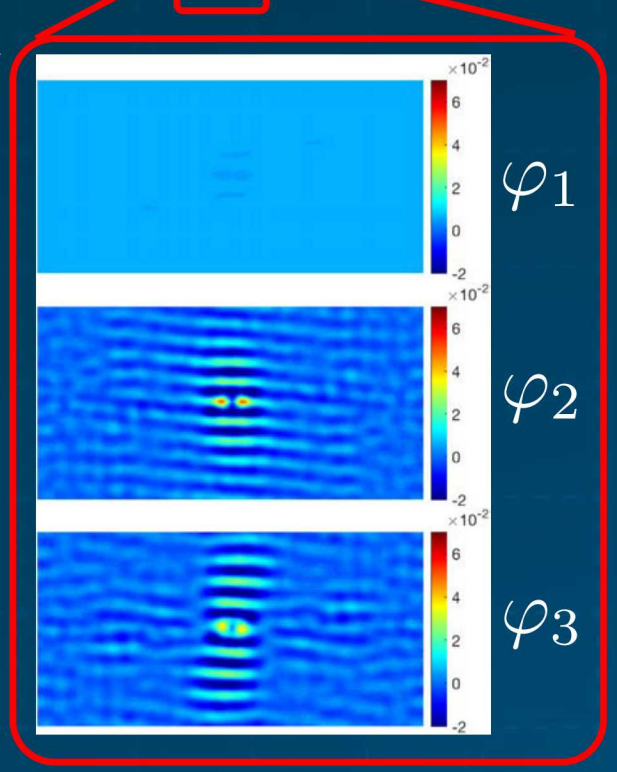
Error	min	max	mean	standard deviation
$\mathrm{err}_1(\boldsymbol{\theta}_{\mathrm{train}})$	0.0030	0.0690	0.0146	0.0123
$\mathrm{err}_1(\boldsymbol{\theta}_{\mathrm{test}})$	0.0030	0.0441	0.0123	0.0091
$\mathrm{err}_3(\boldsymbol{\theta}_{\mathrm{train}})$	0.0032	0.1197	0.0206	0.0223
$\mathrm{err}_3(\boldsymbol{\theta}_{\mathrm{test}})$	0.0032	0.0771	0.0154	0.0139

1. Overall the error is less than 2% for both training and test sets
2. Error values confirm predictiveness and robustness of overall protocol
3. Largest source of error propagation in protocol stems from PCE

Sources of Errors

PCA reduces the dimensionality

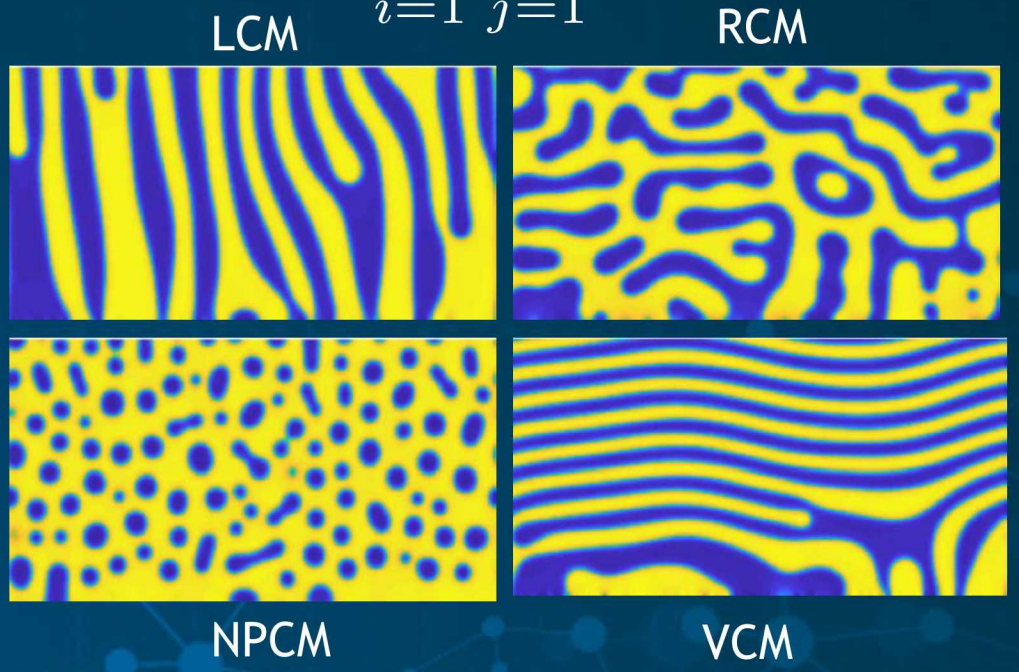
$$S_{\text{pca}} = \sum_{i=1}^{Q} \alpha_i(\theta)\varphi_i + \hat{S}$$



Basis elements capture complexity of morphology and long-range correlation

PCE maps back to processing parameters

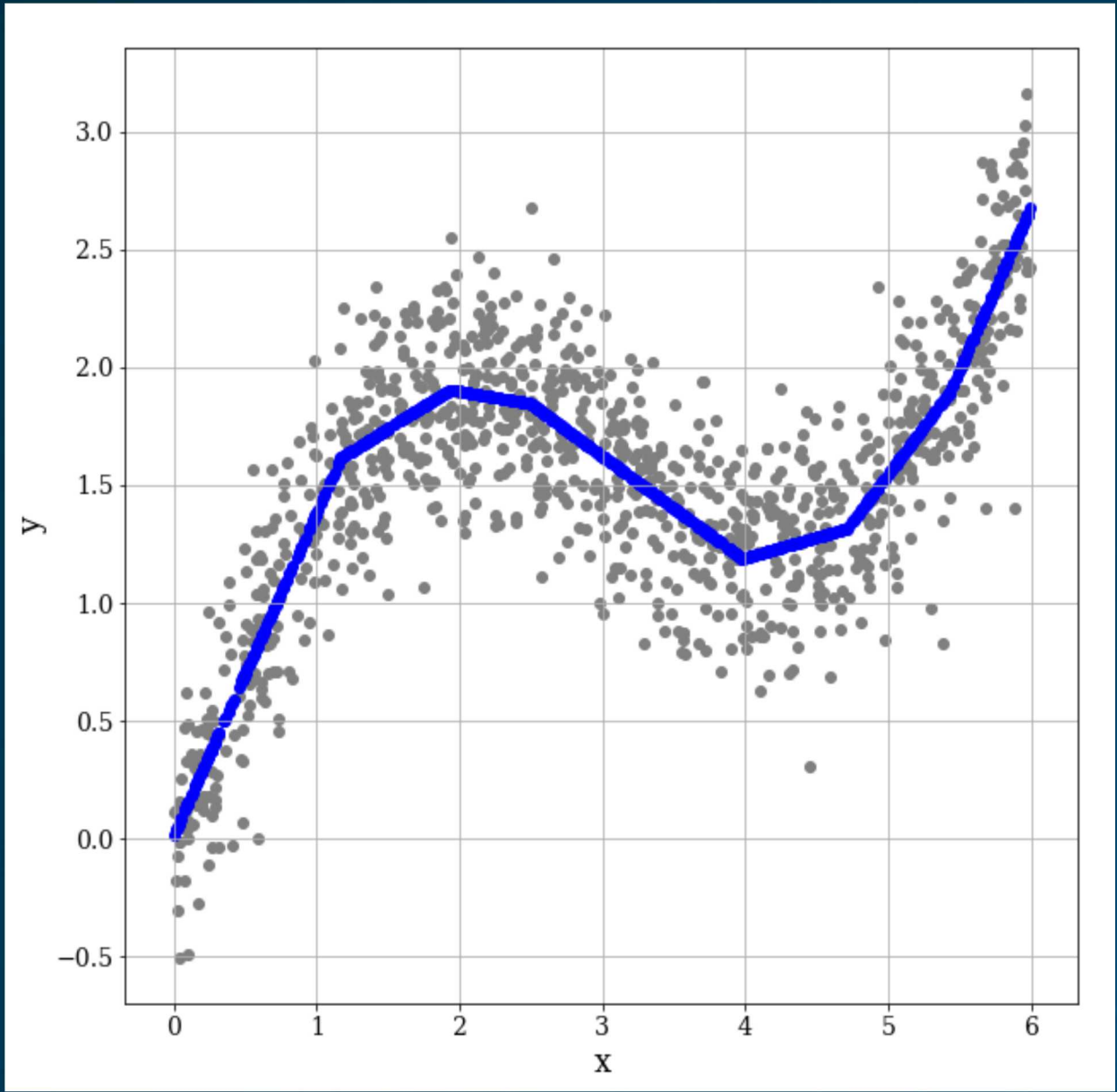
$$S_{\text{predicted}} = \sum_{i=1}^{Q}\sum_{j=1}^{Li} p_j^i \Psi(\theta)\varphi_i + \hat{S}$$



Small changes in processing input space results in drastic change in morphology: but…Legendre polynomial in PCE are continuous polynomials

ACCELERATING MICROSTRUCTURE EVOLUTION

Time-series Forecasting



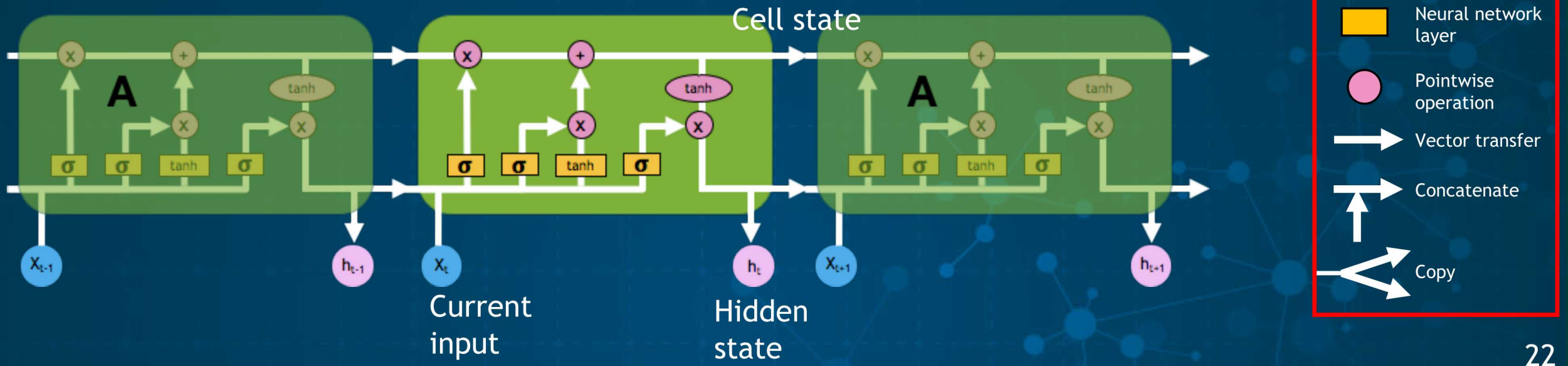
TIME-SERIES MULTIVARIATE ADAPTIVE REGRESSION SPLINES (TSMARS)

1. Useful for identifying nonlinear structure in time series
2. Non-parametric
3. Autoregressive: divide time-series into optimal subdomains to fit splines
4. Predicting microstructure evolution trajectories in PC space
5. Training is done for N previous time steps, predicting N+1, N+2,…

TIME-SERIES FORECASTING

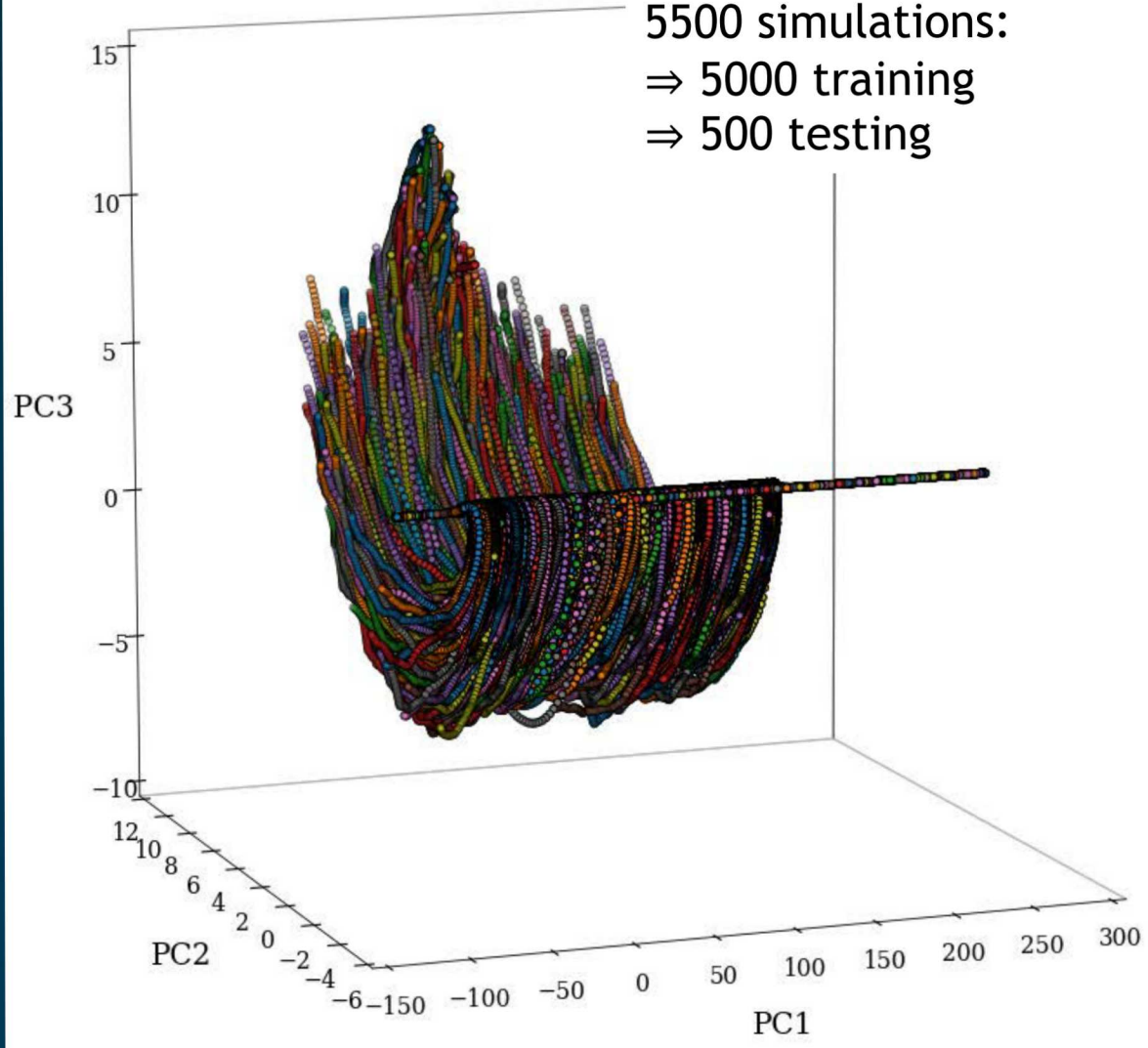
LONG SHORT TERM MEMORY NETWORK (LSTM)

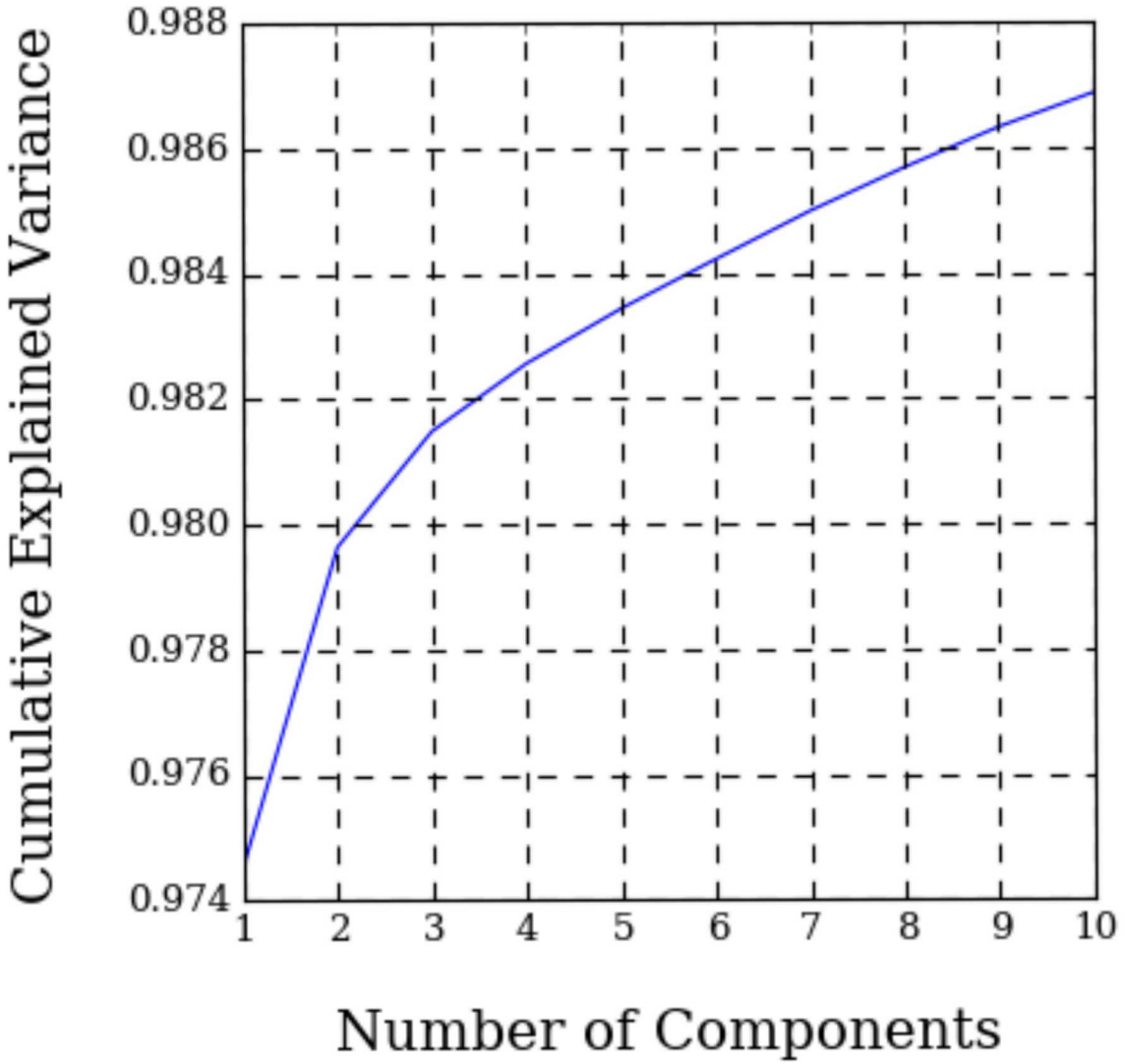
1. LSTM is a special kind of a recurrent neural net (RNN):
   Network with loops in them allowing information to persist (i.e, memory)
2. Looping connect previous information to current: TIME HISTORY!
3. Uses previous states, and current input
4. Learn/forget gate (internal structure) used to form long-term memory (known as cell state)and short-term memory (hidden state)
5. Training is done for N time steps, predicting N+1, N+2,…



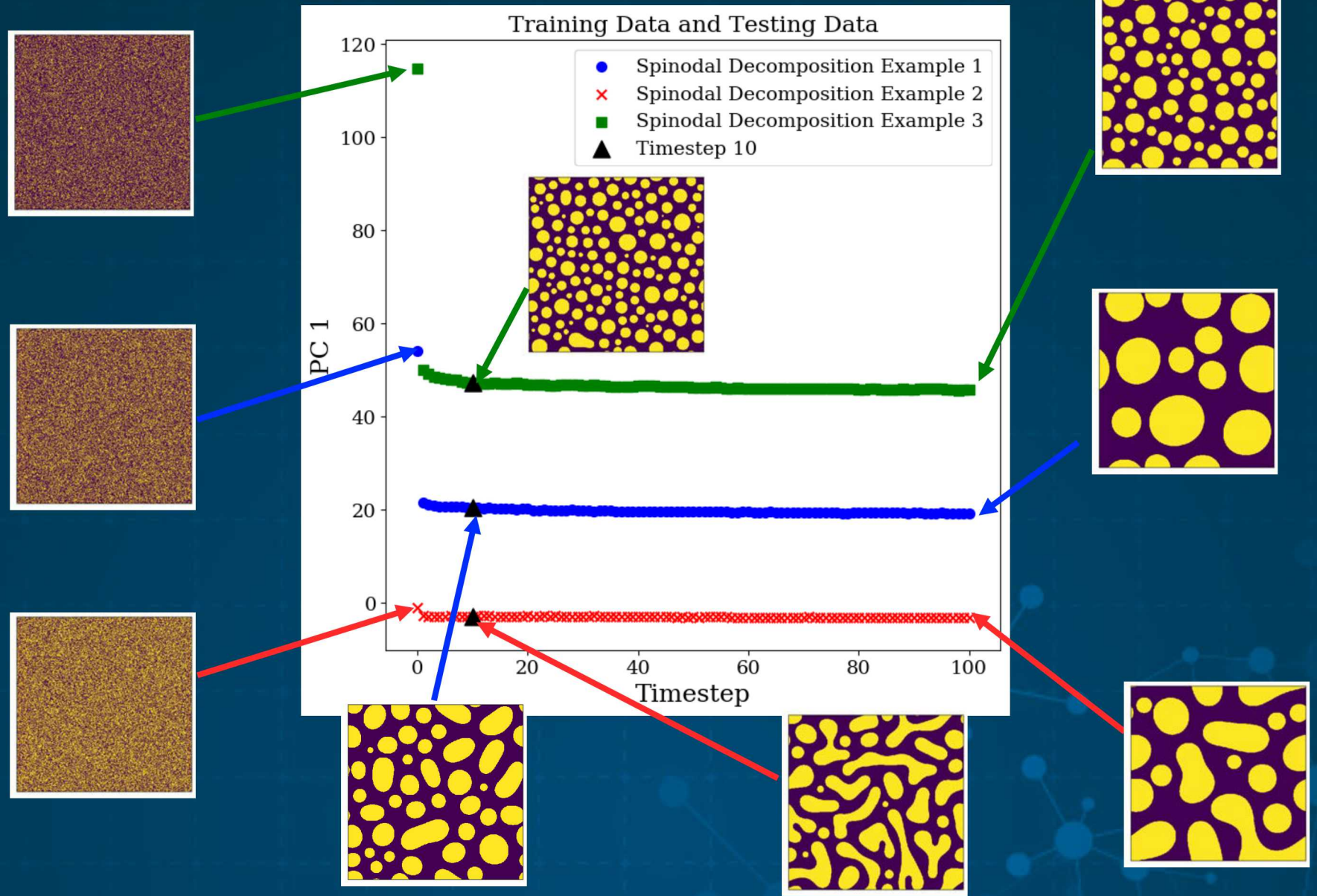
Microstructure Trajectory in PC Space

5500 simulations:
⇒ 5000 training
⇒ 500 testing



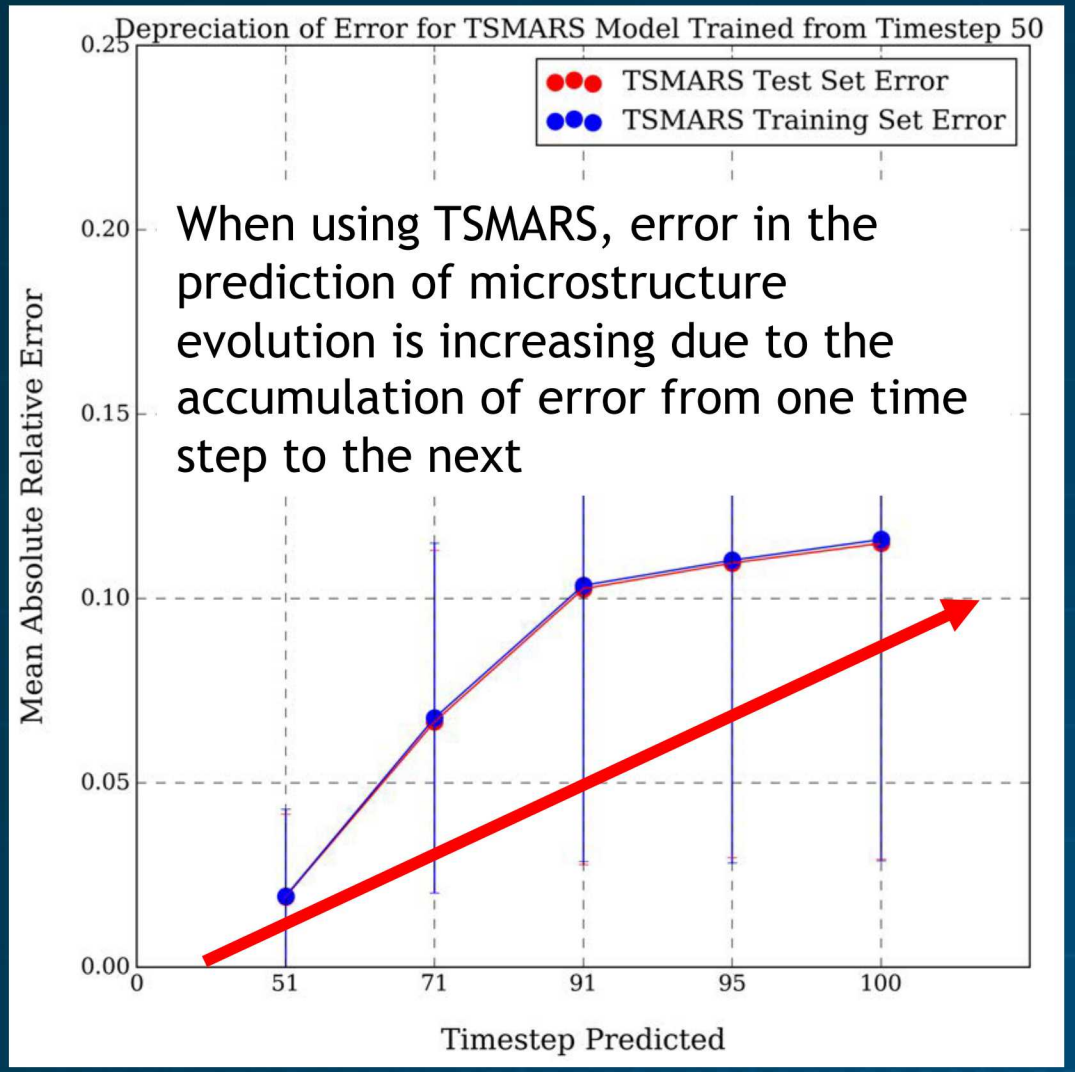


Microstructure Trajectory in PC Space



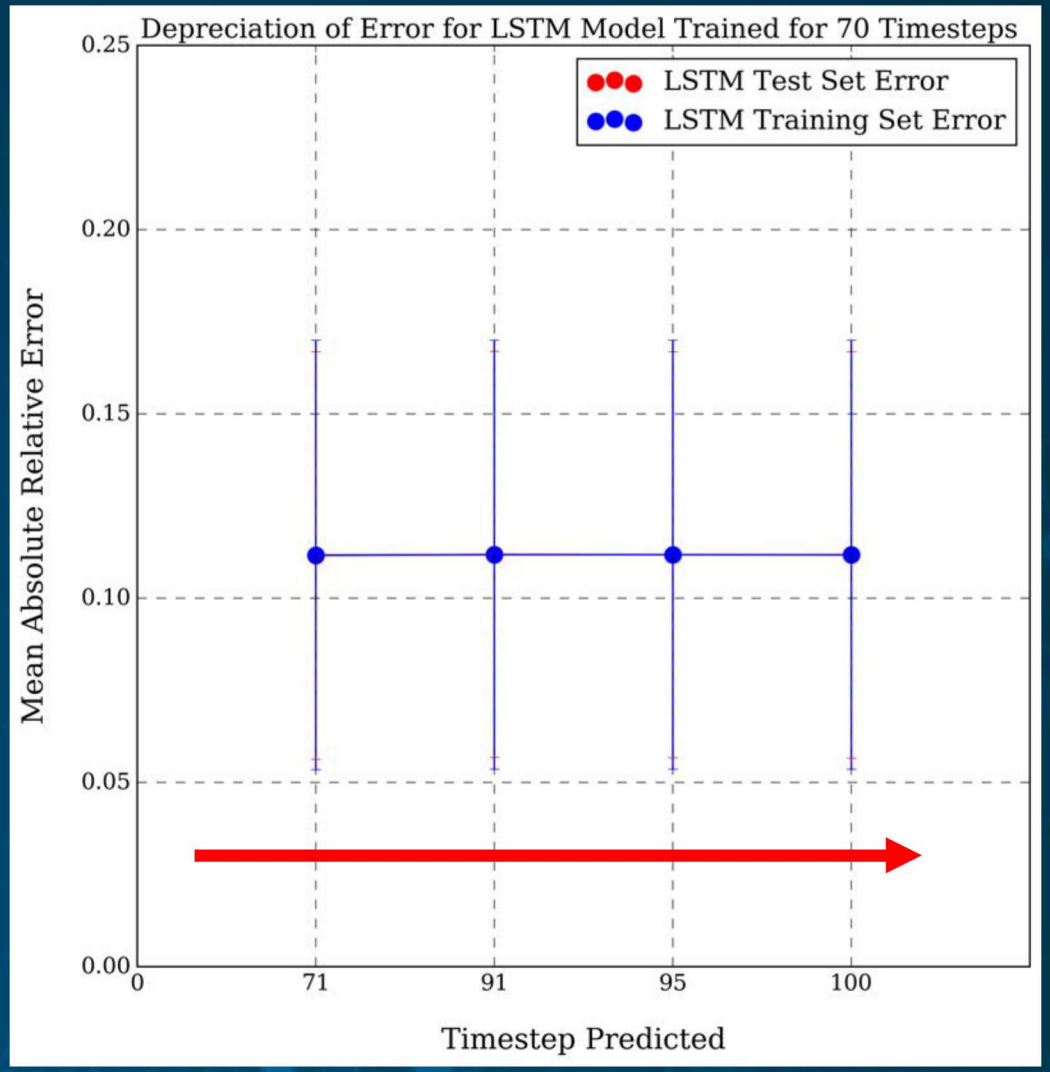
Why Do We Need A Deep Learning Strategy?

TSMARS



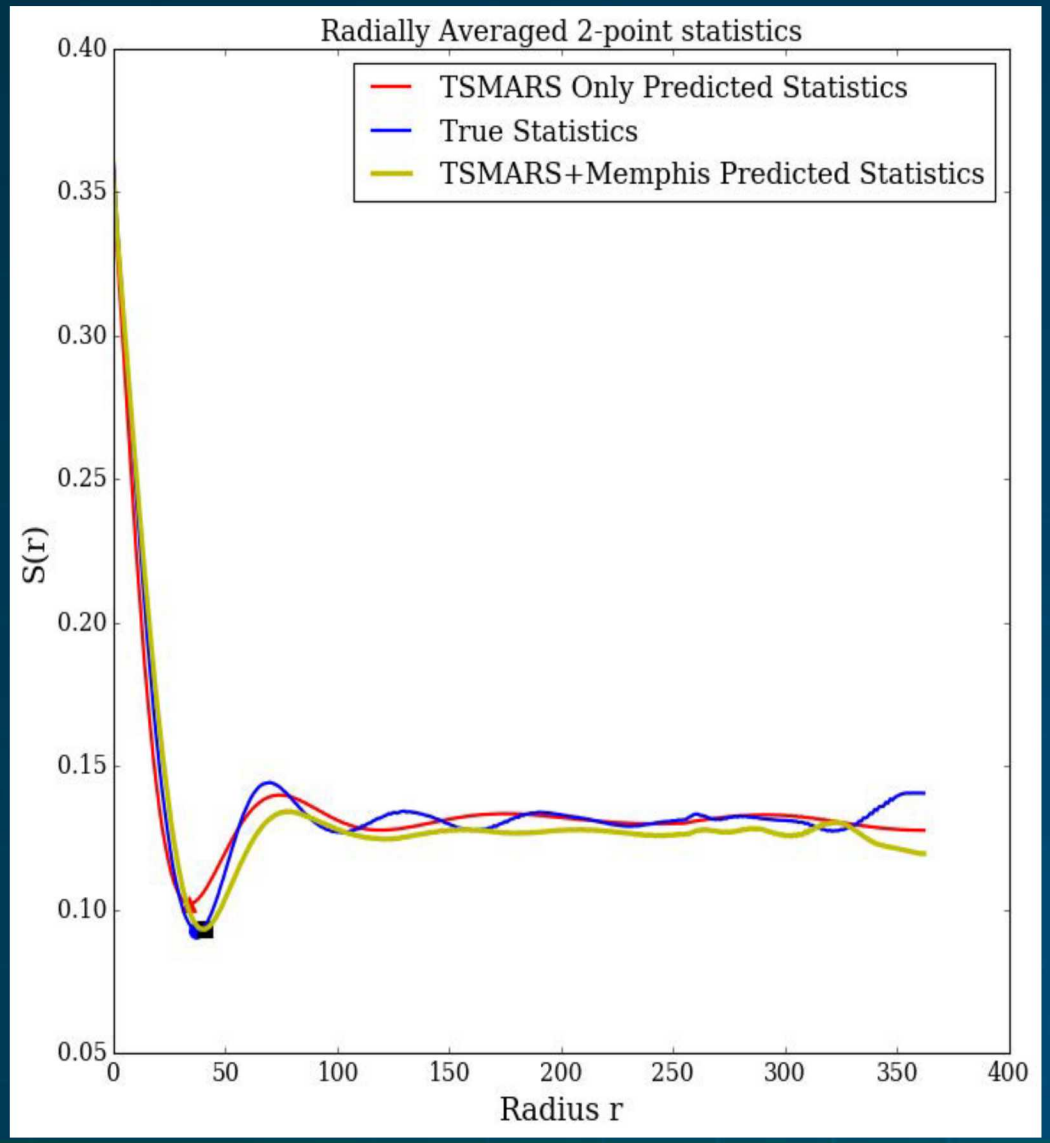
When using TSMARS, error in the prediction of microstructure evolution is increasing due to the accumulation of error from one time step to the next

LSTM

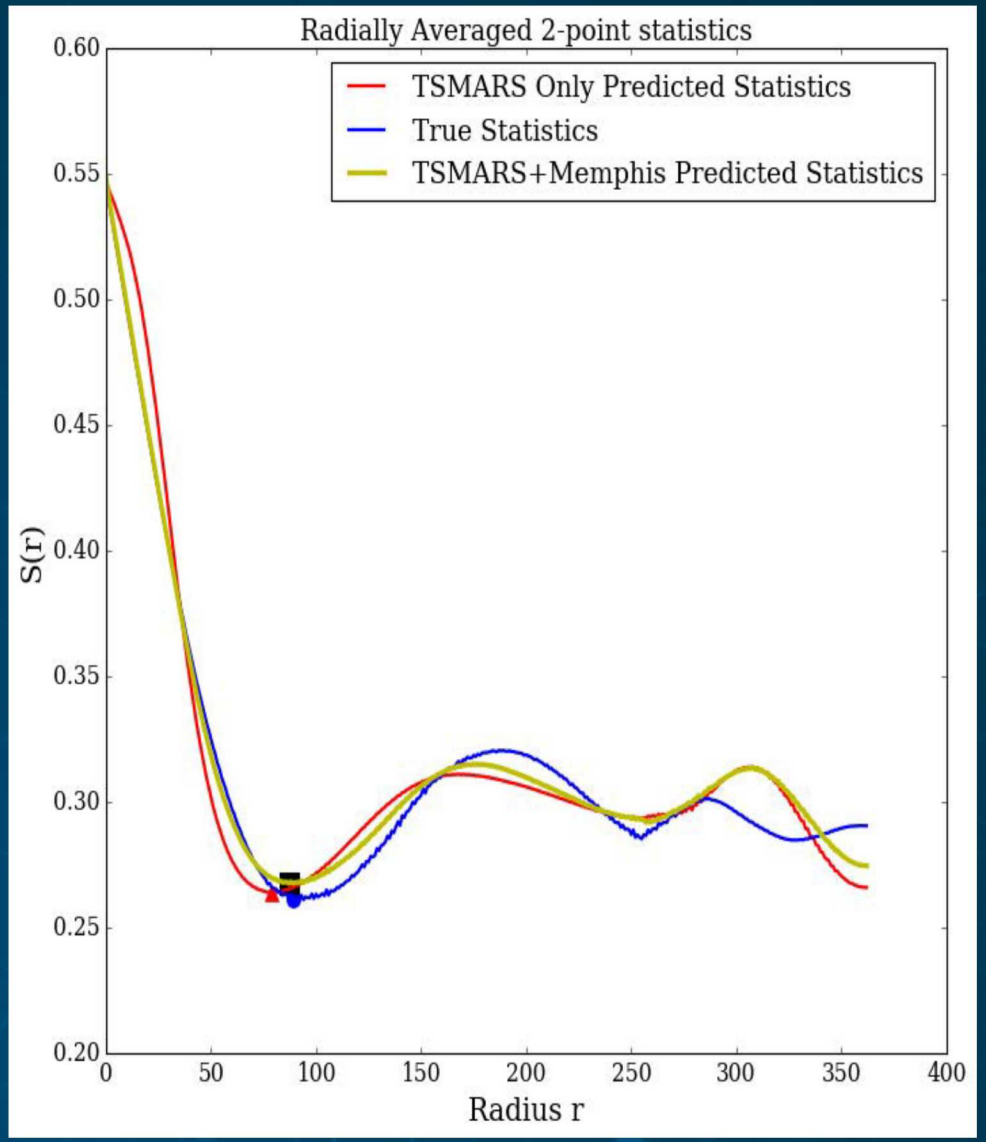


Feeding Predictions Back Into Memphis

TSMARS



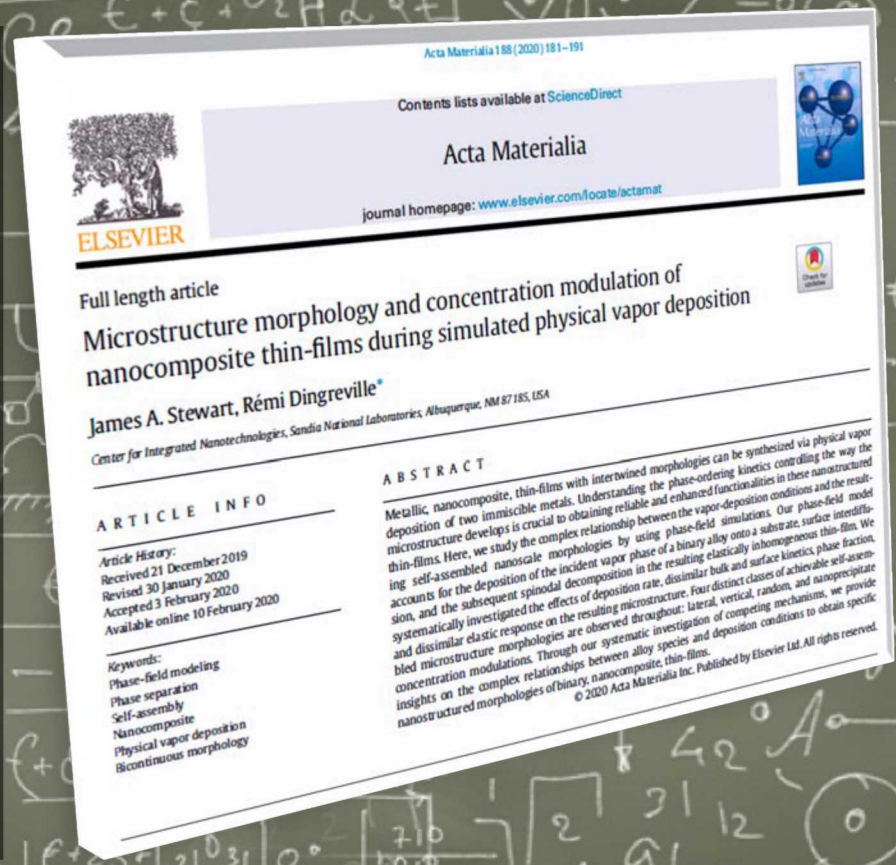
LSTM



MOVING FORWARD

MEMPHIS

- Open-source and available through CINT user program
- Modular implementation
- Performance comparable to other PF capabilities
- Extensions includes LB, SMB
- Development of additional Multiphysics models (e.g., ISCC)



Acta Materialia 188 (2020) 181–191

Contents lists available at ScienceDirect

Acta Materialia

journal homepage: www.elsevier.com/locate/actamat

Full length article

Microstructure morphology and concentration modulation of nanocomposite thin-films during simulated physical vapor deposition

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ARTICLE INFO

Article History:
Received 21 December 2019
Revised 30 January 2020
Accepted 3 February 2020
Available online 10 February 2020

Keywords:
Phase-field modeling
Phase separation
Self-assembly
Nanocomposite
Physical vapor deposition
Bicontinuous morphology

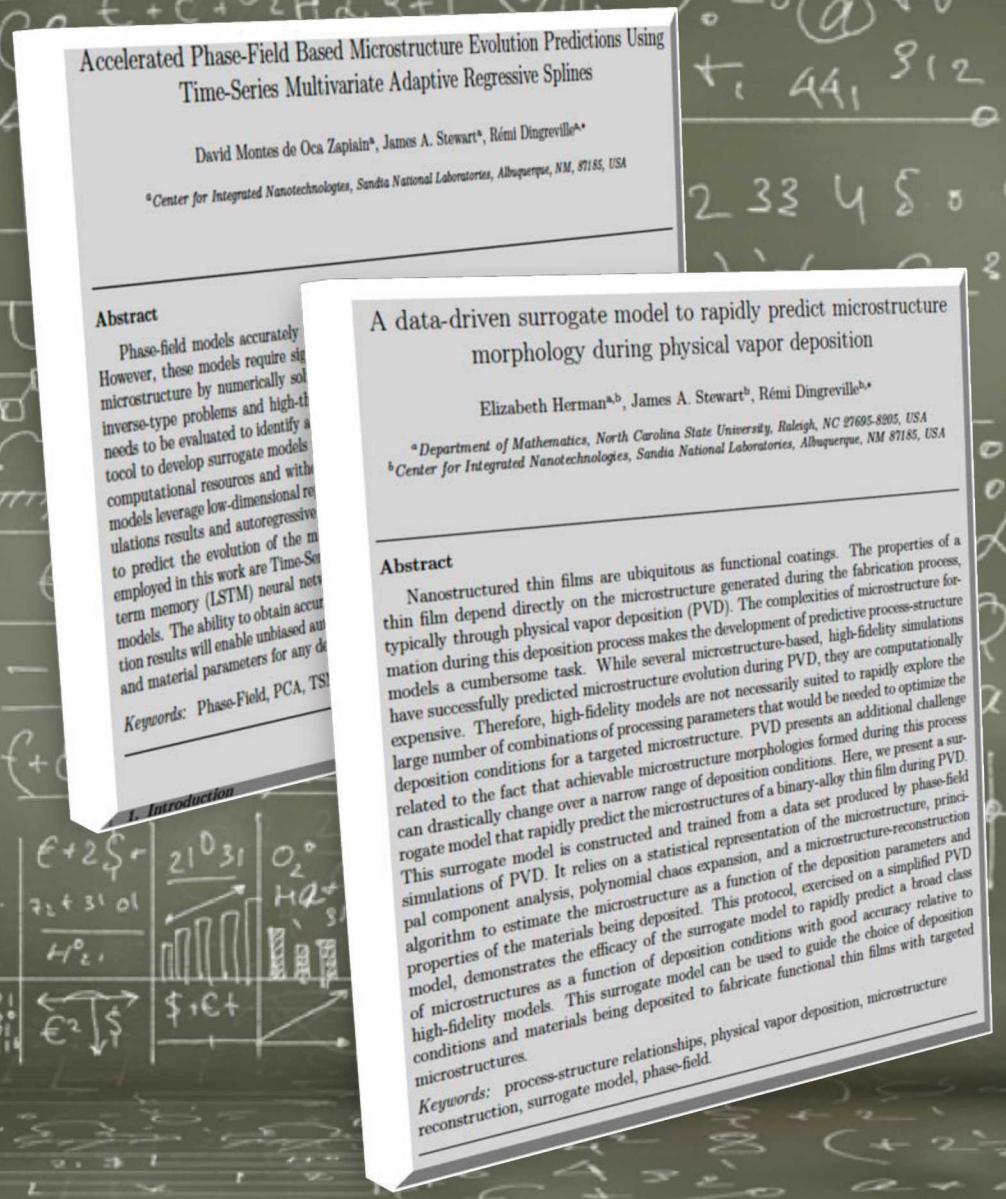
ABSTRACT

Metallic nanocomposite, thin-films with interwined morphologies can be synthesized via physical vapor deposition of two immiscible metals. Understanding the phase-ordering kinetics controlling the way the microstructure develops is crucial to obtaining reliable and enhanced functionalities in these nanostructured thin-films. Here, we study the complex relationship between the vapor-deposition conditions and the resulting self-assembled nanoscale morphologies by using phase-field simulations. Our phase-field model accounts for the deposition of the incident vapor phase of a binary alloy onto a substrate, surface interdiffusion, and the subsequent spinodal decomposition in the resulting elastically inhomogeneous thin-film. We systematically investigated the effects of deposition rate, dissimilar bulk and surface kinetics, phase fraction, and dissimilar elastic response on the resulting microstructure. Four distinct classes of achievable self-assembled microstructure morphologies are observed throughout: lateral, vertical, random, and nanoprecipitate concentration modulations. Through our systematic investigation of competing mechanisms, we provide insights on the complex relationships between alloy species and deposition conditions to obtain specific nanostructured morphologies of binary, nanocomposite, thin-films.

MOVING FORWARD

MACHINE LEARNING

- Regression models and deep-learning algorithms to accelerate predictions of microstructure evolution
- Integration of PF-ML capabilities with PVD chamber at LANL to guide synthesis process



Accelerated Phase-Field Based Microstructure Evolution Predictions Using Time-Series Multivariate Adaptive Regressive Splines

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[a]Center for Integrated Nanotechnologies, Sandia National Laboratories, Albuquerque, NM, 87185, USA

Abstract

Phase-field models accurately
However, these models require sig
microstructure by numerically sol
inverse-type problems and high-th
needs to be evaluated to identify a
tocol to develop surrogate models
computational resources and withe
models leverage low-dimensional re
ulations results and autoregressive
to predict the evolution of the m
employed in this work are Time-Se
term memory (LSTM) neural netw
models. The ability to obtain accur
tion results will enable unbiased au
and material parameters for any de

Keywords: Phase-Field, PCA, TS

1. Introduction

A data-driven surrogate model to rapidly predict microstructure morphology during physical vapor deposition

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Abstract

Nanostructured thin films are ubiquitous as functional coatings. The properties of a thin film depend directly on the microstructure generated during the fabrication process, typically through physical vapor deposition (PVD). The complexities of microstructure formation during this deposition process makes the development of predictive process-structure models a cumbersome task. While several microstructure-based, high-fidelity simulations have successfully predicted microstructure evolution during PVD, they are computationally expensive. Therefore, high-fidelity models are not necessarily suited to rapidly explore the large number of combinations of processing parameters that would be needed to optimize the deposition conditions for a targeted microstructure. PVD presents an additional challenge related to the fact that achievable microstructure morphologies formed during this process can drastically change over a narrow range of deposition conditions. Here, we present a surrogate model that rapidly predict the microstructures of a binary-alloy thin film during PVD. This surrogate model is constructed and trained from a data set produced by phase-field simulations of PVD. It relies on a statistical representation of the microstructure, principal component analysis, polynomial chaos expansion, and a microstructure-reconstruction algorithm to estimate the microstructure as a function of the deposition parameters and properties of the materials being deposited. This protocol, exercised on a simplified PVD model, demonstrates the efficacy of the surrogate model to rapidly predict a broad class of microstructures as a function of deposition conditions with good accuracy relative to high-fidelity models. This surrogate model can be used to guide the choice of deposition conditions and materials being deposited to fabricate functional thin films with targeted microstructures.

Keywords: process-structure relationships, physical vapor deposition, microstructure reconstruction, surrogate model, phase-field.

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- Soft, Biological, and Composite Nanomaterials

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