SAND2022-12858C

Machine learning methods for designing thin films

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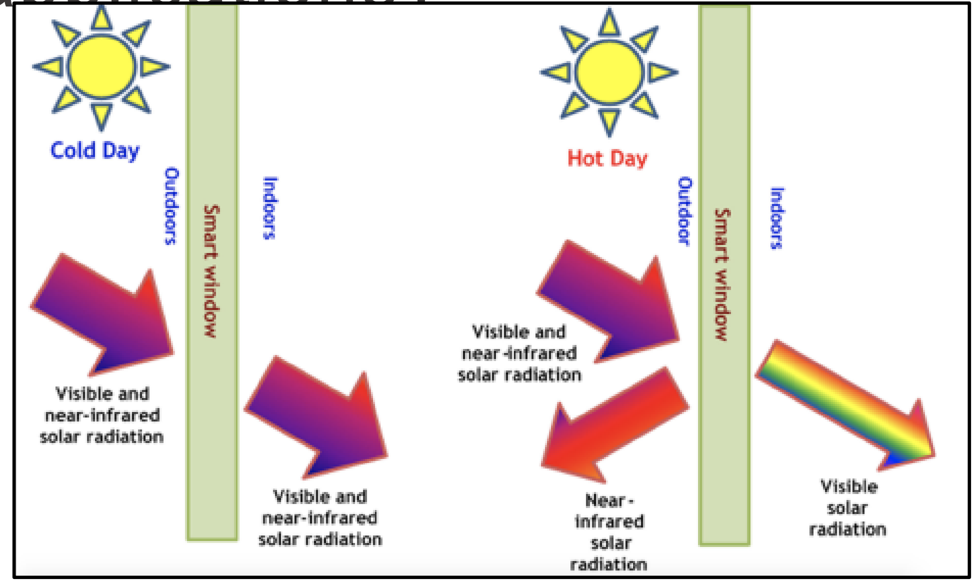
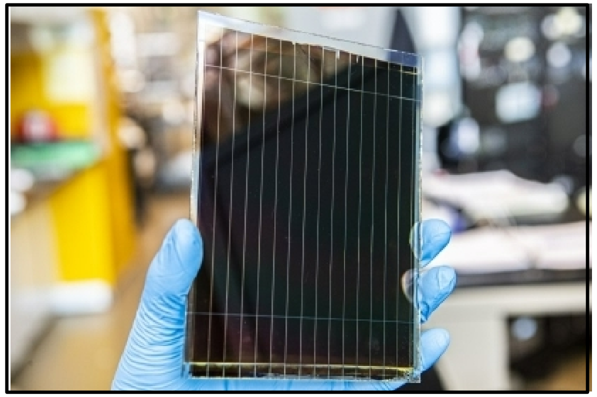
How do we design thin films tailored for specific applications?



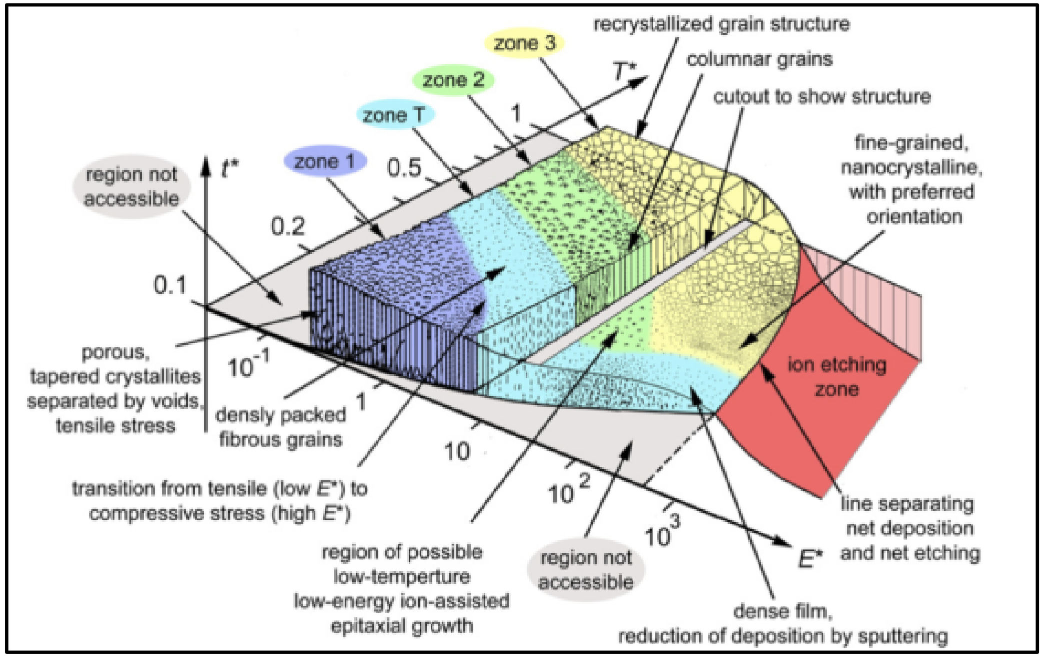
Source: [nist.gov]



Source: [energy.gov]

Source: [certechinc.com]

Designing tailor-made thin films requires an understanding of processing-structure-property linkage



Structure zone diagrams relate processing conditions to microstructure

Anders *Thin Solid Films* (2010)

Microstructure formation in PVD-grown alloy thin films

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

Free energy of system

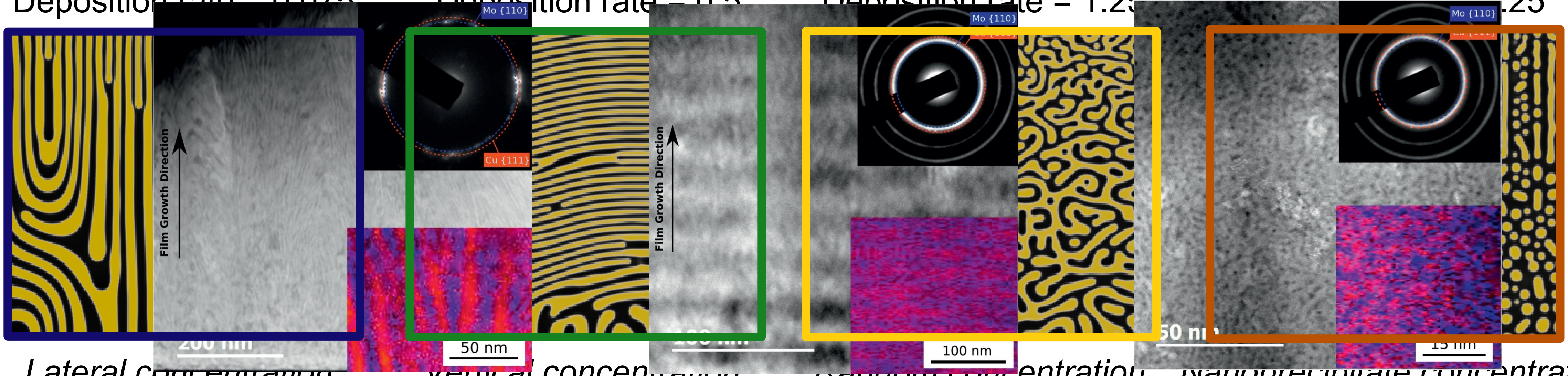
Stewart et al. *Acta Materialia* (2020)

$$\frac{\partial c}{\partial t} = \nabla.\left[M_c(\phi, c)\,\nabla\frac{\delta F}{\delta c}\right] \qquad \frac{\partial \phi}{\partial t} = \nabla.\left[M(\phi)\,\nabla\frac{\delta F}{\delta \phi}\right] + S(n(\phi))$$

$$\frac{\partial \rho}{\partial t} = \nabla.\left[D_\rho\,\nabla\rho\right] - \nabla.\left[\rho\, v\right] - S(n(\phi))$$

Evolution equations

Deposition rate = 0.075 | Deposition rate = 0.5 | Deposition rate = 1.25 | Deposition rate = 1.25 | $A_{65}B_{35}$



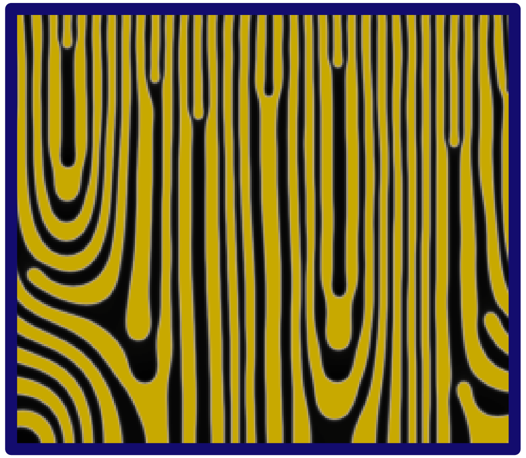
Lateral concentration modulation (LCM) | *Vertical concentration modulation (VCM)* | *Random concentration modulation (RCM)* | *Nanoprecipitate concentration modulation (NPCM)*

Derby et al. Thin Solid Films (2018)

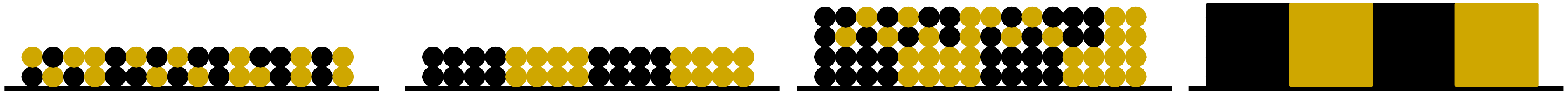
Phase field model simulates microstructure evolution for various deposition conditions

Factors governing microstructure evolution

Deposition rate = 0.075



Low deposition rates / high diffusion times give LCM structures



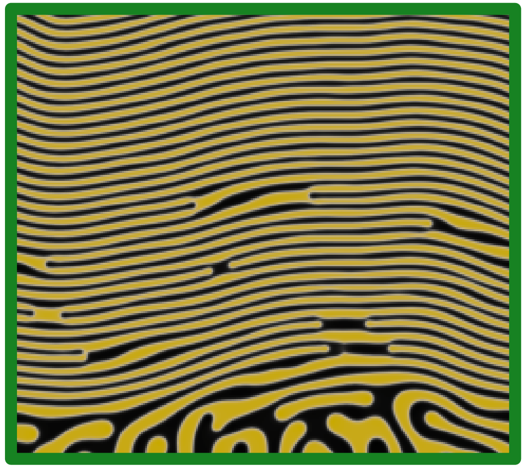
Initial deposition

Spinodal decomposition laterally (complete)

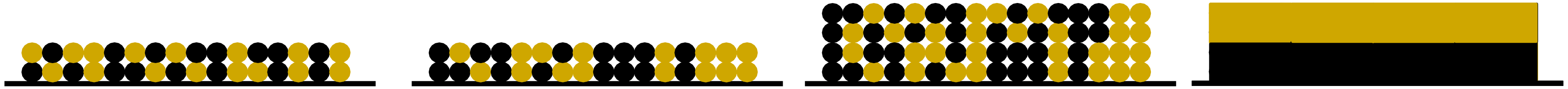
Next deposition

Spinodal decomposition laterally (complete)

Deposition rate = 0.5



High deposition rates / low diffusion times give VCM structures



Initial deposition

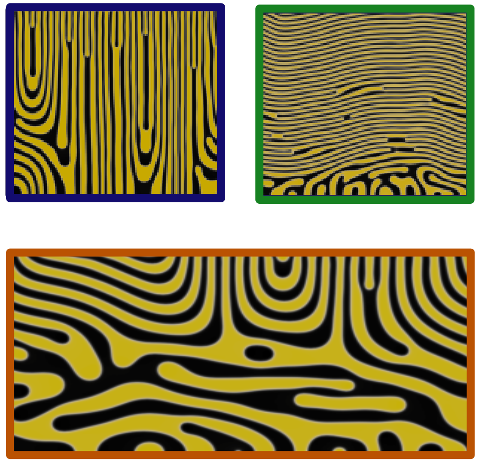
Spinodal decomposition laterally (incomplete)

Next deposition

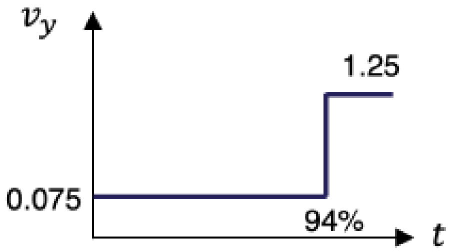
Spinodal decomposition laterally and vertically

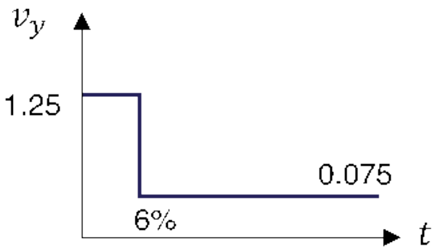
How do we design PVD-grown thin film microstructures?

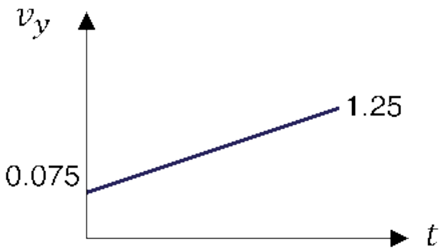
What processing conditions to use to obtain desired film microstructure?

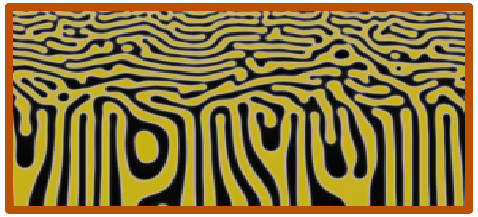


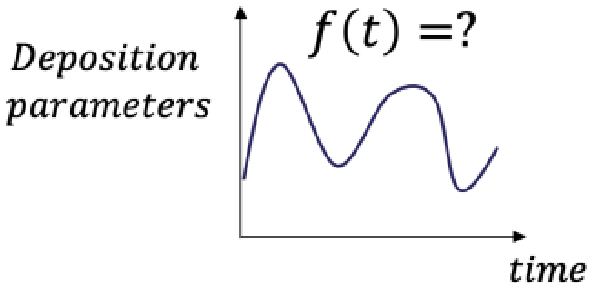
Current SZDs only consider protocols that are constant in time





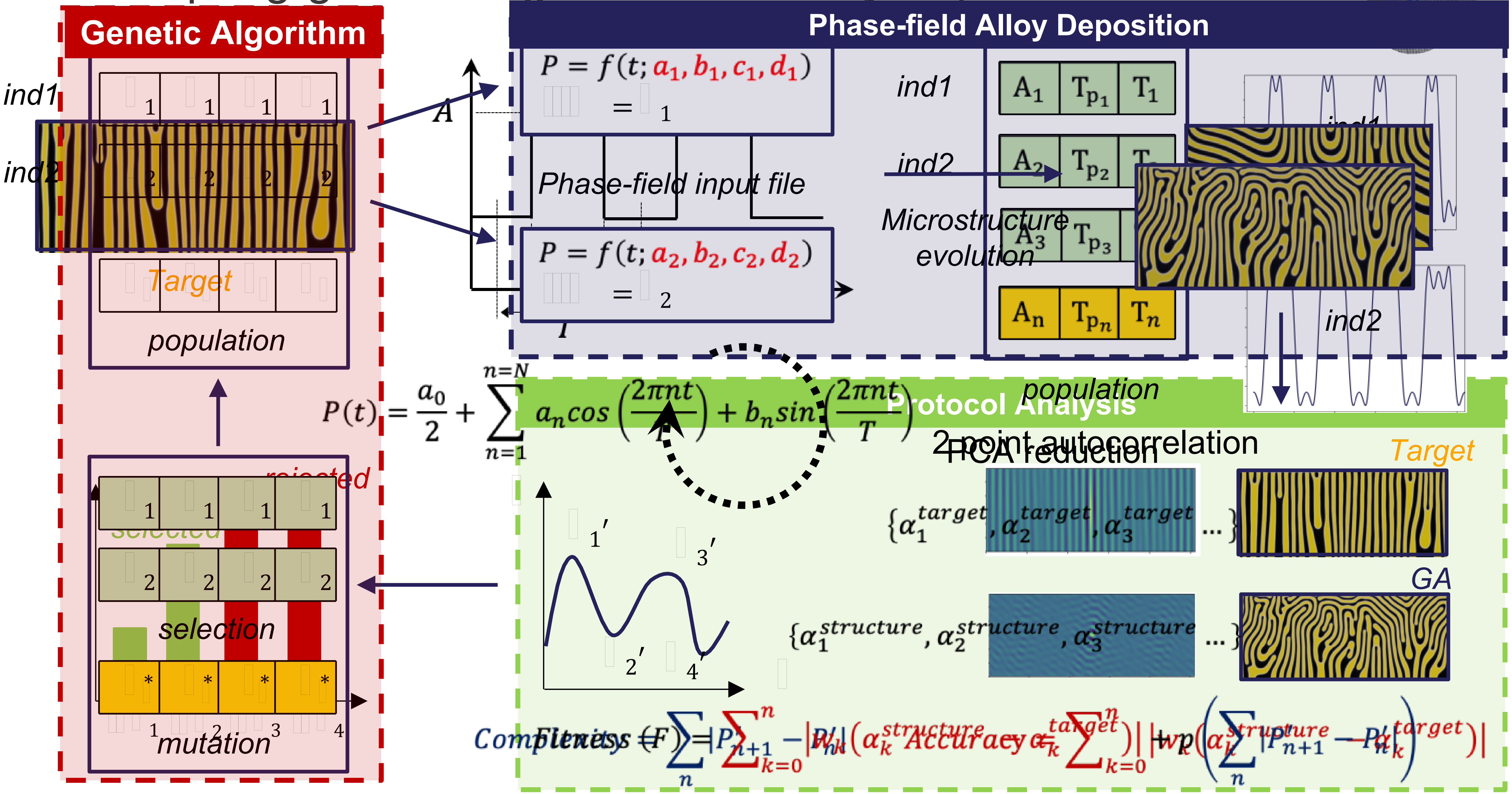




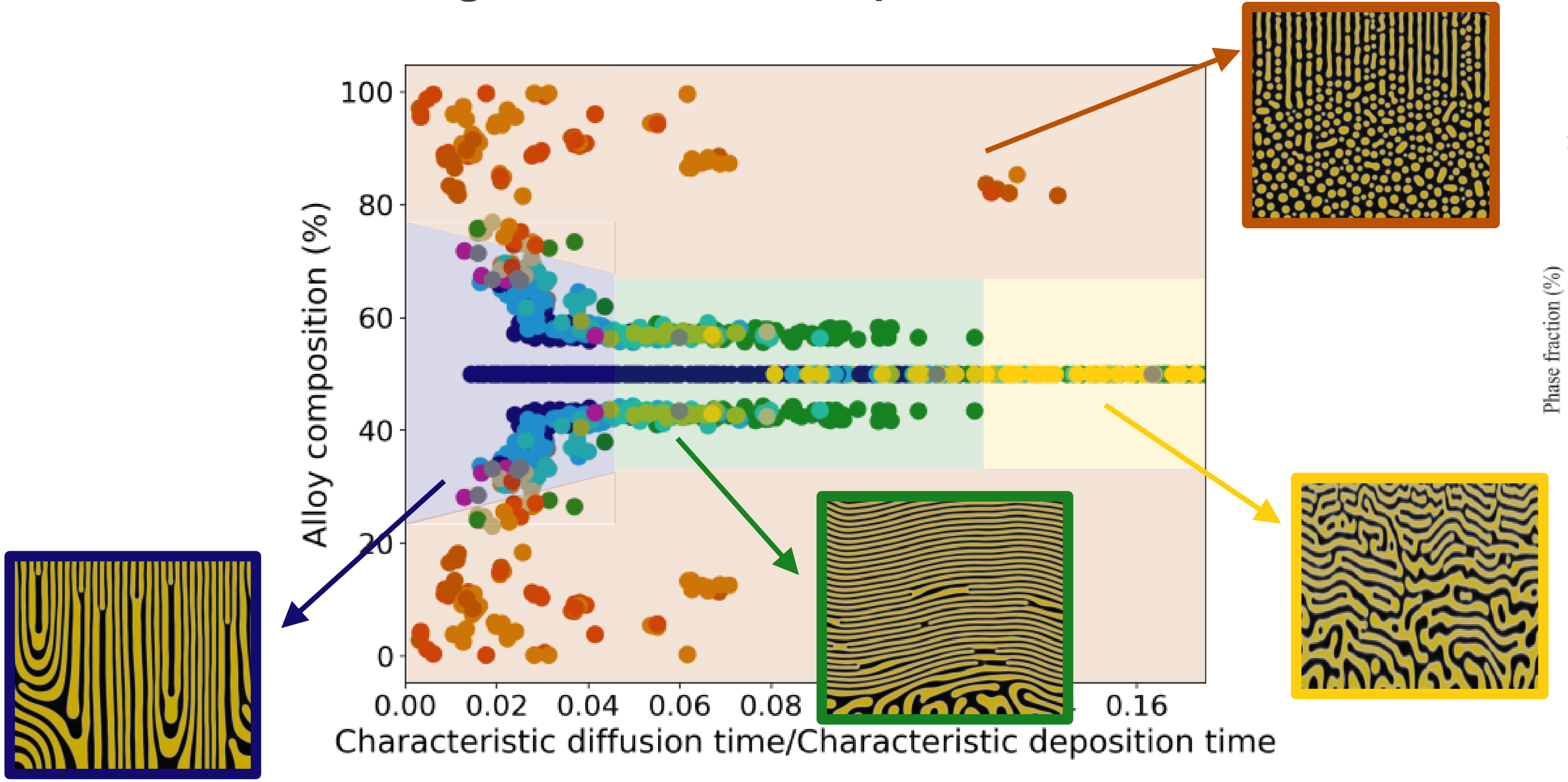


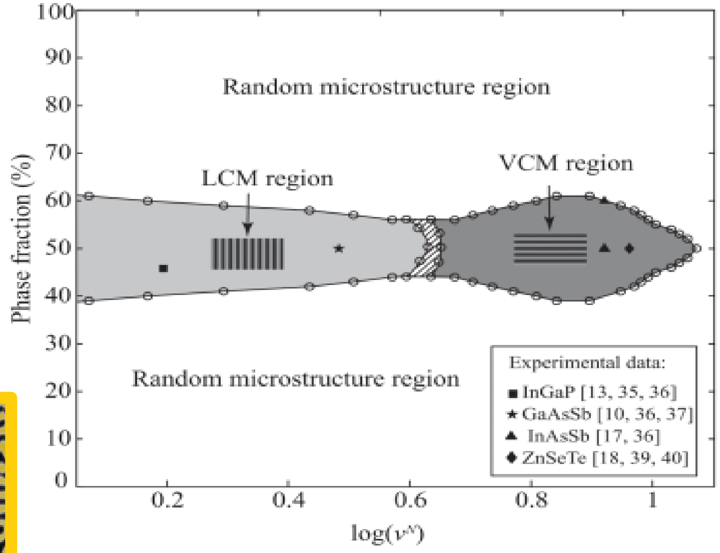
We use a genetic algorithm to discover time-dependent protocols that result in desired microstructure

Coupling genetic algorithms to alloy deposition



Re-creating constant deposition structure zone diagram

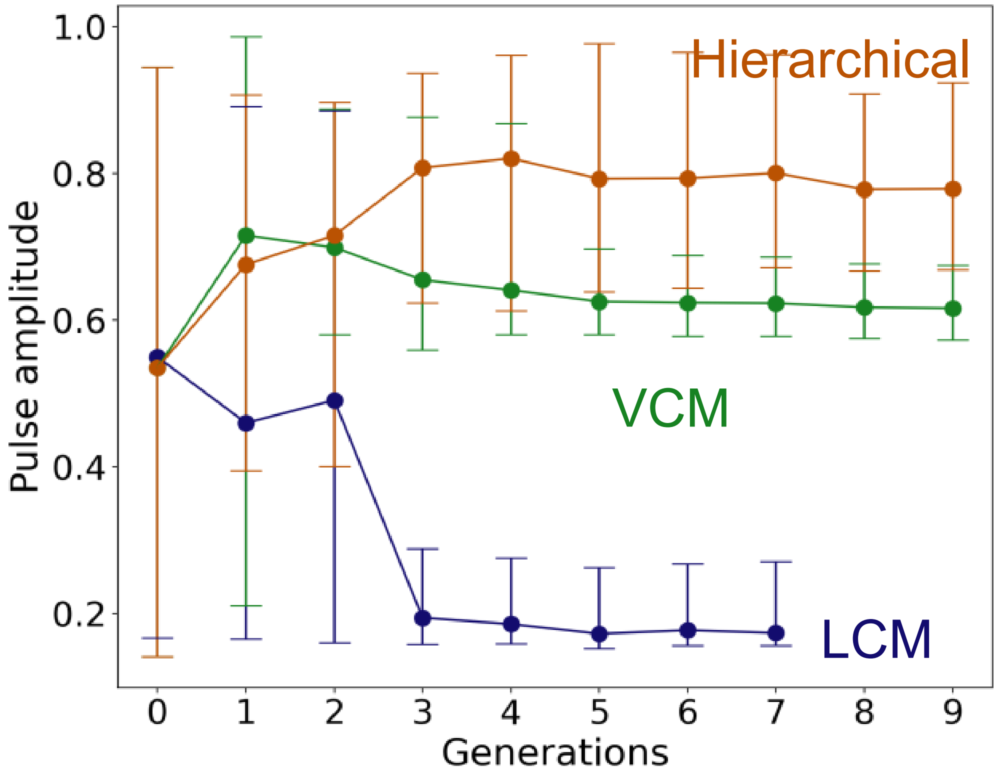


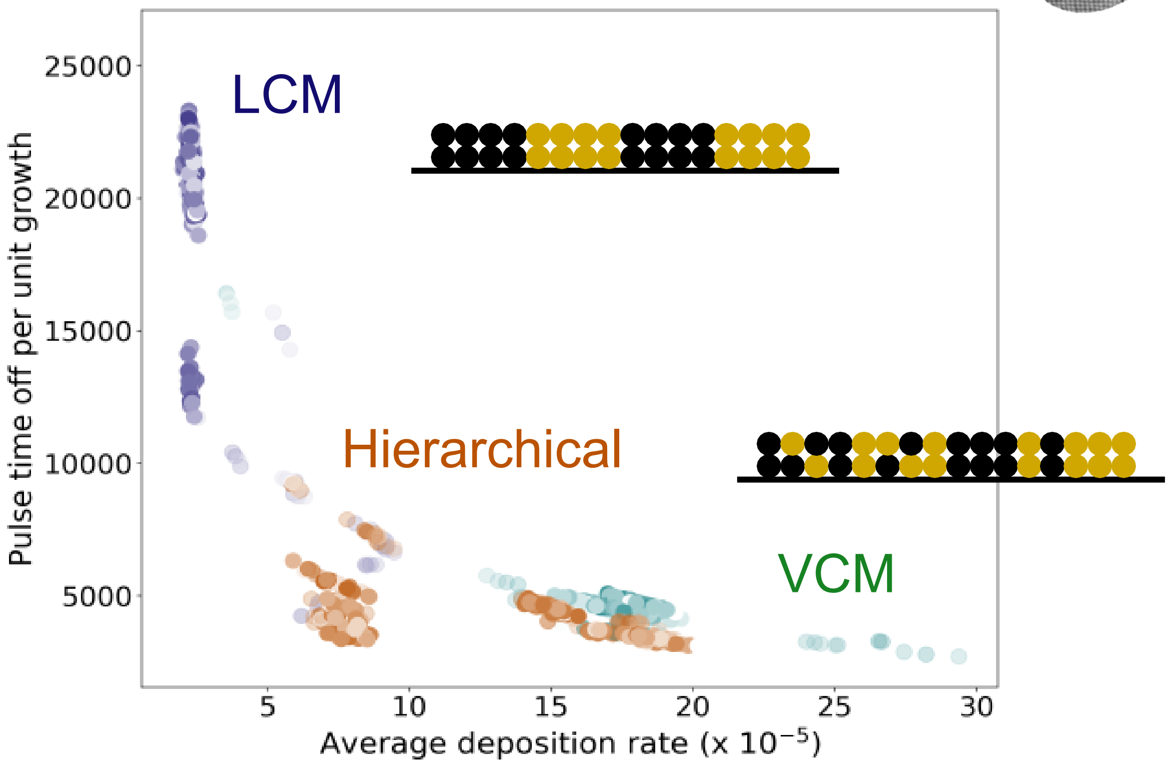


Lu, Yong et al. *Physical review letters* (2012)

- Low deposition/high diffusion rates lead to lateral concentration modulations
- High deposition/low diffusion rates lead to vertical concentration modulations
- Structure zone diagram agrees with previous phase field models and experiments

Understanding the choices of the genetic algorithm





- GA favors low amplitudes to generate LCM structures and high amplitudes for VCM structures
- Range of deposition rates can be used to get hierarchical structures
- Genetic algorithm learns deposition-diffusion trade offs

But…

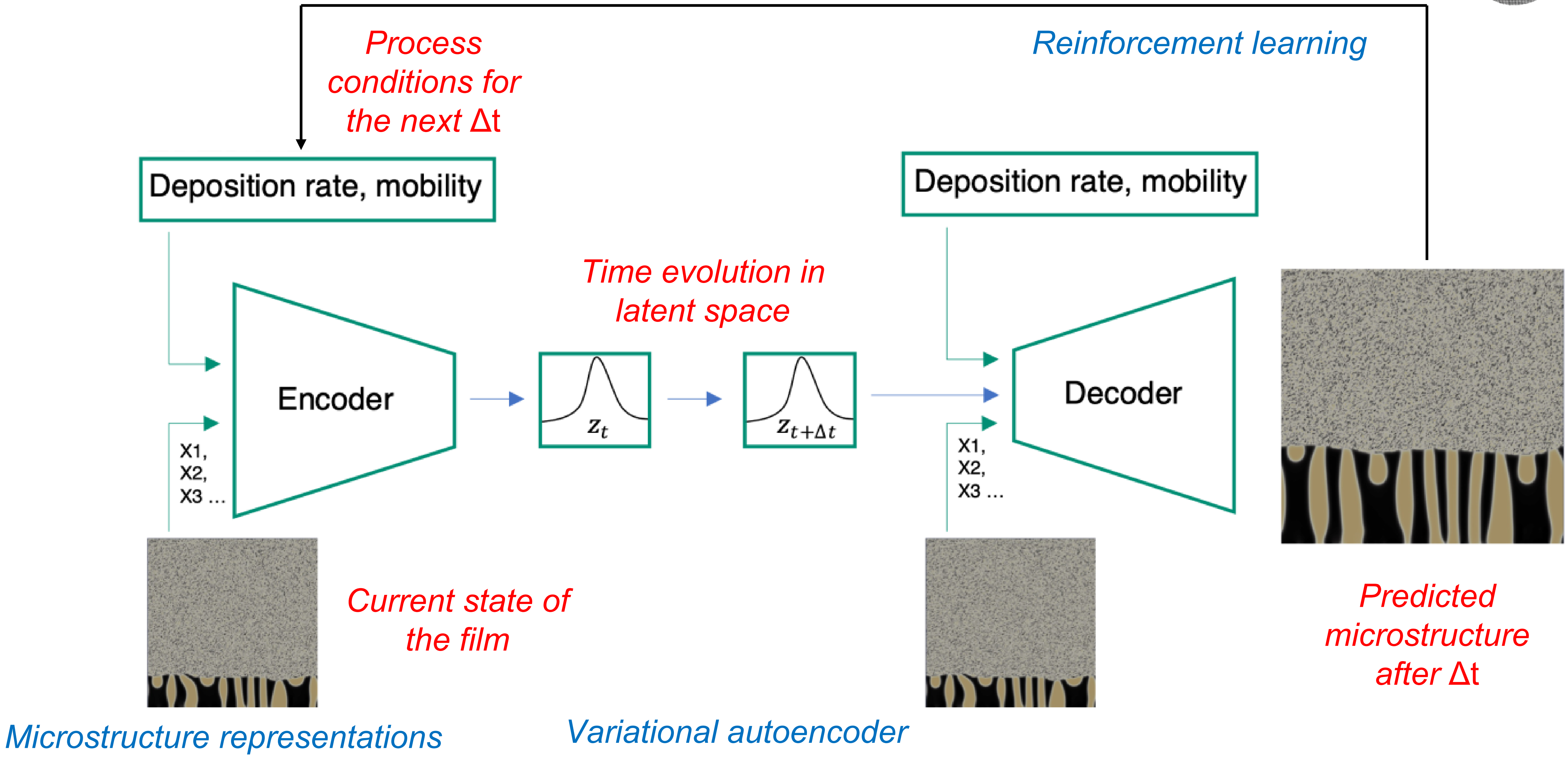
“Online learning” is expensive… we want an “offline” counterpart

Replacing GA-PVD with RL-PVD

Phase field → generative model (VAE) *saves computational time*

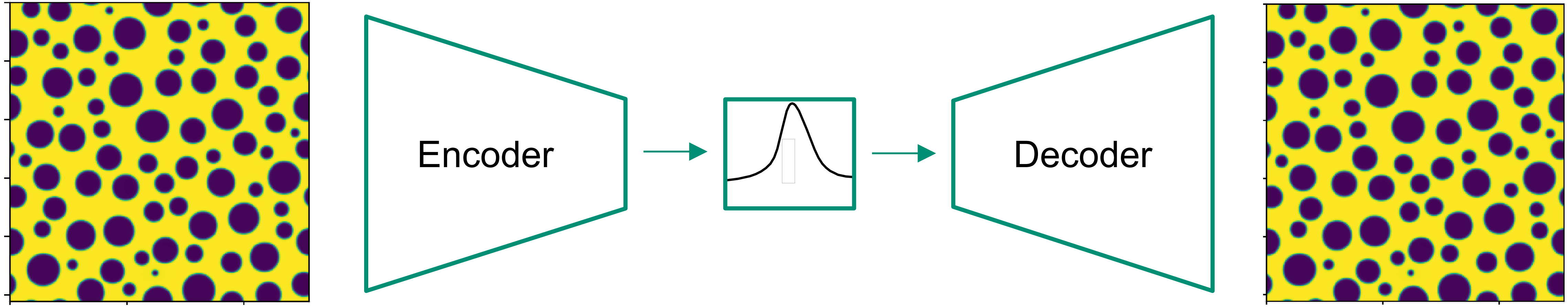
Genetic algorithm → reinforcement learning *explores a broader set of protocols*

RL-PVD framework



Solving simpler problems first…

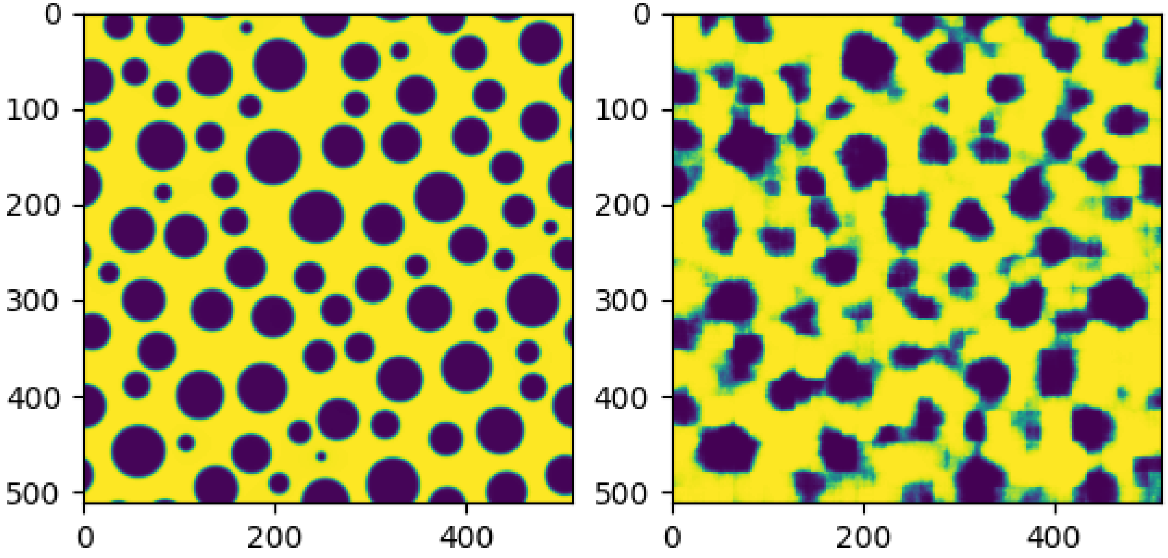
Develop a VAE to generate spinodal decomposition microstructures



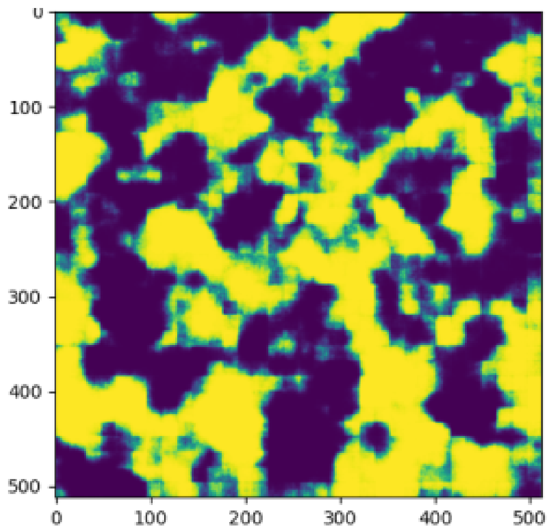
Reconstruction

Generation

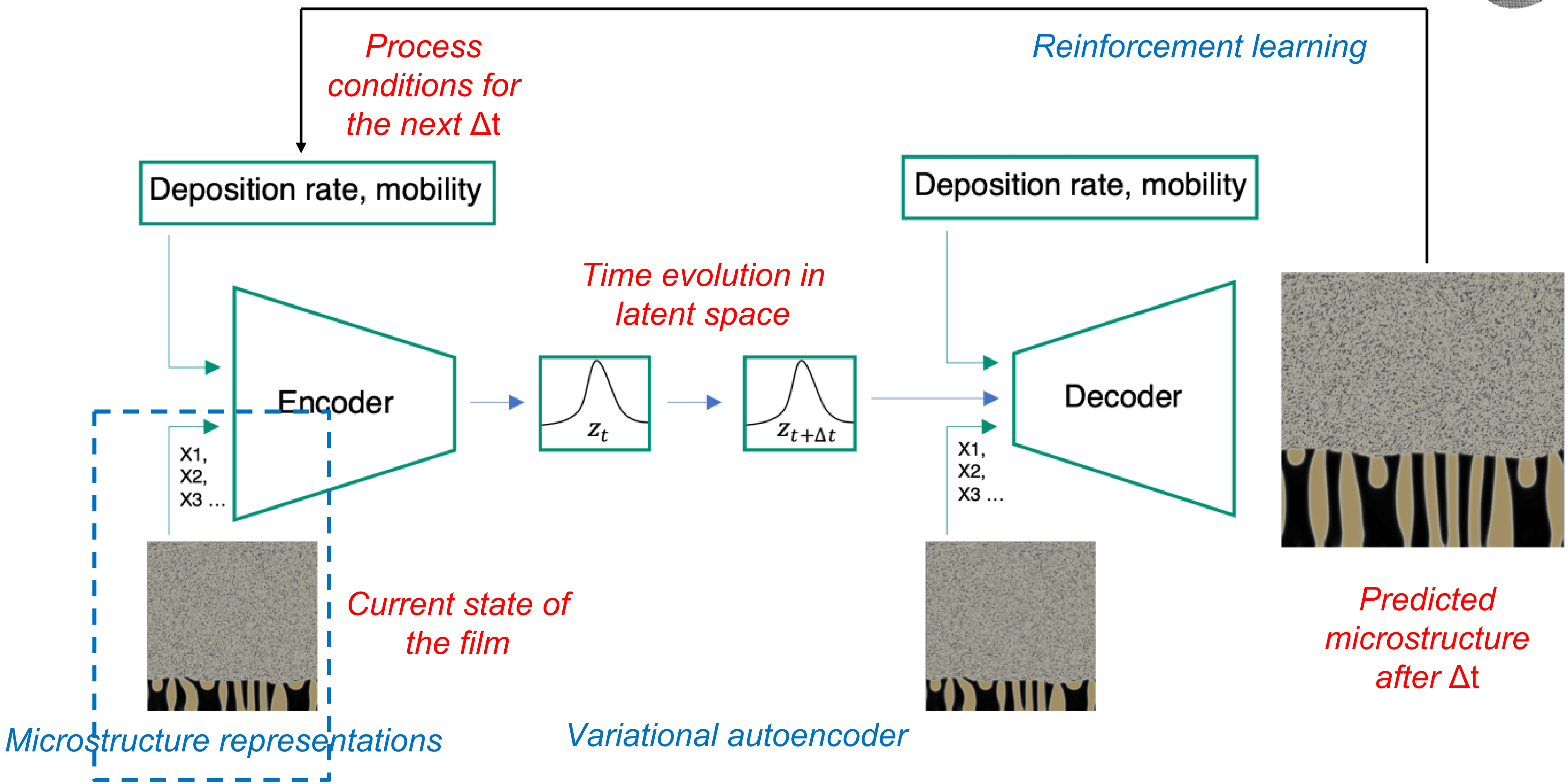
Ground truth



VAE reconstruction



Microstructure encodings?



Comparing low-dimensional embeddings

Represent multi-scale features: Latent dimensions represent features across multiple length/time scales

Low-dimensionality: Small number of latent dimensions accurately represents the microstructure

Smooth time-evolution: Latent dimensions show smooth evolution with time as microstructure evolves (similar microstructures should have similar embeddings)

Methods

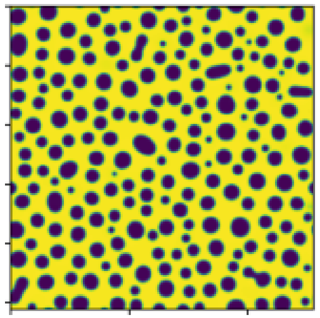
Principal Component Analysis (PCA)

Karhunen Loeve Expansion (KLE)

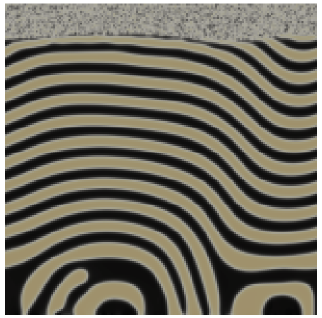
Autoencoders

Diffusion maps

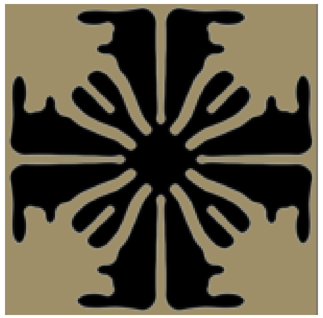
Datasets (2D)



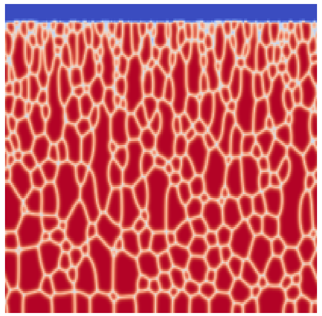
Spinodal decomposition



PVD thin films

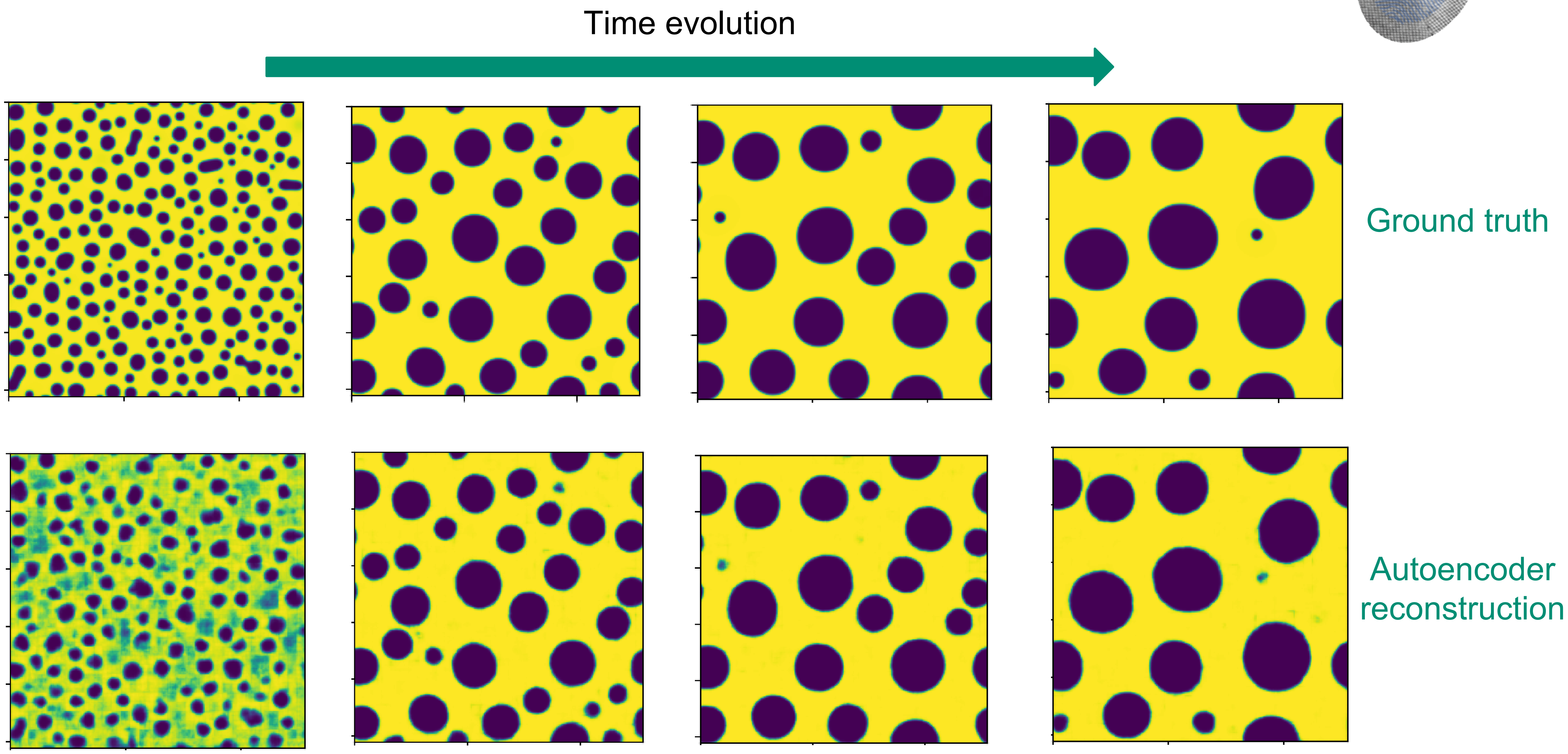


Dendrite growth



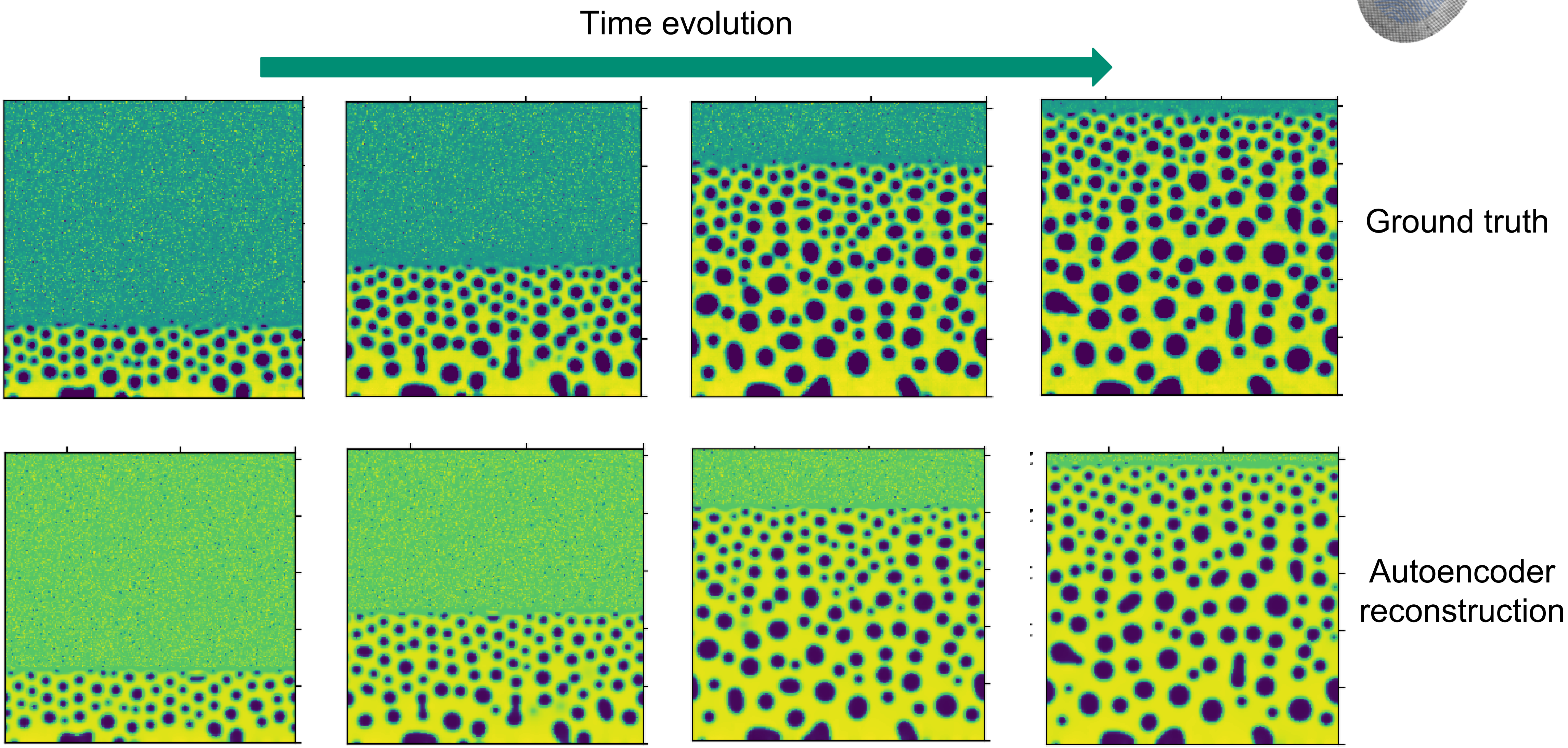
Grain growth

Autoencoders for the spinodal decomposition dataset



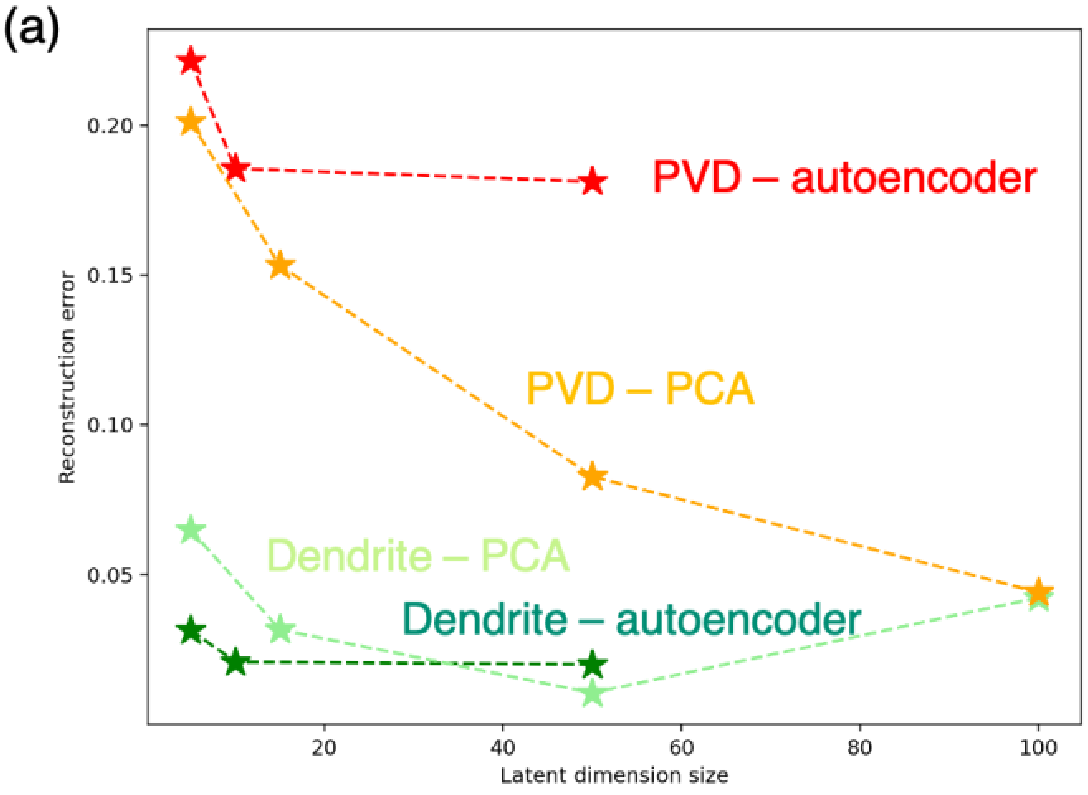
Autoencoders show good general reconstruction for spinodal decomposition data

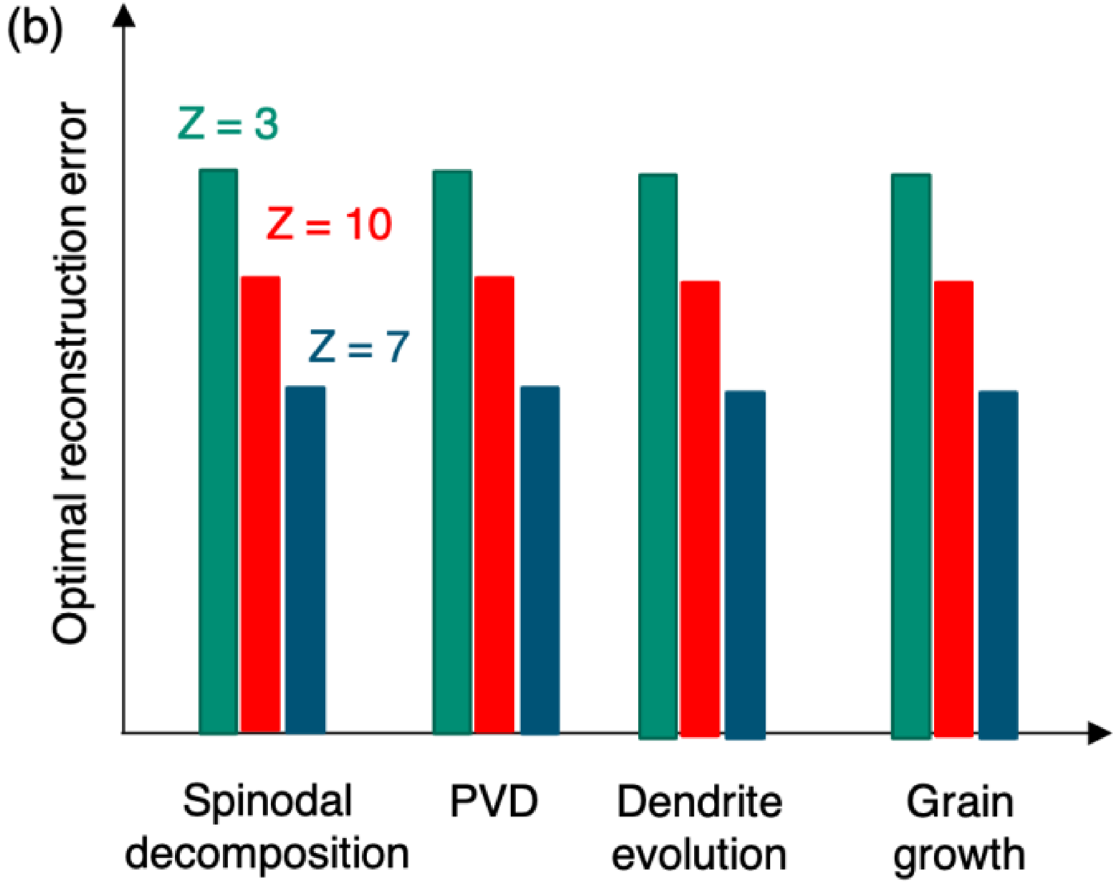
Autoencoders for the PVD dataset



Autoencoders show good reconstruction for PVD data

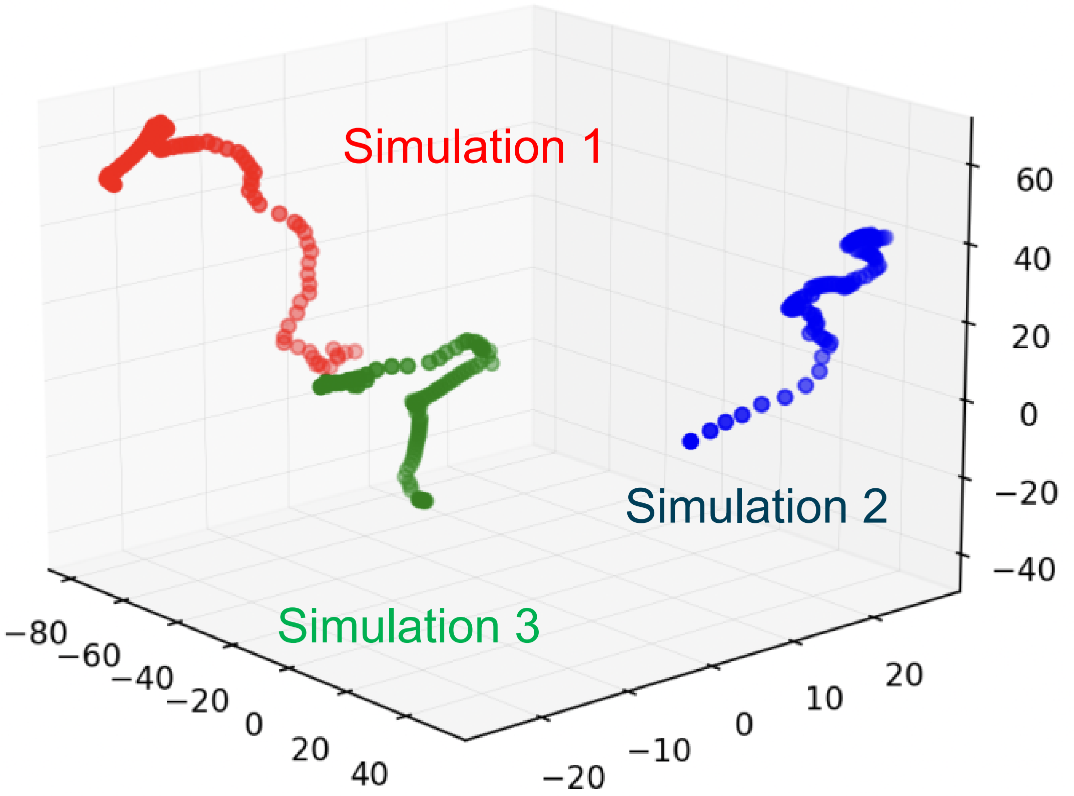
Reconstruction summary





Time evolution in latent space

PCA latent dimension evolution with time



- Is the evolution smooth?
  - Is the evolution linear/non-linear?

- Do all microstructure evolutions have similar latent space evolutions?
  - Do similar processing conditions have similar latent space evolutions?

Time evolution in latent space

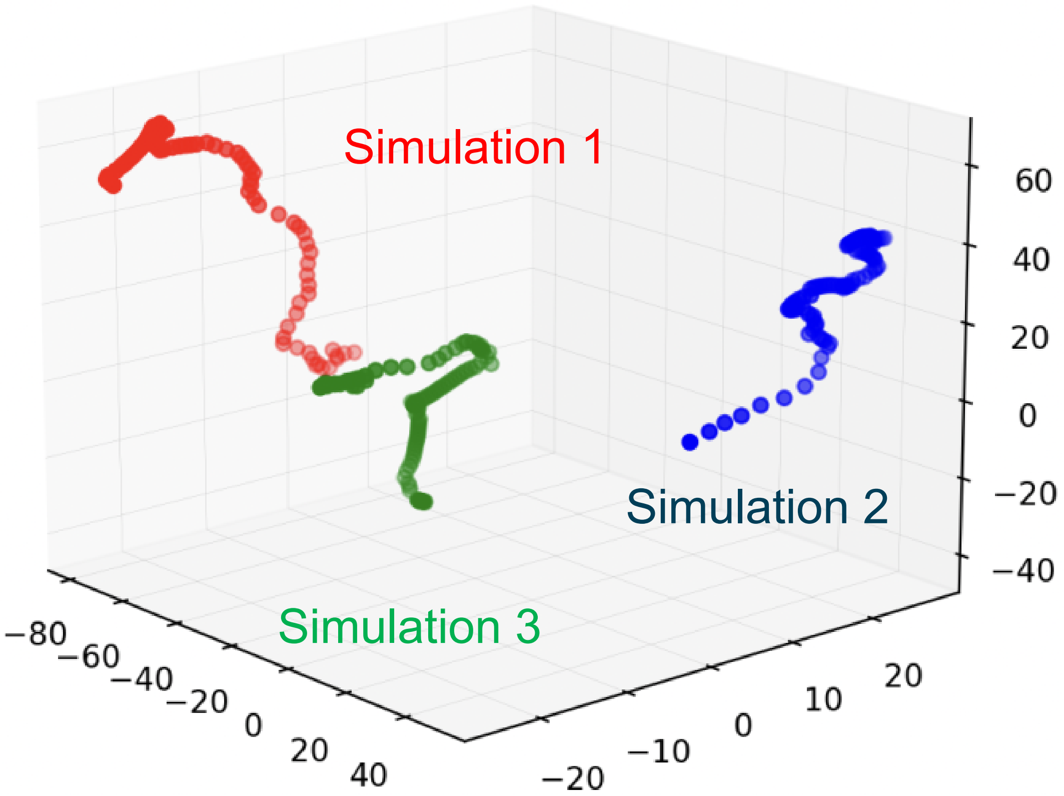
PCA latent dimension evolution with time

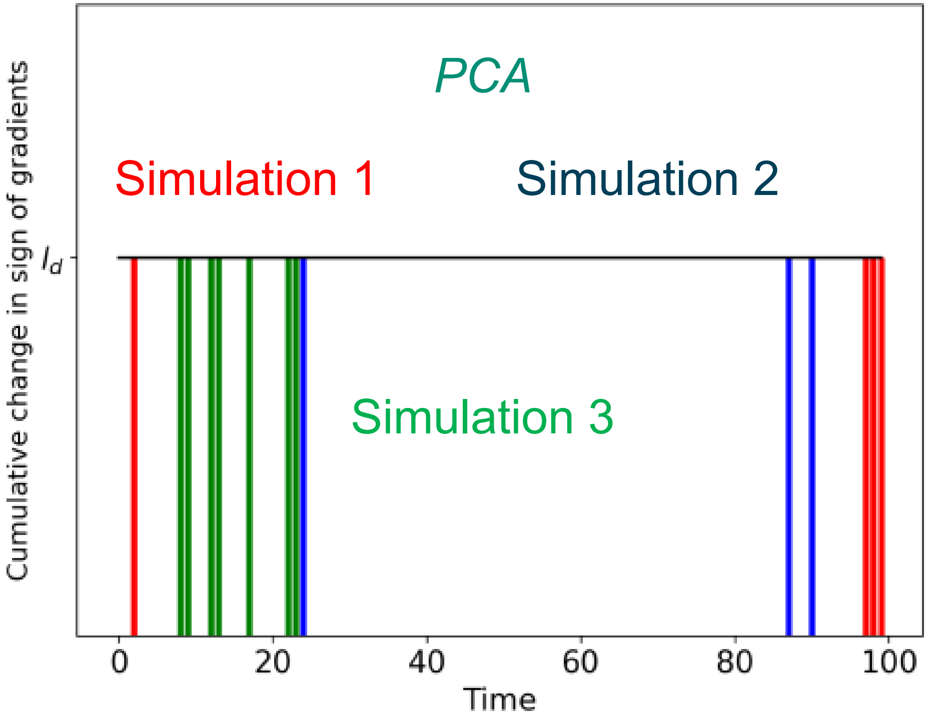
$$C(t) = \big(z_1(t), z_2(t), z_3(t)\big)$$

$C'(t) \neq \vec{0}$

smoothness condition

All derivatives change sign between two points

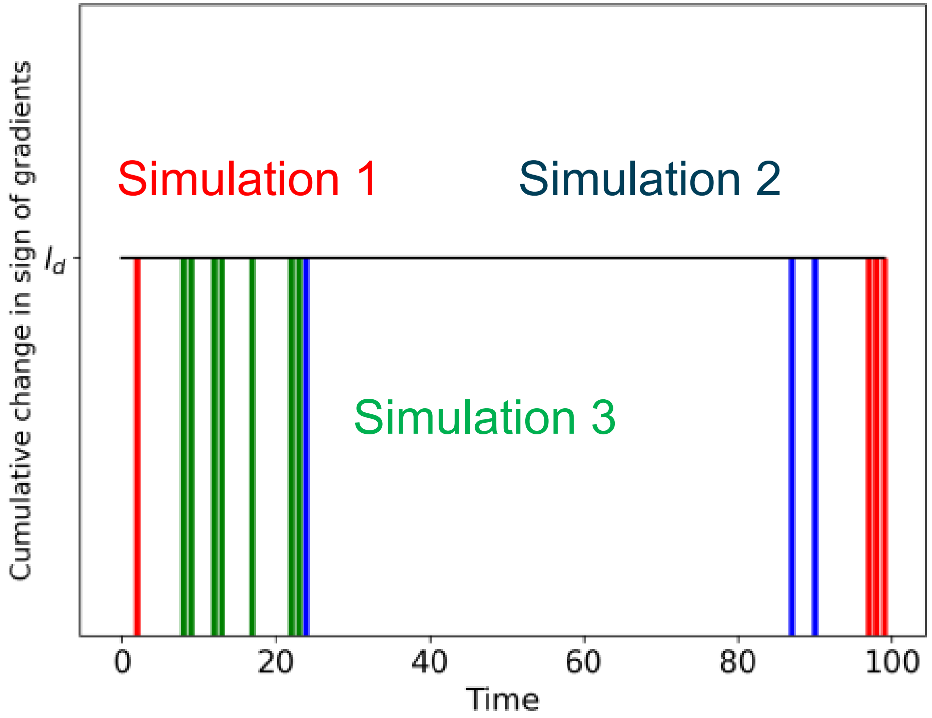




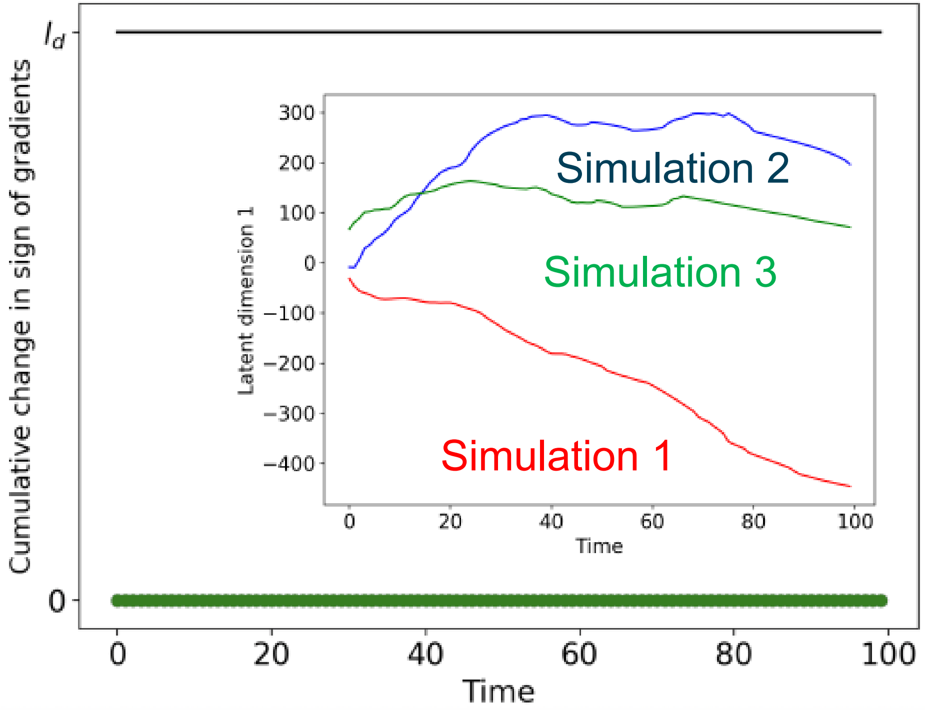
- ➢ Non-Smooth/non-linear evolution with time
- ➢ Similar processing conditions do not have similar latent space evolutions

Time evolution in latent space

PCA latent dimension evolution with time



Autoencoder latent dimension evolution with time

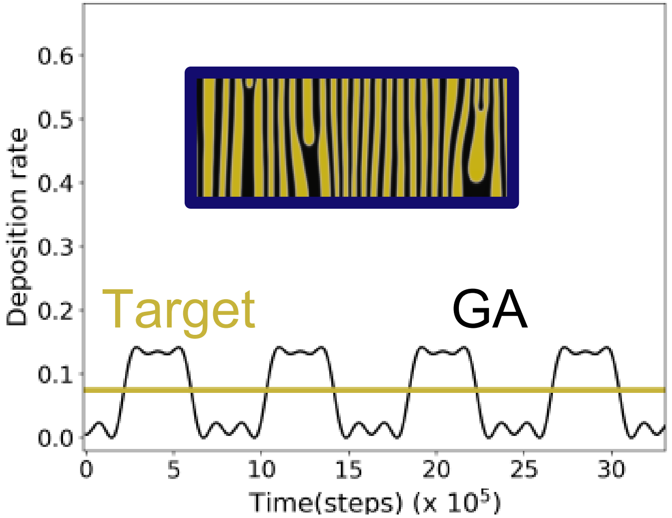


- Non-Smooth/non-linear evolution with time
- Similar processing conditions do not have similar latent space evolutions

- Smooth/non-linear evolution with time
- Similar processing conditions do not have similar latent space evolutions

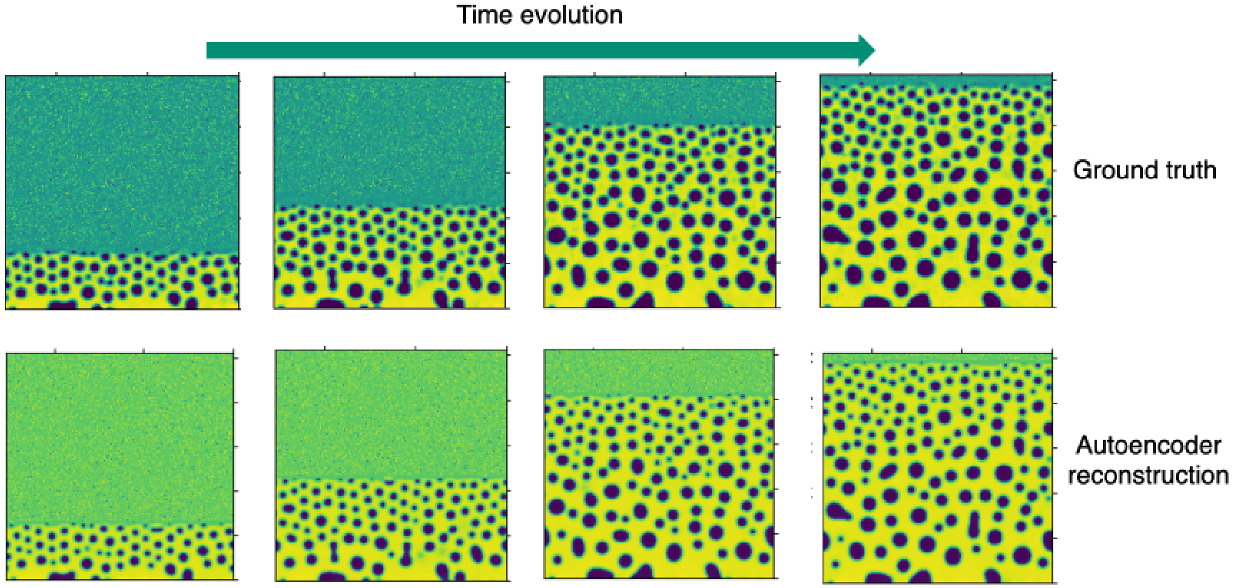
Summary

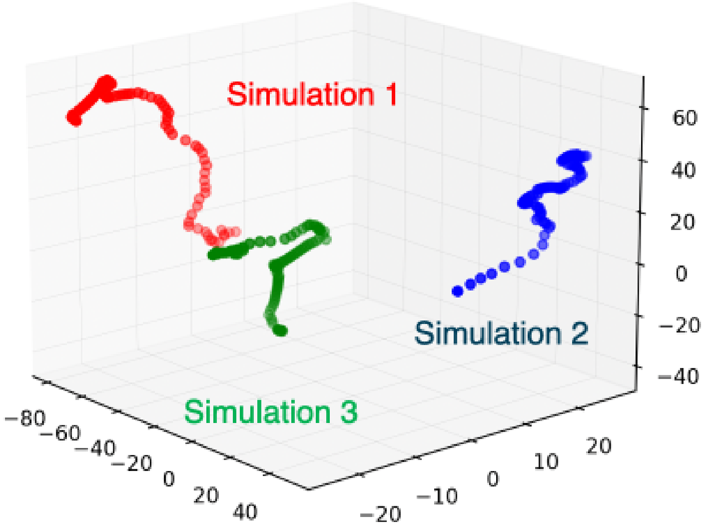
- Genetic algorithm guided PVD protocols suggest alternatives to human-intuition based deposition protocols



- Generative models to generate phase field microstructures show a promising start

- Dimensionality reduction methods show good reconstruction, latent dimension representations vary in trajectory smoothness and linearity





Backup slides

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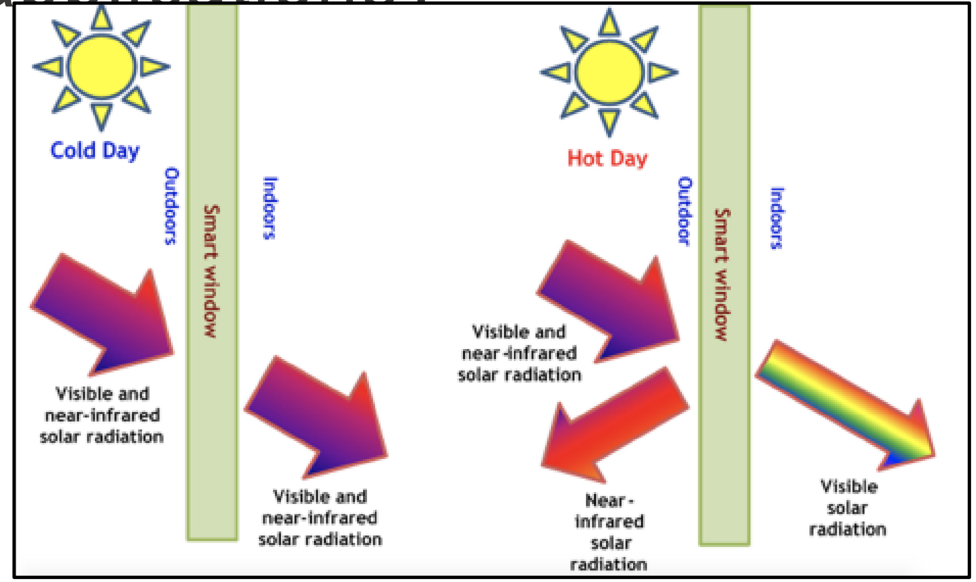
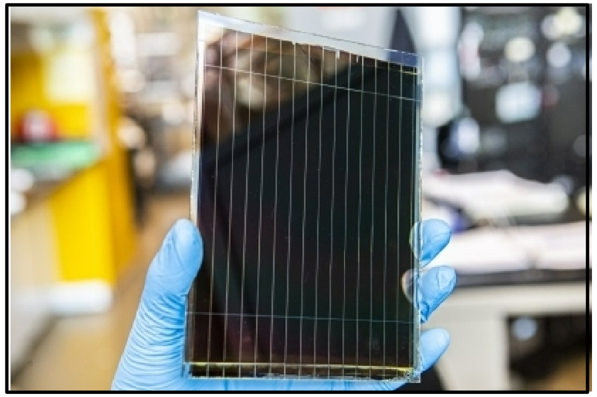
How do we design thin films tailored for specific applications?



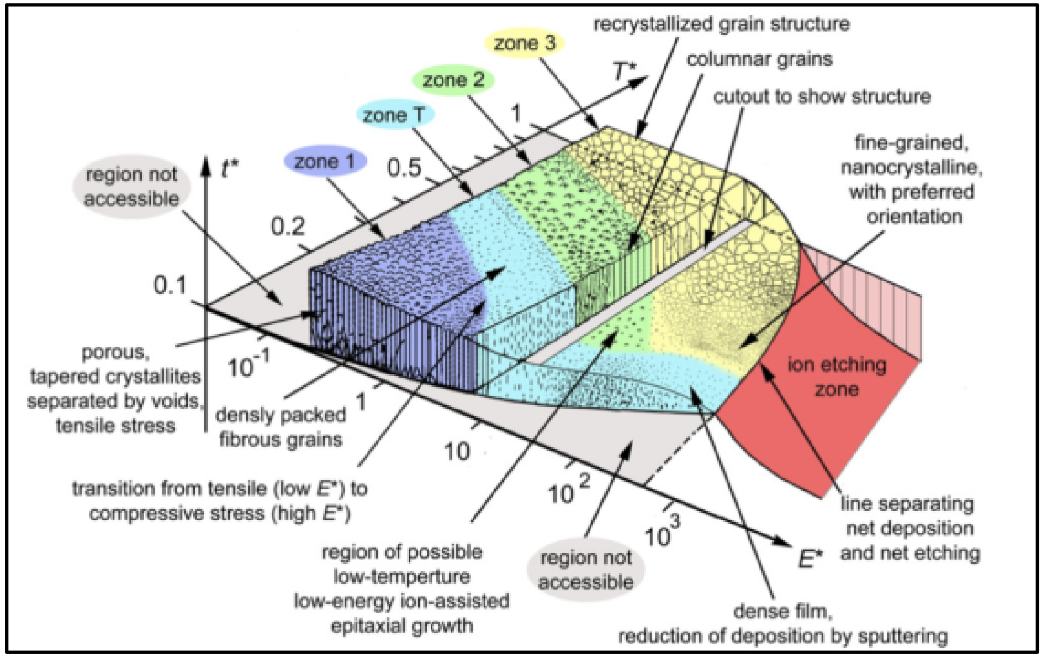
Source: nist.gov



Source: energy.gov

Source: certechinc.com

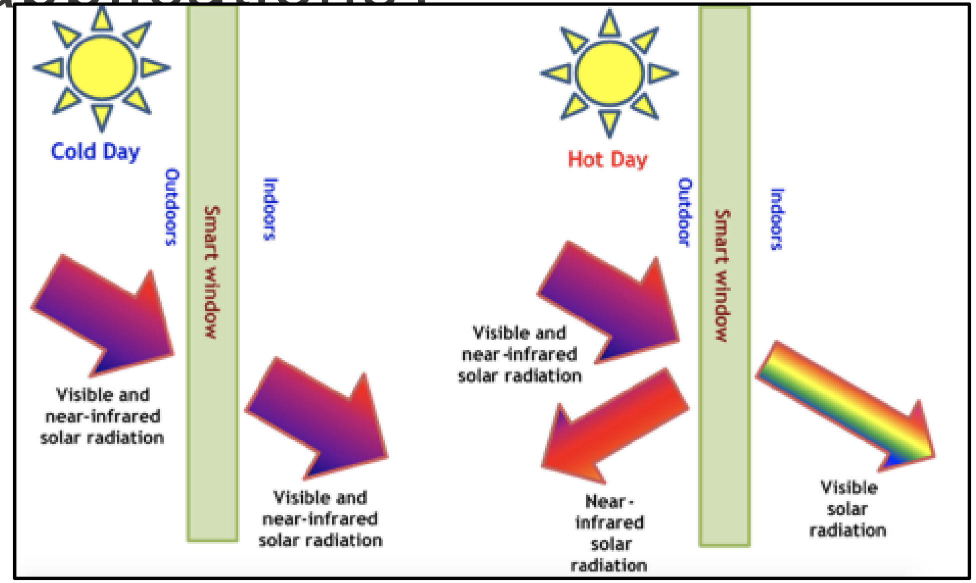
Designing tailor-made thin films requires an understanding of processing-structure-property linkage



Anders *Thin Solid Films* (2010)

Structure zone diagrams relate processing conditions to microstructure

How do we design thin films tailored for specific applications?



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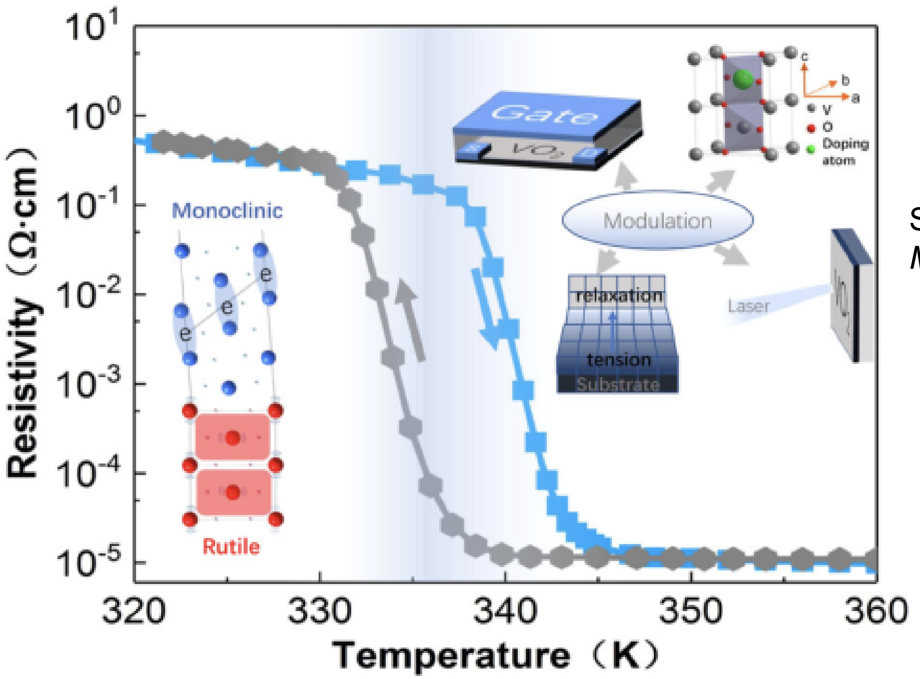
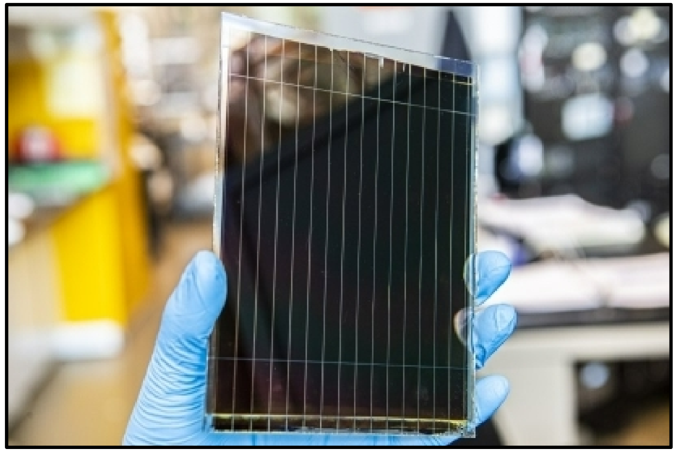


Shao et al. *NPG Asia Materials* (2018)



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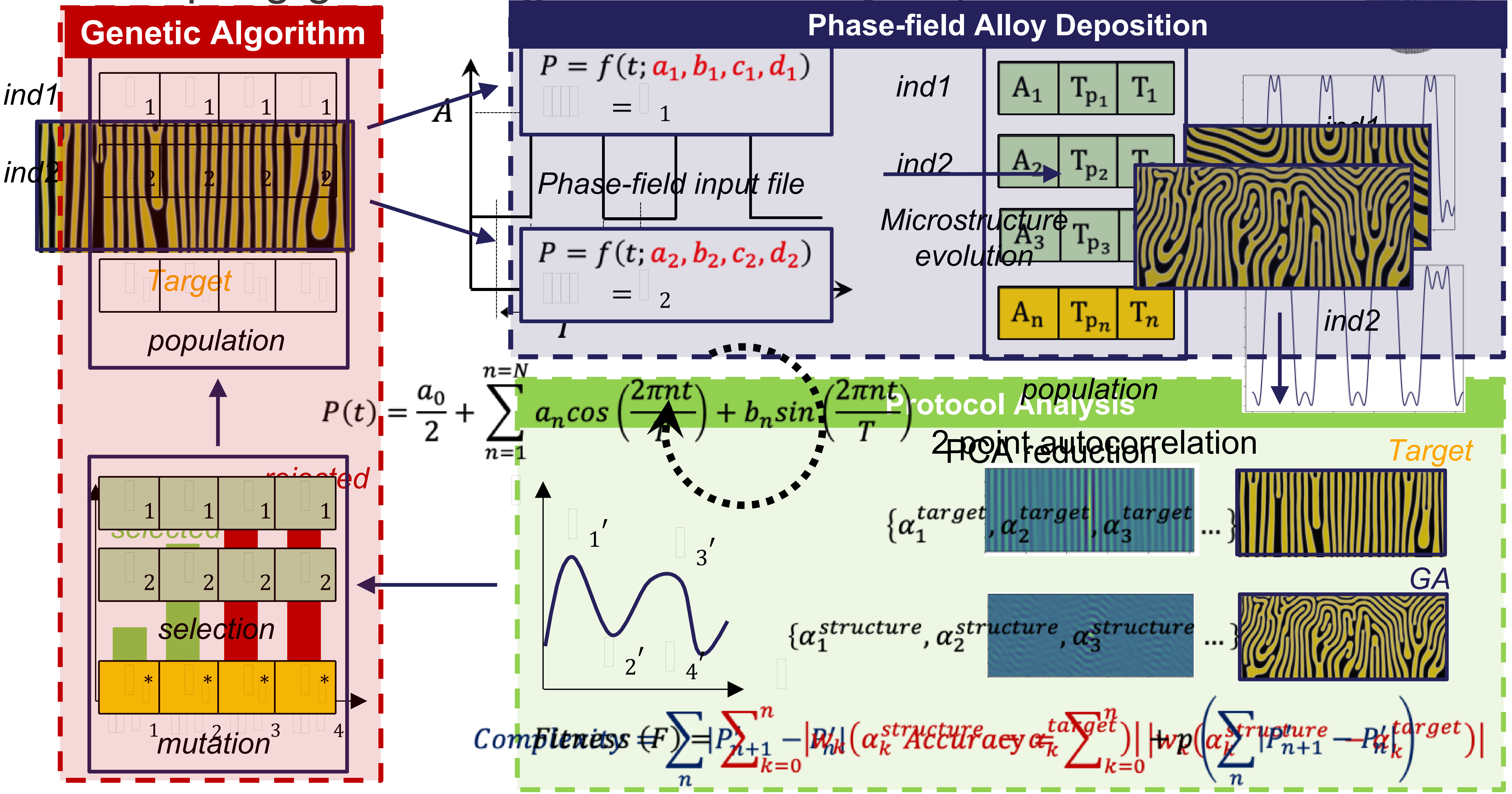
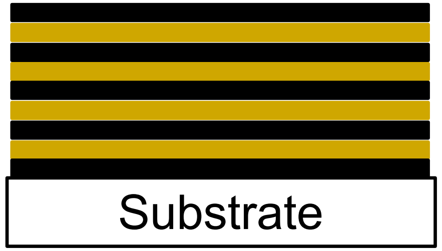
Designing tailor-made thin films requires an understanding of processing-structure-property linkage

Coupling genetic algorithms to alloy deposition



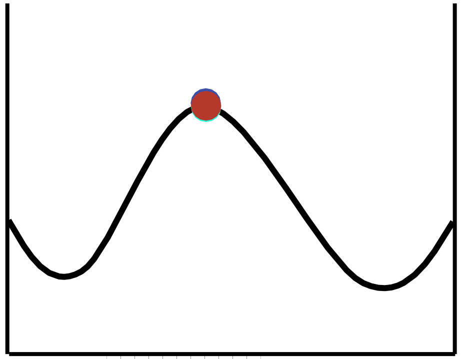
Microstructure formation in metallic alloy thin films

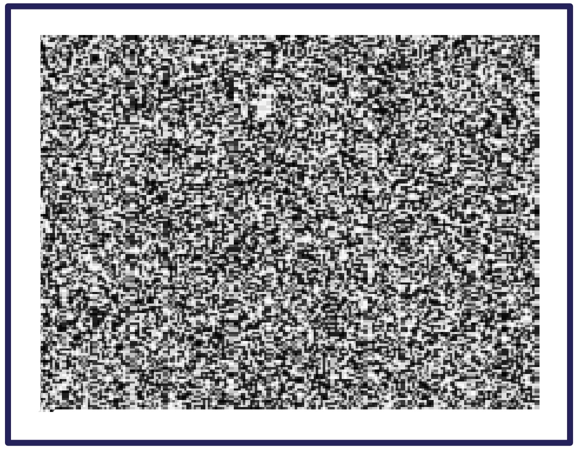
What processing conditions to use to obtain desired film microstructure?



Layer by layer deposition

Self-assembly via spinodal decomposition





Source: [wikipedia]

PVD experiments

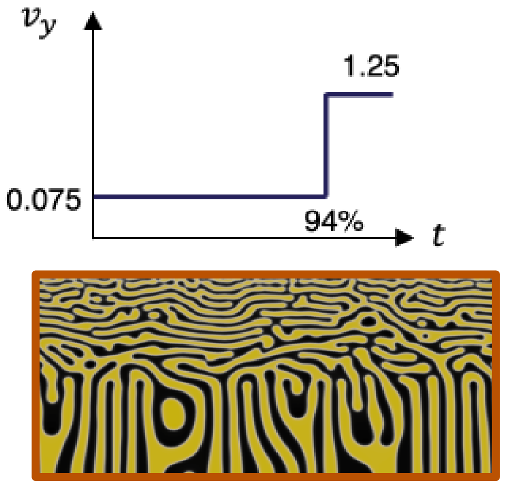
Phase field simulations

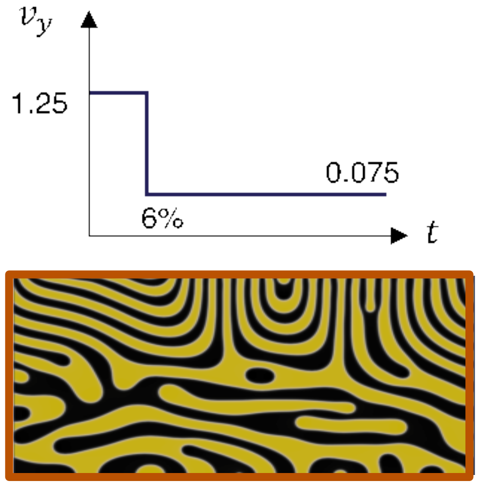
Spinodal decomposition results in spontaneous concentration modulations

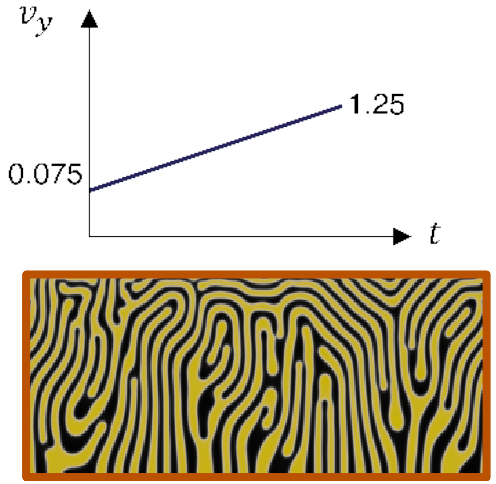
How do we design PVD-grown thin film microstructures?

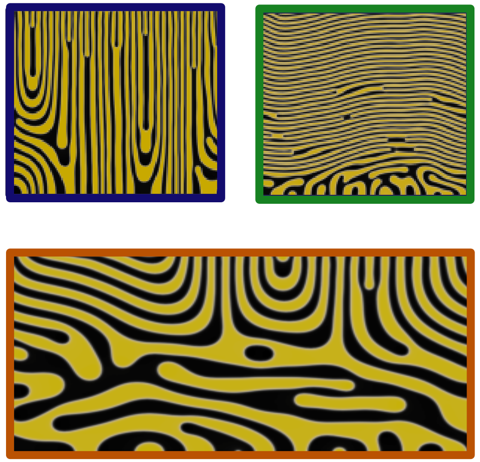
What processing conditions to use to obtain desired film microstructure?

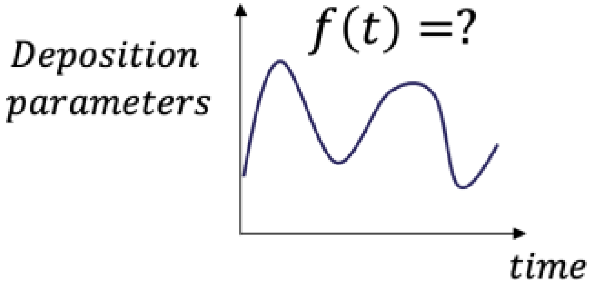
Current SZDs only consider protocols that are constant in time





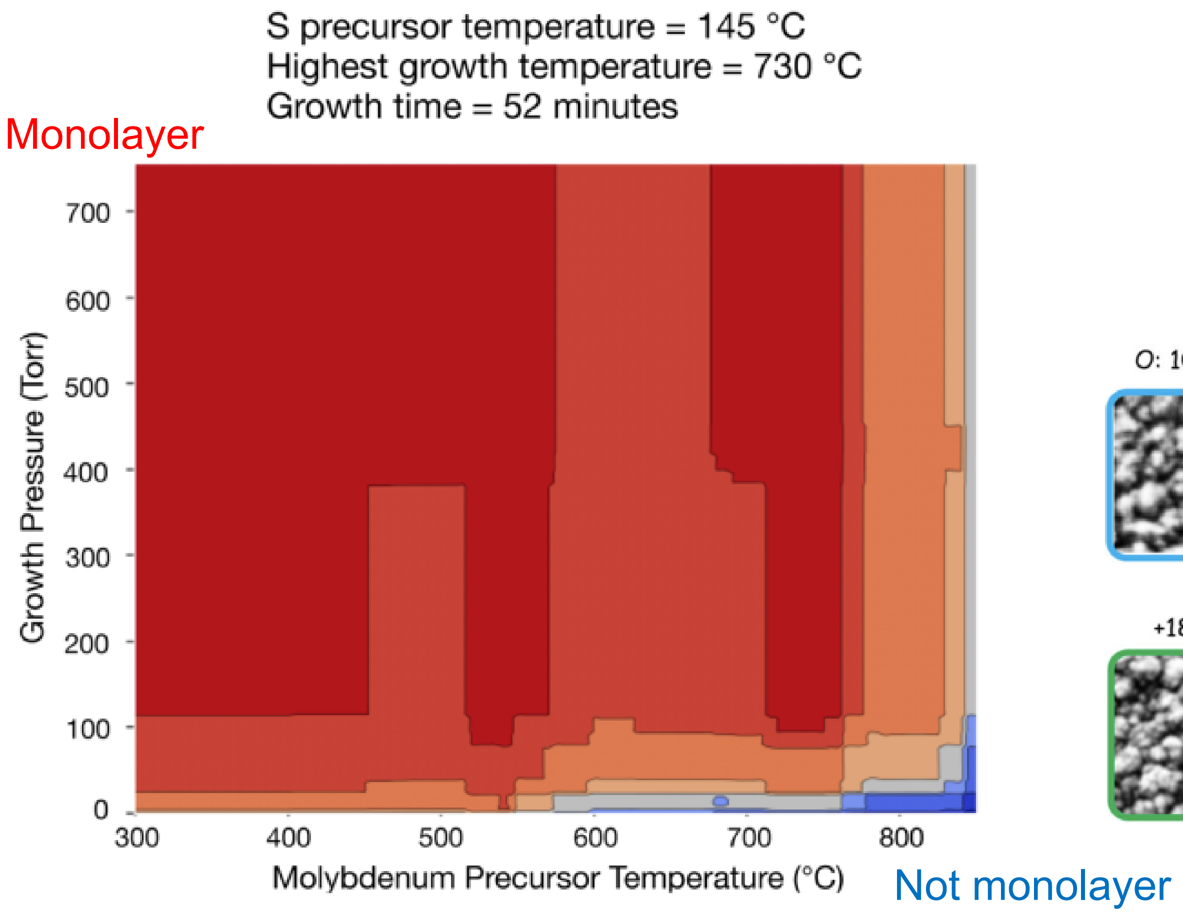






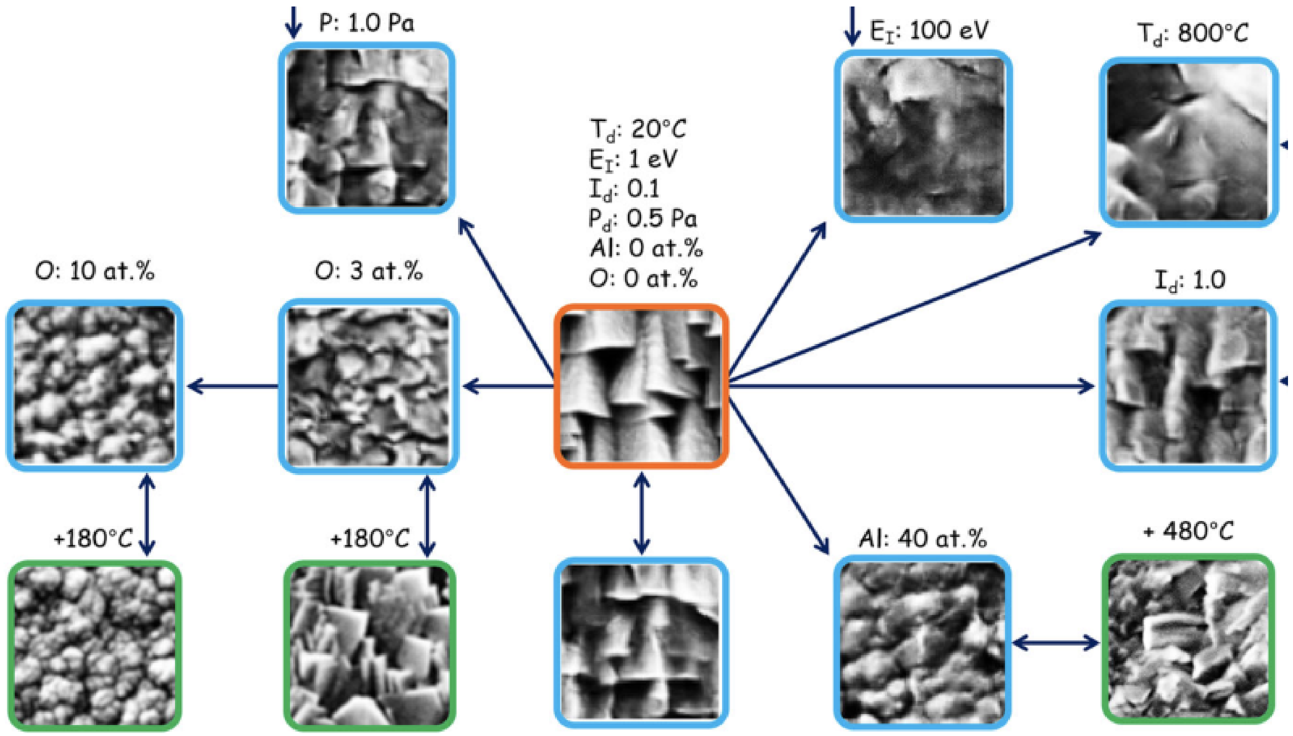
We use a genetic algorithm to discover time-dependent protocols that result in desired microstructure

High dimensional structure zone diagrams



Costine et al. *Journal of Applied Physics* (2020)

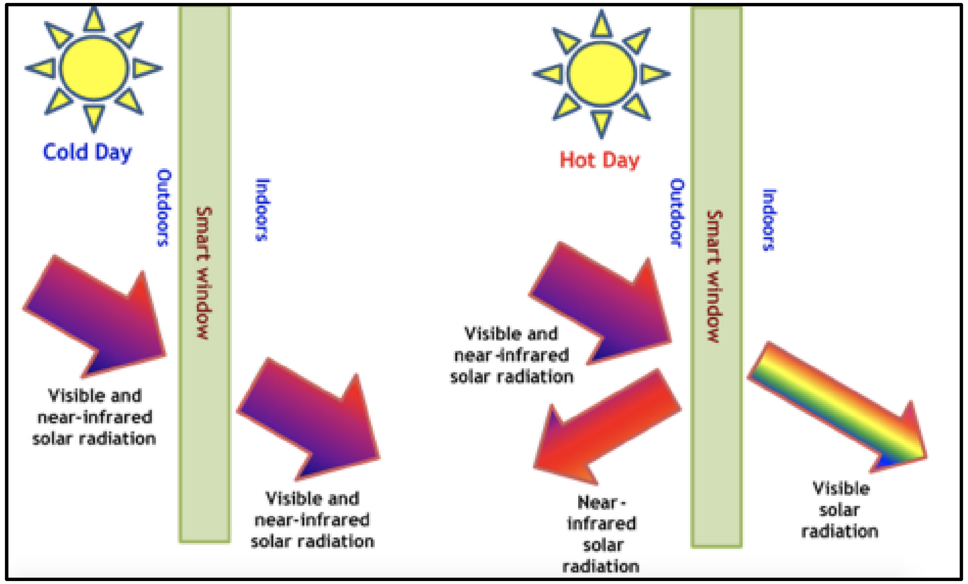
Generative Adversarial Network based SZD



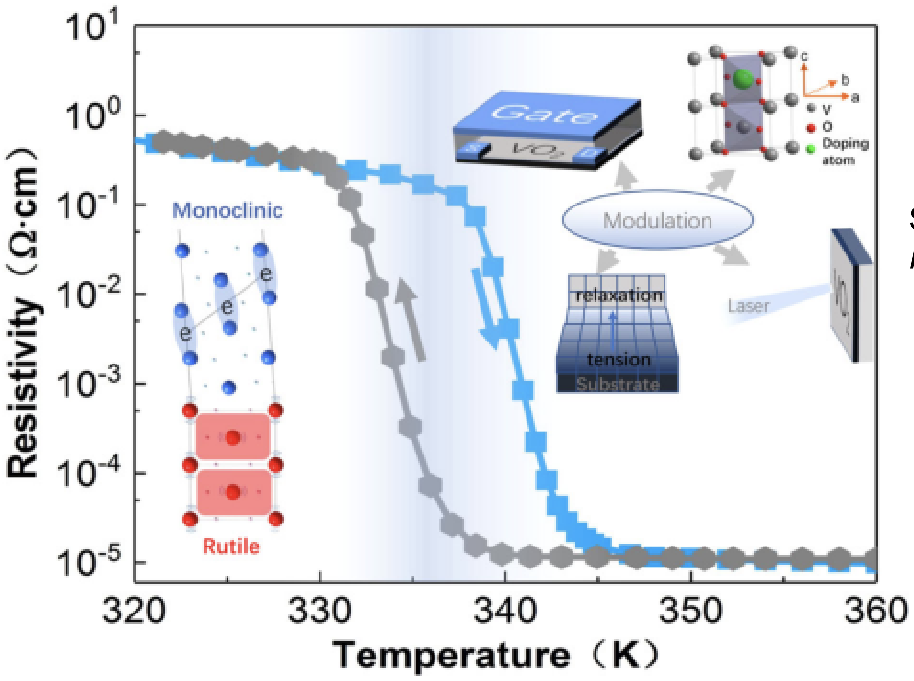
Banko et al. *Communications Materials* (2020)

ML methods can give high dimensional SZDs

Structure-property-processing relationships in thin films



Source: nist.gov



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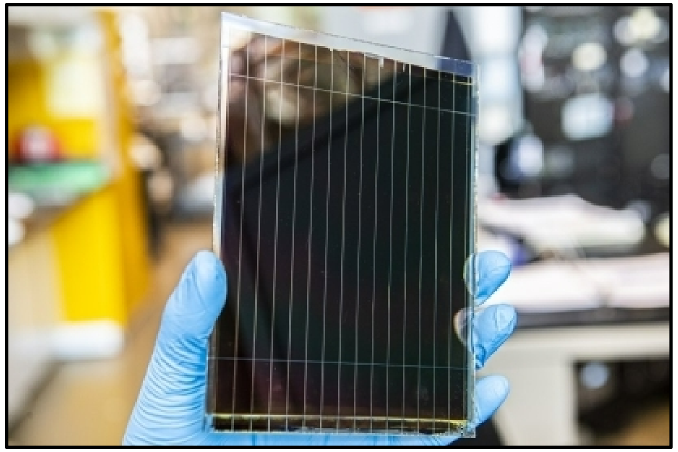


Source: energy.gov

Source: renata.com

Thin film structure decides properties

Phase field simulations of alloy deposition

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

Free energy of system

$$\frac{\partial c}{\partial t} = \nabla.\left[M_c(\phi, c)\,\nabla\frac{\delta F}{\delta c}\right]$$

Evolution equation

$$M_c(\phi, c) = M^{bulk} + M^{surf}$$

Surface mobility dominates microstructure

$$M^{bulk} = \frac{1}{4}(2-\phi)(1+\phi)^2\left[h(c)M_A^{bulk} + (1-h(c))M_B^{bulk}\right]$$

$$M^{surf} = e^{-\left(\frac{\phi}{\sigma^{surf}}\right)^2}\left[h(c)M_A^{surf} + (1-h(c))M_B^{surf} - M^{bulk}\right]$$

$$\frac{\partial \phi}{\partial t} = \nabla.\left[M(\phi)\,\nabla\frac{\delta F}{\delta \phi}\right] + S(n(\phi))$$

Evolution equation with source term

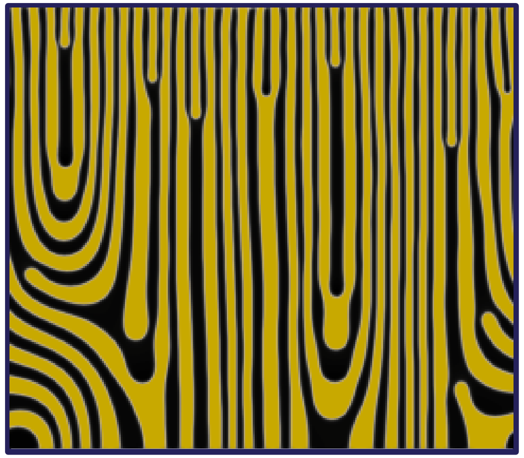
$$\frac{\partial \rho}{\partial t} = \nabla.\left[D_\rho\,\nabla\rho\right] - \nabla.\left[\rho\, v\right] - S(n(\phi))$$

Vapor evolution equation

$\boxed{\text{Phase field model simulates microstructure evolution for various deposition conditions}}$

Microstructures using constant deposition conditions

Deposition rate = 0.075



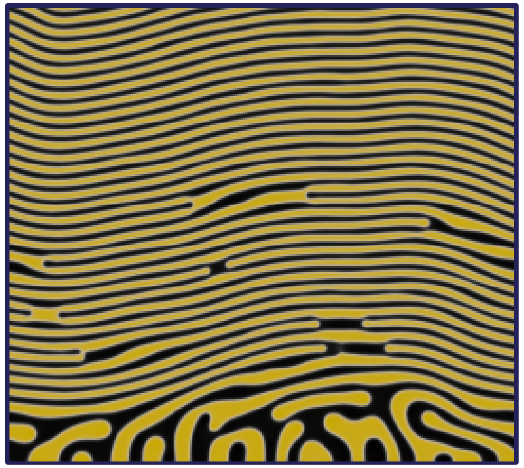
Lateral concentration modulation (LCM)

Deposition rate = 1.25

Random concentration modulation (RCM)

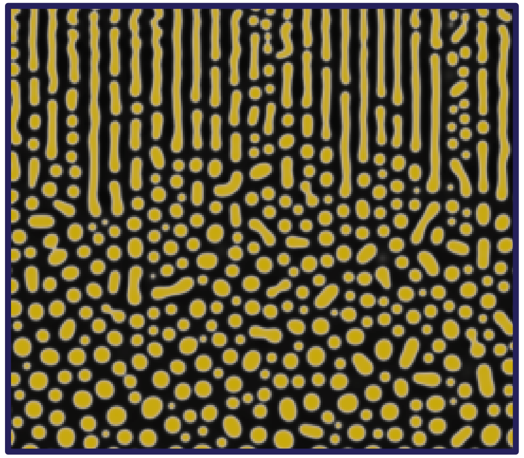
Competition between deposition and diffusion gives different microstructure regimes

Deposition rate = 0.5



Vertical concentration modulation (VCM)

Deposition rate = 1.25

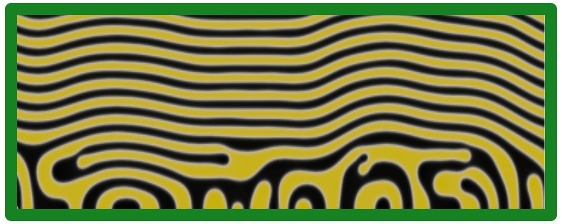


Nanoprecipitate concentration modulation (NPCM)

$A_{65}B_{35}$

Discovering time-dependent protocols

Vertical concentration modulation (VCM)



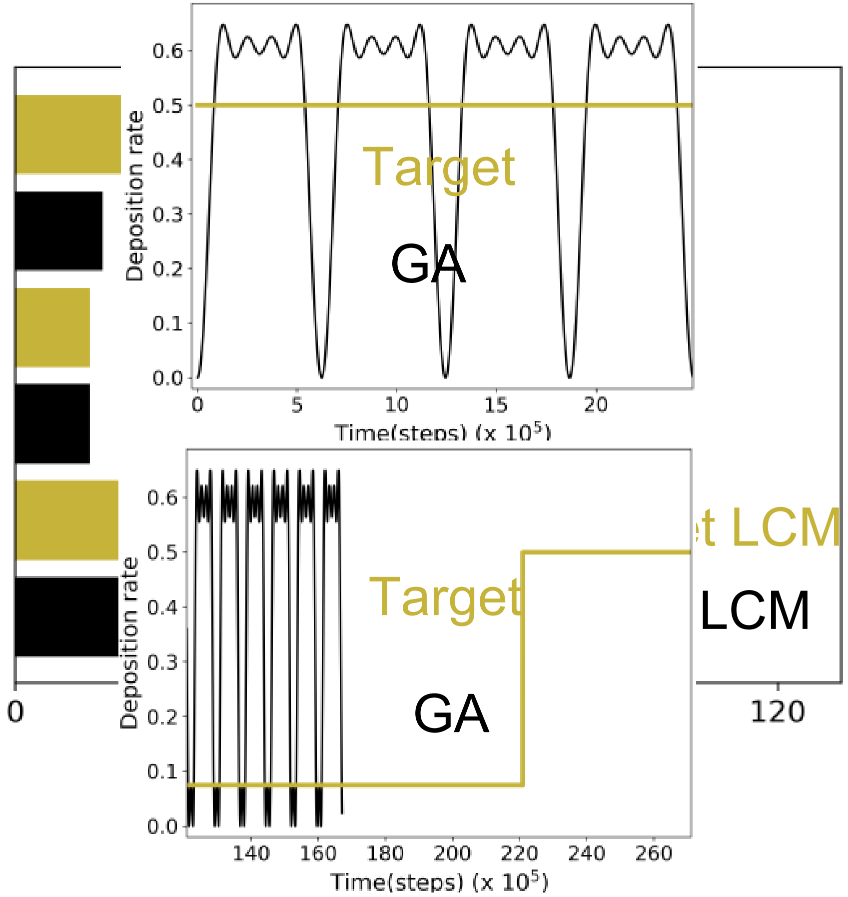
Target

GA

Hierarchical microstructure (HCM)

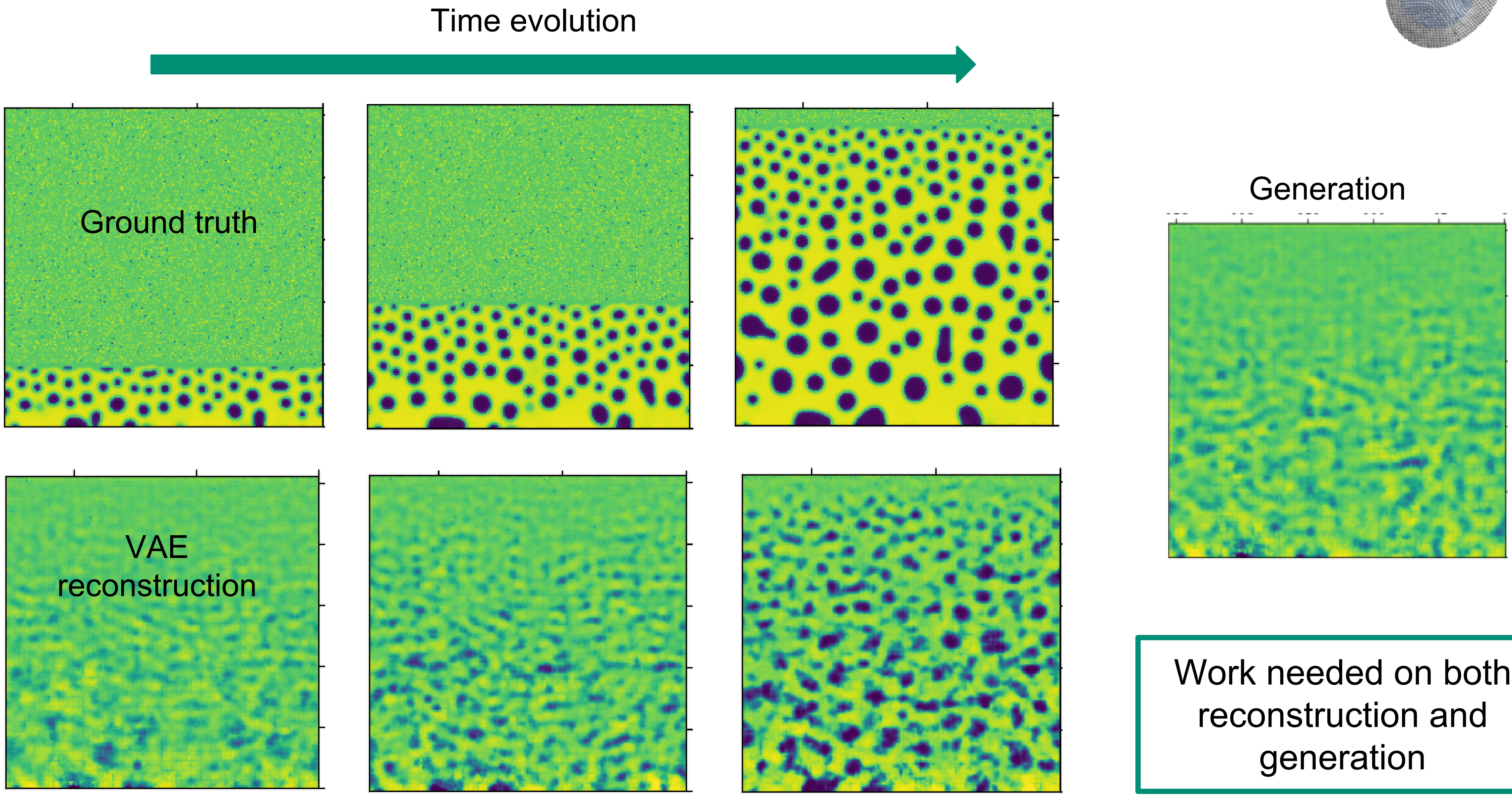
Target

GA



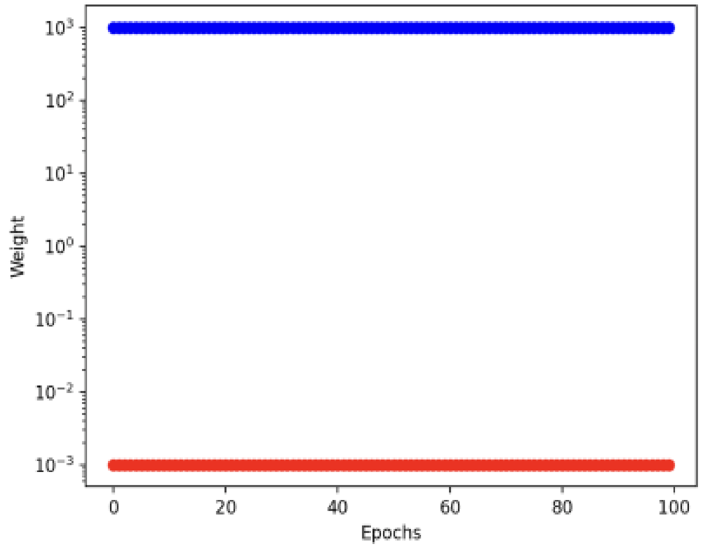
- ➢ Genetic algorithm discovers pulse protocol that results in target structure
- ➢ Time taken to achieve microstructure similar or lower than target protocol

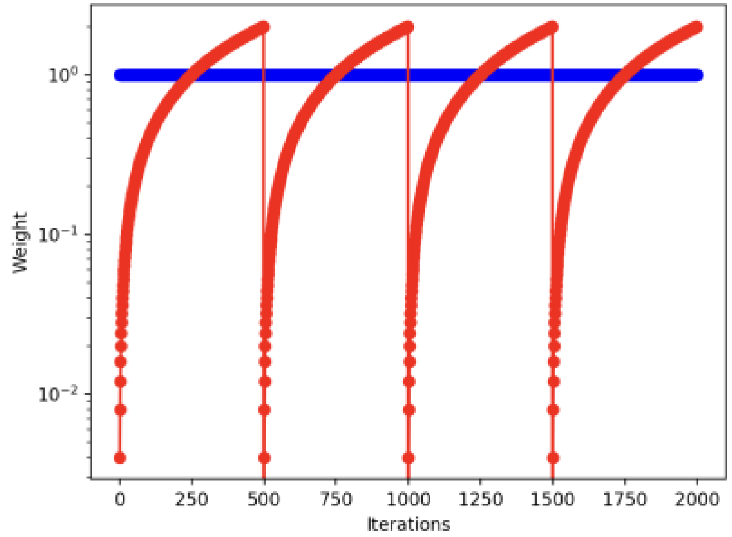
VAE for the PVD dataset



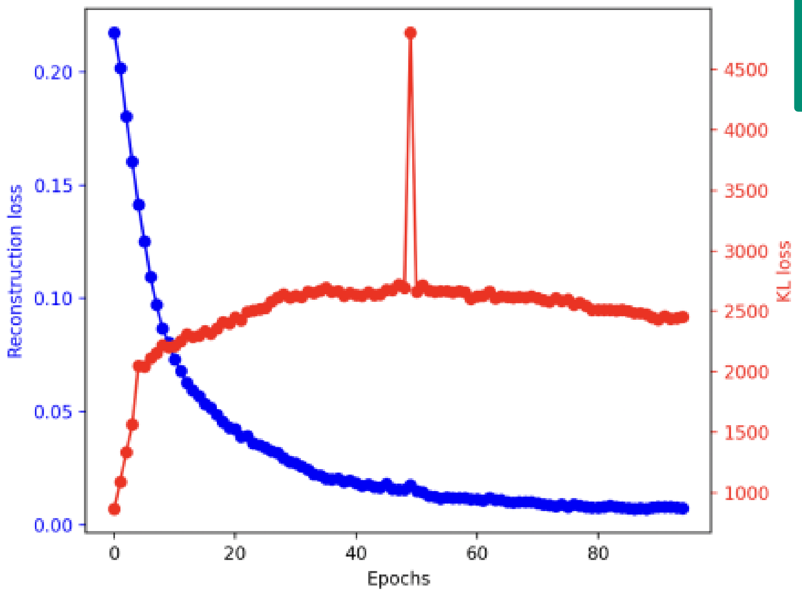
Work needed on both reconstruction and generation

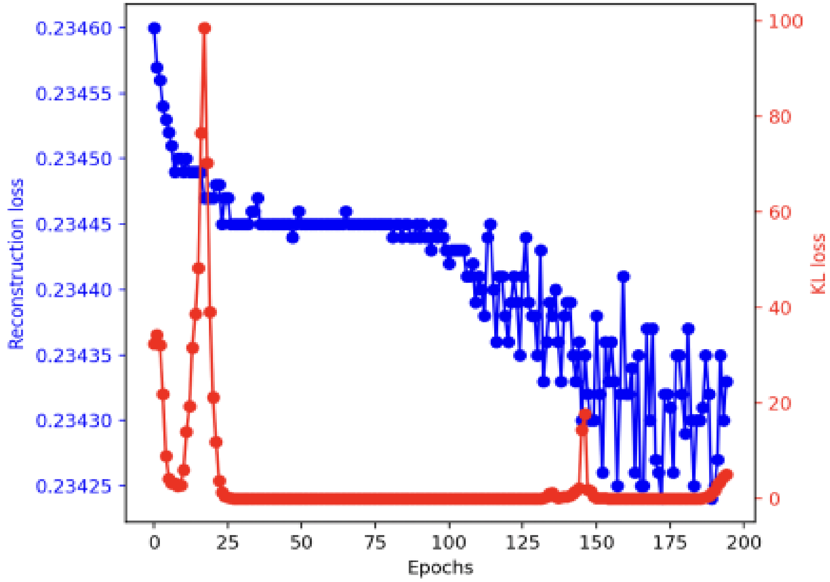
Experiments with the training protocol



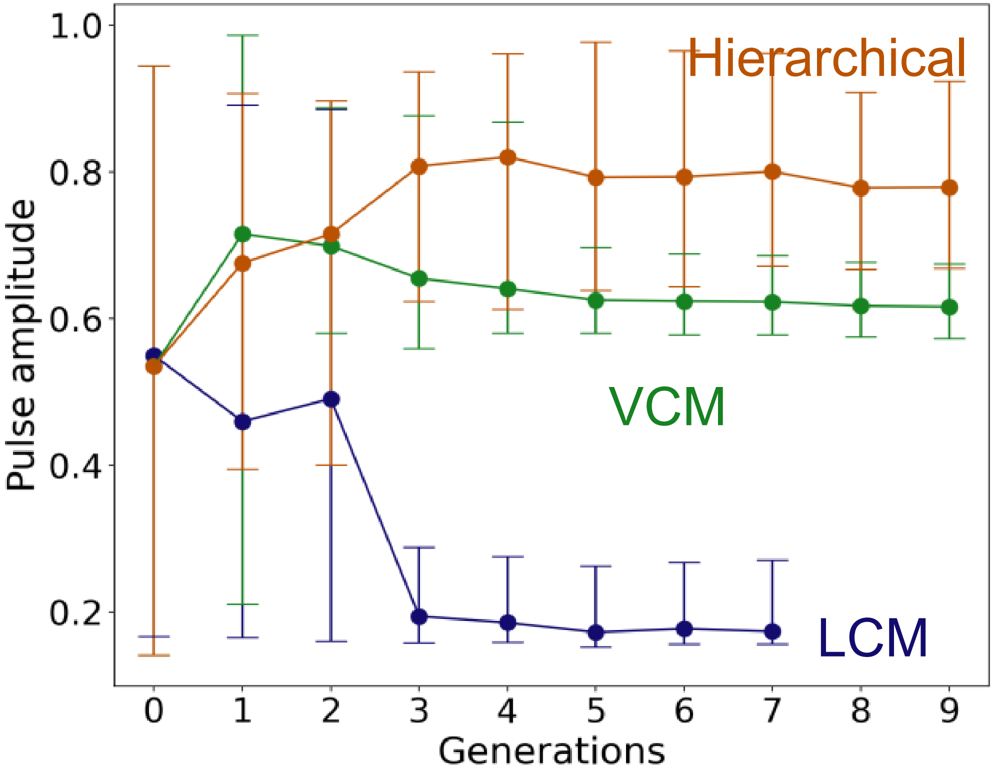


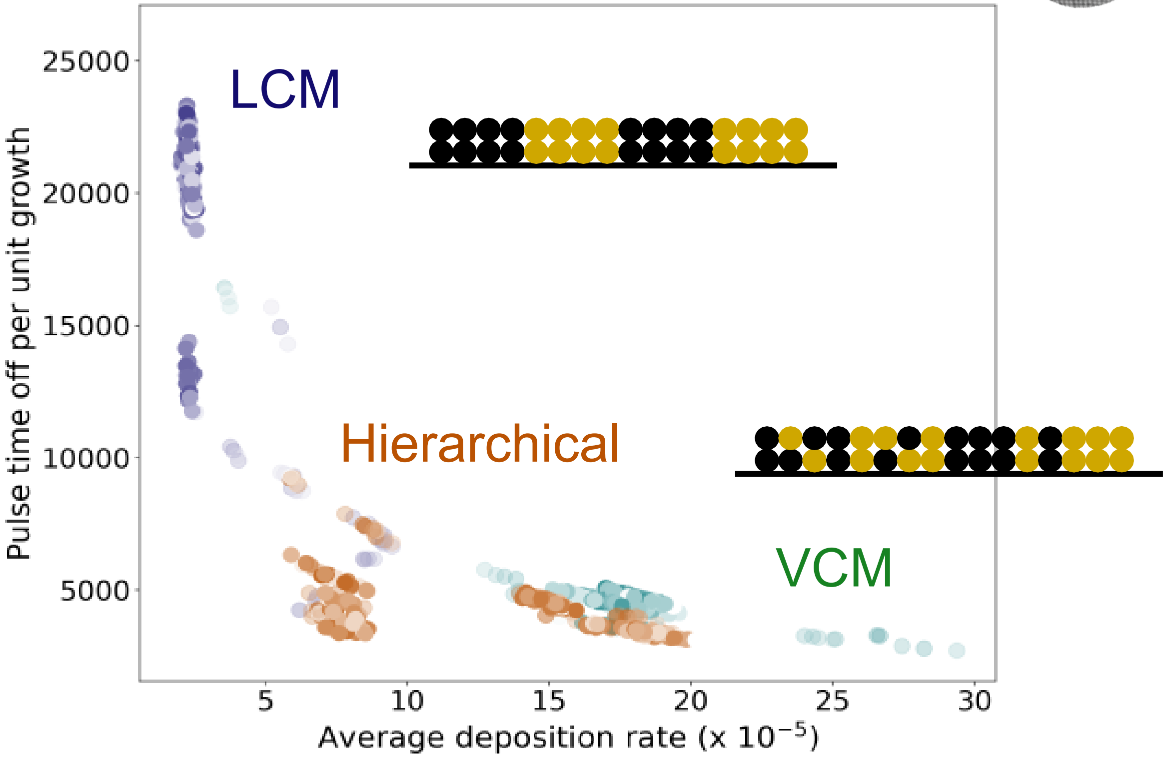
Experimenting with various training protocols to reduce loss





Understanding the choices of the genetic algorithm

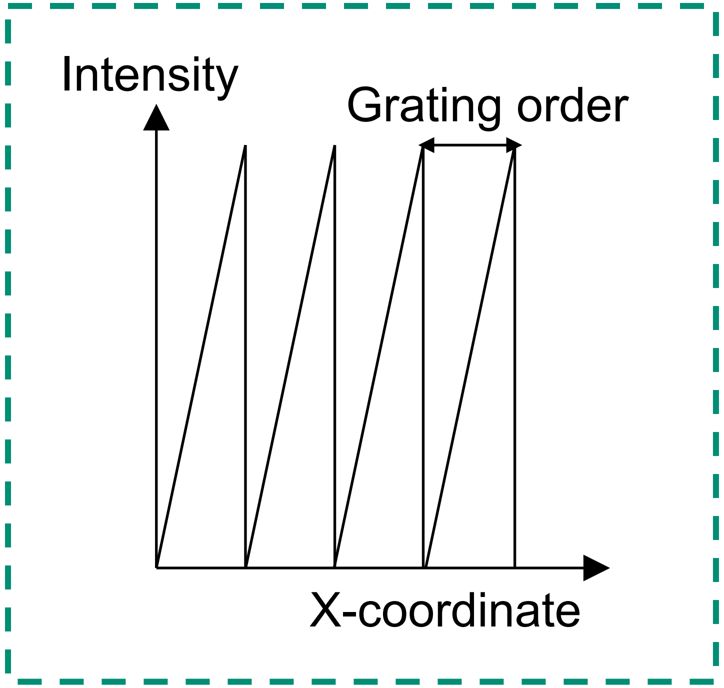




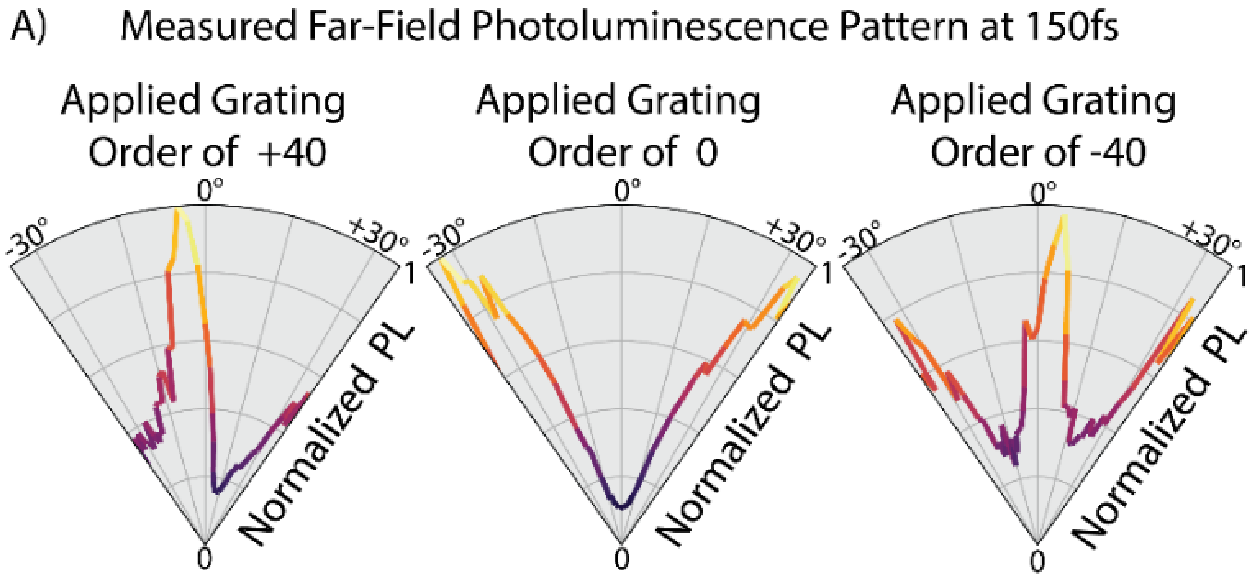
- ➢ GA favors low amplitudes to generate LCM structures and high amplitudes for VCM structures
- ➢ Range of deposition rates can be used to get hierarchical structures
- ➢ Genetic algorithm learns deposition-diffusion trade offs

Solving simpler problems first…

Optimize input optical profiles to steer incoherent light



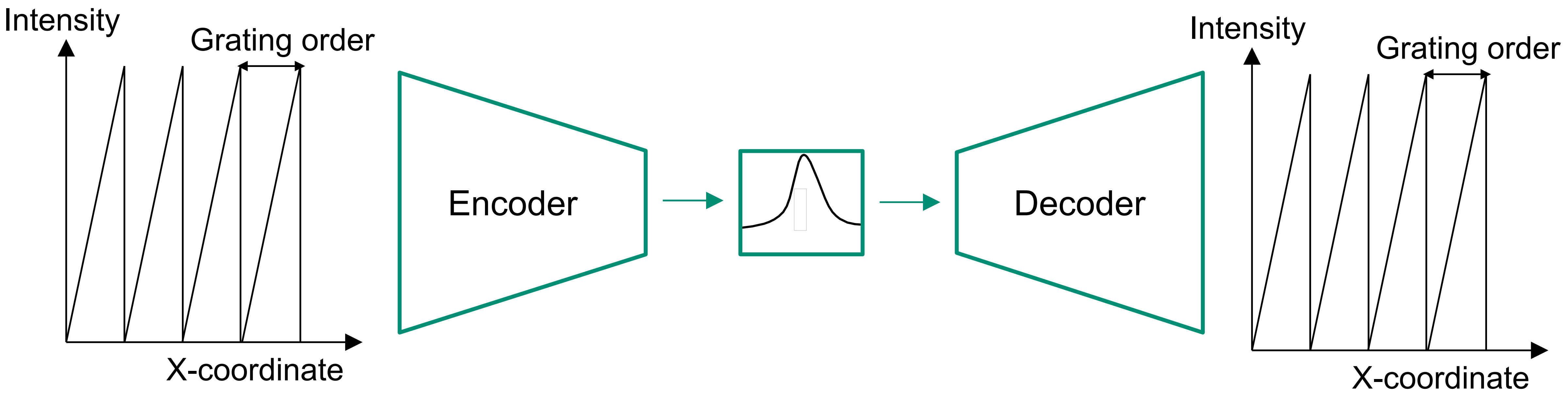
Input optical profile



Output beam steering

Specific input optical profiles result in steering of light

Generating new input profiles



Training data

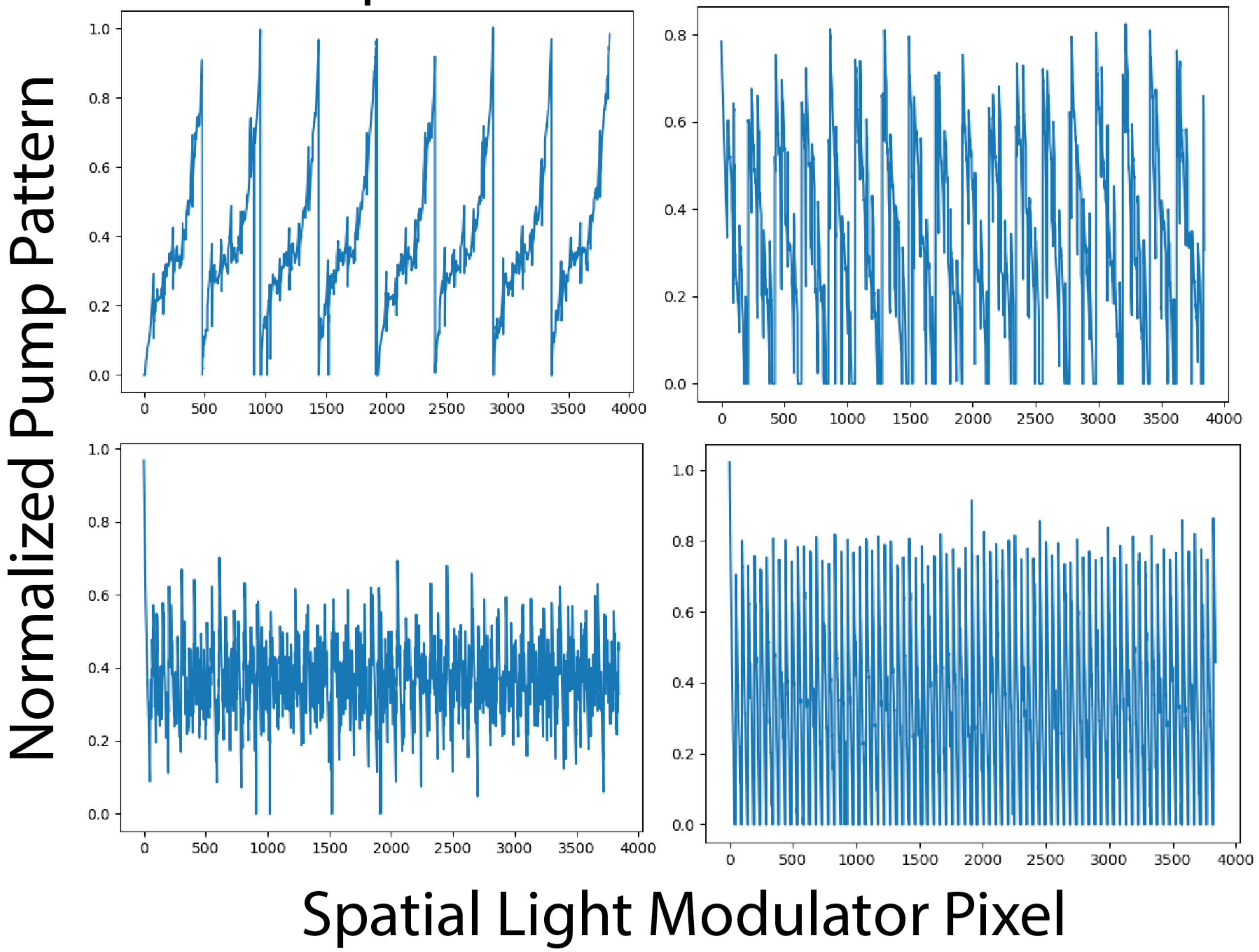
Input: 1D profile – 3840 pixels
Output: 1D profile – 3840 pixels (same as input)
Loss: Reconstruction error (MSE) + KL divergence error (enforce latent space distribution)

Generation (use)

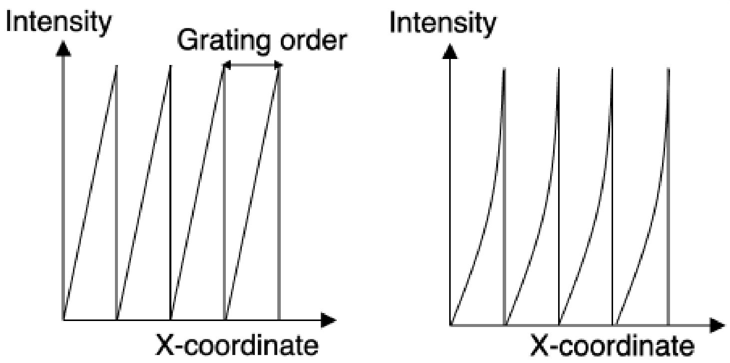
Input: ‘z’ sampled from learnt distribution
Output: 1D profile

Generating new input profiles

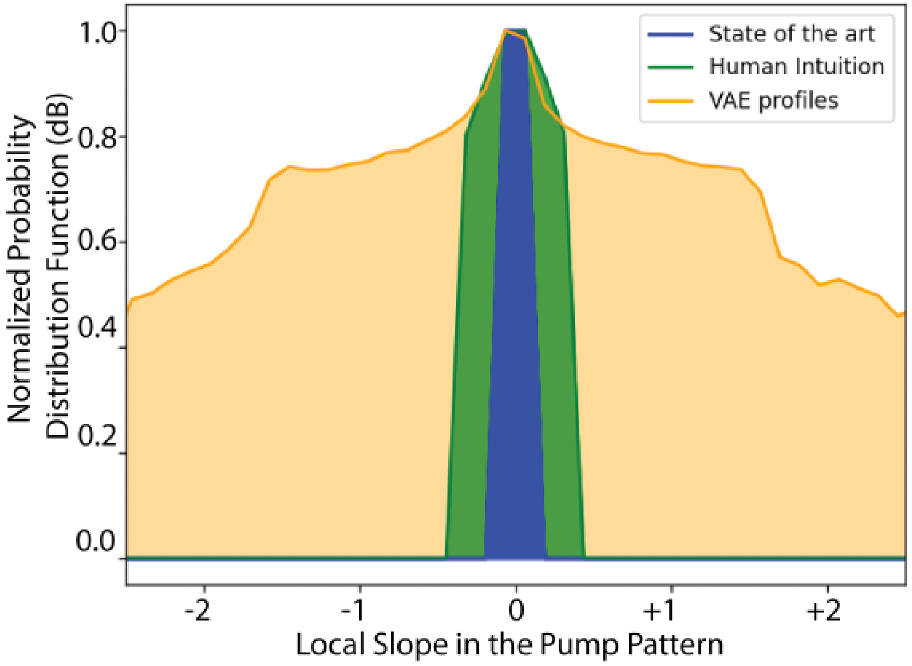
Example VAE Patterns Generated



Training set

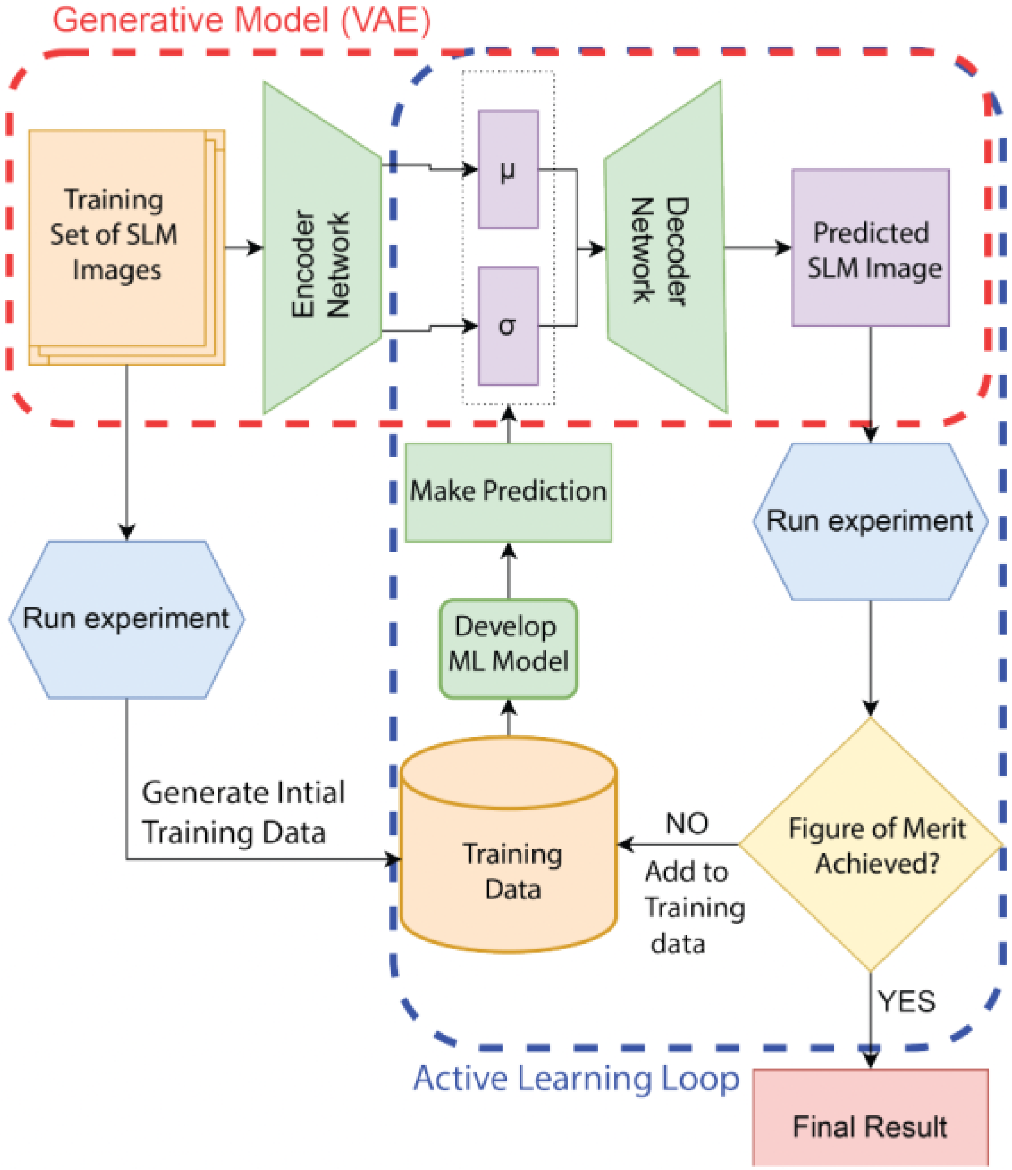


B) Generative Capability of the VAE



VAE generates a variety of input profiles

Finding optimal profiles using VAEs and active learning



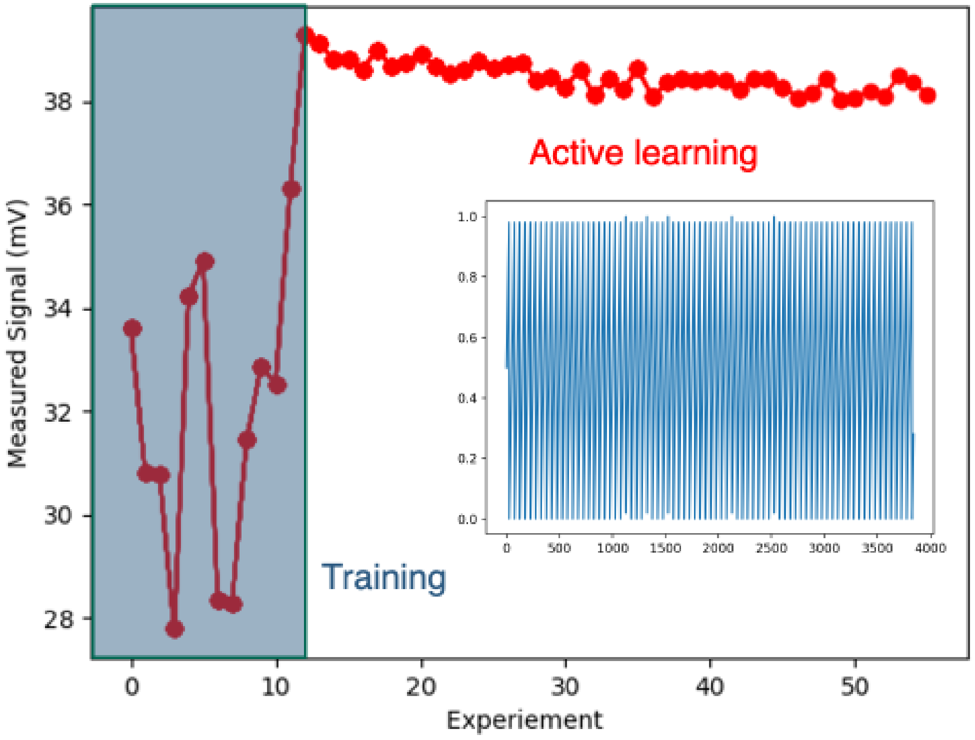
(1) VAE trained to database of input profiles

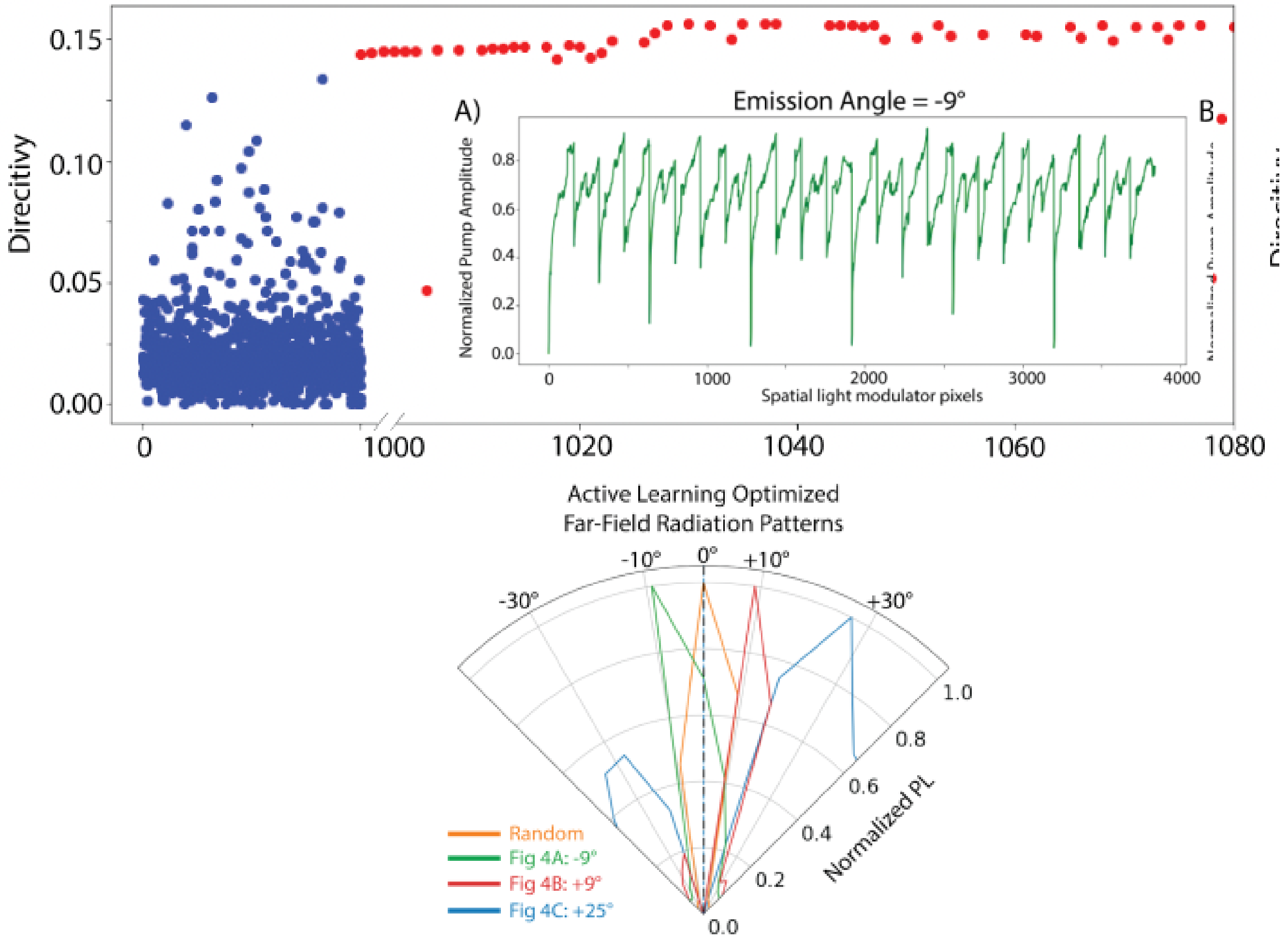
(2) Trained VAE coupled to active learning scheme

(3) Active learning optimizes latent dimension such that figure of merit is achieved

(4) Training data and data explored by active learning coupled to an equation learner to learn underlying physics

Finding optimal profiles using VAEs and active learning





Active learning rediscovers grating order of 80 to have maximum beam steering

Active learning finds multiple profiles beyond human intuition with varying beam steering angles