SAND2022-2200C

Designing thin film microstructures using genetic algorithms

Saaketh Desai, Remi Dingreville

Center for Integrated Nanotechnologies

Sandia National Laboratories

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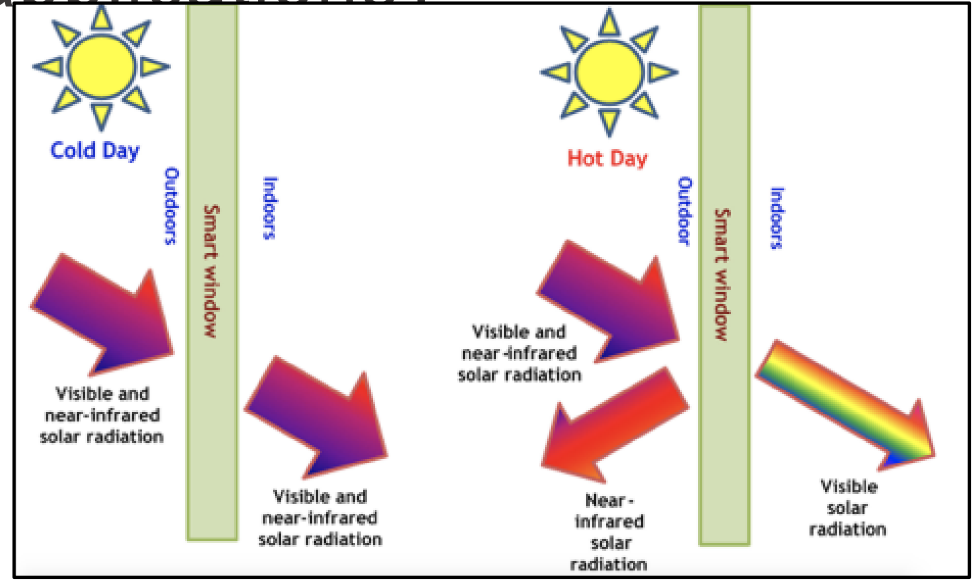
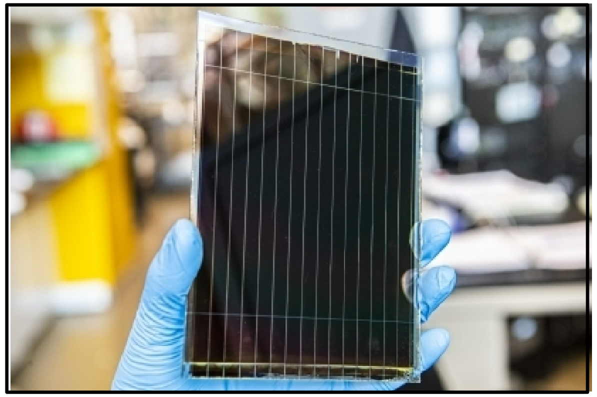
How do we design thin films tailored for specific applications?



Source: nist.gov

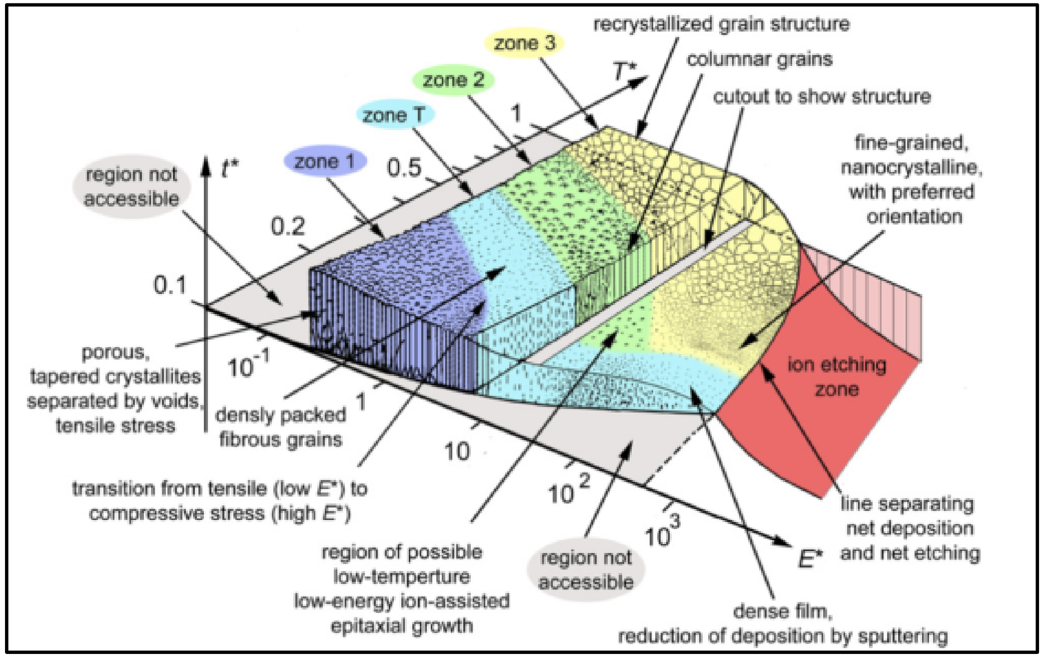


Source: energy.gov

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 metallic alloy thin films

Binary metallic alloy microstructures governed by spinodal decomposition

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

Free energy of system

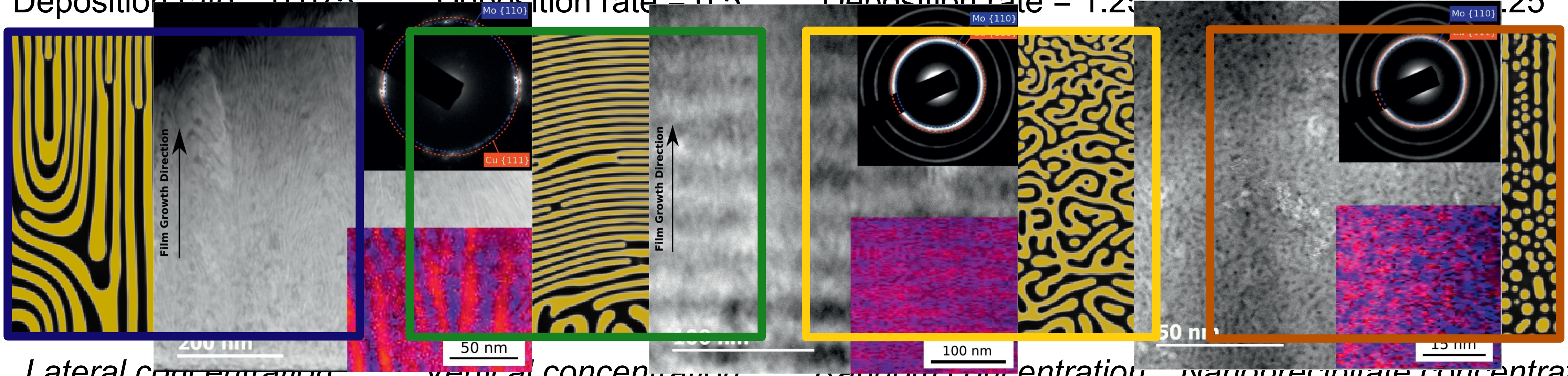
Stewart et al. *Acta Materialia* (2020)

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

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

Evolution equations

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



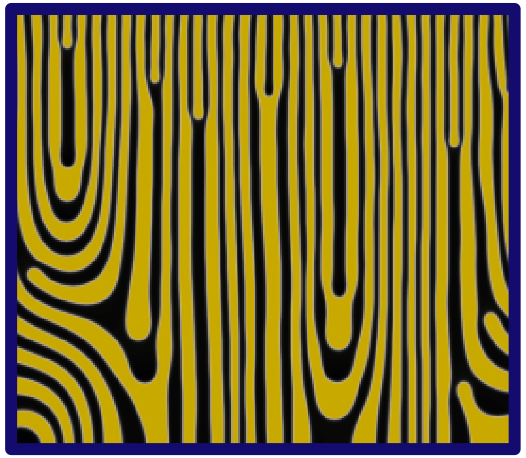
Lateral concentration modulation (LCM) | *Vertical concentration modulation (VCM)* | *Random concentration modulation (RCM)* | *Nanoprecipitate concentration modulation (NPCM)*

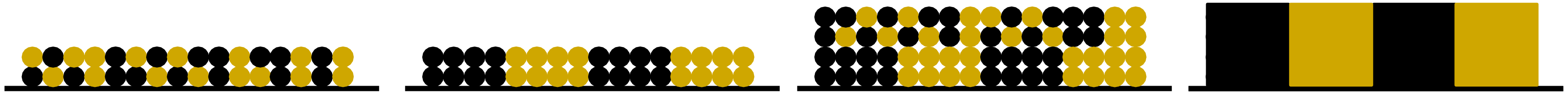
Derby et al. Thin Solid Films (2018)

Phase field model simulates microstructure evolution for various deposition conditions

Microstructures using constant deposition conditions

Deposition rate = 0.075





Initial deposition

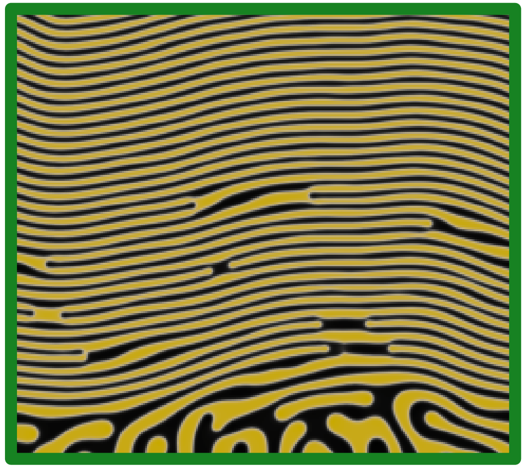
Spinodal decomposition laterally (complete)

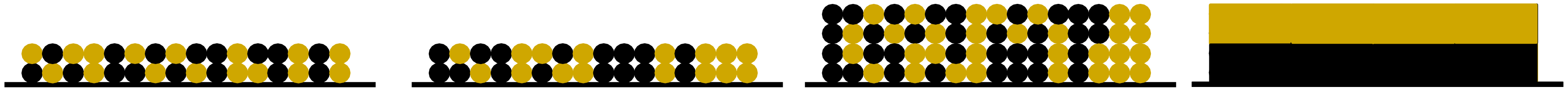
Next deposition

Spinodal decomposition laterally (complete)

Deposition rate = 0.5

Competition between deposition/diffusion gives different microstructures





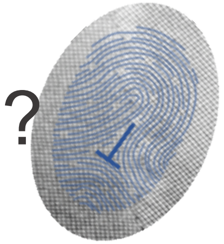
Initial deposition

Spinodal decomposition laterally (incomplete)

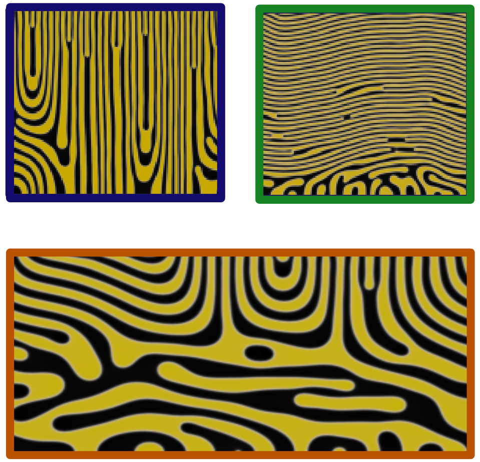
Next deposition

Spinodal decomposition laterally and vertically

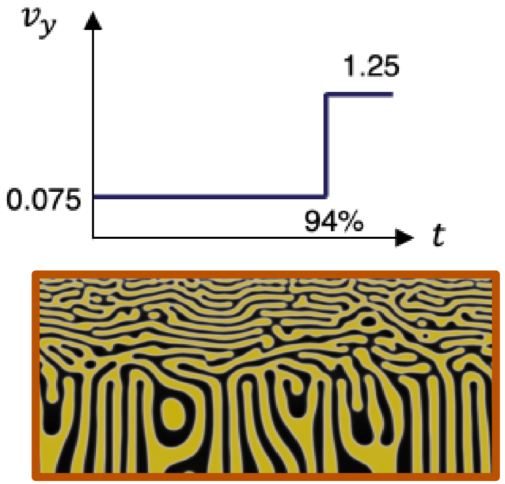
How do we design metallic alloy thin film microstructures?

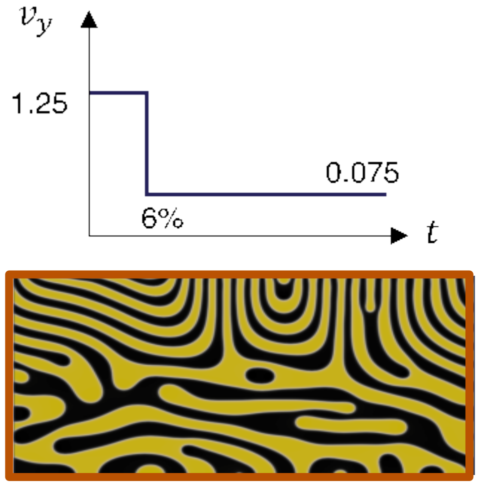


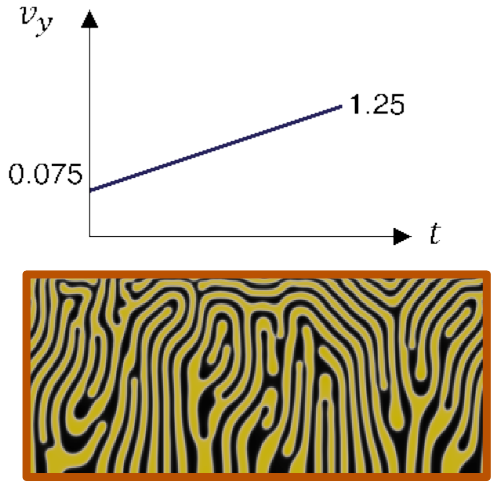
What processing conditions to use to obtain desired film microstructure?

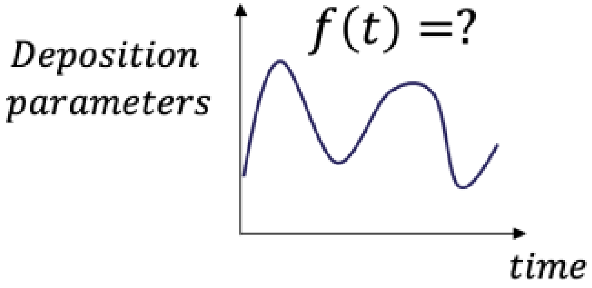


Current SZDs only consider protocols that are constant in time



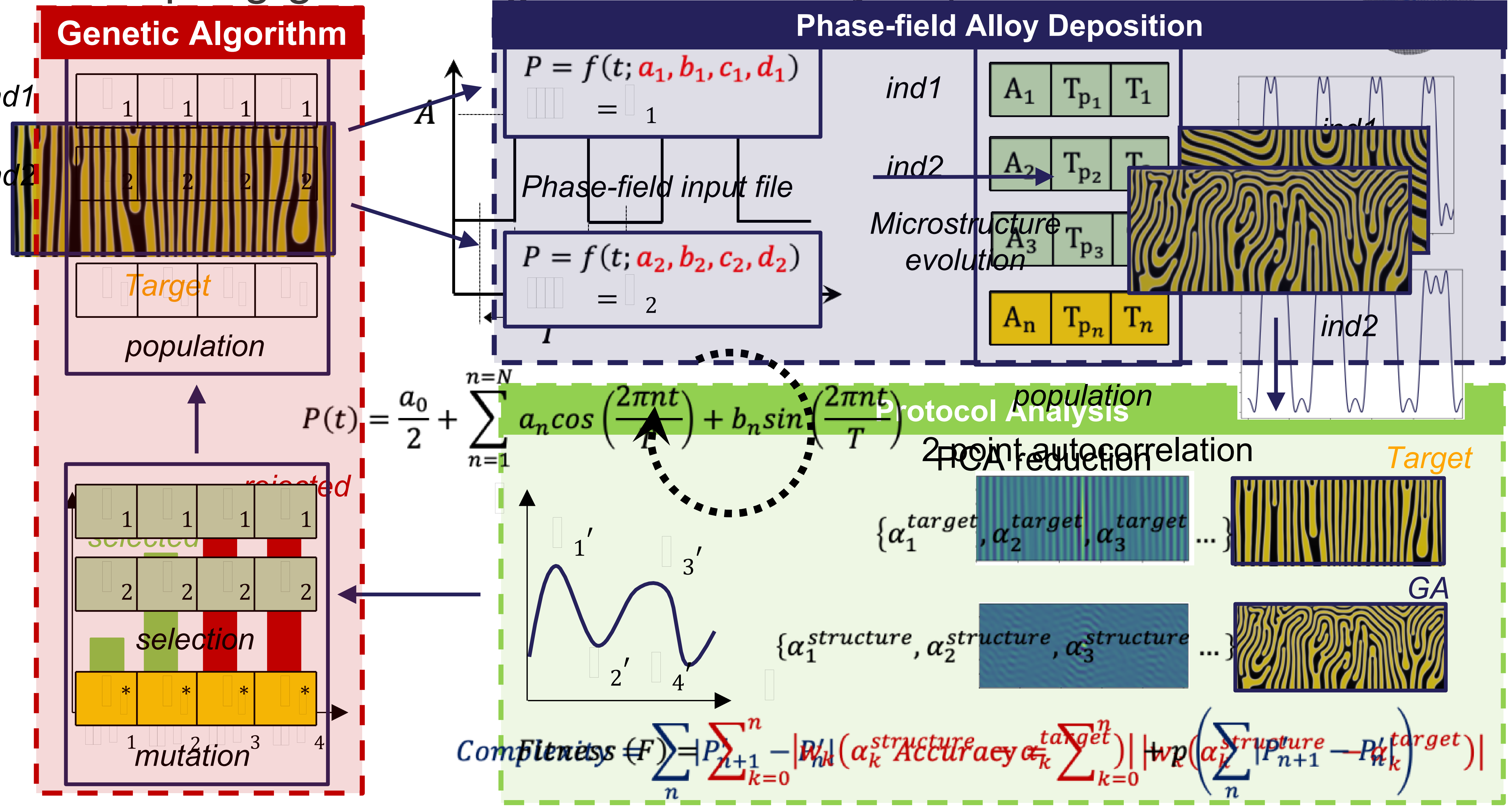




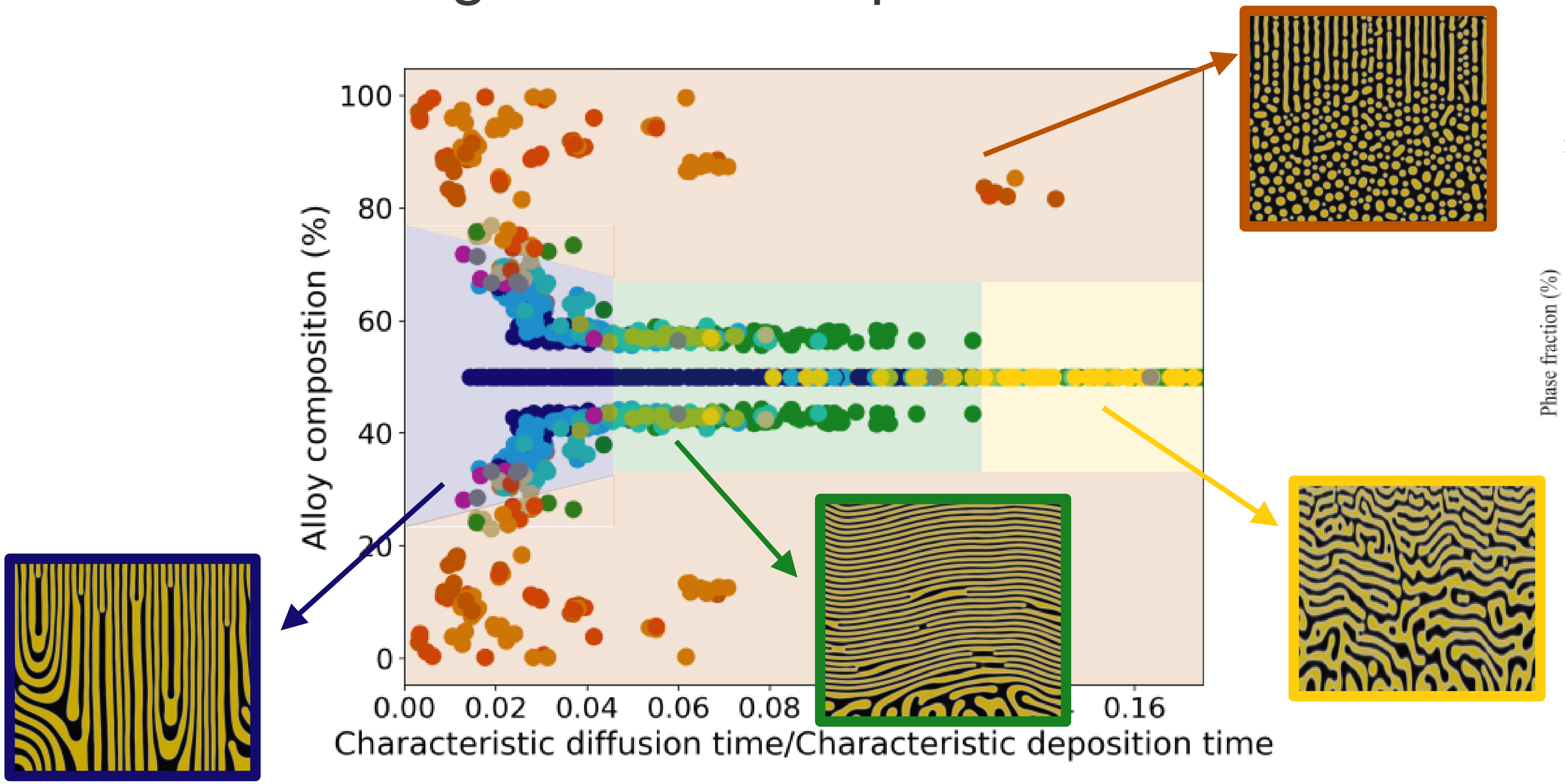


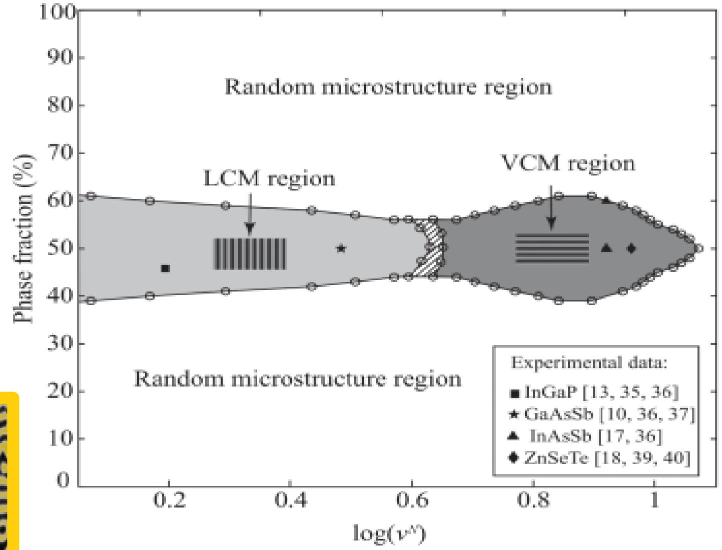
We use a genetic algorithm to discover time-dependent protocols that result in desired microstructure

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

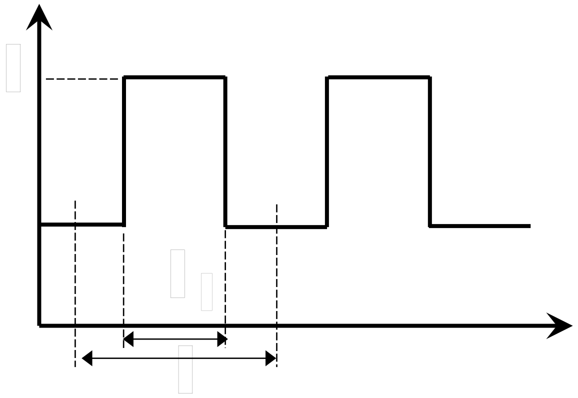
Discovering time-dependent protocols

<u>*Search space*</u>*: pulsed deposition rates*

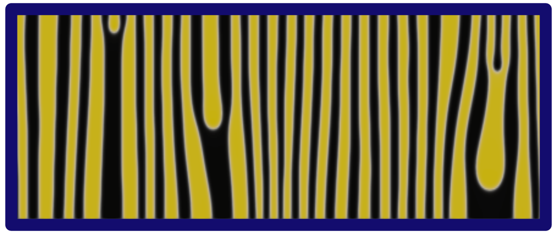
$$v_y^*(t) = \frac{a_0^*}{2} + \sum_{n=1}^{n=N} a_n^* cos\left(\frac{2\pi nt}{T}\right) + b_n^* sin\left(\frac{2\pi nt}{T}\right)$$

$$a_n^* = \frac{2A}{n\pi} \sin\left(\frac{n\pi T_p}{T}\right) \qquad {}^* = 0 \qquad a_0^* = \frac{AT_p}{T}$$

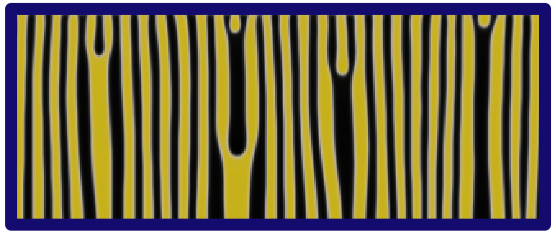
Genetic algorithm parameters: $A, T, f = \frac{T_p}{T}$



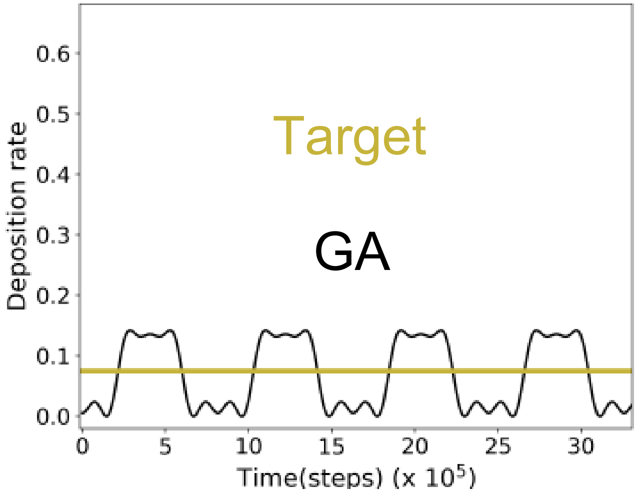
Lateral concentration modulation (LCM)



Target



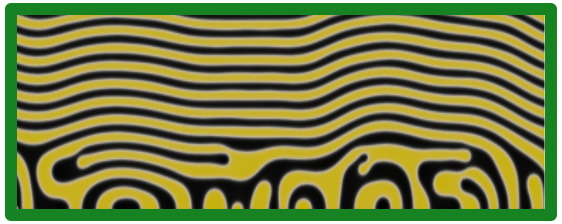
GA



- ➢ Genetic algorithm discovers pulse protocol that results in target structure
- ➢ Deposition amplitude similar to deposition rate used to generate target

Discovering time-dependent protocols

Vertical concentration modulation (VCM)



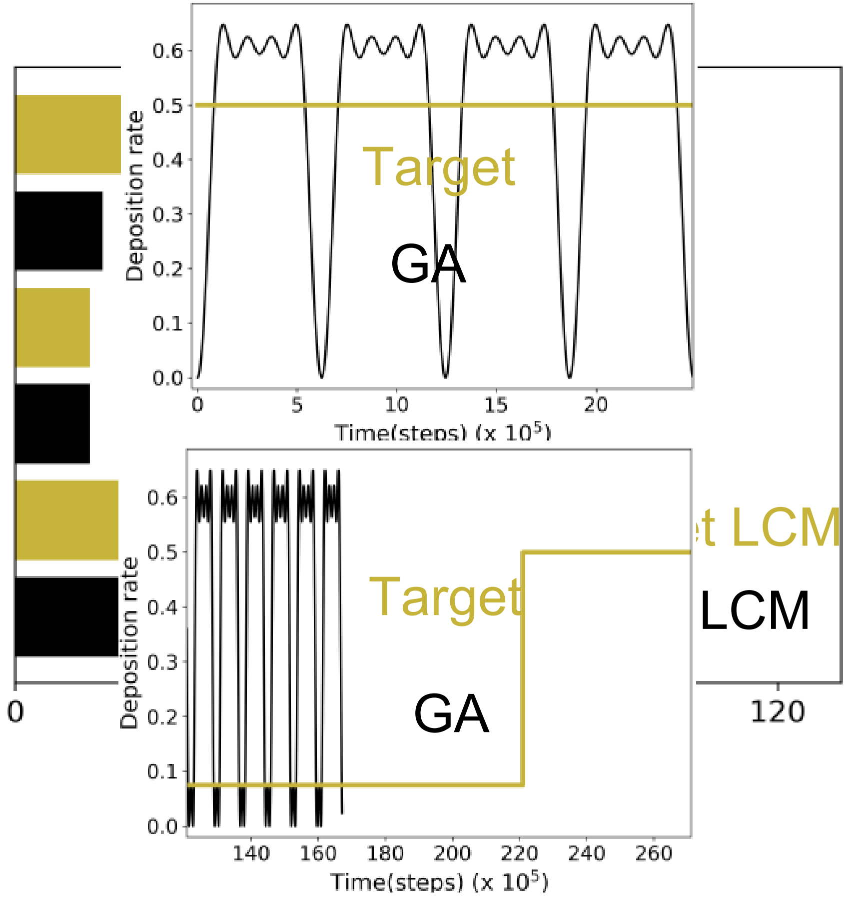
Target

GA

Hierarchical microstructure

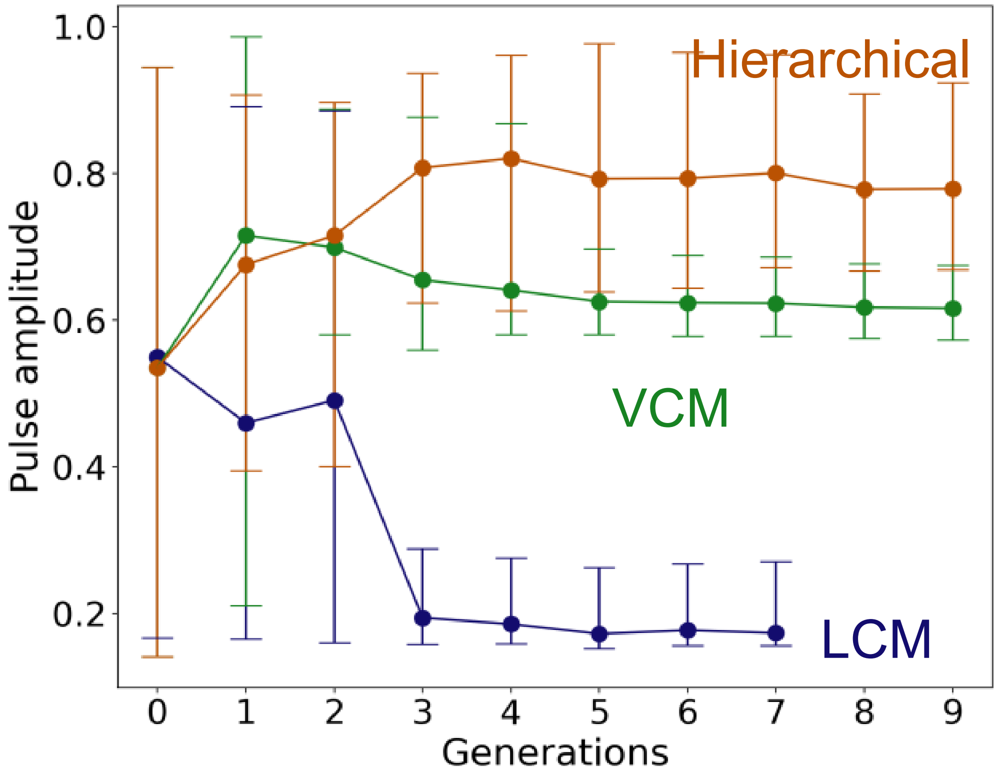
Target

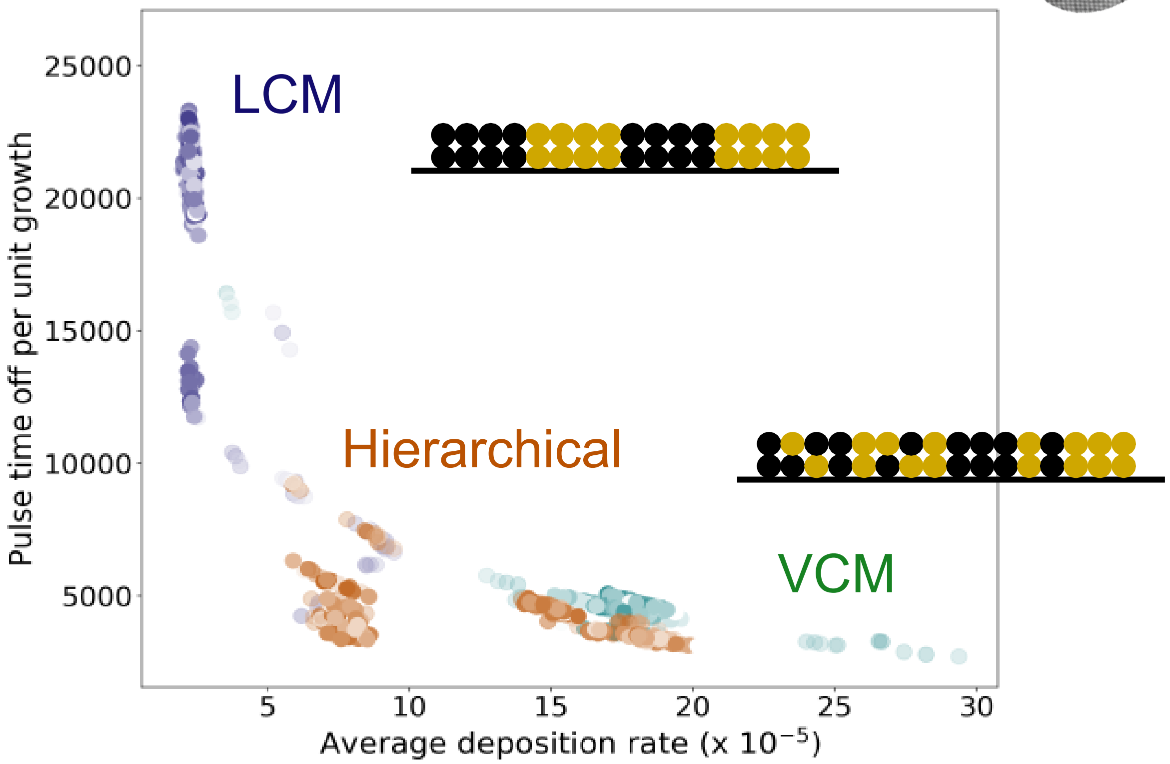
GA



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

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

Optimizing the deposition rate and the mobility

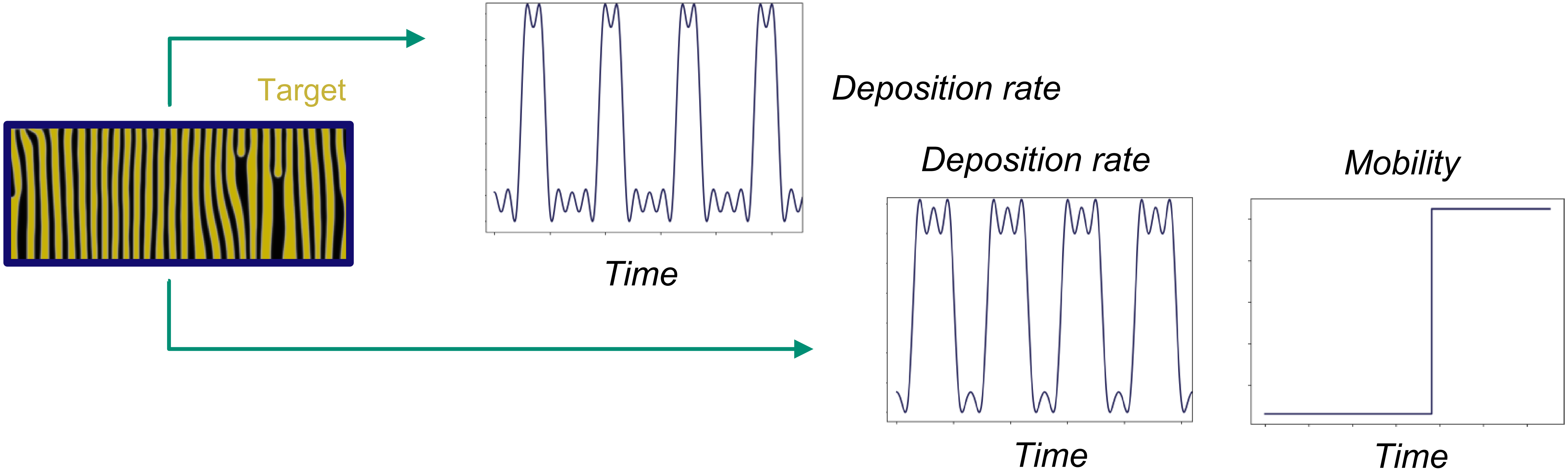
<u>Search space</u>: pulsed deposition rates and step function mobilities

$$v_y^*(t) = \frac{a_0^*}{2} + \sum_{n=1}^{n=N} a_n^* cos\left(\frac{2\pi nt}{T}\right) + b_n^* sin\left(\frac{2\pi nt}{T}\right)$$

$$a_n^* = \frac{2A}{n\pi}\sin\left(\frac{n\pi T_p}{T}\right) \qquad \Box^* = 0 \qquad a_0^* = \frac{AT_p}{T}$$

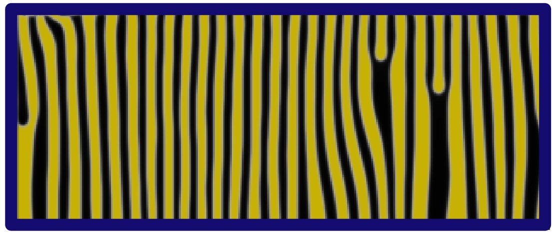
$$M(t) = \begin{cases} a \text{ if } t < t_0 \\ b \text{ if } t \geq t_0 \end{cases}$$

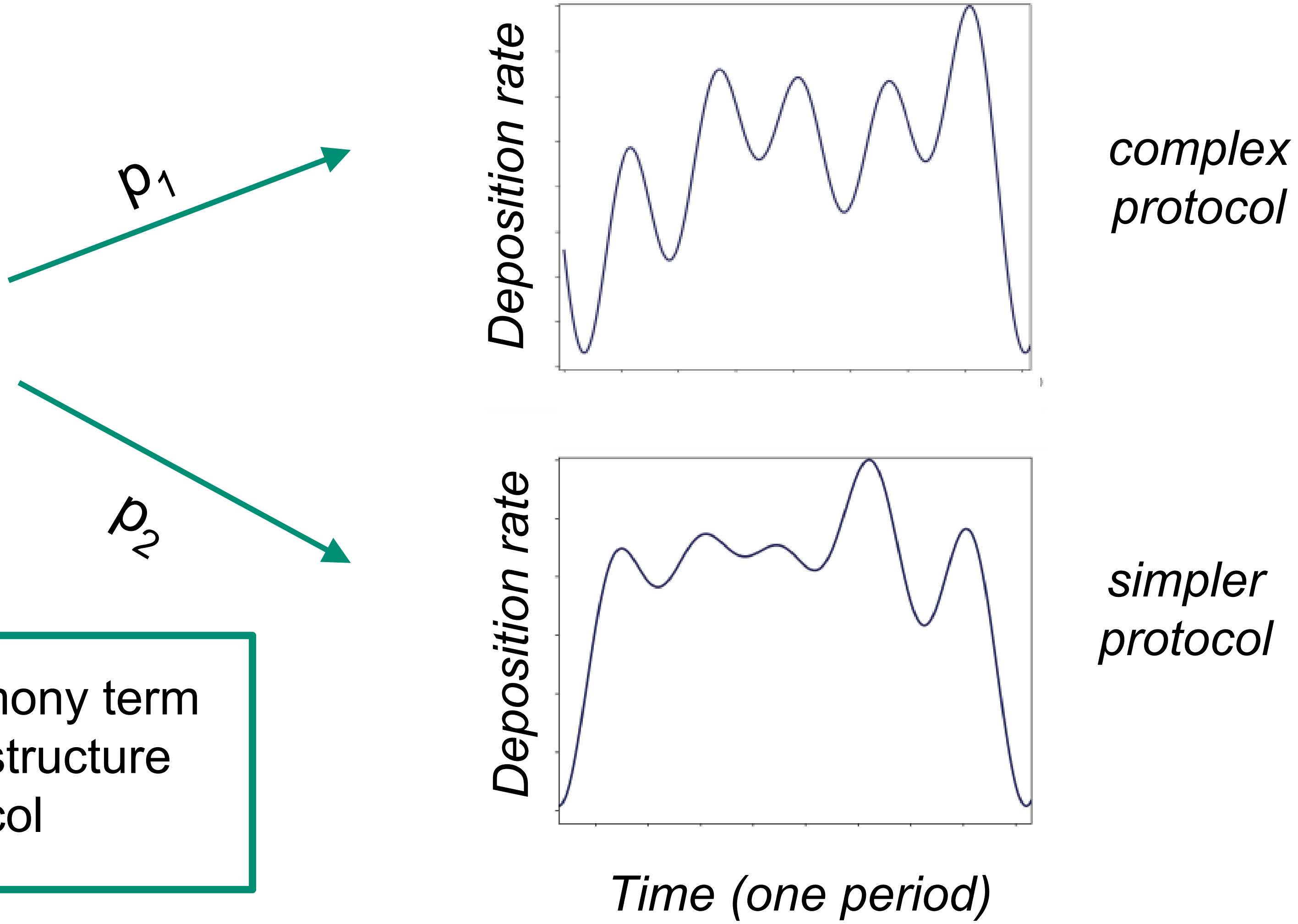
Genetic algorithm parameters: $A,\ T,\ f = \frac{T_p}{T}, a, b, g = \frac{t_0}{T}$



Favoring simple protocols

$$Fitness\ (F) = \sum_{k=0}^{n} \left| w_k \left(\alpha_k^{structure} - \alpha_k^{target} \right) \right| + p(Complexity)$$





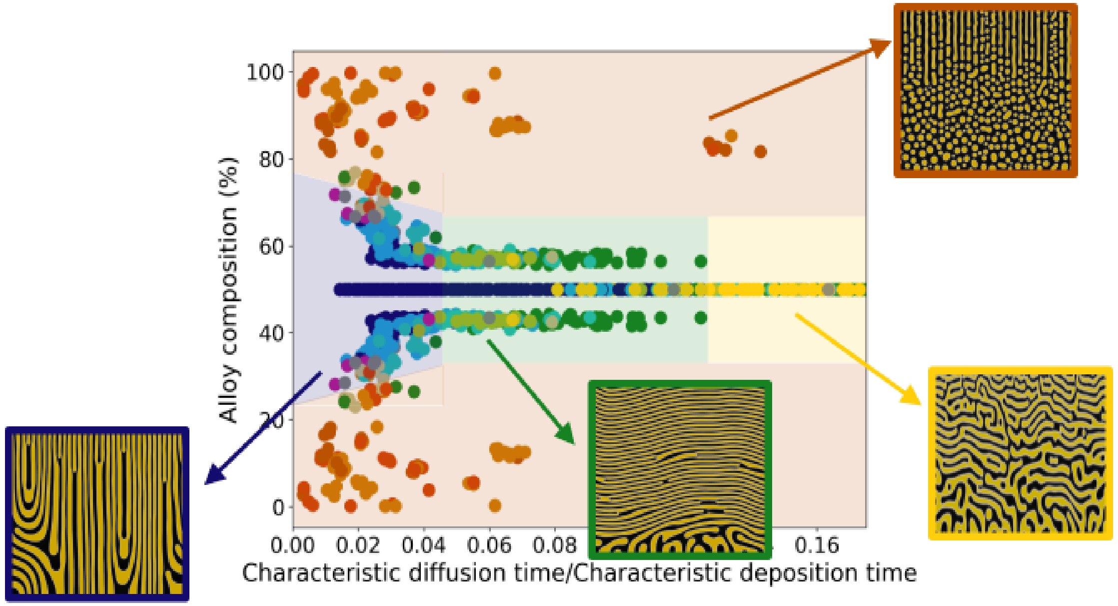
Increasing weight on parsimony term gives near identical microstructure with a simpler protocol

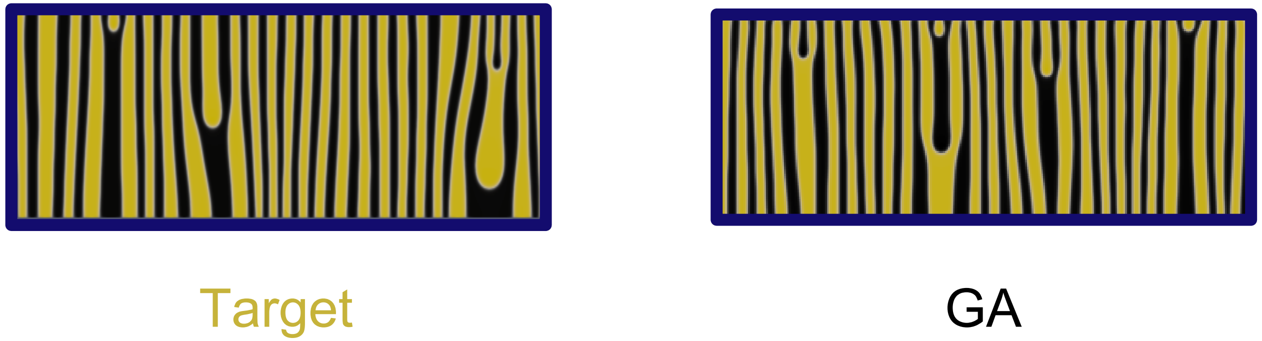
Take home message

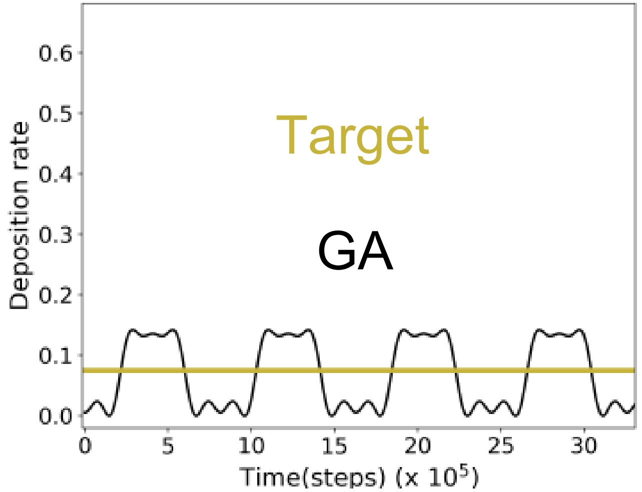
- ➢ Coupled genetic algorithm to phase field simulations to achieve targeted thin film microstructures

- ➢ GA achieves target microstructures for constant deposition scenarios

- ➢ GA discovers time-dependent deposition rates to achieve target microstructures, learning deposition-diffusion tradeoffs







Backup slides

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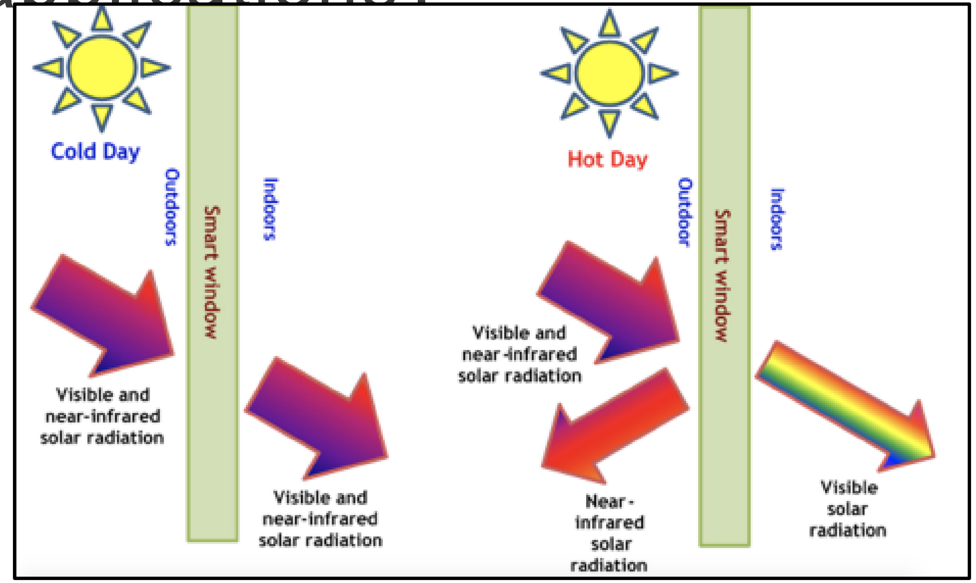
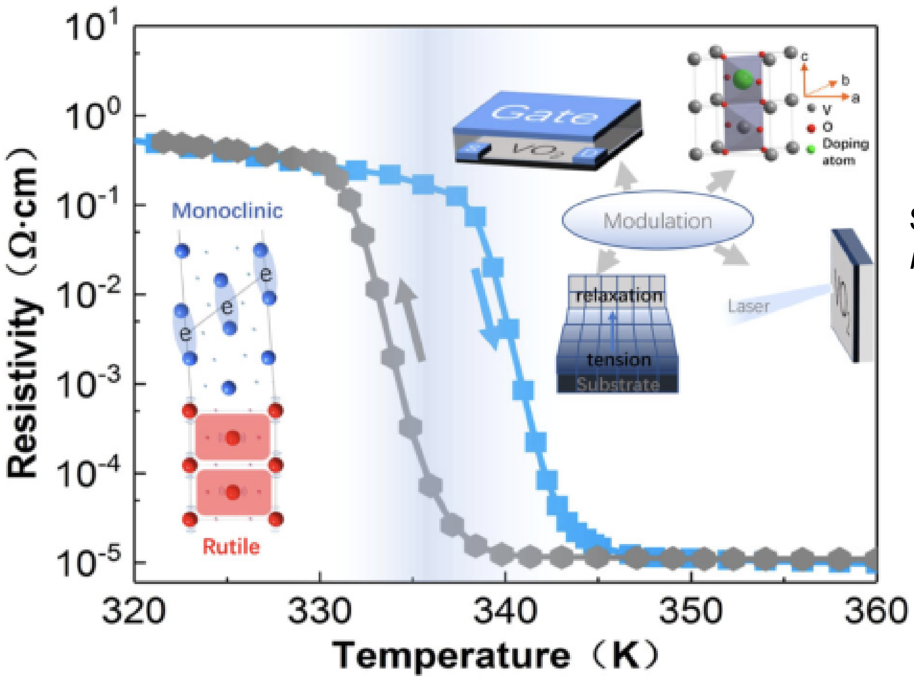
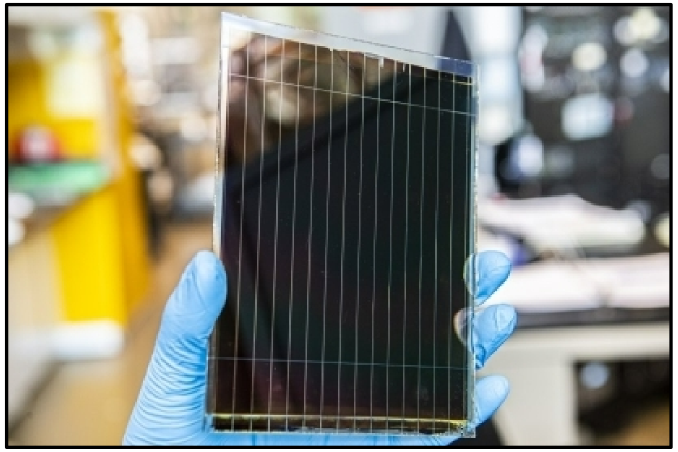
How do we design thin films tailored for specific applications?



Source: nist.gov



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



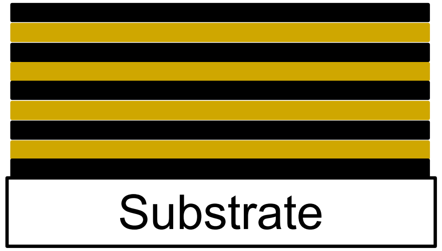
Source: energy.gov

Source: certechinc.com

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

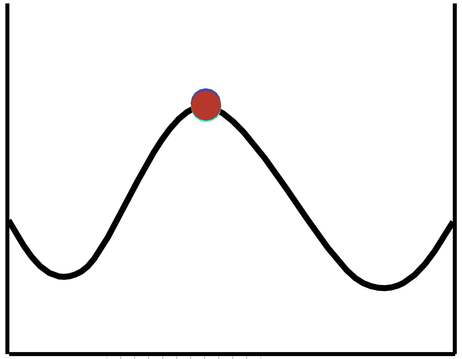
Microstructure formation in metallic alloy thin films

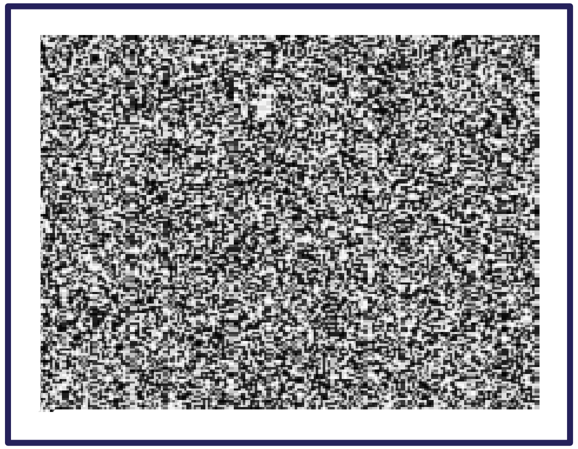
What processing conditions to use to obtain desired film microstructure?



Layer by layer deposition

Self-assembly via spinodal decomposition





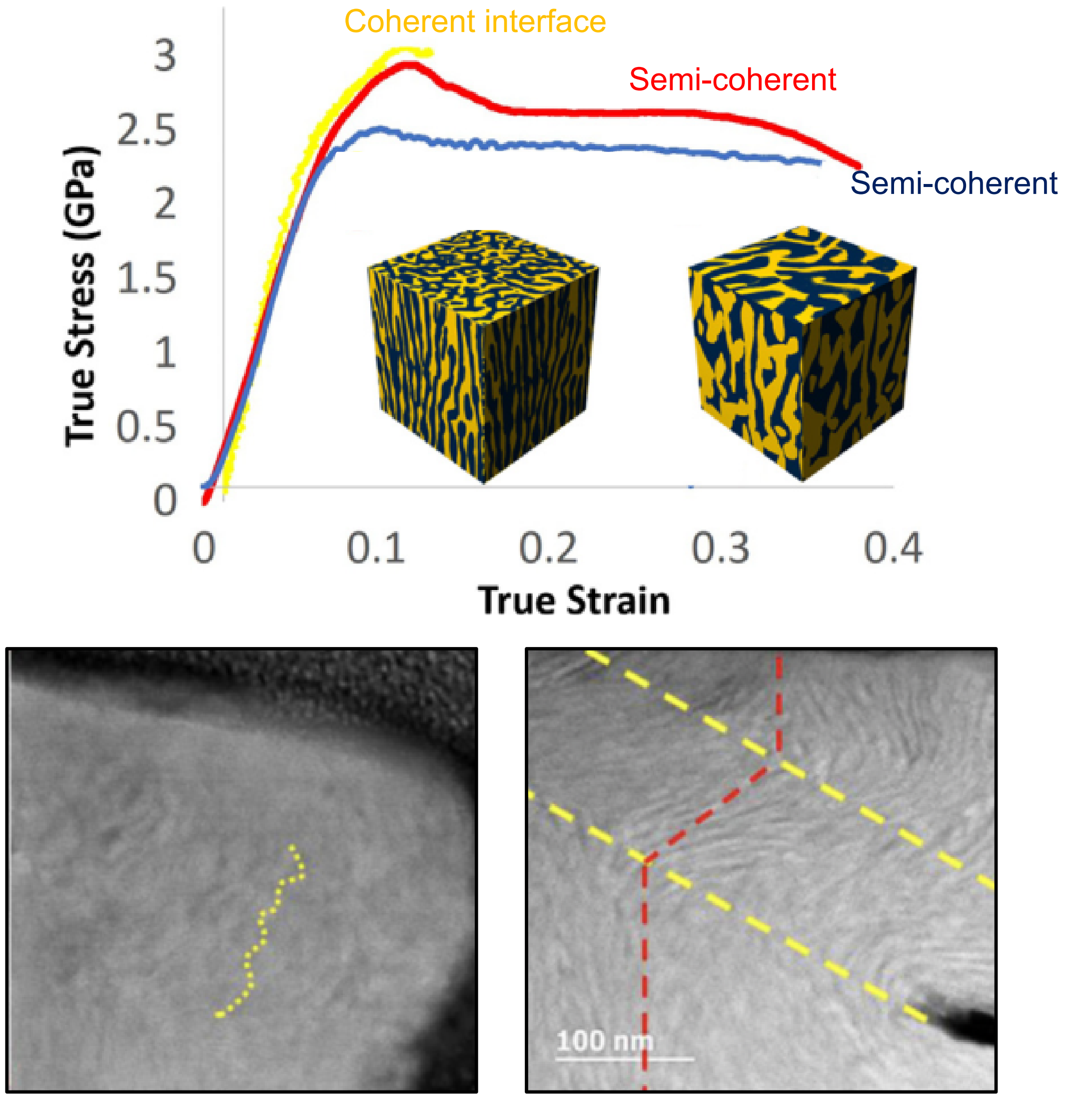
Source: [wikipedia]

PVD experiments

Phase field simulations

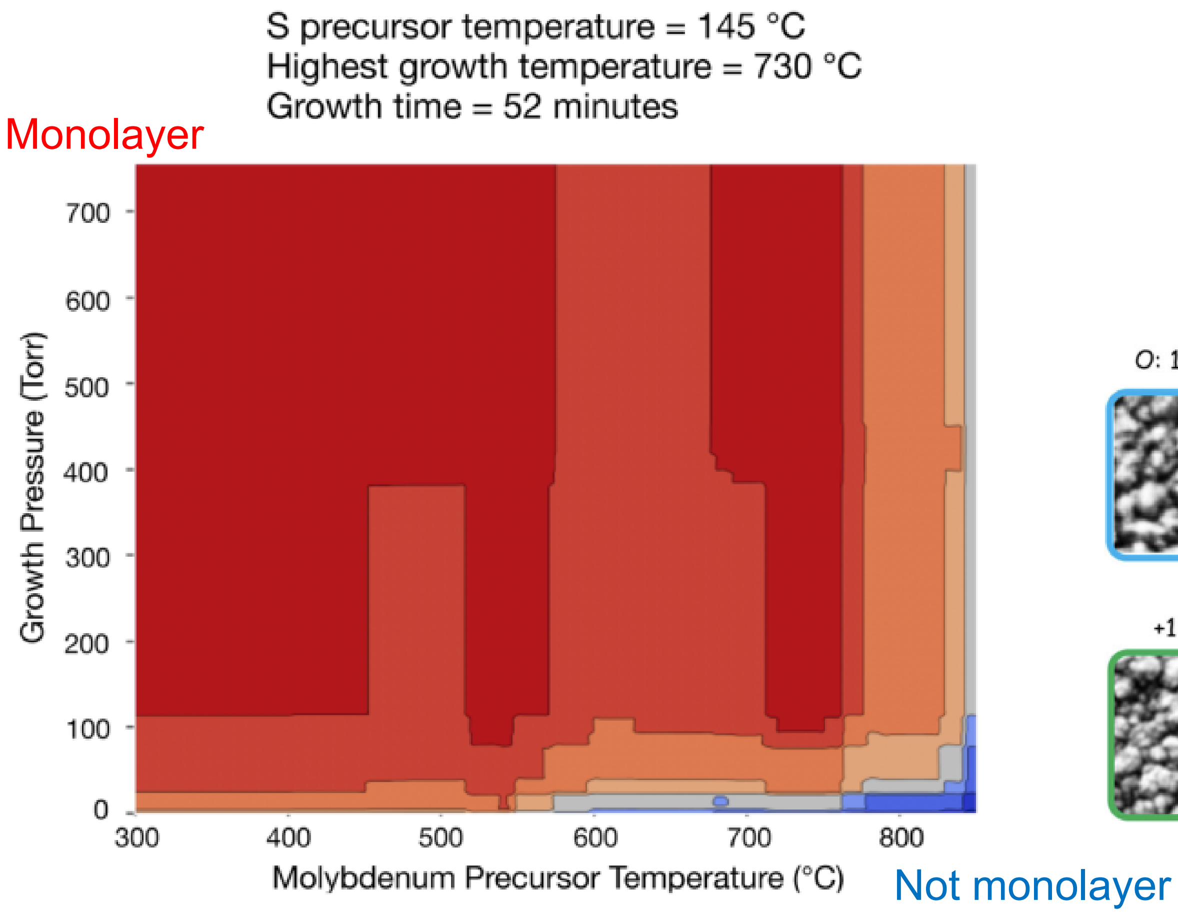
Spinodal decomposition results in spontaneous concentration modulations

Structure-property-processing relationships in thin films



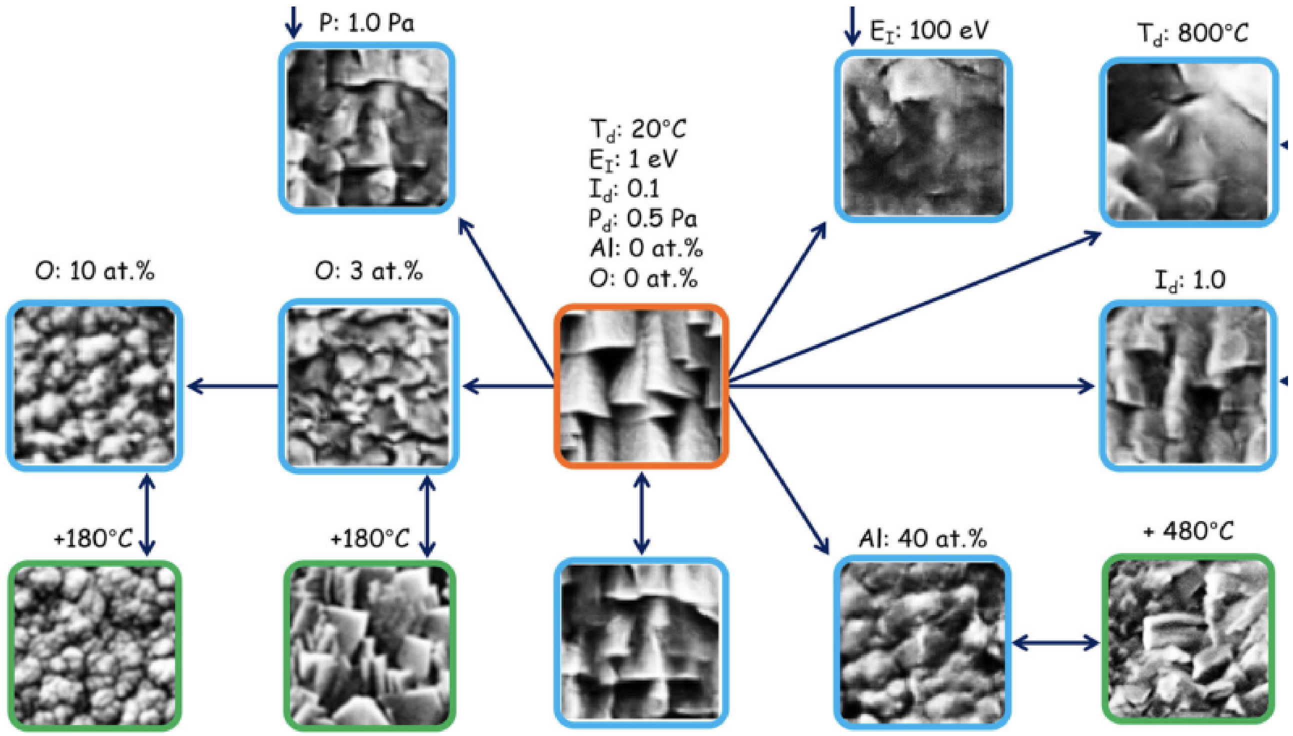
Cui et al. *Materials & Design* (2019)

High dimensional structure zone diagrams



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

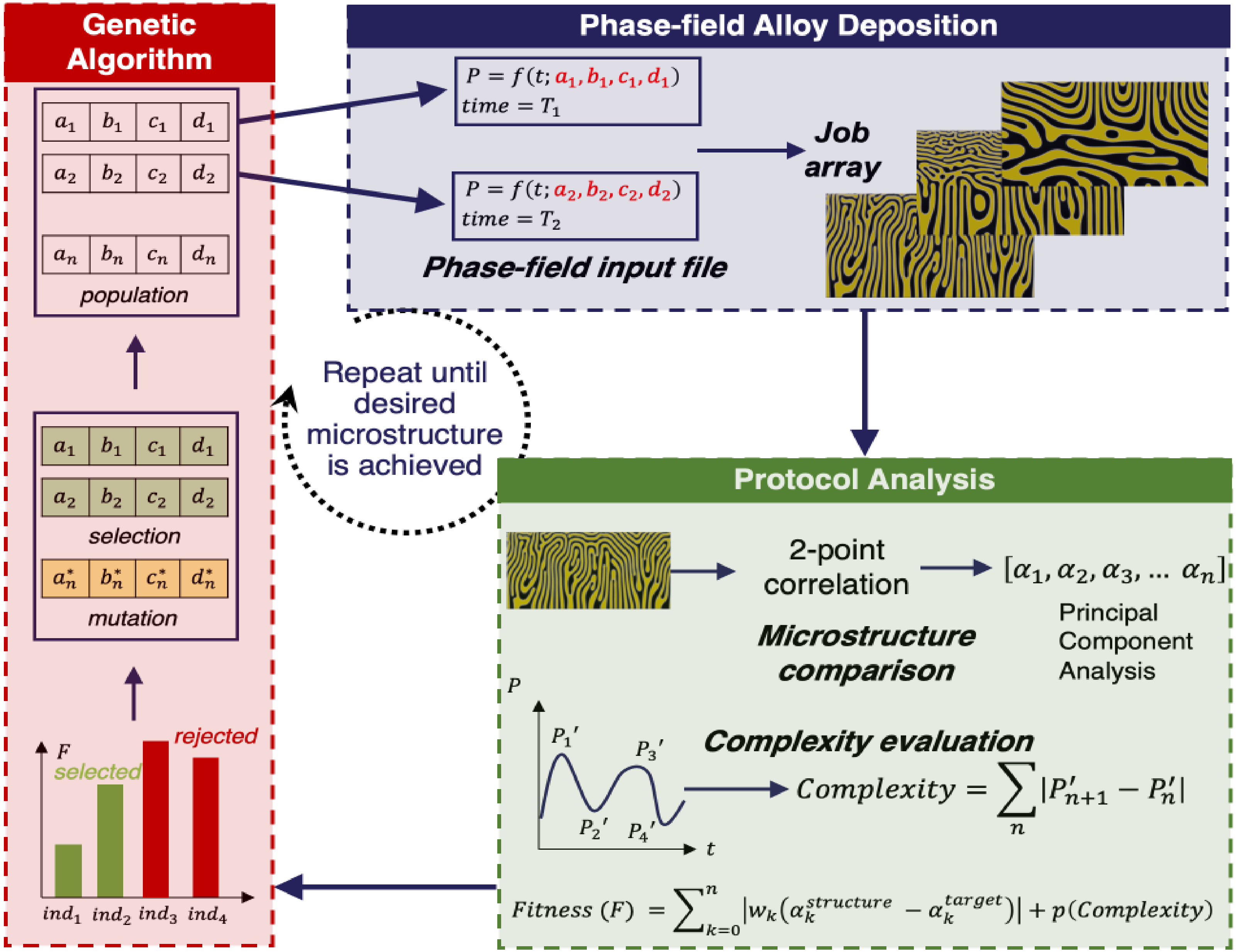
Generative Adversarial Network based SZD



Banko et al. *Communications Materials* (2020)

ML methods can give high dimensional SZDs

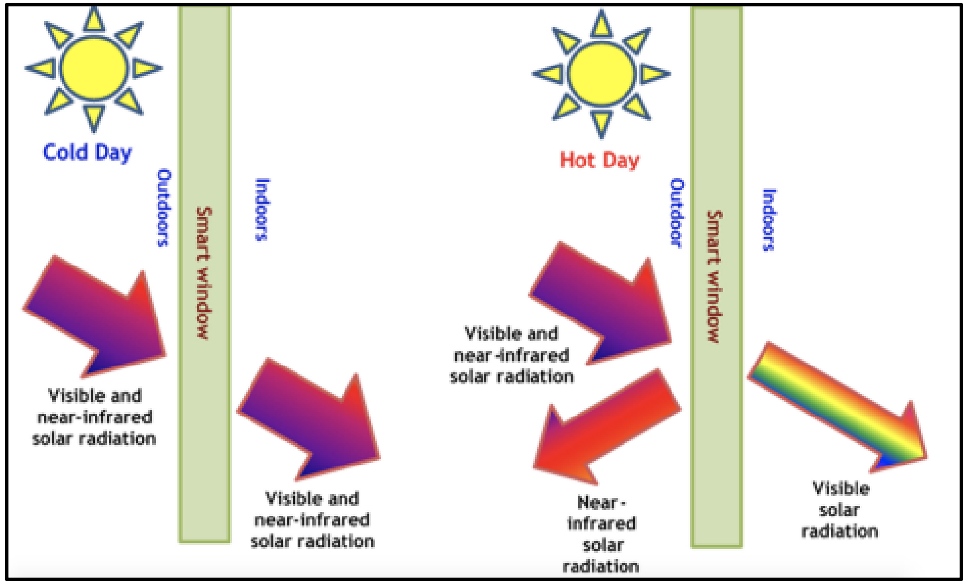
Workflow



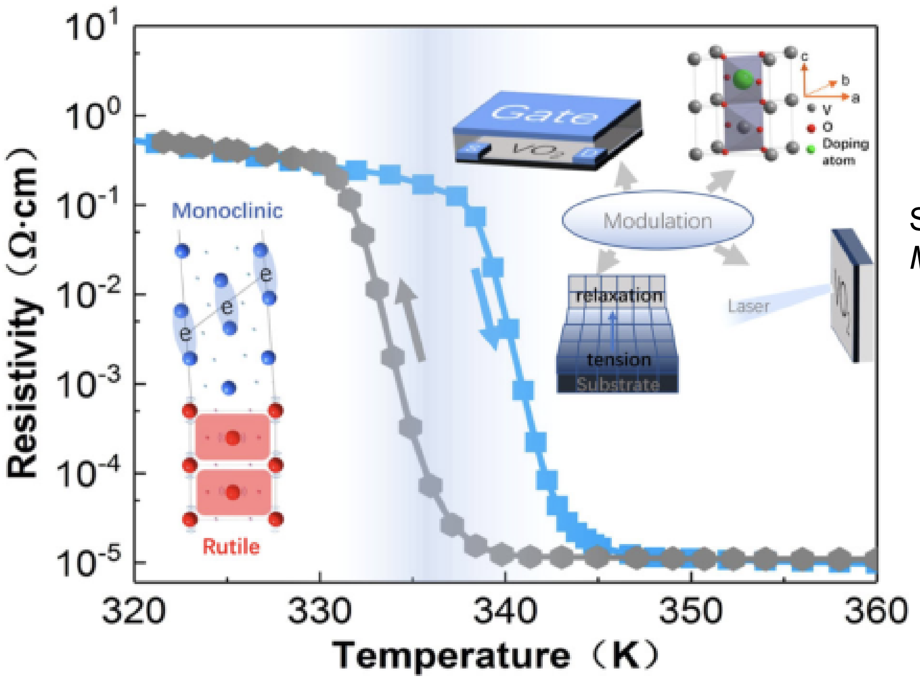
Accuracy

Complexity

Structure-property-processing relationships in thin films



Source: nist.gov



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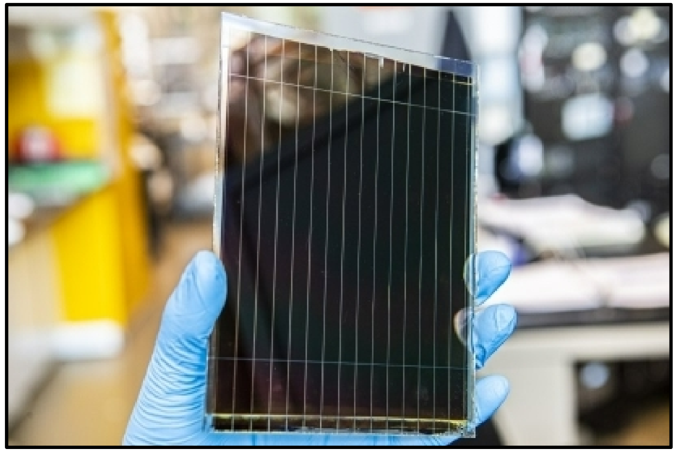
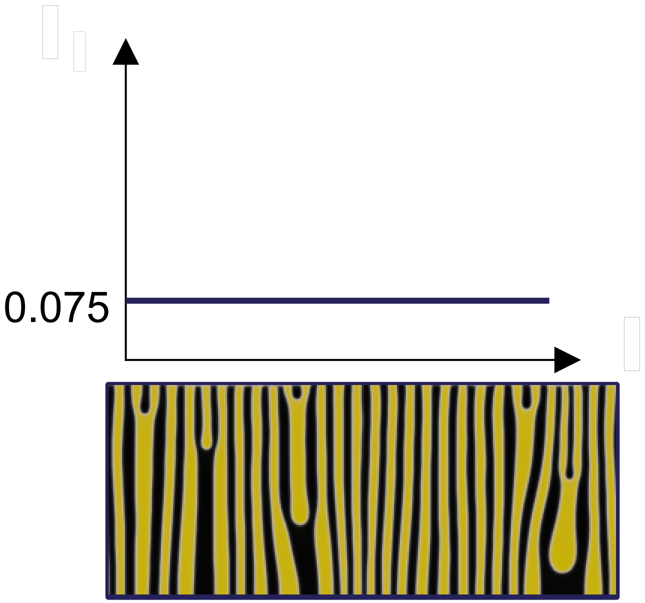


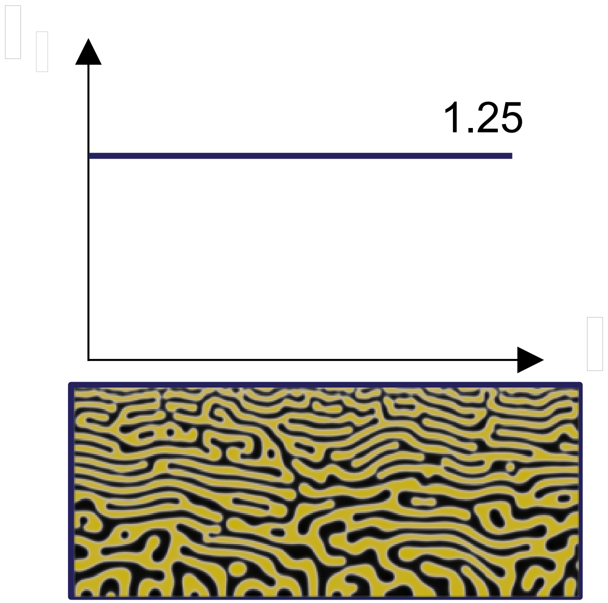
Source: energy.gov

Source: renata.com

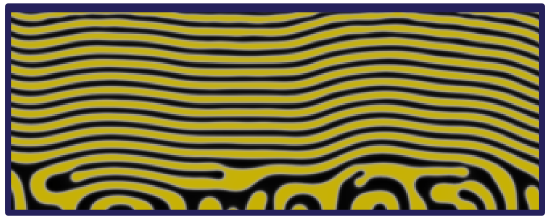
Thin film structure decides properties

Microstructures from simple time-dependent protocols

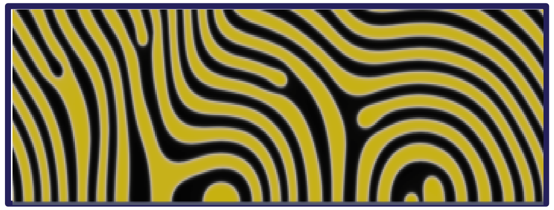




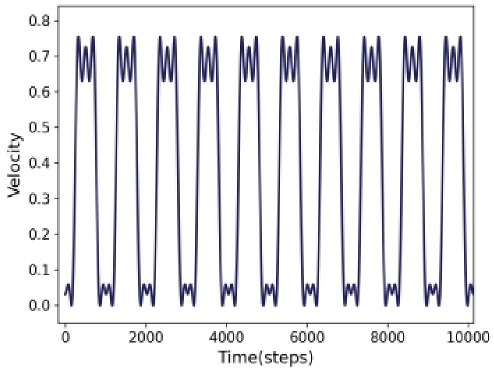
Optimizing the deposition rate and the mobility

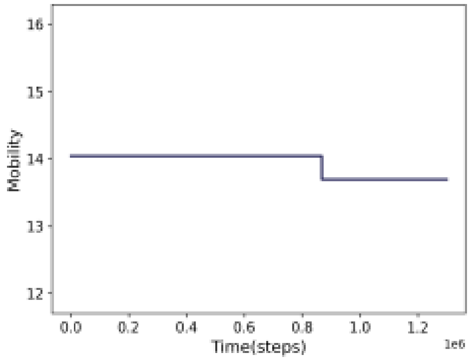


Target

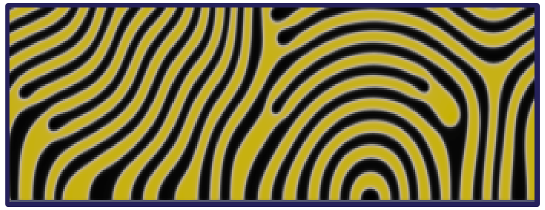


GA

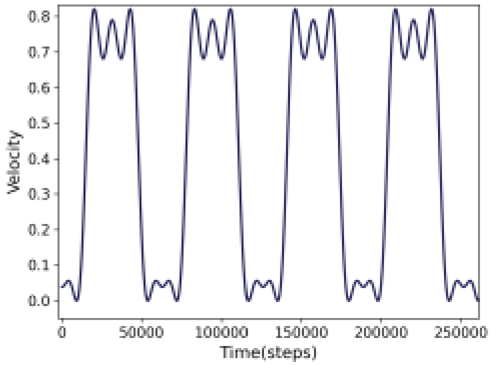


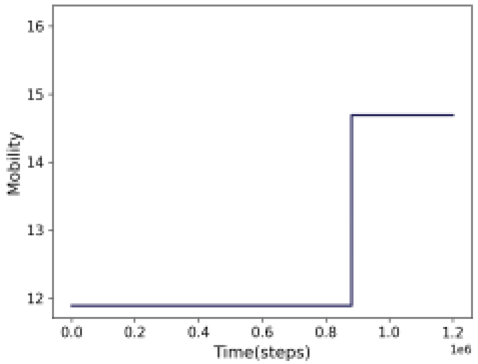


Target

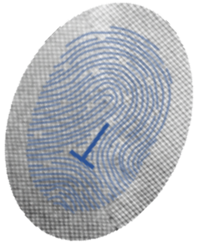


GA





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

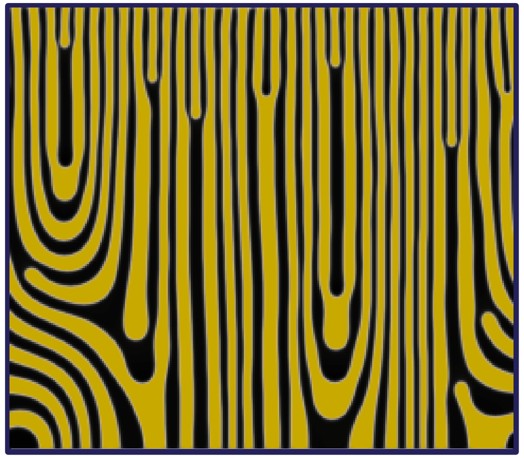
$$\frac{\partial \rho}{\partial t} = \nabla.\left[D_\rho\,\nabla\rho\right] - \nabla.\left[\rho\, v\right] - S(n(\phi))$$

Vapor evolution equation

Phase field model simulates microstructure evolution for various deposition conditions

Microstructures using constant deposition conditions

Deposition rate = 0.075



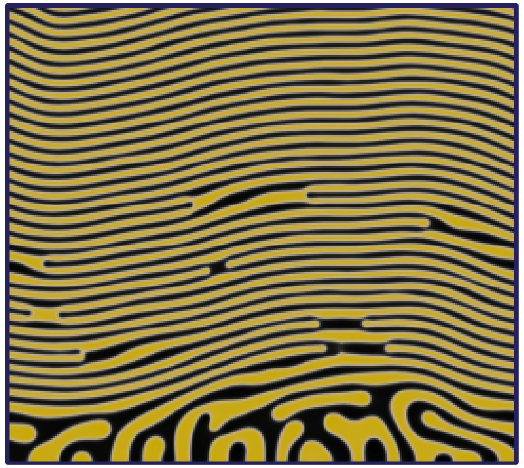
Lateral concentration modulation (LCM)

Random concentration modulation (RCM)

Deposition rate = 1.25

Competition between deposition and diffusion gives different microstructure regimes

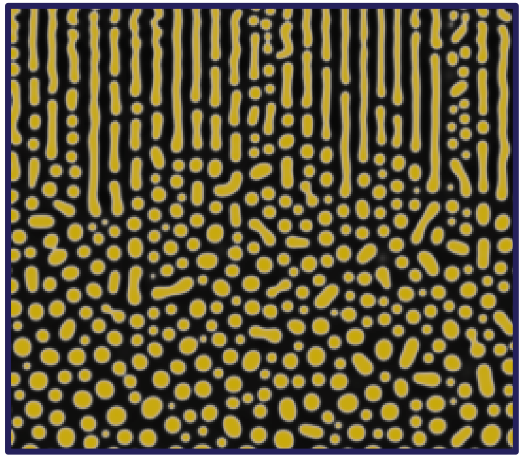
Deposition rate = 0.5



Vertical concentration modulation (VCM)

Nanoprecipitate concentration modulation (NPCM)

Deposition rate = 1.25



$A_{65}B_{35}$