Equations of Motion for Grain Boundaries in Polycrystalline Materials

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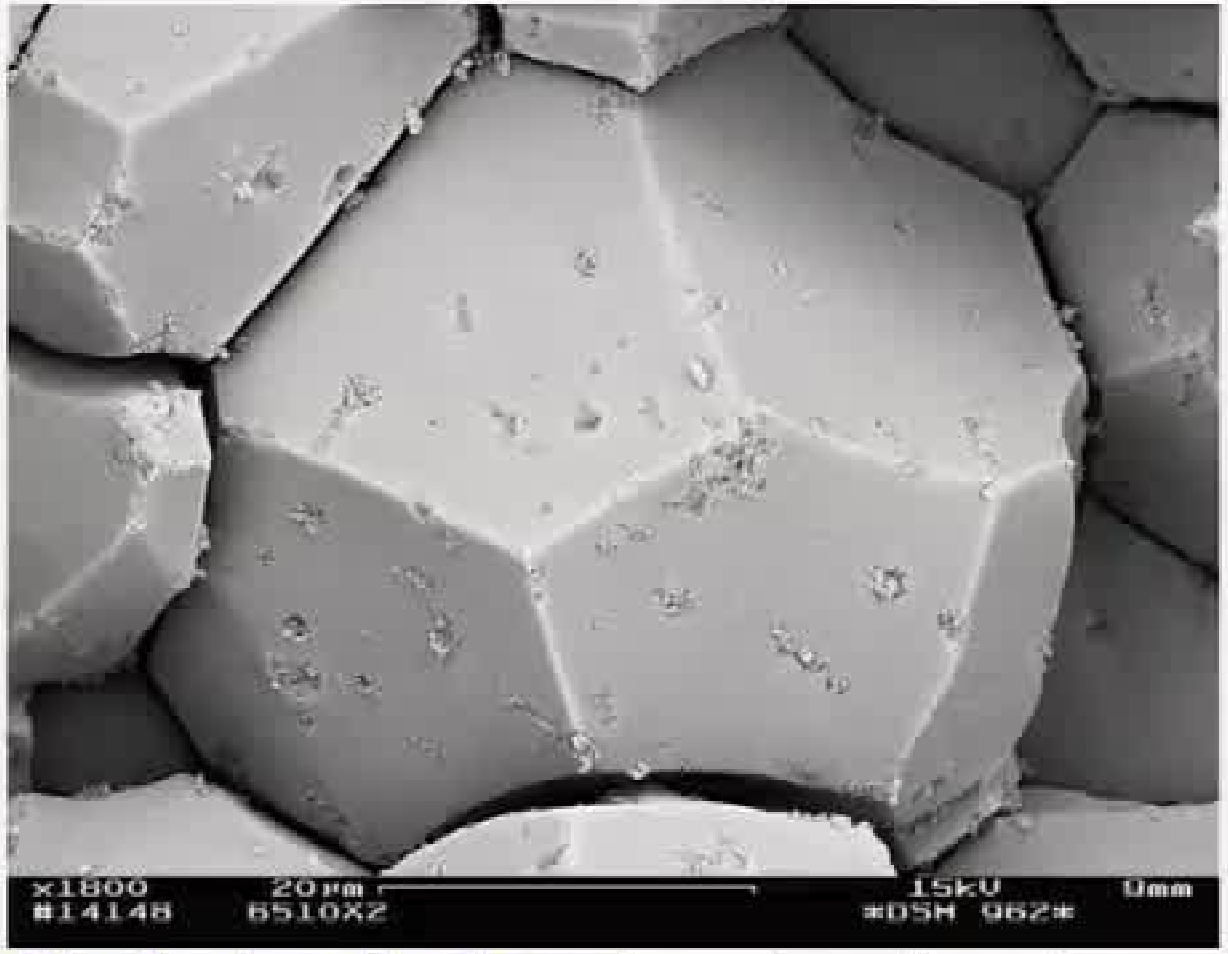
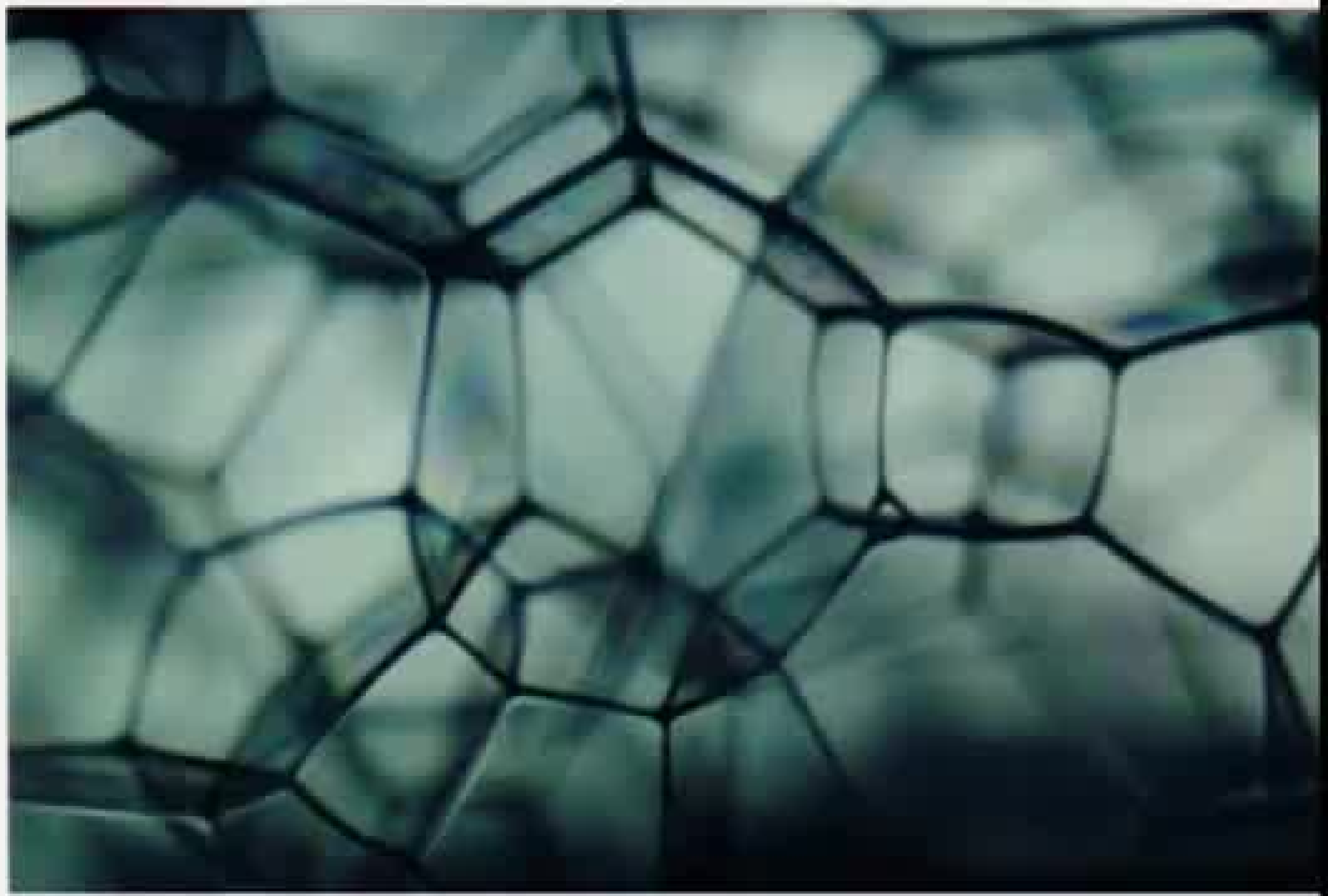
Penn

Polycrystals and Soap Froths

Calcite, $CaCO_3$



Soap froth



Similar topologies: domains, domain walls, triple lines, quadrajunction

Similar evolution: domains coarsen with to decrease domain wall area per unit volume

this naturally leads to $v = -AH$

$R \sim (At)^{1/2}$

H: mean curvature

A: (surface energy γ)$\cdot$(kinetic coef.)

Polycrystals

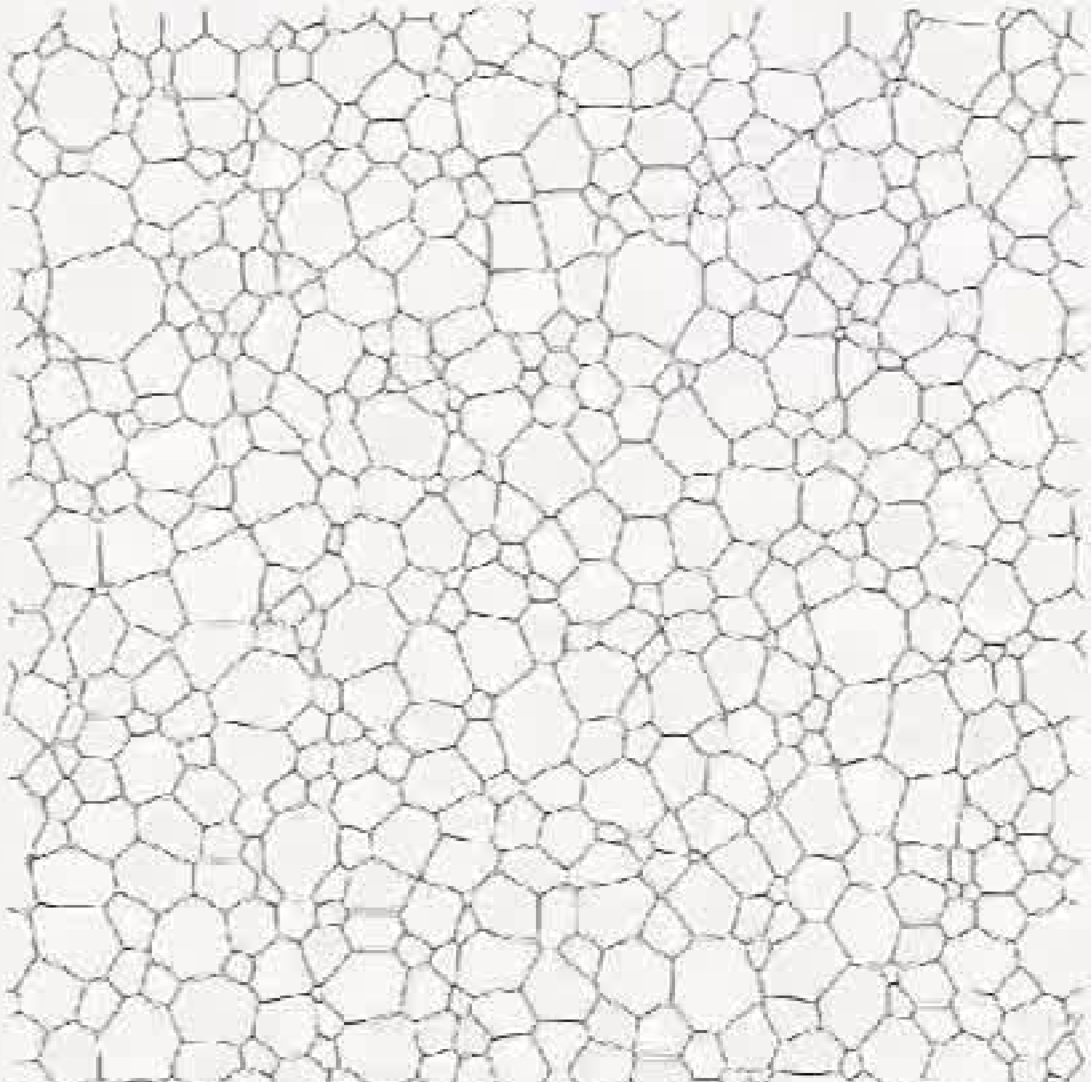
- **Domains:** solids/crystals
- **Walls:** grain boundaries
- **Kinetic coef:** atoms hopping across GB

Soap froths

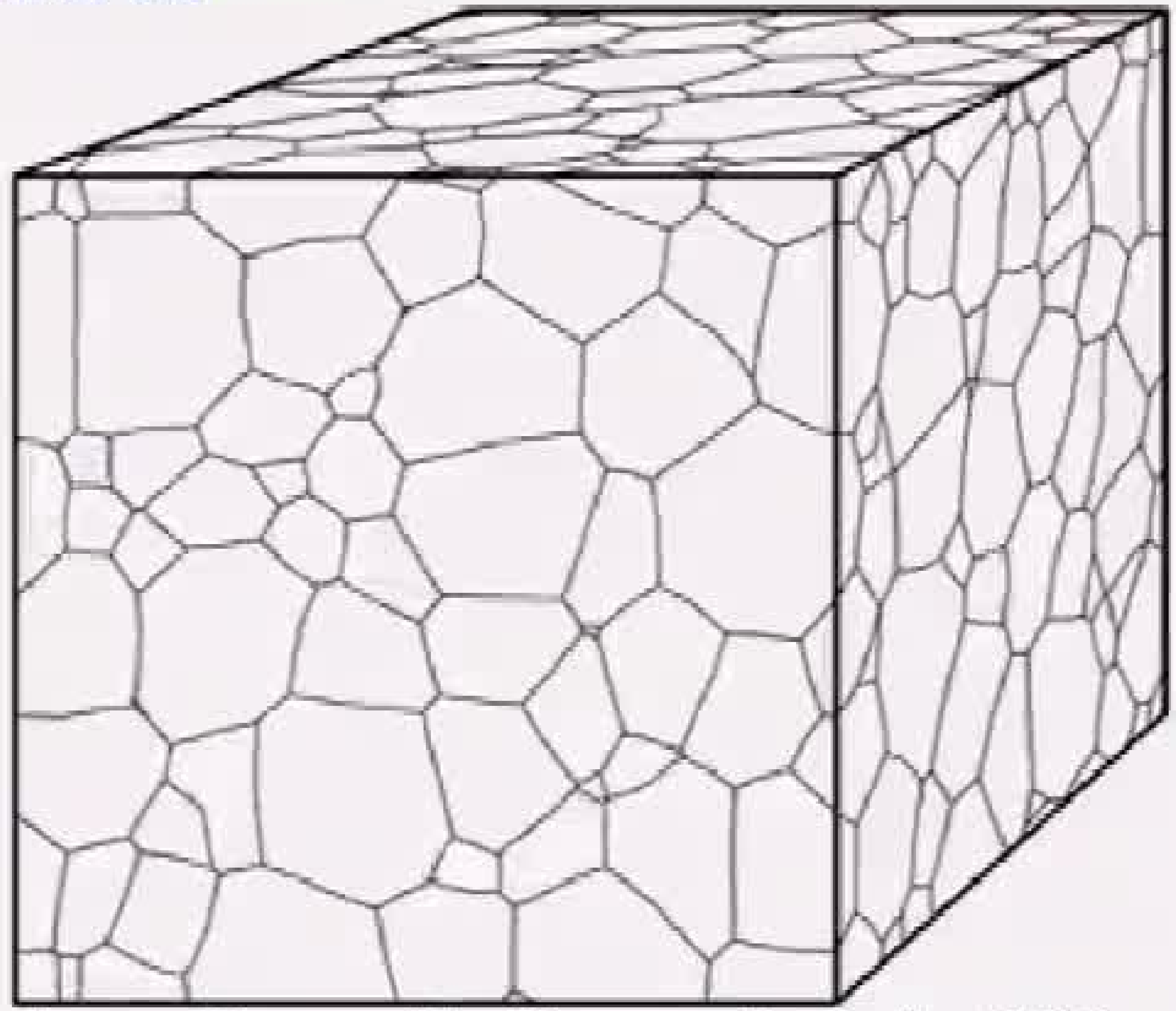
- **Domains:** gas
- **Walls:** liquid films
- **Kinetic coef:** diffusion through film

Motion by Mean Curvature Flow

- (1) evolution is mean curvature H flow
- (2) curvatures balanced at triple junctions → triple junction angles are $2\pi/3$ (isotropic case)
- Font tracking implementations in 2d and 3d



Lazar, MacPherson, Srolovitz, 2010



Lazar, Mason, MacPherson, Srolovitz, 2011

- von Neumann (1951) & Mullins (1952): $\left(\frac{\partial A}{\partial t}\right) = \left(\frac{M\gamma\pi}{3}\right)(n-6)$ 2d
- MacPherson & Srolovitz (2007) all d

Motion by Mean Curvature Flow

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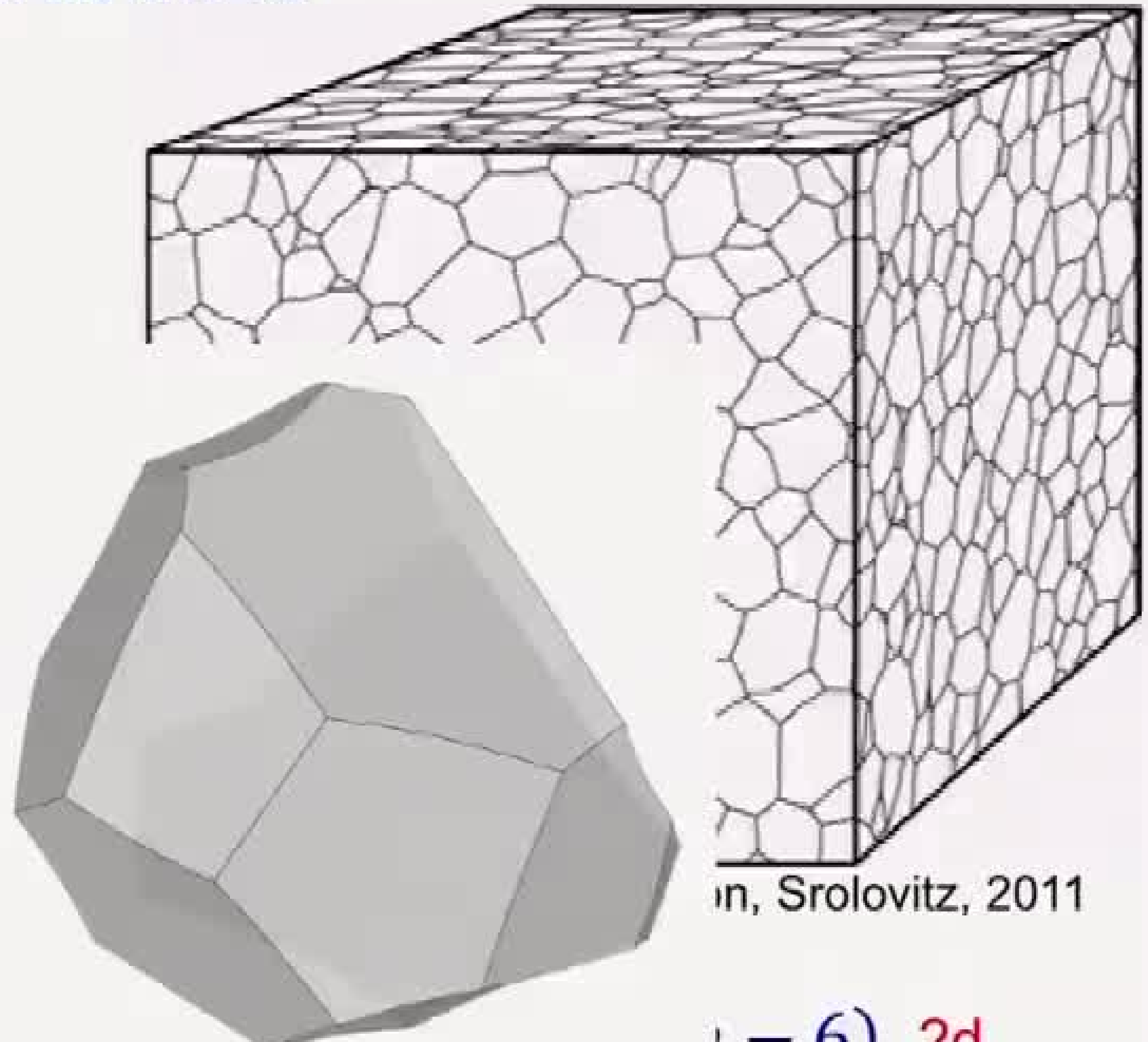


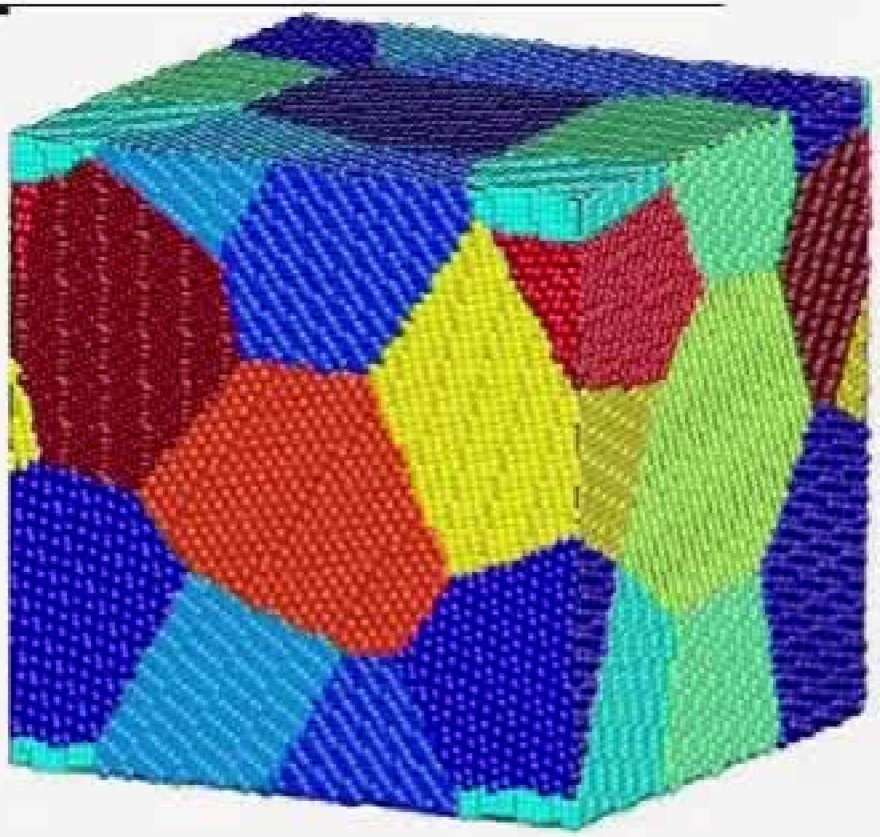
Lazar, MacPhei

n, Srolovitz, 2011

- von Neur ... $- 6)$ 2d
- MacPher

Real Grain Boundaries

- But, grains are crystalline → crystal structure (symmetry) effects & elasticity
- Anisotropy in GB properties and how they move (5 dimensional space)
- Not mean curvature flow – crystal structure matters!



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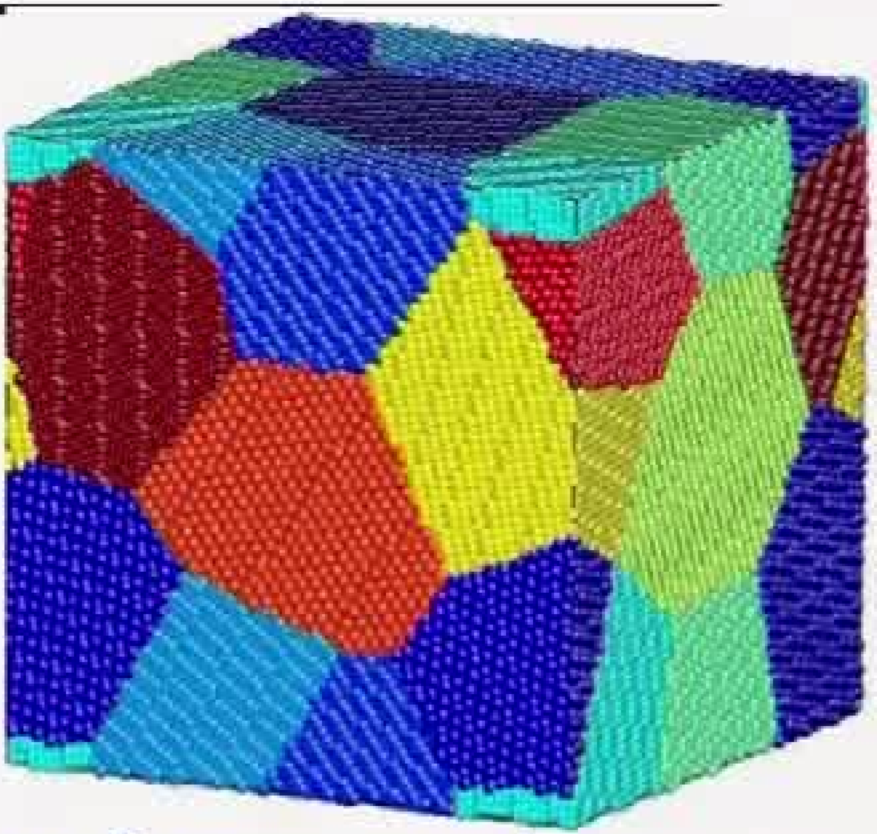
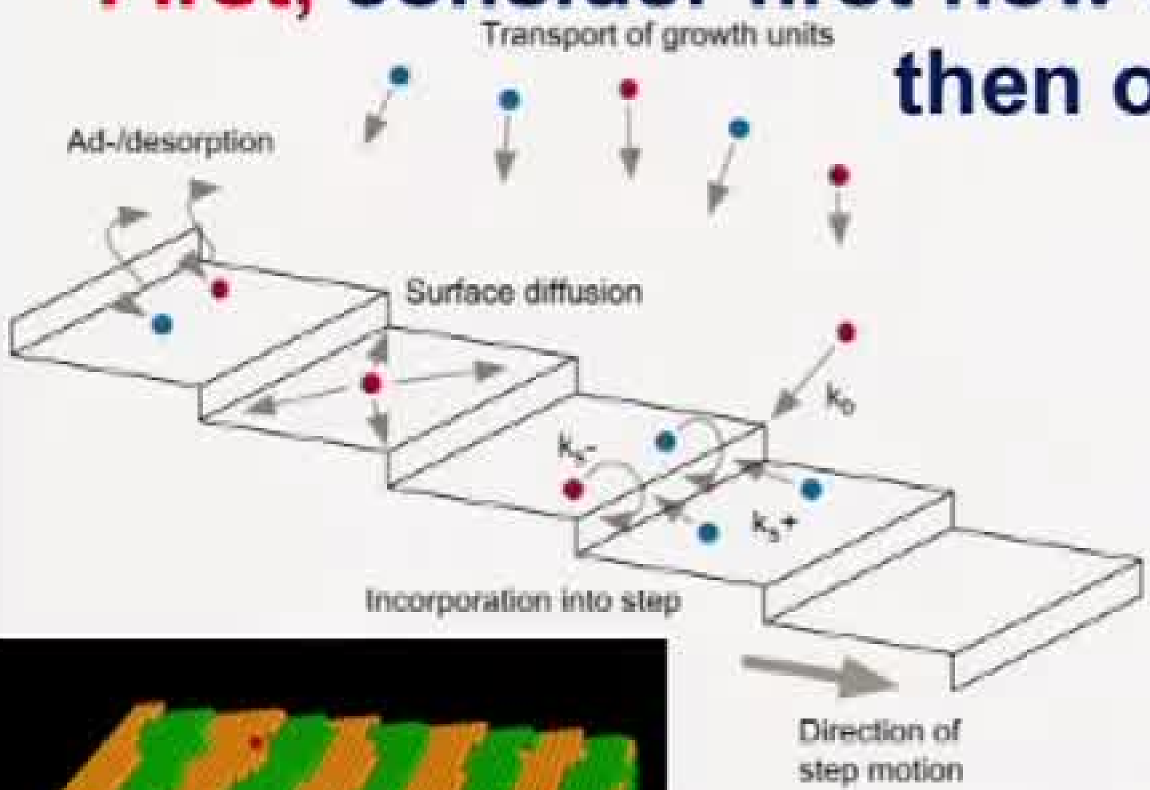


First, consider first how crystals grow/surfaces evolve then on to GBs

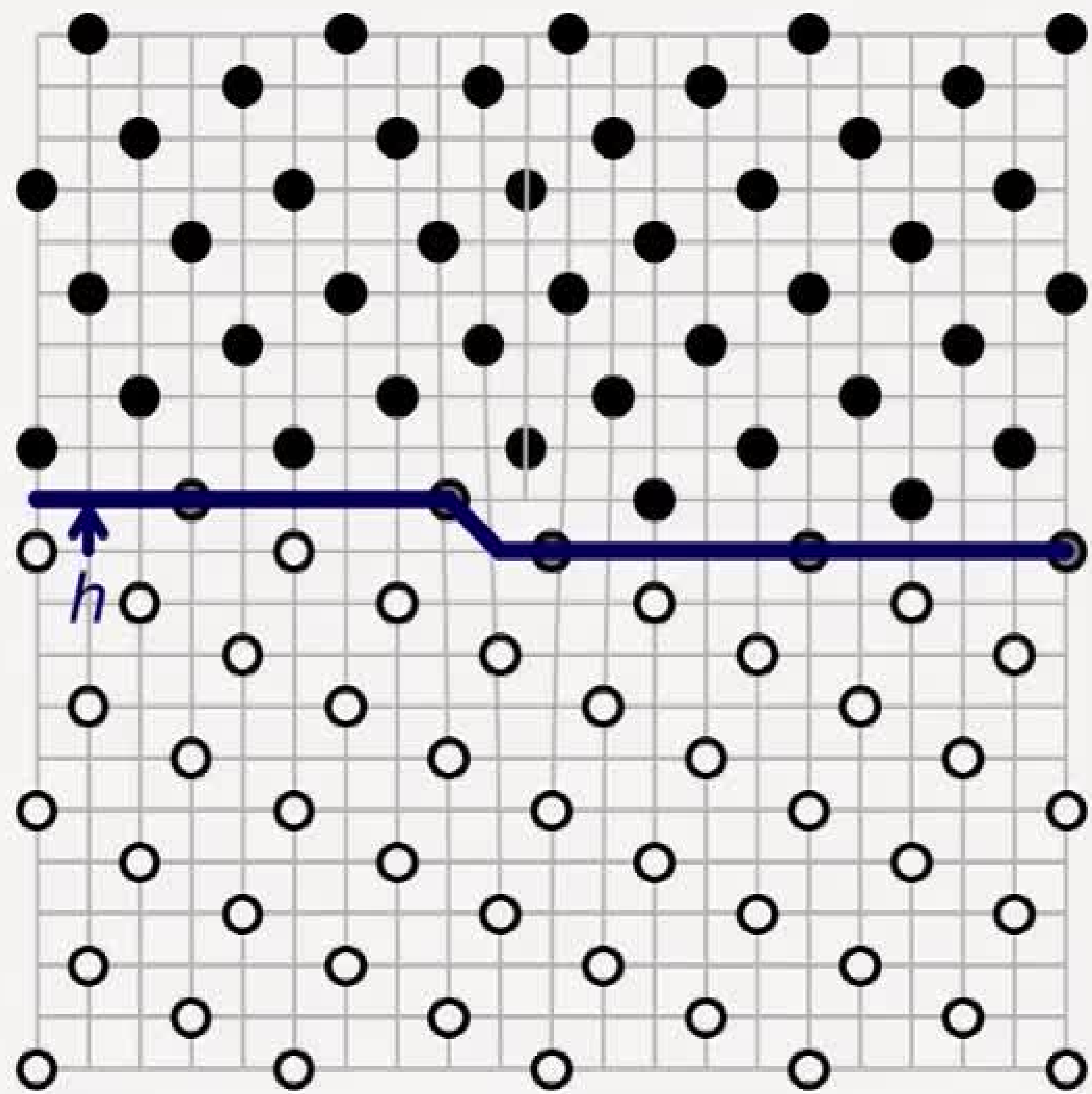


- Normal motion of the surface **is** lateral motion of steps
- Crystallography determines step heights
- Here, steps move by adding atoms from the terraces or from the gas

Real Grain Boundaries

- Now, instead of surfaces, we focus on grain boundaries – interfaces between misoriented crystals

- Like surfaces, GBs have steps (characterized by *h*)



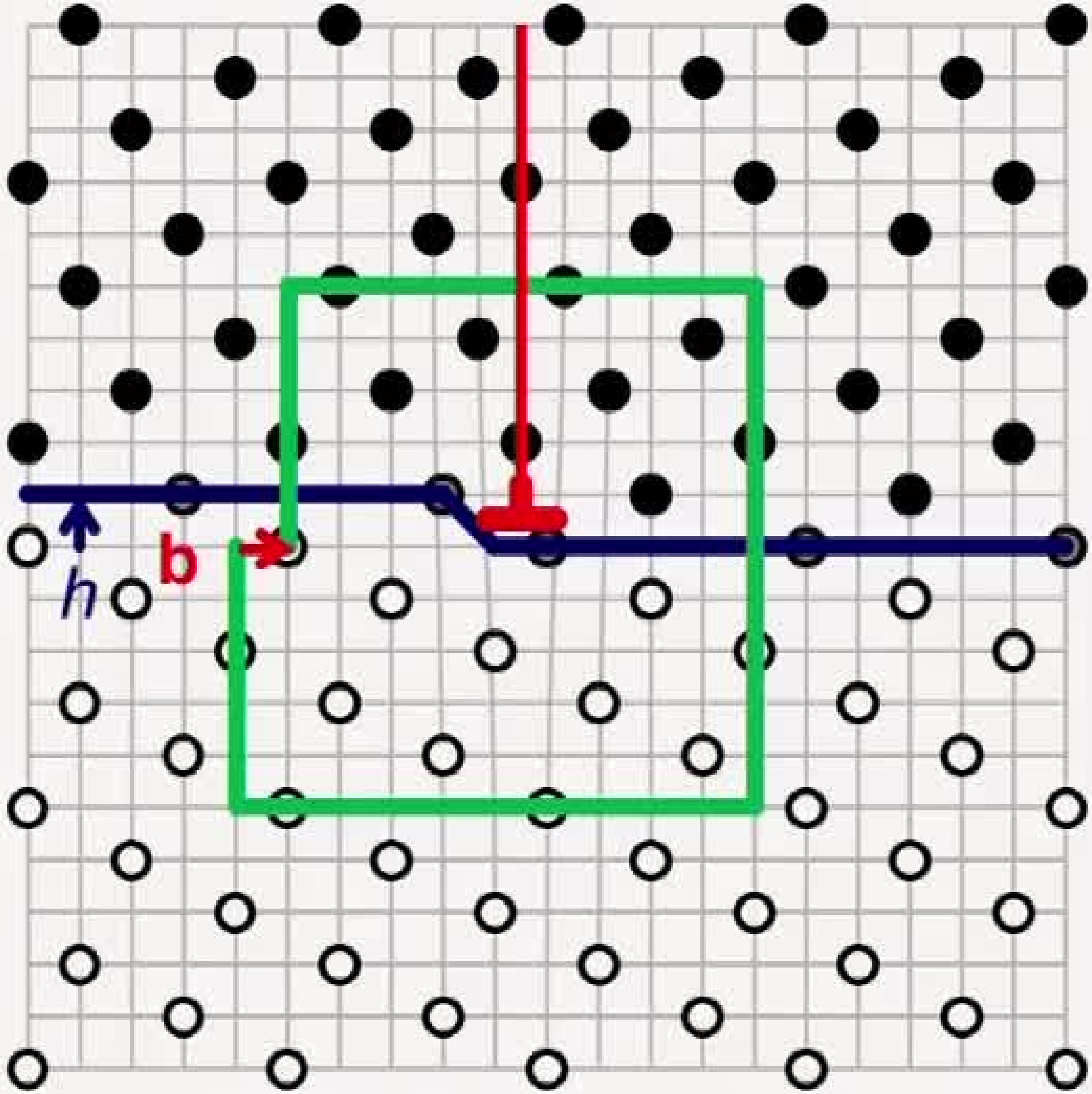
Real Grain Boundaries

- Now, instead of surfaces, we focus on grain boundaries – interfaces between misoriented crystals

- Like surfaces, GBs have steps (characterized by *h*)
- These steps don't fit perfectly into the additional crystal lattice above GB → extra half plane → elastic distortion → dislocations (characterized by Burgers vector, **b**)

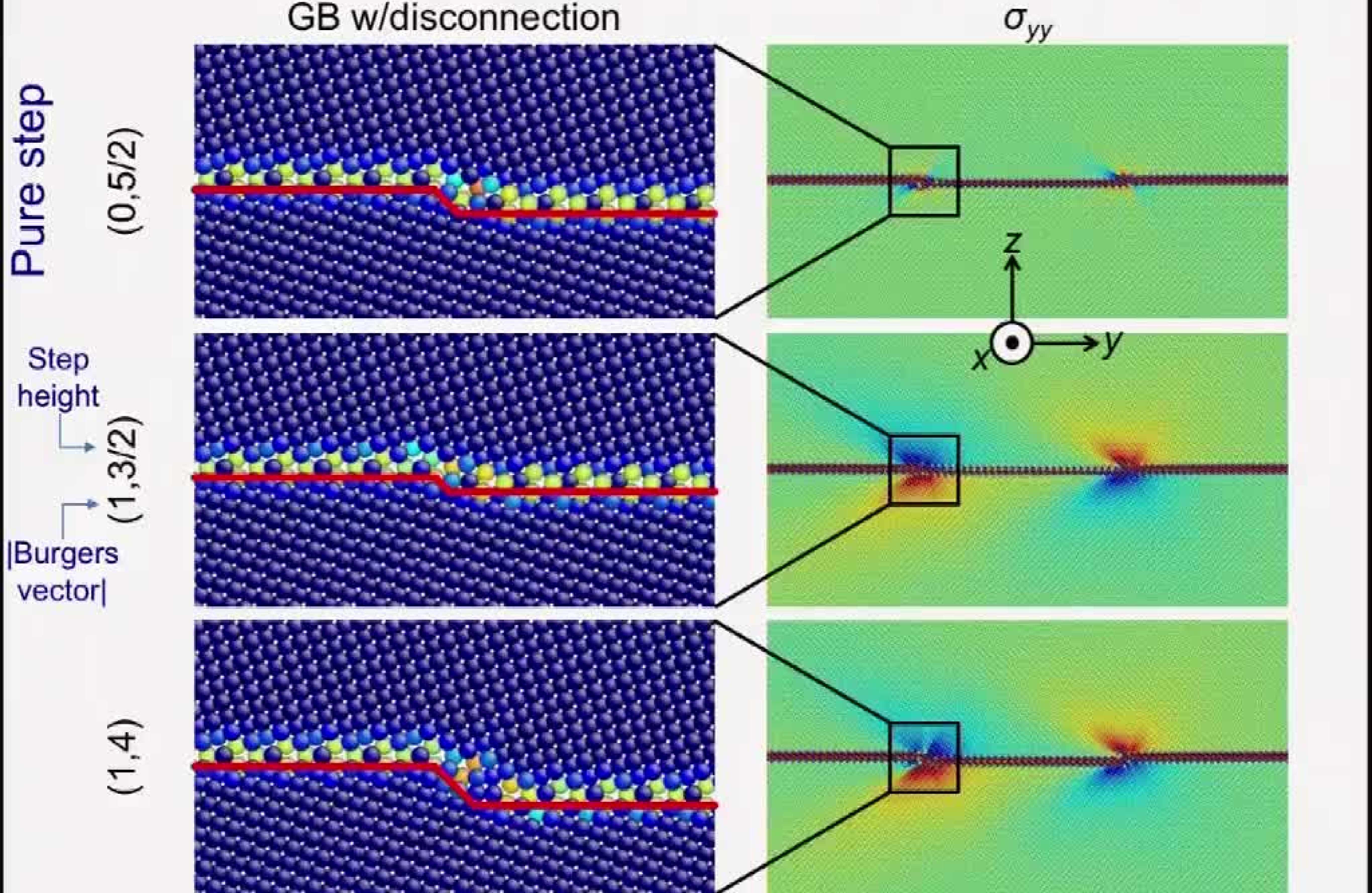
$$\mathbf{b} = \oint \frac{\partial \mathbf{u}}{\partial s} ds$$

- This defect → disconnection; characterized by (**b**,h)

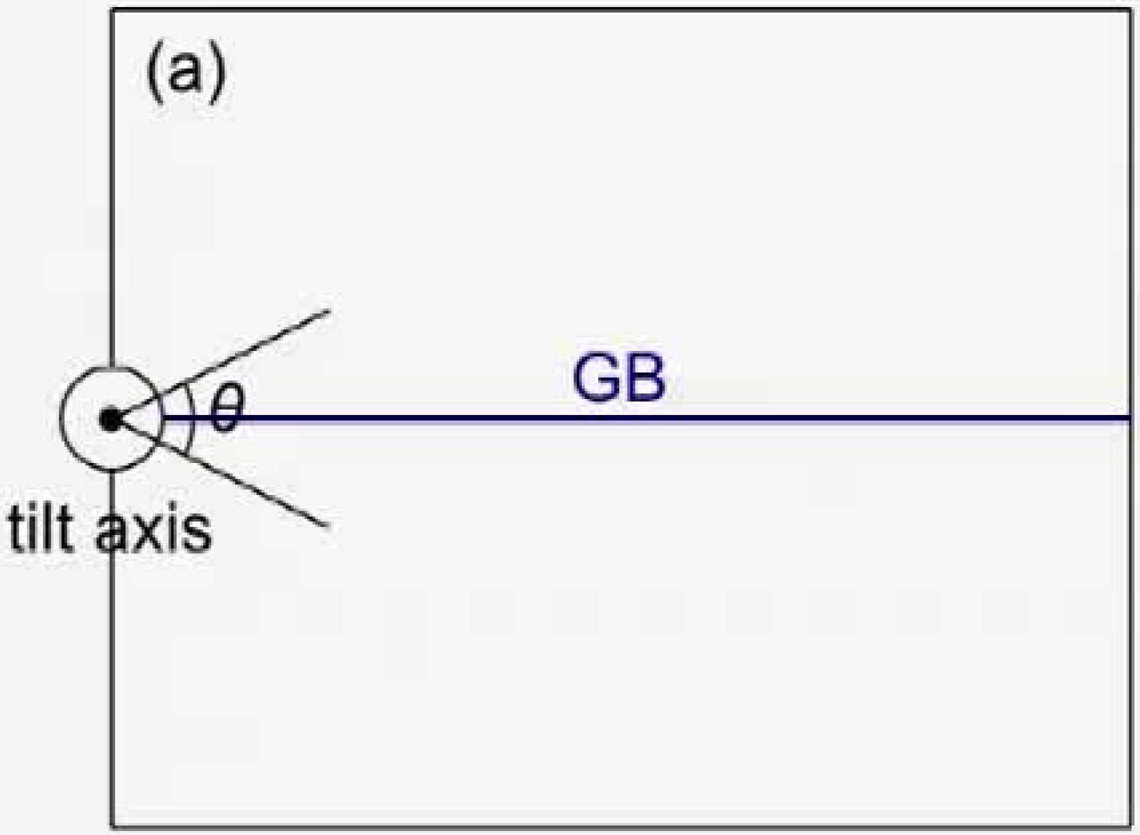


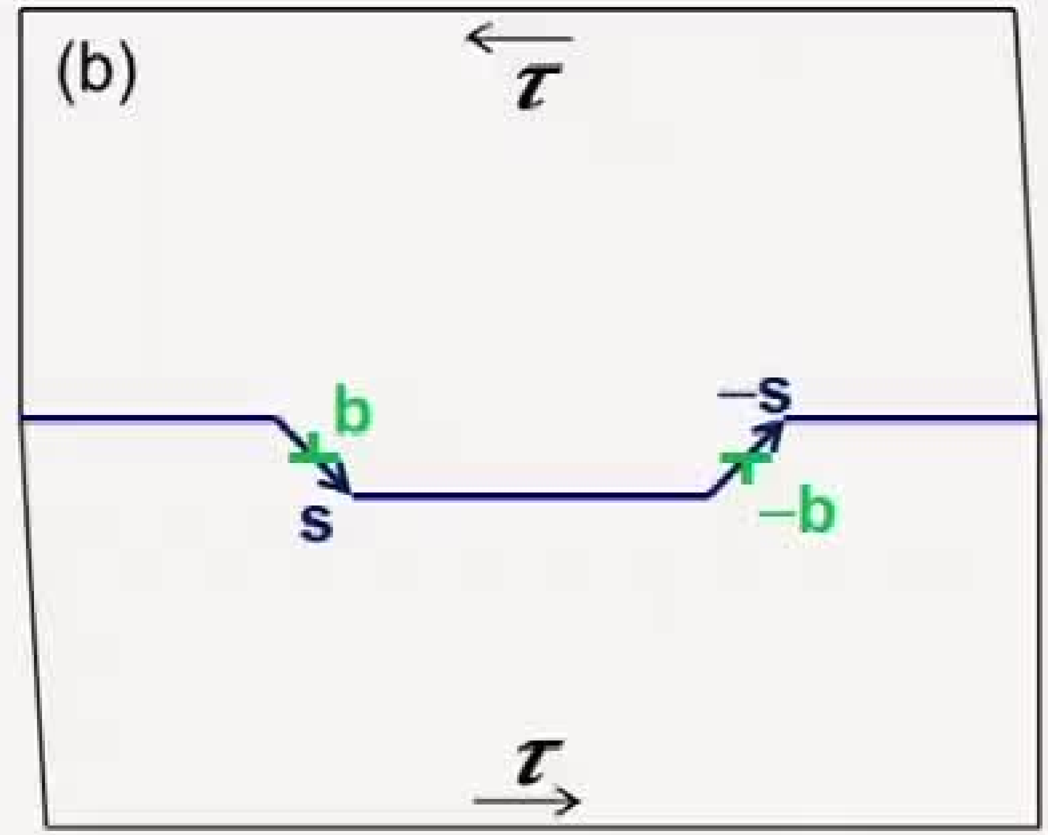
Disconnections

Molecular Dynamics (MD) simulation: Σ5 [100] θ = 36.87° STGB in EAM Cu

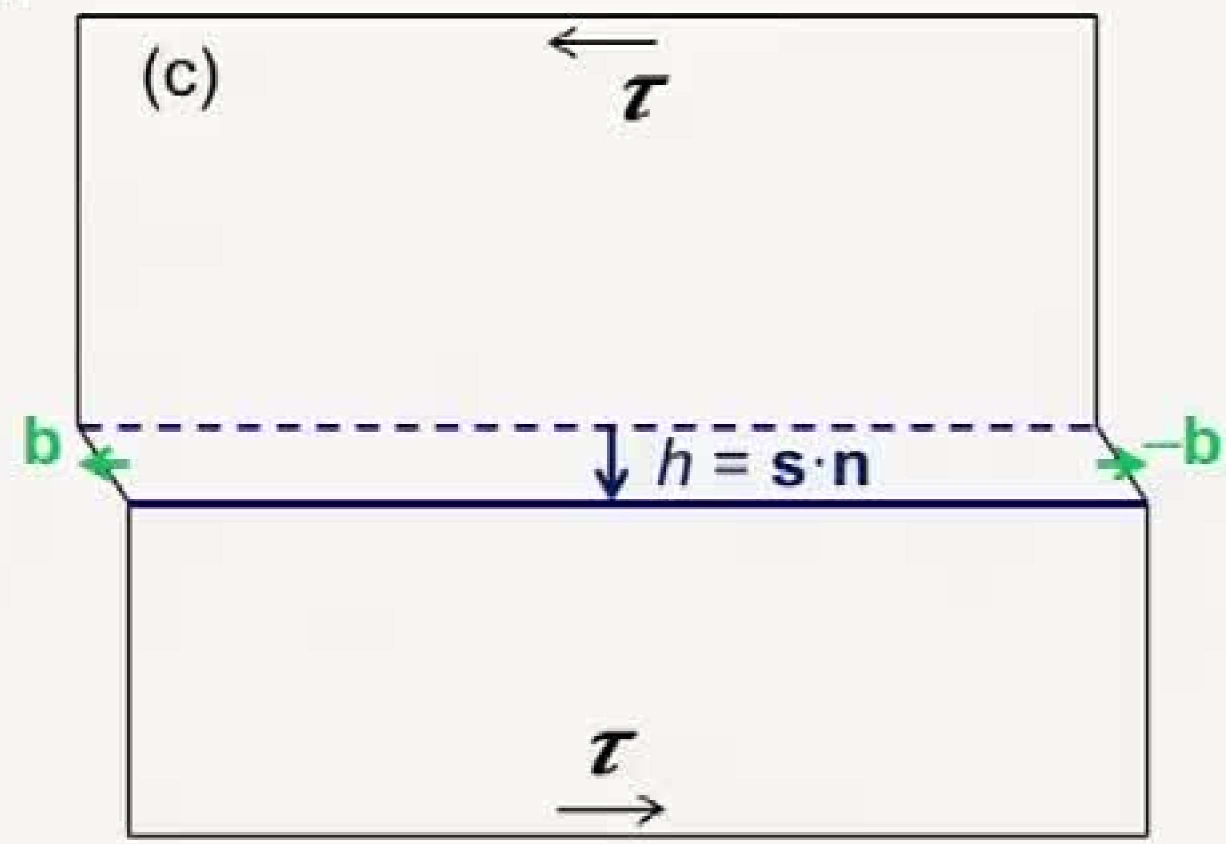


Disconnections: GB Migration & Shear





Shear → nucleation of a a DSC dislocation dipole with $\mathbf{b}\cdot\mathbf{n} = 0$



Coupling factor:

$$\beta = v_{||}/v_n = |\mathbf{b}|/h$$

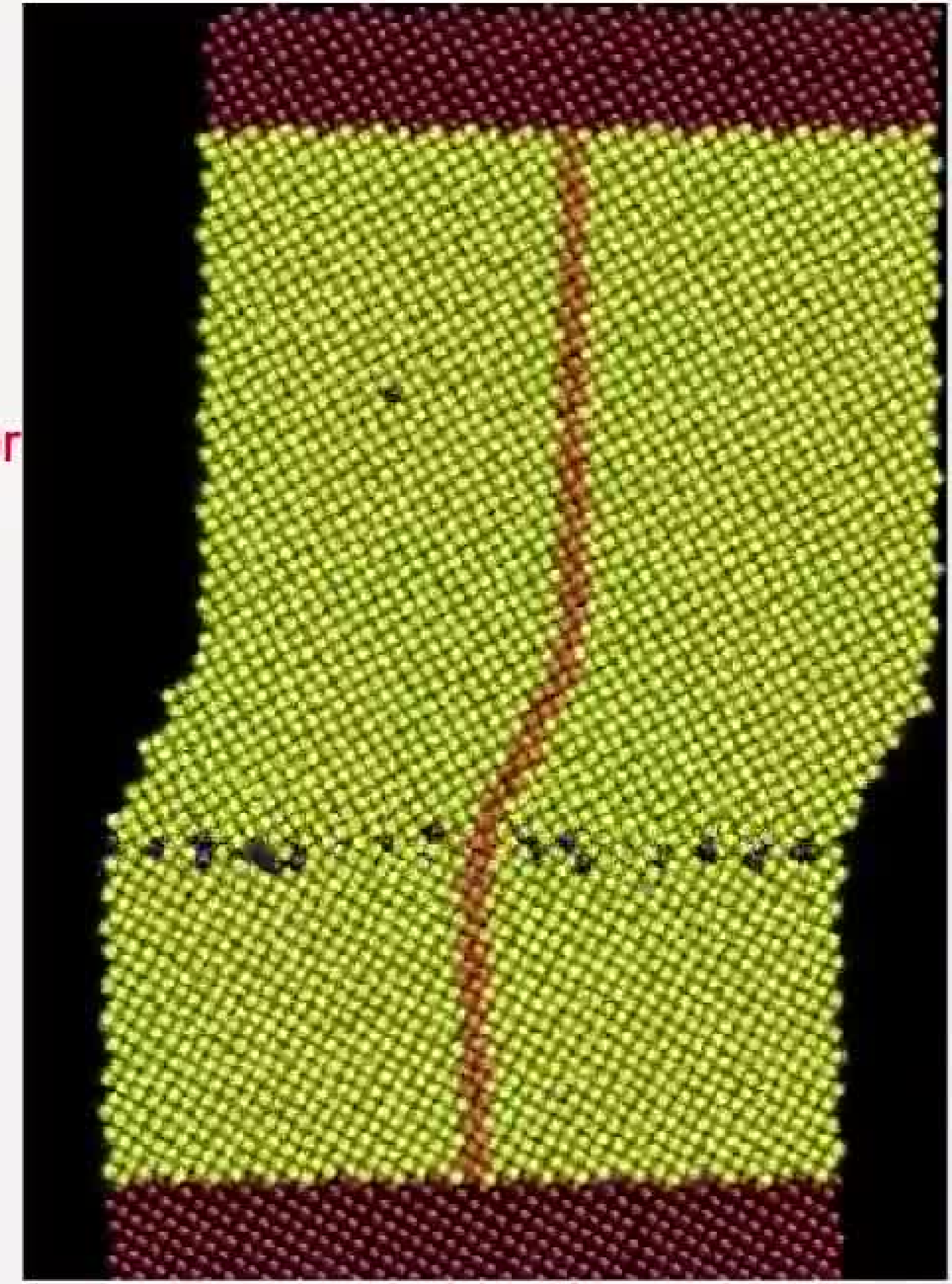
Rae, Smith, Philos. Mag. A**41** (1980)

Stress-Driven GB Dynamics

An example of stress-driven (shear-coupled) migration

Shear coupling factor

$$\beta = |\mathbf{b}|/h$$



Constant Shear Rate MD simulation in Cu

Σ17(530) [001] 61.9°

Cahn, Mishin, Suzuki 2006

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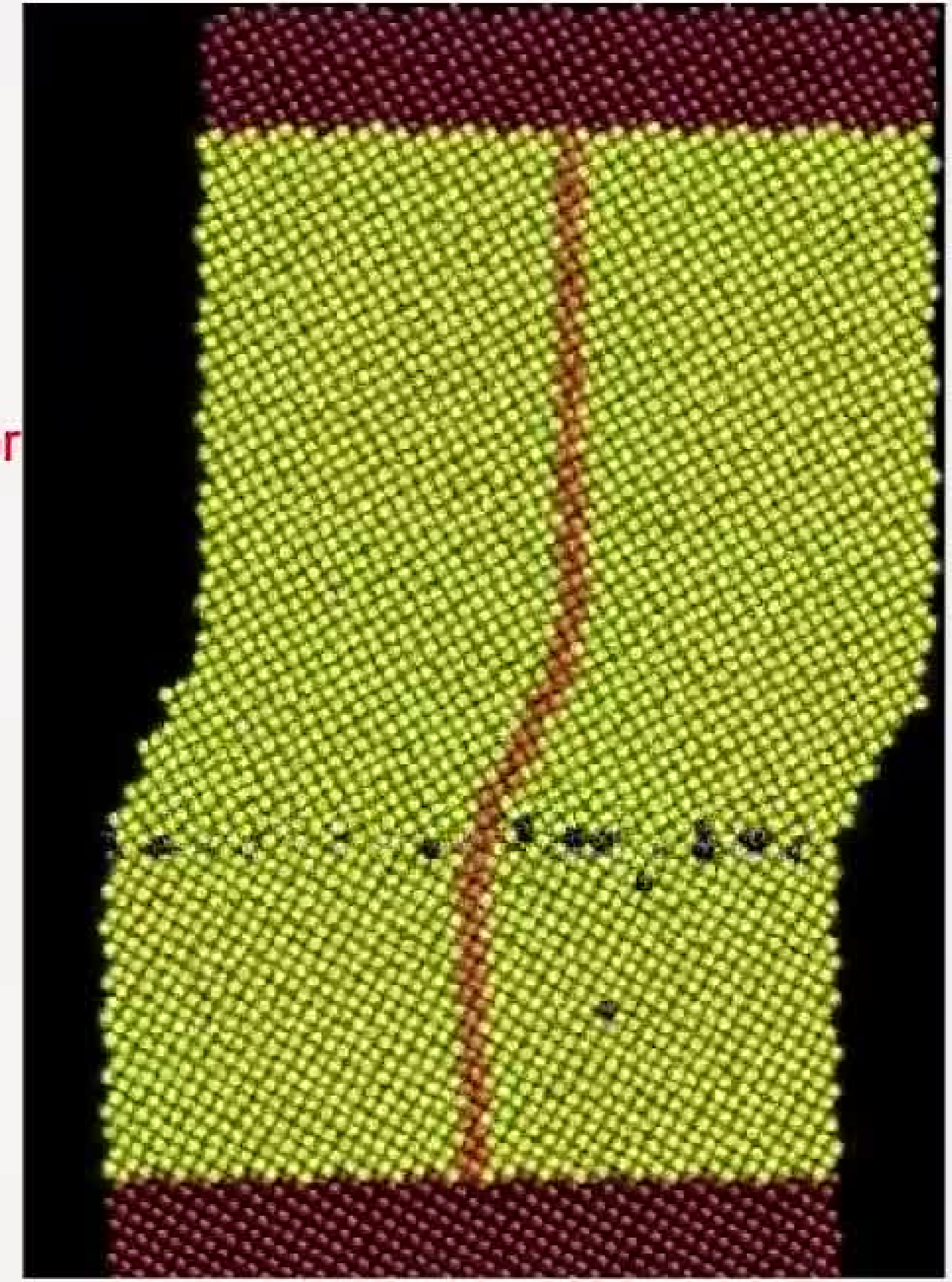

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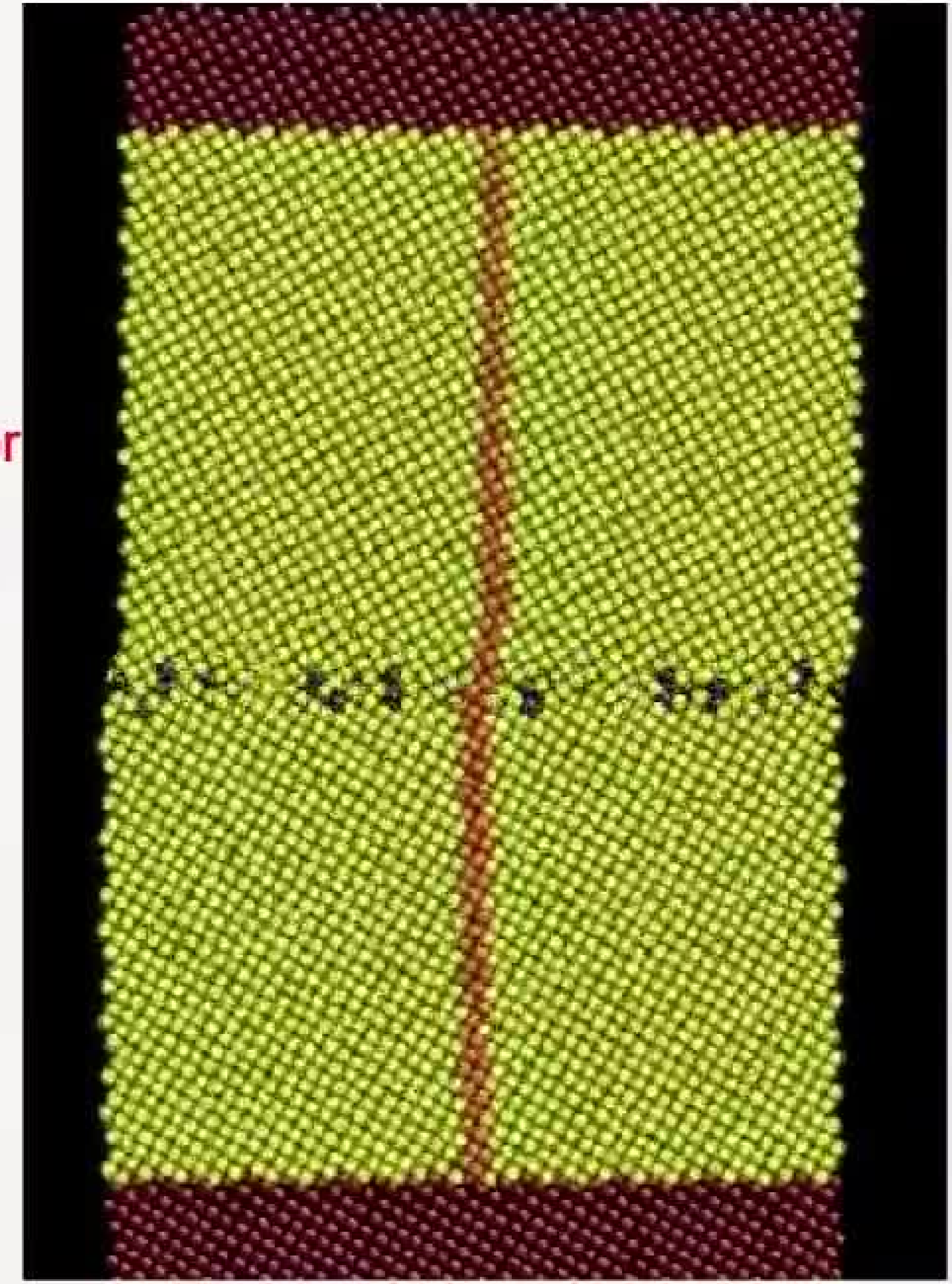

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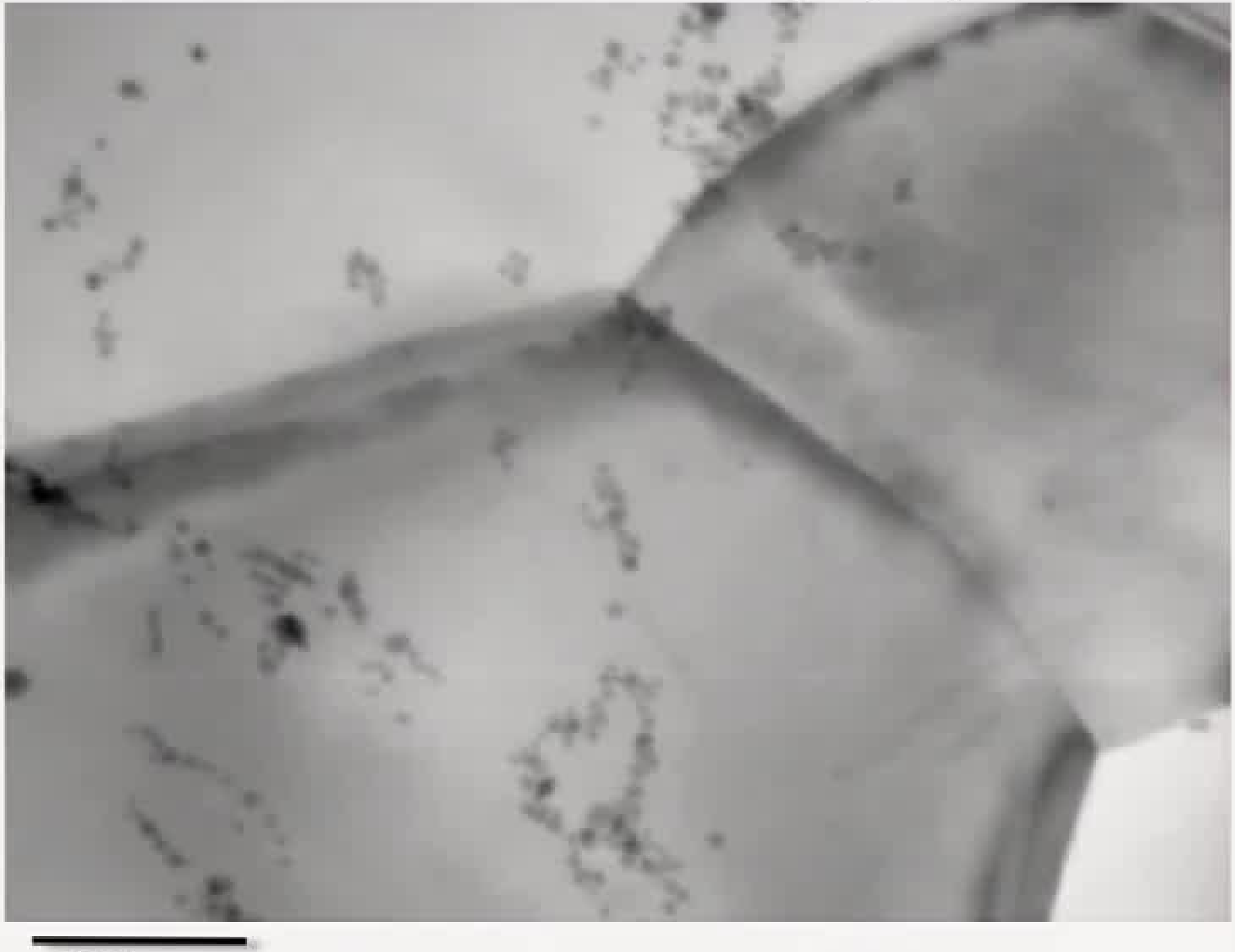
GB Migration is Step Motion

TEM movie of stressed thin film of polycrystalline Al at 420˚C



200 nm

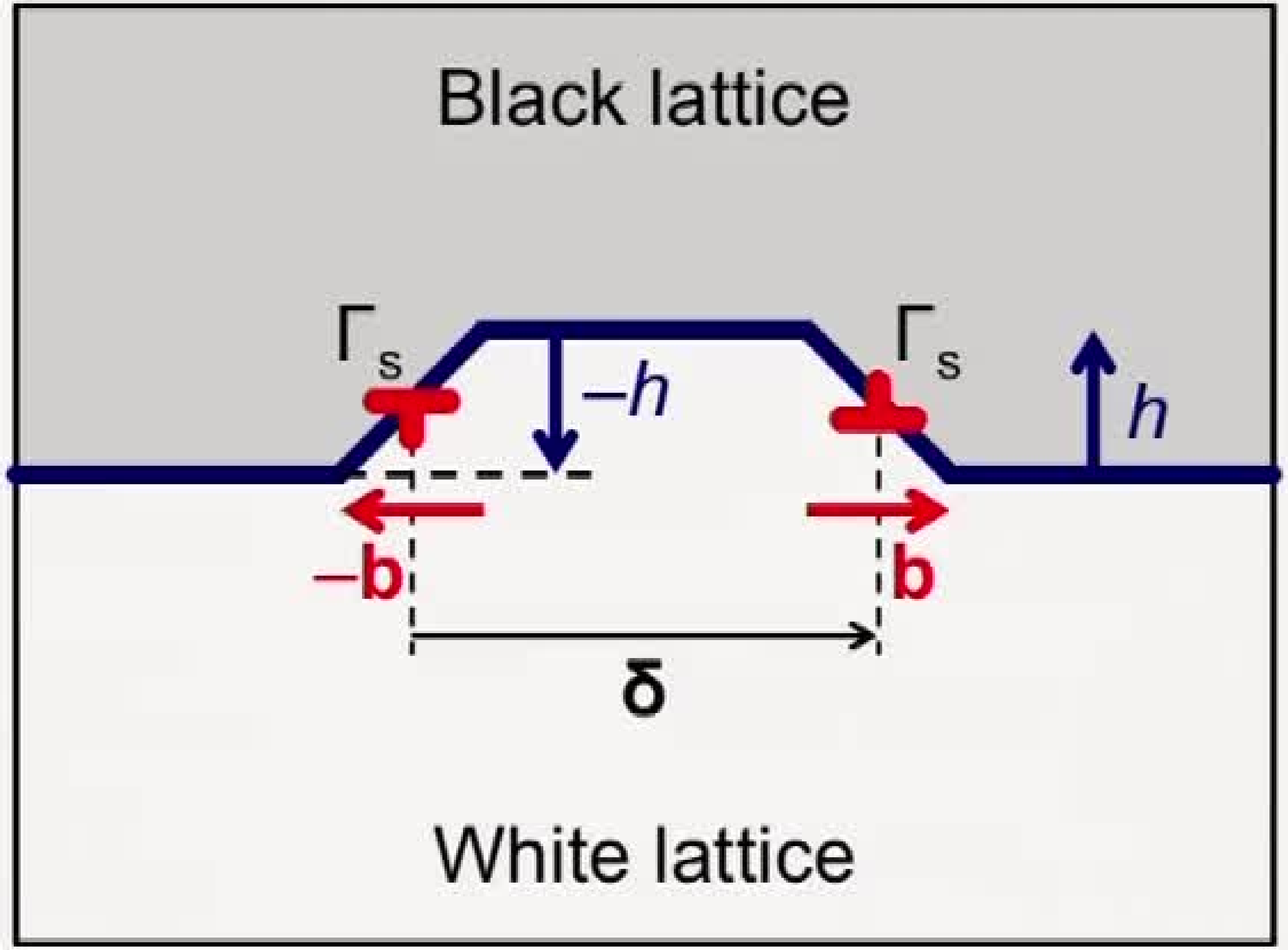
Tensile axis

Rajabzadeh, Legros, Combe, Mompiou, Molodov
Philosophical Magazine **93**, 1299 (2013)

How do GB move?

- **Summary:**
  - GBs move by formation & motion of disconnections
  - Disconnections: line defects that exist only at interfaces
  - Disconnections: both dislocation **b** and step character *h*
  - Bicrystallography tells us that **b** can be any translation vector consistent with both crystals (depends only on type & misorientation of two crystals)
  - Possible step heights *h* are set by the **b** and GB inclination
  - **b** and *h* are conserved quantities
  - Couples stress and chemical potential jump driving forces

Disconnections Pair Energy

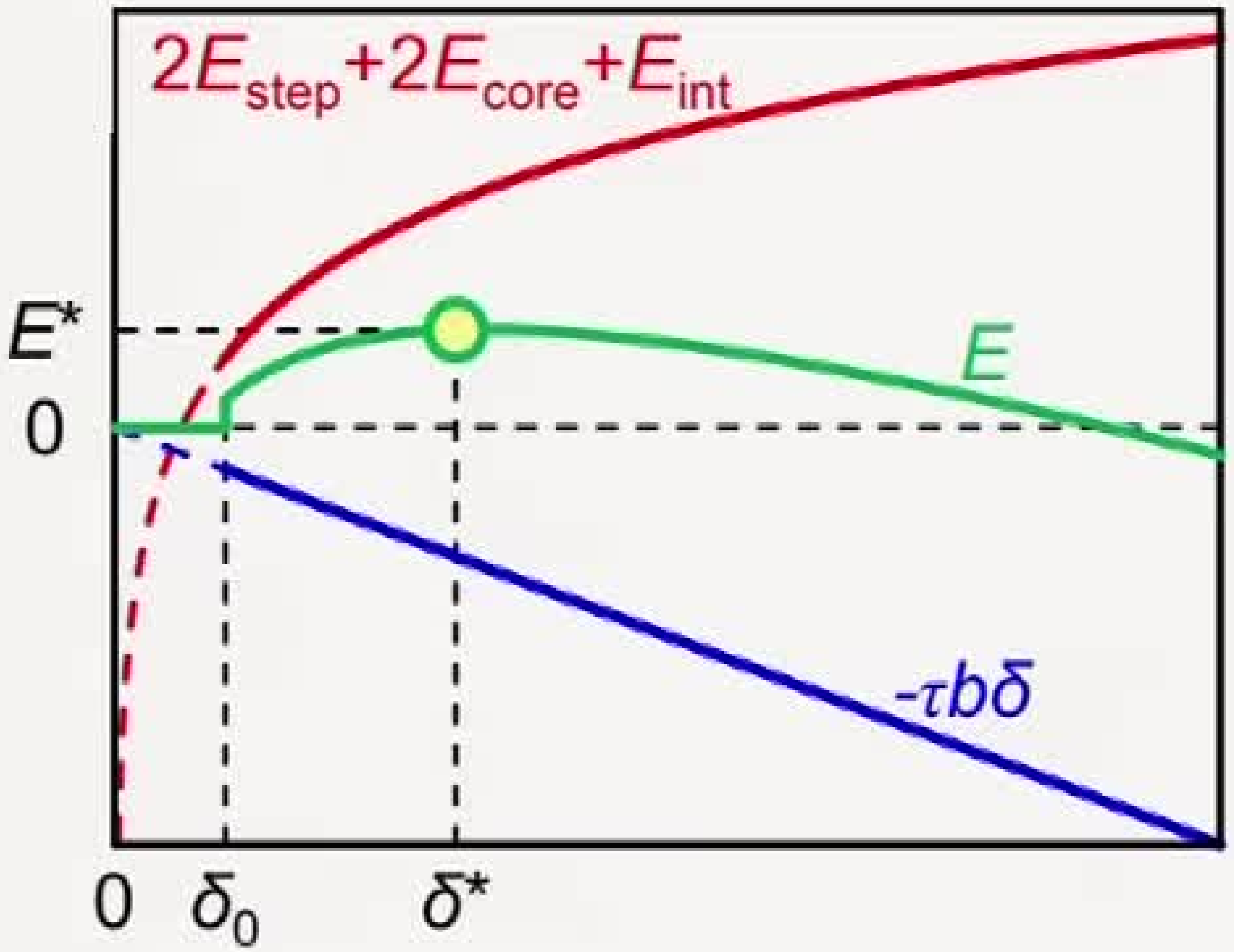


$$E = 2E_{\text{step}} + 2E_{\text{core}} + E_{\text{int}}$$

$$E_{\text{step}} = \Gamma_s \left|h\right|,\ E_{\text{core}} = \zeta K b^2,\ E_{\text{int}} = 2Kb^2 \ln\frac{\delta}{r_0}$$

Disconnection Nucleation-Controlled Migration

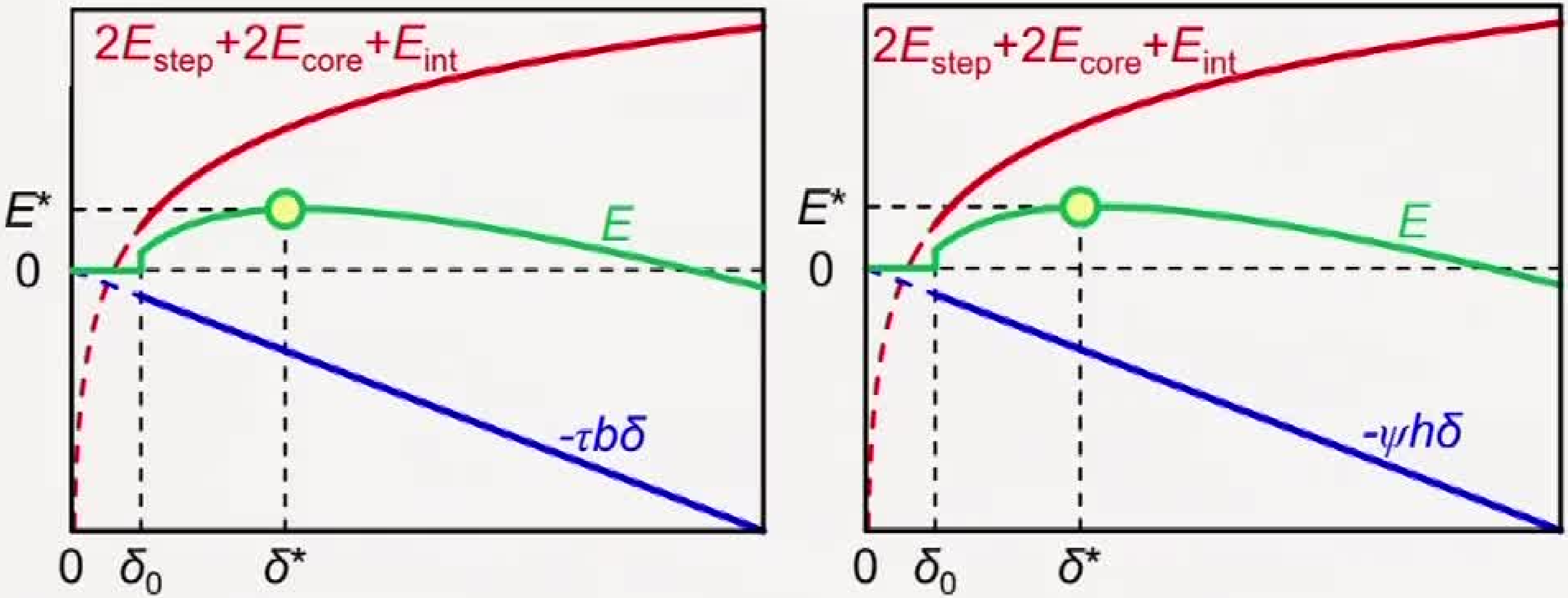
$$E = 2E_{\text{step}} + 2E_{\text{core}} + E_{\text{int}} \boxed{-\tau b\delta}$$



- Stress <u>couples</u> to $|\mathbf{b}_i|$

Disconnection Nucleation-Controlled Migration

$$E = 2E_{\text{step}} + 2E_{\text{core}} + E_{\text{int}} - \tau b \delta \qquad E = 2E_{step} + 2E_{core} + E_{int} - \psi h \delta$$



- Stress couples to $|\mathbf{b}_i|$
- Chemical potential jump driving force couples to $h_{i,j}$

Disconnection Nucleation-Controlled Migration

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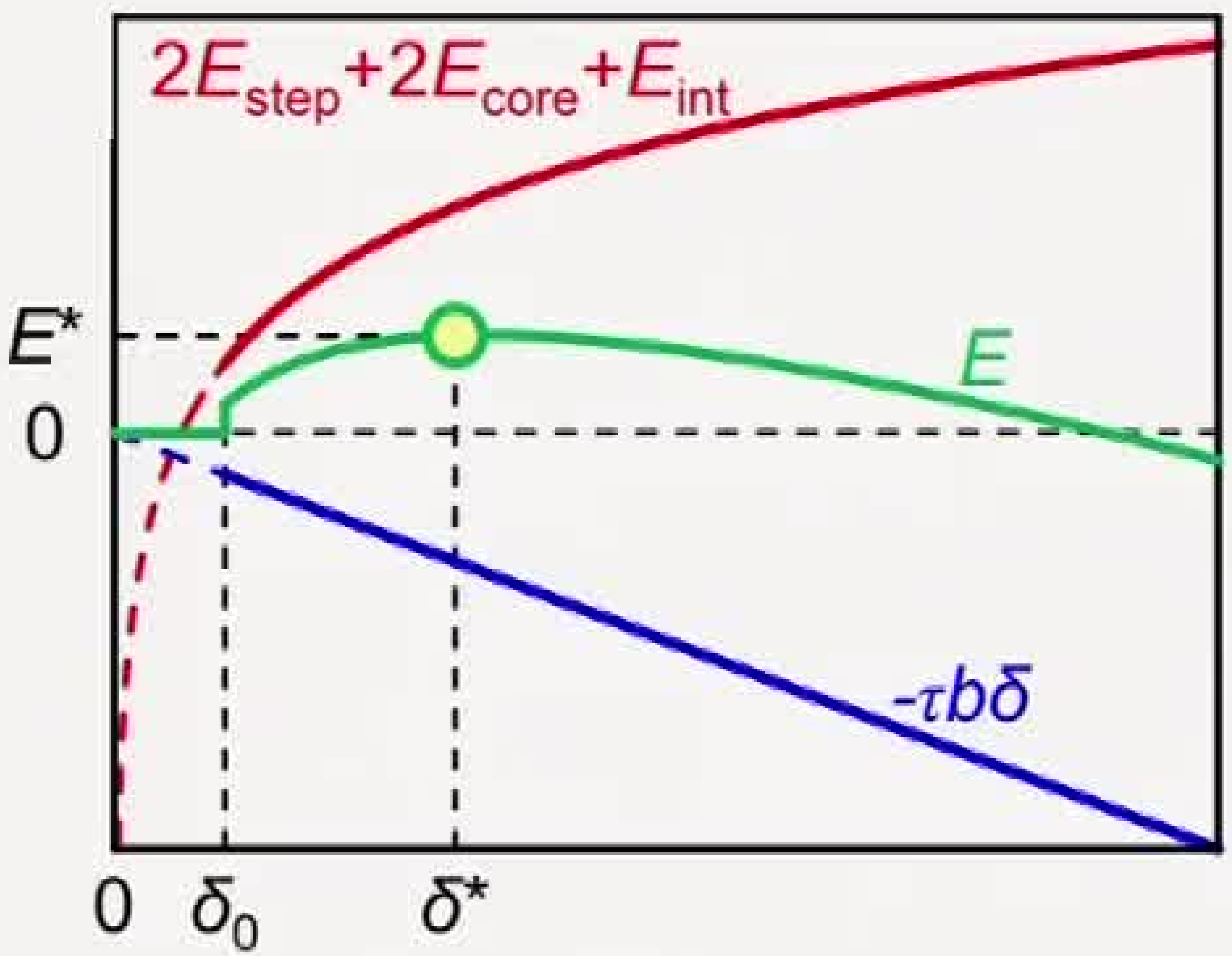
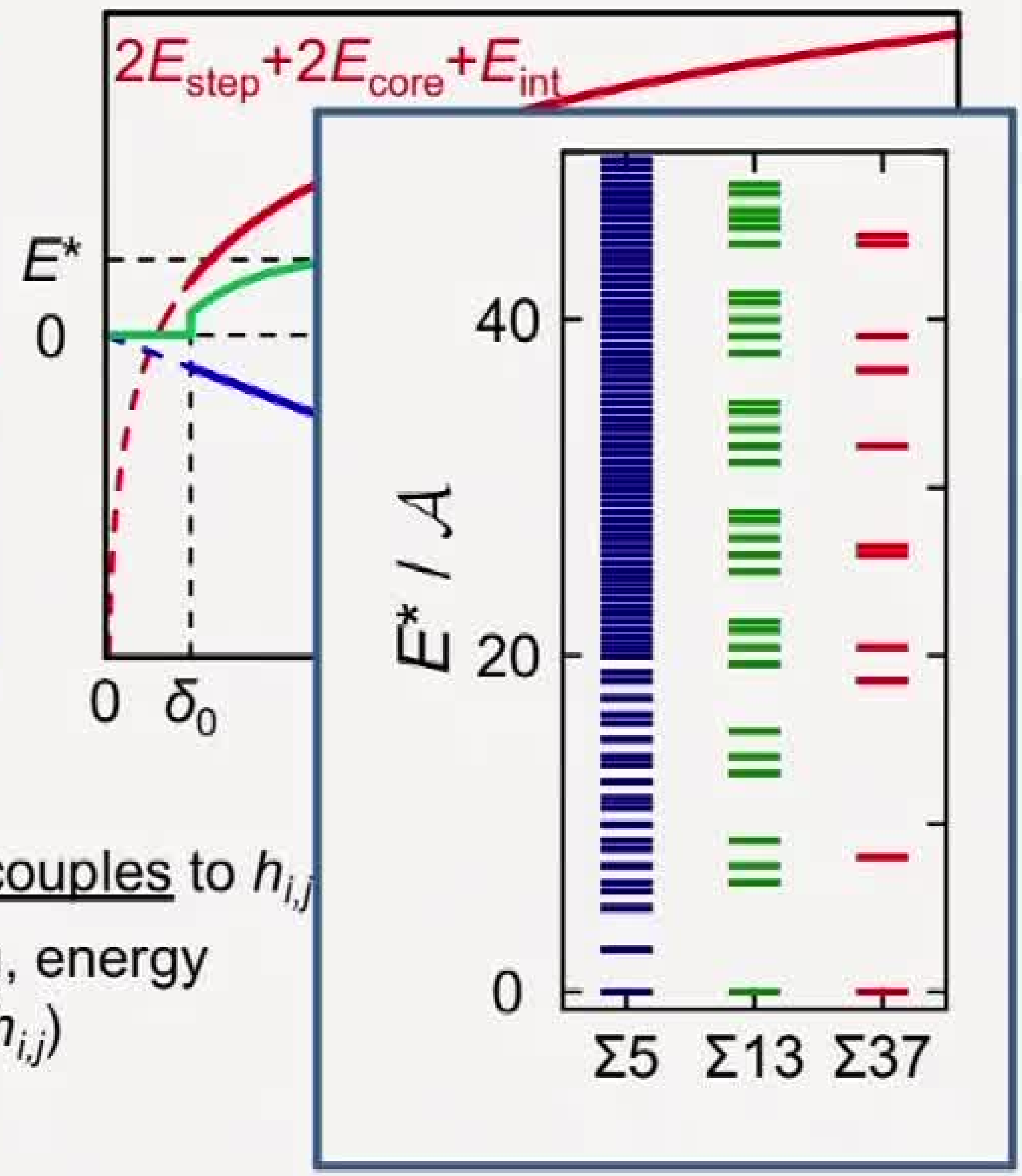
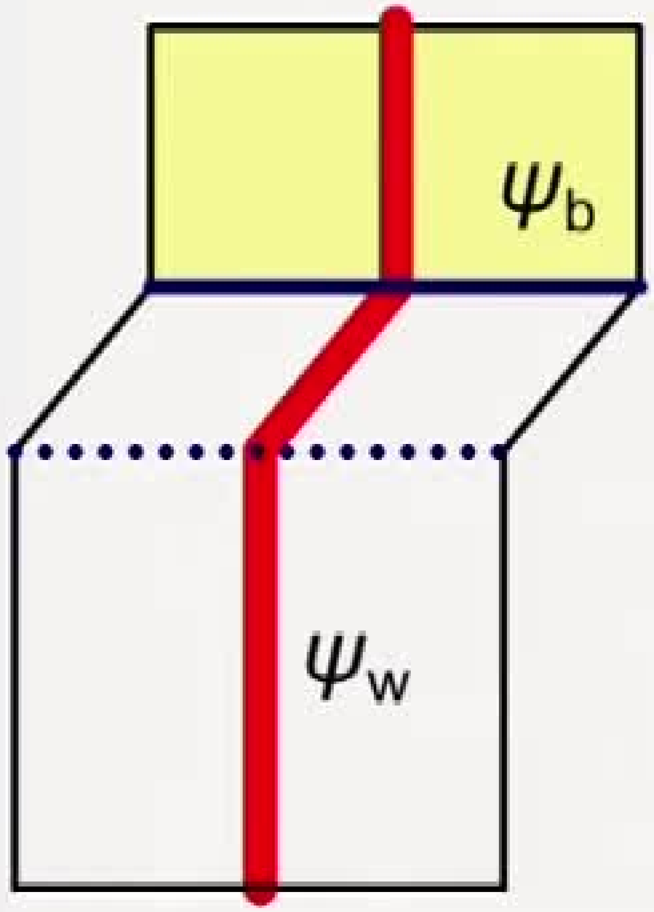




- Stress couples to $|\mathbf{b}_i|$
- Chemical potential jump driving force couples to $h_{i,j}$
- Nucleation favors small $|\mathbf{b}|$ (w/o driving, energy $\sim|\mathbf{b}_i|^2$) & small h (w/o driving, energy $\sim h_{i,j}$)
- Probability $P_{i,j} \sim \exp(-E^*_{i,j}/kT)$

GB Migration: chemical potential jump ψ

Σ39 32.2˚ symmetric tilt GB (free ends)



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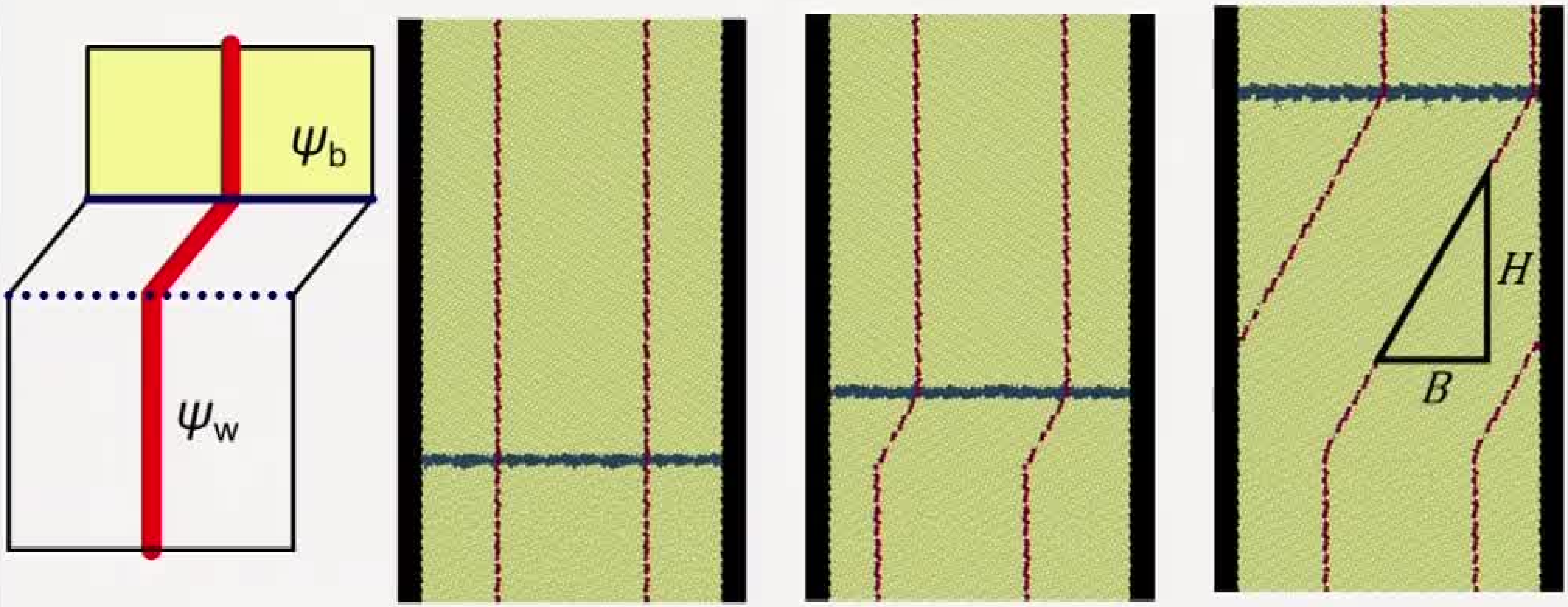
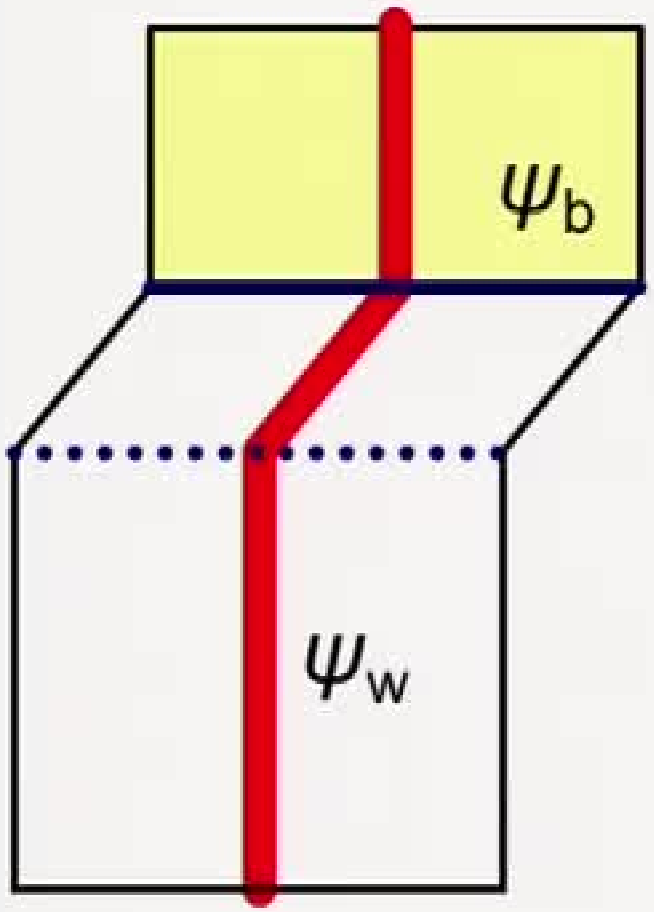


- Measure slope → shear rate/migration rate: $\beta = v_{||}/v_n = B/H = 0.58$
- The corresponding (b,h) is the bicrystallography allowed $\{\mathbf{b}_i, h_{i,j}\}$ with the smallest $E_{i,j}^*$

Thomas, Chen, Han, Purohit, DJS, Nature Comm. (2017)

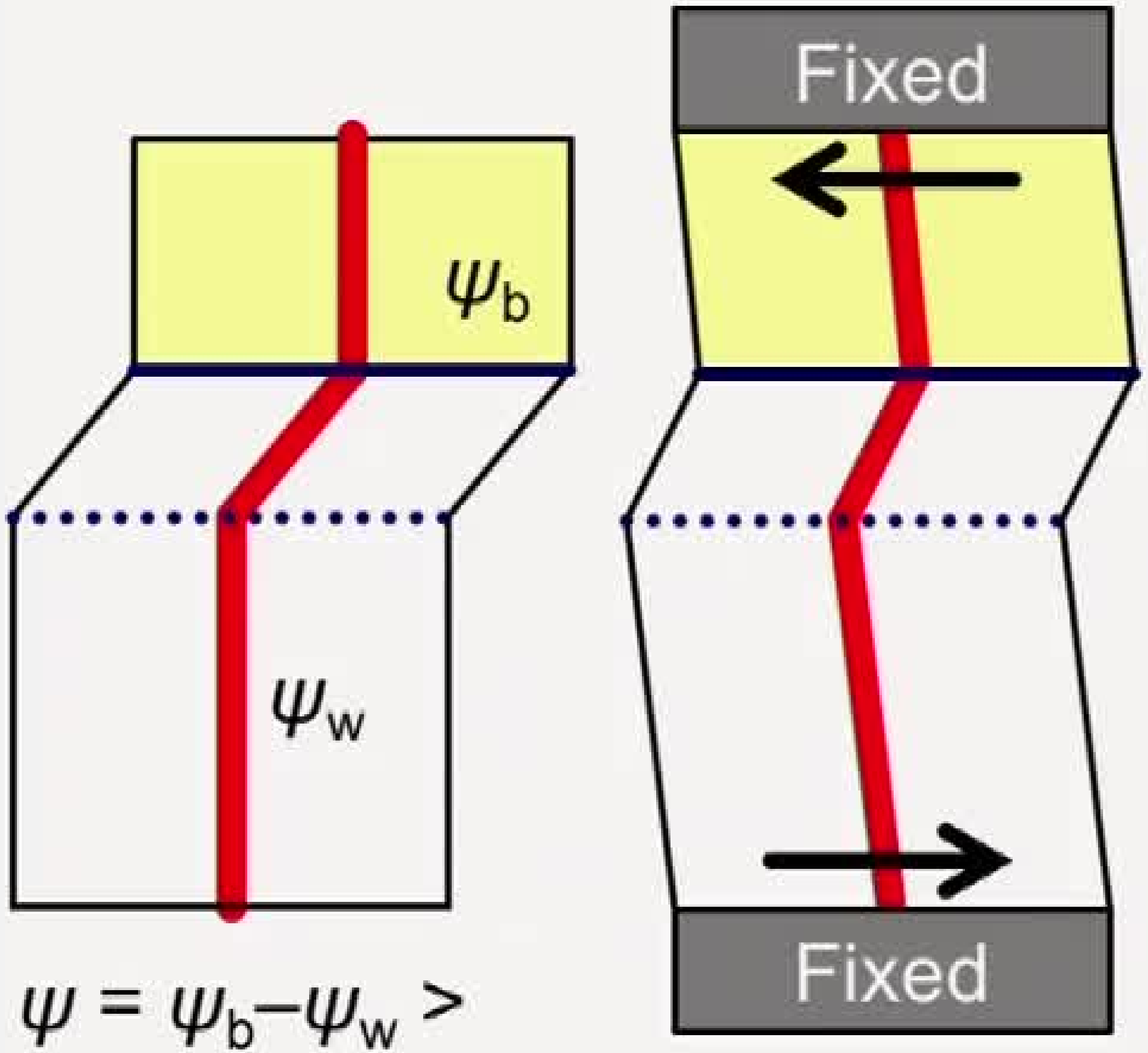
Constrained GB Migration



$\psi = \psi_b - \psi_w > 0$

Unconstrained
Shear coupling

Constrained GB Migration



Unconstrained
Shear coupling

Relaxation
(stagnation)

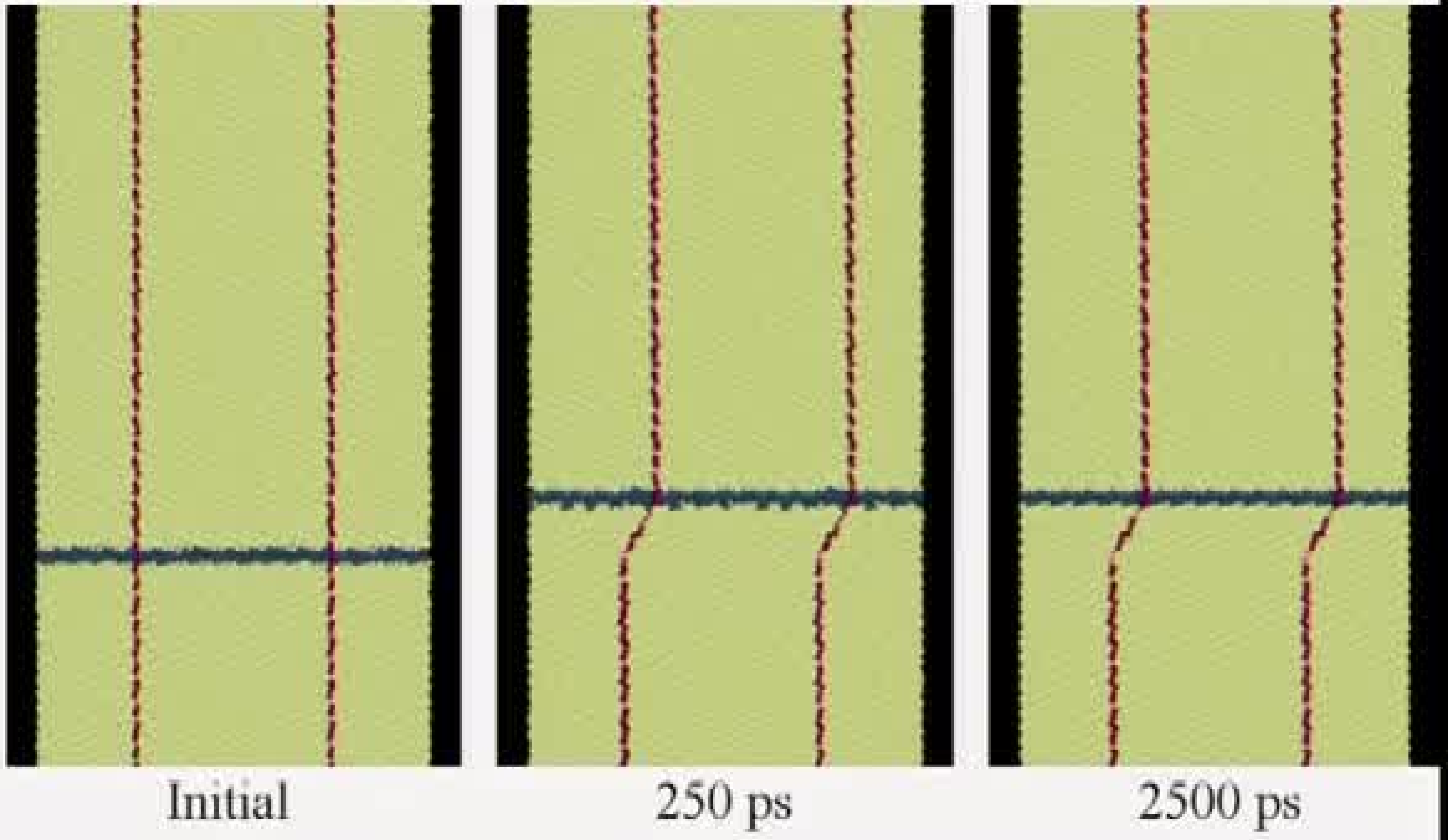
Repeat GB migration simulations using a chemical potential jump driving force, but now, keep the ends fixed

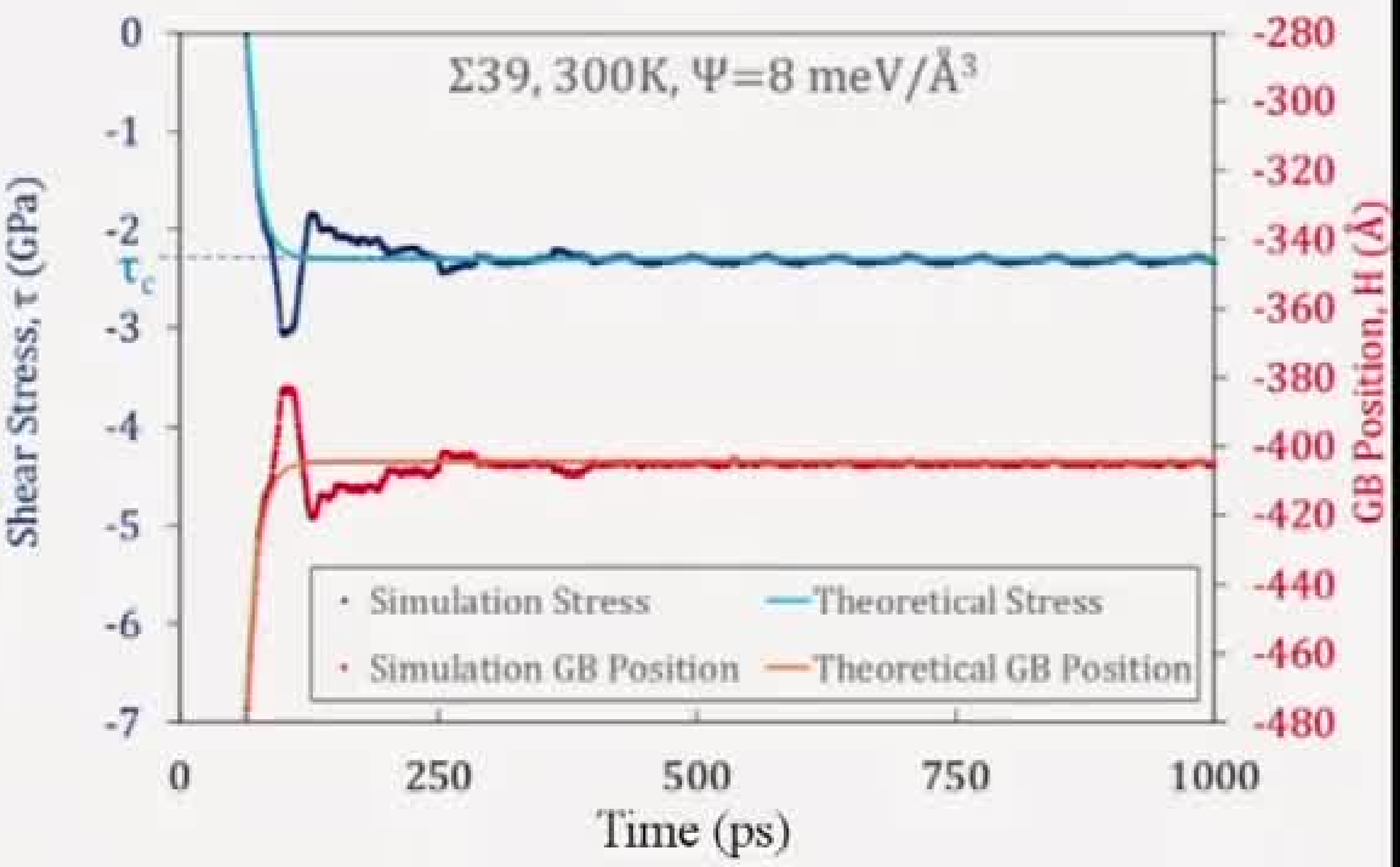
GB migrates under chemical potential jump driving force

Shear coupling means stress develops

Constrained GB Migration

Exactly same GB as previous case but w/ top & bottom edges fixed (rather than free)





Boundary migrates, then stops

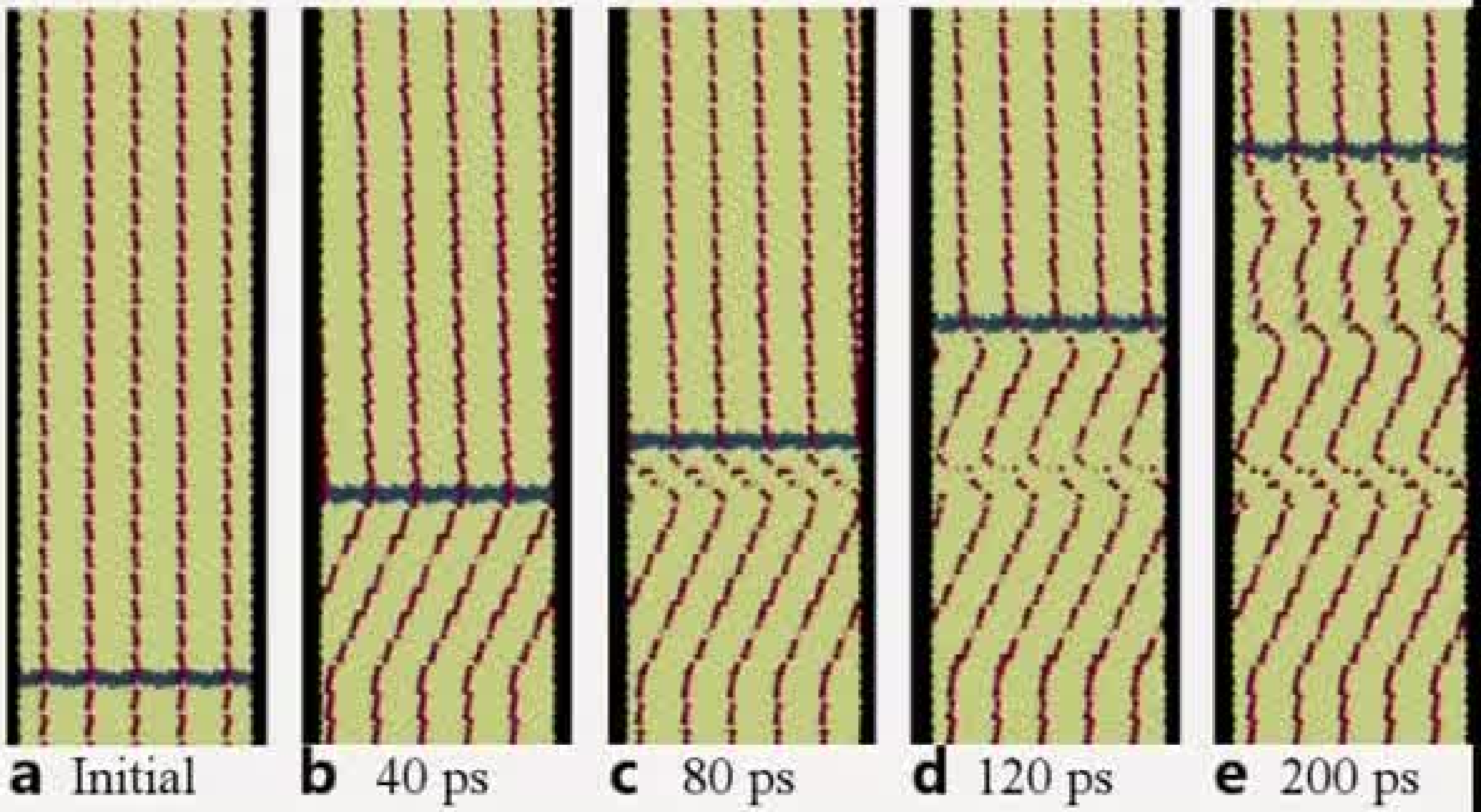
Stresses build during migration

Elastic driving force cancels chemical potential jump driving force

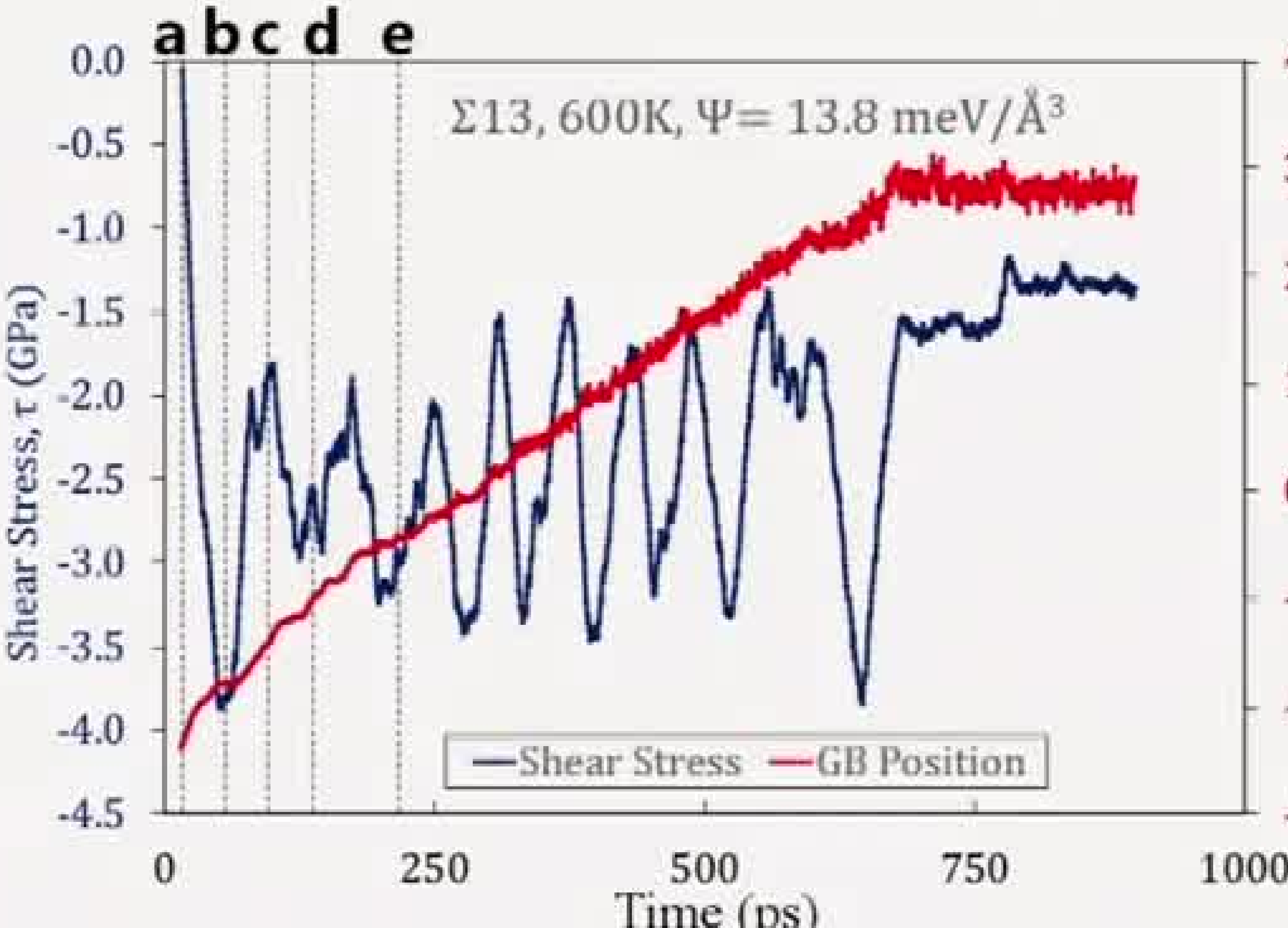
Predict this happens at $\tau = \tau_c$

Constrained GB Migration

Repeat fixed end simulations but with a different GB
$\Sigma 13\ [111]\ (3\bar{4}1)$



- Boundary migrates long distance
- Switches coupling modes as it migrates
- Stresses rise or fall with migration depending on the sign of the coupling mode of the moment
- Net, constant GB migration rate/mobility



Multiple Coupling Modes

- When there are multiple coupling modes, $B \neq b$, $H \neq h$, $\beta(T)$
- The actual shear and migration rates depend on driving force & T

$$\dot{B} = \frac{\omega w}{kT}\left(\tau \sum_i b_i^2 e^{-E_i/kT} + \Psi \sum_i h_i b_i e^{-E_i/kT}\right) = K_{11}\tau + K_{12}\Psi$$

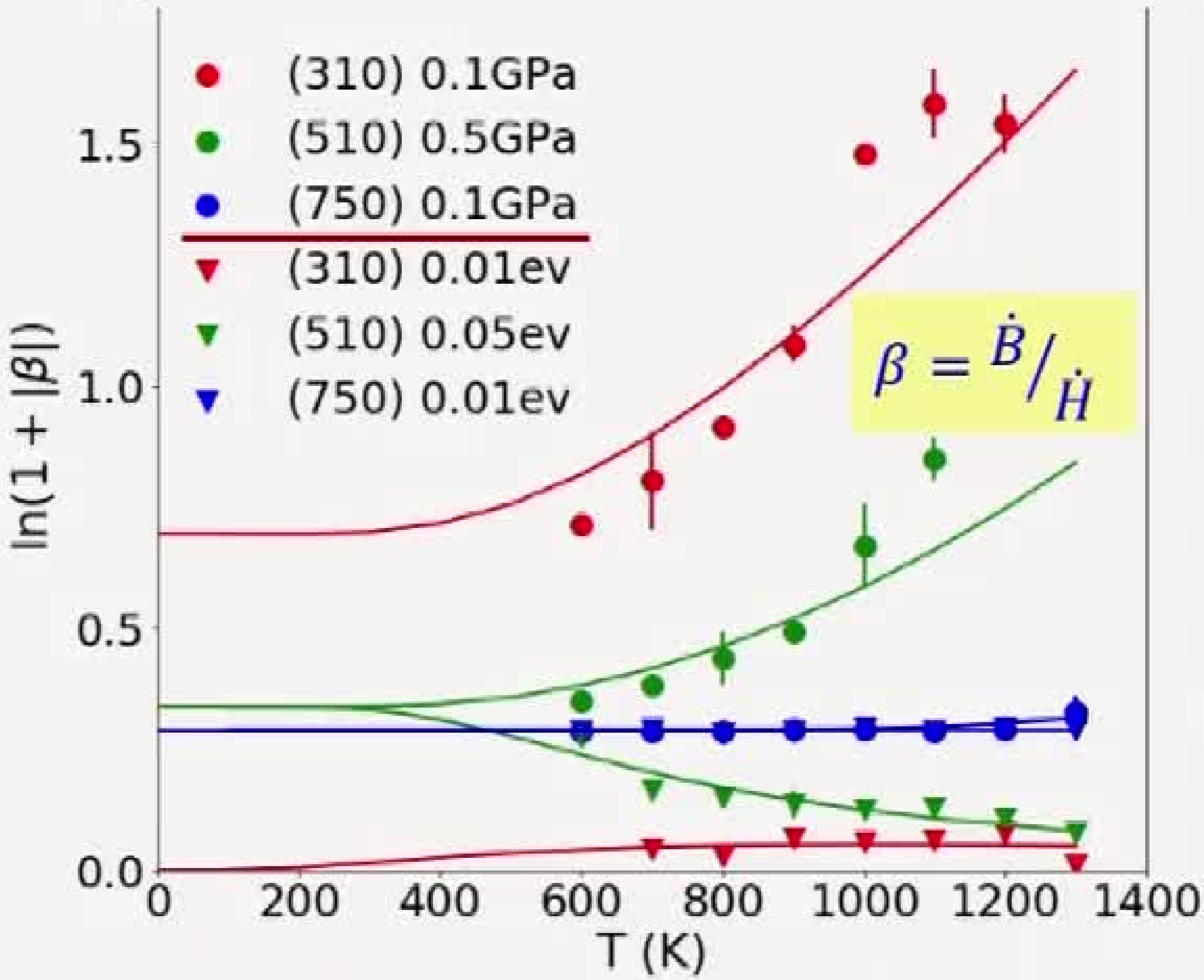
$$\dot{H} = \frac{\omega w}{kT}\left(\tau \sum_i h_i b_i e^{-E_i/kT} + \Psi \sum_i h_i \; e^{-E_i/kT}\right) = K_{21}\tau + K_{22}\Psi$$

(linearized driving force)

- $K_{12} = K_{21}$: the K_{ij} are temperature-dependent Onsager coefficients

$$\frac{1}{\beta} = \frac{\sum_i h_i e^{-\frac{Q_i}{k_B T}} \sinh \frac{b_i S\tau - h_i S\Delta\psi}{k_B T}}{\sum_i b_i e^{-\frac{Q_i}{k_B T}} \sinh \frac{b_i S\tau - h_i S\Delta\psi}{k_B T}}$$

$$\frac{Q_i}{L} = Ab_i^2 + B|h_i|$$



MD + Theory
Chen, Thomas, Han, DJS (2018)

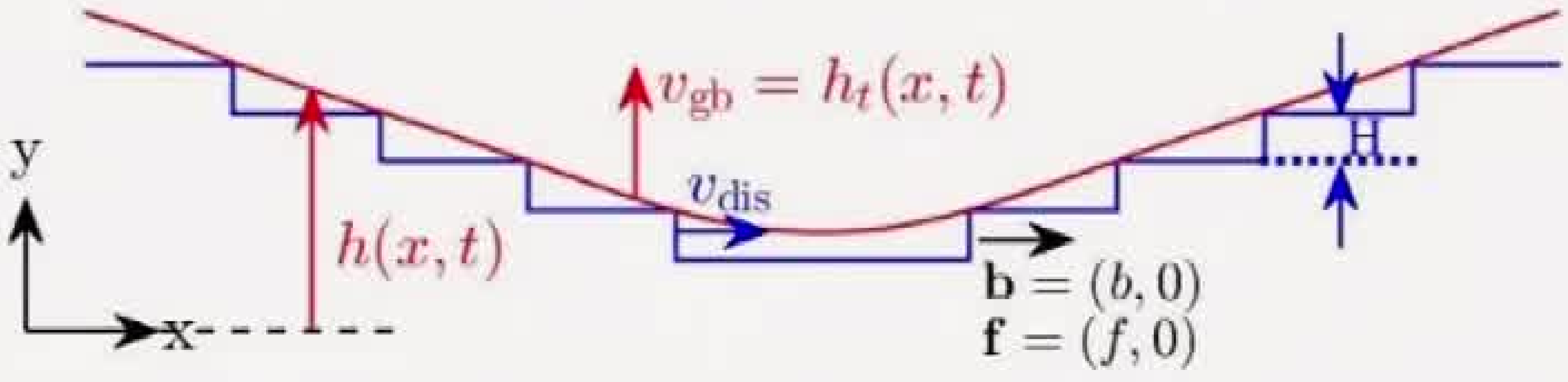
Towards a Continuum GB Equation of Motion

- Disconnection model works well to describe a wide range of GB kinetic phenomena – shear coupling, roughening, effects of different driving forces,… GB mobility (sometimes)
- But, too complex to describe GB migration in the “wild” (polycrystal) rather than “domesticated” GBs (bicrystal)
- Need a continuum equation of motion for GB – replace curvature flow

Zhang, Han, Xiang, DJS, Phys. Rev. Lett. (2017)

Continuum Model for GB Migration

- Replace discrete disconnection steps/dislocation with continuous disconnection density $\rho(x)$ for fixed coupling constant $\beta=b/H$
- Disconnection density is related to GB profile: i.e., $H\rho(x) = h_x(x)$



- Evolution of profile (pure kinematics):

$$h_t + v_d h_x = 0$$

- Disconnection velocity (assume overdamped disconnection motion):

$$v_d = M_d f_d$$

where M_d is the disconnection mobility and f_d is the total force on the disconnection – internal and external stresses, capillarity, chemical potenital driving forces,…

Continuum Model for GB Migration

- This leads to

$$h_t = -M_{\mathrm{d}}[(\sigma_i + \tau)b \ + \Psi H - \gamma h_{xx} H]|h_x|$$

- Term 1: the elastic interaction of the (Burgers vector) of the disconnections with a stress field – i.e., the Peach-Koehler force where σ_i is the stress field associated with other disconnections and τ is an applied stress)

$$\sigma_i = K \int_{-\infty}^{\infty} \frac{\beta h_x(x_1, t)}{x - x_1} dx_1; \qquad K = \frac{\mu}{2\pi(1-\nu)}$$

Continuum Model for GB Migration

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- Terms 2 &3: arise from the variation of the energy with respect to GB displacement $(\delta E / \delta z)$
- Term 3: associated with the decrease in GB energy that occurs upon the mutual annihilation of opposite disconnections as they random walk on the GB – this is the curvature flow term

Continuum Equation of Motion

- This does not yet address the question of where disconnections come from; source term g

$$h_t + v_d h_x = g$$

- Without a source term, a flat GB would always remain flat

Continuum Equation of Motion

- This does not yet address the question of where disconnections come from; source term g

$$h_{\mathrm{t}} + v_{\mathrm{d}} h_{\mathrm{x}} = g$$

- Without a source term, a flat GB would always remain flat
- Source: equilibrium thermal fluctuations in GB profile; i.e., equilibrium concentration of disconnections

$$c_e^+ c_e^- = \frac{1}{a^2} e^{-2F_d/k_B T} \text{ or } c_e = \frac{1}{a} e^{-F_d/k_B T}$$

where F_d is the nucleation barrier for the disconnection pair

- This yields a source term of the form

$$g = -2c_e H v_{\mathrm{d}} \frac{h_x}{|h_x|} \qquad B = (2H/a)\, e^{-F_d/k_B T}$$

- The final equation of motion is

$$h_t = -M_{\mathrm{d}}[(\sigma_i + \tau)b + \Psi H - \gamma h_{xx} H](|h_x| + B)$$

- The main temperature dependences are in B, M_{d}, and (b,H)

Continuum Equation of Motion

- Equation of motion

$$h_t = -M_{\mathrm{d}}[(\sigma_i + \tau)b + \Psi H h_x - \gamma_{GB} h_{xx} H](|h_x| + B)$$

where $B = c_e H = (2H/a)\exp(-F_d/kT)$

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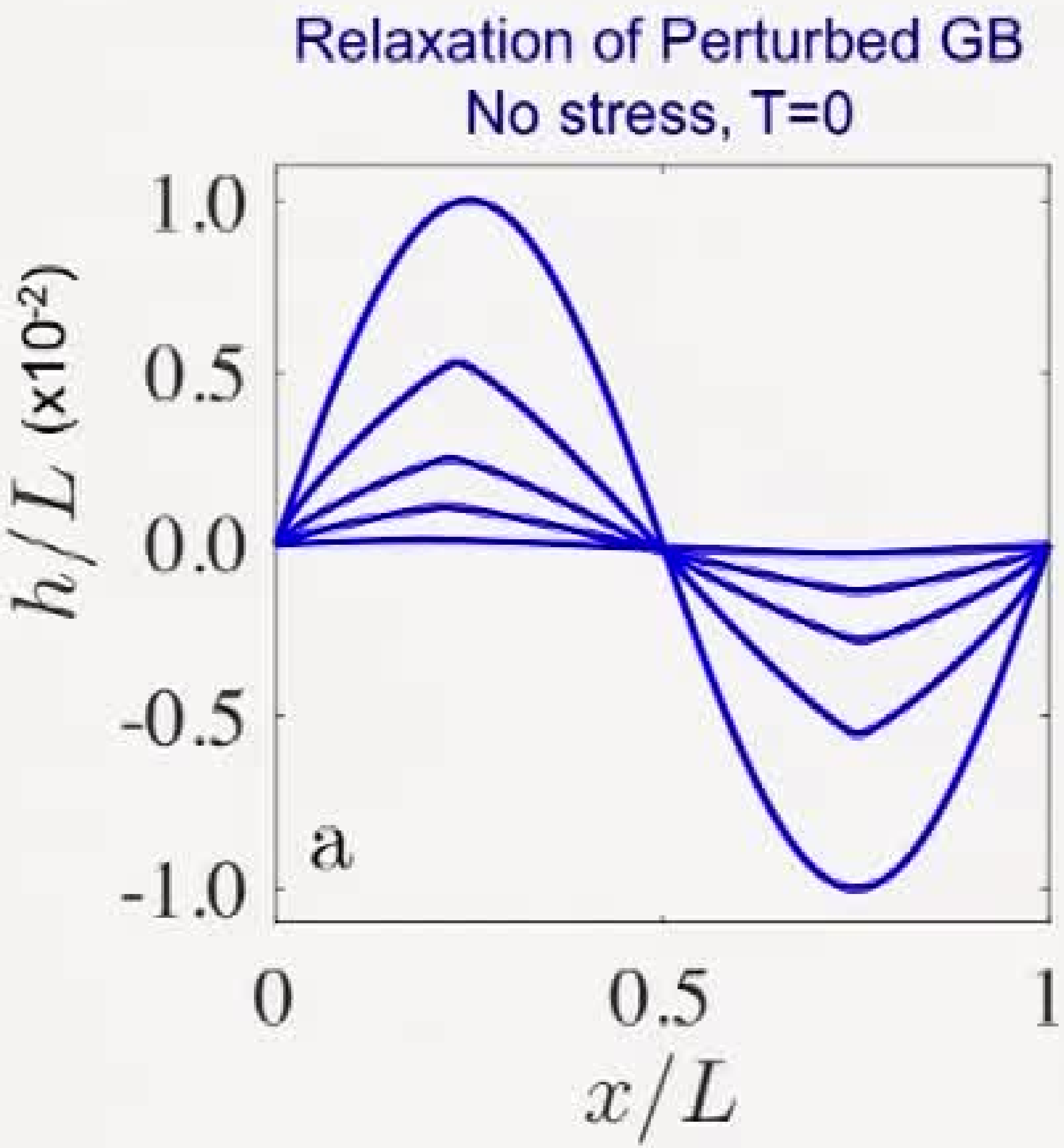
- Two examples (finite difference)



Continuum Equation of Motion

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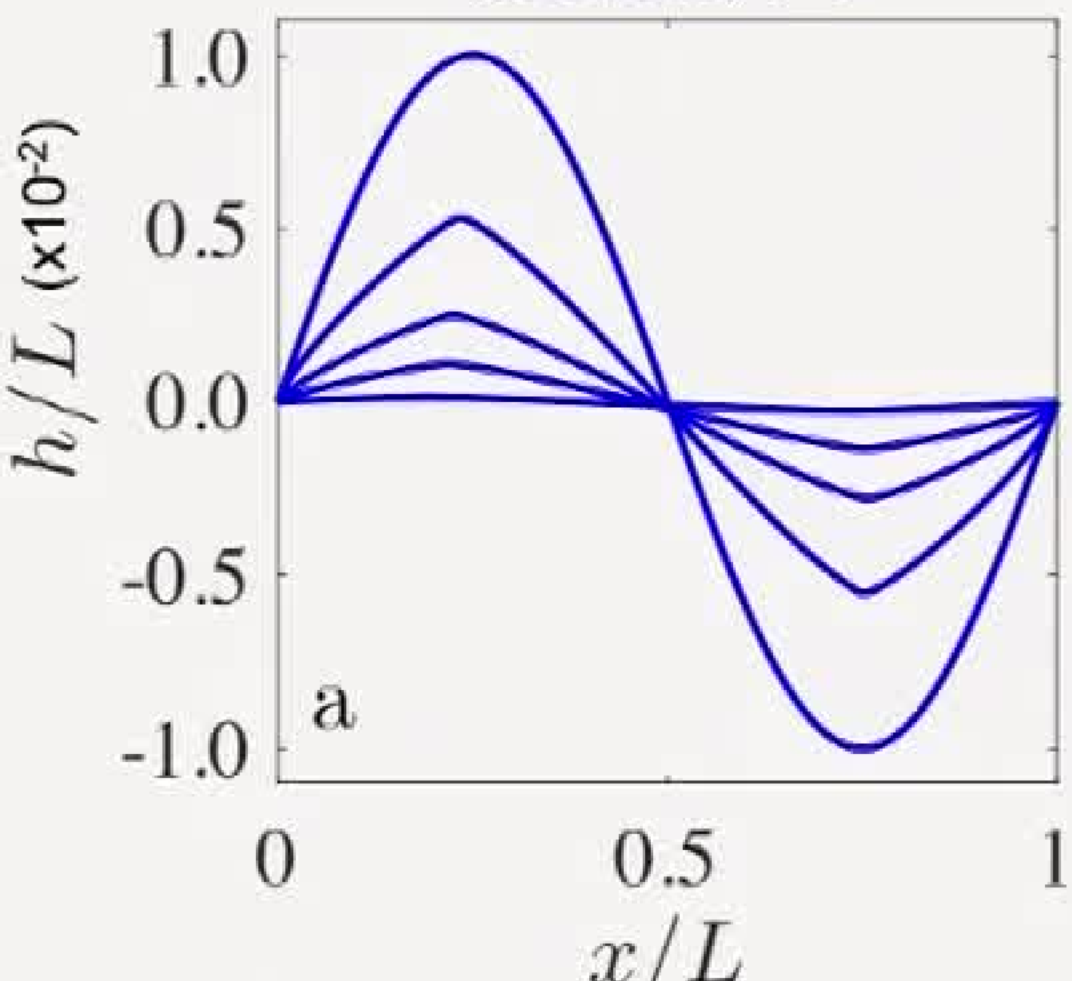
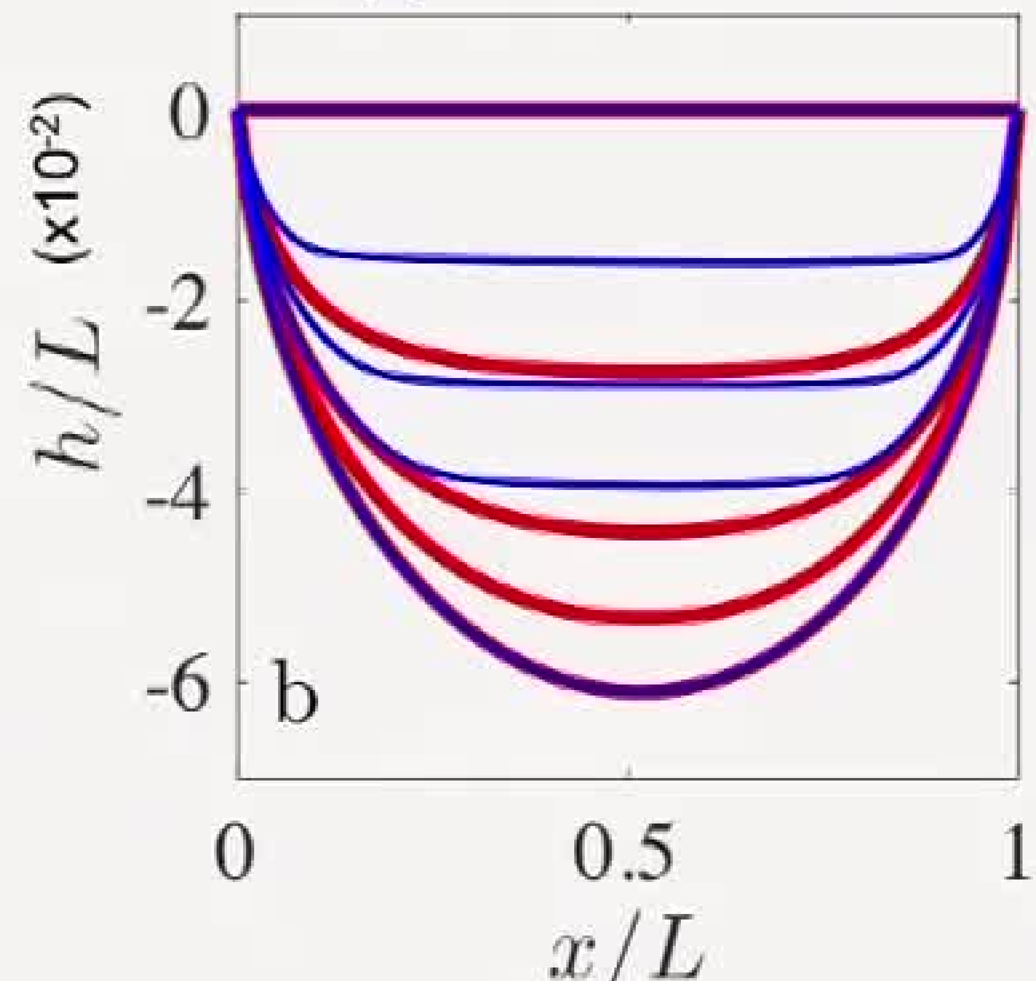
where $B = c_e H = (2H/a)\exp(-F_d/kT)$

- Two examples (finite difference)

Relaxation of Perturbed GB
No stress, T=0

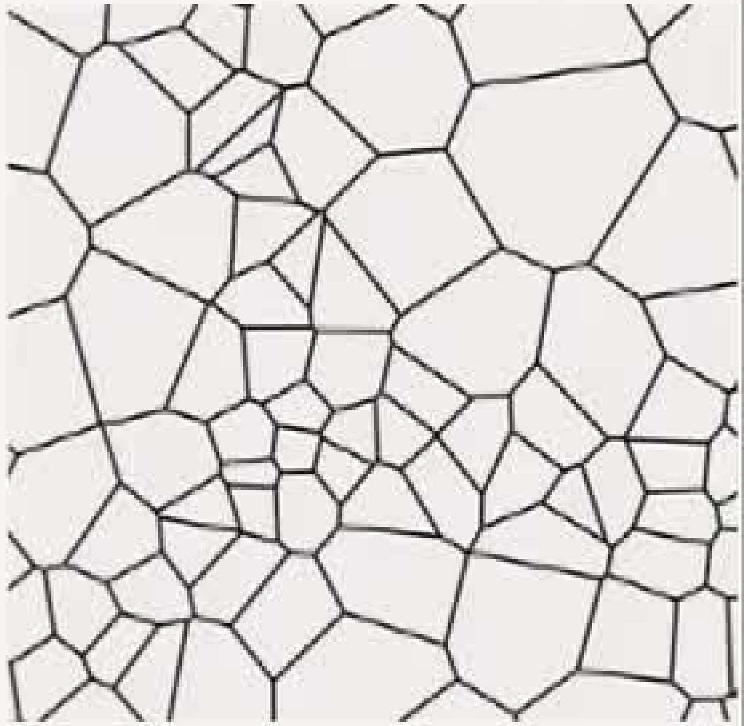


GB evolution
Pinned at ends
Applied stress, 2 T's



Grain Boundary Triple Junctions

- In a polycrystal, GBs are not of infinite extent, but end at vertices where 3 grains meet
- Under an applied stress, disconnections nucleate and propagate to the vertices where they are pinned → disconnections pile-up
- Pile-up generates stress / costs elastic energy



- Continued TJ motion requires nucleation of oppositely signed disconnection to relieve stress

Grain Boundary Triple Junctions

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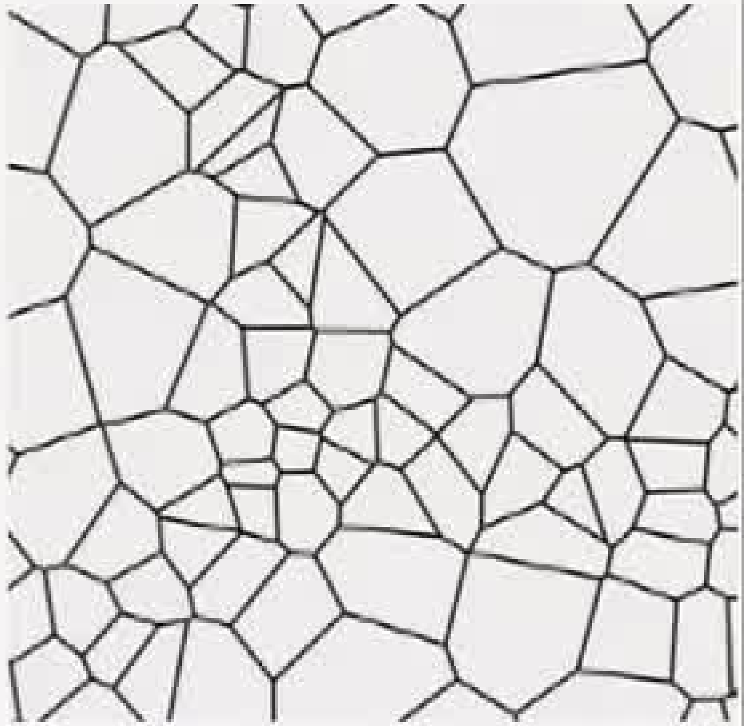
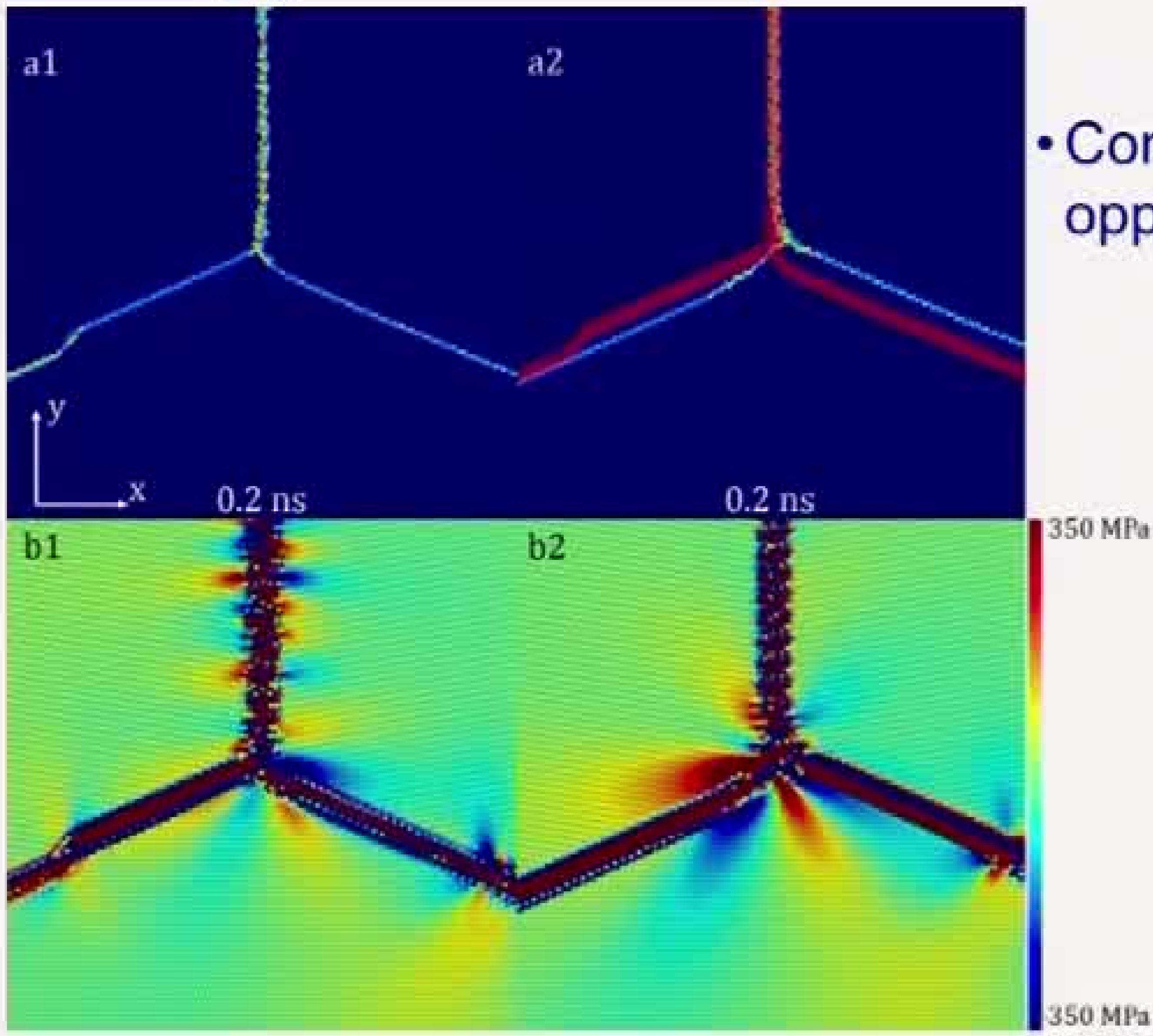
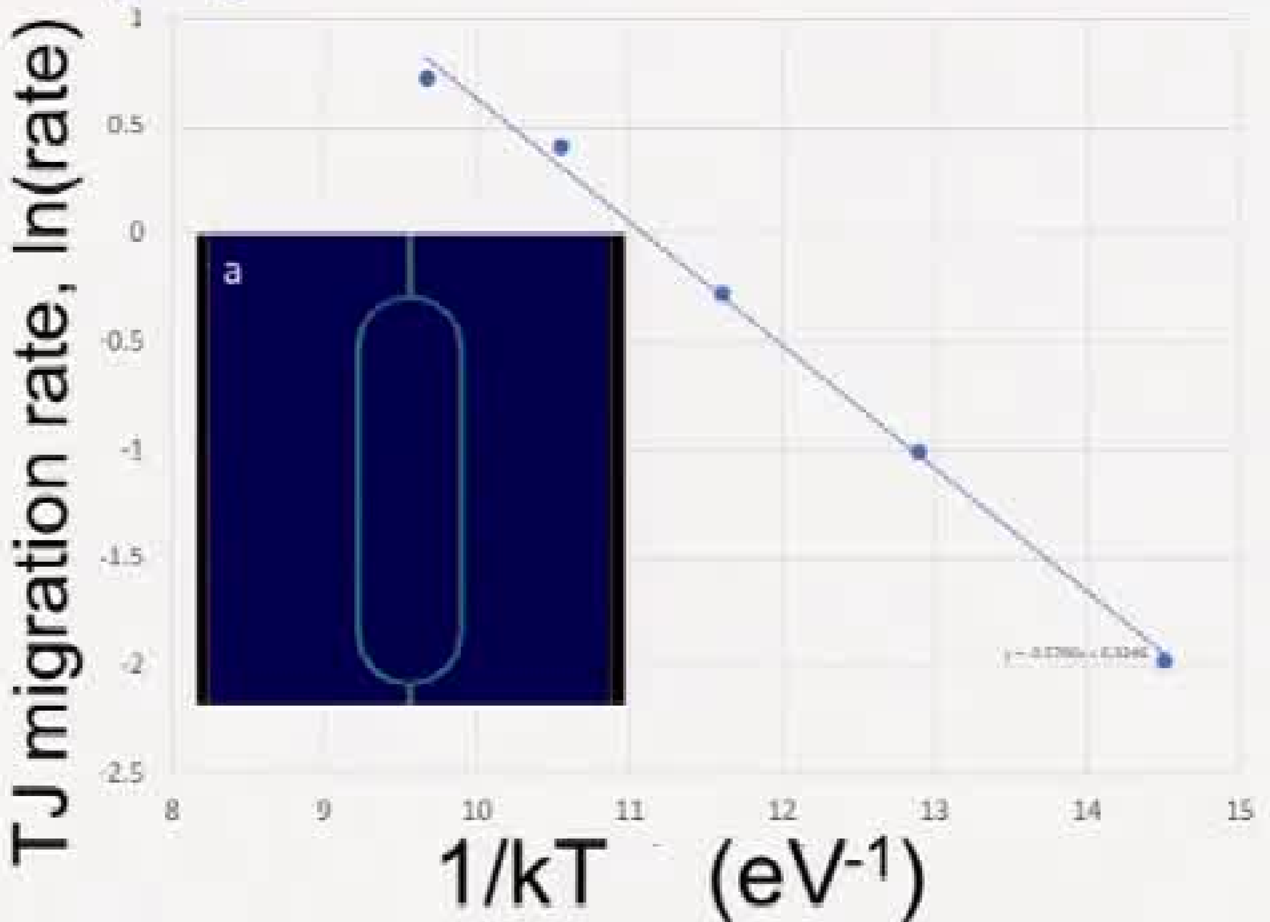
- Under an applied stress, disconnections nucleate and propagate to the vertices where they are pinned → disconnections pile-up
- Pile-up generates stress / costs elastic energy



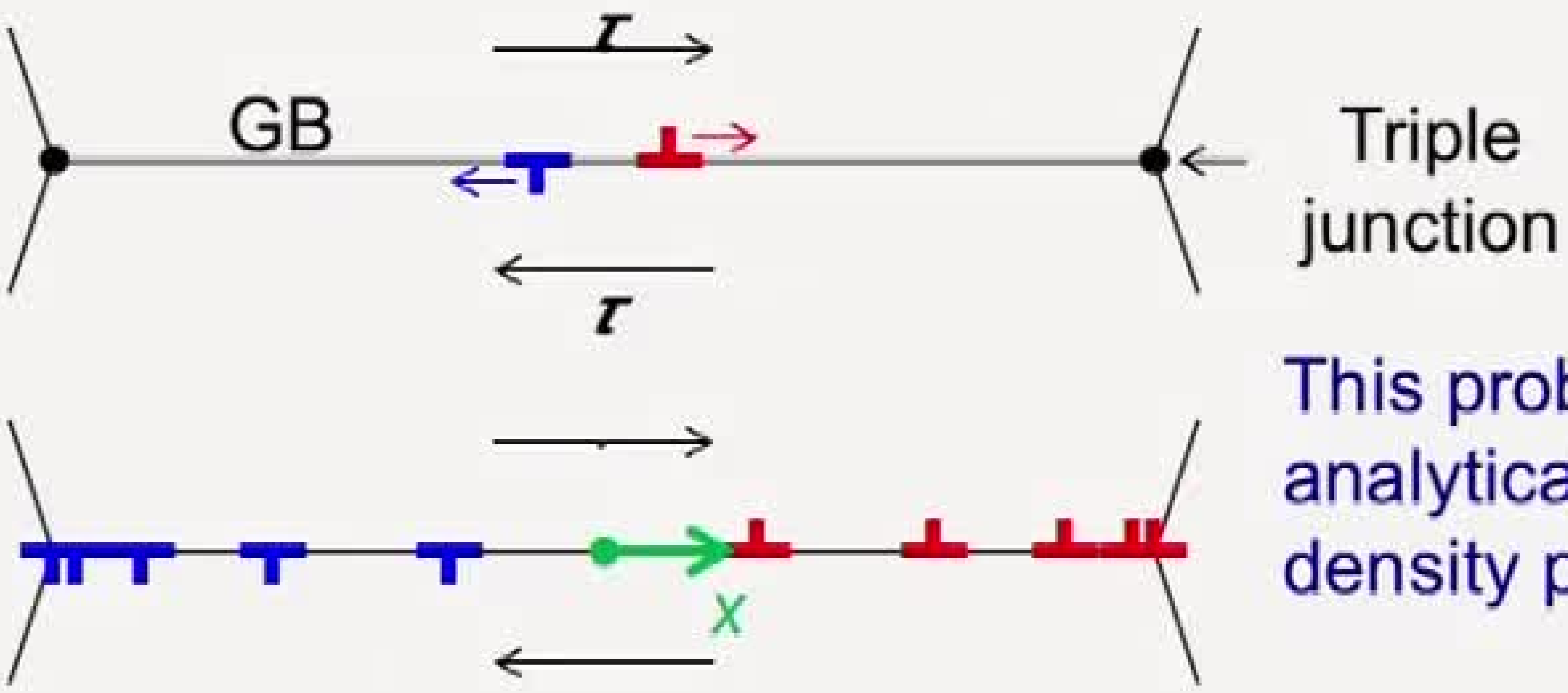


- Continued TJ motion requires nucleation of oppositely signed disconnection to relieve stress

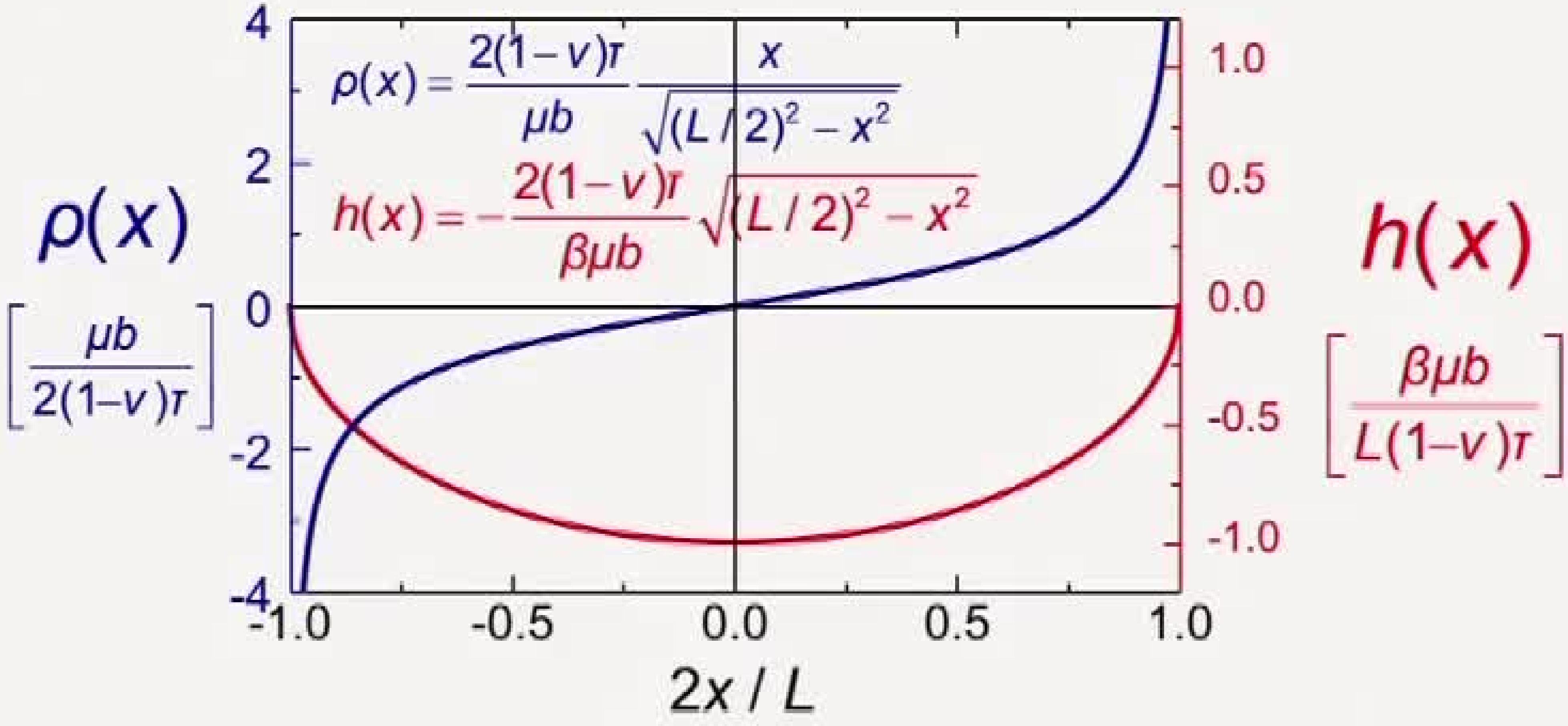


Grain Boundary Triple Junctions

- Single-disconnection mode

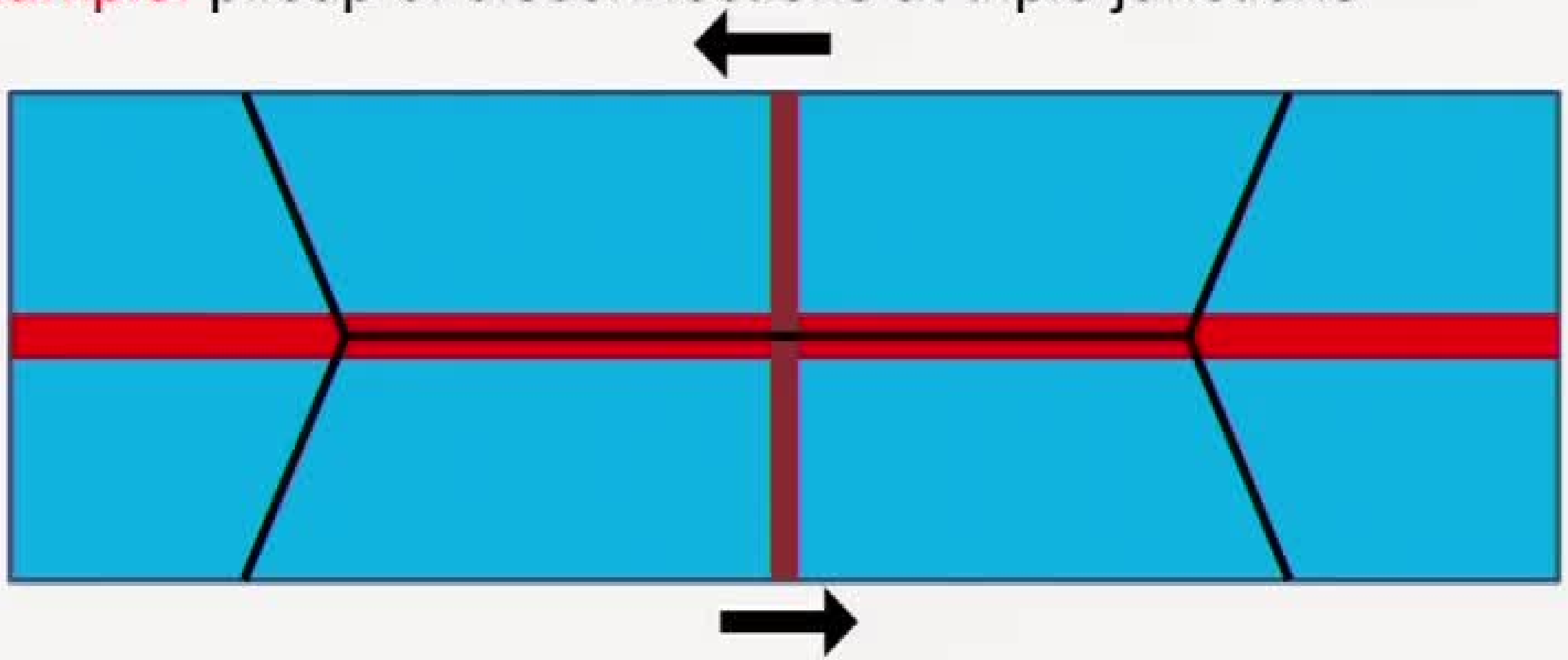


This problem is simple;
analytical solution for disconnection density profile $\rho(x)=h_x(x)/H \rightarrow h(x)$



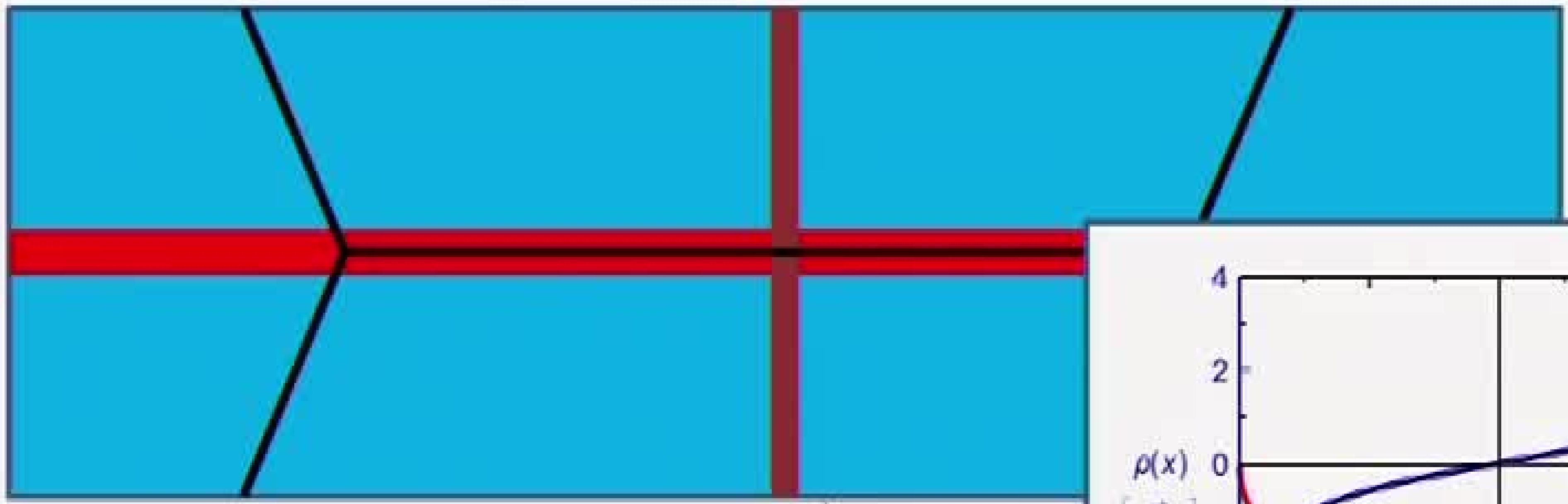
Grain Boundary Triple Junctions

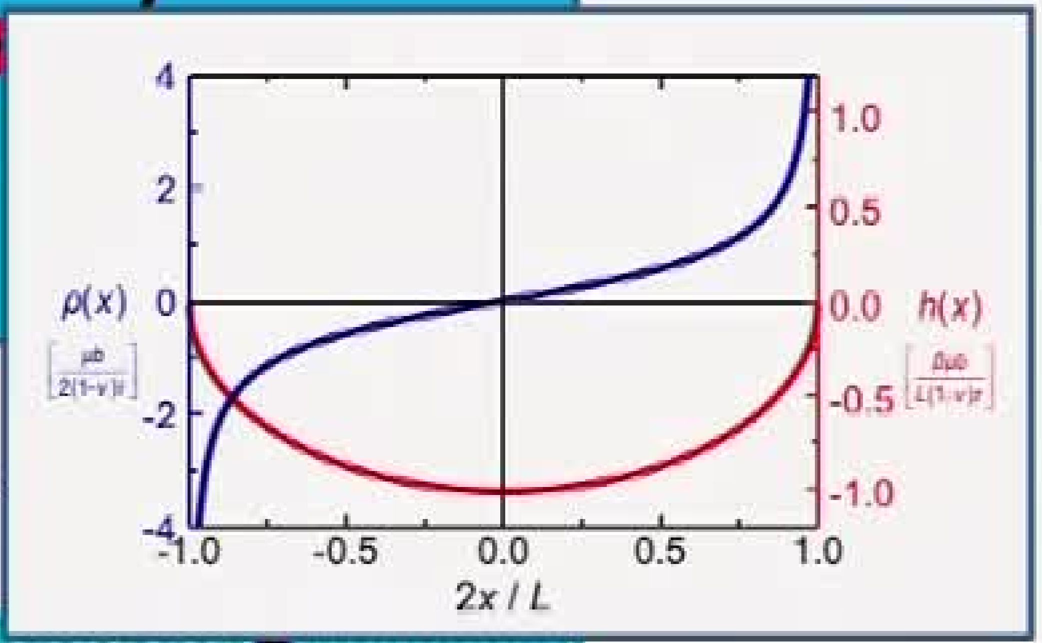
Example: pileup of disconnections at triple junctions



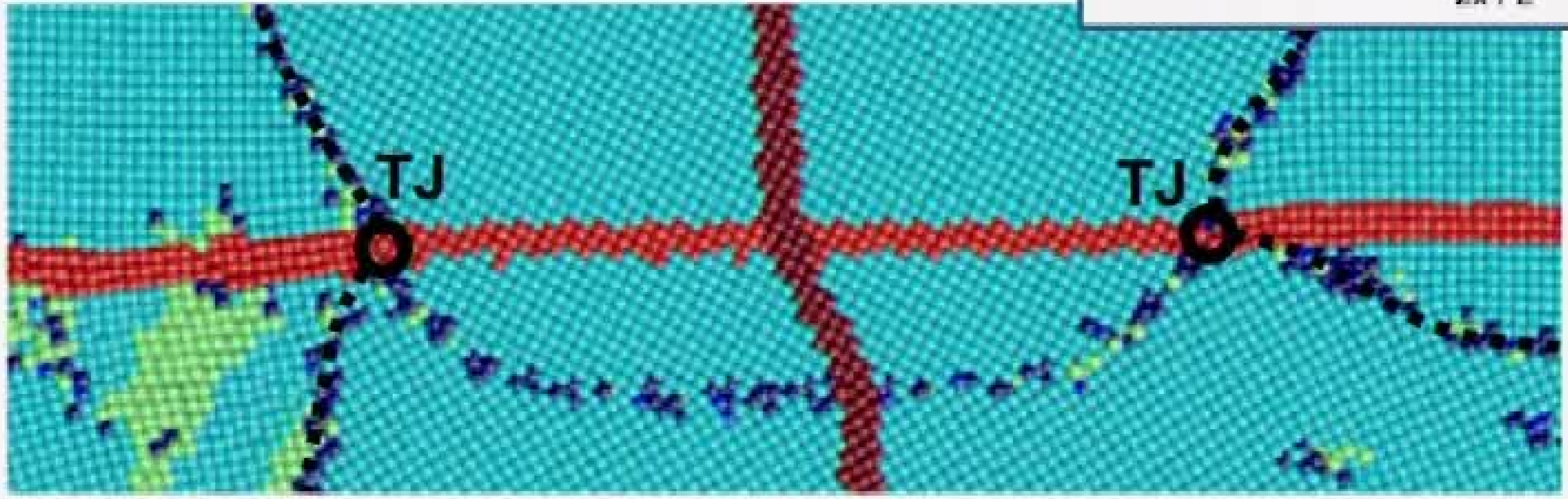
Grain Boundary Triple Junctions

Example: pileup of disconnections at triple junctions



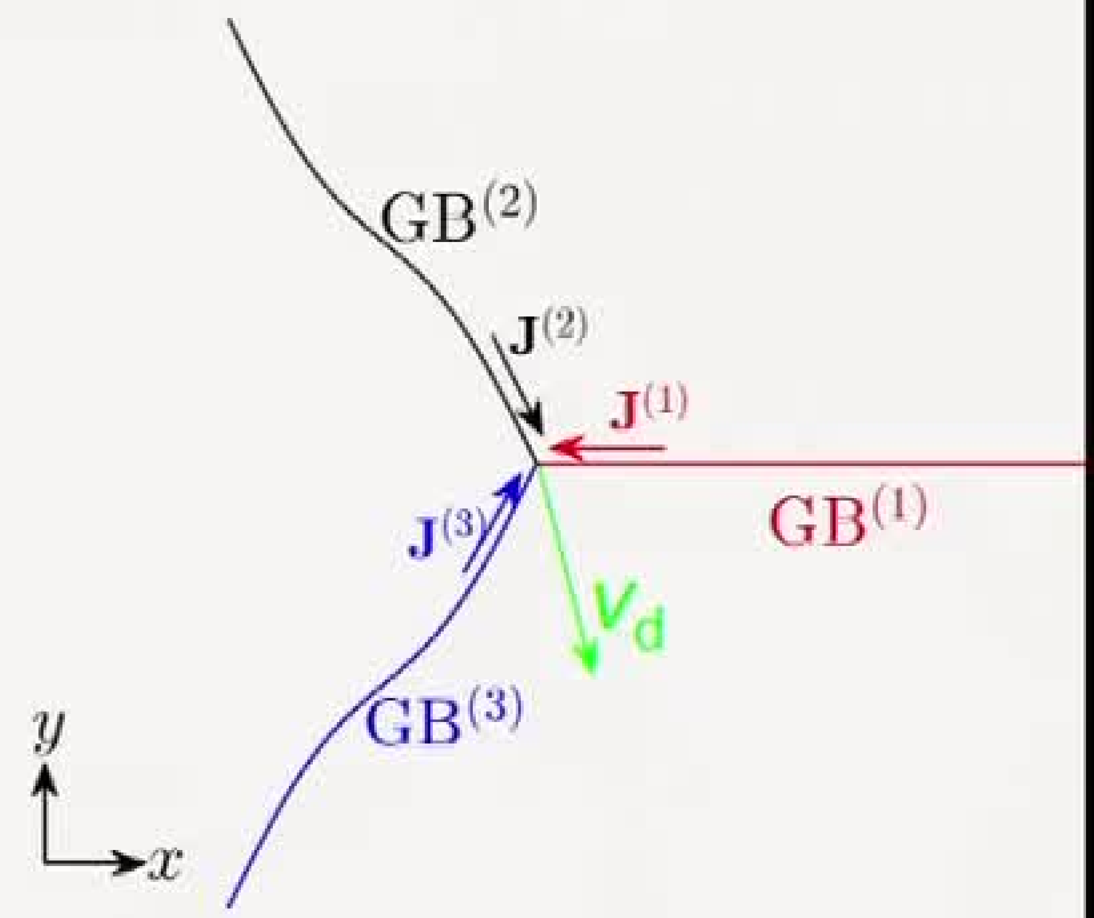


Molecular Dynamics Simulations
Aramfard, Deng (2014)



Grain Boundary Triple Junctions

- If triple junctions (TJs) were really pinned, grain growth would not be possible → triple junctions can move
- Flux of **steps** from disconnections into TJ moves the TJ



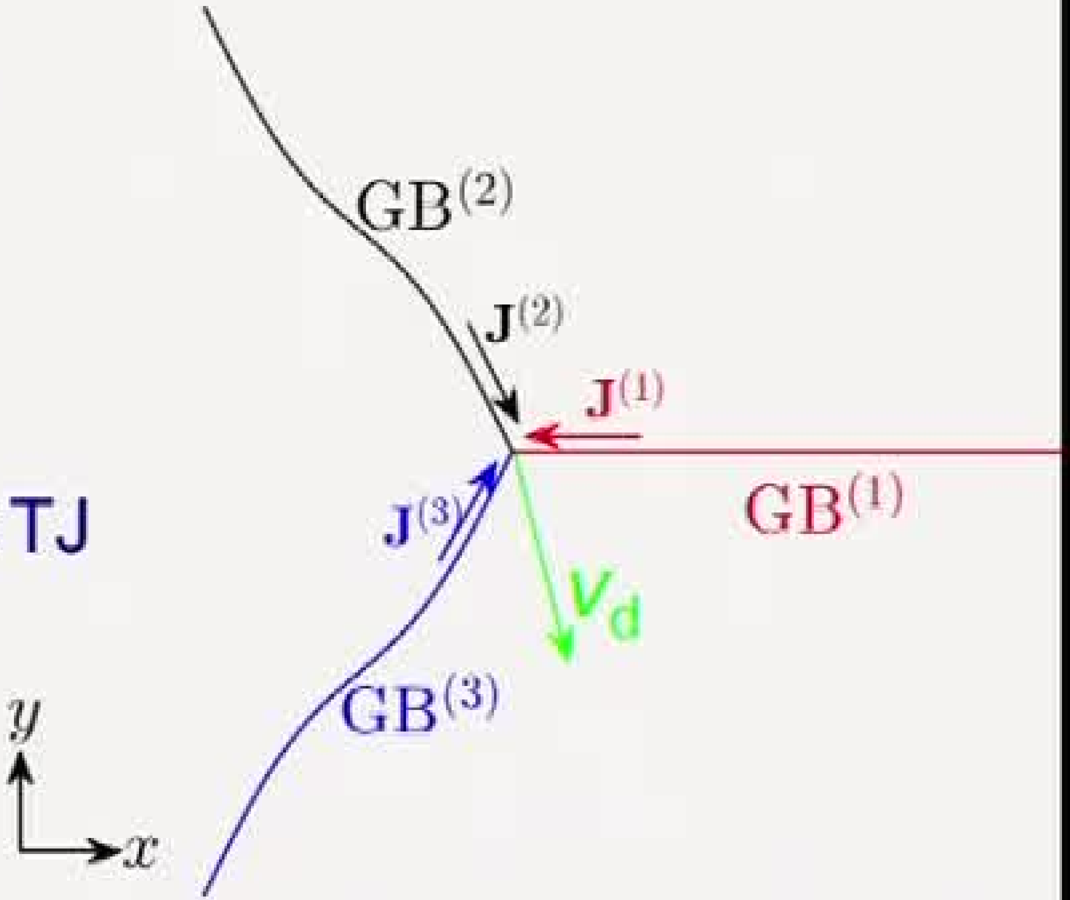
Grain Boundary Triple Junctions

- If triple junctions (TJs) were really pinned, grain growth would not be possible → triple junctions can move
- Flux of **steps** from disconnections into TJ moves the TJ
- But Burgers vectors may or may not be annihilated at TJs → back stress from Burgers vector accumulation pushes disconnections away from TJ

$$\mathbf{v}_{\mathrm{TJ}} = -\sum_{i=1}^{3} H^{(i)} J^{(i)}(\mathbf{x}_0)\mathbf{n}^{(i)}$$

If there is no barrier to disconnection reaction at TJ

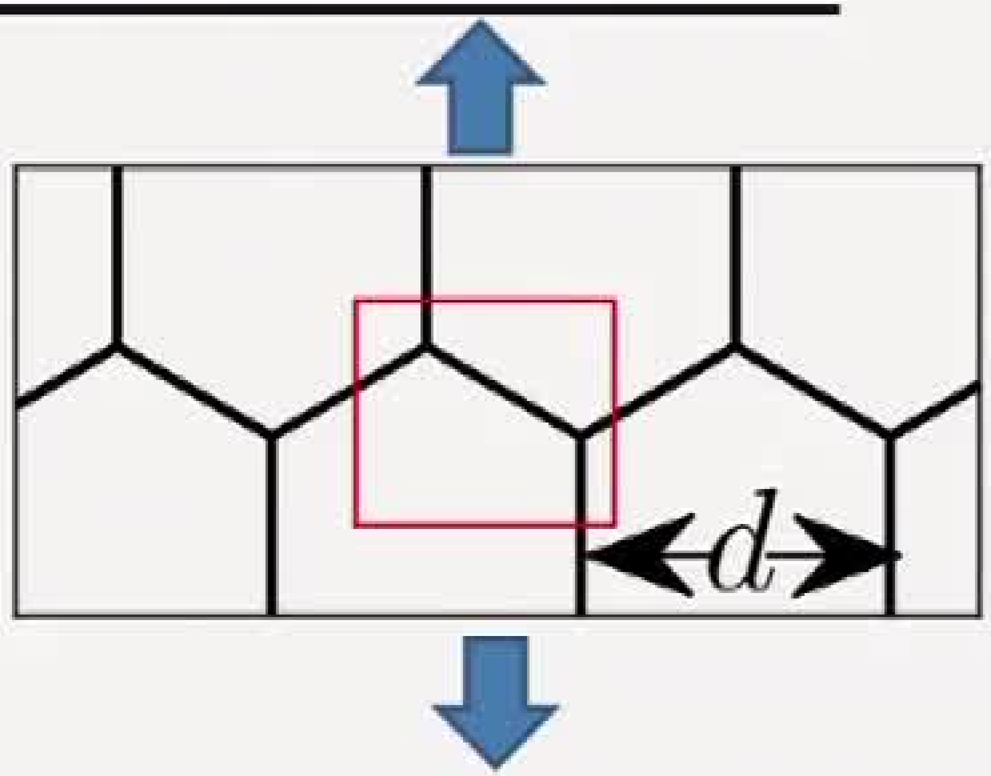
$$J^{(i)}(\mathbf{x}_0) = \left(\rho^{(i)}(\mathbf{x}_0) + \frac{B}{2}\right) v_d^{(i)}(\mathbf{x}_0)$$

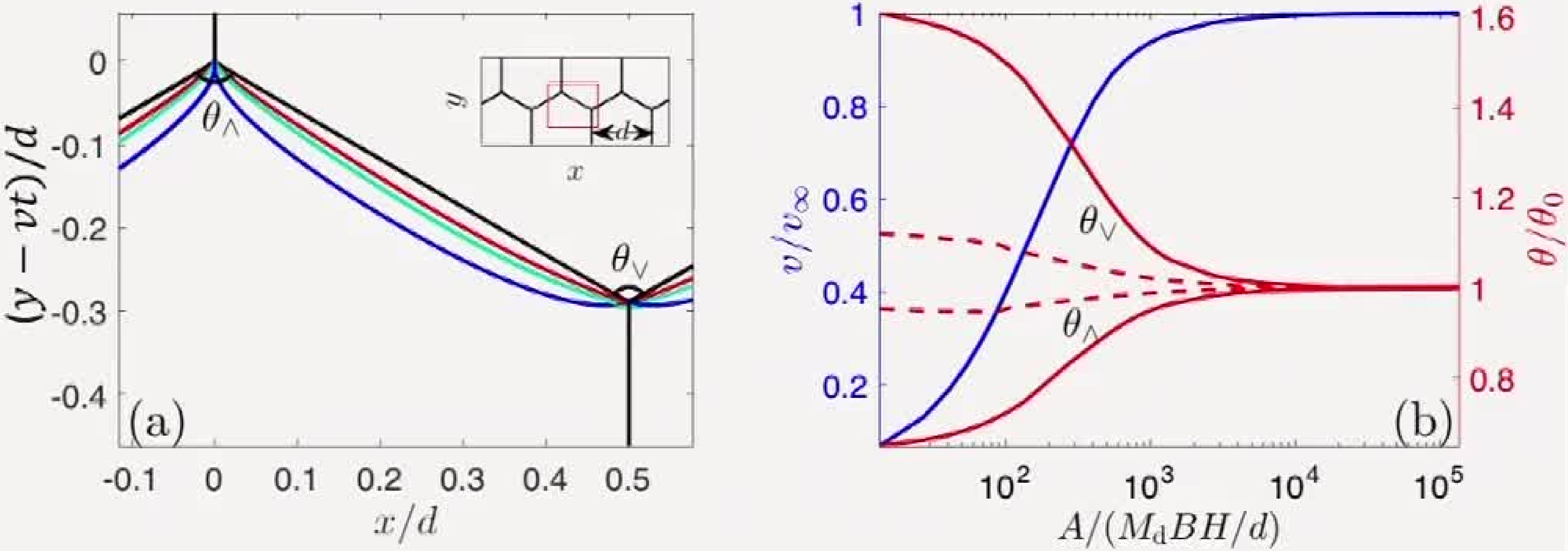


However, if there is a disconnection reaction barrier at the TJ, then $v_d^{(i)}(\mathbf{x}_0)$ should be replaced by the TJ reaction rate/mobility $A^{(i)}$

Grain Boundary Triple Junctions

- Consider a simple example of an idealized polycrystal subject to uniaxial tension
- No shear on vertical GBs; equal and opposite shear on diagonal GBs – mirror symmetry around vertical GBs
- TJs move vertically (steady-state)





Conclusions

- GB migration is controlled by disconnection motion
- Disconnections are characterized by BOTH h and $\mathbf{b}$
- Multiple {**b**,h} pairs for every GB; set by bicrystallography

Conclusions

- GB migration is controlled by disconnection motion
- Disconnections are characterized by BOTH *h* and **b**
- Multiple {**b**,h} pairs for every GB; set by bicrystallography
- Choosing between modes depends on nature of the (local) driving force(s) and temperature
- GB mobility is NOT a material/GB property; disconnection properties are

Opportunities and Challenges

- Extension of the equation of motion to include multiple disconnection types $\{\mathbf{b}, h\}$
- GBs that are not constrained to $h_x \ll 1$; multiple reference states
- More general description of TJs
- Numerical model for microstructure evolution (like our curvature flow code)
- Grain boundary mobility
- Grain rotation