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Learning the Short- and Medium-Range Enthalpic Structural Manifolds of Disordered Materials

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November 29, 2022 ● MRS Fall Meeting DS02.05 (Hynes 210, 3:30 PM)

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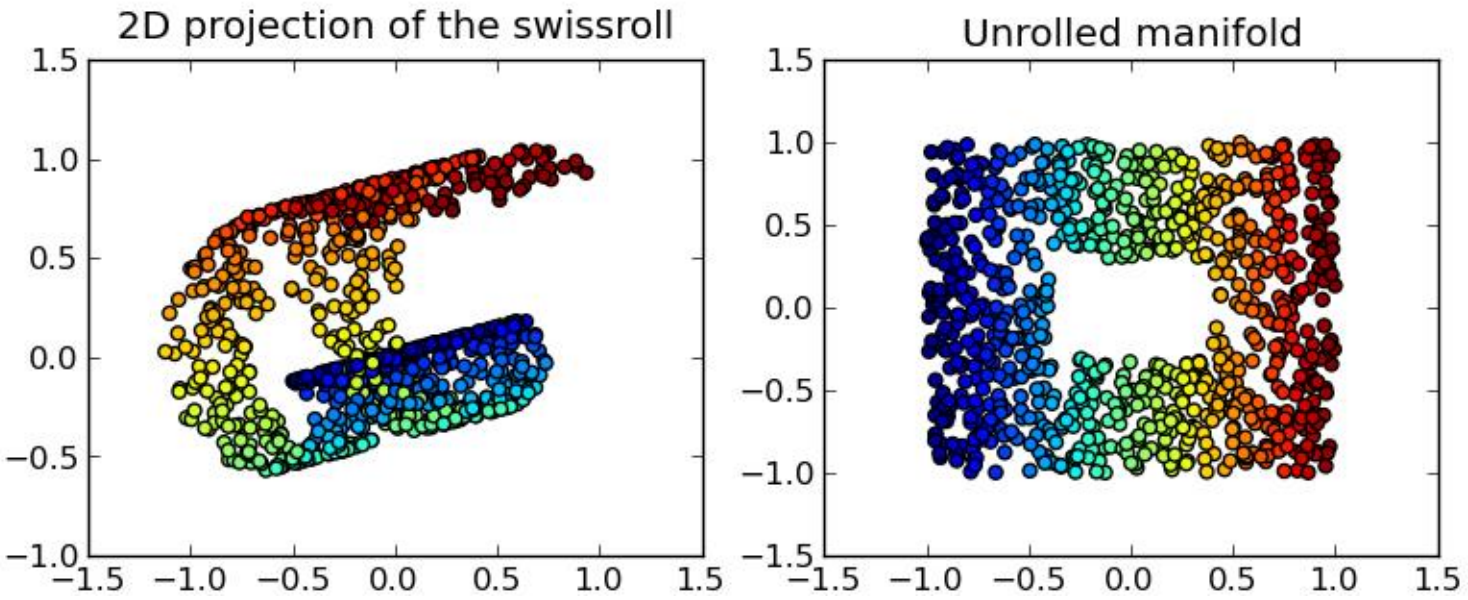
Concept: Manifold Learning / Dimensionality Reduction

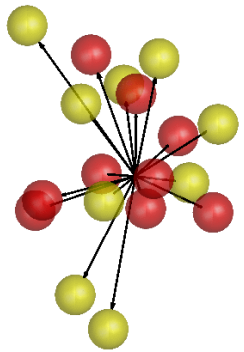


Given a set of points $\mathcal{X} \in \mathbb{R}^n$,
learn a function $f: \mathbb{R}^n \longrightarrow \mathbb{R}^q$ where $q < n$

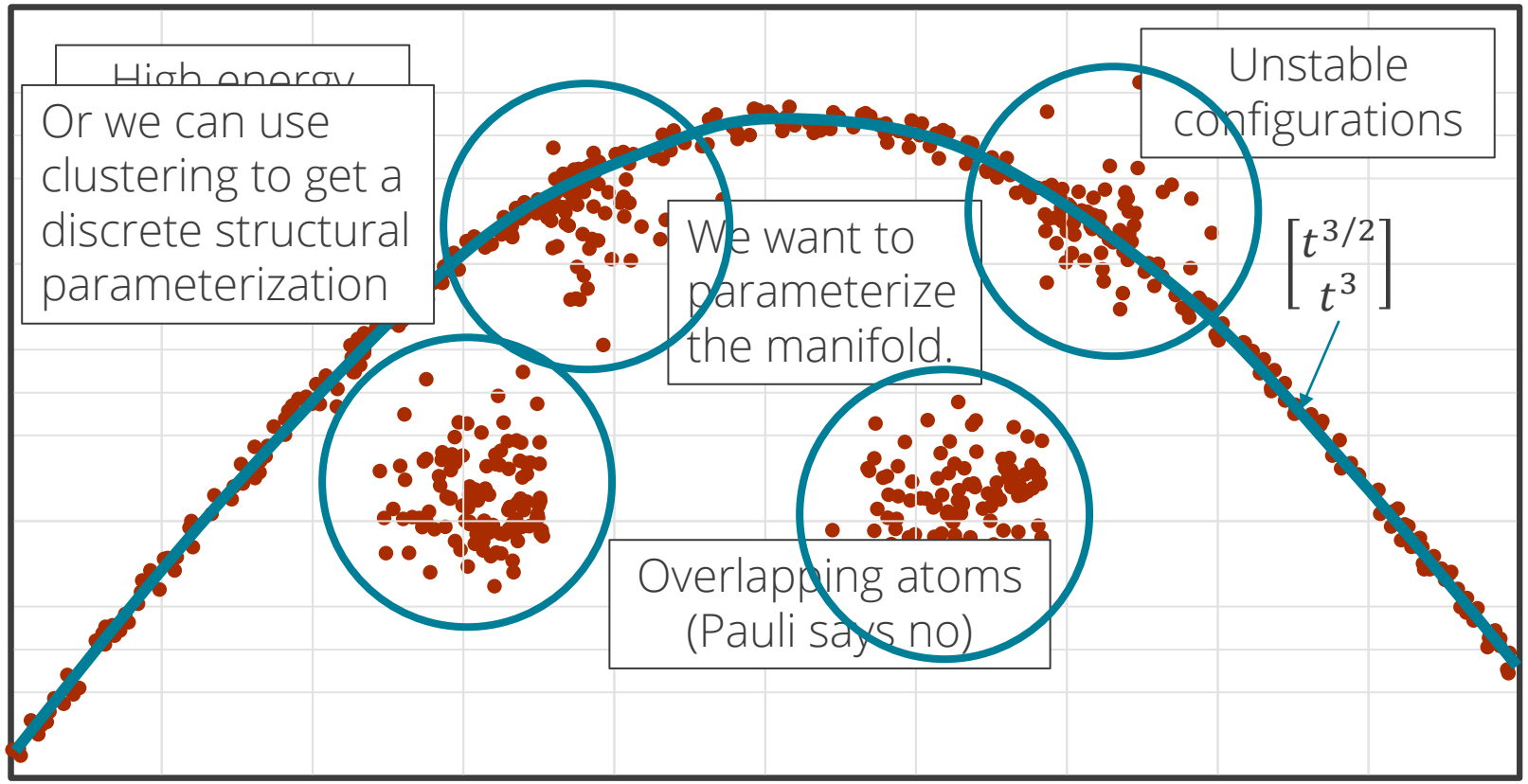
- where neighboring points in $\mathcal{X}$ are neighbors in $f(\mathcal{X})$
- where $f(\mathcal{X})$ captures the important information in $\mathcal{X}$

Concept: Manifold Learning / Dimensionality Reduction

We need 3n real numbers to naively quantify an atomic configuration, plus n integers to encode species information.



So imagine we're plotting configs cut out of some material on axes spanning $\mathbb{R}^{3n} \times \mathbb{N}$.



We only observe atomic configurations in certain regions of $\mathbb{R}^{3n} \times \mathbb{N}$.
Enthalpy pulls atomic configurations onto a manifold.
Entropy and kinetics spread atomic configurations out on that manifold.

Dimensionality reduction strategy

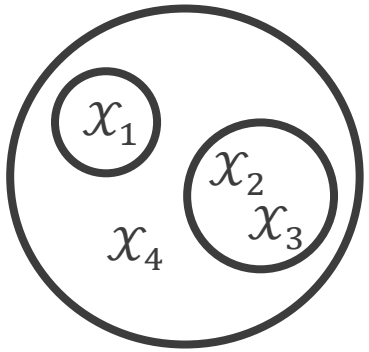
Cut out environments

$$\mathcal{X} = \begin{bmatrix} \boldsymbol{X}_1 & s_1 \\ \vdots & \vdots \\ \boldsymbol{X}_n & s_2 \end{bmatrix}$$

This is the real technical challenge →

Compute **generalized distances** between pairs of environments

$$\boldsymbol{D} = \begin{bmatrix} d(\mathcal{X}_1, \mathcal{X}_1) & \cdots & d(\mathcal{X}_N, \mathcal{X}_1) \\ \vdots & \ddots & \vdots \\ d(\mathcal{X}_1, \mathcal{X}_N) & \cdots & d(\mathcal{X}_N, \mathcal{X}_N) \end{bmatrix}$$



Hierarchical Clustering *discrete parameterization of manifold*

Diffusion Maps *continuous parameterization of manifold*

$$f\left(\begin{bmatrix} \boldsymbol{X}_1 & s_1 \\ \vdots & \vdots \\ \boldsymbol{X}_n & s_2 \end{bmatrix}\right) = \begin{bmatrix} y_1 \\ \vdots \\ y_q \end{bmatrix} \qquad (q \ll 4n)$$

Properties of a Generalized Distance Function

Continuity and **Smoothness**: the generalized distance is stable with respect to small atomic perturbations.

- An atom jumping across the cutoff radius won't dramatically change the result

Completeness: the generalized distance between two configurations is zero iff the two configurations are equivalent.

Rotation Invariance: the generalized distance is the same regardless of frame.

Permutation Invariance: the distance is the same regardless of order of atoms.

Tolerates Variable Numbers of Atoms: calculates meaningful distances between environments with different numbers of atoms.

Differentiable: the distance can be differentiated with respect to atomic positions. Important for empirical potentials.

Speed: the distance can be quickly calculated between a pair of atomic configurations.

Gaussian Integral Inner Product (GIIP) Distance

Integral inner product of two functions

(1) $$\langle a, b \rangle = \int_{\mathbb{R}^3} a(\mathbf{x}') \cdot b(\mathbf{x}') \, d\mathbf{x}'$$

Norm of a function induced by the inner product above

(2) $$||a|| = \sqrt{\langle a, a \rangle}$$

Gaussian function with standard deviation sigma, normalized to 1

(3) $$G_\sigma(\mathbf{x}) = \exp\left[-|\mathbf{x}|^2/(2\sigma^2)\right] / (\pi^{3/4}\sigma^{3/2})$$

Atomic density function consisting of weighted Gaussians centered on atomic positions

(4) $$\rho_{\mathcal{X}}(\mathbf{x}) = \sum_{\mathbf{x}' \in \mathcal{X}} w_{\mathbf{x}'} G_{\sigma_{\mathbf{x}'}}(\mathbf{x} - \mathbf{x}')$$

Gaussian Integral Inner Product (GIIP) between two configurations

(5) $$\left\langle \mathcal{X}^\alpha, \mathcal{X}^\beta \right\rangle = \langle \rho_{\mathcal{X}^\alpha}, \rho_{\mathcal{X}^\beta} \rangle$$

Distance between two configurations can be calculated with three inner products

(6) $$\left|\mathcal{X}^\alpha - \mathcal{X}^\beta\right|^2 = \left\langle \mathcal{X}^\alpha - \mathcal{X}^\beta, \mathcal{X}^\alpha - \mathcal{X}^\beta \right\rangle = \left\langle \mathcal{X}^\alpha, \mathcal{X}^\alpha \right\rangle + \left\langle \mathcal{X}^\beta, \mathcal{X}^\beta \right\rangle - 2 \cdot \left\langle \mathcal{X}^\alpha, \mathcal{X}^\beta \right\rangle$$

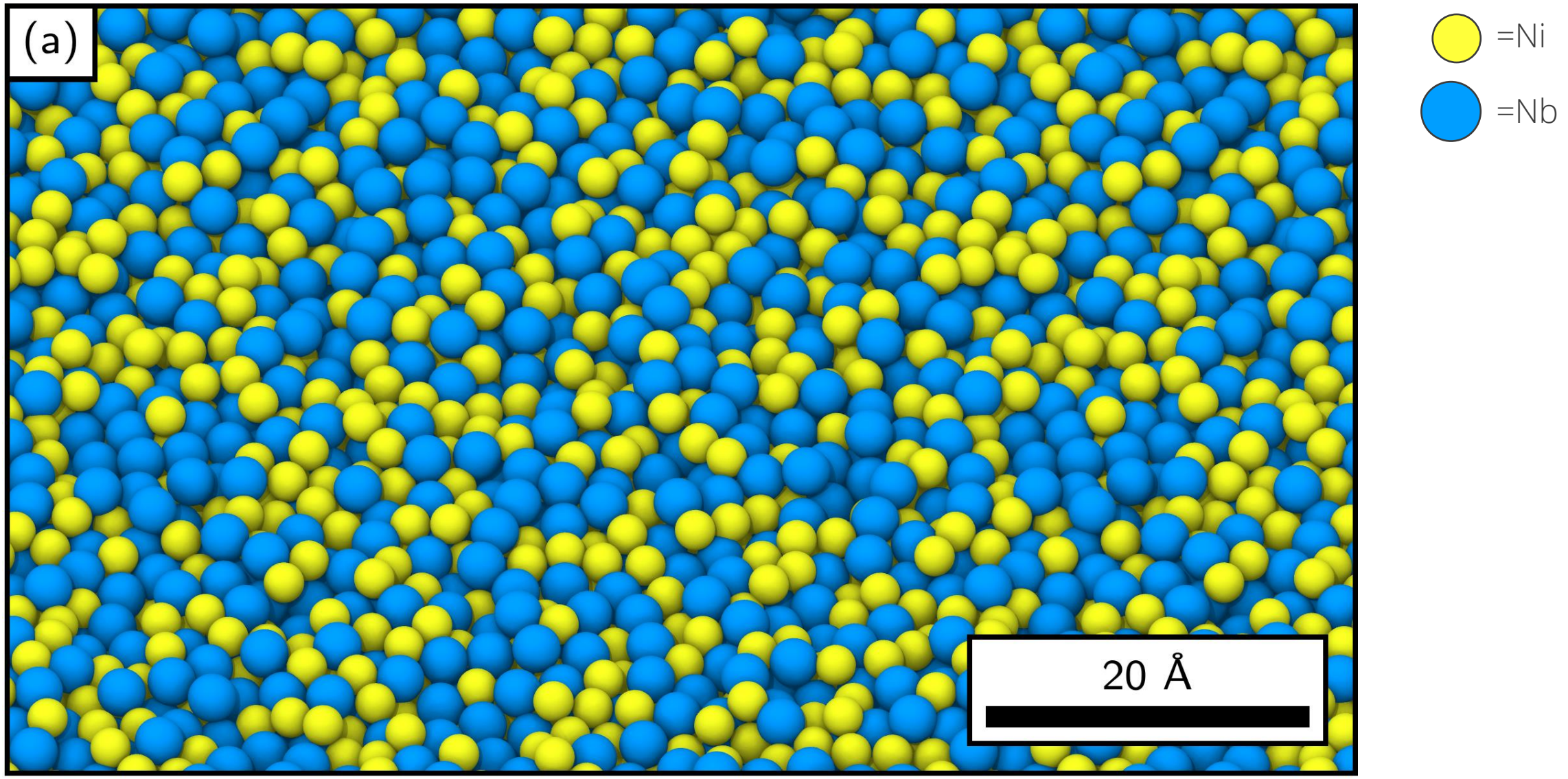
Orientation invariance by minimizing over all possible rotations/rotoinversions

(7) $$\min_{\mathbf{R} \in \mathrm{O}(3)} \left|\mathcal{X}^\alpha - \mathbf{R}\mathcal{X}^\beta\right|^2$$

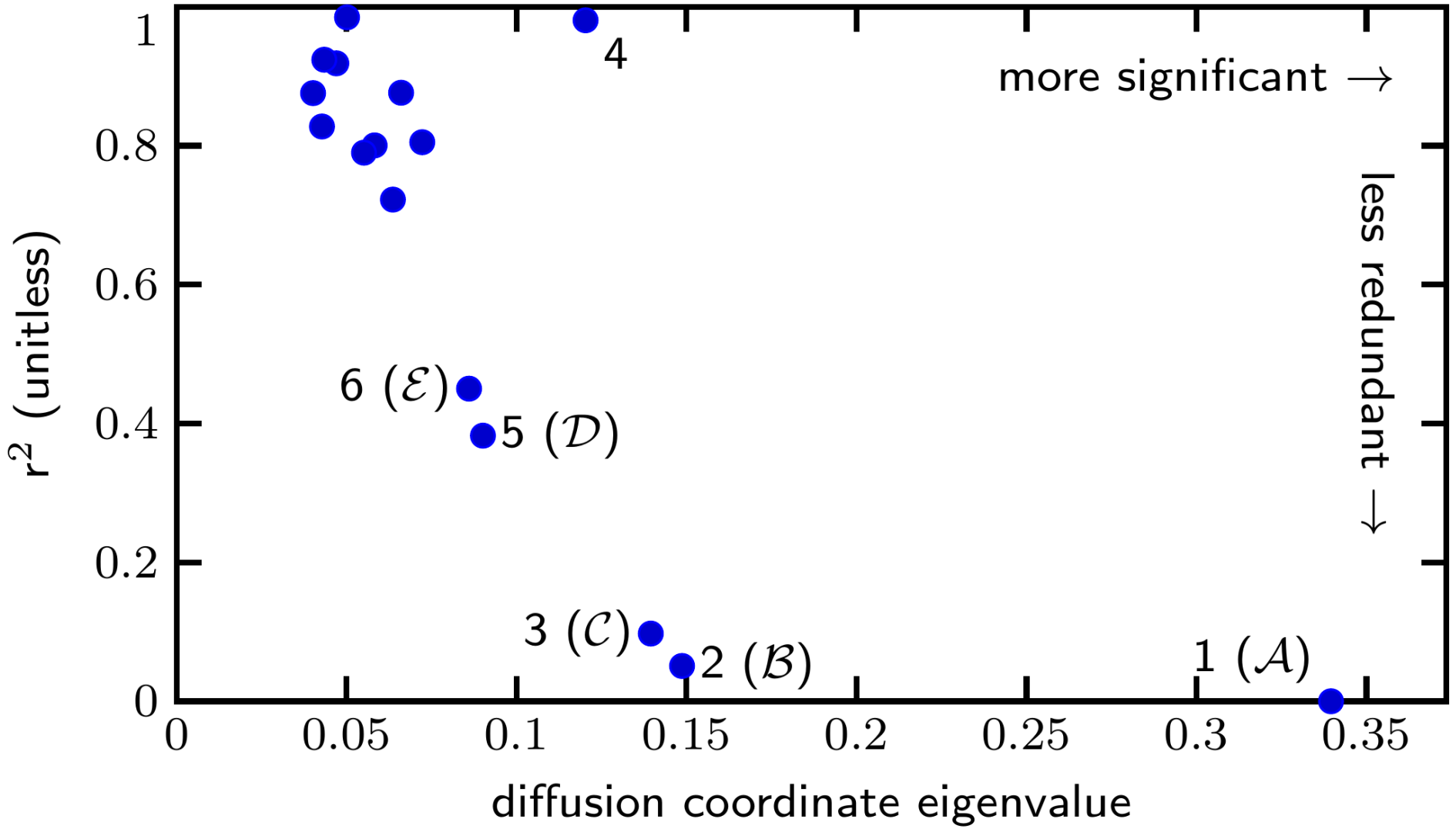
GIIP is analytically tractable in a computationally convenient form

(8) $$\left\langle \mathcal{X}^\alpha, \mathcal{X}^\beta \right\rangle = 2\sqrt{2} \sum_{\mathbf{x}^\alpha \in \mathcal{X}^\alpha} \sum_{\mathbf{x}^\beta \in \mathcal{X}^\beta} w_{\mathbf{x}^\alpha} w_{\mathbf{x}^\beta} \left(\frac{\sigma_{\mathbf{x}^\alpha} \sigma_{\mathbf{x}^\beta}}{\sigma_{\mathbf{x}^\alpha}^2 + \sigma_{\mathbf{x}^\beta}^2} \right)^{3/2} \exp\left[-|\mathbf{x}^\alpha - \mathbf{x}^\beta|^2 / (2\sigma_{\mathbf{x}^\alpha}^2 + 2\sigma_{\mathbf{x}^\beta}^2)\right]$$

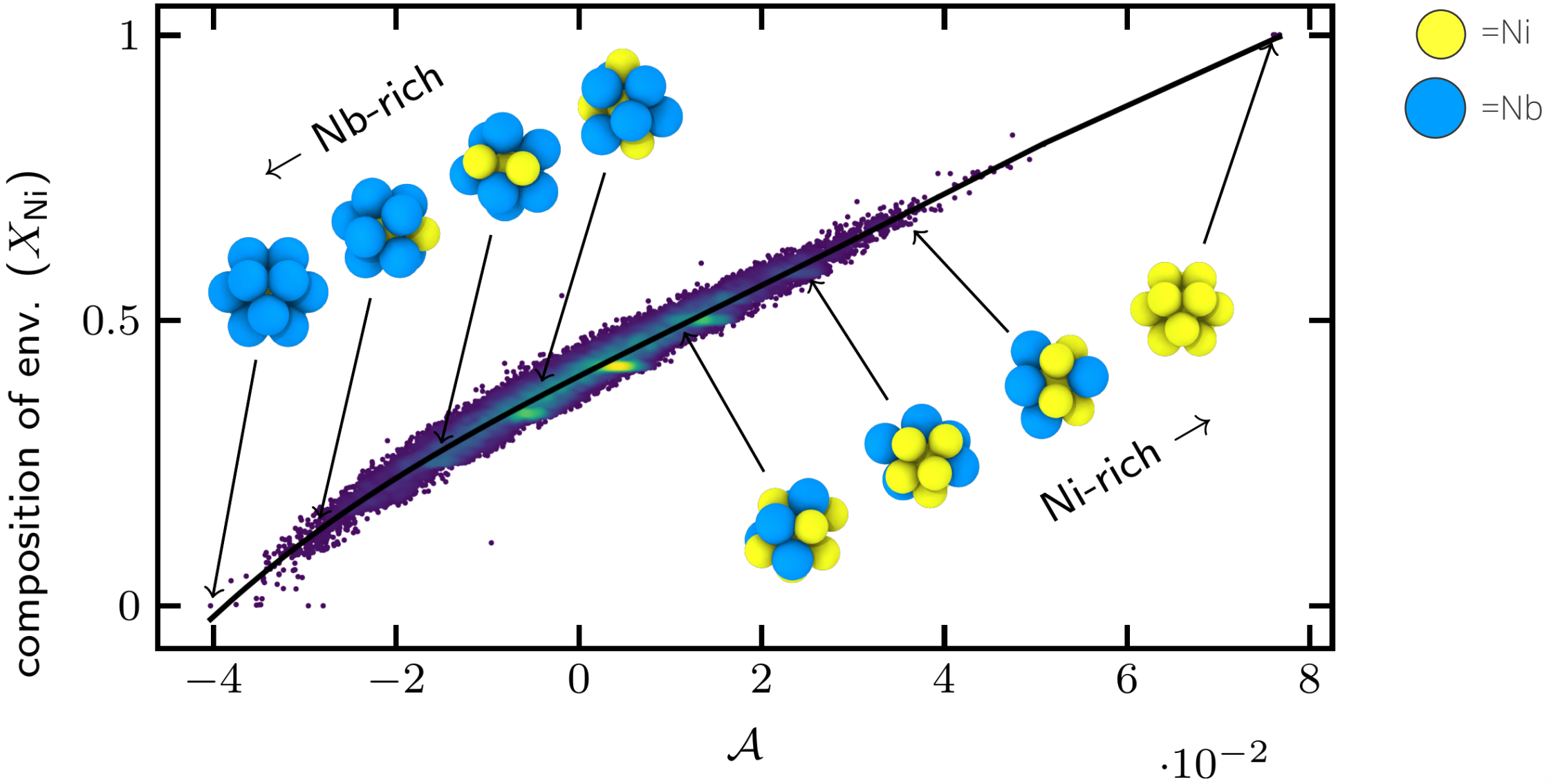
Example: Three-dimensional EAM NiNb metallic glass



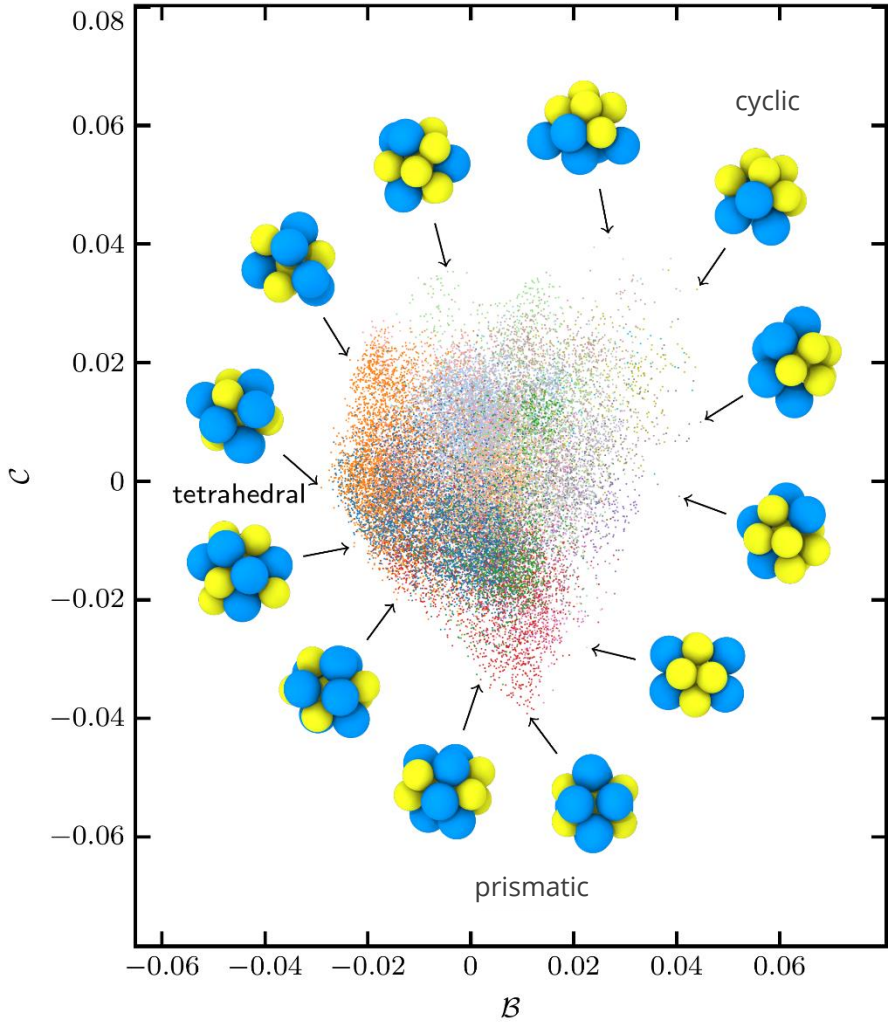
Selecting a parsimonious set of diffusion coords for metallic glass



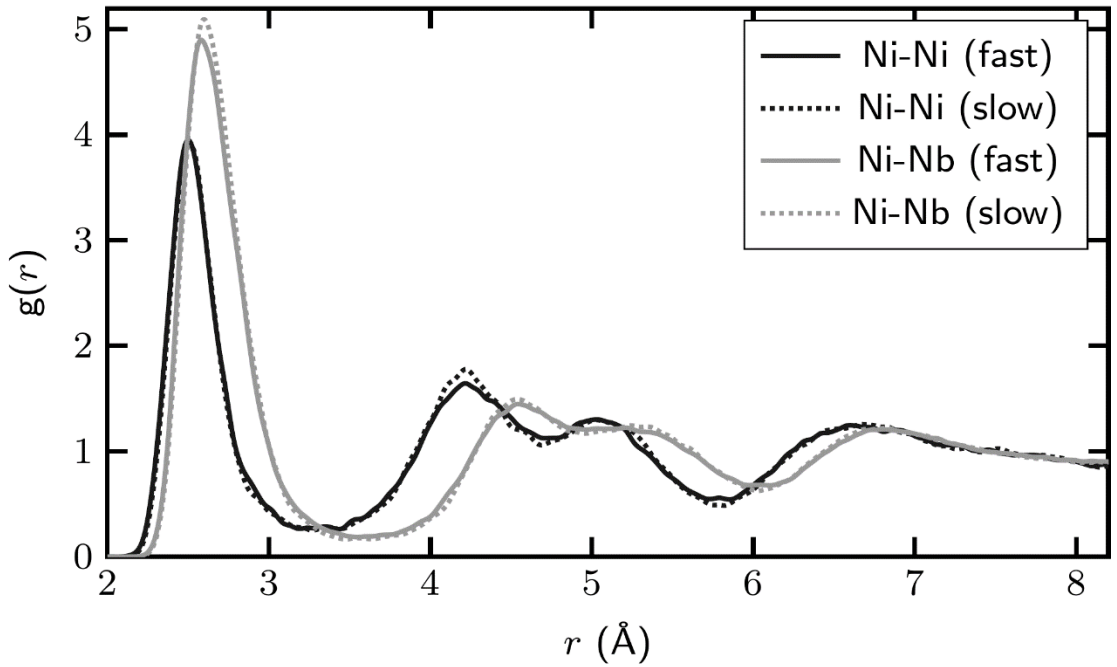
Diffusion coordinate *A* is composition of environment

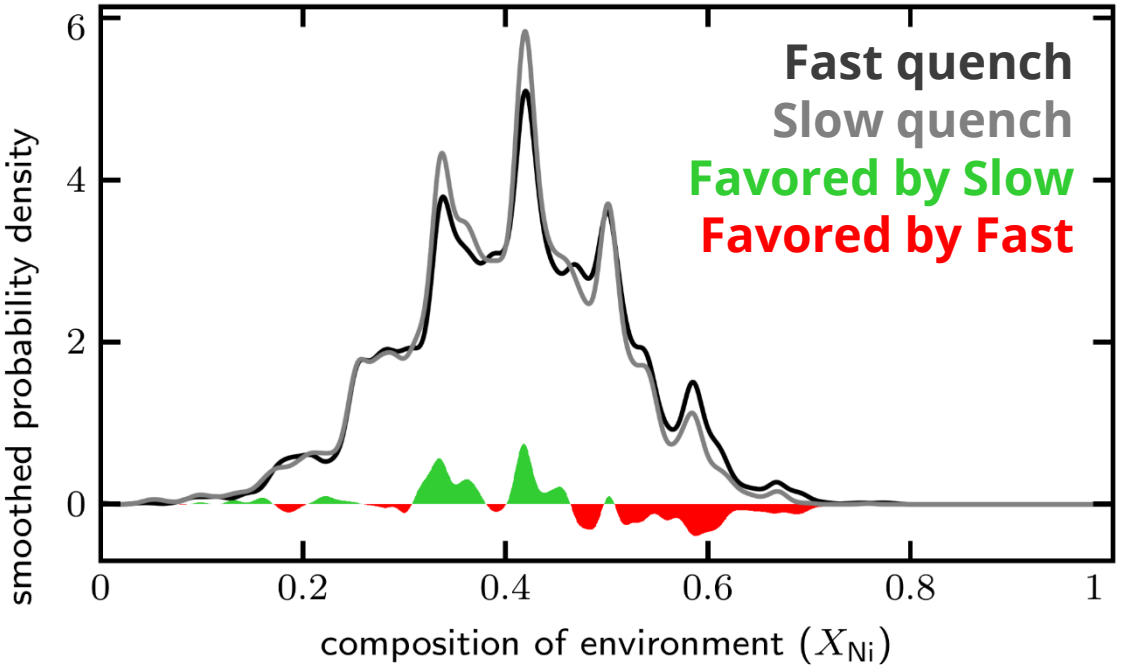


Diffusion coordinates *B* and *C* parameterize local symmetry



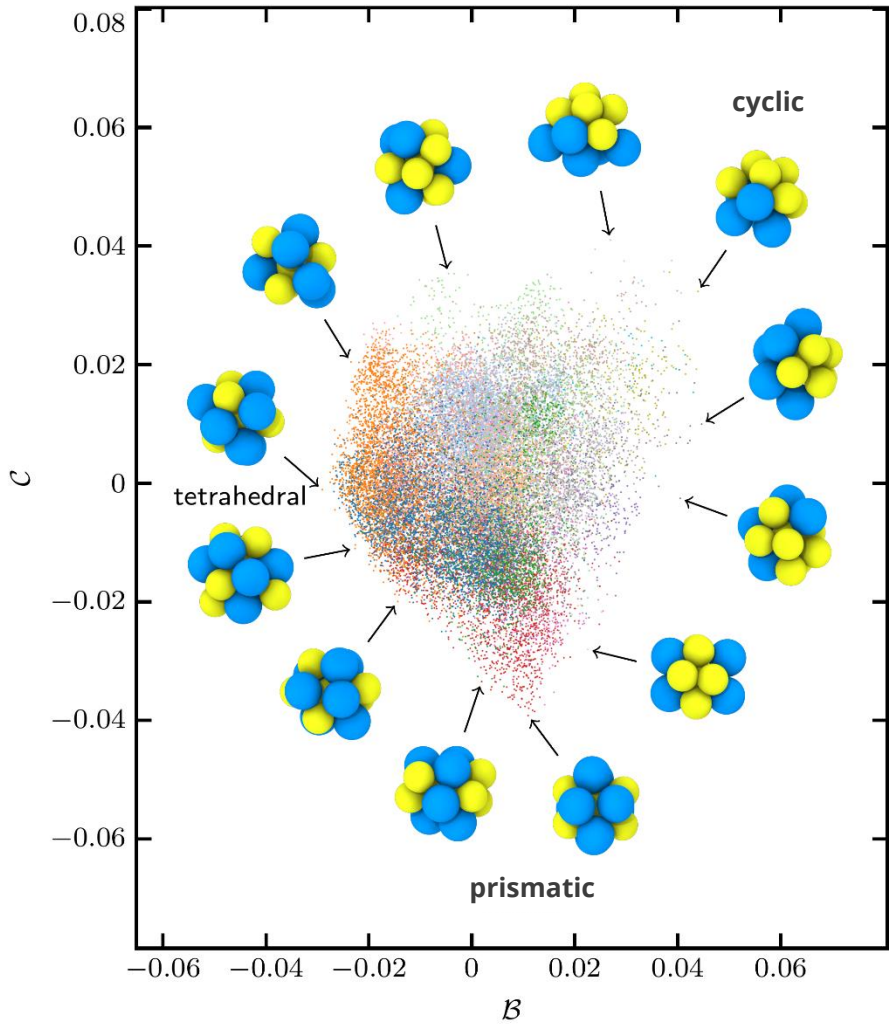
Quench rate impacts structure- but specifics are hazy





Filling in some details around quench rate and structure

All four MG samples plotted in diffusion space, colored by 20 agglomerative clusters



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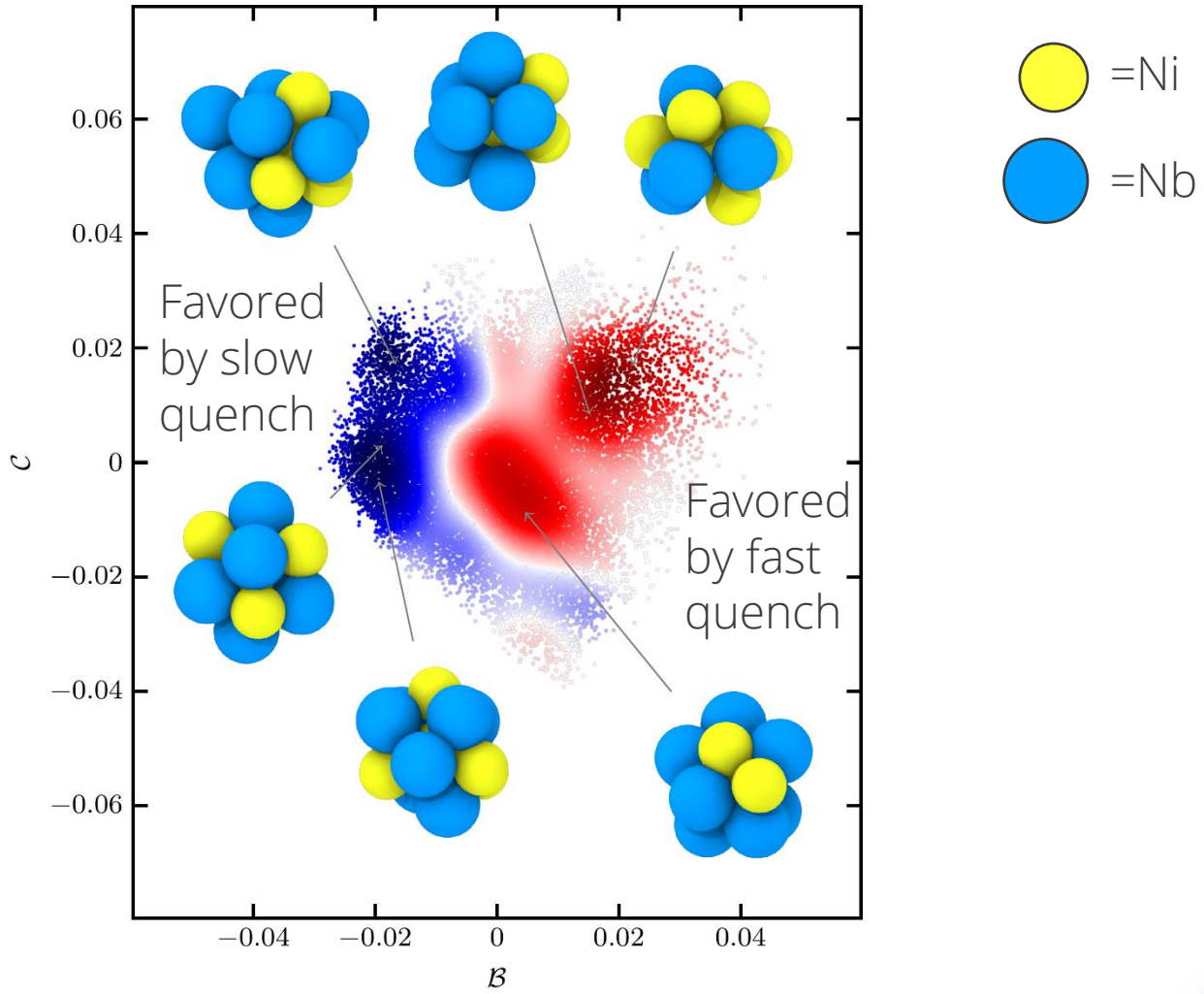

Final thoughts

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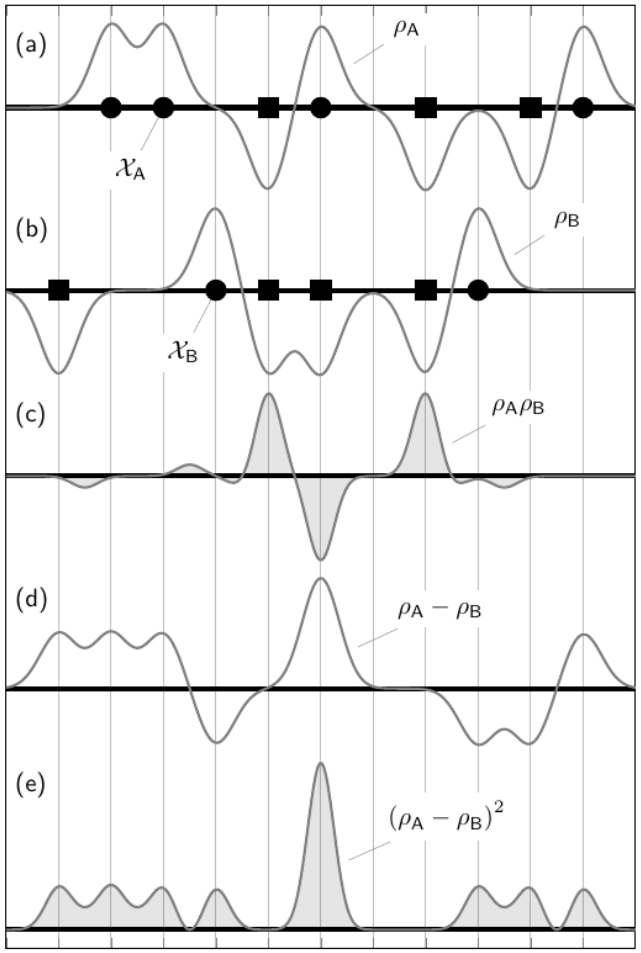
Understanding the GIIP Distance – One-dimensional Example



Atomic density function for atomic configuration 1

Atomic density function for atomic configuration 2

The GIIP between atomic configurations 1 and 2 is the integral of the product of their respective atomic density functions.

To find the squared GIIP distance between atomic configurations 1 and 2, take the difference between their respective atomic density functions, square it, and integrate the squared difference.