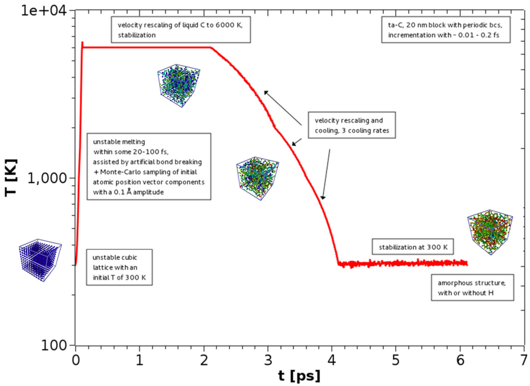
Symmetry-aware generative model for glassy motifs

Martin Uhrín, Anna Paulish

Computational Atomistic Methods and Machine Learning, SIMaP

Motivation



Timo Hakala, Kenneth Holmberg and Anssi Laukkanen. Lubricants. 9. 30. (2021).

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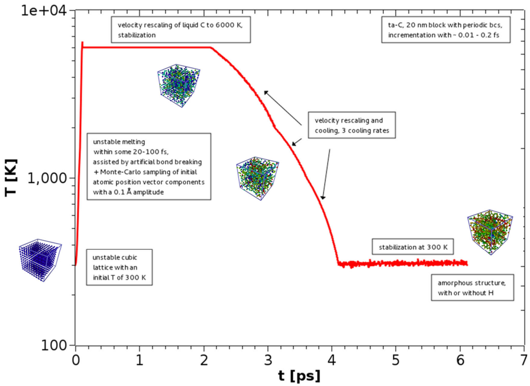
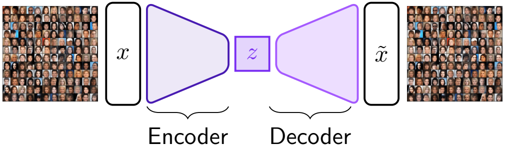


Given an example structure(s), can we teach a generative machine learning model to generate novel examples, bypassing the need for further molecular dynamics?

Timo Hakala, Kenneth Holmberg and Anssi Laukkanen. Lubricants. 9. 30. (2021).

The Variational Autoencoder

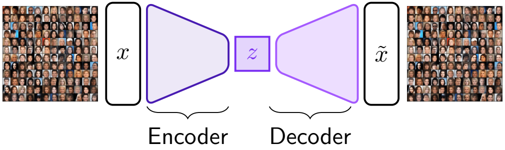
The autoencoder



$$\mathcal{L} = (x - \tilde{x})^2$$

The Variational Autoencoder

The autoencoder



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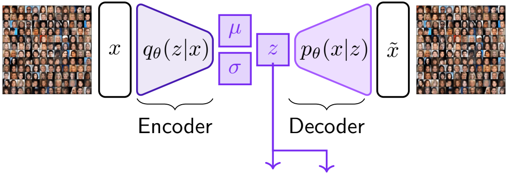
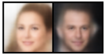
The *variational* autoencoder



$$z \sim \mathcal{N}(\mu, \sigma)$$



$$\mathcal{L} = (x - \tilde{x})^2 + \sum_j KL(q_j(z|x)||p(z))$$

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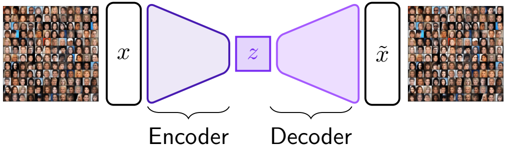

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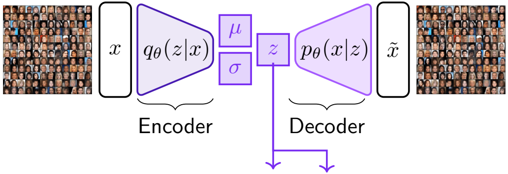

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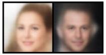
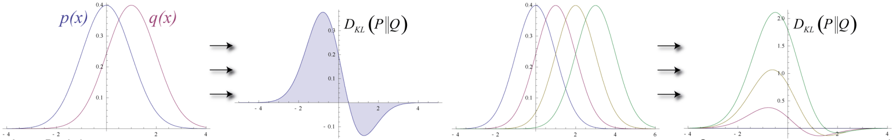


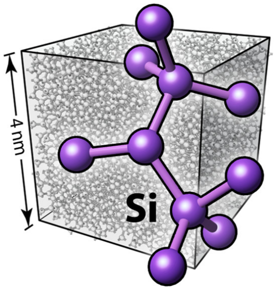
$$\mathcal{L} = (x - \tilde{x})^2 + \sum_j KL(q_j(z|x)||p(z))$$

Kullback-Leibler divergence

$$D_{\mathrm{KL}}(P \parallel Q) = \int_{-\infty}^{\infty} p(x) \ \log\left(\frac{p(x)}{q(x)}\right) \ \mathrm{d}x$$



Symmetry-aware representation of local atomic environments



V. L. Deringer et al., Journal of Physical Chemistry Letters **9**, 2879–2885 (2018)

Direct representation

$$x = \begin{bmatrix} x_1 & y_1 & z_1 \\ x_2 & y_2 & z_2 \\ & \vdots & \\ x_N & y_N & z_N \end{bmatrix}$$

Not a suitable input to a learning model. Consider

$$x' = xQ$$

where Q is a rotation matrix.

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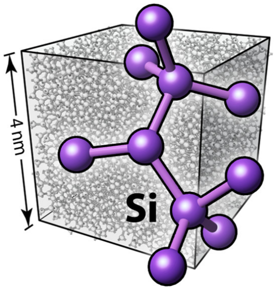

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Symmetry-aware representation

$$G = \begin{bmatrix} x^1 \cdot x^1 & \cdots & x^1 \cdot x^N \\ \vdots & \ddots & \vdots \\ x^N \cdot x^1 & \cdots & x^N \cdot x^N \end{bmatrix} = \begin{bmatrix} \|x^1\|^2 & \cdots & \|x^1\|\|x^N\| \cos\theta_{1N} \\ \vdots & \ddots & \vdots \\ \|x^N\|\|x^1\| \cos\theta_{N1} & \cdots & \|x^N\|^2 \end{bmatrix}$$

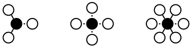
This representation is rotationally invariant.

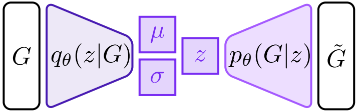
$$G = xx^T = (xQ)(xQ)^T = xQQ^Tx^T = xIx^T$$

Variational autoencoder for atomic motifs

Training

1. For each atom in unit cell, extract local atomic environment up to r_{cut}. Keeps closest n atoms





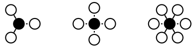
minimise

$\mathcal{L} = (G - \tilde{G})^2 + \sum_j KL(q_j(z|G)||p(z))$

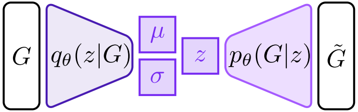
Variational autoencoder for atomic motifs

Training

❶ For each atom in unit cell, extract local atomic environment up to r_{cut}. Keeps closest n atoms



❷ Calculate Gram matrix $x^i \cdot x^j$, keep upper triangular part, $j \geq i$



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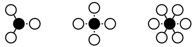
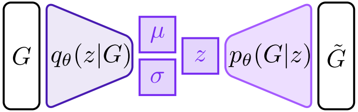


2. Calculate Gram matrix $x^i \cdot x^j$, keep upper triangular part, $j \geq i$
3. Generate permutation copies of atom labels i (data augmentation) e.g. $[1, 2, 3], [1, 3, 2], [2, 1, 3]$, etc



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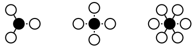

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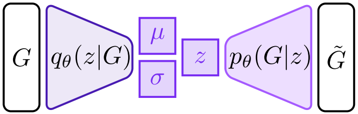
4. Train VAE using gradient-based optimisation



minimise
$\mathcal{L} = (G - \tilde{G})^2 + \sum_j KL(q_j(z|G)||p(z))$

Variational autoencoder for atomic motifs

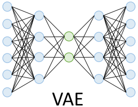
$$E = \begin{bmatrix} r_1 \\ r_2 \\ \vdots \\ r_N \end{bmatrix} = \begin{bmatrix} x_1 & y_1 & z_1 \\ x_2 & y_2 & z_2 \\ \vdots & \vdots & \vdots \\ x_N & y_N & z_N \end{bmatrix}$$

Initial atomic environment

Loss function on the invariants: $\|G - G'\|^2 + D_{KL}$

input

$$G = \begin{bmatrix} \langle \mathbf{r}_1, \mathbf{r}_1 \rangle & \langle \mathbf{r}_1, \mathbf{r}_2 \rangle & \dots & \langle \mathbf{r}_1, \mathbf{r}_N \rangle \\ \langle \mathbf{r}_1, \mathbf{r}_1 \rangle & \langle \mathbf{r}_1, \mathbf{r}_2 \rangle & \dots & \langle \mathbf{r}_1, \mathbf{r}_N \rangle \\ \vdots & \vdots & \ddots & \vdots \\ \langle \mathbf{r}_N, \mathbf{r}_1 \rangle & \langle \mathbf{r}_N, \mathbf{r}_2 \rangle & \dots & \langle \mathbf{r}_N, \mathbf{r}_N \rangle \end{bmatrix}$$



reconstructed

$$\begin{bmatrix} \langle \mathbf{r'}_1, \mathbf{r'}_1 \rangle & \langle \mathbf{r'}_1, \mathbf{r'}_2 \rangle & \dots & \langle \mathbf{r'}_1, \mathbf{r'}_N \rangle \\ \langle \mathbf{r'}_1, \mathbf{r'}_1 \rangle & \langle \mathbf{r'}_1, \mathbf{r'}_2 \rangle & \dots & \langle \mathbf{r'}_1, \mathbf{r'}_N \rangle \\ \vdots & \vdots & \ddots & \vdots \\ \langle \mathbf{r'}_N, \mathbf{r'}_1 \rangle & \langle \mathbf{r'}_N, \mathbf{r'}_2 \rangle & \dots & \langle \mathbf{r'}_N, \mathbf{r'}_N \rangle \end{bmatrix} = G'$$

SVD

$$\begin{bmatrix} x'_1 & y'_1 & z'_1 \\ x'_2 & y'_2 & z'_2 \\ \vdots & \vdots & \vdots \\ x'_N & y'_N & z'_N \end{bmatrix} = \begin{bmatrix} r'_1 \\ r'_2 \\ \vdots \\ r'_N \end{bmatrix} = E'$$

Reconstructed atomic environment

The problem with invariants space

$$\begin{bmatrix} x_1 & y_1 & z_1 \\ x_2 & y_2 & z_2 \\ & \vdots & \\ x_N & y_N & z_N \end{bmatrix} \quad \text{vs} \quad \begin{bmatrix} x^1 \cdot x^1 & \cdots & x^1 \cdot x^N \\ \vdots & \ddots & \vdots \\ x^N \cdot x^1 & \cdots & x^N \cdot x^N \end{bmatrix}$$

$$3N(-6) \quad \text{DoFs} \quad \text{vs} \quad \frac{N(N+1)}{2} \quad \text{DoFs}$$

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$$\begin{bmatrix} x_1 & y_1 & z_1 \\ x_2 & y_2 & z_2 \\ & \vdots & \\ x_N & y_N & z_N \end{bmatrix} \quad \text{vs} \quad \begin{bmatrix} x^1 \cdot x^1 & \cdots & x^1 \cdot x^N \\ \vdots & \ddots & \vdots \\ x^N \cdot x^1 & \cdots & x^N \cdot x^N \end{bmatrix}$$

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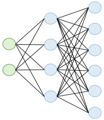
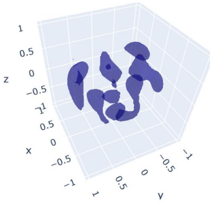


Decoder

→ $\tilde{\boldsymbol{\Phi}}$ →

Generated fingerprints



$\tilde{f}(r_i, \ldots)$

Density grid

The problem with invariants space

$$\begin{bmatrix} x_1 & y_1 & z_1 \\ x_2 & y_2 & z_2 \\ & \vdots & \\ x_N & y_N & z_N \end{bmatrix} \quad \text{vs} \quad \begin{bmatrix} x^1 \cdot x^1 & \cdots & x^1 \cdot x^N \\ \vdots & \ddots & \vdots \\ x^N \cdot x^1 & \cdots & x^N \cdot x^N \end{bmatrix}$$

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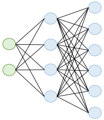
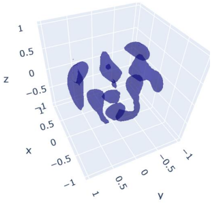


Decoder

$\longrightarrow$ $\tilde{\boldsymbol{\Phi}}$ $\longrightarrow$

Generated fingerprints



$\tilde{f}(r_i, \ldots)$

Density grid

The solution: synchronisation

X X’

The problem with invariants space

$$\begin{bmatrix} x_1 & y_1 & z_1 \\ x_2 & y_2 & z_2 \\ & \vdots & \\ x_N & y_N & z_N \end{bmatrix} \quad \text{vs} \quad \begin{bmatrix} x^1 \cdot x^1 & \cdots & x^1 \cdot x^N \\ \vdots & \ddots & \vdots \\ x^N \cdot x^1 & \cdots & x^N \cdot x^N \end{bmatrix}$$

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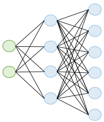

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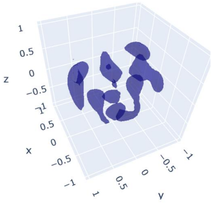
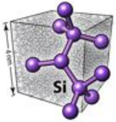


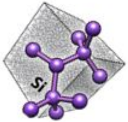
$\tilde{f}(r_i, \dots)$

Density grid

The solution: synchronisation



$\Rightarrow$ $\boldsymbol{\Phi}$ $\Leftarrow$



X X'

We know

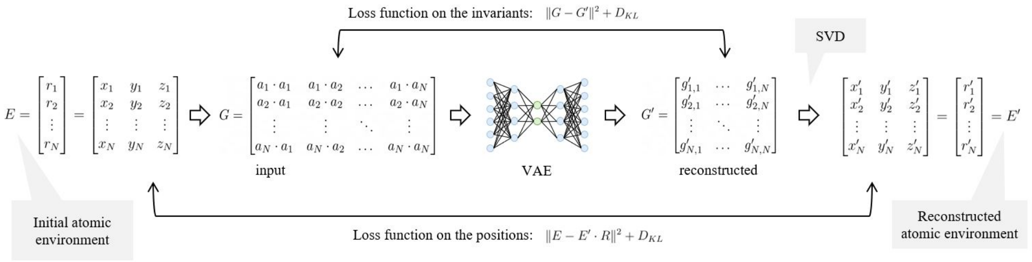
$$X = QX'$$

with some rotation matrix Q.
We can solve for this using:

$$\min_Q \|X - QX'\|_F$$

Training and generating

Training



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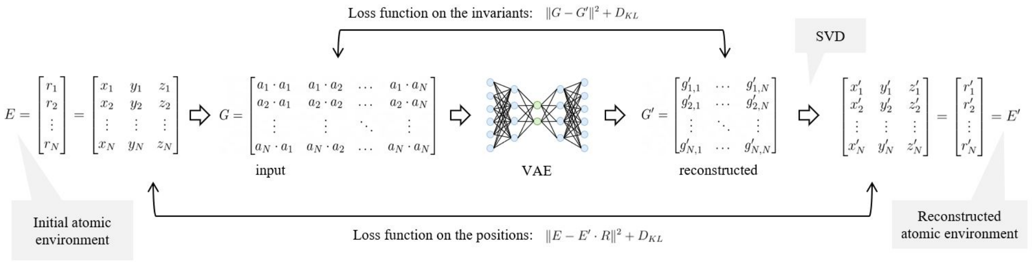
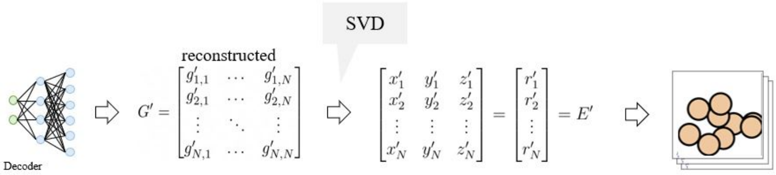


Generating

Draw n_Z samples from $\mathcal{N}(0, 1)$



Variational autoencoder for atomic motifs

All tests performed in amorphous Si, 512 atom unit cells generated using ML potential trained on DFT[1].

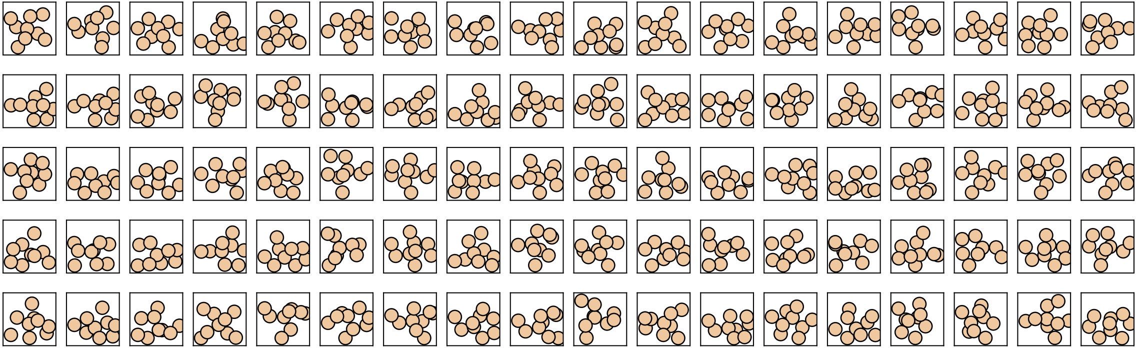
- Radial cutoff: 4 Å
- 8 atoms per environment
- latent space: 8 neurons (compared to $3n - 6 = 18$ DoFs)
- Encoder architecture 36-28-18-8 with tanh activations

Data normalisation

- $G'_{ii} = (G_{ii} - \mu_{\text{diag}})/\sigma_{\text{diag}}$
- $G'_{ij} = (G_{ij} - \mu_{\text{off-diag}})/\sigma_{\text{off-diag}}, i < j$

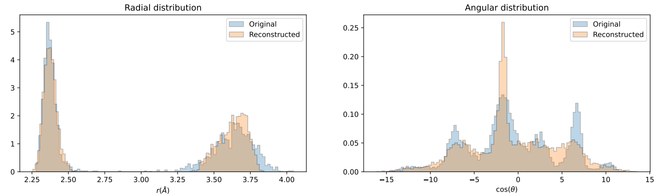
V. L. Deringer et al., Journal of Physical Chemistry Letters **9**, 2879–2885 (2018)

Example generated environments



Model performance

Training reconstruction



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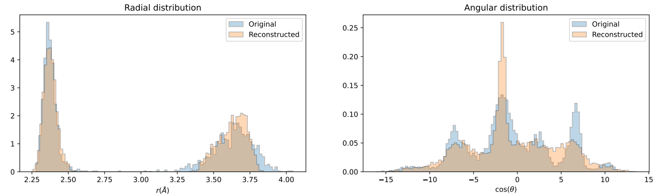

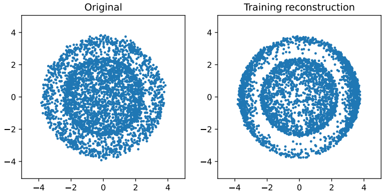

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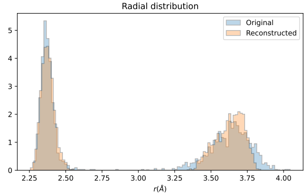
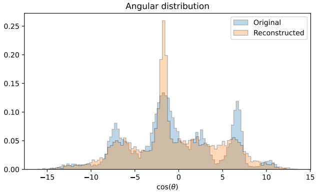
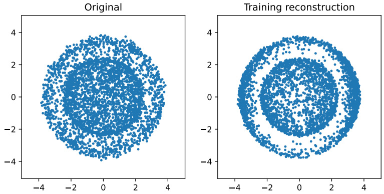
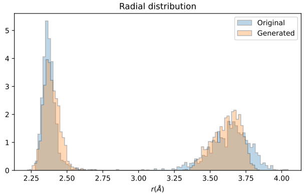
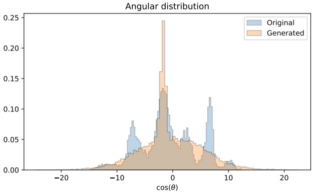






Generated samples





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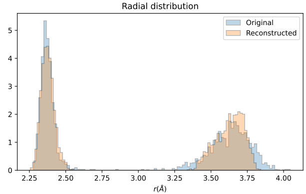

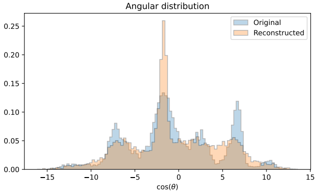

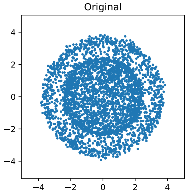

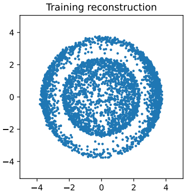

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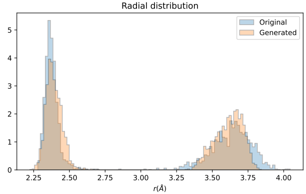
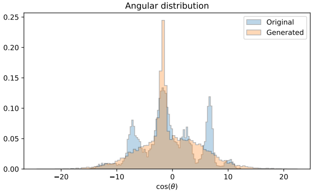
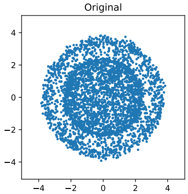
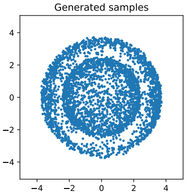


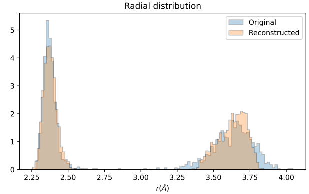


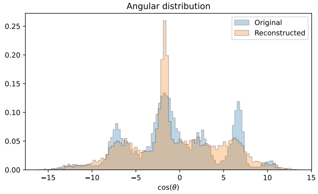


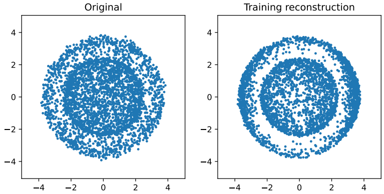


Model performance

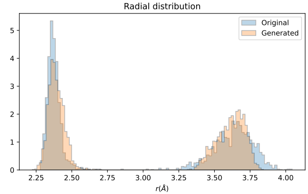
Training reconstruction

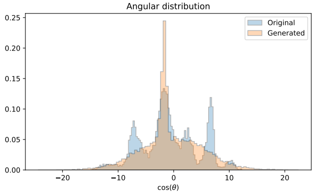


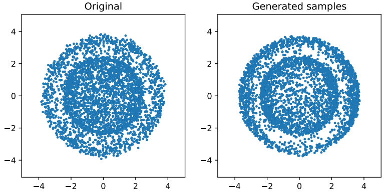


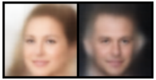


Generated samples

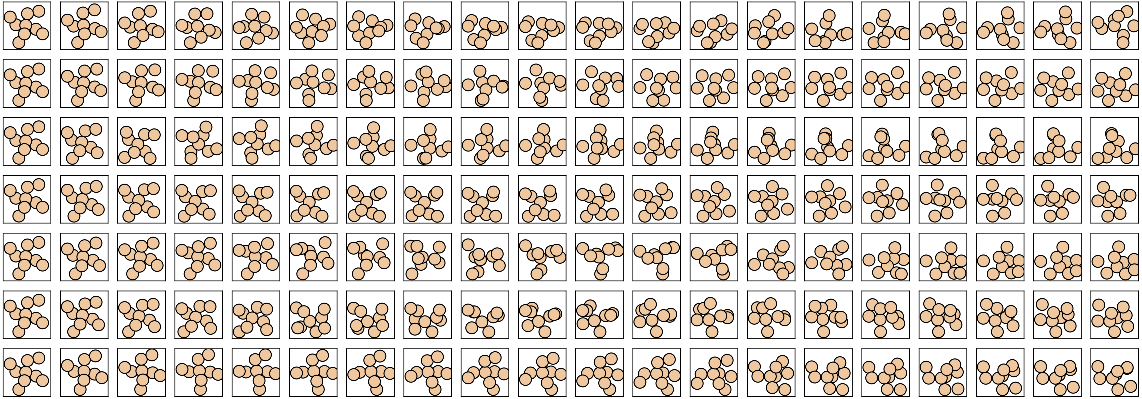




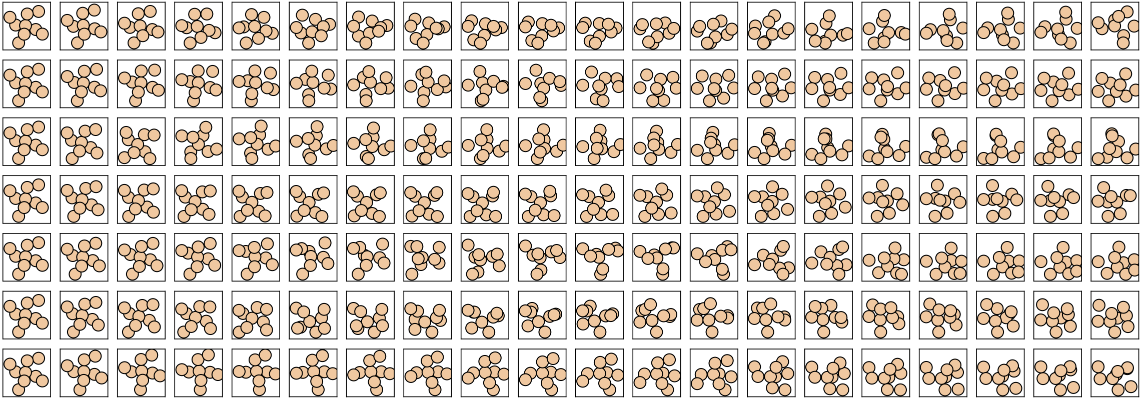


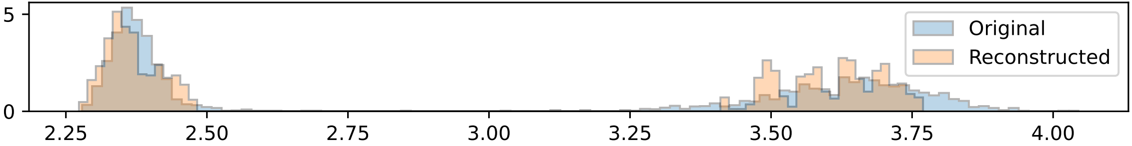


Interpolating between motifs



Interpolating between motifs





Inpainting

Atom infilling: Building complete unit cells

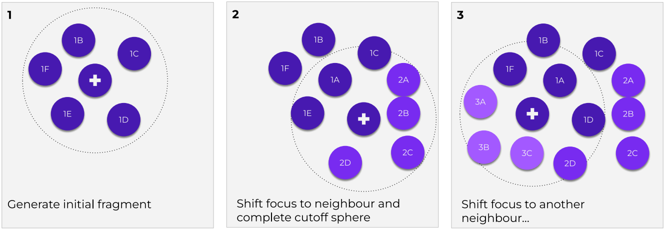
Image inpainting

T. F. Chan and J. Shen, Communications on Pure and Applied Mathematics **58**, 579–619 (2005)

Atom infilling: Building complete unit cells

Image inpainting

Environment infilling



T. F. Chan and J. Shen, Communications on Pure and Applied Mathematics **58**, 579–619 (2005)

Atom infilling

Setup

- Calculate initial Gram matrix with known atoms, fill in rest with noise G^I
- Project into latent space $z^I = f_{\text{encode}}(G^I)$

Atom infilling

Setup

- Calculate initial Gram matrix with known atoms, fill in rest with noise G^I
- Project into latent space $z^I = f_{\text{encode}}(G^I)$

Objective function

$$\min_z \| f_{\text{decode}}(z) - G \circ f_{\text{decode}}(z) \|^2$$

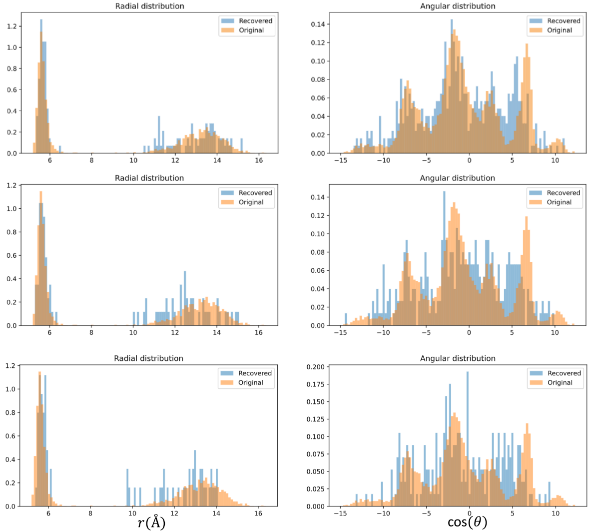
i.e. find point that lies on latent space manifold *and* where known atoms are in their original positions.

Atom infilling results

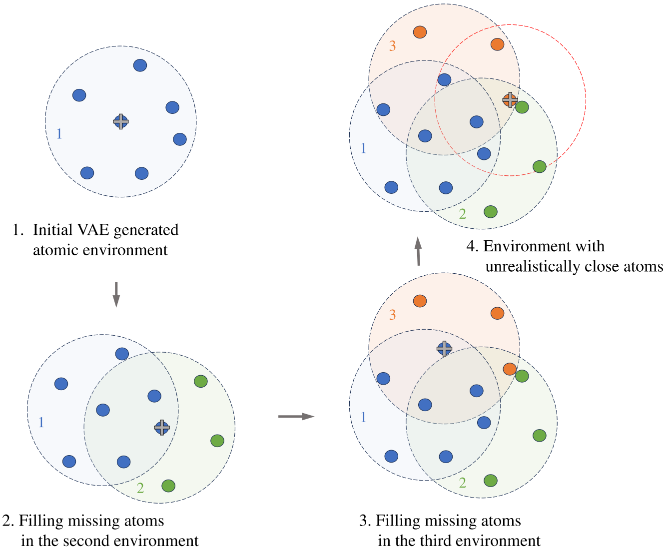
Fixed atoms: 7

Fixed atoms: 6

Fixed atoms: 5



Atom infilling results



Conclusion

Take aways

Symmetry-aware representations allow us to make efficient models that respect isometries of atomic gemetries

Using relatively little data we can learn to generate novel motifs and interpolate between them

Variational Autoencoder provides a relatively simple and powerful generative architecture

Infilling will require *longer sightedness*

Take aways

Symmetry-aware representations allow us to make efficient models that respect isometries of atomic gemetries

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Infilling will require *longer sightedness*

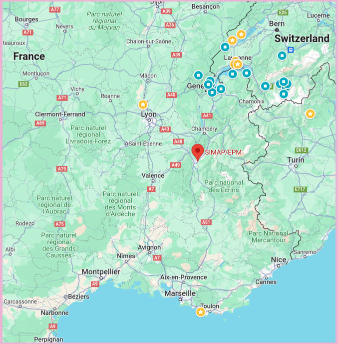
Next Steps

Migrate model onto graph network

Diffusion

Cartesian → spherical harmonic representation

We're hiring!



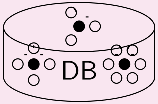
Invariant and equivariant machine learning

milad

e3nn

- Theory of atomistic structure representations
- Inversion of invariant fingerprints
- Highly data-efficient equivariant neural networks

Generative models



- Linking 3D geometry to properties
- Motif based construction of molecules and materials
- Coupling experiment and theory

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