Kinetic Langevin Diffusion for Crystalline Materials Generation

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TL;DR

We propose a novel diffusion model for crystalline materials generation, where the key innovation is a **diffusion process to model the fractional coordinates, inspired by kinetic Langevin dynamics.**

Problem setting

Data

Unit cell $\boldsymbol{x} = (\boldsymbol{f}, \boldsymbol{l}, \boldsymbol{a})$

Fractional coordinates $\boldsymbol{f} = (\boldsymbol{f}_1, \dots, \boldsymbol{f}_K) \in [0,1)^{3\times K}$

Lattice vectors $\boldsymbol{l} = (\boldsymbol{l}_1, \boldsymbol{l}_2, \boldsymbol{l}_3) \in \mathbb{R}^{3\times 3}$

Atomic species $\boldsymbol{a} = (a_1, \dots, a_K) \in \mathbb{Z}^K$

Tasks

Crystal Structure Prediction (CSP) $p_\theta(\boldsymbol{f}, \boldsymbol{l} | \boldsymbol{a}) \approx p_{\text{data}}(\boldsymbol{f}, \boldsymbol{l} | \boldsymbol{a})$

De-Novo Generation (DNG) $p_\theta(\boldsymbol{f}, \boldsymbol{l}, \boldsymbol{a}) \approx p_{\text{data}}(\boldsymbol{f}, \boldsymbol{l}, \boldsymbol{a})$

Symmetries

Permutation of atom indices

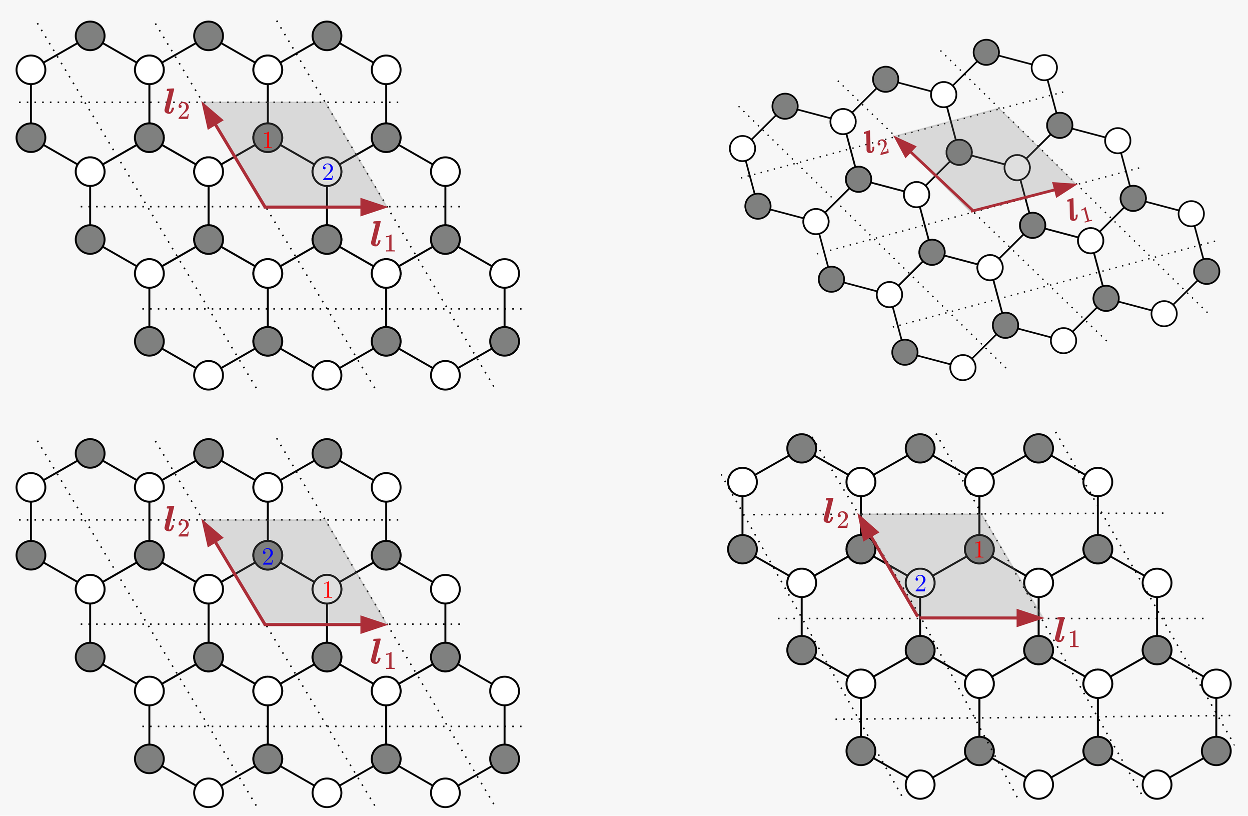
$$p(\boldsymbol{f}, \boldsymbol{l}, \boldsymbol{a}) = p(g \cdot \boldsymbol{f}, \boldsymbol{l}, g \cdot \boldsymbol{a}), \quad \forall g \in S_K$$

Rotation of lattice vectors

$$p(\boldsymbol{f}, \boldsymbol{l}, \boldsymbol{a}) = p(\boldsymbol{f}, g \cdot \boldsymbol{l}, \boldsymbol{a}), \quad \forall g \in \mathrm{SO}(3)$$

Periodic translation of fractional coordinates

$$p(\boldsymbol{f}, \boldsymbol{l}, \boldsymbol{a}) = p(g \cdot \boldsymbol{f}, \boldsymbol{l}, \boldsymbol{a}) \quad \forall g \in \mathbb{T}^3$$



Previous Work - Riemannian score-matching

$$\boldsymbol{f} \in [0,1)^{3\times K} \cong \mathbb{T}^{3\times K}$$

Brownian motion on a torus

$$\mathrm{d}\boldsymbol{f}_t = \sqrt{\mathrm{d}\sigma^2(t)/\mathrm{d}t}\,\mathrm{d}\boldsymbol{w}_t$$

Transition kernel

$$p_{t|0}(\boldsymbol{f}_t | \boldsymbol{f}_0) \propto \sum_{\boldsymbol{k} \in \mathbb{Z}^{3\times K}} \exp\left(-\frac{\|\boldsymbol{f}_t - \boldsymbol{f}_0 + \boldsymbol{k}\|^2}{2\sigma^2(t)}\right)$$

Wrapped-Normal distribution

Kinetic Langevin Diffusion for Materials (KLDM)

Auxiliary variables

$$(\boldsymbol{f}_t, \boldsymbol{v}_t)$$

Velocities living on the Lie algebra (Euclidean space)

Forward process

$$\begin{cases} \mathrm{d}\boldsymbol{f}_t &= \boldsymbol{f}_t \boldsymbol{v}_t \mathrm{d}t, \\ \mathrm{d}\boldsymbol{v}_t &= -\gamma \boldsymbol{v}_t \mathrm{d}t + \sqrt{2\gamma}\mathrm{d}\boldsymbol{w}_t^{\mathfrak{g}}, \end{cases}$$

Linear ODE on the manifold

Usual SDE on the Lie algebra

$$\boldsymbol{f}\exp(\boldsymbol{v}\mathrm{d}t) \to w(\boldsymbol{f} + \boldsymbol{v}\mathrm{d}t)$$

Reverse process

$$\begin{cases} \mathrm{d}\boldsymbol{f}_t = \boldsymbol{f}_t \boldsymbol{v}_t \mathrm{d}t, \\ \mathrm{d}\boldsymbol{v}_t = [-\gamma \boldsymbol{v}_t - 2\gamma \nabla_{\boldsymbol{v}_t} \log p_t(\boldsymbol{f}_t, \boldsymbol{v}_t)]\,\mathrm{d}t + \sqrt{2\gamma}\,\mathrm{d}\boldsymbol{w}_t^{\mathfrak{g}}\,. \end{cases}$$

Trivialized Diffusion Model [Zhu et al., 2024]

Transition kernel

Procedure remains simulation-free

$$p_{t|0}(\boldsymbol{f}_t, \boldsymbol{v}_t | \boldsymbol{f}_0, \boldsymbol{v}_0) = \mathrm{WN}_{\boldsymbol{r}}\left(\mathrm{logm}(\boldsymbol{f}_0^{-1}\boldsymbol{f}_t) | \boldsymbol{\mu}_{\boldsymbol{r}_t}, \boldsymbol{\sigma}^2_{\boldsymbol{r}_t}\right) \cdot \mathcal{N}_{\boldsymbol{v}}(\boldsymbol{v}_t | \boldsymbol{\mu}_{\boldsymbol{v}_t}, \boldsymbol{\sigma}^2_{\boldsymbol{v}_t})$$

$$\nabla_{\boldsymbol{v}_t} \log p_{t|0} = \nabla_{\boldsymbol{\mu}_{\boldsymbol{r}_t}} \log \mathrm{WN}\left(\boldsymbol{r}_t | \boldsymbol{\mu}_{\boldsymbol{r}_t}, \sigma^2_{\boldsymbol{r}_t}\mathbf{I}\right) \frac{\partial \boldsymbol{\mu}_{\boldsymbol{r}_t}}{\partial \boldsymbol{v}_t} + \nabla_{\boldsymbol{v}_t} \log \mathcal{N}_{\boldsymbol{v}}\left(\boldsymbol{v}_t | \boldsymbol{\mu}_{\boldsymbol{v}_t}, \sigma^2_{\boldsymbol{v}_t}\mathbf{I}\right)$$

Design choices

1) Distribution of the initial velocities $p(\boldsymbol{v}_0) = \delta(\boldsymbol{v}_0)$ "Atoms are at rest"

2) Improved score parameterisation

$$\nabla_{\boldsymbol{v}_t} \log p_{t|0}(\boldsymbol{f}_t, \boldsymbol{v}_t | \boldsymbol{f}_0, \boldsymbol{v}_0) = \frac{1 - e^{-t}}{1 + e^{-t}} \nabla_{\boldsymbol{\mu}_{\boldsymbol{r}_t}} \mathrm{WN}\left(\boldsymbol{r}_t | \boldsymbol{\mu}_{\boldsymbol{r}_t}, \sigma^2_{\boldsymbol{r}_t}\mathbf{I}\right) \boxed{- \frac{\boldsymbol{\varepsilon}_{\boldsymbol{v}}}{\sigma_{\boldsymbol{v}_t}}}$$

"With 1) in place"

$$s_\theta^{\boldsymbol{v}}(\boldsymbol{x}_t, t) = \frac{1 - e^{-t}}{1 + e^{-t}} s_\theta^{\boldsymbol{f}}(\boldsymbol{x}_t, t) - \boxed{\frac{\boldsymbol{v}_t}{\sigma^2_{\boldsymbol{v}_t}}}, \qquad \boldsymbol{v}_t = \boxed{\boldsymbol{\mu}_{\boldsymbol{v}_t}} + \boldsymbol{\sigma}_{\boldsymbol{v}_t}\varepsilon_{\boldsymbol{v}}$$

3) Mean-free velocity field $\sum_k^K \boldsymbol{v}_k = \mathbf{0}$ $\qquad \boldsymbol{\mu}_{\boldsymbol{v}_t} = \mathbf{0}, \forall t$

Other modalities

(C) Continuous diffusion on one-hot encodings

(C-AB) Continuous diffusion on Analog Bits

(D) Discrete (masking) diffusion

Experiments

CSP

KLDM is competitive on MP-20 and MPTS-52 using an EM sampler, and provides improved performance with a PC samples.

Model	Perov-5 MR [%] ↑	Perov-5 RMSE ↓	MP-20 MR [%] ↑	MP-20 RMSE ↓	MPTS-52 MR [%] ↑	MPTS-52 RMSE ↓
			Metrics @ 1			
CDVAE	45.31	0.1138	33.90	0.1045	5.34	0.2106
DiffCSP (PC)	52.02	0.0760	51.49	0.0631	12.19	0.1786
EquiCSP (PC)	52.02	0.0707	57.59	0.0510	14.85	**0.1169**
FlowMM	**53.15**	0.0992	61.39	0.0566	17.54	0.1726
KLDM-ε (EM)	53.14 ±.6	0.0758 ±.002	61.72 ±.2	0.0686 ±.001	17.71 ±.3	0.2023 ±.005
KLDM-ε (PC)	52.72 ±.8	0.0678 ±.002	65.37 ±.1	**0.0455** ±.001	21.46 ±.2	0.1339 ±.002
KLDM-$\boldsymbol{x}_0$ (EM)	52.44 ±.7	0.0698 ±.002	62.92 ±.2	0.0833 ±.002	21.13 ±.2	0.1800 ±.003
KLDM-$\boldsymbol{x}_0$ (PC)	52.14 ±.9	**0.0647** ±.002	**65.83** ±.2	0.0517 ±.001	**23.93** ±.2	0.1276 ±.002
			Metrics @ 20			
CDVAE	88.51	0.0464	66.95	0.1026	20.79	0.2085
DiffCSP (PC)	98.60	**0.0128**	77.93	0.0492	34.02	0.1749
FlowMM	98.60	0.0328	75.81	0.0479	34.05	0.1813
KLDM-ε (EM)	**99.97**	0.0152	**83.68**	0.0532	39.04	0.1865
KLDM-ε (PC)	99.94	0.0226	81.08	**0.0440**	**39.81**	0.1462
KLDM-$\boldsymbol{x}_0$ (EM)	99.89	0.0186	82.94	0.0575	37.77	0.1673
KLDM-$\boldsymbol{x}_0$ (PC)	99.92	0.0255	80.18	0.0453	37.10	**0.1394**

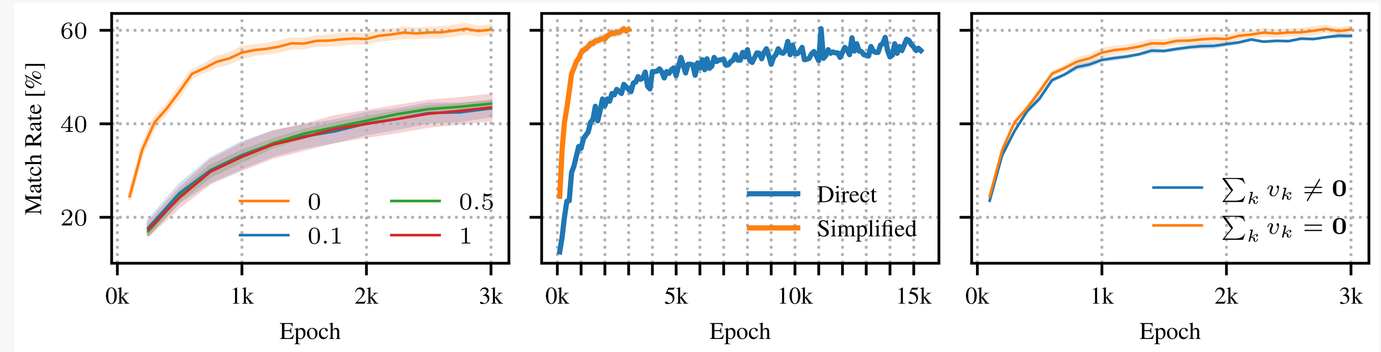
DNG

KLDM produces samples that are more stable and closer to the convex hull on average, but with slightly higher RMSD and fewer unique and stable samples.

	RMSD [Å] ↓	Avg. above hull [eV/atom] ↓	Stable [%] ↑	S.U.N. [%] ↑
MatterGen-MP*	0.147	0.201	47.05	25.76
DiffCSP*	0.413	0.189	41.25	20.13
KLDM-$\boldsymbol{x}_0$ (C)	0.371 ±.01	0.269 ±.01	38.62 ±.1	16.67 ±.1
KLDM-$\boldsymbol{x}_0$ (C-AB)	0.296 ±.01	0.187 ±.01	49.84 ±.1	17.91 ±.1
KLDM-$\boldsymbol{x}_0$ (D)	0.283 ±.01	0.155 ±.01	59.21 ±.1	18.52 ±.1

Ablation study

(**left**) Variance of initial velocity, (**center**) simplified parametrization, and (**right**) zero-net translation velocity field.



Conclusion

In **KLDM**, fractional coordinates are modelled through a coupling with auxiliary variables representing velocities, leading to improved generative performance on both CSP and DNG tasks.