LLM Meets Diffusion: A Hybrid Framework for Crystal Material Generation

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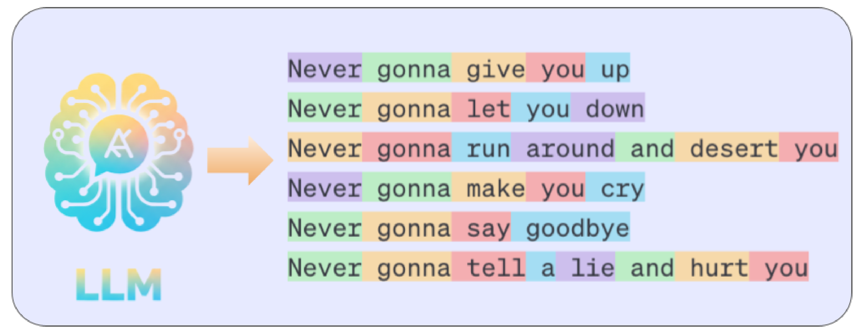
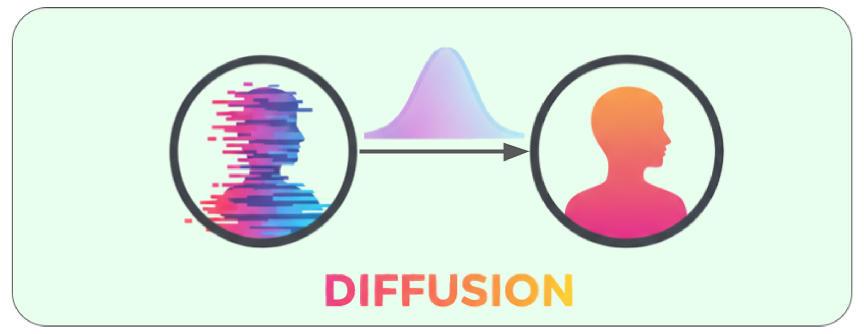
Motivation

The Challenge: Designing novel periodic crystal structures requires handling both discrete atomic compositions and continuous spatial features (positions, lattice).

- LLMs excel at discrete, compositional features (Atom Types).
- Diffusion Models are effective at continuous variables (Coordinates, Lattice).
- Their integration combines these strengths for crystal generation.





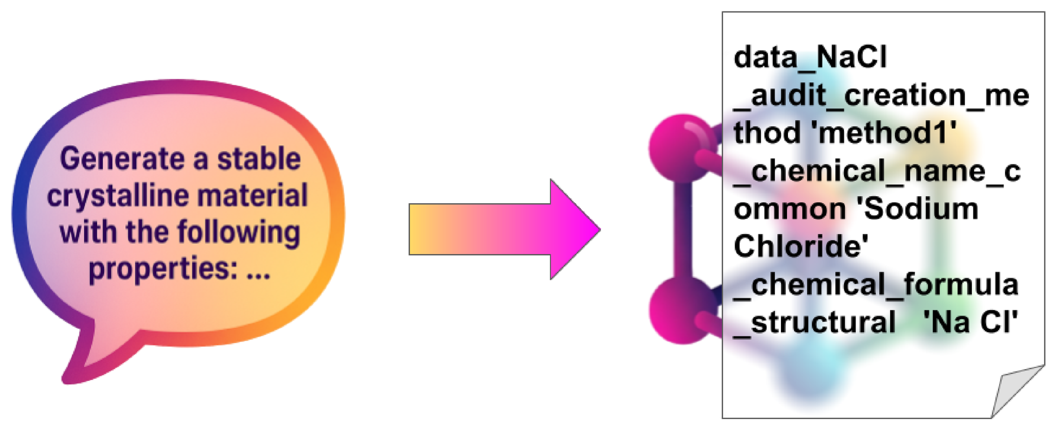
Problem Setting

A material is described by three parameters $M = (A, X, L)$

M : Crystalline Material Structure = A : Atom types — Discrete integer (1 to 93); X : Fractional coordinates — Continuous periodic (0 to 1); L : Lattice parameters — Continuous (real number)

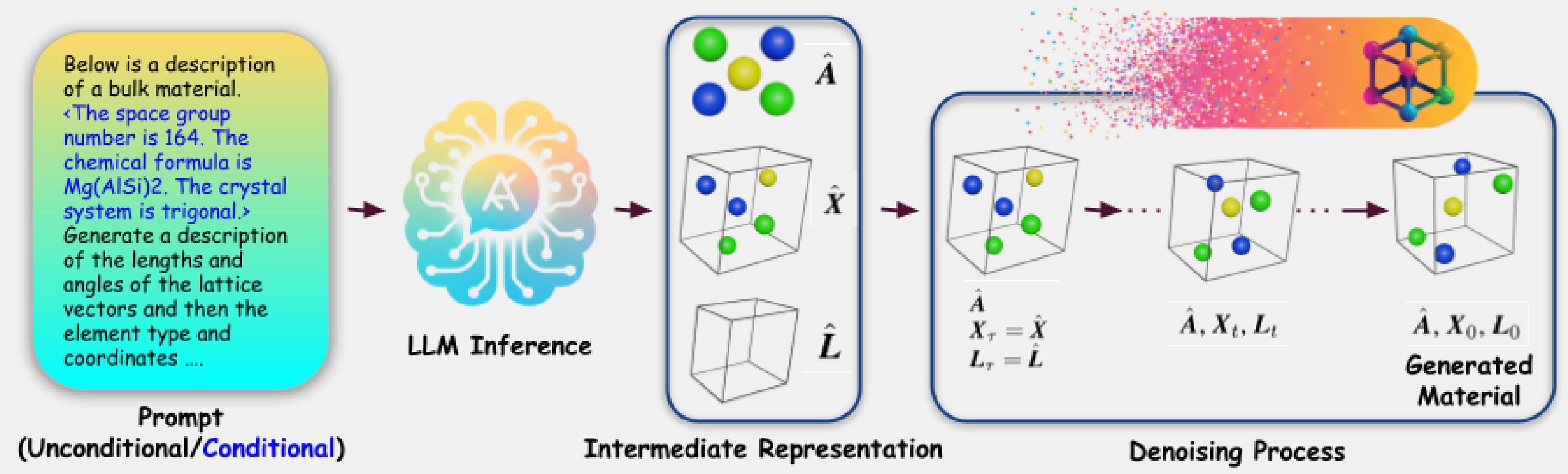
Given: A dataset of stable crystalline materials along with properties.
Outcome: A set of novel crystalline material structures that are also stablei.

LLM training objective:
Input: A text prompt optionally containing a set of desired properties (like space group, crystal symmetry, formation energy, band gap, etc.)
Fine tuning objective: CIF representation of the target material.



Diffusion training objective:
Input: A valid crystalline material structure. Training objective (CSP task): Structure (coordinates and lattice parameters) of the given material from its composition (set of atoms) only.

CrysLLMGen Architecture



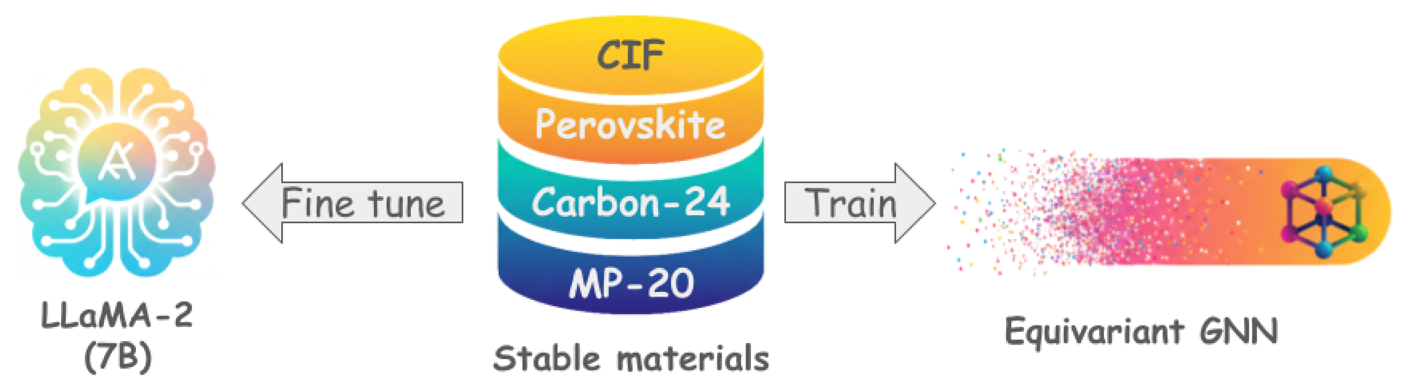
Method Overview

LLM

- Pre-trained LLM - LLaMA-2 (7B)
- Fine-tuned on dataset of stable crystal structures.
- Captures compositional patterns.

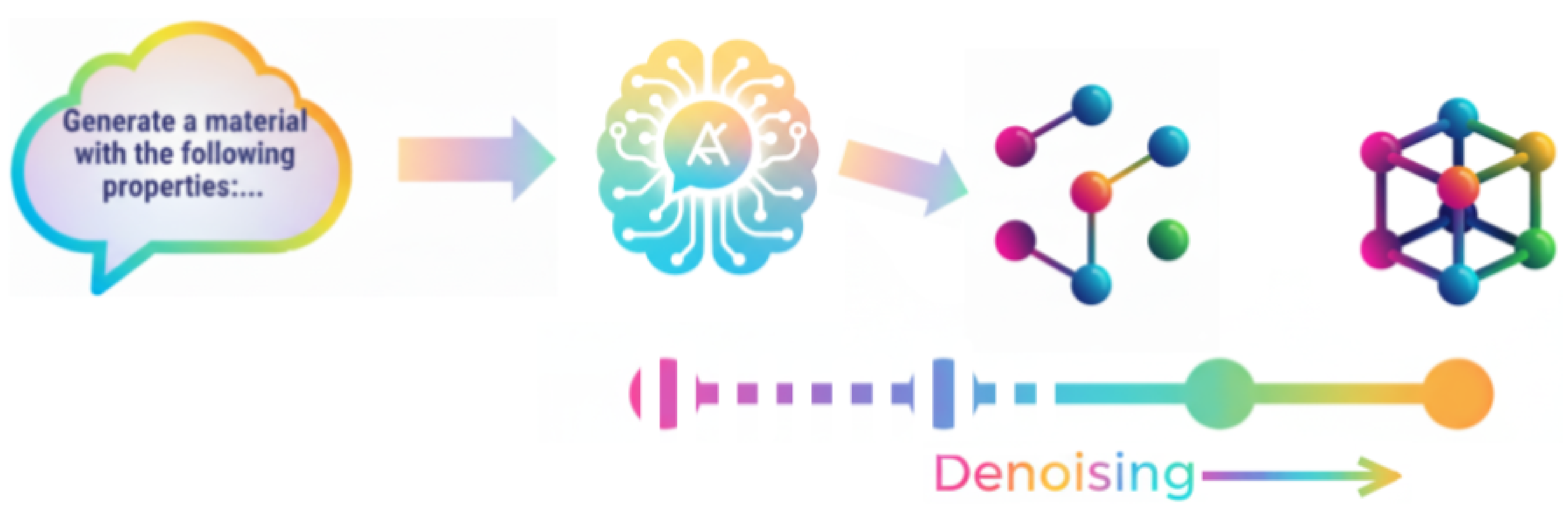
Diffusion

- EGNN-based denoising model.
- Guarantees periodic E(3) equivariance.
- Wrapped normal diffusion on fractional coordinates.



Sampling Procedure

- LLM produces candidate materials with high compositional validity but low structural validity.
- Diffusion Model refines candidate material keeping composition fixed.
- Diffusion denoising process starts from an intermediate timestep because it is starting from candidate materials instead of Gaussian prior.

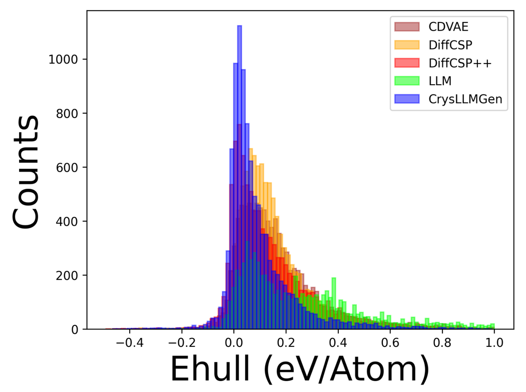


Results

Dataset	Category	Model	Validity ↑		Coverage ↑	
			Struct	Comp	Prec	Rec
MP-20	Diffusion	CDVAE	100	86.70	99.49	99.15
		DiffCSP	100	83.25	99.76	99.71
		DiffCSP++	99.94	85.12	99.59	99.73
		MatterGen	100	86.34	99.45	99.59
		UniMat	97.20	89.40	99.70	99.80
		SymmCD	92.30	87.13	98.78	97.33
	Flow Matching	FlowMM	96.85	83.19	99.58	99.49
		FlowLLM	99.94	90.84	99.82	96.95
	Bayesian Flow Networks	CrysBFN	100	87.51	99.79	99.09
	LLMs	LLaMA-2 (7B)	97.70	93.55	99.32	96.95
	LLM + Diffusion	CrysLLMGen (7B)	99.94	93.55	99.84	98.52

Table 1. Performance of different categories of models on unconditional generation task

Stability & Novelty



Method	% MS ↑	% MSUN ↑	% Stable ↑	% SUN ↑
CDVAE	23.58	21.99	3.08	2.56
DiffCSP	35.04	32.19	7.36	5.61
DiffCSP++	42.39	30.56	8.58	6.55
FlowMM	31.64	22.46	4.76	3.06
SymmCD	40.01	31.69	9.99	6.76
Llama-2 (7B)	56.60	26.66	12.67	4.84
CrysLLMGen (7B)	62.02	35.94	16.79	9.21

Conclusions

- CrysLLMGen bridges discrete–continuous divide.
- Enables text-conditional crystal generation.
- Paves the way for multi-modal generative materials design.

Presentation Recording