LLM Meets Diffusion: A Hybrid Framework for Crystal Material Generation

Subhojyoti Khastagir, Kishalay Das, Pawan Goyal, Seung-Cheol Lee, Satadeep Bhattacharjee, Niloy Ganguly

CNeRG
NEURAL INFORMATION PROCESSING SYSTEMS

Introduction and Motivation

Significance of Material Generation

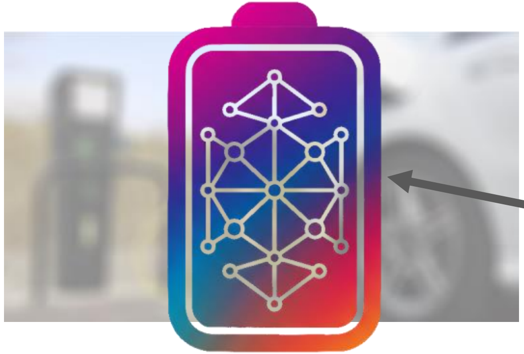
Low energy
density

Flammable
electrolytes

Costly catalytic
converters

Low efficiency
solar cells

Significance of Material Generation



Solid state batteries with
high energy density

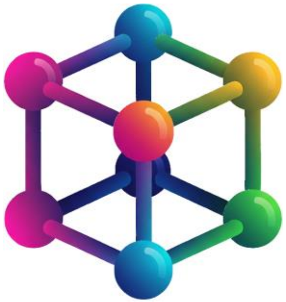
Electrolytes with low
thermal conductivity

Materials

Rare-earth element
free catalysts

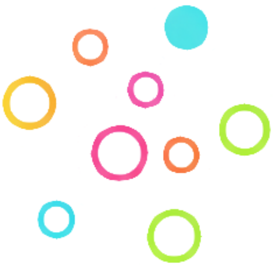
High efficiency
perovskite solar cells

The Material Generation Challenge



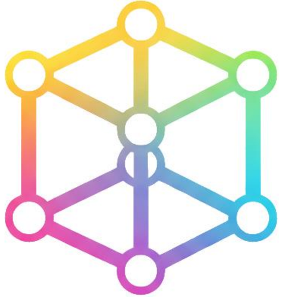
M = (A, X, L)

=



A: Atom Types

X: Fractional Coordinates

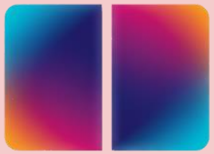


L: Lattice parameters

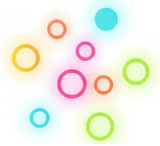
Crystal structure involves multimodal components:

- Discrete (Atom types, A)
- Continuous (Coordinates X, Lattice L)

Difficult to capture material symmetries



Limitation of Existing Models



Large Language Models

- High compositional validity (discrete features)
- Low structural validity (continuous features)

Diffusion Models

- High structural validity (continuous features)
- Low compositional validity (discrete features)

Validity Metric	Denoising Models CDVAE	DiffCSP	FlowMM	LLMs LLaMA-2(7B)
Structural Validity	100	100	96.85	96.40
Compositional Validity	86.70	83.25	83.19	93.30

CrysLLMGen: The Hybrid Framework

Highlights of CrysLLMGen

Novel hybrid architecture

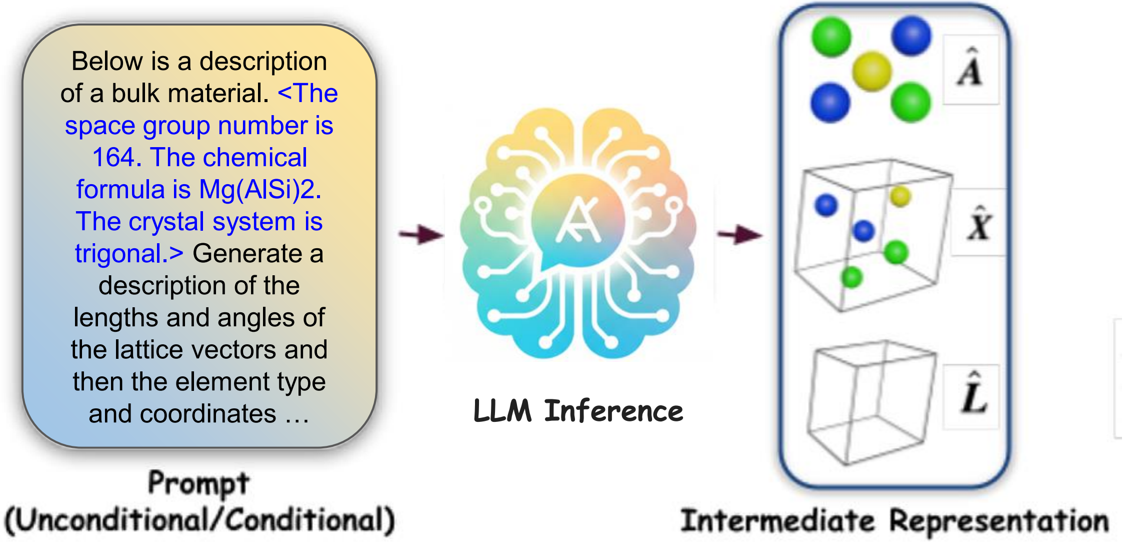
High structural validity

High compositional validity

Validity Metric	Denoising Models CDVAE	DiffCSP	FlowMM	LLMs LLaMA-2(7B) [7]	LLM+Diffusion CrysLLMGen(7B)
Structural Validity	100	100	96.85	96.40	99.94
Compositional Validity	86.70	83.25	83.19	93.30	93.55

2 stage sampling process

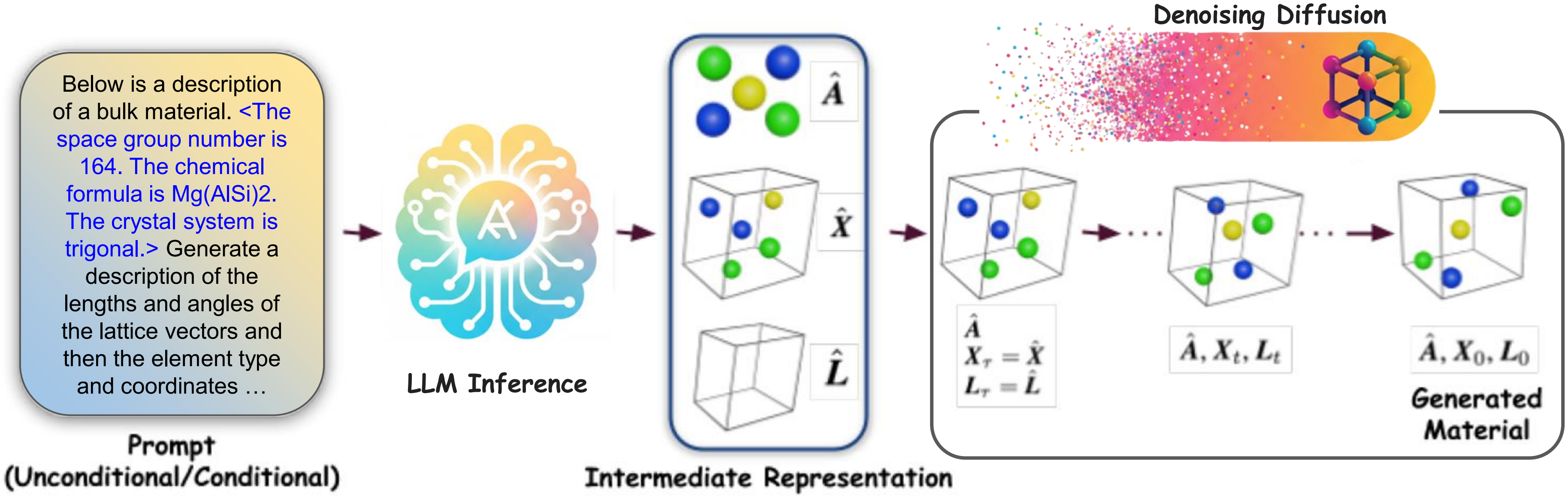
Stage 1: Fine tuned LLM generates intermediate representation



2 stage sampling process

Stage 1: Fine tuned LLM generates intermediate representation

Stage 2: Retain A and refine X, L



Implementation

Fine tune

CIF
Perovskite
Carbon-24
MP-20

Train

Built upon CSPNet
EGNN architecture

LLaMA-2 (7B)
Fine-tuned on textual representation of crystal materials

Stable crystal materials

Experimental results

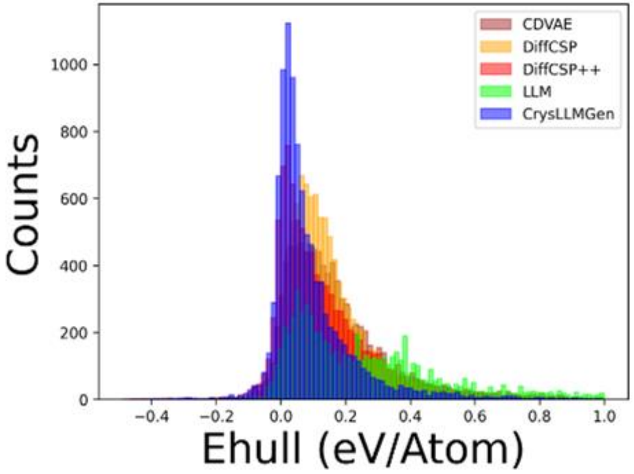
Balanced validity metrics

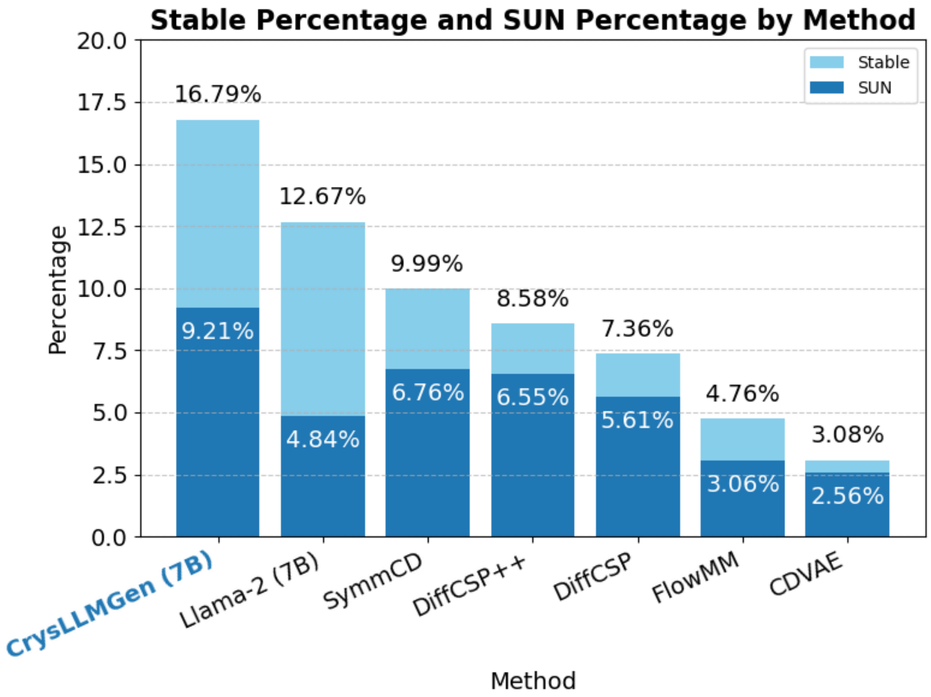
Category	Model	Validity ↑		Coverage ↑	
		Struct	Comp	Prec	Rec
Diffusion	CDVAE	100	86.70	99.49	99.15
	DiffCSP	100	83.25	99.76	99.71
	DiffCSP++	99.94	85.12	99.59	99.73
	MatterGen	100	86.34	99.45	99.59
	UniMat	97.20	89.40	99.70	99.80
	SymmCD	92.30	87.13	98.78	97.33
Flow Matching	FlowMM	96.85	83.19	99.58	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

Best
2nd best

Performance of different categories of models on unconditional generation task

Stability, Uniqueness and Novelty



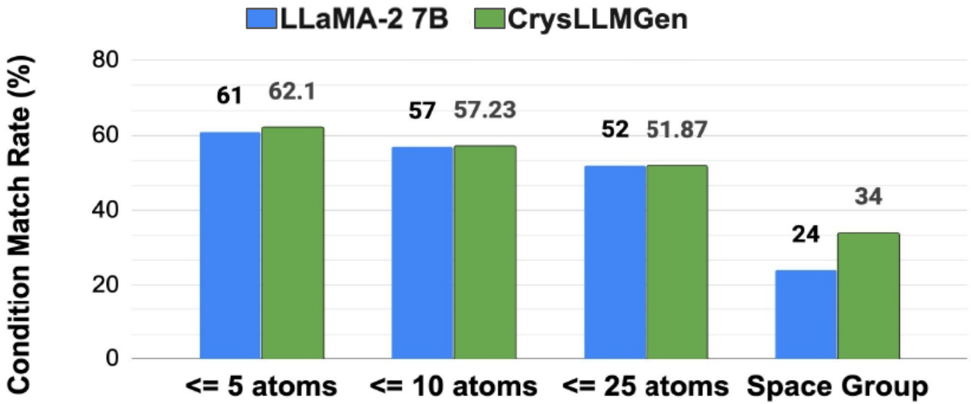


Conditional generation capabilities

Conditional Prompt

Below is a description of a bulk material. The chemical formula is SnO. The space group number is 31. Generate a description of the lengths and angles of the lattice vectors and then the element type and coordinates ....

Conditions Satisfied

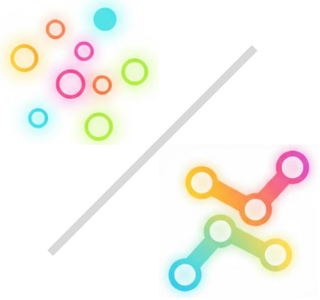


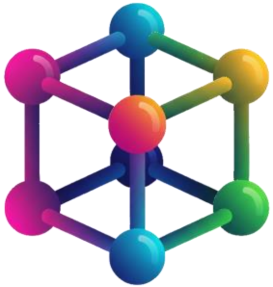
CrysLLMGen matches the criteria in the prompt better than LLM-based approach

Conclusion

Summary of contributions

- First hybrid model combining LLMs and DMs for crystal generation.
- Successfully mitigates the trade-off between compositional and structural validity.
- Generates structures that are significantly more stable, unique, and novel than state-of-the-art models.





Thank you