Learning Superconductivity from Ordered and Disordered Material Structures

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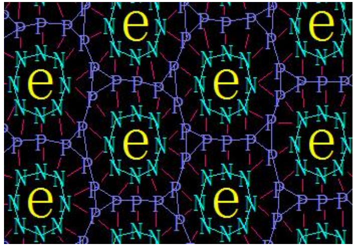
Background

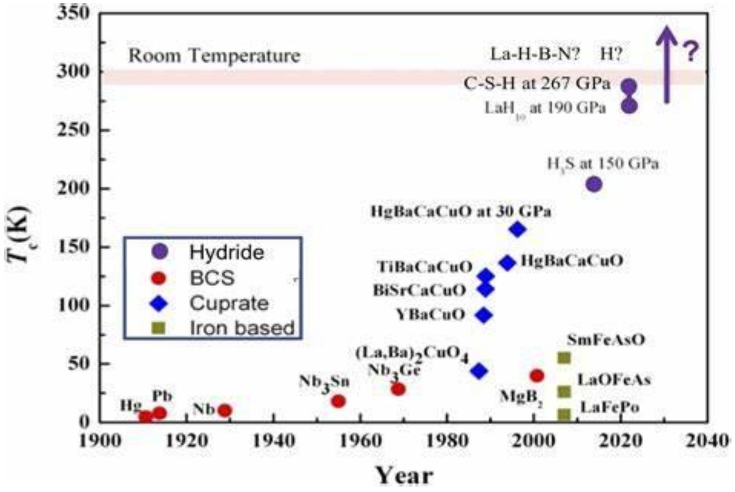
❑ High-temperature Superconductors(HSC)

- Zero resistance, Meissner effect
- Energy transmission, advanced electromagnetics, and quantum computing, etc.



❑ Challenges for Designing HSC

- Theoretical calculation: HSC mechanism unclear/BCS theory is limited.
- Hunt for HCS: "Holy Grail" of physics, a century-old challenge.



The First Room-Temperature Ambient-Pressure Superconductor

Sukbae Lee, Ji-Hoon Kim, Young-Wan Kwon

For the first time in the world, we succeeded in synthesizing the room-temperature superconductor ($T_c \geq 400$ K, 127°C) working at ambient pressure with a modified lead-apatite (LK-99) structure. The superconductivity of LK-99 is proved with the Critical temperature (T_c), Zero-resistivity, Critical current (I_c), Critical magnetic field (H_c), and the Meissner effect. The superconductivity of LK-99 originates from minute structural distortion by a slight volume shrinkage (0.48 %), not by external factors such as temperature and pressure. The shrinkage is caused by Cu^{2+} substitution of Pb^{2+}(2) ions in the insulating network of Pb(2)-phosphate and it generates the stress. It concurrently transfers to Pb(1) of the cylindrical column resulting in distortion of the cylindrical column interface, which creates superconducting quantum wells (SQWs) in the interface. The heat capacity results indicated that the new model is suitable for explaining the superconductivity of LK-99. The unique structure of LK-99 that allows the minute distorted structure to be maintained in the interfaces is the most important factor that LK-99 maintains and exhibits superconductivity at room temperatures and ambient pressure.

We need new method...

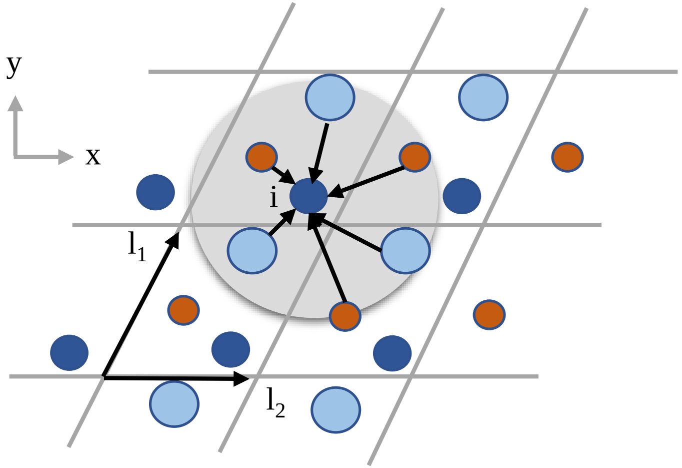
Background

❑ Data Driven Method

- Deep Learning: Bypass complex physical theories
- GNN extensively applied to model materials
    - Properties prediction
    - 3D structures generation

❑ Inverse Materials Design

- Given target properties to generate 3D structures
    - CDVAE/DiffCSP/SyMat



GNN: Represent atom/bond as node/edge

We need data to train models...

Related Dataset

❑ SuperCon

- 33,000, only chemical formulas

❑ Jarvis-DFT

- 1058, DFT calculated with BSC theory

❑ S2S

- 1,065, label materials with Superconductivity (Yes or No)

❑ 3DSC

- 9,150, elemental matching and manual doping (some not experimental observation)

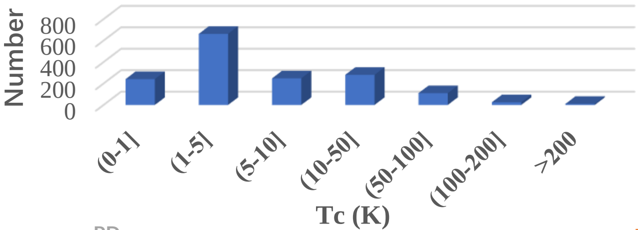
SuperCon3D Dataset

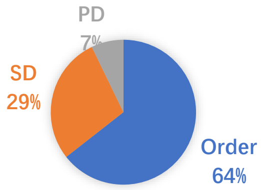
❑ Collection Methods

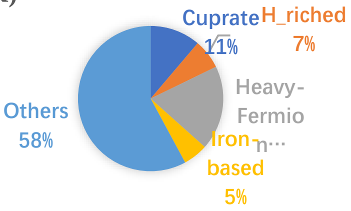
- Formula matching between SuperCon and ICSD
- Manually collection from references

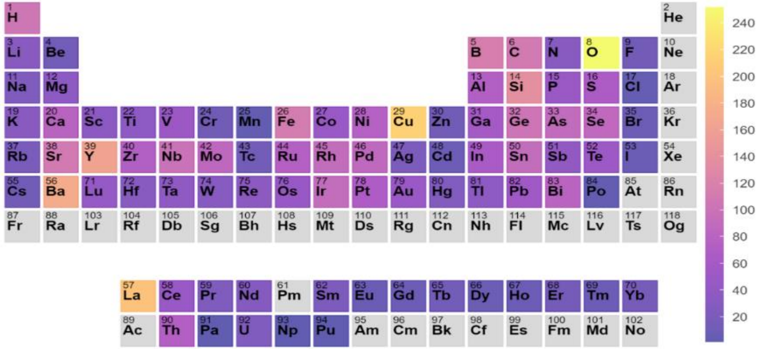
❑ Data Distribution

- Cover 83 elements in periodic table
- Contain ordered and disorder structures
- Five Types:
  - ✓ Cuprate, H-riched, Heavy fermion, Iron-based, others
- T_c values range from (0, 290] K









How to use SuperCon3D?

❑ Real-world Superconducting Materials

➢ Imperfection or disorder for tuning T_c.

❑ Common disordered structures

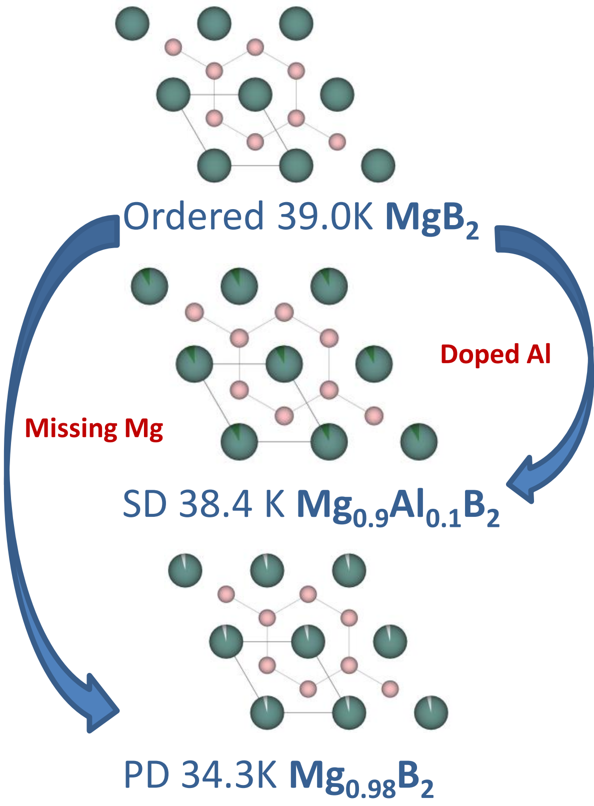
➢ **Substitutional Disorder** (SD): a site is occupied by more than on atomic species.

➢ **Positional Disorder** (PD): one atom in the unit cell occurs position shift.

➢ **SD + PD** (SPD): both SD and PD can occur simultaneously.

➢ **Interstitial Disorder (ID)**: atoms occupying interstitial sites outside regular lattice positions in a crystal, unseen in SuperCon3D dataset.

➢ **Random**: unseen in SuperCon3D dataset.



Graph Representation: Order → Disorder

❑ Introduce atomic occupancy to redefine material structure

SD

$$\begin{cases} m_i > 1, \\ a_{i,1} \neq a_{i,2} \neq \cdots \neq a_{i,m_i}, \\ w_{i,1} + w_{i,2} + \cdots + w_{i,m_i} = 1 \end{cases}$$

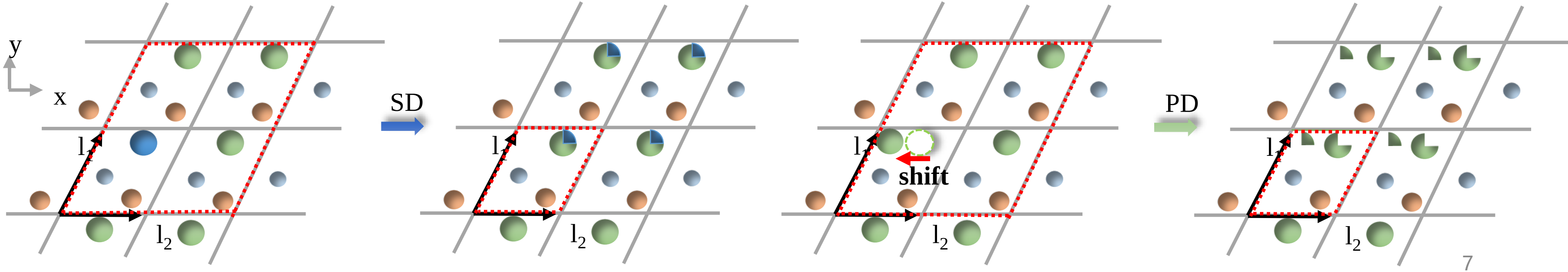
PD

$$\begin{cases} m_i = 1, \\ w_{i,1} < 1. \end{cases}$$

ID

$$w_{i,1} + w_{i,2} + \cdots + w_{i,m_i} + w_{i,\text{interstitial}} = 1 + \Delta$$

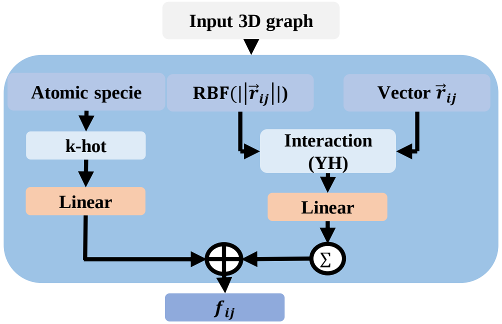
- Unit cell: $\mathcal{M} = (L, S)$
- Lattice: $L = [l_1, l_2, l_3] \in \mathbb{R}^{3\times 3}$
- Site $S_i = (A_i, \mathbf{w}_i, \mathbf{x}_i)$
- Conposition $A_i = [a_{i,1}, \ldots, a_{i,m_i}] \in \mathbb{R}^{m_i \times h}$
- Atomic occupancy: $\mathbf{w}_i \in \mathbb{R}^{m_i}$
- Cartesian coordinate: $\mathbf{x}_i \in \mathbb{R}^3$

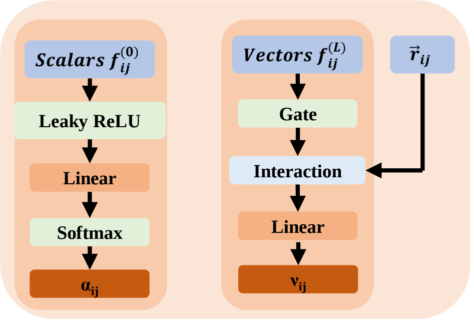


SODNet: Structures -> T_c

❑ SODNet:

- ➢ Transformer-based GNN framework for representing ordered and disordered graphs.
- ➢ SE(3)-equivariance through irreducible representation-based vector space features





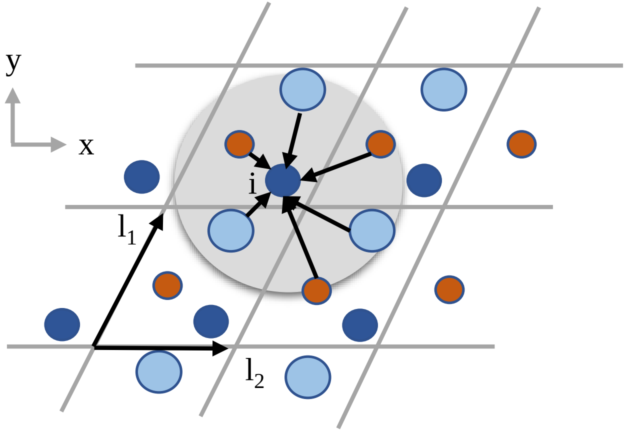
❑ Ordered and Disorder Graph Representation

- ➢ Node embedding:
- ➢ Edge embedding:

$$h_i = \begin{cases} a_{i,1}, & S_i \text{ is ordered,} \\ \sum_k w_{i,k} a_{i,k}, & S_i \text{ is SD or SPD,} \\ w_{i,1} a_{i,1}, & S_i \text{ is PD.} \end{cases}$$

$$\|\vec{r}_{ij}\| > R_i + R_j \qquad E = w_i w_j RBF(\|\vec{r}_{ij}\|),$$

$$x_{ij} = \varphi(h_i) + \varphi(h_j), \qquad f_{ij} = \varphi_f(x_{ij} \otimes c_E^{TP} SH(\vec{r}_{ij}))$$

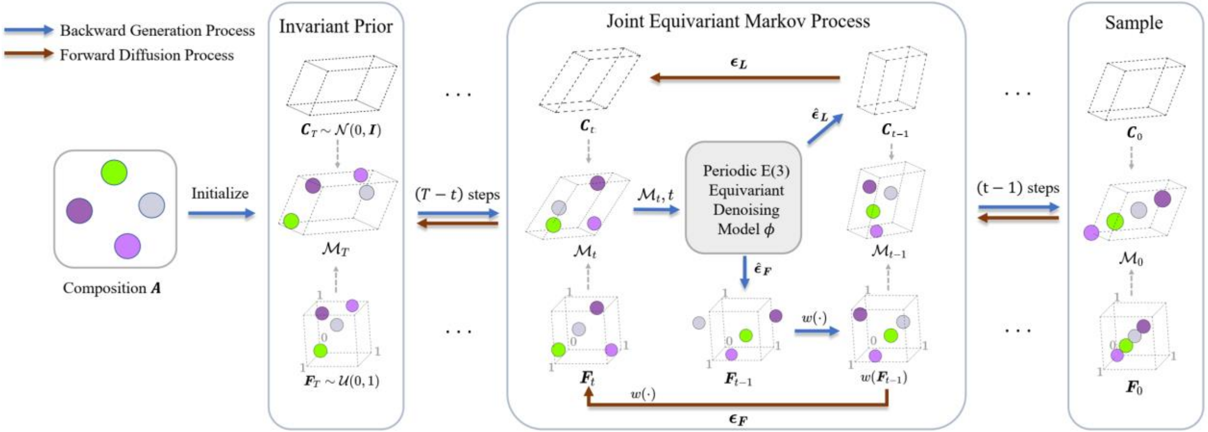


KNN Edges

DiffCSP-SC: Targeting T_c -> Structures

❑ DiffCSP-SC: Equivariant diffusion for superconducting crystal structure generation

- Transformer-based architecture
- Diffusion on $\boldsymbol{C}$
  - ✓ Gaussian Prior
  - ✓ DDPM-based Markov Chain
- Diffusion on $\boldsymbol{F}$
  - ✓ Uniform Prior
  - ✓ Score Matching + Wrapped Normal Distribution



Previous work

DiffCSP-SC: Superconductors Generation

❑ DiffCSP-SC: Equivariant diffusion for superconducting crystal structure generation

➢ Transformer-based architecture

Periodic E(3) Equivariant Denoising Model ϕ

➢ Input Feature

$$h_i^{(0)} = \rho(f_{\text{atom}}(\boldsymbol{a}_i), f_{\text{pos}}(t))$$

➢ Message-Passing Blocks

$$\boldsymbol{m}_{ij}^{(s)} = \varphi_m(\boldsymbol{h}_i^{(s-1)}, \boldsymbol{h}_j^{(s-1)}, \boldsymbol{L}^\top\boldsymbol{L}, \psi_{\text{FT}}(\boldsymbol{f}_j - \boldsymbol{f}_i)),$$

$$\boldsymbol{m}_i^{(s)} = \sum_{j=1}^{N} \boldsymbol{m}_{ij}^{(s)},$$

$$\boxed{\boldsymbol{h}_i^{(s)} = \boldsymbol{h}_i^{(s-1)} + \varphi_h(\boldsymbol{h}_i^{(s-1)}, \boldsymbol{m}_i^{(s)}).}$$

$$h_i^{(s)} = h_i^{(s-1)} + \sum_{j=1}^{N} \theta_{ij}^{(s)} v_{ij}^{(s)}$$

$$\theta_{ij}^{(s)} = Softmax\left(\frac{\mathbf{q}_i^{(s)\top}\mathbf{k}_{ij}^{(s)}}{\sqrt{d}}\right)$$

$$\mathbf{q}_i^{(s)} = \varphi_q\left(h_i^{(s-1)}\right),$$

$$\mathbf{k}_{ij}^{(s)} = \varphi_k\left(h_i^{(s-1)}, L^\top L, \psi_{\text{FFT}}(f_j - f_i)\right),$$

$$\mathbf{v}_{ij}^{(s)} = \varphi_v\left(h_i^{(s-1)}, L^\top L, \psi_{\text{FFT}}(f_j - f_i)\right)$$

➢ Lattice Denoising Term

$$\hat{\boldsymbol{\epsilon}}_{\boldsymbol{L}} = \boldsymbol{L}\varphi_L\left(\frac{1}{N}\sum_{i=1}^{N}\boldsymbol{h}_i^{(S)}\right)$$

$$\hat{\boldsymbol{\epsilon}}_{\boldsymbol{F}}[:, i] = \varphi_F(\boldsymbol{h}_i^{(S)})$$

Experiment Results

❑ SODNet

Method	Data: Train	Data: Test	Performance: MAE (logK)↓	Performance: R^2 ↑
RF-c	O	O	0.738±0.165	0.711±0.050
SVM-c	O	O	0.632±0.094	0.801±0.041
RF-geo	O	O	0.741±0.115	0.759±0.051
SVM-geo	O	O	0.578±0.114	0.827±0.042
SchNet	O	O	0.891±0.041	0.401±0.032
CGCNN	O	O	0.879±0.047	0.405±0.022
DimeNet++	O	O	0.811±0.058	0.434±0.092
SphereNet	O	O	0.762±0.048	0.467±0.096
ALIGNN	O	O	0.755±0.049	0.479±0.090
Matformer	O	O	0.748±0.043	0.570±0.135
MEGNet	O	O	0.794±0.006	0.497±0.009
	O/SD	O/SD	0.889±0.049	0.431±0.058
SODNet	O	O	0.622±0.112	0.595±0.101
	O/SD/PD/SPD	O	**0.584±0.119**	**0.634±0.117**
	O/SD	O/SD	**0.518±0.084**	**0.716±0.064**
	O/SD/PD/SPD	O/SD/PD/SPD	**0.505±0.055**	**0.748±0.032**

Model Performance

Method	Performance: MAE (logK)↓	Performance: R^2↑
w/o Occupancy Embedding		
w/o disorder node embedding	0.990±0.033	0.365±0.044
w/o disorder edge embedding	0.592±0.087	0.655±0.046
w/o O(3) Equivariance		
w/o equivariant operations	0.611±0.046	0.618±0.027
SODNet	**0.505±0.055**	**0.748±0.032**

Ablation Studies

Experiment Results

❑ DiffCSP-SC

Model	Data	Performance SR10	SR30	SR50
CDVAE	O	0.03	0.03	0.03
SyMat	O	0.03	0.04	0.04
DiffCSP	O	0.04	0.05	0.05
DiffCSP-SC	O	0.05	0.05	0.10
CDVAE	Pre-training + O	0.25	0.25	0.30
SyMat	Pre-training + O	0.28	0.28	0.35
DiffCSP	Pre-training + O	0.30	0.30	0.45
DiffCSP-SC	Pre-training + O	**0.37**	**0.37**	**0.50**

Model Performance

Method	Performance SR10	SR30	SR50
w/o Transformer			
w/o attention	0.28	0.28	0.45
w/o Pre-training			
w/o pre-training	0.05	0.05	0.10
DiffCSP-SC	**0.37**	**0.37**	**0.50**

Ablation Studies

Pretrain on **1.1 million** stable material structures

Experiment Results

❑ Real-world Superconductors Validation

Material	O/SD/PD	T_c^{exp} (K)	T_c^{pred} (K)	Relative Error(%)
CaH_6	O	215 [35]	242.25	12.67
Ti	O	26 [66]	8.50	67.31
CsV_3Sb_5	O	2.3 [18]	2.36	6
$Cs(V_{0.93}Nb_{0.07})_3Sb_5$	SD	4.45 [29]	4.71	5.84
Zr_4Rh_2O	O	3.73 [58]	4.12	10.45
Zr4Pd2O	O	2.73 [58]	2.82	3.3
$LaFeSiO_{0.9}$	PD	10 [23]	7.93	20.7

Outlier: Extreme pressure (248 GPa)

Experiment Results

❑ Application: Screening Known Structures

Type	ICSD code	Chemical formula	O/SD/PD	T_c (K)	Reported SC.
Cuprate	68675	$CuO_2Sr_{0.075}$	PD	93.42	CuO_2Sr (91K)
	50774	$Ca_{0.779}CuO_2Y_{0.041}$	PD	65.70	-
	50773	$Ca_{0.82}CuO_2$	PD	64.72	$CaCuO_2$ (89K)
	68217	Ba_2CuO_3	O	59.89	$Ba_2CuO_{3.2}$ (70K)
H-riched	187375	ErH_3	O	193.03	-
	635802	GdH_3	O	143.19	-
	623739	$H_{2.57}Co_{0.14}U_{0.84}$	PD	136.76	-
	42009	$TbH_{2.25}$	SD	135.13	-
Heavy-Fermion	168466	$LaMg_{12}$	O	23.83	-
	161141	$LaMg_{11.196}Al_{0.804}$	SD	21.13	-
	69897	$C_2Ce_{0.75}U_{0.25}$	PD	11.88	-
	647197	$Np_{1.1}Pu_{0.9}$	SD	11.75	-
Iron-based	427163	$Ba_{0.83}Fe_2Rb_{0.17}As_2$	SD	23.21	$Ba_{0.6}Fe_2Rb_{0.4}As_2$ (37.5k)
	188347	$BaFe_2As_2$	O	23.27	-
	39530	$FeCl_7Te$	O	19.57	-
	633401	$FeSb_{0.4}Te_{1.6}$	SD	16.83	-
Others	96031	$Ba_{1.1432}Co_{0.1429}$-$O_{3.0009}Rh_{0.8574}$	PD	202.12	-
	58639	$Ba_{0.515}Ca_{0.485}$	SD	160.95	-
	616160	$BaSr$	SD	123.51	-
	106111	$SrTl_2$	O	63.52	-

Screening entire ICSD, selecting 20 entries

parent compounds exhibit superconductivity[1][2]

Disordered compound shows superconductivity[3]

Disordered compound presents superconductivity[4]

[1]. *Physica C: Superconductivity*, 227(3-4): 395–398, 1994

[2]. Nature, 414(6862):434–436, 2001.

[3]. PNAS, 116(25):12156–12160, 2019.

[4]. Z Anorg Allg Chem, 640(5): 830–835, 2014.

Experiment Results

❑ Application: Generating Novel Structures

Type	Index	Chemical formula	T_c (K)	Reported SC.
Cuprate	1	$Ba_2CuCl_2O_2$	33.56	-
	2	$Tl_2Ca_2Ba_2Cu_3O_{10}$	14.09	-
	3	$Ba_3CaLa_2GdCu_7O_{17}$	10.12	-
	4	YCu_3O_7	9.73	-
H-riched	5	TbH_3	164.33	TbH_3(20K)
	6	SeH_3	139.89	SeH_3(113K)
	7	$CaGe_2H_9$	103.55	-
	8	Ca_2MnCrH_6	58.07	-
Heavy-Fermion	9	Ba_3Pu	44.81	-
	10	Th	43.61	-
	11	ThC_3	17.96	-
	12	Lu	4.86	-
Iron-based	13	$BaFe_2Se_2$	11.99	-
	14	SmFeAsO	4.42	$SmFeAsO_{0.8}F0.2$ (54K)
	15	KFe_2As_2	4.23	KFe_2As_2 (20K)
	16	NdFeAsF	4.13	-
Others	17	Ba_3Ca	80.04	-
	18	Ba_2Se	60.70	-
	19	Ba	52.26	-
	20	Mg_3B	43.96	-

Calculated by DFT[1]

Predicted by ML[2]

Disorders display superconductivity[3] [4]

[1]. JPCCC, *125(6):3640–3649, 2021.*

[2]. arXiv preprint arXiv:2301.10474, 2023

[3]. IEEE T APPL SUPERCON, 2023.

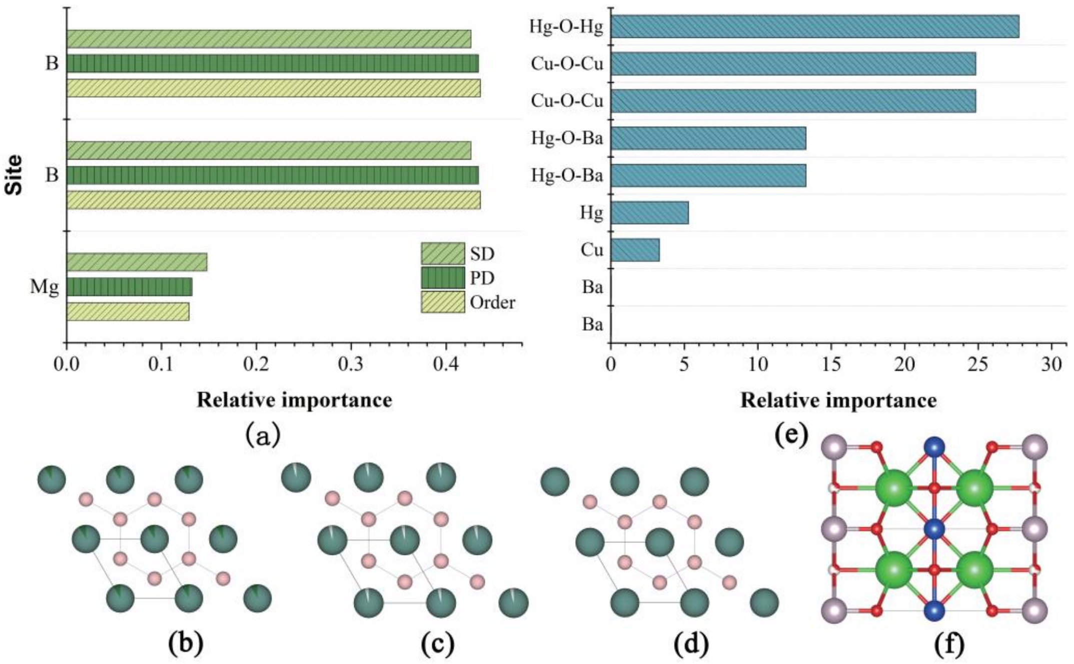
[4]. J SUPERCOND NOV MAGN, 33(8):2347–2354, 2020.

Generating Novel Structures (selecting 20 entries)

Why Transformer?

❑ Relationship Between Structures and T_c

- Characteristics of superconductors: large number of atoms and diverse elements.
- Identify key atomic contributions to Tc.



Shows potential for atomic-level superconductor design.

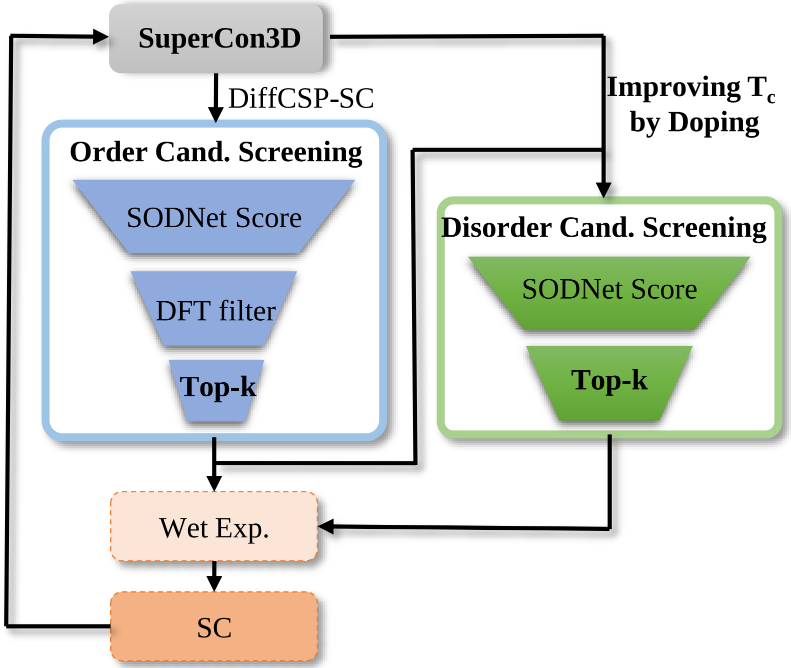
Limitations & Solutions

❑ Limitations

- ➢ Data unevenness
  - ✓ Scarce High Tc data, uneven across 5 material types.
- ➢ Elemental skewness
  - ✓ Especially in Cu and O

❑ Solutions

- ➢ More high-quality data
- ➢ Proposed pipeline
  - ✓ SuperCon3D + DiffCSP-SC + DFT + SODNet + Wet Exp.



Conclusion

- ❑ **A new dataset SuperCon3D** containing both ordered-and-disordered crystal structures and experimental Tc

- ❑ We propose **two deep learning models** to showcase the possible methods for exploring
  - ➢ SODNet: Tc predictor
  - ➢ DiffCSP-SC: Crystal Structures generator targeting high Tc

- ❑ Based on our proposed models, we present **a list of candidate superconductors** for future experimental validation
  - ➢ First report of candidate disordered superconductors using GNN methods.

Thanks

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