WYCKOFF TRANSFORMER: GENERATION OF SYMMETRIC CRYSTALS

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ABSTRACT

We propose Wyckoff Transformer, a generative model for materials conditioned on space group symmetry. Most real–world inorganic materials have internal symmetry beyond lattice translation. Symmetry rules that atoms obey play a fundamental role in determining the physical, chemical, and electronic properties of crystals. These symmetries form energy configurations, determine stability, and influence key material structural and functional properties such as electrical and thermal conductivity, optical and polarization behavior, and mechanical strength. And yet, despite the recent advancements, state-of-the-art diffusion models struggle to generate highly symmetric crystals. We use Wyckoff positions as the basis for an elegant, compressed, and discrete structure representation. To model the distribution we develop a permutation–invariant autoregressive model based on Transformer and absence of positional encoding. Our experiments demonstrate that Wyckoff Transformer has the best performance in generating novel diverse stable structures conditioned on the symmetry space group, while also having competitive metric values when compared to model not conditioned on symmetry. We also show that it is able to predict formation energy and band gap within DFT accuracy.

1 INTRODUCTION

Space of all possible combinations of atoms forming periodic structures is intractably large. It is not possible to screen it fully, even with a fast machine learning algorithm. Practical materials, however, occupy only a small part of it. Firstly, they must correspond to an energy minimum. Secondly, occupying an energy minimum is not sufficient to establish if the material is synthesizable or indeed experimentally stable. Having a generative model that outputs a priori stable materials is a step towards speeding up automated material design by orders of magnitude.

1.1 SPACE GROUPS AND WYCKOFF POSITIONS

A crystal structure can be systematically described through its lattice and atomic basis. The lattice provides a repeating geometric framework, defined as an infinite periodic arrangement of points in space. Based on interactions between the constituent electrons and nuclei, atoms rearrange into such a lattice and, therefore, follow a finite set of symmetries: the group of all such symmetry operations that uniquely define the periodic arrangement is called the space group of the crystal. These arrangements in a crystal are governed by a finite set of symmetry operations, such as rotations, reflections, inversions, and translations. These operations combine to form the 230 distinct space groups, which serve as a comprehensive classification system for all possible crystal symmetries in three dimensions. Each space group defines the unique symmetry properties of a crystal structure, defining the allowable positions for atoms within the unit cell. This ensures that every crystal possesses at least the simplest level of symmetry, referred to as P1 symmetry, which involves only translational symmetry. The atomic basis specifies the arrangement of atoms associated with each lattice point, thus defining the overall crystal structure.

Within a given space group, a subgroup forms the site symmetry, referring to the set of symmetry operations that leave a specific point in the crystal invariant. These operations describe the local symmetrical environment, such as mirror, screw axis, or inversions centered on that a given region.

Higher site symmetry is in regions where multiple symmetry elements intersect, while those with lower site symmetry include only one symmetry operation. Taking space group 225 Fm-3m as an example, site symmetry subgroup m-3m represents a highly symmetric environment like the center of a cubic unit cell, where multiple symmetry elements intersect, including mirror planes and a 3–fold rotoinversion axis. In contrast, another lower site symmetry subgroup .3m corresponds to a less symmetric environment with only a threefold rotation axis and a mirror plane.

These site symmetry points, classified by their symmetry properties, are grouped into Wyckoff positions (WPs) (Wyckoff, 1922). Mathematically, a WP encompasses all points whose site symmetry groups are conjugate subgroups of the full space group Kantorovich (2004). Each WP is characterized by two key attributes:

1. Site symmetry
2. Symmetry equivalence: two different Wyckoff positions in the same space group can share the same site symmetry but may still be symmetry equivalent. This equivalence arises when the Wyckoff positions can be mapped onto each other using higher–order symmetry operations, such as those defined by the Euclidean normalizer of the space group. These symmetry–equivalent WPs form the basis for enumeration and augmentation in the subsequent sections of this work.

WPs for a given space group are commonly enumerated by Latin letters in the order of multiplicity, the number of equivalent atomic positions in a crystal structure that are related by the symmetry operations of the space group. WPS are denoted by a combination of the multiplicity value and the letter, e. g. `2a`. The number of distinct WPs in a space group is finite, ranging from a single WP in the simplest symmetry group P1 to as many as 27 in the most complex space groups. These classifications enable the description of not only discrete points but also more complex geometric features. For example, some Wyckoff positions represent 1D lines, 2D planes, or even open 3D regions within the unit cell, depending on the symmetry constraints. This flexibility underscores the utility of Wyckoff positions in describing diverse crystallographic arrangements. By introducing these fundamental concepts – lattice, atomic basis, space groups, site symmetry, and Wyckoff positions – this framework provides a foundation for understanding crystal structures.

Our work relies on a crucial insight: most ($\approx$ 98%) known crystals have symmetry beyond the lattice translation, see figure 1. Those symmetries are not merely a mathematical observation; optical, electrical, magnetic, structural and other properties are determined by symmetry, as shown by Malgrange et al. (2014); Yang et al. (2005), as well as our results in section 3.4.

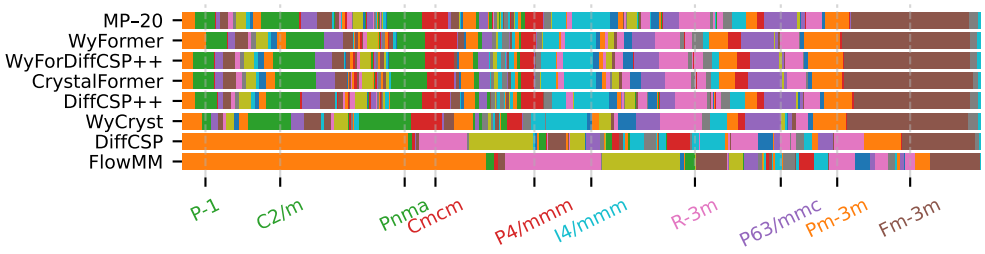

Figure 1: Distribution of space groups in MP–20 dataset Xie et al. (2021) and the generated samples. 10 space groups most frequent in MP–20 are labeled, 98% of MP–20 structures belong to symmetry groups other that P1, with internal symmetries. Plot design by Levy et al. (2024)

1.2 Our contribution

Our contribution can be summarized as follows:

1. Representing a crystal as an unordered set of tokens fused from the chemical element and Wyckoff position; section 2.1.
2. Encoding Wyckoff positions using their universally-defined symmetry point groups; section 2.1.

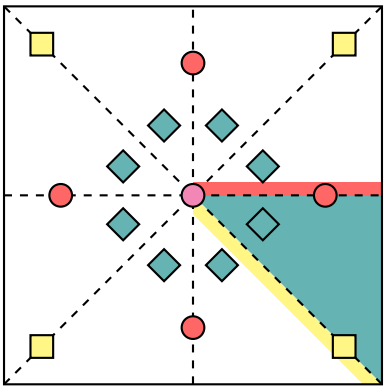

Figure 2: Wyckoff positions illustration of a toy 2D crystal Goodall et al. (2020). It contains 4 mirror lines, and one rotation center. There are four Wyckoff positions, illustrated by shading. Magenta is the Wyckoff position that is invariant under all the transformations, it only contains a single point; red and yellow lie on the mirror lines, and teal is only invariant under identity transformation and occupies the rest of the space. Markers of the corresponding colors show one of the possible locations of an atom belonging to the corresponding Wyckoff position.

3. Wyckoff Transformer architecture and training protocol that combine autoregressive probability factorization with permutation invariance; section 2.3.
4. Model invariance with the respect to the arbitrary choice of the coset representative of the space group affine normalizer; sections 2.1, 2.3.
5. Empirically, our model outperforms past methods in generating novel diverse materials conditioned on space group symmetry; section 3.4.
6. Despite not using the information about atom coordinates, our model achieves performance in band gap prediction competitive with the machine learning models that use it; Wyckoff Transformer is also able to predict formation energy prediction within the level of DFT accuracy; section 3.4.

1.3 Related work

Crystal generation is a burgeoning field, with a plethora of models operating in the 3D point cloud space (Jiao et al., 2024a;b; Cao et al., 2024; Yang et al., 2023; Zeni et al., 2024; Xie et al., 2021). Our approach complements them naturally by providing discrete symmetry constraints and/or initial structure approximation.

Application of Wyckoff positions in machine learning. The concept of Wyckoff positions in crystalline materials emerged from Ralph W. G. Wyckoff's work on crystallography, originally published more than a 100 years ago (Wyckoff, 1922), which laid the groundwork for understanding equivalent positions in space groups, serving as a precursor to the International Tables for Crystallography. Given their elegant representation, naturally, in modern times WPs have found their way into material informatics and machine learning, first as feature engineering approach for property prediction (Goodall et al., 2020; Jain & Bligaard, 2018; Möller et al., 2018; Goodall et al., 2022), and recently for generative models. The main limiting factor was the ability of machine learning algorithms to handle discrete structured data which is formed by WPs. Our work is inspired by Zhu et al. (2024), the first generative model to utilize Wyckoff positions. It uses a VAE over one–hot–encoded information about WPs. A concurrent work by Cao et al. (2024) independently explores an approach similar to ours. The main differences between our and all other approaches is that they use representations based on Wyckoff letters. Wyckoff letter definitions depends on the space group, unlike site symmetry, leading to data fragmentation. Zhu et al. (2024); Cao et al. (2024) also don't take into account dependency of the Wyckoff letters on the arbitrary choice of the coset representative of the space group Euclidean normalizer. Finally, Cao et al. (2024) use positional encoding to establish the relationship between the chemical elements and Wyckoff positions they occupy, while we combine them in one token.

2 WYCKOFF TRANSFORMER (WYFORMER)

2.1 TOKENIZATION

A crystal can be represented as a space group, a set of WPs and chemical elements occupying them, the fractional coordinates of the WP degrees of freedom, and free lattice parameters. Such representation reduces the number of parameters by an order of magnitude without information loss. For example, see figure 3.

```
Group: I4/mmm (139)
Lattice: a = b = 8.9013,  c = 5.1991,  α = 90.0,  β = 90.0,  γ = 90.0
Wyckoff sites:
Nd @ [ 0.0000  0.0000  0.0000], WP [2a] Site [4/m2/m2/m]
Al @ [ 0.2788  0.5000  0.0000], WP [8j] Site [mm2.]
Al @ [ 0.6511  0.0000  0.0000], WP [8i] Site [mm2.]
Cu @ [ 0.2500  0.2500  0.2500], WP [8f] Site [..2/m]
```

Figure 3: Wyckoff representation of $Nd(Al_2Cu)_4$ (mp-974729), variable parameters in **bold**. If represented as a point cloud, the structure has $13[\text{atoms}] \times 3[\text{coordinates}] + 6[\text{lattice}] = 42$ parameters; if represented using WPs, it has just 4 continuous parameters (WPs `8i` and `8j` each have a free parameter, and the tetragonal lattice has two), and 5 discrete parameters (space group number, and WPs for each atom).

Of work is based on the observation that for stable materials space group symmetry and Wyckoff sites almost completely define the structure – 98.3% of the structures in MP–20 dataset have unique Wyckoff representations. Therefore our model only generates the discrete part; one could argue that the symmetry captured by this discrete part is sufficient to determine a desired property of a material (for example piezoelectricity via non-centrosymmetry, direct/indirect band gap via positions of the valence/conduction bands in the Brillouin Zone, etc.), while the fractional coordinates can be linked to the magnitude of that property. Given a Wyckoff representation that reflects the lattice symmetry, coordinates can be determined as discussed in section 2.4.

We represent each structure as a set of tokens, as shown in figure 4. The first token contains the space group, the others chemical elements and WPs. We encode a WP as a tuple containing site symmetry and so-called *enumeration*. Several WPs can correspond to the isomorphic site symmetries, for example both yellow and red in figure 2 correspond to reflection, but with different axes. To differentiate those WPs we enumerate them separately within each space group and site symmetry according to the conventional WP order Aroyo et al. (2006). For example, in space group 225 present in figure 4 WP `4a` is encoded as `(m-3m, 0)`, `4b` as `(m-3m, 1)`, and `8c` as `(-43m, 0)`. The purpose of this encoding is to take advantage of the fact that, unlike Wyckoff letters, site symmetry definition is universal across different space groups.

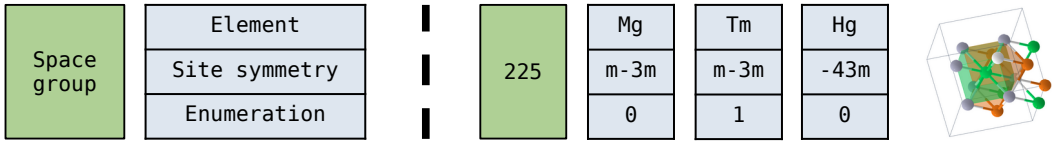

Figure 4: An example of structure tokenization, $TmMgHg_2$ mp-865981

The two-part encoding has another advantage. For some crystals *enumerations* part, and only this part, of Wyckoff representation is not uniquely defined, as it depends on the arbitrary choice of the coset representative of the space group Euclidean normalizer. See the example in figure 5.

2.2 MODEL ARCHITECTURE

Token embeddings are constructed by concatenating the embeddings for every part of the token (element, site symmetry, enumeration). To each structure we add a `STOP` token. We use those

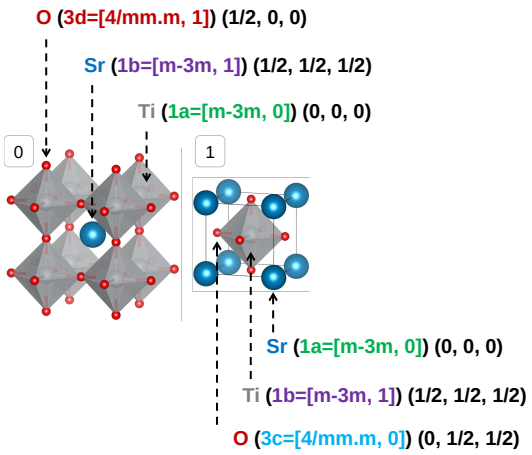

Figure 5: Two possible equivalent Wyckoff representations of $SrTiO_3$, depending on the lattice center choice:
[(Ti, (m−3m, 0)), (Sr, (m−3m, 1)), (O, (4/mm.m, 1))]
[(Ti, (m−3m, 1)), (Sr, (m−3m, 0)), (O, (4/mm.m, 0))]

sequences as an input for an encoder-only Transformer Vaswani (2017). Wyckoff representation is permutation–invariant, so is Transformer; we don't use positional encoding.

De novo generation To represent states where some parts of token are known and others are not, we replace those values with `MASK`. We take the output of Transformer on the token containing `MASK` value(s), concatenate it with a one-hot vector encoding presence of each token in the input sequence, and use the result as the input for three fully-connected neural networks, one for each predicted part of token.

Property prediction We take the average of the Transformer outputs and use it as input for a fully-connected neural network that outputs a scalar predicted value.

2.3 Training

We use data augmentation to encourage invariance with the respect to the choice of the coset representative of the space group affine normalizer, by picking a randomly selected equivalent representation at every training epoch. It is made possible by the low number of variants; in MP–20 (Xie et al., 2021) dataset for 96% structures there are less than 10.

De novo generation We use cross–entropy loss. We predict the next part of token in cascade: first the chemical element, then, conditioned on it, site symmetry and, finally, enumeration.

Unlike Transformer itself, auto–regressive generation is not permutation–invariant. Fortunately for us, the number of WPs is small, the average in MP-20 is just 3.0; this allows us to achieve permutation invariant generation with just training augmentation. We shuffle the order of every Wyckoff representation at every training epoch. We use multi–class loss when training to predict the fist cascade part, chemical element, further reducing learning complexity. Model is trained for 9×10^5 epochs; due to the efficiency of the representation, entire dataset fits into GPU memory. We use the loss on the validation dataset for early stopping, learning rate scheduling, and manual hyperparameter tuning.

Property prediction Training as a regular regression model using MSE loss over 150k epochs. Accordingly, the model uses the entire sequence of tokens, i.e. all atom types and WPs.

2.4 STRUCTURE GENERATION

We generate crystals conditioned on space group number. It is sampled from the combination of training and validation datasets. Wyckoff representation is then autoregressively sampled using the Wyckoff Transformer. We use two ways to generate the final crystal structure conditioned on the representation, the details are described in appendix section A. They both start with sampling a structure conditioned on the Wyckoff representation with pyXtal (Fredericks et al., 2021), and then relaxing it with CHGNet (Deng et al., 2023) and CrySPR (Nong et al., 2024) or DiffCSP++ (Jiao et al., 2024b).

3 EXPERIMENTAL EVALUATION

Dataset We use MP-20 Xie et al. (2021), which contains almost all experimentally stable materials in Materials Project Jain et al. (2013) with a maximum of 20 atoms per unit cell, within 0.08 eV/atom of the convex hull, and formation energy smaller than 2 eV/atom, 45229 structures in total.

3.1 METRICS

Structure property similarity metrics Coverage and Property EMD (Wasserstein) distance, have been proposed as a low–cost proxy metric for de novo structure generation by Xie et al. (2021) and then followed by most of the subsequent work.

Validity Xie et al. (2021) proposed verifying crystal feasibility according to two criteria:

> **Structural** validity means that no two atoms are closer than 0.5Å. All structures in MP–20 and almost all structures produced by state–of–the–art model fulfill it.
>
> **Compositional** validity means having neutral charge (Davies et al., 2019). Only 90% of MP–20 structures pass this test meaning that nonconforming structures are physically possible if somewhat rare.

Novelty and uniqueness The purpose of de novo generation is to obtain new materials. Generating materials that already exist in the training dataset increases the model performance according to structure stability and similarity metrics, but such structures are useless for material design and just increase the gap between the proxy metrics and the model fitness for its purpose. Therefore we exclude generated materials that are not novel and unique from metric computation. On a deeper level, generative models for materials are subject to exploration/exploitation trade–off: the more physically similar are the sampled materials to the training dataset, the more likely they are stable and distributed similar to the data, but the less useful they are for the purpose of material design. From a purely machine learning point of view, novelty percentage serves a proxy metric for overfitting.

Stability is estimated by computing energy above convex hull, and comparing it to a threshold $E_{\text{hull}} < 0.08$ eV, same as used during construction of MP–20 dataset. Then we compute S.U.N. Zeni et al. (2024) – the fraction of stable unique novel structures.

Due to DFT computational costs, we use CHGNet (Deng et al., 2023) for stability estimation of the generated structures, and then compute DFT for a manageable sample from the novel structures generated by the strongest models. Materials Project (Jain et al., 2013) is the source of the structures for the hull; we computed CHGNet predicted energies for it to use as references.

Symmetry of the structures has paramount physical importance. Controlling symmetries also leads to control over physical, electronic, and mechanical behavior, which is desirable in property–directed inverse design of materials. For example, in electronic materials, higher symmetry can improve carrier mobility and uniformity in electronic band structure, enhancing performance in applications such as semiconductors or optoelectronics. Furthermore, high–symmetry structures often exhibit isotropic properties, meaning their behaviors are the same in all directions, making them more versatile for industrial use. From a computational perspective, for a fixed set of atoms that constitute a crystal, enforcing symmetries (beyond the basic P1 translation symmetry) allows for computing permutations to search for useful materials while maintaining a focus on practical, synthesizable crystal structures.

This combination of stability, desirable properties, and computational efficiency makes symmetry consideration in crystals especially valuable in generative models for materials discovery. While higher symmetry is more tractable to compute, experimental realization could require external energy inputs (higher temperatures and pressures: think diamond vs graphite); most databases computed with DFT today are only at 0K and hence do not include this degree of freedom. Keeping this in mind, to evaluate the models according to their ability to reproduce symmetry properties we propose four new metrics:

P1 is the percentage of the structures that have symmetry group $P1$. In MP–20 the corresponding number is just 1.7%, and yet more than a third of the structures generated by some state–of–the–art models lack symmetry beyond lattice translation. We argue that presence of symmetry is good proxy value for structure feasibility that is difficult to capture in standard DFT computations, and would require finite–temperature calculations and/or improved methodologies.

Novel Unique Templates (#) is the number of the novel uniquen element-agnostic Wyckoff representations (section 2.1) in the generated sample. Element-agnostic means that we remove the chemical element, while retaining the symmetry information. For example, for the $TmMgHg_2$ in figure 4, it will be as follows:{`[(X, (m-3m, 0)), (X, (m-3m, 1)), (X, (-43m, 0))]`; `[(X, (m-3m, 1)), (X, (m-3m, 0)), (X, (-43m, 0))]`}. The metric provides a lower limit on overfitting and physically meaningful sample novelty: if two materials have different symmetry templates, their physical properties will be different, while inverse is not always true. It serves as an addition to the strict structure novelty, which provides the upper bound. As discussed above, and previously in section 2.1, symmetry by itself affects physical properties of materials. Crystals with higher symmetry tend to have lower energy states due to more uniform atomic arrangements, which contributes to their thermodynamic stability. This makes them more likely to be synthesizable and usable in real–world applications.

Space Group χ^2 is the χ^2 statistic of difference of the frequencies of space groups between the generated and test datasets.

S.S.U.N. is the percentage of the structures that are symmetric (space group not P1), stable, unique and novel.

3.2 METHODOLOGY

Wyckoff Transformer was trained using MP–20 dataset following the original train/test/validation split. We sampled 10^4 Wyckoff representations, then obtained 10^3 structures using pyXtal+CHGNet and DiffCSP++ approaches described in section 3.2.

WyCryst (Zhu et al., 2024) only supports a limited number of unique elements per structure, therefore we trained it on a subsection of MP–20 containing only binary and ternary compounds, 35575 in total. Evaluation of Wyckoff Transformer trained on the same dataset as WyCryst is present in Appendix E. As WyCryst also produces Wyckoff representations, and not structures, the same pyXtal+CHGNet procedure was used to obtain them.

CrystalFormer (Cao et al., 2024) code and weights published by the authors were used by us to produce the sample, conditioned on the space groups sampled from MP–20.

DiffCSP (Jiao et al., 2024a), DiffCSP++ (Jiao et al., 2024b), and FlowMM (Miller et al., 2024) samples were provided by the authors.

Every data sample contained 1000 structures and was relaxed using CHGNet. The generated samples were filtered for uniqueness, more than 99.5% of structures for every method passed the filtering, therefore its impact is minimal and not further discussed.

We computed for DFT for ~ 90 novel structures for WyFormer and the baselines leading according to CHGNet–based metrics; detailed description of the settings is available in Appendix C.

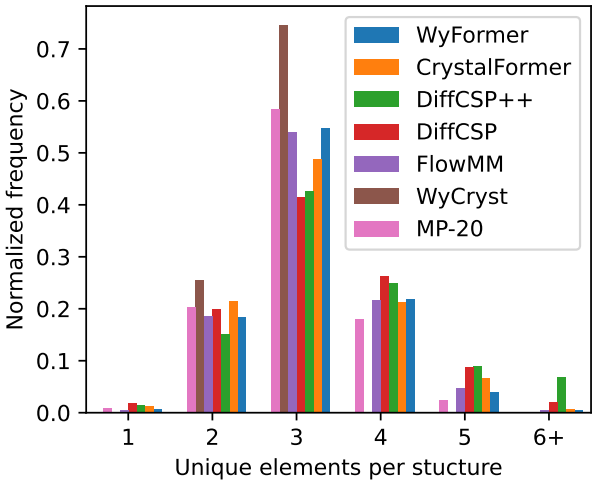

Figure 6: Distribution of the number of unique elements per structure for MP–20 and novel generated structures.

3.3 De novo structure generation results

Metric values are present in table 1. Wyckoff Transformer achieves the best template novelty, fraction of asymmetric structures and space group distribution reproduction. Wyckoff Transformer and DiffCSP have similar S.S.U.N. (T–test $p = 0.8$) and S.U.N. (T–test $p = 0.2$). Given the limited DFT sample size, and DiffCSP's superior S.U.N. computed with CHGNet, we are prepared to believe that on a larger DFT sample it will surpass WyFormer. The correlation of CHGNet–determined stability with DFT–determined is 0.33 – 0.44, meaning that CHGNet is a blunt, but still useful tool for stability estimation.

Proxy metrics are present in table 1c. Every model wins in at least one category, with the second place usually being close. We therefore would like to point out to some of the largest differences. WyCryst and CrystalFormer have significantly lower novelty compared to the other models. While manageable per se, it also means that the models have been overfitted, and their structures are more similar to the training dataset. DiffCSP++ oversamples the structures with the large number of unique elements, WyFormer matches the distribution most closely, as depicted in figure 6.

3.4 Material property prediction results

The results are shown in Table 2. As can be seen from the first column of the table, although our model is inferior in quality to neural networks specifically trained to predict energy, its error is within the difference between DFT calculations and experimental data, i.e. its results for predicting energy are within the DFT error and can be used to assess the properties of the material in practice.

Our model outperforms 4 out of 6 the baselines in predicting the band gap – a much more difficult property to predict.

From this we can conclude that the symmetries and composition of the crystal alone already carry a considerable amount of information about its properties, and while knowledge of the coordinates of the atoms is essential for predicting the energy, the symmetries play a much greater role for predicting the band gap, since the Brillouin zones depend on them. This is evidence supporting the notion that symmetries can determine the properties of a crystal virtually independently of the coordinates.

Table 1: Evaluation of the generated structures, symmetric properties in 1a, stability in 1b, distribution similarity in 1c; structures were relaxed with CHGNet Deng et al. (2023). $E_{\text{hull}} < 0.08$ stability threshold is used, the same as in the training dataset, MP–20. WyForDiffCSP++ refers to Wyckoff Transformer with DiffCSP++ structure generation.

(a) Evaluation of the methods according to the symmetry metrics. Sample size is 1000; the metrics are computed only using novel structurally valid examples.

Method	**Novel Unique** Templates (#) ↑	**P1 (%)** ref $= 1.7$	**Space Group** $\chi^2 \downarrow$
WyFormer	180	3.24	0.223
WyForDiffCSP++	**186**	**1.46**	**0.212**
DiffCSP++	*10*	2.57	0.255
CrystalFormer	74	0.91	0.276
WyCryst	165	4.79	0.710
DiffCSP	76	36.57	7.989
FlowMM	51	44.27	12.423

(b) Stability of the generated structures, as estimated by DFT and CHGNet. Due to limited resources, DFT was only computed for the baselines with the strongest CHGNet S.U.N. and S.S.U.N.; # refers to the number of DFT samples; $\boldsymbol{r}$ is the Pearson correlation between structures' stability determined by DFT and CHGNet. **Bold** indicates the values within $p = 0.1$ statistical significance threshold from the best.

Method	**DFT** ↑			$\boldsymbol{r}$	**CHGNet** ↑	
	#	S.U.N. (%)	S.S.U.N. (%)		S.U.N. (%)	S.S.U.N. (%)
WyFormer	96	7.5	7.5	0.33	39.2	38.2
WyFormerDiffCSP++	95	**14.1**	**14.1**	0.44	36.7	36.0
DiffCSP++	94	8.5	**8.5**	0.32	41.4	**40.8**
CrystalFormer	–	–	–	–	33.9	33.8
WyCryst	–	–	–	–	36.6	35.2
DiffCSP	82	**20.8**	**13.1**	0.36	**57.4**	40.6
FlowMM	–	–	–	–	49.2	29.9

(c) Evaluation of the methods according to validity and property distribution metrics. Following the reasoning in section 3.1, we apply filtering by novelty and structural validity, and do not discard structures based on compositional validity. An evaluation following the protocol proposed by Xie et al. (2021) is available in Appendix D.

Method	**Novelty** (%) ↑	**Validity (%)** ↑		**Coverage (%)** ↑		**Property EMD** ↓		
		Struct.	Comp.	COV-R	COV-P	ρ	E	N_{elem}
WyFormer	90.00	99.56	80.44	98.67	96.72	0.74	0.053	**0.097**
WyForDiffCSP++	89.50	99.66	80.34	99.22	96.79	0.67	0.050	0.098
CrystalFormer	*76.92*	86.84	82.37	**99.87**	95.13	0.52	0.100	0.163
DiffCSP++	89.69	**100.00**	**85.04**	99.33	95.80	**0.15**	0.036	0.504
WyCryst	*52.62*	99.81	75.53	98.85	87.10	0.96	0.113	0.286
DiffCSP	**90.06**	**100.00**	80.94	99.55	96.21	0.82	0.052	0.294
FlowMM	89.44	**100.00**	81.93	99.67	**99.64**	0.49	**0.036**	0.131

Table 2: MAE values for different methods (Xie & Grossman, 2018; Schütt et al., 2017; Chen et al., 2019; Louis et al., 2020; Choudhary & DeCost, 2021; Deng et al., 2023) All models use Materials Project data, but a larger subsample was used by the baseline models compared to our calculations, that used only MP–20. Results by Lin et al. (2023)). Xie & Grossman (2018); Jha et al. (2019) report the error between DFT–computed and experimental results ≈ 0.08 eV for energy, and ≈ 0.6 eV for band gap.

Method	**Energy, eV**	**Band Gap, eV**
CGCNN	0.031	0.292
SchNet	0.033	0.345
MEGNet	0.030	0.307
GATGNN	0.033	0.280
ALIGNN	0.022	0.218
CHGNet	0.030	-
PotNet	**0.019**	**0.204**
WyFormer	0.044	0.247

4 Conclusions and Limitations

Our work shares the limitation of most generative models for materials Zeni et al. (2024); Zhu et al. (2024); Xie et al. (2021); Jiao et al. (2024a): we learn the distribution from the training dataset, so there must be stable structures that are out-of-domain and won't be generated. E_{hull} as a proxy for stability is commonly used, but is imperfect, as it doesn't take into account entropy, and the hull determination relies on known structures. Using CHGNet for stability estimation is less precise compared to DFT, even thought it's one of the best models available, its energy prediction MAE on matbench discovery benchmark (Riebesell et al., 2023) is ≈ 0.06 eV.

Novelty and diversity evaluation is crucial. A model can generate structures that are same or similar to the ones in the training dataset, and are valid, but not very useful for material design. Counting complete duplicates is a step in the right direction, but doesn't measure substantial sample diversity Hicks et al. (2021).

In conclusion, we show that our Wyckoff Transformer approach represents a novel advancement in the generation of symmetric crystal structures by leveraging Wyckoff positions to encode material symmetries more efficiently. Unlike previous methods, Wyckoff Transformer achieves a higher degree of structure diversity while maintaining stability, by encoding the discrete symmetries of space groups without relying on atomic coordinates. This unique tokenization of symmetry elements enables the model to explore a reduced, yet highly representative space of possible configurations, resulting in more stable and purportedly synthesizable crystals. The model respects the inherent symmetry of crystalline materials, outperforms existing models in generating both novel and physically meaningful structures. These innovations underscore the method's potential in accelerating material discovery while maintaining accuracy in predicting key properties like formation energy and band gap, comparable to complementary methods.

Reproducibility Statement

The code and trained model weights will be published with the paper under an open source license.

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APPENDIX

A STRUCTURE GENERATION DETAILS

The process of obtainining crystal structures from Wyckoff representations using PyXtal Fredericks et al. (2021) begins by specifying a space group and defining WPs. PyXtal allows users to input atomic species, stoichiometry, and symmetry preferences. Based on these parameters, PyXtal generates a random crystal structure that respects the symmetry requirements of the space group. Once the initial structure is generated, we then perform energy relaxation using CHGNet. CHGNet is a neural network-based model designed to predict atomic forces and energies, significantly speeding up calculations that would traditionally require density functional theory (DFT). Energy relaxation involves optimizing the atomic positions to reach a minimum energy configuration, which represents the most stable form of the material. CHGNet, trained on vast DFT datasets, can efficiently relax crystal structures by adjusting atomic positions to reduce the total energy. This approach ensures that the final structure is not only symmetrical but also physically realistic in terms of energy stability.

For the 2nd structure generation method, DiffCSP++ is a diffusion-based crystal structure prediction model that focuses on generating purpotedly stable crystal structures by sampling from an energy landscape in a physically consistent manner. DiffCSP++ generation also starts with PyXtal sampling.

B ENERGY ABOVE HULL CALCULATIONS

To obtain the E_{hull}, we firstly constructed the reference convex hull data by querying all 153235 structures from the Materials Project (MP), and then using CHGNet (Deng et al., 2023) with using CrySPR interface (Nong et al., 2024) to do structure relaxations for all MP structures by relaxing both lattice cells and atomic positions (vc-relax), which renders 153,226 valid entries for relaxed structures and energies; secondly, for each 1,000 generated structures from each generative model, we followed the same vc-relax procedure to get the relaxed structures and energies; finally, using the `pymatgen.analysis.phase_diagram` sub–module the E_{hull} for each entry of generated structure was computed by referencing to the MP convex hull, $E_{\text{hull}} = \max\{\Delta E_i\}$, where ΔE_i is the decomposition energy of any possible path for a structure decomposing into the reference convex hull.

C DFT DETAILS

All DFT structure relaxations were performed using the Vienna ab-initio simulation package (VASP) with the plane-wave basis set. Kresse & Furthmüller (1996) The electron-ion interaction is described by the projector augmented wave (PAW) pseudo-potentials. Kresse & Joubert (1999) The pseudo-potentials recommended by the VASP team are used. The exchange-correlation of valence electrons is treated with the Perdew-Burke-Ernzerhof (PBE) functional within the generalized gradient approximation (GGA). Perdew et al. (1996) The cutoff for kinetic energy of plane waves was set to 520 eV. Convergence thresholds of 10^{-8} eV for total energy and 10^{-4} eV Å^{-1} atom^{-1} for force were set. The Monkhorst-Pack scheme of $\boldsymbol{k}$-points sampling in the Brillouin zone with spacing of 0.15 Å^{-1} is used Monkhorst & Pack (1976), in which the Γ point is included. The Dudarev et al.

simplified DFT+U scheme Dudarev et al. (1998) was adopted for the oxides and fluorides that contain one or more of the following transition metals: Co (3.32 eV), Cr (3.7 eV), Fe (5.3 eV), Mn (3.9 eV), Mo (4.38 eV), Ni (6.2 eV), V (3.25 eV), W (6.2 eV), consistent with the MP. Spin-polarized relaxations initialized with ferromagnetic, high-spin valence configurations were also performed to check if there is any magnetic atom with magnetism $\geq 0.15\ \mu_{\mathrm{B}}$.

The MP convex hull (v2023.11.1) was used as the reference hull. To do so comparably, additional DFT relaxations and self-consistent field (SCF) calculations using the VASP settings from `MPRelaxSet` and `MPStaticSet` in pymatgen were further performed based on the previously relaxed structures. The raw total energies of SCF calculations using the `MPStaticSet` are then corrected using the correction scheme of `MaterialsProject2020Compatibility` before putting into the `PhaseDiagram` to obtain the DFT E_{hull}. What should be emphasized here is that the precision parameters, which are generated by `MPRelaxSet` and `MPStaticSet`, are too coarse, regarding especially the convergence thresholds (2×10^{-4} eV for energy, and 2×10^{-3} eV Å^{-1} for cumulative force) and the density of $\boldsymbol{k}$-points sampling (equivalent to a spacing of only 0.35 Å^{-1}). The `MPRelaxSet` is not strictly appropriate for direct structure relaxations for generated structures that typically are far off equilibrium.

D Legacy metrics

For completeness sake, in table 3 we present the metrics computed following the protocol set up by Xie et al. (2021). We would like to again reiterate the issues with it. Firstly, the metrics are negatively correlated with structure novelty, the raison d'être for material generative models. Secondly, filtering by charge neutrality aka compositional validity means discarding viable structures.

Table 3: Method comparison according the protocol set up by Xie et al. (2021).

(a) Directly using structures produced by the methods, without additional relaxation. Note that CHGNet is an integral part of generating structures with Wyckoff Transformer and WyCryst, so it's used.

Method	**Validity (%)** ↑		**Coverage (%)** ↑		**Property EMD** ↓		
	Struct.	Comp.	COV-R	COV-P	ρ	E	N_{elem}
WyckoffTransformer	99.60	81.40	98.77	95.94	0.39	0.078	0.081
WyFormerDiffCSP++	99.80	81.40	99.51	95.81	0.36	0.083	**0.079**
CrystalFormer	93.39	84.98	99.62	94.56	0.19	0.208	0.128
DiffCSP++	99.94	**85.13**	99.67	**99.54**	0.31	**0.069**	0.399
WyCryst	99.90	82.09	99.63	96.16	0.44	0.330	0.322
DiffCSP	**100.00**	83.20	**99.82**	99.51	0.35	0.095	0.347
FlowMM	96.87	83.11	99.73	99.39	**0.12**	0.073	0.094

(b) All structures have been relaxed with CHGNet.

Method	**Validity (%)** ↑		**Coverage (%)** ↑		**Property EMD** ↓		
	Struct.	Comp.	COV-R	COV-P	ρ	E	N_{elem}
WyckoffTransformer	99.60	81.40	98.77	95.94	0.39	0.078	0.081
WyTransDiffCSP++	99.70	81.40	99.26	95.85	0.33	0.070	**0.078**
CrystalFormer	89.92	84.88	**99.87**	95.45	0.19	0.139	0.119
DiffCSP++	**100.00**	**85.80**	99.42	95.48	**0.13**	**0.036**	0.453
WyCryst	99.90	82.09	99.63	96.16	0.44	0.330	0.322
DiffCSP	**100.00**	82.50	99.64	95.18	0.46	0.075	0.321
FlowMM	**100.00**	82.83	99.71	**99.56**	0.17	0.046	0.093

E Evaluation on MP–20 binary & ternary

Comparison of WyFormer to WyCryst is presented in tables 4 and 5. Both models were trained on a subset of MP–20 training data containing only binary and ternary structures, and similarly selected

subset of MP–20 testing dataset is used as the reference for property distributions. All generated structures were relaxed with CHGNet. CHGNet was used for the formation energy computation for both generated and hull reference structures.

WyFormer outperforms WyCryst across the board. S.U.N. values are close, but this is achieved by WyCryst sacrificing sample diversity and property similarity metrics, with about half of the generated structures already existing in the training dataset.

Method	**Template Novelty** (%) ↑	**P1 (%)** ref = 1.7	**Space Group** $\chi^2 \downarrow$	**S.S.U.N.** (%) ↑
WyFormer	**25.63**	**1.43**	**0.224**	**37.9**
WyCryst	18.51	4.79	0.815	35.2

Table 4: Evaluation of the methods according to the symmetry metrics. Aside from Template Novelty, metrics are computed only using novel structurally valid structures.

Method	**Novelty** (%) ↑	**Validity (%)** ↑		**Coverage (%)** ↑		**Property EMD** ↓			**S.U.N.** (%) ↑
		Struct.	Comp.	COV-R	COV-P	ρ	E	N_{elem}	
WyFormer	**91.19**	**99.89**	**77.28**	**98.90**	**96.75**	**0.83**	**0.064**	0.084	**38.4**
WyCryst	52.62	99.81	75.53	98.85	89.27	1.35	0.128	**0.003**	36.6

Table 5: Evaluation of the methods according to validity and property distribution metrics. Following the reasoning in section 3.1, we apply filtering by novelty and structural validity, and do not discard structures based on compositional validity. Validity is also computed only for novel structures.

F Hyperparameters

F.1 Optimizer

We use SGD optimizer with starting learning rate 0.2, and `ReduceLROnPlateau` scheduler with `factor=0.8` and patience of 40k epochs monitoring the validation dataset loss.

G Fine-tuning LLM with Wyckoff representation

To challenge Wyckoff Transformer's architecture, we compared it with pre–trained language models that were used in vanilla mode as well as after fine–tuning. We explored two different textual representations of crystals corresponding to a given space group:

- **Naive**, which contains the specifications of atoms at particular symmetry groups encoded by Wyckoff symmetry labels: `Na at a, Na at a, Na at a, Mn at a, Co at a, Ni at a, O at a, O at a, O at a, O at a, O at a, O at a`
- **Augmented**, which contains the specifications of atom types with its' symmetries and site enumerations: `Na @ m @ 0, Na @ m @ 0, Na @ m @ 0, Mn @ m @ 0, Co @ m @ 0, Ni @ m @ 0, O @ m @ 0, O @ m @ 0, O @ m @ 0, O @ m @ 0, O @ m @ 0, O @ m @ 0`, where the set of valid symmetries is: `['2.22', '4/mmm', '1', '-3..', '6mm', 'm-3m', '2', '3mm', '.m', '-6mm2m', '4mm', '.32', '322', '.2/m.', '-1', '.m.', '..m', 'm.2m', '.3m', '3m', 'm2m.', '2mm', '-32/m.', '2..', '..2', '.3.', '2/m', '-43m', '4/mm.m', '.2.', '2/m2/m.', '23.', '222', 'm..', 'mm.', '-3.', 'm-3.', '3.', '4/m..', '.-3m', '2m.', '-32/m', '-42m', 'm.mm', '4..', 'm.m2', '422', '32.', '22.', '-622m2', '3m.', '.-3.', 'mmm..', '222.', 'mm2..', '-4m2', '2/m..', 'mm2', '-3m2/m', '-4m.2', '2mm.', '3..',`

```
'-42.m', '..2/m', '4m.m', '-4..', '6/mm2/m', 'm2m', 'm2.',
'2.mm', 'mmm.', 'mmm', '32', 'm', '-6..']
```

We fine-tuned the OpenAI `chatGPT-4o-mini-2024-07-18` model using different representations and compared it with the vanilla OpenAI `gpt-4o-2024-08-06` model. For each of the cases prompt looked like: `Provide example of a material for spacegroup number X`. The table below contains details of the model training:

Model	Base Model	Representation	Hyperparameters	Training Time	Inference Time	Number of Parameters
WyLLM-vanilla	gpt-4o-2024-08-06	Naive	–	–	74m	$\approx$ 200B
WyLLM-naive	gpt-4o-mini-2024-07-18	Naive	epochs: 1, batch: 24, learning rate multiplier: 1.8	51m	51m	$\approx$ 8B
WyLLM-site-symmetry	gpt-4o-mini-2024-07-18	Site Symmetry	epochs: 1, batch: 24, learning rate multiplier: 1.8	95m	37m	$\approx$ 8B

Table 6: Comparison of different models and their characteristics. Number of parameters is not known exactly and is taken from public sources as an approximate estimation. For reference, WyFormer has 150k parameters.

Both training and inference times were measured using batch job execution on OpenAI's cloud. The fine-tuned model returned a JSON string that was easy to parse, while the vanilla model required additional parsing of its output.

Method	Novelty	Validity (%) $\uparrow$		Coverage (%) $\uparrow$		Property EMD $\downarrow$		
	(%) $\uparrow$	Struct.	Comp.	COV-R	COV-P	ρ	E	N_{elem}
WyFormer	89.50	99.66	80.34	99.22	**96.79**	0.67	**0.050**	0.098
WyLLM-naive	94.67	99.79	82.89	98.72	94.97	0.39	0.067	**0.015**
WyLLM-vanilla	**95.59**	99.82	**88.75**	94.46	59.67	2.23	0.234	0.253
WyLLM-site-symmetry	89.58	**99.89**	83.89	**99.44**	96.32	**0.29**	nan	0.039

Method	Wyckoff Validity (%) $\uparrow$	Novel Unique Templates (#) $\uparrow$	$P1$ (%) ref $= 1.7$	Space Group $\chi^2 \downarrow$
WyFormer	**97.8**	186	1.46	0.212
WyLLM-naive	94.9	**237**	**1.38**	0.167
WyLLM-vanilla	*28.7*	87	2.03	0.621
WyLLM-site-symmetry	89.6	191	2.24	**0.158**

Table 7: Comparison for WyFormer to different variant of WyLLM. All structures have been relaxed with DiffCSP++. Sample size is 1000 structures per model. The metrics described in section 3.1. `nan` is placed where the generated structures contained a rare element that crashed the property computation code. Wyckoff Validity refers to the percentage of the generated outputs that are valid Wyckoff representations. Aside from LLM–specific problems, such as non–existent elements, a Wyckoff representation can be invalid if it places several atoms at Wyckoff position without degrees of freedom, or refers to Wyckoff positions that do not exist in the space group.

Comparison the WyFormer to WyLLM is present in table 7. When fine–tuned, an LLM using Wyckoff representations shows similar performance to WyFormer – at a much greater computational cost. Using site symmetries instead of Wyckoff letters doesn't unequivocally increase the LLM performance, a possible explanation is that since this representation is our original proposition, the LLM is less able to take advantage of pre–training that contained letter–based Wyckoff representations. Without fine-tuning, the majority of LLM outputs are formally invalid, and the distribution of the valid ones doesn't match MP–20.