Crystal Structure Prediction with Improved Initialization

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Crystal Structure Prediction from **First-principles**

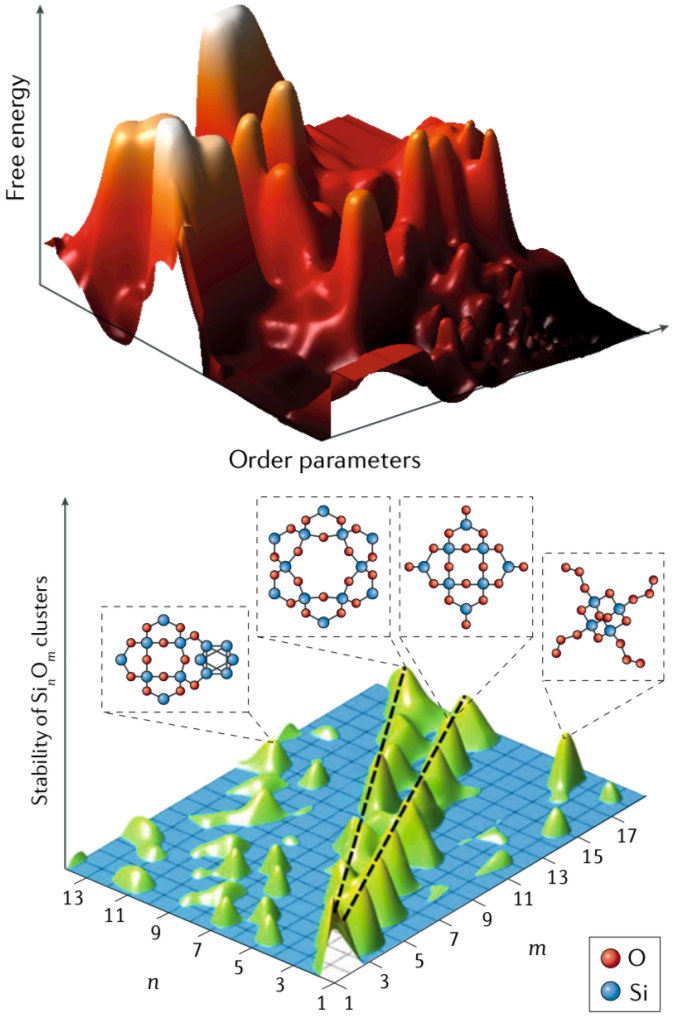
Structure prediction drives materials discovery

Artem R. Oganov[1,2,3], Chris J. Pickard[4,5]*, Qiang Zhu[6] and Richard J. Needs[7]*

Nature Review Materials, 2019

- **Structure navigation**
  - Evolutionary algorithm (USPEX)
  - Random search (AIRSS)
  - Minima hopping
  - Some others ……

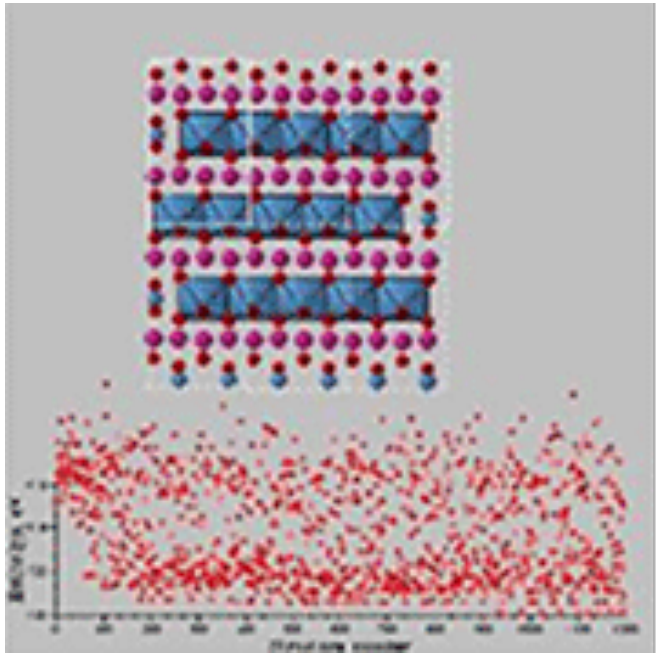
- **Energy & Properties Evaluation**
  - Evolutionary algorithm (USPEX)
  - Random search (AIRSS)
  - Minima hopping
  - Some others ……



Universal Structure Predictor: Evolutionary Xtallography

Inorganic crystals

Oganov, JCP, 2006

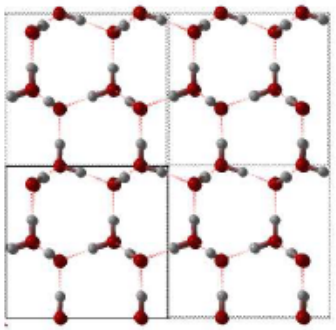


$MgSiO_3$

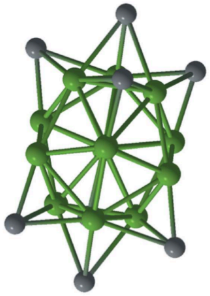
Organic crystals

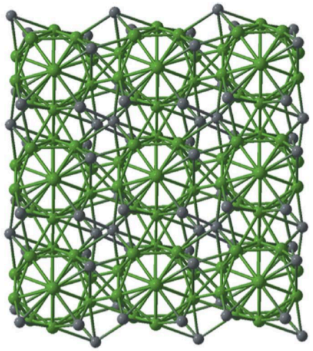
Zhu, Acta Cryst B, 2012

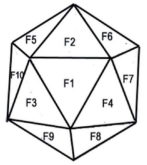
H_2O



Methane A



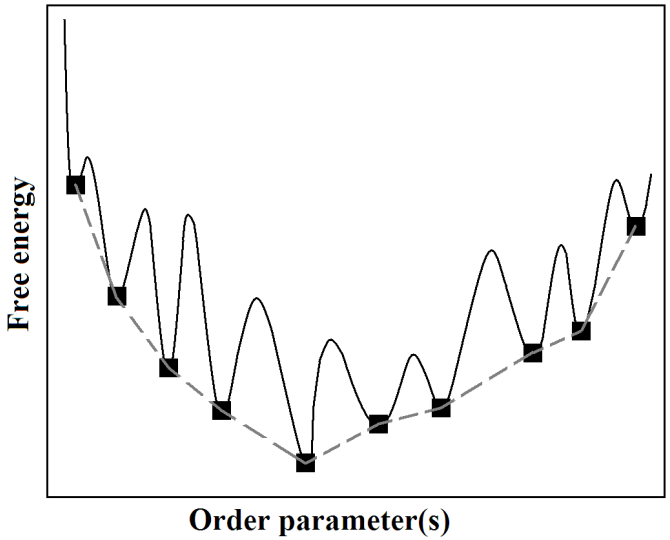




CSP Strategy

Global optimization Structural Navigation *derivative-free*

- (Quasi)-random search
- (Quantum) simulated annealing
- Minima/Basin hopping
- Bio-inspired algorithms
  - Evolutionary/genetic
  - Particles swarm
  - Ant colony
  - Firefly



Local optimization Geometry Optimization *Convex optimization*

- DFT
- Semi-classical
- Generic (customized) force field
- Machine learning

CSP Strategy (Population based methods)

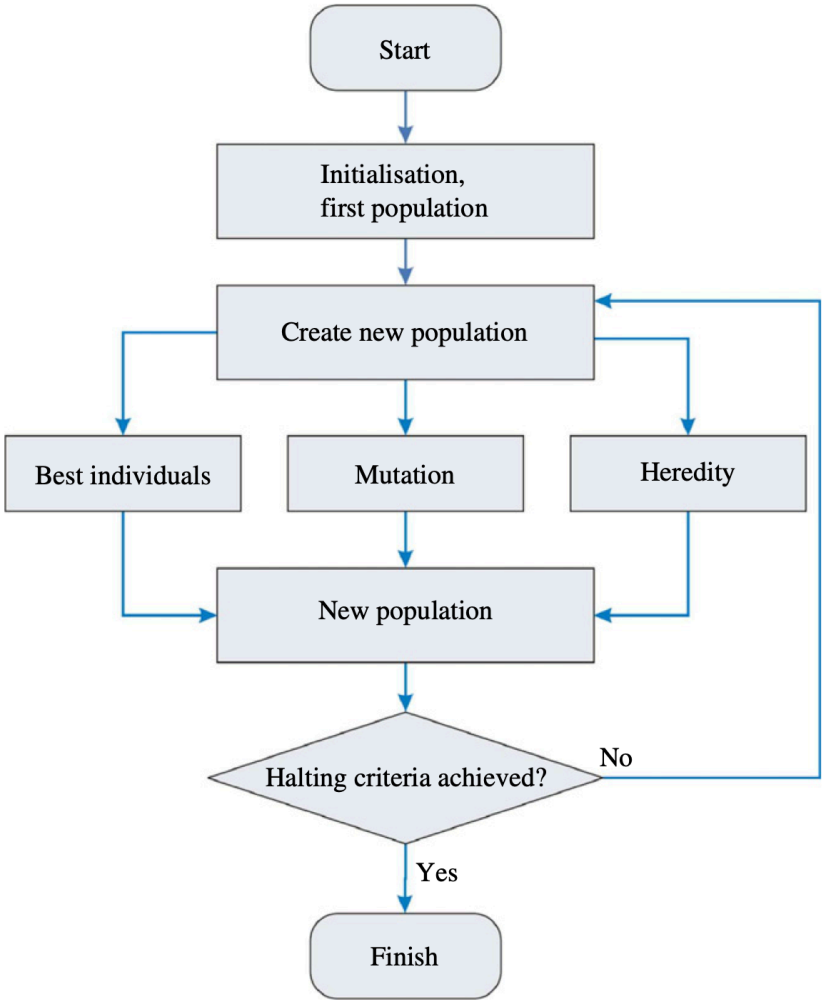


Fig. 1. Flowchart of a typical evolutionary algorithm.

Lyakhov, CPC, 2013

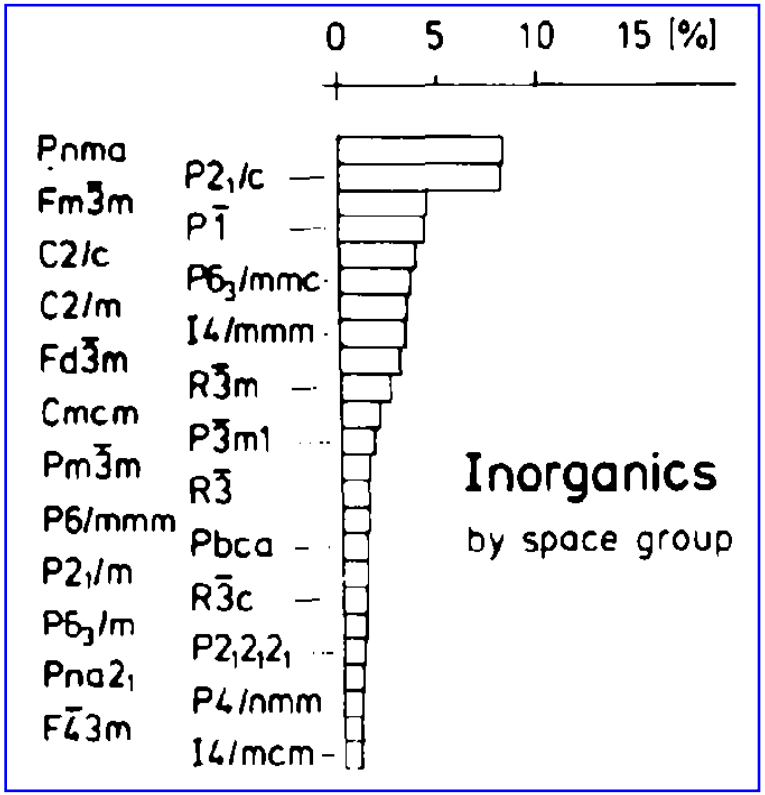
All population based methods requires an structure initialization

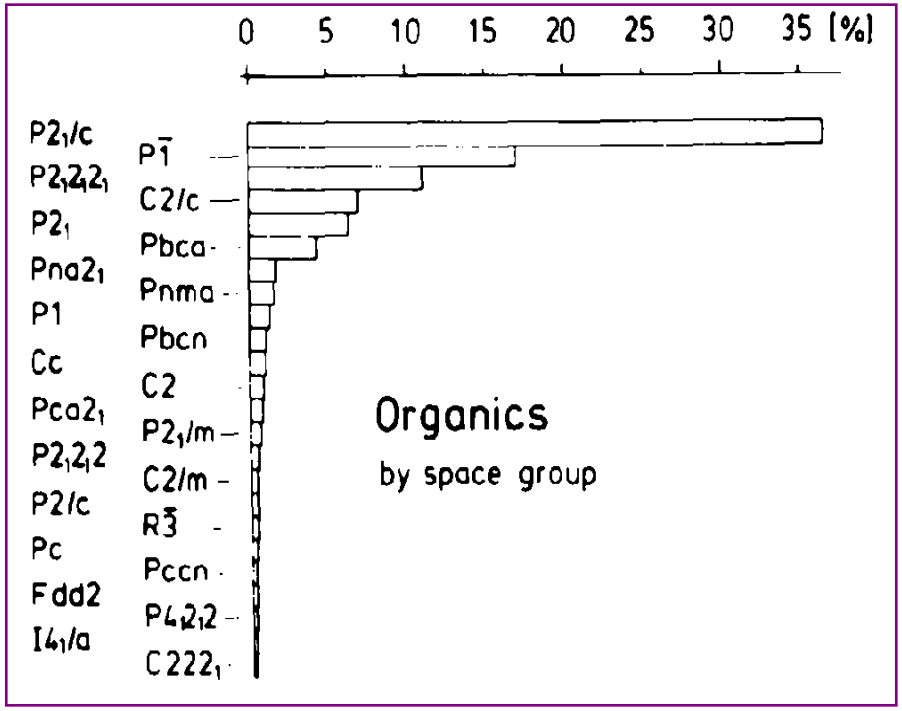
- Purely random
- Random symmetrization
- Random topology (Bushlanov, CPC, 2019)

In this talk, I will focus on the random symmetrization based on the open source code PyXtal.

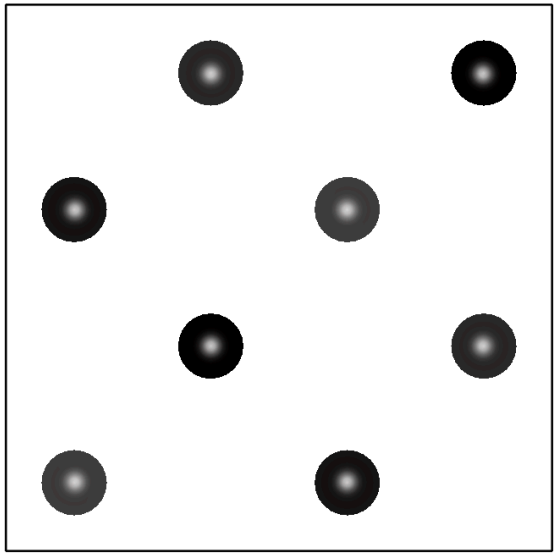
PyXtal

Symmetry Preference





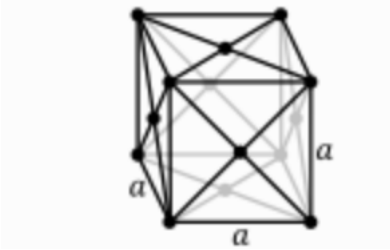
How to describe a crystal by symmetry?



A diamond structure

Space group: Fd-3m (227)

Bravi lattice: Face-centered cubic



Atomic Positions

1/8	1/8	1/8
7/8	3/8	3/8
1/8	5/8	5/8
7/8	7/8	7/8
5/8	1/8	5/8
11/8	3/8	7/8
5/8	5/8	1/8
11/8	7/8	3/8

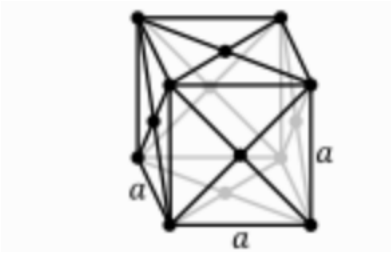
Wyckoff Sites

192i
96h
96g
48f
32e
16d
16c
8b
8a

How to randomly generate a diamond-like crystal?

Space group: Fd-3m (227)
8 carbon atoms

1. Build a FCC lattice



2. Choose Wyckoff sites

Wyckoff Sites
192i
96h
96g
48f
32e
16d
16c
8b
8a

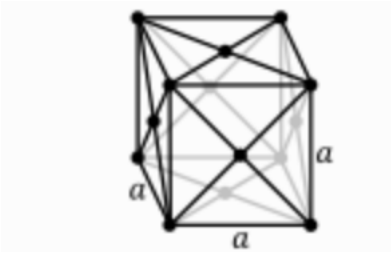
3. Fill atomic positions

Atomic Positions		
1/8	1/8	1/8
7/8	3/8	3/8
1/8	5/8	5/8
7/8	7/8	7/8
5/8	1/8	5/8
11/8	3/8	7/8
5/8	5/8	1/8
11/8	7/8	3/8

How to randomly generate a diamond-like crystal?

Space group: Fd-3m (227)
16 carbon atoms

1. Build a FCC lattice



2. Choose Wyckoff sites

Wyckoff Sites

192i
96h
96g
48f
32e
16d
16c
8b
8a

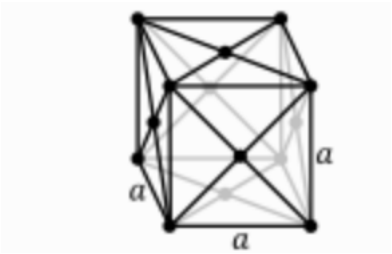
3. Fill atomic positions

- 16c
- 16d
- 8a + 8b

How to randomly generate a diamond-like crystal?

Space group: Fd-3m (227)
32 carbon atoms

1. Build a FCC lattice



2. Choose Wyckoff sites

Wyckoff Sites
192i
96h
96g
48f
32e
16d
16c
8b
8a

3. Fill atomic positions

- 32e
- 16c + 16d
- 8a + 8b + 16c
- 8a + 8b + 16d

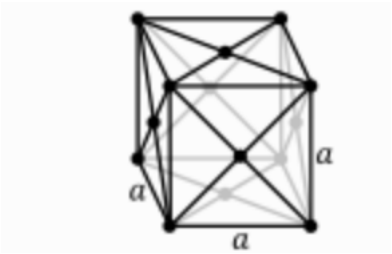
32e		
x	x	x
-x	-x	-x
…	…	…

High multiplicities means more freedoms

How to randomly generate a diamond-like crystal?

Space group: Fd-3m (227)
32 carbon atoms

1. Build a FCC lattice



2. Choose Wyckoff sites

Wyckoff Sites

192i
96h
96g
48f
32e
16d
16c
8b
8a

3. Fill atomic positions

- 32e
- 16c + 16d
- 8a + 8b + 16c
- 8a + 8b + 16d

192i

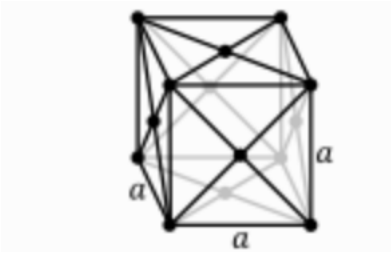
x	y	z
-x	-y	-z
…	…	…

High multiplicities means more freedoms

How to randomly generate a diamond-like crystal?

Space group: Fd-3m (227)
32 carbon atoms

1. Build a FCC lattice



2. Choose Wyckoff sites

Wyckoff Sites
192i
96h
96g
48f
32e
16d
16c
8b
8a

3. Fill atomic positions

- 32e
- 16c + 16d
- 8a + 8b + 16c
- 8a + 8b + 16d

192i

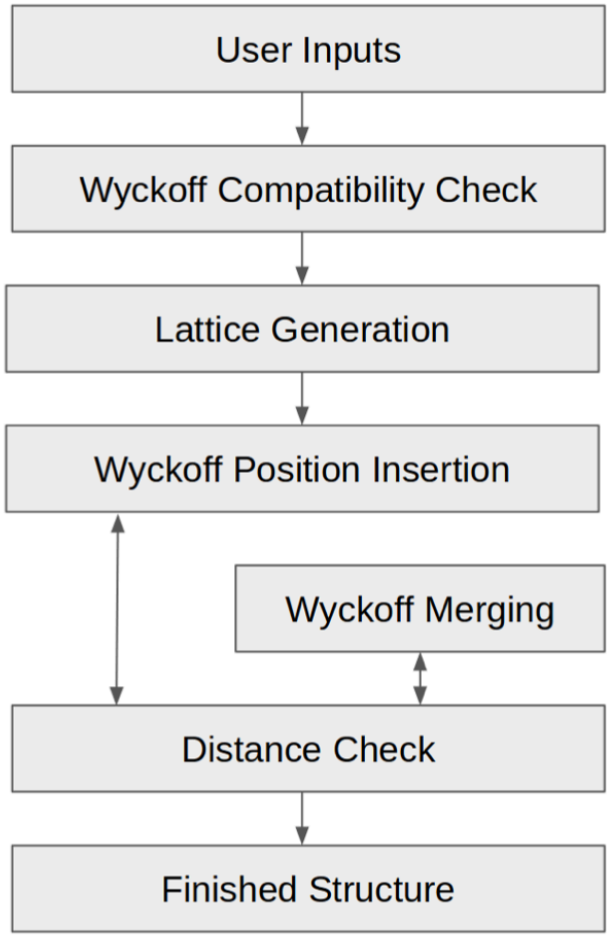
x	y	z
-x	-y	-z
…	…	…

High multiplicities means more freedoms

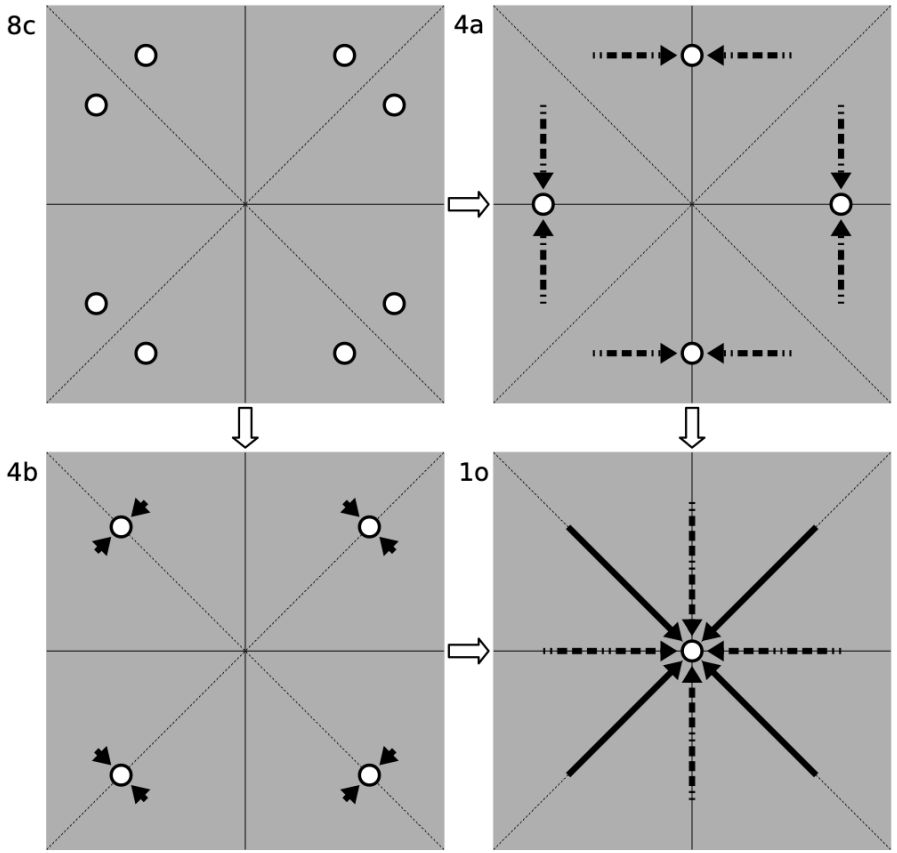
How to choose the right combination of Wyckoff sites?

- Precompute all the allowed solution, then randomly pick one
- **Start with the highest possible sites, then merge it (to avoid selecting too high-symmetrical results)**

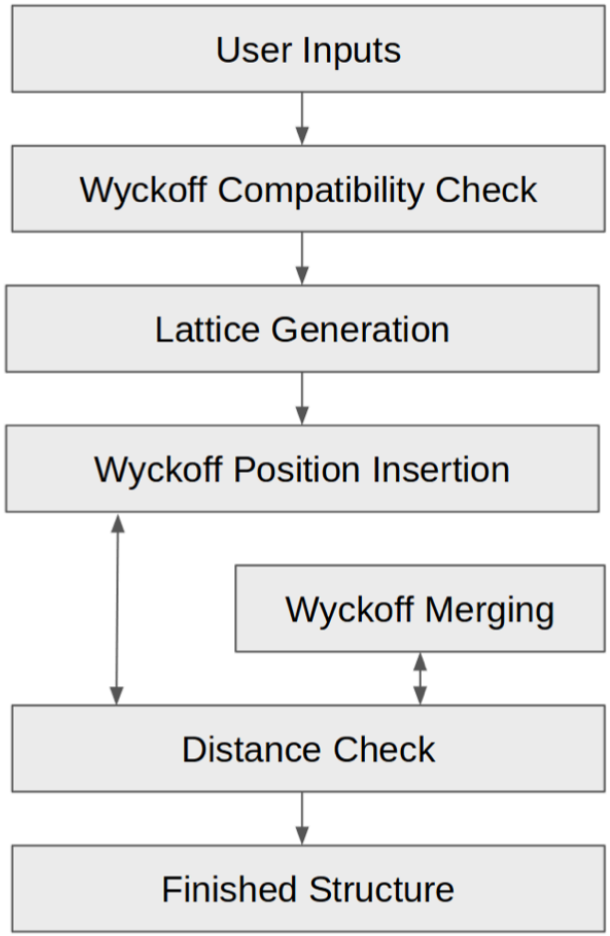
Wyckoff Site Selection & Merging

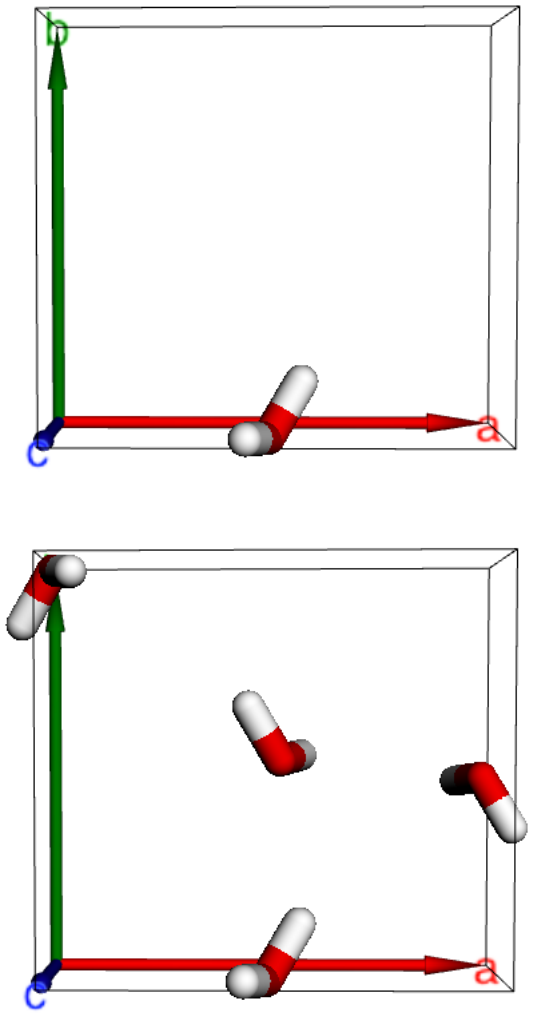


Space group: C_{4v} *4mm*



How to generate a molecular crystal?



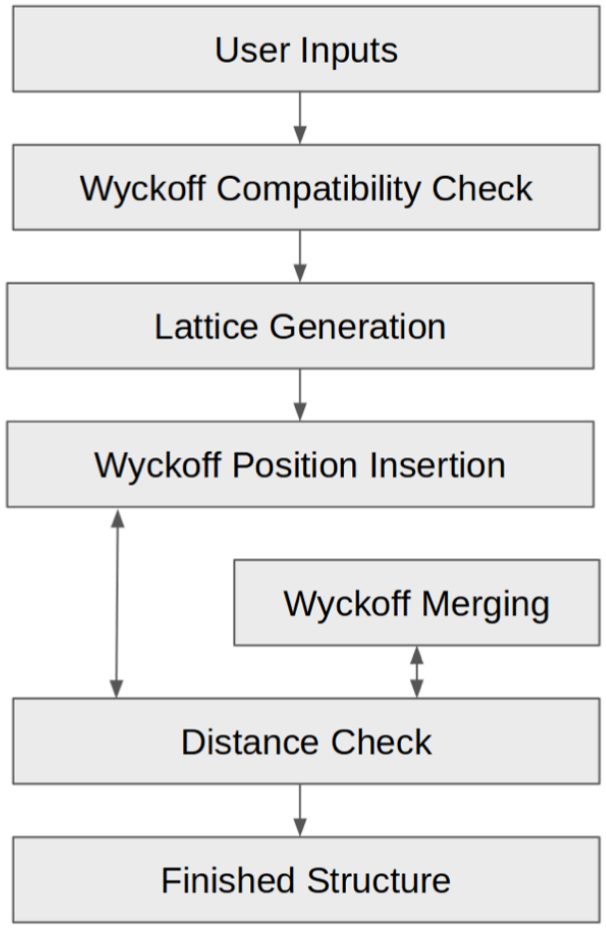


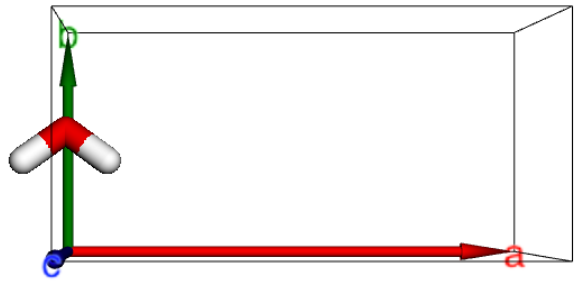
Space group: $P2_12_12_1$
# of molecules: 4 H_2O
Cell params: optional
Wyckoff sites: optional

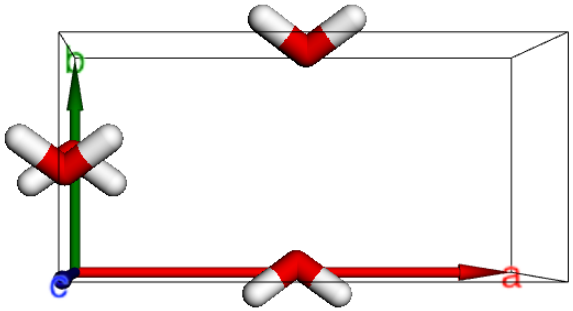
Site	Symmetry
4a	m..

x	y	z
-x+1/2	-y	z+1/2
-x	y+1/2	-z+1/2
x+1/2	-y+1/2	-z

How to generate a molecular crystal with Z`<1?







Space group: *Cmc*2_1
# of molecules: 4 H_2O
Cell params: optional
Wyckoff sites: optional

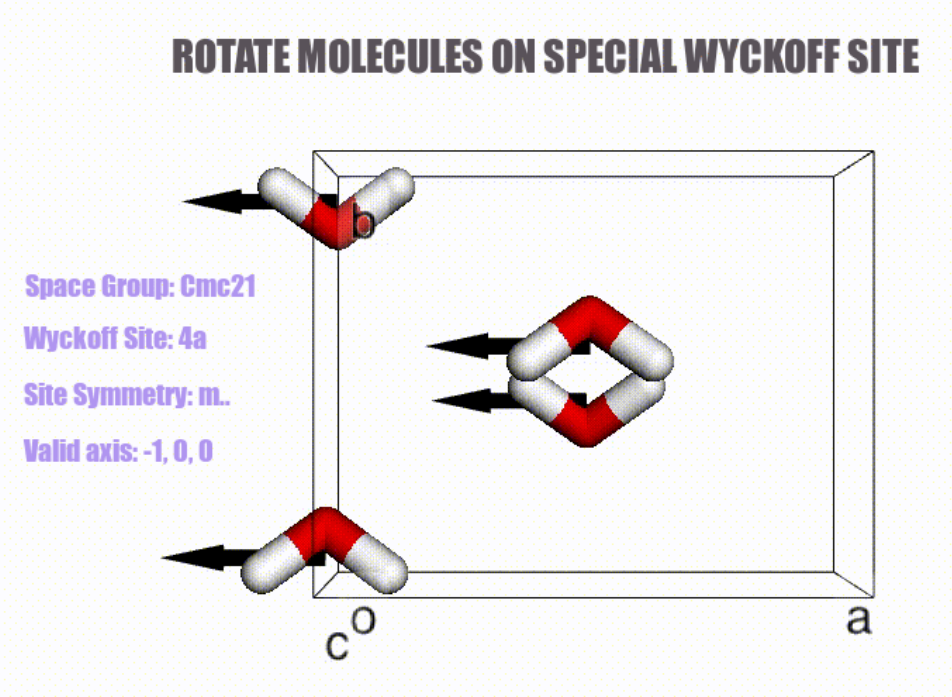
Site	Symmetry
8b	1
4a	m..

0	y	z
0	-y	z+1/2
1/2	y+1/2	z
1/2	-y+1/2	z+1/2

Molecular structures occupying Wyckoff Positions

- (Special) Wyckoff positions have site symmetry
- Molecular symmetry must be equal or greater
- Orientation must be correct
- All valid orientations will be calculated by PyXtal

- Z' can be any integer and fractional numbers



Molecular structures occupying Wyckoff Positions

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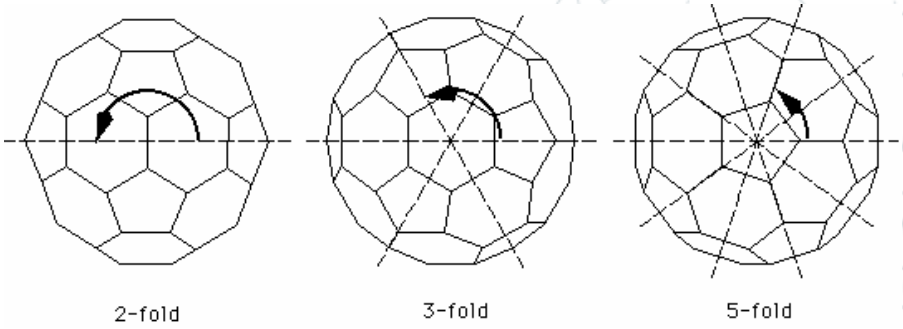
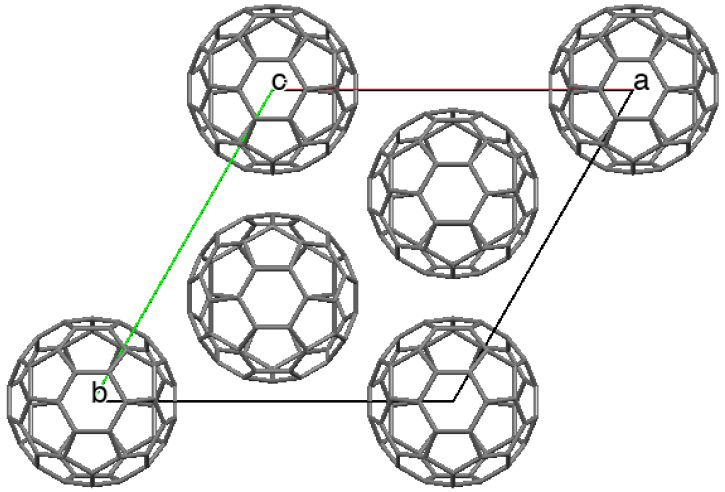
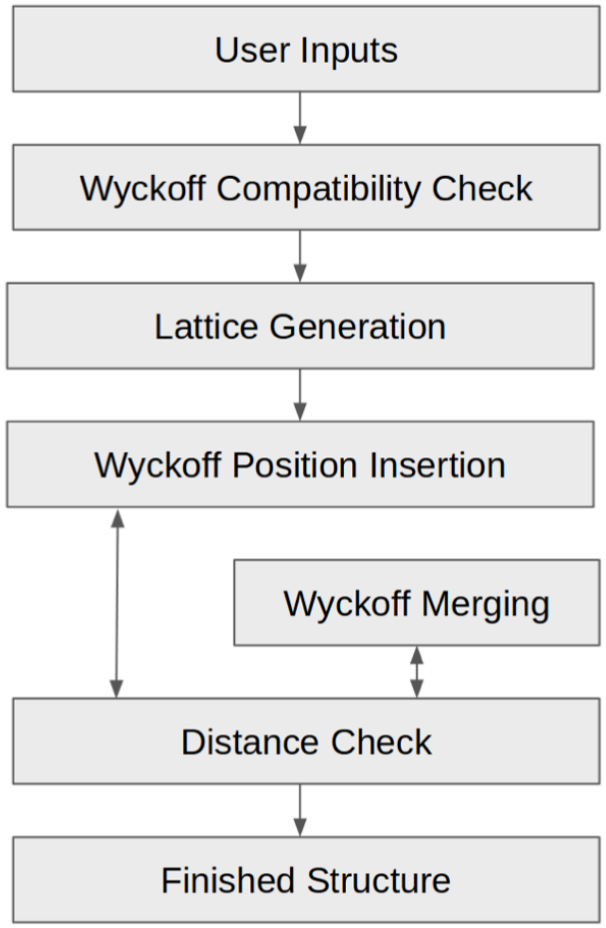


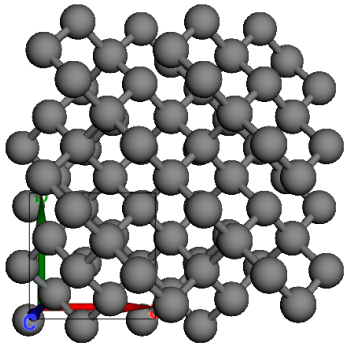


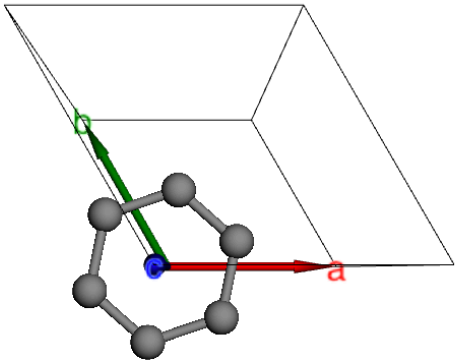
C_{60}: $P6_3mc$ (Z=4, Z'=1/3)

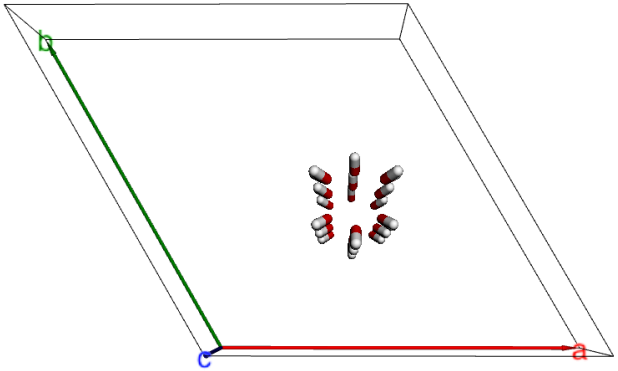
More examples on symmetric generation

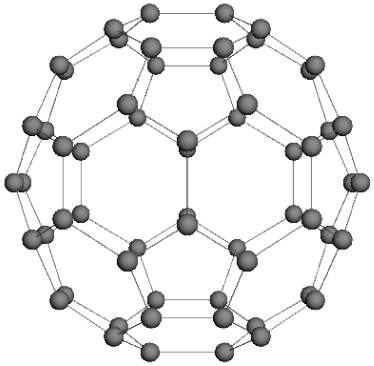


- Atomic crystal (0D, 1D, 2D, 3D)
- Molecular crystal (1D, 2D, 3D)









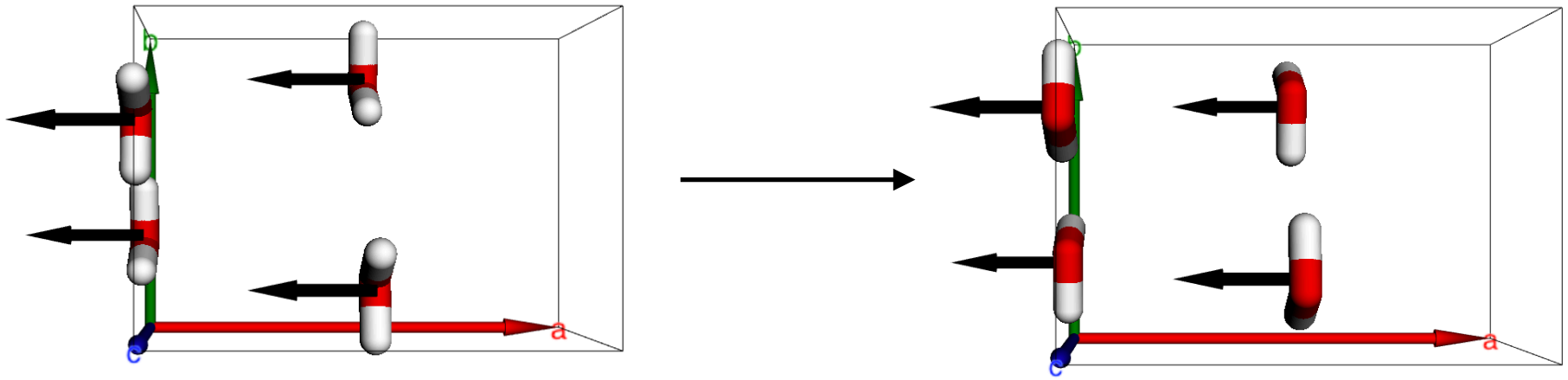
https://pyxtal.readthedocs.io

Vary the crystals: Symmetry invariant

```python
from pyxtal.molecular_crystal import molecular_crystal

h2o = molecular_crystal(36, ['H2O'], [2])
print(h2o)
```

```
------Random Molecular Crystal------
Dimension: 3
Group: Cmc21
Volume factor: 1.1
orthorhombic lattice:   4.9933   5.6751   6.1478  90.0000  90.0000  90.0000
Wyckoff sites:
        H2O1 @ [0.0000 0.4828 0.4315]  WP:  4a, Site symmetry m.. ==> Euler: -172.390 -47.970 -108.176
```



Rotation & translation according to the given constraints

Vary the crystals: Lower the symmetry

```
1 from pyxtal.crystal import random_crystal
```

```
1 C = random_crystal(227, ['C'], [2], 1.0)
```

```
1 print(C)
```

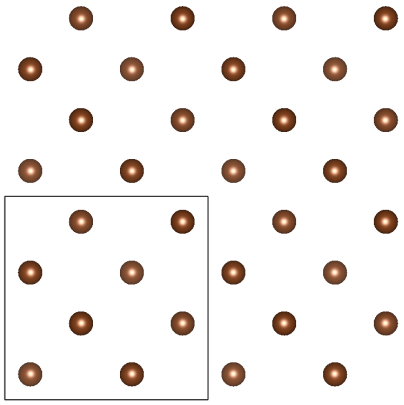
```
------Random Crystal------
Composition: C8
Dimension: 3
Group: Fd-3m (227)
Volume factor: 1.0
cubic lattice:   5.1495   5.1495   5.1495  90.0000  90.0000  90.0000
Wyckoff sites:
        C @ [0.1250 0.1250 0.1250], WP:  8a, Site symmetry: -4 3 m
```

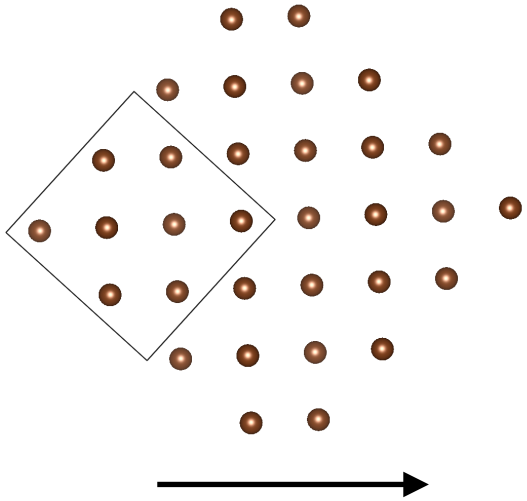
Supported since PyXtal-v0.1.2

```
1 C1 = C.subgroup(H=141)
2 print(C1)
```

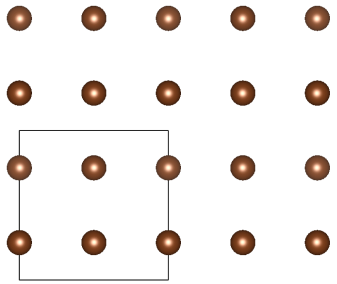
```
------Random Crystal------
Composition: C8
Dimension: 3
Group: I41/amd (141)
Volume factor: 1.0
tetragonal lattice:   3.6413   3.6413   5.1495  90.0000  90.0000  90.0000
Wyckoff sites:
        C @ [0.0200 0.7718 0.1204], WP:  4a, Site symmetry: -4 m 2
```

Fd-3m
8a





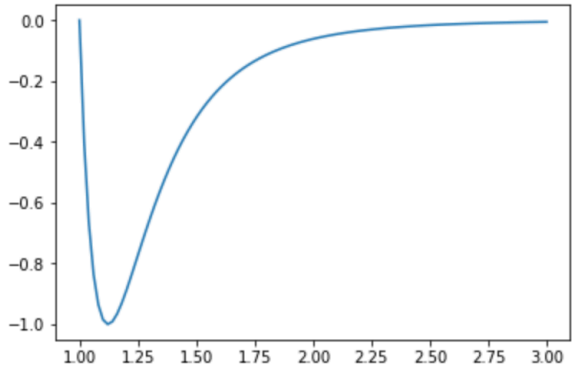
I41/amd
4a

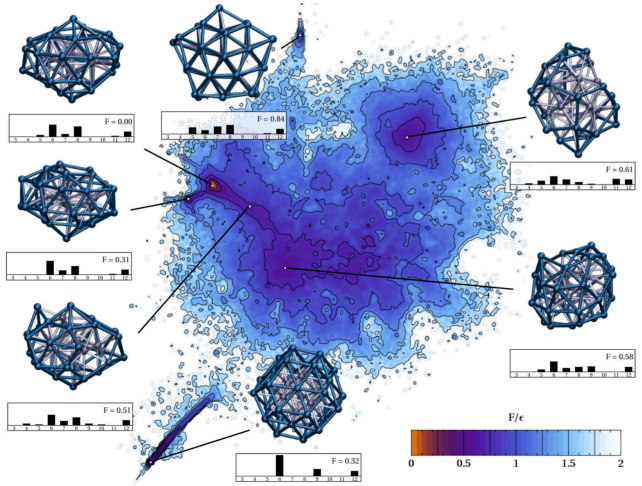


Transformation matrix
Wyckoff splitting scheme

Structure Optimization: Lennard-Jones Clusters

$$V(r) = 4\epsilon\left[\left(\frac{\delta}{r}\right)^{12} - \left(\frac{\delta}{r}\right)^{6}\right]$$



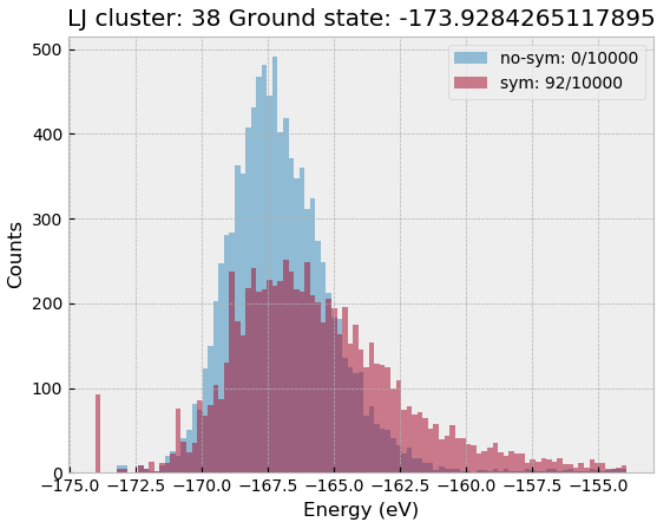


Ceriotti, Tribello, Parrinello, JCTC, 2013

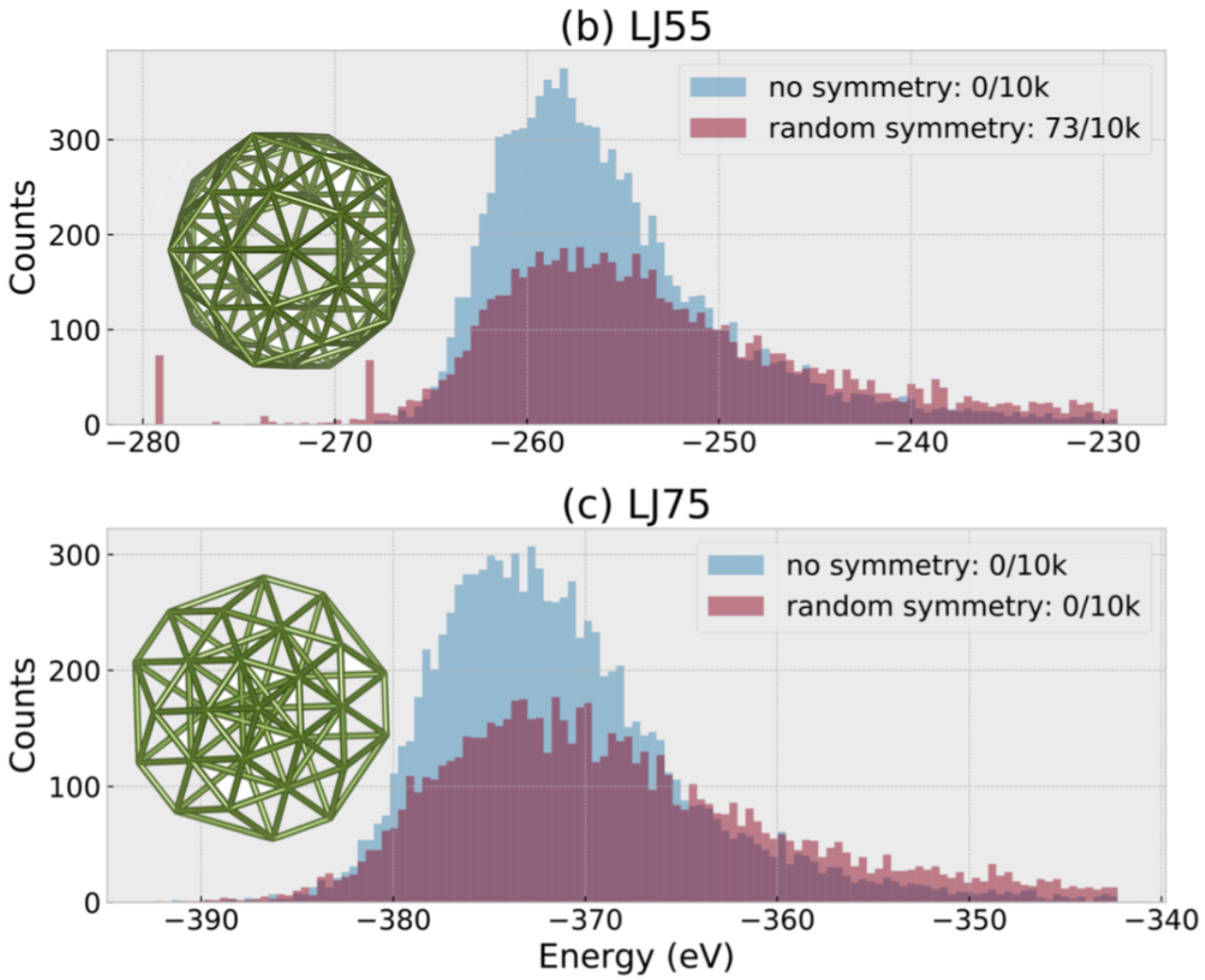
Random sampling

PyXtal

- 10000 random structures
- then optimize



Structure Optimization: Lennard-Jones Clusters

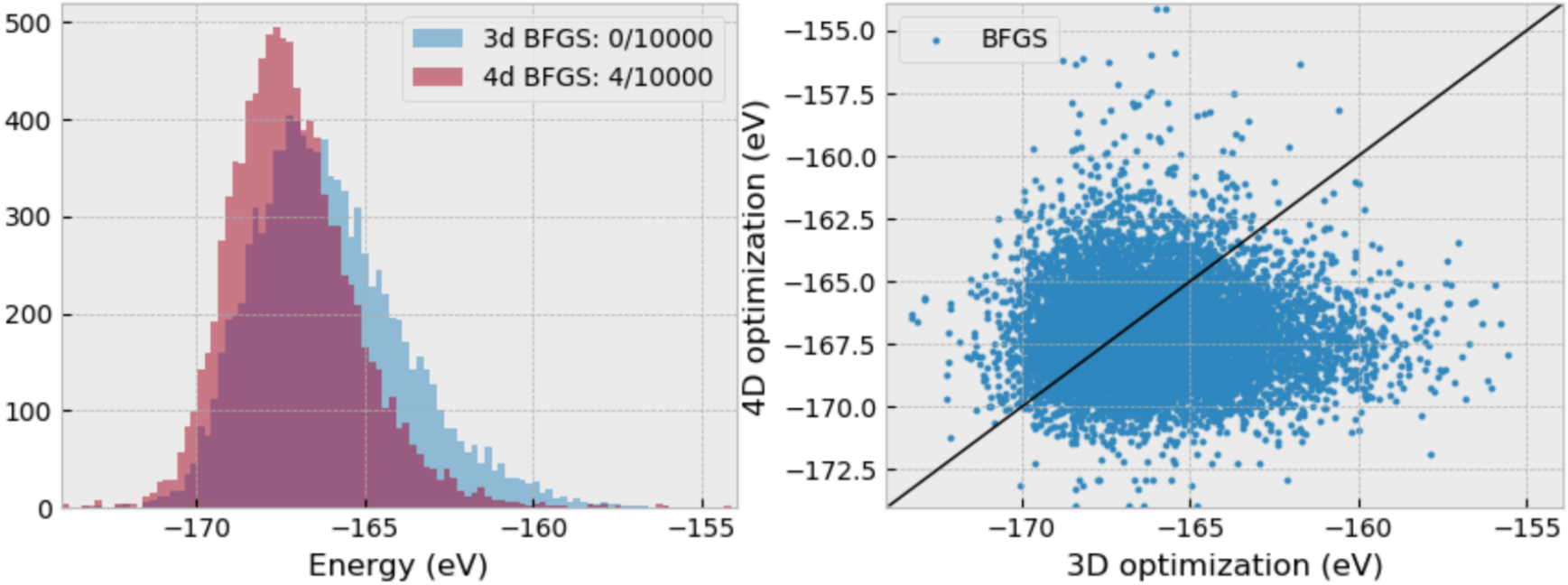


Structure prediction can be improved by handling structure generation.

Structure Optimization: Exploratory algorithms

- 10000 random structures with *C1* symmetry
- Optimize them differently
  - Ordinary L-BFGS
  - Stochastic hyperspatial optimization

C. J. Pickard, PRB, 2019

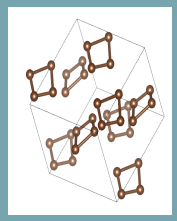


PyXtal *Python library for handling crystal symmetry*

Input Constraints

* Stoichiometry
* Cell Parameters
* Space Group

Random Structures

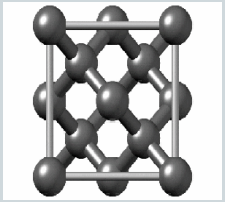


Optimization

DFT/FF/ML

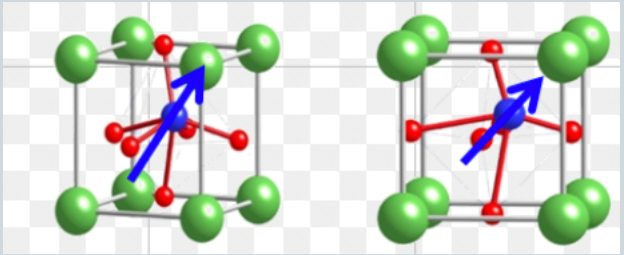
- *Gradient decent*
- *Conjugate gradient*
- *Quasi newton*
- *Stochastic methods*

Optimized Structures



Variation

- Perturbation by group/subgroup relation
- Identify pseudo symmetry



Atomic/Molecular

4D: superspace group
3D: space group
2D: Layer group
1D: Rod group
0D: Point group

Summary

PyXtal is an open source code to deal with structures

Generation
- Generation of atomic/molecular crystals from 0D, 1D, 2D to 3D
- Crystals with molecules in special Wyckoff positions (Z`<1)

Modulation
- Group-subgroup relation
- Interface with Pymatgen/ASE for structural manipulation
- Exploratory optimization algorithms

Analysis
- Symmetry information
- PXRD

PyXtal

2018/04

Launched by Scott

Supervised by Zhu

2019/07

Dee-y created the logo

Scott graduated

Dean/Stanley

implemented XRD

2019/09

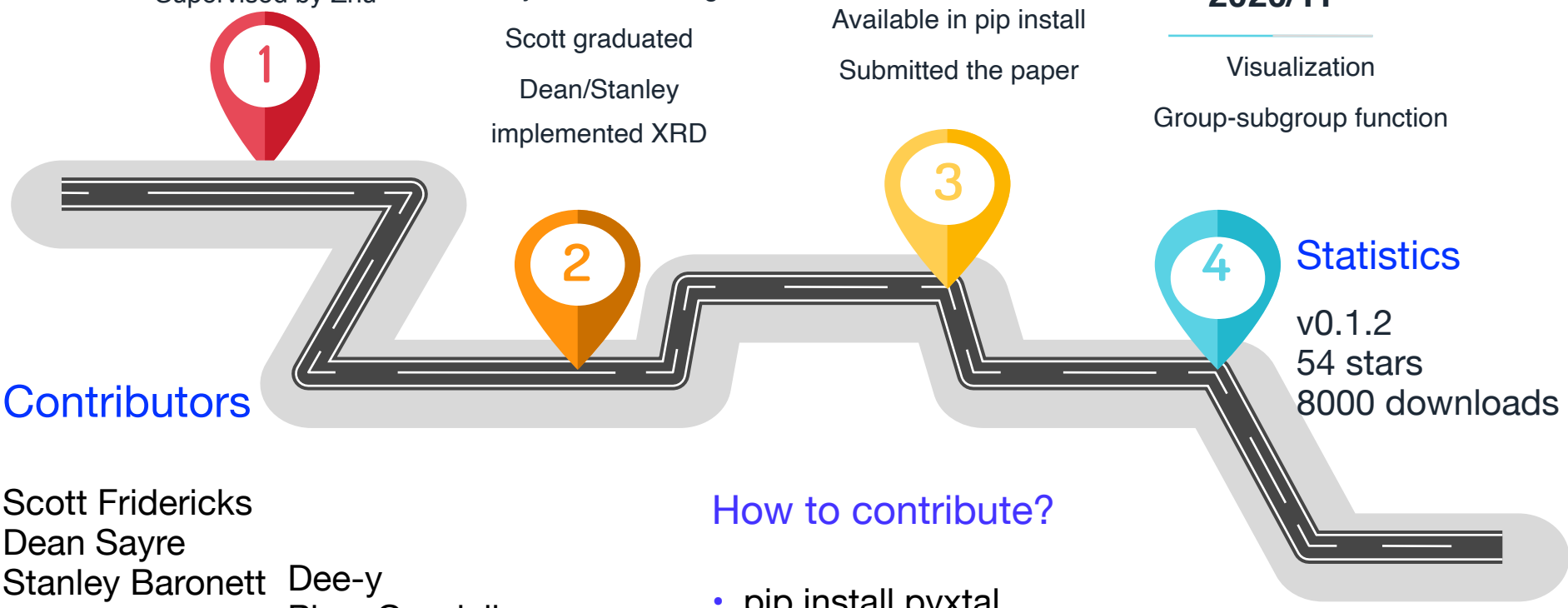
Available in pip install

Submitted the paper

2020/11

Visualization

Group-subgroup function



Statistics

v0.1.2
54 stars
8000 downloads

Contributors

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Janosh Riebesell

Outside

How to contribute?

- pip install pyxtal
- Star our project in Github
- Suggestions
- Fork the repository
- Send your pull request

Acknowledgement

Merci
谢谢
Спасибо
Grazie
Thank You
ありがとう

Skoltech
- A. R. Oganov

UNLV
- Scott Fredericks
- Howard Yanxon
- David Zagaceta
- Kevin Parish

1-2 **postdocs opening** in
- Molecular simulation
- Code development
- Forcefield development

Feel free to contact **qiang.zhu@unlv.edu**