Annealing for the prediction of grand canonical structures

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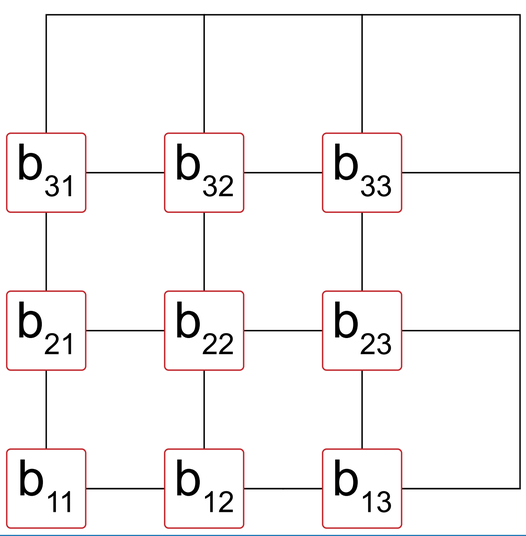
Background

Goal: Solve crystal structure prediction (CSP) problem using quantum annealing (QA) or simulated annealing (SA).



1. Fix mesh size g and discretize unit cell by equipartitioning base vectors into g parts giving lattice r_{ij} (here g=3).
2. Per species in the crystal, place binary variable b_{ij} on lattice points r_{ij} (here single species).
3. Define equivalence: $b_{ij} = 1 \leftrightarrow$ place atom of corresponding species on r_{ij}.
4. Given interatomic potential of any order (here second) we get

$$\min_{\{r\}} \frac{1}{2} \sum_{n,m}^{N} E_2(r_n, r_m) = \min_{\{b\}} \frac{1}{2} \sum_{i,j,k,l}^{g} E_2(r_{ij}, r_{kl}) b_{ij} b_{kl} + \Big(\sum_{ij} b_{ij} - N\Big)^2 + \Big(\sum_{ij} b_{ij} - c \sum_{ij} \tilde{b}_{ij}\Big)^2$$

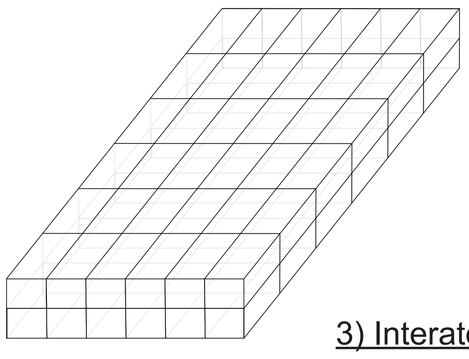
i.e. the equivalence: optimal configuration on lattice ↔ optimal binary string.

5. The resulting quadratic unconstrained binary optimization problem (QUBO) is solved using Ising machines.
6. No need to specify N, possible by adding orange penalty (fix atom number), or blue penalty (fix atom ratios).

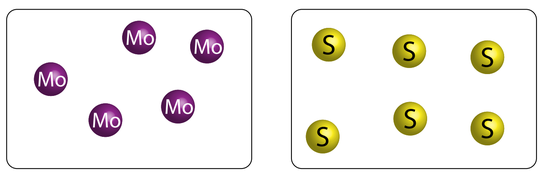
Algorithm & Results

INPUT

1) Unit Cell + Mesh size



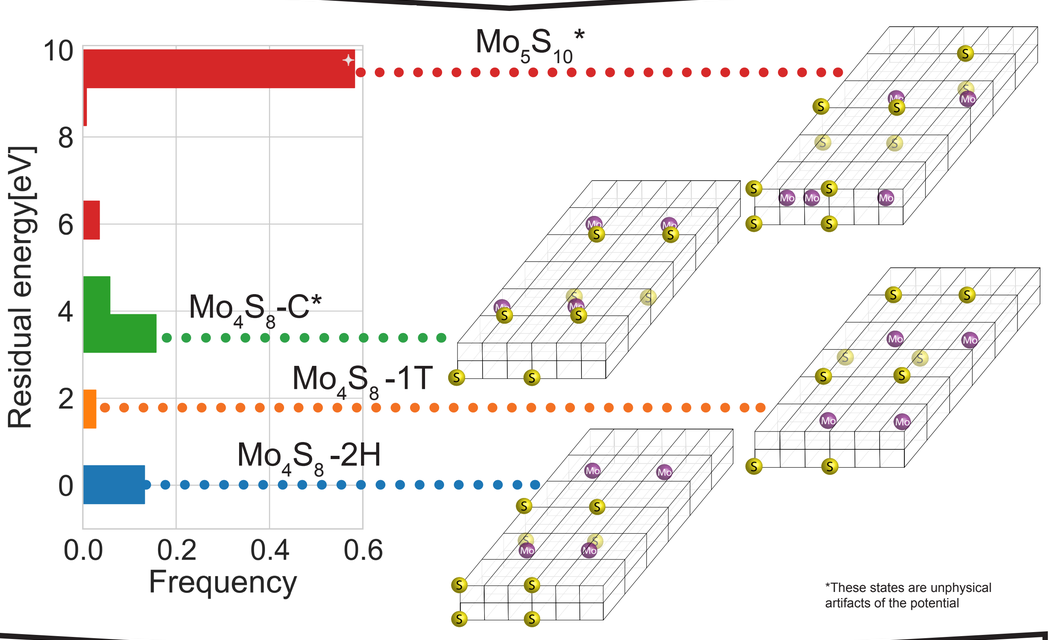
2) Atom Species



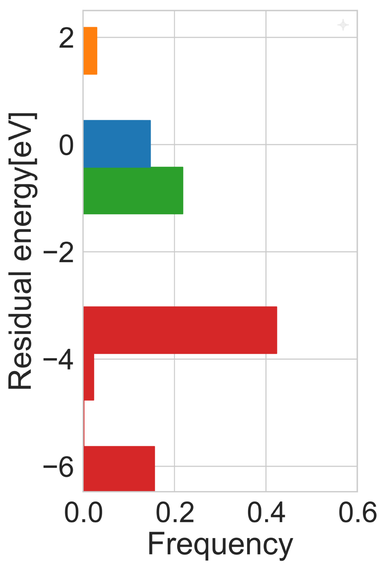
3) Interatomic potential (Stillinger-Weber)

$$E(\vec{r}_1, \ldots, \vec{r}_N) = \sum_i E_1(\vec{r}_i) + \sum_{i,j} E_2(\vec{r}_i, \vec{r}_j) + \sum_{i,j,k} E_3(\vec{r}_i, \vec{r}_j, \vec{r}_k)$$

ANNEALING MACHINE



LOCAL OPTIMIZATION



» Higher probability for locally optimized configurations with lower energy despite higher energy on discretized lattice.

» Simultaneous optimization for both configuration and density.

» Identify minima beyond the initial discretization, minimizing the significance of discretization choice.

OUTPUT CONFIGURATIONS

Top: Input to the algorithm, 1) dimensions and dimensions of unit cell together with mesh size for discretization 2) the atom species to put on the grid points of the lattice and 3) a parametrized interatomic potential.

Middle: The histogram of returned energies over 1000 SA runs, together with example configurations of the peaks on the discretized lattice. The 2H and 1T configurations correspond to the physical ground state and first excited state, the Mo_4S_8-C and Mo_5S_{10} states are local minima of the potential but not physical states and as such are artifacts of the potential. In particular the Mo_5S_{10} local minimum has a different density highlighting the density optimization aspect of our algorithm.

Bottom: The histogram after applying BFGS on the configurations returned from SA. The frequencies reflect the locally optimized with the lowest having the highest probability, and shows that our algorithm correctly identified locally optimized minima with optimal density (even though in this case it is not physical).

Figure: Outline of the algorithm together with results from a MoS_2 monolayer calculation where only the ratio of atoms is fixed.

LJ-Cluster: Following the algorithm outline of the left figure with the Lennard-Jones pair potential as the underlying potential we also optimize for the structure of a Krypton cluster where the ground state is the FCC configuration. In particular we use no penalty.

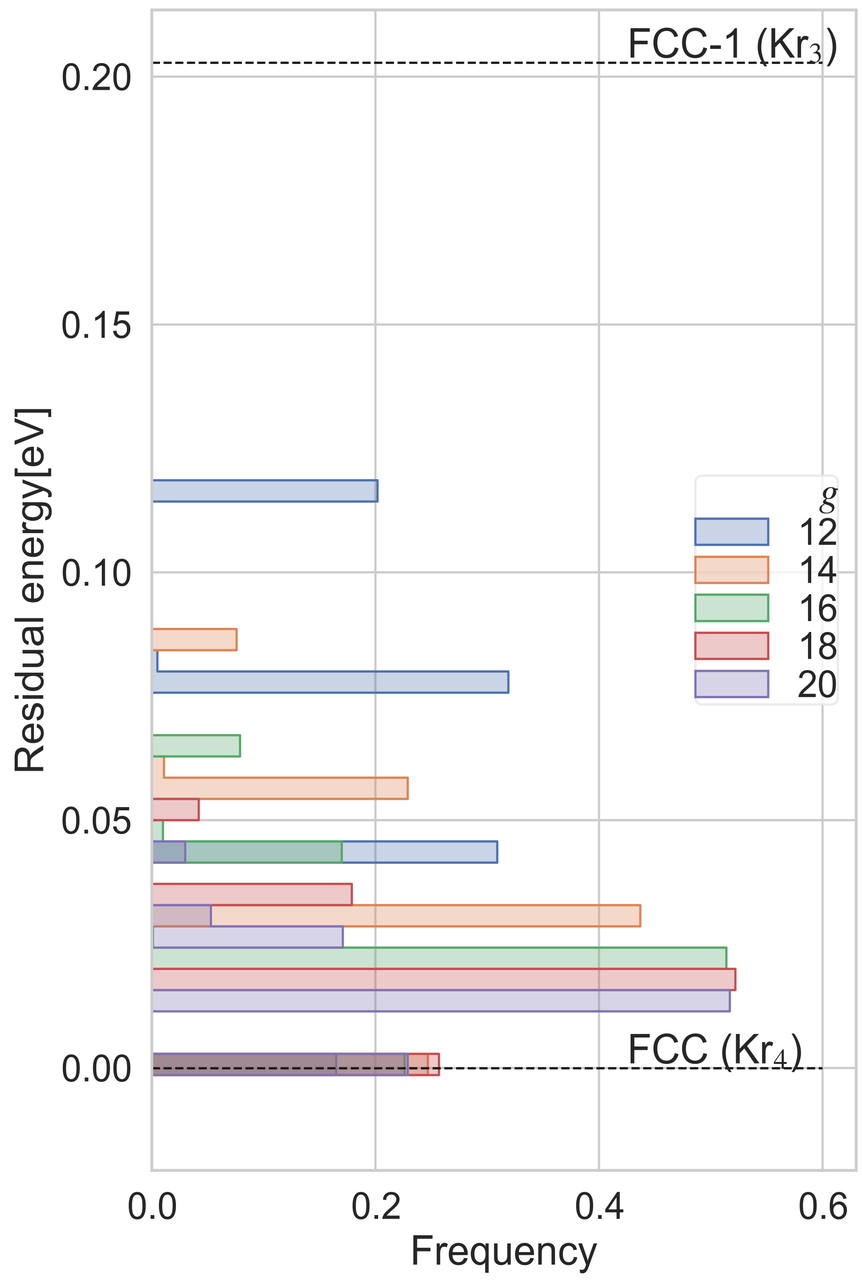


Figure: Energy histogram at the "Annealing Machine" stage of the algorithm for different mesh sizes g with the ground state (FCC) and local minimum (FCC-1) marked by dotted lines. Performing local optimization leads to only FCC states.

» All configurations have correct density.

» Mesh size has no impact on difficulty.

Conclusions

- Lennard-Jones allows optimization of density in absence of any penalty.
- Difficulty of solving the CSP does not scale with the mesh size.
- Successfully reproduces ground states, even when not present in the discretization.
- Note: A related proposal for ionic crystal QUBO has been published during our work on this [2].

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References:

[1] Y. Couzinie, Y. Nishiya, H. Nishi, T. Kosugi, H. Nishimori, Y. Matsushit,. *Annealing for prediction of grand canonical crystal structures: Efficient implementation of n-body atomic interactions.* arXiv preprint arXiv:2307.03123 (2023).

[2] V. V. Gusev, D. Adamson, A. Deligkas, D. Antypov, C. M. Collins, P. Krysta, I. Potapov, G. R. Darling, M. S. Dyer, P. Spirakis, and M. J. Rosseinsky, *Optimality guarantees for crystal structure prediction*, Nature 619, 68 (2023).