Annealing for prediction of grand canonical crystal structures

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Abstract

We propose an annealing scheme usable on modern Ising machines for crystal structures prediction (CSP) by taking into account the general n-body atomic interactions between atoms. The crystal structure is represented by discretizing a unit cell and placing binary variables which express the existence of an atom on every grid point. The resulting higher-order unconstrained binary optimization (HUBO) problem is solved using simulated and quantum annealing. Using the example of Lennard-Jones clusters we show that we can simultaneously optimize both the particle density and the configuration. We show for a covalently bonded monolayer MoS2 crystal that we can simultaneously optimize for the particle density as well as the crystal structure using simulated annealing. We also show that we reproduce ground states of the interatomic potential with high probability that are not represented on the discretized cell.

1. Introduction

CSP from chemical composition alone is still one of the most difficult problems in materials science, due to the exponential scaling with the system size. In this work we propose a scheme to encode CSP problems into a HUBO form usable on modern Ising machines and do a first analysis using quantum and simulated annealing (QA resp. SA). We show that the algorithm performs well for simultaneous optimization of both density and configuration.

2. Theory

Discretize a given unit cell into a set of lattice points X and consider the CSP of optimally placing atoms of species S onto this lattice, where optimality is given by minimizing the energy of a given interatomic potential. Introduce a set b_x^s of binary variables which express the existence ($b_x^s = 1$) or non-existence ($b_x^s = 0$) of an atom of type s on x. For an order-M potential it is then possible to calculate coefficients $V_M^{s_1,\dots,s_M}(x_1,\dots,x_M)$ such that for example in the case $M = 3$ the cohesive energy of the configuration encoded by $\{b_x^s\}_{x,s}$ is calculated by the Hamiltonian

$$H = \sum_{\substack{x\in X\\ s\in S}} V_1^s(x) b_x^s + \frac{1}{2!}\sum_{\substack{x_1,x_2\in X\\ s_1,s_2\in S}} V_2^{s_1,s_2}(x_1,x_2) b_{x_1}^{s_1} b_{x_2}^{s_2} + \frac{1}{3!}\sum_{\substack{x_i\in X\\ s_i\in S}} V_3^{s_1,s_2,s_3}(x_1,x_2,x_3) b_{x_1}^{s_1} b_{x_2}^{s_2} b_{x_3}^{s_3}.$$

This Hamiltonian generalizes directly to order M potentials. Solving the CSP is then equivalent to finding a minimizing binary string, a task which modern Ising machines excel at. Note that optimizing the binary string means that we optimize for both the optimal density (number of 1s

in the string) and the configuration (placement of the 1s). By adding a positive function $f(\{b_x^s\})$ that takes large values if there is not a set target number of 1s in the input binary string it is possible to penalize configurations that do not have a target density, or ratio of species.

3. Results and discussion

We look at a Krypton Lennard-Jones cluster and covalently bound hexagonal MoS_2 monolayer.

3.1 Krypton system

The Lennard-Jones pair potential HUBO for the Krypton system is solved on D-Waves Advantage 4.1 Quantum annealer. The main finding (Fig. 1) is that simultaneous density optimization performs similar to the fixed density one, in terms of the probability of finding the optimal configuration (~20% probability after local optimization with Broyder-Fletcher-Goldfarb-Shanno (BFGS)).

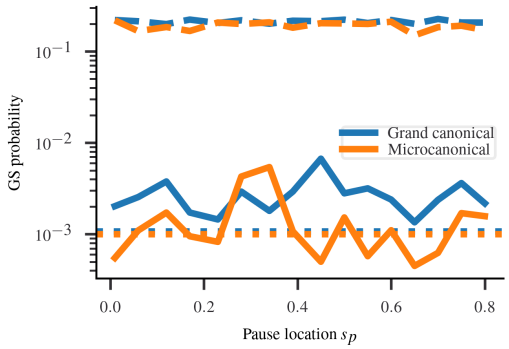

Fig.1 Ground state probability results for the optimization of a Krypton primitive cell using QA (dotted), QA+pausing (solid) and QA+pausing+BFGS (dashed) plotted against the pause location. Grand canonical is with density optimization, microcanonical without.

3.2 Hexagonal MoS_2 monolayer system

For the three-body interactions in MoS_2, we find with SA (Fig. 2) that both with and without fixing the density we find the ground state 2H and 1T configurations in a minority of cases. Using BFGS we see that the states that do not correspond to 2H or 1T converge to a configuration that has even lower energy. We call these lower energy states Mo_5S_{10} and Orthorombic. These are artifacts of the interatomic potential and do not correspond to physical reality. The main finding is, that while the 2H and 1T configuration are encodable on our discretized unit cell, the other two are not and thus our scheme managed to find the true ground state of the interatomic potential despite them not being the expected density or on the discretized primitive cell.

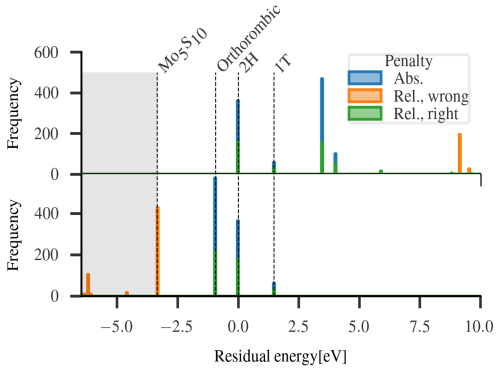

Fig.2 Histogram of obtained configurations using fixed ratio of species (Rel. penalty) or fixed number of atoms (Abs. penalty). The top histogram is SA while the bottom is SA+BFGS. The orange energies have the wrong densities while the green ones the correct density.

4. Summary

We presented an algorithm to solve the CSP on modern Ising machines. We optimize for both the density and configuration at the same time so that we need not specify atom numbers, a distinct advantage over existing algorithms. Further we have provided evidence that the discretization, necessary to encode the problem for Ising machines, is not detrimental to the quality of results received, by showing that we find ground states outside the discretized lattice. This algorithm could pave the way for applications of quantum computing in materials science.

References

1) Y. C., Y. Nishiya, H. Nishi, T. Kosugi, H. Nishimori and Y. Matsushita, “Annealing for prediction of grand canonical crystal structures: Efficient implementation of n-body atomic interactions”. *arXiv:2307.03123 (2023)*