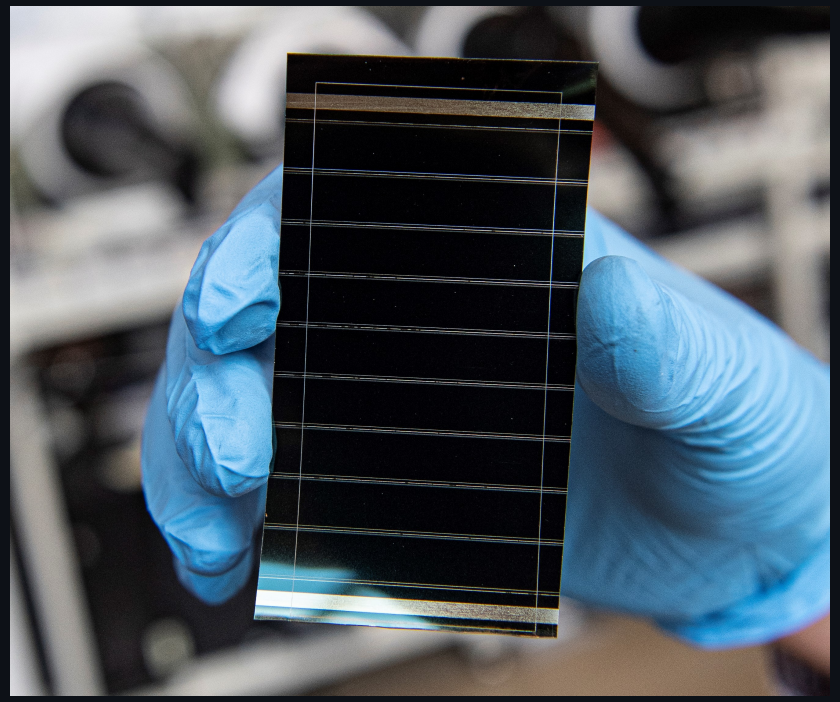
Optimizing Lattice Energy in Irregular CM

Ethan Meltzer

UToledo REU 2023

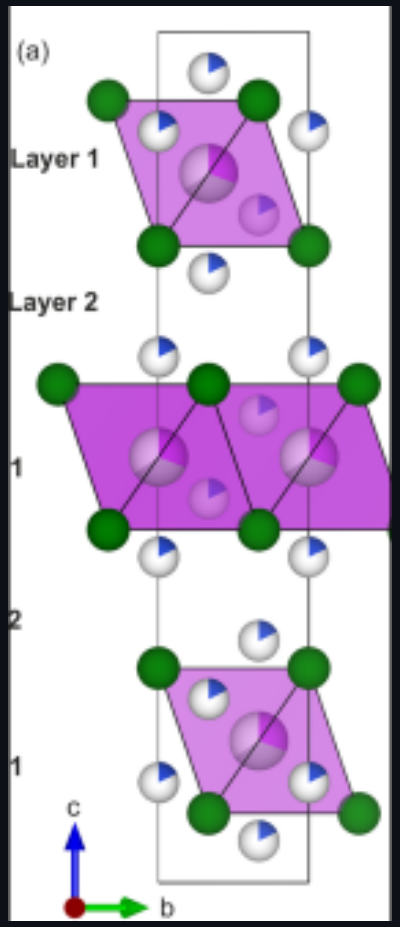
Background

- Traditional solar panels use lead-based semiconductors (perovskites) like $\mathrm{CH_3NH_3PbX_3}$
- ((Lead is bad)), and solar panels are only going to be more in demand



https://www.energy.gov/eere/solar/perovskite-solar-cells

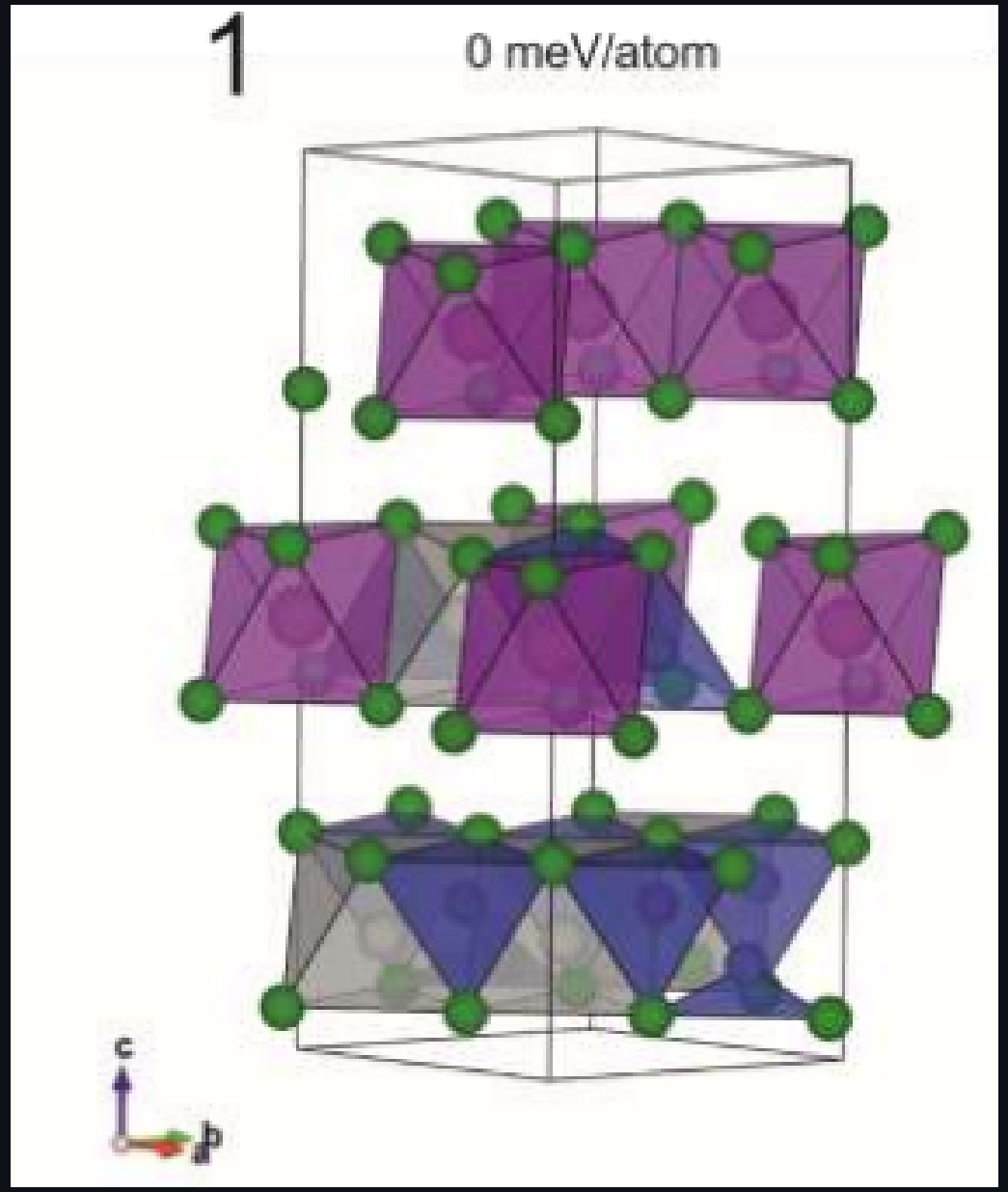
- Oct 2021, new research from University of Liverpool reports synthesis of lead-free semiconductor promising for photovoltaics, $\mathrm{Cu_2AgBiI_6}$
- Paper reports irregular internal structure in addition to electronic properties, structure was analyzed using ChemDASH/VASP: serial minimization, slow
    - Parallelizing could mean more accurate results
- Worth researching: helps us understand why it's effective, could make commercial synthesis easier in future



Journal of the American Chemical Society 2021 143 (10), 3983-3992 DOI: 10.1021/jacs.1c00495

The Goal

- Use *Vienna Ab-initio Simulation Package* (VASP) to simulate a 9-cell chunk of crystal using electron density functionals (DFT)
- Explore different feasible configurations of the crystal
- Determine which configurations result in the lowest lattice energies
- Verify that those configs produce similar electronic properties to measurements made in lab



Journal of the American Chemical Society 2021 143 (10), 3983-3992 DOI: 10.1021/jacs.1c00495

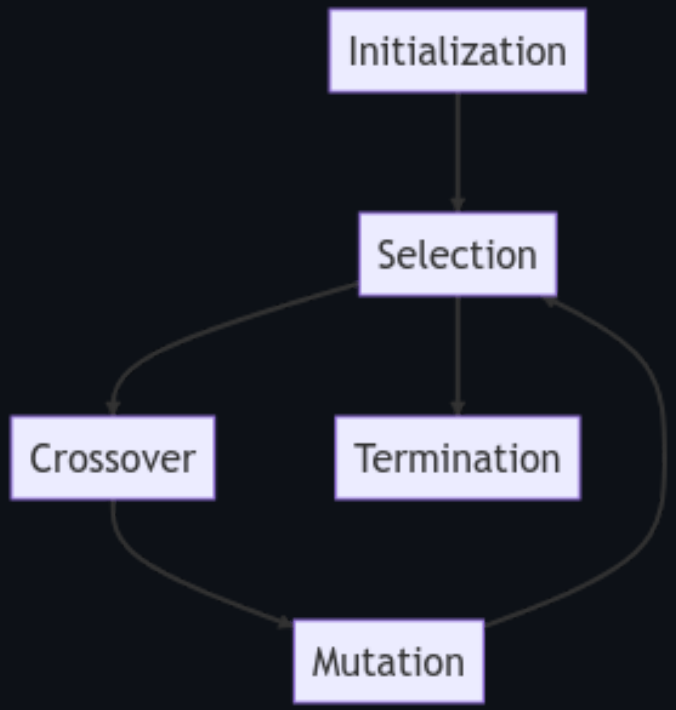
The problem

How can we find the global minimum of a discrete function with unpredictable behavior and a very large finite domain?

$$\binom{108}{18}\mathrm{Cu} * \left[\binom{27}{9} + \binom{18}{9}\right]\mathrm{Ag/Bi} = 6.595 * 10^{26}\ \text{configurations}$$

Big number

- $\mathrm{TB} = 10^{12}$ bytes, would need 600 trillion 1 TB storage drives to represent each configuration with 1 bit
- In meters, roughly the diameter of the observable universe 🪐🌌

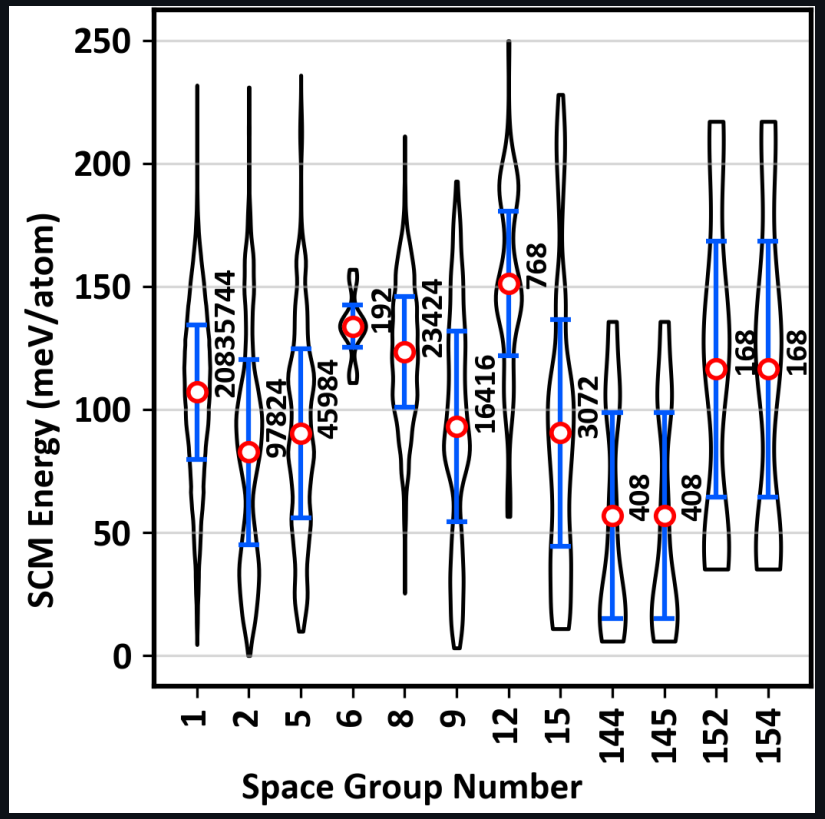


My approach

- Genetic algorithim converges on minimum while only looking at a small sample of the possible configurations
- Easily parallelizable, can run as many simulations as CPU threads available at once
- Use in this field is an active research area, especially with irregular materials

First generation

- Some correlation between crystal symmetry/regularity and lattice energy in similar materials, many exceptions
- Skewing the first generation of configurations towards highly symmetric (high space group) configurations could lead to a faster convergence on a minimum.
- Accomplish this by binning the randomly generated first generation
  - Each bin is equal size but
  - Number of space groups represented in each bin varies



Generating child configurations

- Configs for future generations are generated from the best of the previous generation
- Crossover: site filling is determined by indexing the occupied sites of the parents and selecting at random
- Mutation: A few occupied atomic sites in the child configuration will be swapped with ones chosen at random, most often resulting in an atom switching with a vacancy.