**Ferroelectric 2D van der Waals materials based on CuInP2Se6 (CIPSe)**

A diagram of a molecule

Description automatically generatedStructurally this material consists of layers of edge-sharing Se6 octahedra separated by a van der Waals gap. The cations sit inside the Se6 octahedra as Cu(1+) and In(3+) cations, and as dimeric P2(8+) units where each P has a formal charge of 4+. In the ferroelectric phase, each Cu cation displaces from the center of each Se6 octahedron by approximately 1.2 to 1.4 Angstroms along the c-axis, while the In cation displaces in an opposite manner along the c-axis by approximately 0.1 Angstroms. This creates a polarization, which is switchable by an applied electric field. Neither the P cation or Se anion displace much. The general structure type for CIPSe, AgInP2S6, is depicted in Figure 1.

**Figure 1:** the CIPSe structure is classified in the AgInP2S6 structure prototype, where the pink sphere is the 3+ cation (In) and the large silver sphere is the 1+ cation (Cu).

CIPSe has the following crystallographic parameters. It is a hexagonal unit cell with a=b=6.392 Angstroms, c=13.3380 Angstroms, and alpha=beta=90 degrees, while c = 120 degrees. It is classified in space group P3bar1c (#163) with the following reduced coordinates:

Cu (2d) 2/3 1/3 ¼  
In (2a) 0 0 ¼  
P (4f) 1/3 2/3 0.1655  
Se (12i) 0.0095 0.3401 0.1201

This means that there are 20 atoms in the unit cell and that we need to use symmetry to come up with the remainder of positions. A typical set of coordinates looks like this in our DFT simulations:

ATOMIC\_POSITIONS (crystal)

Cu 0.666665143 0.333334857 0.250000000

Cu 0.333334857 0.666665143 0.750000000

In -0.000001293 0.000001293 0.250000000

In 0.000001293 -0.000001293 0.750000000

P 0.333332479 0.666667630 0.166077293

P 0.333332370 0.666667521 0.333922707

P 0.666667521 0.333332370 0.833922707

P 0.666667630 0.333332479 0.666077293

Se -0.007906859 0.330413064 0.122599907

Se 0.669587753 0.661683099 0.122599849

Se 0.338316087 1.007906856 0.122599738

Se 0.669586936 1.007906859 0.377400093

Se 0.338316901 0.330412247 0.377400151

Se -0.007906856 0.661683913 0.377400262

Se 1.007906859 0.669586936 0.877400093

Se 0.330412247 0.338316901 0.877400151

Se 0.661683913 -0.007906856 0.877400262

Se 0.330413064 -0.007906859 0.622599907

Se 0.661683099 0.669587753 0.622599849

Se 1.007906856 0.338316087 0.622599738

What we would like is a) to generate random and scaled displacements for these 20 atoms that would also include ferroic and antiferroic patterns that can be pasted into an input file for a single point energy calculation. The other piece we would need is a b) slight variation in the c/a ratio for this hexagonal structure, where a and c/a are represented like this in the input file:

celldm(1) = 11.89280,

celldm(3) = 2.10809,

All of this would need to c) be pasted into Quantum Espresso input files and then each run as a self-consistent field single point energy calculation. We would need on the order of 1000 total scf calculations for each ABP2Se6 composition, so d) automating the submission with unique names is also necessary. The first 800 would be used to train a machine learning model that could predict the ground state structure of CIPSe, and we could put 8 to 10 into each 24 hour runscript. The remaining 200 would be used to test and verify/refine the model. The model and its ingredients are still TBD, but we are circling PyTorch.

We also have structure types that do not fit our criteria, and could feed them into the model as well, to obtain total energies of materials that we don’t want.

Ultimately, we would want a machine learning model for use in any unknown composition to sidestep having to do 100s of DFT simulations to find a ground state.

For now we need python to do the following:

1. Generate random and scaled displacements for a 20 atom unit cell
2. Generate scaled a and c/a values to accompany the random displacements
3. Create input files for Quantum Espresso
4. Submit the DFT calculations (8 to 10 at once. Give each unique names).