# Predicting thermal effects in optoelectronic properties of solid solutions with crystal graph neural networks

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Characterizing the optoelectronic properties of inorganic semiconductor materials with first principles methods such as Kohn-Sham Density Functional Theory (DFT) is crucial for optoelectronic and energy applications, for example photovoltaics or microelectronic devices. Using DFT and hybrid exchange-correlation functionals is usually sufficient to obtain reliable band gap and optical properties values in accordance with experiments, but this methodology requires significant supercomputer simulation time, even for crystals structures with few atoms in the unit cell. Thus, when trying to study the optoelectronic properties of solid solutions, since these compounds feature unit cells with a large number of atoms (hundreds or thousands), standard DFT computations are not feasible and other methodologies, usually less accurate, should be used. In this work we are interested in studying the solid solutions of a novel family of inorganic materials, the chalcohalide anti-perovskite compounds (CA) [1]. These compounds present the following chemical expression $Ag_3BC$ (where B=S, Se, and C=Br, I) and they are expected to be great candidates for energy applications [2], helping with the transition towards a more sustainable and greener future. We previously characterized the optoelectronic properties of the pure compounds of the family with first principles methods, revealing giant electron-phonon interactions effects (thermal effects) [3] in the band gap values. In order to overcome this obstacle and achieve a good agreement with experiments, we performed several ab-initio molecular dynamics simulations (AIMD) at various temperatures, and computed the thermally re-normalized band gap as the mean band gap of different uncorrelated states reached with the AIMD. Proceeding this way, the simulations took one hundred times more computational time than without taking into account the electron-phonon interactions. Therefore, it is impossible to take into account the thermal effects in the optoelectronic properties of CA solid solutions using standard DFT methodologies. Here is where advanced machine learning methodologies could help us to study such a complex problem. We propose using crystal graph neural networks (CGNN) [4]. These are graph neural networks that can be used to predict properties of crystal structures. The unit cell of a material can be mapped to a graph structure that encodes the periodicity of the unit cell as well as the main ion chemical properties (atomic number, atomic mass, ionic radius, etc.) and chemical bonding information (Euclidean length of the bonding). Fig.1 represents this. Using data from the Materials Project database and our own DFT results we trained a model capable to predict band gaps for CA solid solutions, without taking into account thermal effects. In order to consider these effects, we decided to apply the same strategy that we used for pure compounds. As it would not be possible to perform AIMD of the solid solutions due to their big unit cells, we decided to use M3GNet [5] to perform classical molecular dynamic simulations with machine learning interatomic potentials (MLMD). In order to ensure good performance of the MLMD we re-trained M3GNet with our DFT data. Computing the band gaps with CGNN of several states reached with MLMD, we were able to obtain thermally re-normalized band gaps for CA solid solutions consistent with the experiments.

## References

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## Figures

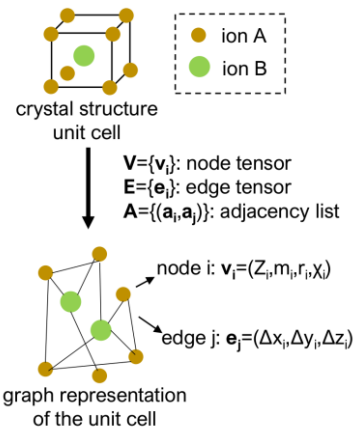


**Figure 1.** Representation of the transformation of a given unit cell of a material into a graph encoding chemical information.