Thermal Effects on Optoelectronics: A Graph Neural Network Approach

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1. Introduction

The effects of temperature on the optoelectronic properties of semiconductor materials have been extensively studied for many years [1, 2]. Understanding these effects is crucial for the development of optoelectronic and energy-related applications, such as photovoltaic devices and LEDs.

Temperature-induced lattice vibrations lead to electron-phonon coupling (EPC), which not only impacts the optoelectronic properties of polar materials but also plays a key role in other fundamental physical phenomena, such as conventional superconductivity and carrier mobility [3].

First-principles methods, such as Density Functional Theory (DFT), are widely used to compute the optoelectronic properties of materials. However, systems exhibiting strong EPC and anharmonic behavior often require more sophisticated and computationally expensive approaches [4]. This challenge is further exacerbated in complex materials, such as solid solutions, where unit cells can contain hundreds or even thousands of atoms.

In this work, we propose leveraging machine learning techniques to model the effects of temperature on the optoelectronic properties of anharmonic semiconductor systems, as illustrated in Fig. 1a. As a case study, we investigate silver chalcohalide antiperovskites (CAP), e.g., Ag_3SBr and Ag_3SI, which exhibit pronounced anharmonicity and strong EPC [5, 6].

2. Methods

We employed crystal graph convolutional neural networks (CGCNN) [7] and *ab initio* molecular dynamics (AIMD) simulations to determine the temperature dependence of the band gap. After performing AIMD calculations at a given temperature T, the renormalized band gap is computed with [8]:

$$E_g(T) = \lim_{t_0\to\infty} \frac{1}{t_0} \int_0^{t_0} E_g^{\mathbf{R}(t)} dt, \qquad (1)$$

where $\mathbf{R}(t)$ represents the atomic positions at time t in the AIMD simulation at T.

The band gaps of the structures corresponding to $\mathbf{R}(t)$ are computed using a CGCNN model. This model is initially trained on the band gap dataset from the Materials Project and subsequently retrained on a DFT-based dataset specific to our study.

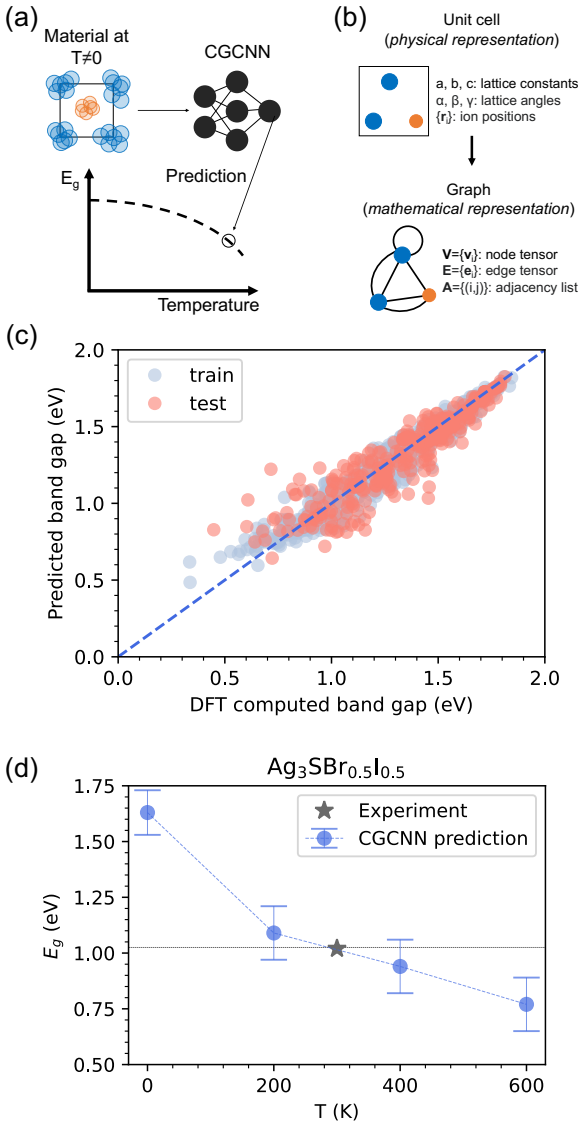

Fig. 1: (a) Schematic illustrating how our machine learning model predicts the band gap of a material at a given temperature. (b) Comparison between the unit cell representation and the graph representation of a material. (c) Predicted band gaps after retraining our graph neural network, compared with Density Functional Theory (DFT) calculations. (d) Band gap prediction for the CAP solid solution $Ag_3SBr_{0.5}I_{0.5}$ using our CGCNN model. The experimental value at $T = 300$ K is shown for reference.

To make predictions using CGCNN, the material's structure must be represented as a graph, as illustrated in Fig. 1b.

Additionally, molecular dynamics simulations using machine learning interatomic potentials (MLIP) [9] can serve as an alternative to AIMD, enabling the study of larger systems across a wider temperature range.

3. Results

The retrained CGCNN model achieved a mean absolute error (MAE) of less than < 0.1 eV in band gap prediction for both the training and test sets (Fig. 1c). The DFT-computed structures were generated using a Monte Carlo approach, incorporating uniform noise into the original structure as well as phononic noise at different temperatures to approximate the configuration space explored during molecular dynamics simulations.

Our model successfully reproduced a temperature-induced band gap reduction of several hundred meV in CAP compounds, in agreement with DFT computations. Moreover, it accurately captured the band gap reduction with temperature for a more complex system, the solid solution $Ag_3SBr_{0.5}I_{0.5}$, Fig. 1d, demonstrating excellent agreement with experimental observations [10].

4. Conclusions

In this work, we demonstrated the effectiveness of machine learning models, specifically graph neural networks, in predicting the temperature dependence of the band gap in anharmonic semiconductor systems. Our approach successfully reproduced the temperature-induced band gap reduction in CAP compounds and extended its predictive capability to more complex solid solutions, showing excellent agreement with experimental data.

These results highlight the potential of machine learning techniques as a powerful alternative to traditional computational methods, enabling efficient band gap predictions across a wide range of temperatures.

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