Hessisches Kompetenzzentrum
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Electronic Structure Calculations on the Influence of Alkali Metals on the Properties of Ag-Containing Cu(In, Ga)(S, Se)$_2$ Absorber Materials

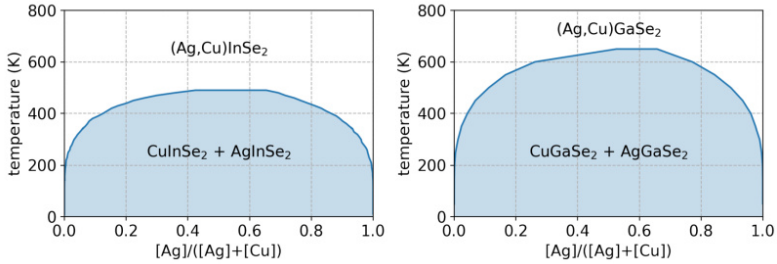

Figure 1: The phase diagrams of (Ag, Cu)InSe$_2$ and (Ag, Cu)GaSe$_2$ calculated using semi-grand canonical Monte Carlo simulations.

Introduction

The compound semiconductor system Cu(In, Ga)(S, Se)$_2$ (short: CIGS) forms the basis for the currently most efficient thin-film solar cells with record laboratory efficiencies in the range of 22% to 23%. In general, Ga and S are used to optimize the band gap in the absorber. An alternative way of increasing the band gap is to substitute Ag for Cu. Ag-containing CIGSe cells that were produced by co-evaporation were able to achieve efficiencies of up to 21% under laboratory conditions. The main goal of this project is the examination of the influence of Cu substitution by Ag on the electronic and structural properties of CIGS solar cells absorber materials. For this objective density-functional theory (DFT) calculations with the VASP software will be used to parameterize a cluster expansion model. The cluster expansion model can then be used in semi-grand canonical Monte Carlo simulations to investigate the phase diagram of the system. The parameterization of the cluster expansion model requires an extensive set of training structures to learn the relevant interactions within the system.

Methods

In this project we employed density-functional theory (DFT) calculations to train a cluster expansion model. These calculations were done using the software package VASP, which is already successfully in use on the Lichtenberg cluster as well as in our group and has proven to be a reliable and high-performance code. The training set calculated using VASP is going to be used to train a cluster expansion model using the ICET software package (https://icet.materialsmodeling.org/index.html).

Project Manager
Dr. Markus Mock

Principal Investigator
Prof. Dr. rer. nat. Karsten Albe

Project Term
2021 - 2022

Clusters
Lichtenberg Cluster Darmstadt

Software
VASP

Additional Software
ICET

Institute
Fachgebiet Materialmodellierung

University
Technische Universität Darmstadt

JUSTUS-LIEBIG-UNIVERSITÄT GIESSEN
TECHNISCHE UNIVERSITÄT DARMSTADT
UNIKASSEL VERSITÄT
Philipps Universität Marburg

Results

A cluster expansion (CE) model was constructed based on a database of 242 training structures in the (Ag, Cu)$InSe_2$ case and 108 training structures in the (Ag, Cu)$GaSe_2$ case. The training structures were generated by enumeration of possible arrangements of Ag and Cu on the (Ag, Cu)(In/Ga)Se_2 lattice. The software package ICET was used to fit the cluster expansions to this database. These cluster expansions were used in the following sections to perform semi-grand canonical Monte Carlo simulations to investigate the phase diagram of the system. Figure 1 finally shows the comparison between the phase diagrams of (Ag, Cu)$InSe_2$ and (Ag, Cu)$GaSe_2$. Both show a similar behaviour with a solid solution at elevated temperatures and phase seperation at low temperatures. The upper consolute temperature of 650 K is higher for (Ag, Cu)$GaSe_2$ compared to the 490 K for (Ag, Cu)$InSe_2$.

Discussion

In this work we examined the influence of Cu substitution by Ag on the electronic and structural properties of CIGS solar cells absorber materials. We parameterized a cluster expansion model of the (Ag, Cu)$InSe_2$ and of the (Ag, Cu)$GaSe_2$ system based on density-functional theory calculations. The cluster expansion model was then be used in semi-grand canonical Monte Carlo simulations to investigate the phase diagrams of both systems, showing that the system has a tendency to phase separate at low temperatures, while it shows a solid solution at high temperatures. The transition temperature between these states is significantly higher for (Ag, Cu)$GaSe_2$ compared to (Ag, Cu)$InSe_2$.

Last Update: 2023-02-13 17:21