Hessisches Kompetenzzentrum
für Hochleistungsrechnen

Fermi Level Engineering of Doped Perovskites: From the Dilute Limit to Phase Formation



Project Manager
Rachele Sciotto

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

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Project Areas
Theoretical Condensed Matter Physics

Clusters
Lichtenberg II Cluster Darmstadt

Software
VASP

Institute
Fachgebiet Materialmodellierung

University
Technische Universität Darmstadt

Introduction

Mixed ionic–electronic conductors (MIECs) can transport both ions and electronic charge carriers, making them key materials for electrochemical energy conversion devices such as solid oxide fuel cells (SOFCs). In SOFCs, chemical energy is directly converted into electricity, bypassing combustion and achieving higher efficiency than conventional methods. Unlike batteries, fuel cells operate continuously as long as fuel and oxidant are supplied. These fuel cells offer a clean and efficient way to convert chemical energy into electricity, making them highly relevant in the shift toward sustainable energy systems. In this project, we aim to understand and predict the behavior of specific atomic defects in barium indate ($Ba_2In_2O_5$, BIO), a promising material for solid oxide fuel cells. In the low-temperature orthorhombic phase of BIO, vacancy ordering restricts oxygen mobility. At higher temperatures, BIO undergoes an order–disorder transition that enhances ionic conductivity. A major challenge in optimizing these materials lies in understanding how certain defects, such as oxygen interstitials, contribute to conductivity.

Methods

Our results show that six distinct oxygen interstitial configurations are stable in BIO. Among these, only two have comparatively low formation energies and therefore play a significant role in the defect chemistry of the material. In addition to the expected negatively charged oxygen interstitial,

we identify a neutral, dumbbell-shaped interstitial configuration, in which the interstitial oxygen forms a bond with a neighboring lattice oxygen atom. As expected, barium and indium vacancies behave as acceptors and display low formation energies under n-type conditions. Using the calculated defect formation energies, we estimated the defect concentrations as a function of oxygen partial pressure. The concentrations of oxygen vacancies and negatively charged oxygen interstitials were found to reach values on the order of 10^{18} cm^{-3}. At high oxygen partial pressures, the neutral dumbbell oxygen interstitial is also predicted to occur in substantial concentrations.
In addition, we initiated the development of a machine learning interatomic potential for BIO. This potential will enable simulations at finite temperatures to investigate dynamic processes such as oxygen diffusion over a longer timescale and at different temperatures.

Results

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Discussion

Our results indicate that oxygen-related defects dominate the defect chemistry of BIO. The presence of a neutral interstitial suggests that more complex mechanisms may also play a role in oxygen transport. To further elucidate the role of these defects in oxygen transport, future work will focus on calculating oxygen migration barriers using the Nudged Elastic Band (NEB) method. In parallel, the development of a machine-learning interatomic potential will enable the investigation of oxygen diffusion mechanisms at realistic operating temperatures. Together, these approaches will provide a more comprehensive understanding of oxygen mobility in BIO and of how it is influenced by the underlying defect chemistry.

Publications

Rachele Sciotto: Fermi-level pinning by oxygen interstitials in $Ba_2In_2O_5$? Insights from ab initio defect chemistry, European Materials Research Society (EMRS) Spring Meeting, Strasbourg (France), 26-30 April

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