First-Principles Studies of Perovskite Compounds for Thermoelectric Applications

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Computational modeling techniques have been used to provide detailed insights into the structure-property relationships of materials. We have collaborated with the National Institute of Standards and Technology (NIST) to develop various perovskite compounds with desired thermoelectric properties. We have used density functional theory-based approaches to study structural stability and electrical properties of \( R_2(\text{FeCo})_O_6 \) perovskite compounds \((R = \text{Pr, Nd, Sm, Eu and Gd})\), for which Fe and Co randomly occupy the B-site. Superlattice and locally disordered phases have been compared through a total energy minimization approach.

**Introduction**

**Methods**

Vienna ab-initio simulation package (VASP) has been performed to optimize geometry and structure. Due to the ferromagnetic natures of Fe and Co, we have supplied initial magnetic moments of \( 4\mu_B \) and \( 1\mu_B \) respectively. Additional input parameters include a conjugate electronic minimization algorithm and a 400 eV cutoff energy for plane-wave basis set. In order to study electronic properties, DFT+U has been used to account for d and f orbitals of certain elements.

Below are theoretical lattice parameters, cell volumes, and bond lengths for \( \text{Pr}_2(\text{FeCo})_O_6 \) options as well as experimental values from a TEM. The local density of states (LDOS) is shown for stable disordered structure II.

**Results**

**Discussion and Conclusions**

- Increasing disorder yields lattice parameters and bond lengths similar to experimental values.
- Total energy minimization calculations indicate that disordered phase of \( \text{Pr}_2(\text{FeCo})_O_6 \) is more stable than its superlattice, agreeing well with experimental observation.
- Overlapping peaks in the DOS plot suggest bonding covalency in the \( \text{R}_2(\text{FeCo})_O_6 \) structure.

Our electronic structure calculations suggest that \( \text{Pr}_2(\text{FeCo})_O_6 \) is semiconducting with an electronic band gap of 0.43 eV.

**Material Application: Thermoelectrics**

- Thermoelectric (TE) effect: direct conversion of temperature differences to electric voltage
- TE figure of merit: \( ZT = S^2\sigma\tau/k \), where \( S \) is the Seebeck coefficient, \( \sigma \) is the electrical conductivity, \( T \) is the temperature and \( k \) is the thermal conductivity
- TE material uses: power generation and refrigeration
- Factors affecting TE advancement: cost, thermal stability, and toxicity [2]

**References**


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