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論 文 要 旨

THESIS SUMMARY

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要旨（英文 800 語程度）
Thesis Summary（approx.800 English Words ）

First-principles calculations have been widely used for ideal crystals to design functional materials. However, various defects, which breaks the three-dimensional periodicity of ideal crystals, exist in real materials and govern their physical properties. Usually, static defects such as substitutions, vacancies, interstitials, impurities, and their complex defects are focused on, while dynamical motions of atoms, i.e., phonons, are also regarded as important defects. Herein I focus on these static and dynamical defects in this thesis to design real materials. In this study, I performed the first-principles design of electronic and thermal functional materials with both the statistic and dynamical defects on the material systems of the layered alkaline earth transition metal nitrides (*AETM*N_2), the ultra-widegap alkaline earth oxides (*AE*O), strontium titanate ($SrTiO_3$), and tin selenide (SnSe).

Chapter 1 General introduction and **Chapter 2 Computational methods**

The background and objective of this study, and the main computational methods including defect formation enthalpy calculations and phonon transport calculations are described.

Chapter 3 Design of high-purity layered nitride semiconductor, *AETM*N_2 (*AE* = Ca, Sr, Ba and *TM* = Ti, Zr, Hf)

$SrTiN_2$ is expected to exhibit unique carrier transport properties since it has a two-dimensional (2D) electron transport layer in its natural layered crystal structure and its conduction band minimum mainly consists of Ti 3d_{xy} orbitals similar to that of the $SrTiO_3$-based 2D electron gas system. However, only nearly metallic conduction has been reported for $SrTiN_2$, which is probably due to the difficulty in synthesizing the high purity and low defect density samples. In this chapter, I theoretically clarified that the issue of the unintentional high carrier generation in layered $SrTiN_2$ semiconductor would originate from the incorporation of the oxygen (O) impurities at the nitrogen (N) sites even at the N-rich condition. On the other hand, I clarified that alkaline earth (*AE*) ion and transition metal (*TM*) ion substitutions are effective to suppress the incorporation of O impurities in *AETM*N_2. The much stronger Ca–N and Hf–N bonds significantly suppress the incorporation of O impurities in $CaTiN_2$ and $BaHfN_2$, which would exhibit more intrinsic semiconductors with *TM* d_{xy} 2D bands.

Chapter 4

Unpublished

Chapter 5

Unpublished

Chapter 6 Heavy hole doping and thermal conductivity reduction of SnSe by isovalent Te ion substitution

SnSe exhibits the world record of thermoelectric figure-of-merit *ZT* in the single crystal form, but the performance of polycrystalline SnSe has been restricted by low electronic conductivity and high κ, compared to the single crystal. In this chapter, I estimated the effect of Te doping in the electronic and thermal transport properties of layered SnSe. Generally, isovalent doping does not increase *n* in semiconductors, while isovalent Te substitution at the Se site largely increases *n* in SnSe. I clarified the origin by first-principles calculations, showing that it is easier to form the hole-donating Sn vacancy (V_{Sn}) by the large-size Te doping from the elongated and weak Sn−Te bonds in $Sn(Se_{1-x}Te_x)$ lattice. In addition, I found that the weak Sn−Te bond strength would induce phonon softening and strong phonon scattering, leading to the significant κ_{Lat} reduction in SnSe.

The present study demonstrates that theoretical calculations including defect calculations and phonon calculations can provide a great insight to understand material properties, which would be a powerful approach to design further novel functional materials.