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Understanding the Ultralow Thermal Conductivity and Strong Anharmonicity of a Lanthanum-Based Germanium Halide Monolayer for Possible Thermoelectric Applications

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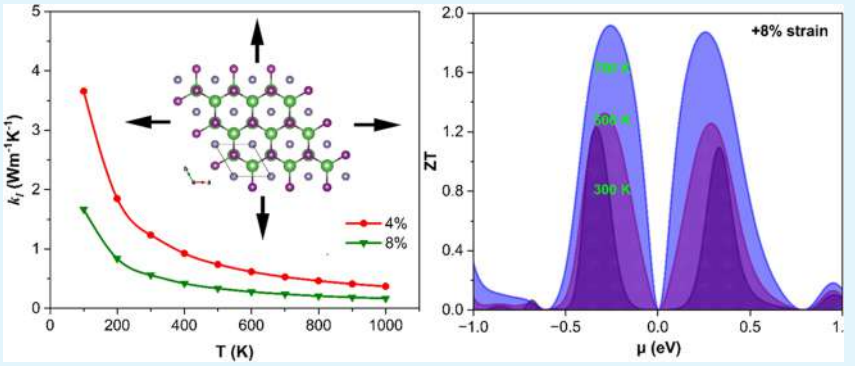

ABSTRACT: Low-dimensional materials outperform their bulk equivalents in terms of thermal and electronic charge transport phenomena. Ultralow thermal conductivity in thermoelectric (TE) semiconductors is rare and plays a crucial role in obtaining promising TE performances. Their performance can be effectively improved via strain engineering, which allows the modulation of geometrical parameters as well as electronic energy levels of a material. With this concept in mind, we systematically studied the effect of biaxial tensile strain on the structure, stability, mechanics, and thermoelectric properties of a novel La_2GeI_2 monolayer by using the hybrid density functional theory and solving Boltzmann transport equations. The strain-induced distortion manipulates the electronic band characteristics with an increase in the band gap, effective mass, and relaxation time of carriers. In principle, La_2Ge is a metal, while the functionalized La_2GeI_2 structure becomes a semiconductor. Two temperature-dependent adsorption structures have been reported in experiments with the $R\bar{3}m$ phase as the most stable ground-state structure. HSE06 calculations predict an indirect gap of 0.69 eV appearing at the Γ−M symmetry points of the Brillion zone in this monolayer. La−Ge bands being prominent around the Fermi level emerge out of p−d covalent hybridization, providing an edge to enhanced conductivities. The calculated transport coefficients and thermal conductivity (k_l) seem to be better than those of available two-dimensional TE materials such as phosphorene, arsenene, etc. We find that a significantly low k_l value (3.22 W/mK) at 300 K can be reduced to an ultralow value of 0.57 W/mK under strain. Owing to the strain-engineered low thermal conductivity, small band gap, significant Seebeck coefficient (∼1100 μV/K), and ZT(∼2), we can rule out the enhanced TE conversion potentials of this monolayer in comparison to traditional TE materials.

KEYWORDS: *2D materials, electronic structure, thermal conductivity, transport coefficients*

INTRODUCTION

2D materials have gained immense interest in recent years due to their multifunctional physical properties and miniature effects (size, quality, cost), making them prospective materials for technological applications.[1−4] Searching for new materials with profound electronic properties has triggered the discovery of new 2D materials from their bulk counterparts. Besides the experimental realization of 2D monolayers or chains, machine learning or density functional theory (DFT)-based simulations have proven to be the fastest accessing tool and one of the most efficient methods to propose novel materials with desired properties.[2−5] Nanosheets/heterostructures of different families are being intensively discovered for 2D TE materials such as 2D sheets of ScX (X═P, As),[6] SnSe,[7,8] MoSSe,[9,10] MoS_2,[11,12] Ti_2VGe,[13] $Cr_2Ge_2Te_6$,[14] Nb_3GeTe_6,[15] Ga_2SeTe,[16] etc. Such novel low-dimensional materials possess promising capabilities to meet the challenges for new technological

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