Stacking-Dependent Anisotropic Thermoelectric Properties of $NbSe_2$ Polymorphs based on First-Principles Band Calculations

Kyoto University of Advanced Science, Graduate School of Engineering

○Mark Edwin Cleofe, Jr., Koichi Nakamura, Tetsuro Habe

E-mail: 2023mm02@kuas.ac.jp

I. Introduction

Interlayer stacking sequences lead to variations in the physical properties of 2D transition metal dichalcogenides (TMDCs) compared to their bulk counterparts. Various configurations can be realized primarily due to weak van der Waals (vdW) interactions in-between the layers [1]. Additionally, the stacking orders can determine crystal symmetry which significantly influences the electronic band dispersions [2]. Layered Niobium Diselenide ($NbSe_2$), one of the emerging TMDCs, has attracted significant attention due to its unique physical properties. However, much of the current literature has focused on $2H_a$, its most stable configuration, leaving behind a limited attention to several metastable structures, such as $2H_c$, 3R, and 2H-3R. Here, we investigate the in-plane and out-of-plane thermoelectric transport properties of $2H_a$, $2H_c$, 3R, and 2H-3R phases of $NbSe_2$, including Seebeck coefficient S, electrical conductivity σ, electronic thermal conductivity κ_e, and thermoelectric power factor $S^2\sigma$.

II. Methods

The optimized crystal and electronic band structures of the various $NbSe_2$ stacking configurations were obtained using density functional theory (DFT). The total electronic energy for each configuration was numerically computed using Quantum ESPRESSO [3]. The calculations are performed using the projector augmented wave method with the Perdew-Burke-Ernzerhof exchange-correlation (XC) functionals. The effects of the vdW interactions were accounted for by incorporating vdw-df-C6 as a correction to the XC functional [4]. The lattice structure and atomic positions were optimized with thresholds set at 1×10^{-5} hartree and 1×10^{-4} hartree/bohr for the total energy and forces on atoms, respectively. The electronic band structures and density of states (DOS) are calculated using the optimized structure for further transport coefficients calculations. The thermoelectric coefficients are calculated by applying the first-principles band structures to the semi-classical Boltzmann transport equation with constant relaxation time approximation. The Fermi velocity v_F calculations are numerically calculated by using the multi-orbital tight-binding Hamiltonian reproducing the first-principles band structure on the Wannier function basis.

III. Results and Discussions

FIG. 1 illustrates the strong anisotropic transport behavior along the in-plane and out-of-plane directions of the layered $NbSe_2$ materials. Aside from a change of sign, the charge density at which the in-plane Seebeck coefficient crosses the zero value varies with the stacking sequence; however, no significant change in amplitude was observed. In contrast, the out-of-plane Seebeck coefficient did not undergo a sign change, but the amplitude was strongly influenced by the stacking structure. Analysis of the Fermi velocities reveals that while having higher v_F in the out-of-plane direction, the transverse electrical and electronic thermal conductivities of the various structures remain limited due to the weak vdW coupling. On the other hand, the low out-of-plane v_F in the $2H_c$ configuration leads to the highest out-of-plane Seebeck coefficient among the stacking orders. These differences are due to the variations in the electronic structure among the stacking sequences. In-depth analysis of the relationship between the transport coefficients and the Fermi velocities will be discussed in the presentation.

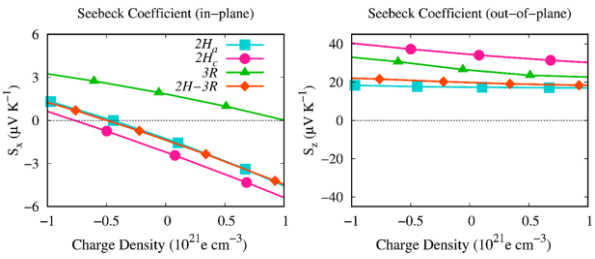

FIG. 1 Seebeck coefficient of $2H_a$, $2H_c$, 3R, and 2H-3R stacking configurations of $NbSe_2$ at 300°K as functions of charge density.

References:

[1] M. Marezio, *et al.*, J. Solid State Chem. **4**, 425 (1972).
[2] F. Zhang, *et al.*, Phys. Rev. B. **82**, 035409 (2010).
[3] P. Giannozzi *et al.*, J. Phys.: Condens. Matter **21**, 395502 (2009).
[4] R. Sabatini, *et al.*, J. Phys.: Condens. Matter **24**, 424209 (2012)