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Novel two-dimensional semiconductors ZrNCl and HfNCl: Stability, electric transport, and thermoelectric properties

Due to the development of the exfoliation technique, it has been possible to obtain atomic-layered sheets from various layered bulk materials [1]. Thereafter, enormous research efforts have been focused on two-dimensional (2D) layered semiconductors due to the potential applications in nanoscale optoelectronics, photonics, valleytronics, photovoltaics, spintronic devices, and so on [2,3]. For this reason, searching for novel 2D semiconducting materials is a challenging issue in the field of low-dimensional systems. Ternary transition-mental nitride halides (TMNH) are known to be changed to superconductors with moderately high transition temperatures up to 25.5 K upon electron doping by means of intercalation through the interlayer space [4]. During the last few decades, as a matter of fact, ternary TMNH have been intensively studied mainly for their superconductivity. Hence, not only their 2D structures but also the realization possibilities have received little attention. Nevertheless, a single-layer (1L) TMNH has been recently suggested simply from the fact that the layers of TMNH are bonded by van der Waals (vdW) interaction like graphite and transition-metal dichalcogenides. However, scientific details of 1L-TMNH including its stability are absolutely lacking. In this work, performing the first-principles calculations to examine extensively the stability, electronic structure, electric transport, and thermoelectric properties, we propose and discover novel promising 2D semiconductors 1L-ZrNCl and 1L-HfNCl. Both of them are shown to be easily isolated from the parent bulk materials and also thermodynamically stable based on the *ab initio* molecular dynamics and phonon dispersion calculations. In addition, strain engineering is found to be available for both 1L-ZrNCl and 1L-HfNCl, where a transition from an indirect to direct band gap is attained under a tensile strain. Note that this is important for optoelectronic device applications. It is also found that 1L-ZrNCl has an outstanding electron mobility of about 1.2×10^3 $cm^2V^{-1}s^{-1}$ in the *a*- or *b*-direction. Such electron mobility is significantly higher than that of 1L-MoS_2, whereas the hole mobility is about two orders of magnitude lower. Lastly, it is indicated that these systems have good thermoelectric properties, i.e., high Seebeck coefficient and high power factor [5]. With these findings, 1L-ZrNCl and 1L-HfNCl would be novel promising 2D materials for a wide range of optoelectronic and thermoelectric applications.

References

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