Supplementary Information for

A promising high temperature 2D thermoelectric material: Novel single-layer $ZrHfS_4$

Won Seok Yun[a], Sang Wook Han[b,*], Hyeon-Jun Lee[a], June-Seo Kim[a], and Myoung-Jae Lee[a,*]

[a]Division of Nanotechnology, DGIST, Daegu 42988, Republic of Korea

[b]Basic Science Research Institute and EHSRC, University of Ulsan, Ulsan 44610, Republic of Korea

[*]Authors to whom correspondence should be addressed: swhan72@ulsan.ac.kr (S.W.H.) and myoungjae.lee@dgist.ac.kr (M.-J.L.)

1. Calculation of cumulative lattice thermal conductivity for 1L-$ZrHfS_4$

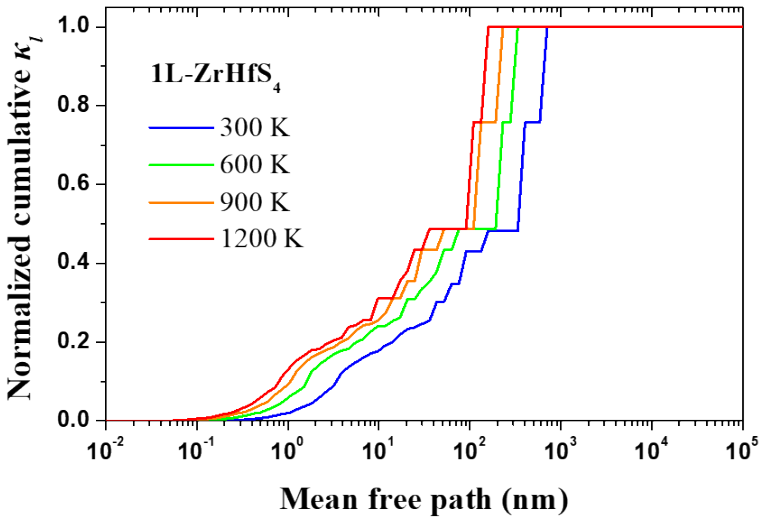

Fig. S1. Calculated normalized cumulative κ_l behaviors with respect to the phonon mean free path (MFP) for 1L-$ZrHfS_4$ at different temperatures.

As shown in Figure S1, we plot the κ_l accumulation as a function of the phonon mean free path (MFP) at various temperatures (300 K, 600 K, 900 K, and 1200 K), offering the insight into how the grain size can affetct κ_l. Note that κ_l is mostly affected by phonons with MFP ranging from 0.1 to 1000 nm. In addition, κ_l accumulation shifts to the low side of MFP when the temperature increases, as phonons with shorter MFPs contribute much at higher temperatures. Quantitively, MFPs at 50% of κ_l accumulation are approximately 159, 76, 52, and 36 nm at 300, 600, 900, and 1200 K, respectively.

2. Scattering rate and phase space calculations for 1L-$ZrHfS_4$

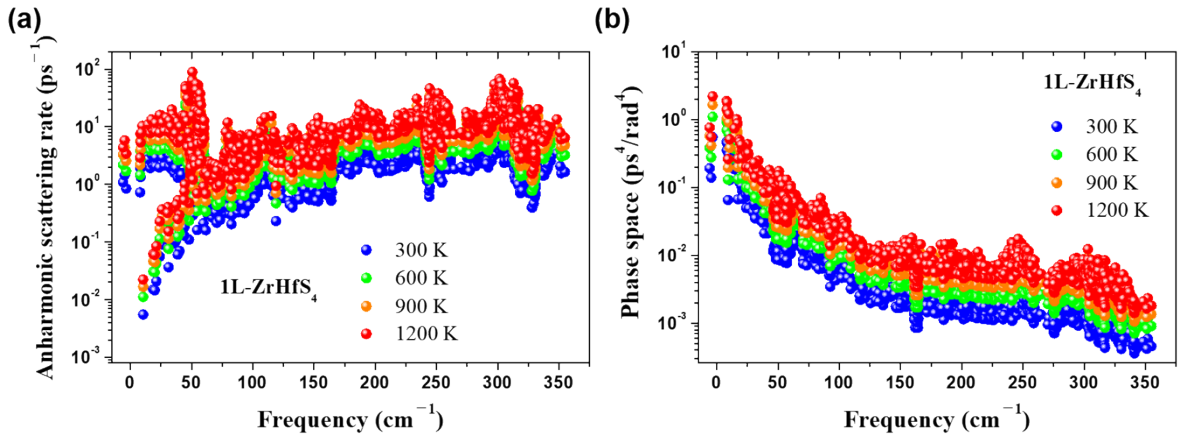

Fig. S2. (a) The frequency dependence of anharmonic scattering rate for 1L-$ZrHfS_4$ at different temperatures. (b) Calculated weighted three-phonon scattering phase space as a function of phonon frequency for 1L-$ZrHfS_4$ at different temperatures.

To gain more insight into the thermal transport of 1L-$ZrHfS_4$, we examined the phonon scattering mechanisms. Figure S2(a) illustrates the calculated anharmonic scattering rate with respect to the phonon frequency at 300 K, 600 K, 900 K, and 1200 K. One can observe that as the temperature increases, calculated phonon-phonon scattering rate for 1L-$ZrHfS_4$ rises. For this reason, the calculated κ_l decreases as the temperature increases, as confirmed in Fig. 3(a). Subsequently, the strength of phonon scattering is depends not only on anharmonicity but also on phase space. Therefore, Fig. S2(b) depicts the phonon frquency dependence of three-phonon scattering phase space at several temperatures. In the low-frequency range (0 to 175 cm^{-1}), the calculated phase space for 1L-$ZrHfS_4$ is larger compared to that in the high-freqency range (above 175 cm^{-1}). This means that the decrease in κ_l is more affected by acoustic phonons than by optical phonons. In addition, similar to the anharmonic scattering rate, the phase space for 1L-$ZrHfS_4$ increases with rising temperature, contributing to the decrease in κ_l.