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Janus 2H-MXTe (M = Zr, Hf; X = S, Se) monolayers with outstanding thermoelectric properties and low lattice thermal conductivities

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Abstract: Two-dimensional (2D) materials have been one of the most popular objects in the research field of thermoelectric (TE) materials, which have attract substantial attention in recent years. Inspired by the synthesized 2H-MoSSe and numerous theoretical studies, we systematically investigated the electronic, thermal, and TE properties of Janus 2H-MXTe (M = Zr, Hf; X = S, Se) monolayers by using first-principles calculations. The phonon dispersion curves and AIMD simulations confirm the thermodynamic stabilities. Moreover, Janus 2H-MXTe were evaluated as indirect band-gap semiconductors with band gaps ranging from 0.56 to 0.90 eV under the HSE06+SOC method. To evaluate the TE performance, firstly, we calculated the temperature-dependent carrier relaxation time with acoustic phonon scattering τ_{ac}, impurity scattering τ_{imp}, and polarized scattering τ_{pol}. Secondly, the calculation of lattice thermal conductivity (κ_l) shows these monolayers possess relatively poor κ_l with the

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values of 3.4-5.4 W/mK at 300 K, which is caused by the low phonon lifetime and group velocity. After computing the electronic transport properties, we found that the n-type doped Janus 2H-MXTe monolayers exhibit a high Seebeck coefficient exceeding 200 μV/K at 300 K, resulting in a high TE power factor. Eventually, combining the electrical and thermal conductivities, the optimal dimensionless figure of merit (zT) at 300 K (900 K) can be obtained, which are 0.94 (3.63), 0.51 (2.57), 0.64 (2.72), and 0.50 (1.98) for n-type doping of ZrSeTe, HfSeTe, ZeSTe, and HfSTe monolayers. Particularly, the ZrSeTe monolayer shows the best TE performance with the maximal zT value. These results indicate the excellent application potential of Janus 2H-MXTe (M = Zr, Hf; X = S, Se) monolayers in TE materials.

Key words：first principle, thermoelectric materials, figure of merit, thermal transport properties

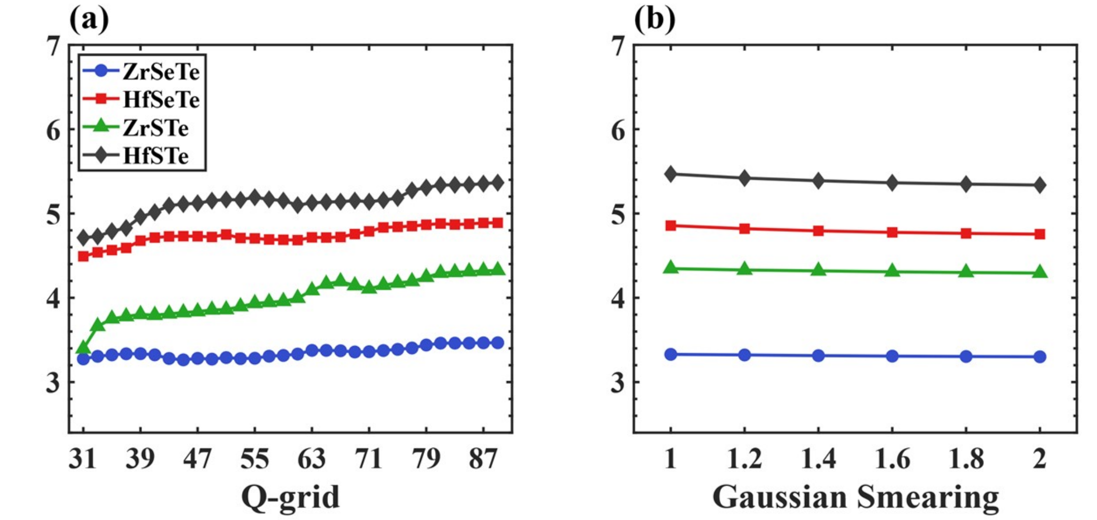

Fig. 1 The convergence test of the thermal conductivity with the increase in the Q-grid and Gaussian Smearing for ZrSeTe, HfSeTe, ZrSTe, and HfSTe monolayers.

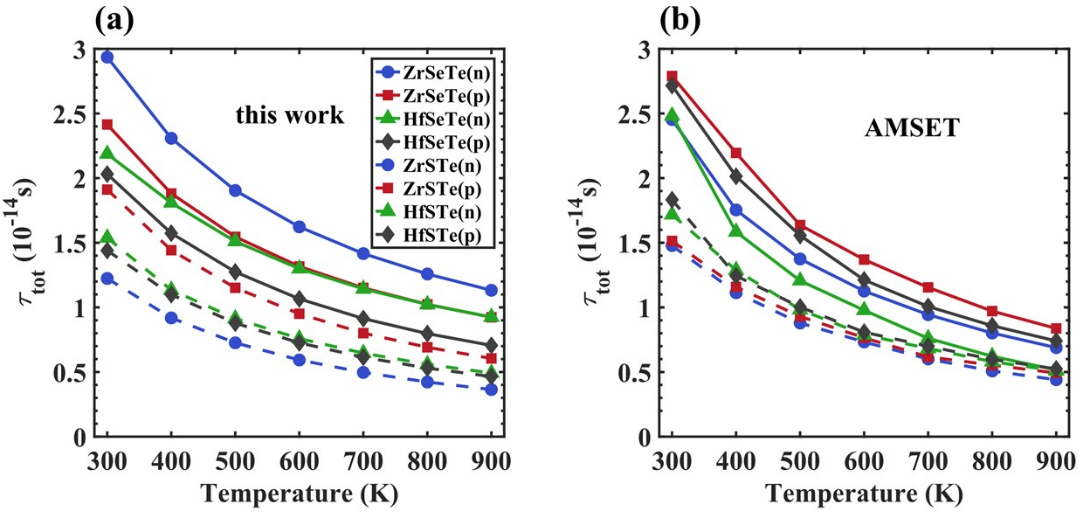

Fig. S2 The total carrier relaxation time τ_{tot} of (a) this work and (b) AMSET.