ARTICLE TYPE

Prediction of high thermoelectric potential in AMN_2 layered nitrides : electronic and vibrational properties †

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Band structures, electronic transport coefficients, harmonic and anharmonic vibrational properties of novel layered nitrides have been studied to evaluate the potential for thermoelectric applications. Using first principles theoretical methods we predict that AMN_2 compounds with A=Ca, Sr, and Ba, and M=Ti, Zr, Hf may exhibit Seebeck coefficients in excess of 150 μVK and good electrical conductivities. The phonon dispersions indicate the presence of low lying optic modes that can lead to low thermal conductivity. The analysis of the mode resolved Grüneisen parameter points to large anharmonicity. In addition, we show that the A-site substitution controls the degeneracies at the top of the valence band and the anisotropy of the Seebeck tensors.

1 Electronic Supplementary Information

Fig. 4 Thermoelectric transport coefficients of $CaTiN_2$ as a function of temperature at the optimal carrier concentration (n): (a) p-type Seebeck coefficient; (b) p-type electrical conductivity divided by the constant relaxation time; (c) n-type Seebeck coefficient; (d) n-type electrical conductivity divided by the constant relaxation time.

⁽xx'/y (zz') Total (a) S (µV/K) S (J/V/K) .40 (C) 800 оо Т (К) т (к 20 (xx'/yy' (b) (xx'/yy (zz') (d) 15 σ.10^{18/τ} (Ω⁻¹m⁻¹s⁻¹) σ.10^{18/τ} (Ω⁻¹m⁻¹s⁻¹) 10 50 50 400 T (K) 200 400 T (K) 600 800 1000 200 600 800 1000

[†] Electronic Supplementary Information (ESI) available: Band structures for CaHfN₂, SrHfN₂ and BaHfN₂; Densities of states for CaZrN₂, SrZrN₂, BaZrN₂, CaHfN₂, SrHfN₂ and BaHfN₂; Thermoelectric transport coefficients of CaTiN₂, BaTiN₂, SrZrN₂, CaHfN₂, SrHfN₂, and BaHfN₂; Vibration density of states of CaTiN₂; Mode-resolved Grüneisen parameters versus mode frequency for SrTiN₃. See DOI: 10.1039/b000000x/

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Fig. 5 Thermoelectric transport coefficients of $BaTiN_2$ as a function of temperature at the optimal carrier concentration (n): (a) p-type Seebeck coefficient; (b) p-type electrical conductivity divided by the constant relaxation time; (c) n-type Seebeck coefficient; (d) n-type electrical conductivity divided by the constant relaxation time.



Fig. 7 Thermoelectric transport coefficients of $CaHfN_2$ as a function of temperature at the optimal carrier concentration (n): (a) p-type Seebeck coefficient; (b) p-type electrical conductivity divided by the constant relaxation time; (c) n-type Seebeck coefficient; (d) n-type electrical conductivity divided by the constant relaxation time.



Fig. 6 Thermoelectric transport coefficients of $SrZrN_2$ as a function of temperature at the optimal carrier concentration (n): (a) p-type Seebeck coefficient; (b) p-type electrical conductivity divided by the constant relaxation time; (c) n-type Seebeck coefficient; (d) n-type electrical conductivity divided by the constant relaxation time.



Fig. 8 Thermoelectric transport coefficients of $SrHfN_2$ as a function of temperature at the optimal carrier concentration (n): (a) p-type Seebeck coefficient; (b) p-type electrical conductivity divided by the constant relaxation time; (c) n-type Seebeck coefficient; (d) n-type electrical conductivity divided by the constant relaxation time.



Fig. 9 Thermoelectric transport coefficients of $BaHfN_2$ as a function of temperature at the optimal carrier concentration (n): (a) p-type Seebeck coefficient; (b) p-type electrical conductivity divided by the constant relaxation time; (c) n-type Seebeck coefficient; (d) n-type electrical conductivity divided by the constant relaxation time.



Fig. 11 Mode-resolved Grüneisen parameters versus mode frequency for ${\rm SrTiN}_2.$



Fig. 1 Band structures for CaHfN₂, SrHfN₂ and BaHfN₂. Energies are refereed to the top of the valence band (black solid lines at 0 eV).



Fig. 2 Density of states projected on the atomic spheres for CaZrN₂, SrZrN₂ and BaZrN₂. Energies are referred to the top of the valence band.



Fig. 3 Density of states projected on the atomic spheres for CaHfN₂, SrHfN₂ and BaHfN₂. Energies are refereed to the top of the valence band.



Fig. 10 Vibration density of states of CaTiN₂. (a) Total, in-plane, and out-of-plane VDOSs; (b) in-plane VDOS projected on each element; (c) out-of-plane VDOS projected on each element; (d) total VDOS projected on each element.

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