Supplementary Materials

Hydrogen Functionalization and Strain Tuning of Superconductivity and Charge Density Waves in MXene-Derived Chalcogenide Monolayers

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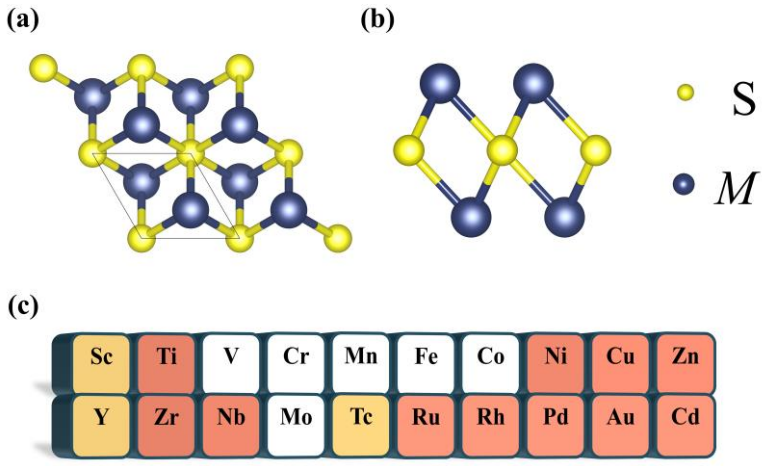

Fig. S1 (a) and (b) are the top view and side view of the 1T-M_2S structure, respectively. (c) shows the screening results of 1T-M_2S monolayers; the white-filled monolayers are magnetic, the red-filled monolayers are neither magnetic nor dynamically stable, and the yellow-filled monolayers are stable

structures.

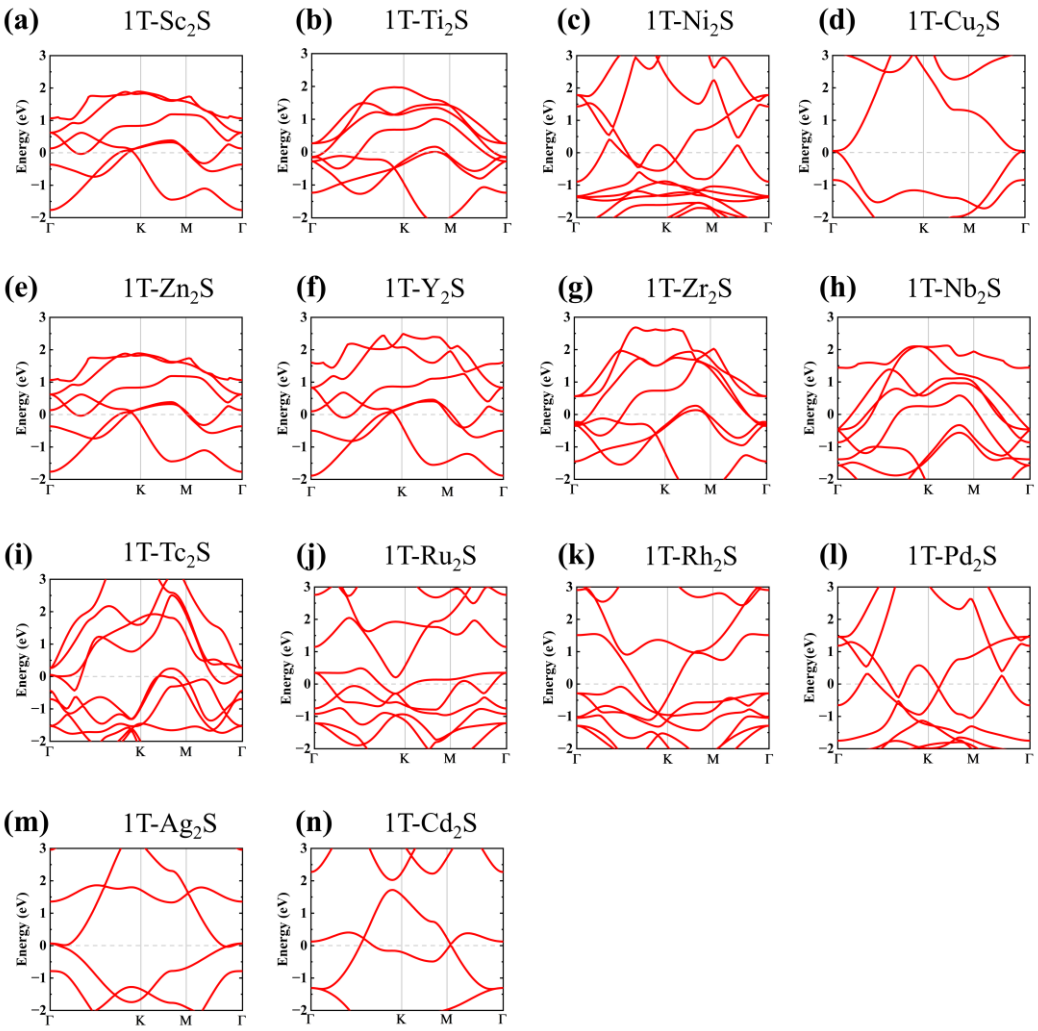

Fig. S2 The band structure diagrams of 1T-M_2S after screening.

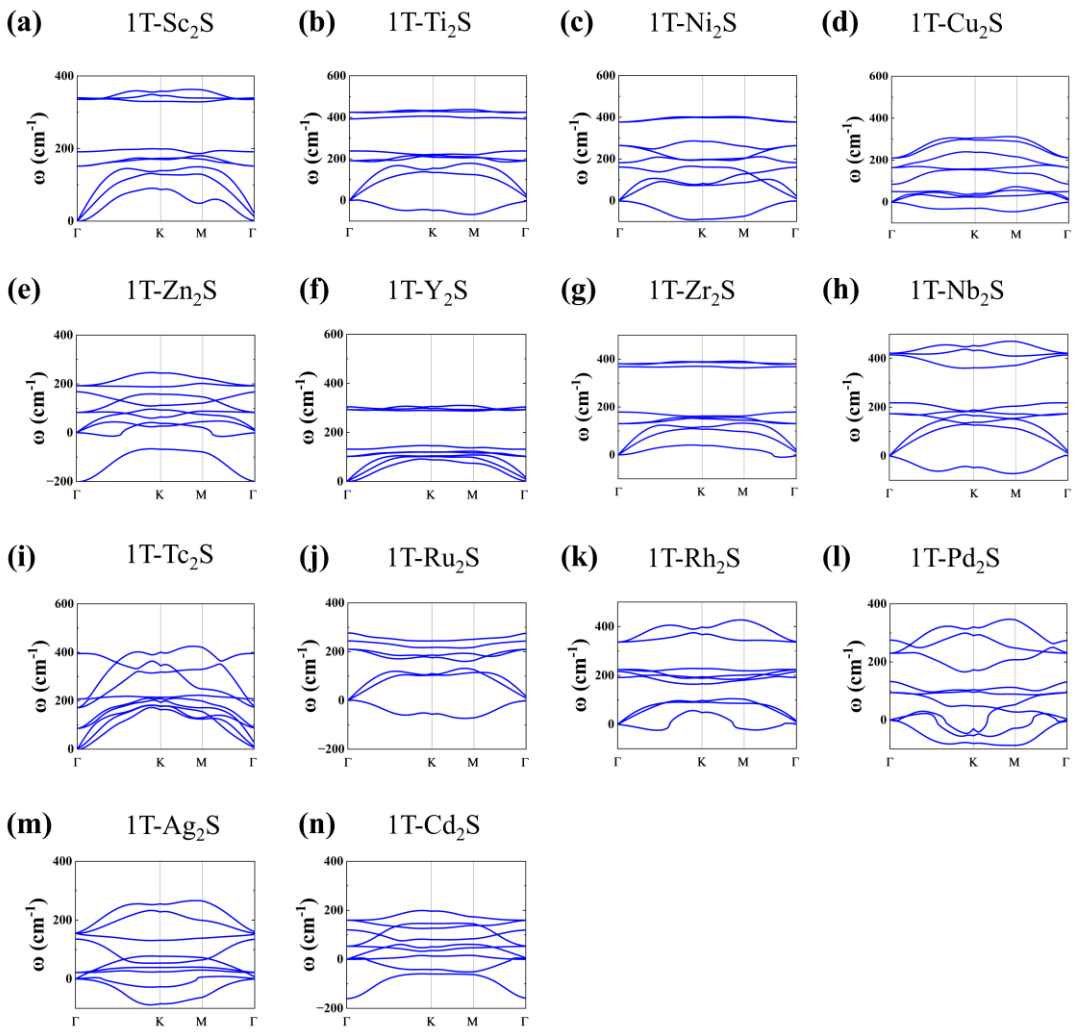

Fig. S3 The phonon dispersions of 1T-M_2S after screening.

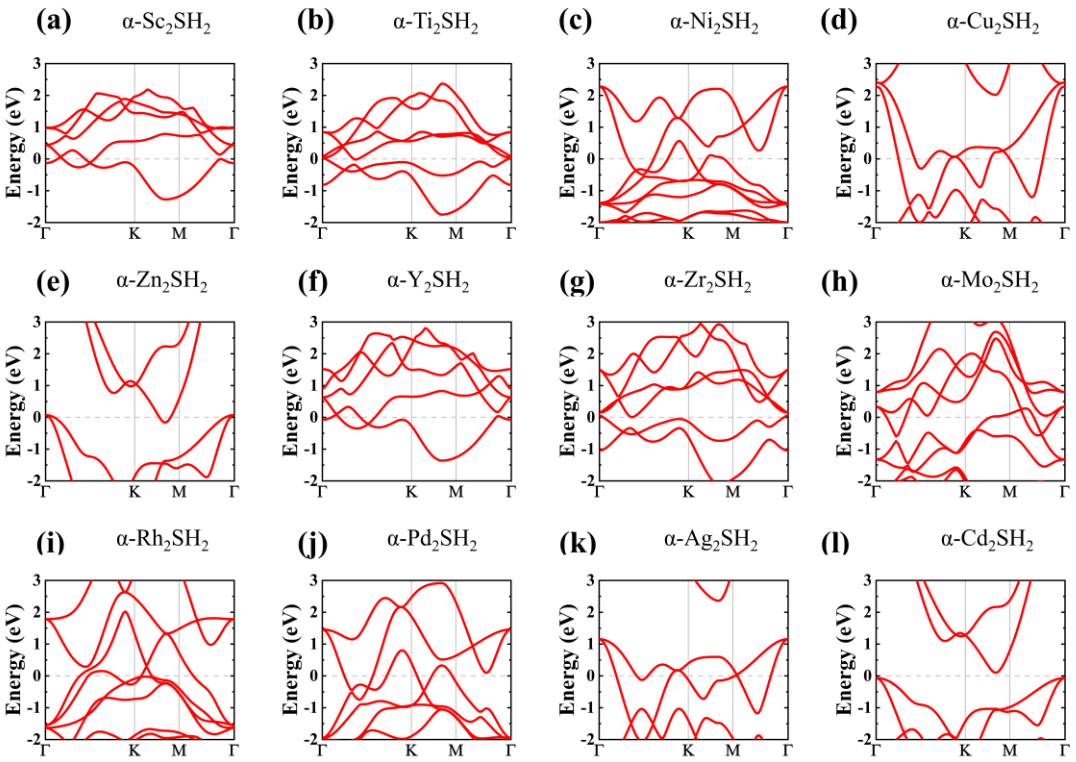

Fig. S4 The band structure diagrams of α-M_2SH_2.

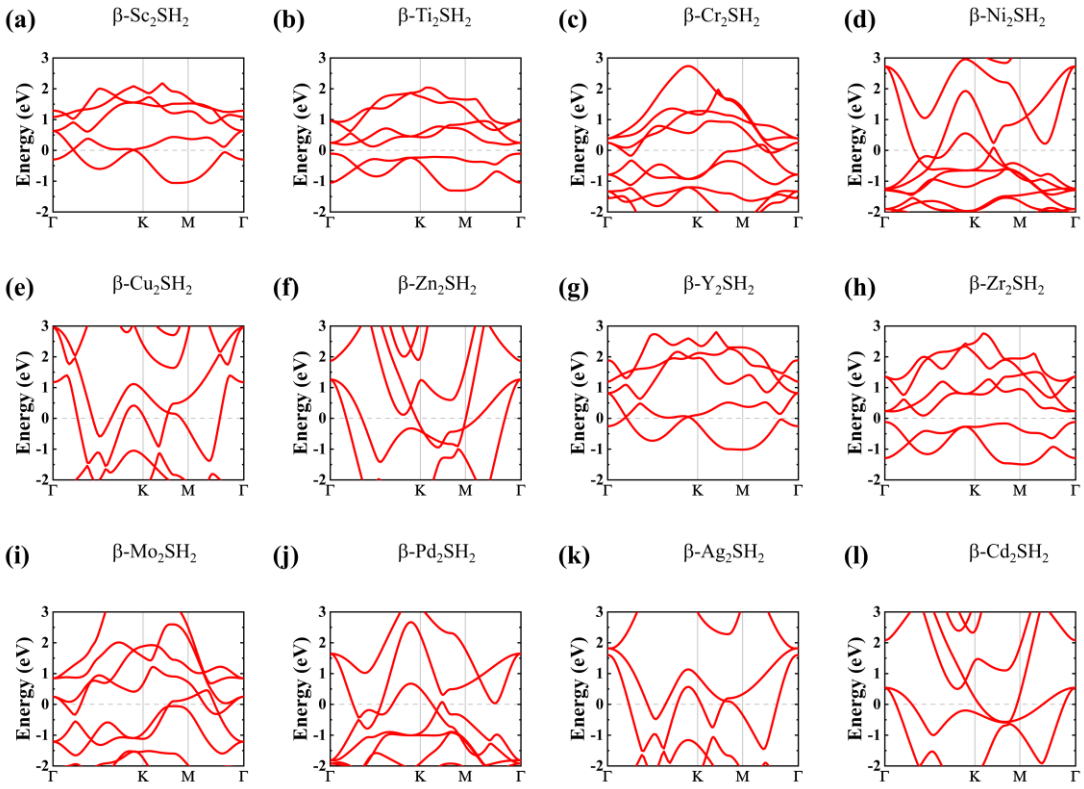

Fig. S5 The band structure diagrams of β-M_2SH_2

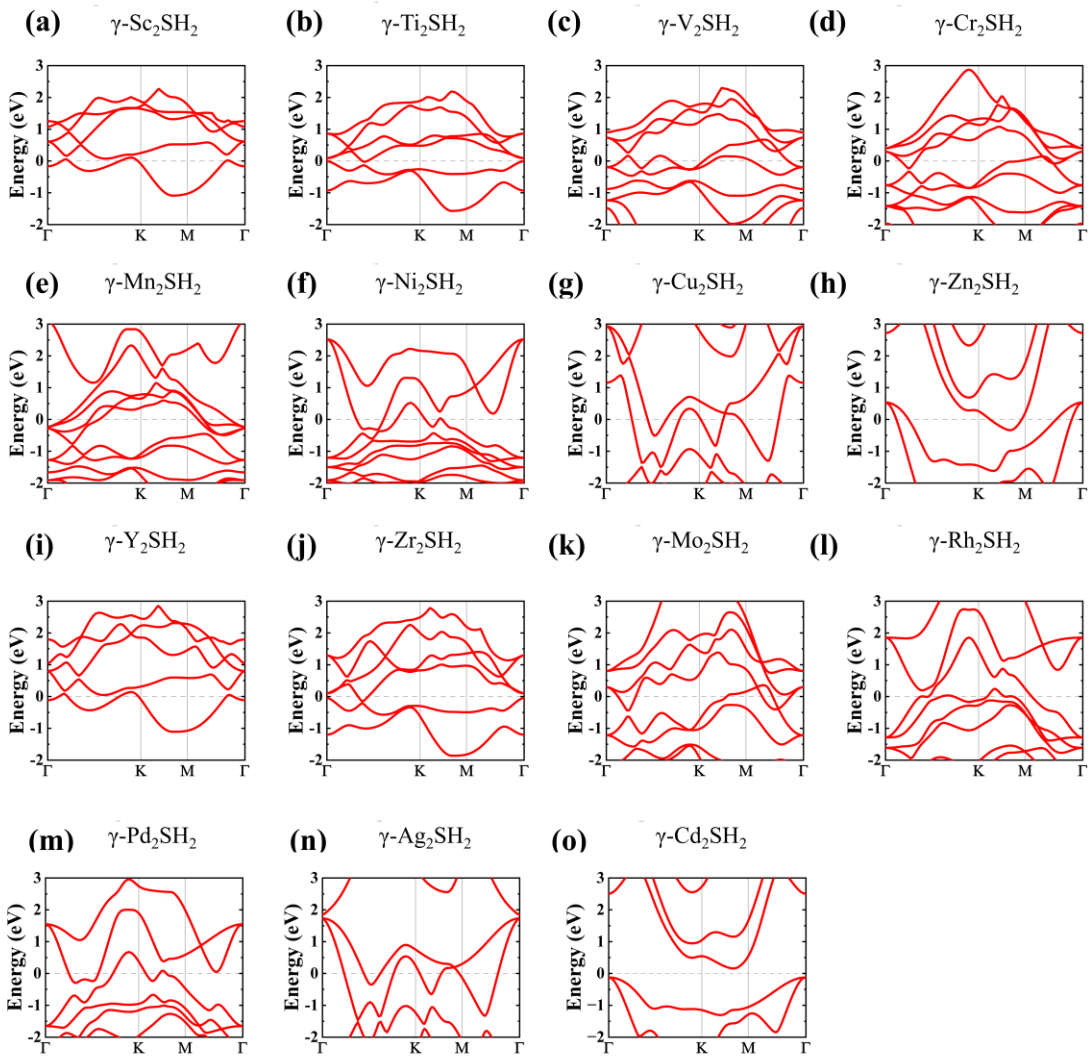

Fig. S6 The band structure diagrams of γ-M_2SH_2

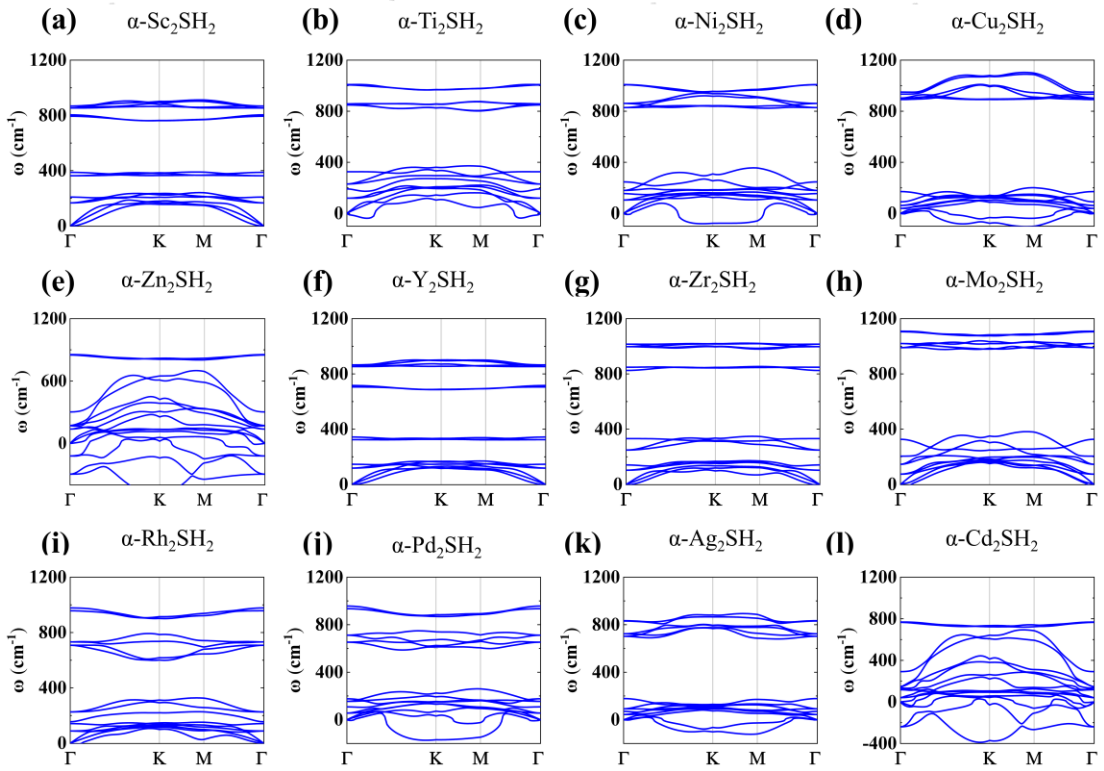

Fig. S7 The phonon dispersions of α-M_2SH_2.

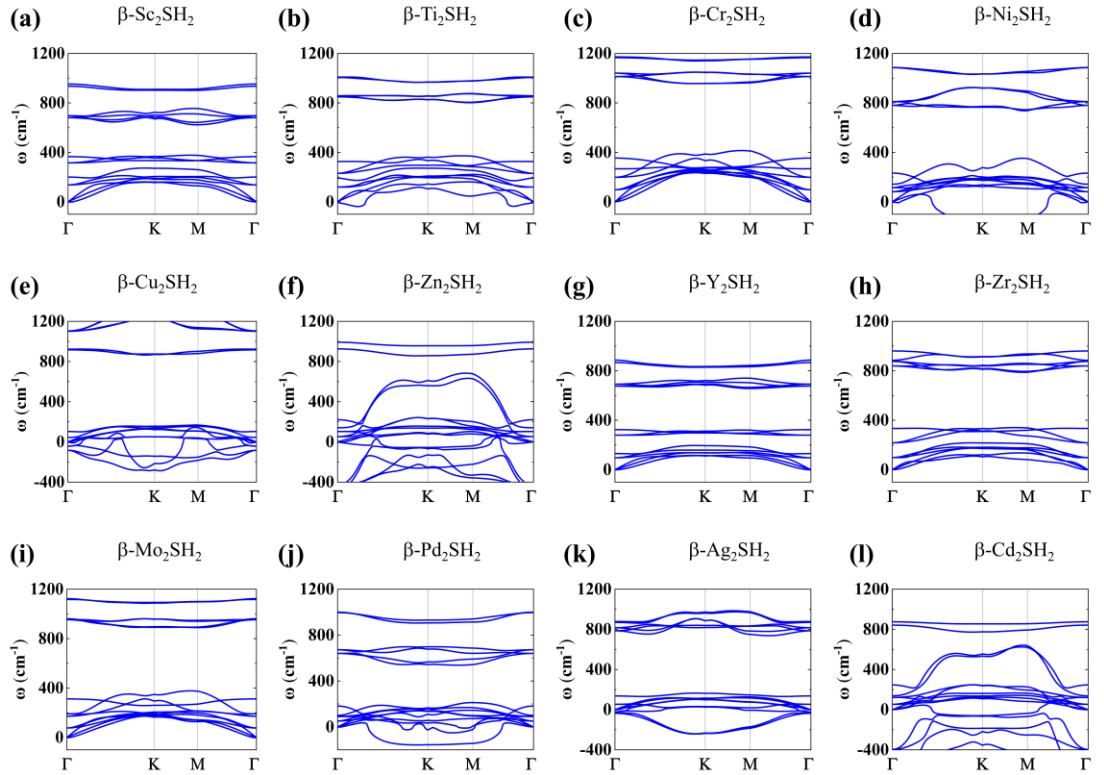

Fig. S8 The phonon dispersions of β-M_2SH_2.

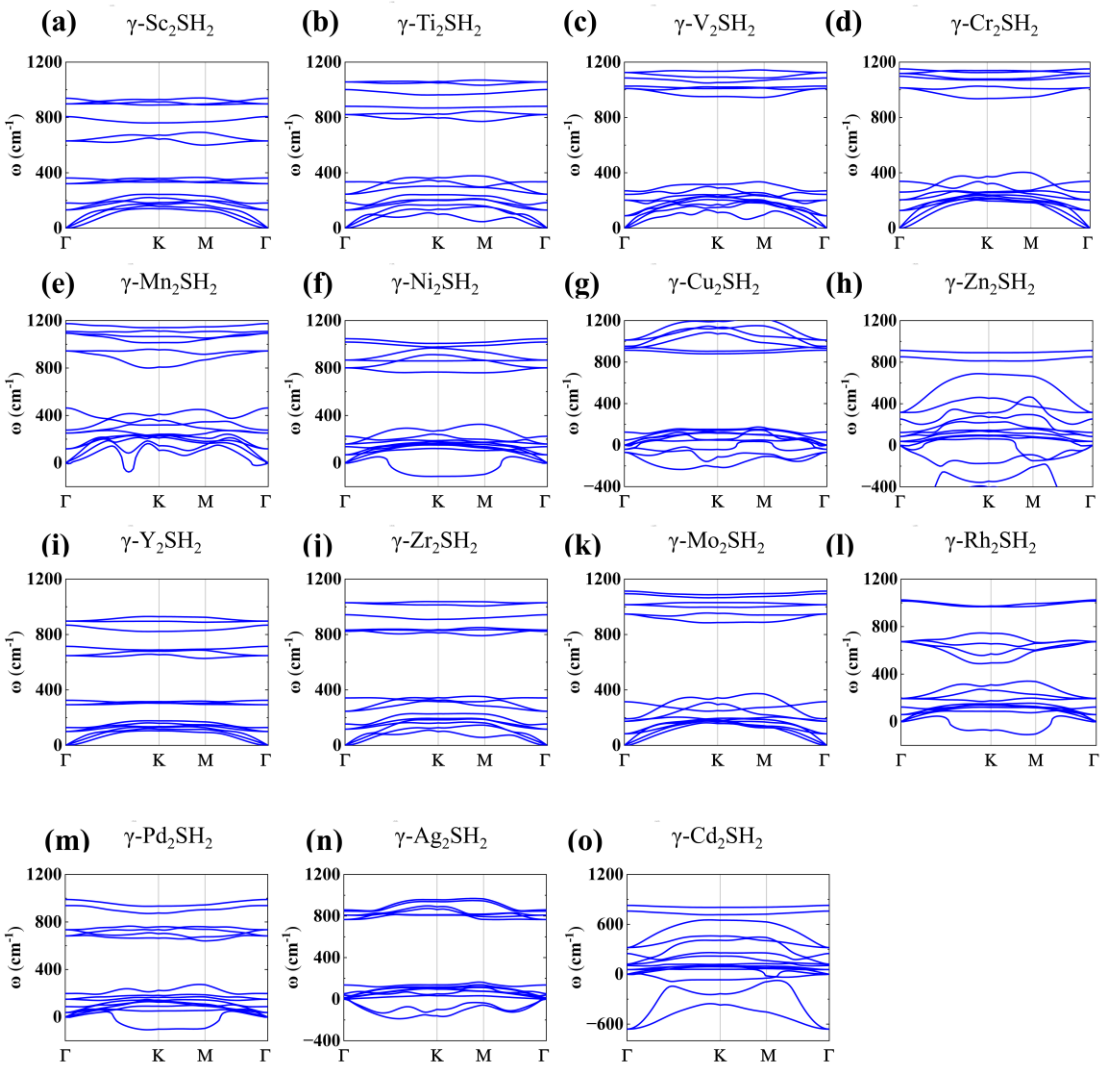

Fig. S9 The phonon dispersions of γ-M_2SH_2

Table S1. Spreads and the RMS of the Wannier functions

Spreads/ Monolayer	α-Tc_2SH_2	β-Tc_2SH_2	γ-Tc_2SH_2	α-Ru_2SH_2	β-Ru_2SH_2	γ-Ru_2SH_2
1	2.25608	5.76747	5.32813	1.34732	1.41177	1.26504
2	3.26768	1.85761	1.58623	1.33598	1.31980	1.88642
3	4.80775	3.54602	4.07875	1.24527	1.37790	1.30251

Spreads/ Monolayer	α-Tc_2SH_2	β-Tc_2SH_2	γ-Tc_2SH_2	α-Ru_2SH_2	β-Ru_2SH_2	γ-Ru_2SH_2
4	2.88629	1.90403	1.99642	2.05587	1.34565	1.30670
5	1.66963	1.62870	5.03137	1.65441	1.37081	1.43676
6	1.61016	1.82799	3.44895	1.72929	1.78773	1.36872
7	5.52226	3.41329	1.58475	2.05991	1.35333	4.44530
8	1.6766	1.85658	5.00516	1.40668	1.34232	1.35980
9	5.72795	3.36230	1.84310	1.71586	1.33583	2.35784
10	2.24368	3.43780	1.60295	1.36879	1.32726	1.70946
11	4.05083	1.63146	3.54917	1.74702	1.70316	5.49063
12	2.26712	1.96754	1.88324	1.62543	1.93744	1.32578
13	4.59013	4.04700	1.59932	2.69237	1.31713	2.05453
14	1.73025	1.95207	2.11160	1.78210	1.74647	1.25678
15	/	/	/	1.25543	1.70075	1.69438
RMS	0.00655	0.00704	0.04141	0.00399	0.02811	0.06392

Table S2. Convergence tests of the K-point density, μ*, and smearing for β-Tc_2SH_2 and β-Ru_2SH_2, evaluated using λ.

Monolayer	β-Tc_2SH_2	β-Ru_2SH_2
k-mesh=120×120×1	1.50	2.42
k-mesh=100×100×1	1.51	2.54
q-mesh=120×120×1	1.50	2.42
q-mesh=100×100×1	1.52	2.42
smear = 0.05	1.50	2.41
smear = 0.1	1.52	2.43

μ^* = 0.1	1.51	2.44
μ^* = 0.12	1.50	2.42

Table S3. The parameters for building Wannier functions

Monolayer	dis_win_max (Ry)	temps (K)
α-Tc_2SH_2	4.1	14-22
β-Tc_2SH_2	5.0	21-27
γ-Tc_2SH_2	4.9	27-32
α-Ru_2SH_2	5.1	16-26
β-Ru_2SH_2	5.0	20-50
γ-Ru_2SH_2	5.0	28-36

The input file for EPW

System-dependent parameters, including the Wannier disentanglement window (dis_win_max),and temperature grids, vary among different materials and are explicitly listed in Supplementary Table S3.

```
&inputepw
   prefix           = ''
   outdir           = './'
   max_memlt        = 40
   ep_coupling      = .true.
   elph             = .true.
   epbwrite         = .false.
   epbread          = .false.
```

```
epwwrite          = .false.
epwread           = .true.
etf_mem           = 1
nbndsub           = 15
proj(1)           = 'random'
wannierize        = .true.
num_iter          = 500
dis_win_min       = -10.0
dis_win_max       = x         ! system-dependent, see Table S3
iverbosity        = 2
fermi_plot        = .false.
eps_acustic       = 2.0       ! THz, lower bound for phonon frequencies
ephwrite          = .true.
fsthick           = 0.5       ! eV, Fermi-surface energy window
degaussw          = 0.05      ! eV, electronic Gaussian broadening
delta_smear       = 0.04      ! eV, electronic delta-function smearing
degaussq          = 0.05      ! meV, phonon Gaussian broadening
nqstep            = 500
eliashberg        = .true.
laniso            = .true.
limag             = .true.
conv_thr_iaxis = 1.0d-4
wscut             = 1
nstemp            = 1
temps             = y         ! K, temperature grid (Table S3)
nsiter            = 500
muc               = 0.10
```

```
    dvscf_dir          = 'save'
    nk1 = 12; nk2 = 12 ; nk3 = 1
    nq1 = 6 ; nq2 = 6 ; nq3 = 1
    mp_mesh_k = .true.
    nkf1 = 120 ; nkf2 = 120 ; nkf3 = 1
    nqf1 = 120 ; nqf2 = 120 ; nqf3 = 1
/
```