Supplemental Information

Inverse Design of Stable Spinel Compounds with High Optical Absorption via Materials Genome Engineering

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The decomposition energies of the four examples of four types of spinel compounds, i.e. $CdBi_2S_4$ (type-I), $PbBi_2S_4$ (type-II), $PbIn_2S_4$ (type-III) and $CdIn_2S_4$ (type-IV) are calculated in common decomposition pathways (decomposing into binary compounds), as shown in Fig. S1. The positive values indicate that they are thermodynamically stable. The phonon spectrum of these four cases are shown in Fig. S2. No virtual frequencies exist in $CdIn_2S_4$ and $CdBi_2S_4$, indicating that the are dynamically stable in the spinel structure. Indeed, $CdIn_2S_4$ and $CdBi_2S_4$ have been synthesized in experiment[1,2]. The weak negative frequencies of $PbBi_2S_4$ and $PbIn_2S_4$ indicates that there may exist some local distortions for these compounds in these spinel structure. We notice that the alloy $(Cd,Pb)Bi_2S_4$ has also been synthesized in experiment[3].

The electronic properties such as band gaps and effective mass of $CdBi_2S_4$, $PbBi_2S_4$, $PbIn_2S_4$ and $CdIn_2S_4$ are shown in Table S1. $CdIn_2S_4$ has pseudodirect band gap, while the other three have indirect band gaps. We conduct the HSE calculations with the parameter α =0.28 to align the benchmark of the experimental band gap of $CdIn_2S_4$ (2.2 eV).[4]

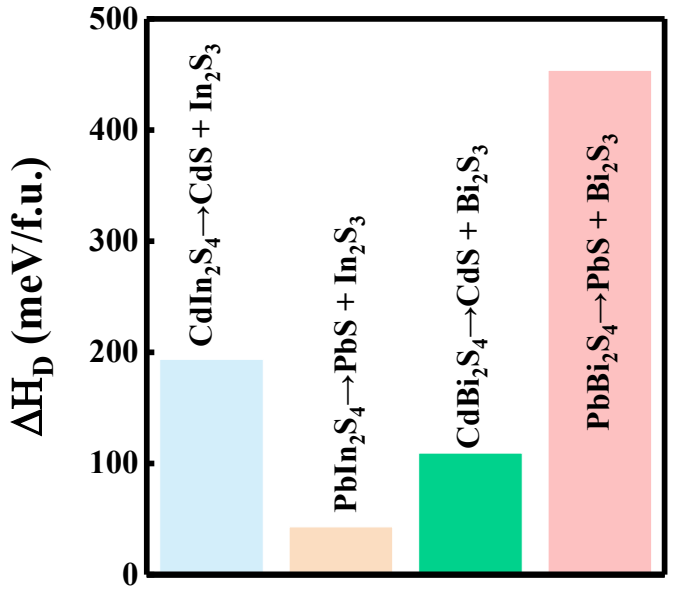

Figure S1. The calculated decomposition energies of $CdBi_2S_4$ (type-I), $PbBi_2S_4$ (type-II), $PbIn_2S_4$ (type-III) and $CdIn_2S_4$ (type-IV) along the common decomposition pathways.

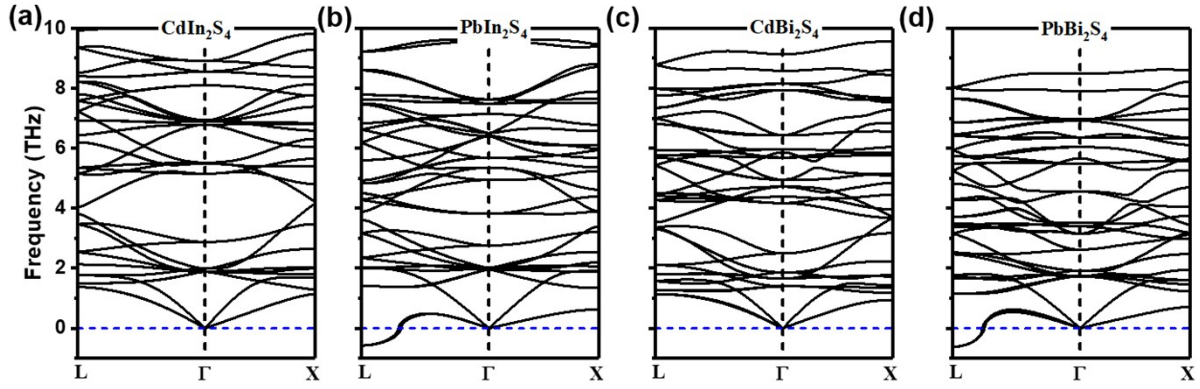

Figure S2. The calculated phonon spectrum of (a) $CdBi_2S_4$, (b) $PbBi_2S_4$, (c) $PbIn_2S_4$ and (d) $CdIn_2S_4$ at 0K.

Table S1. The PBE-calculated and HSE-calculated (α = 0.28) band gaps, electron and hole effective mass for four examples of four types of spinel compounds, i.e. $CdBi_2S_4$ (type-I), $PbBi_2S_4$ (type-II), $PbIn_2S_4$ (type-III) and $CdIn_2S_4$ (type-IV).

Spinel compounds	**PBE gap(eV)**	**HSE gap(eV)**	**Electron effective mass (m_e)**	**Hole effective mass (m_e)**
$CdIn_2S_4$	1.11 (pseudodirect)	2.20	0.14	2.75
$PbIn_2S_4$	0.80 (indirect)	1.48	0.68	0.37
$CdBi_2S_4$	0.74 (indirect)	1.68	0.18	0.47
$PbBi_2S_4$	1.55 (indirect)	2.26	0.59	0.87

References

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