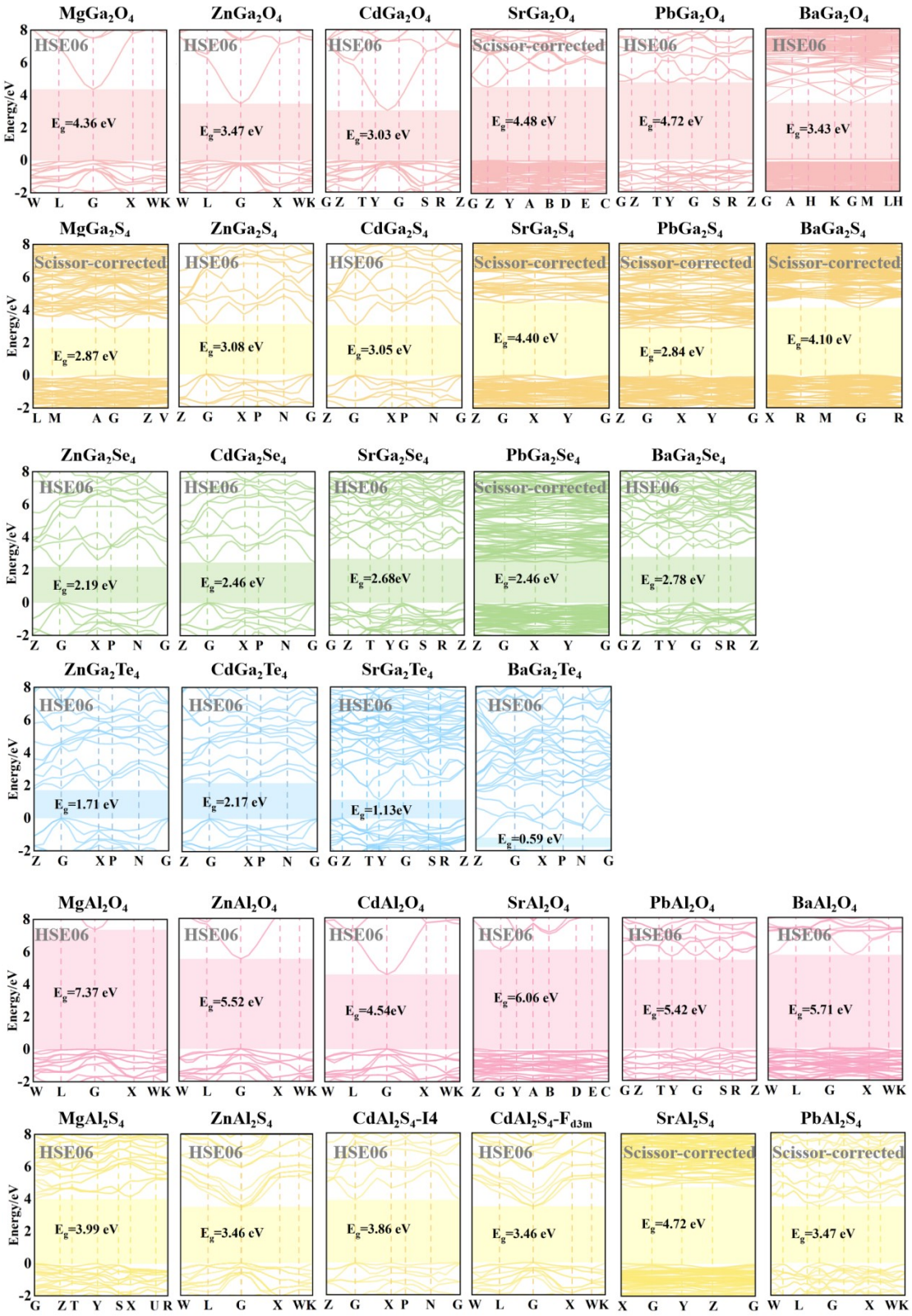
Supporting Information



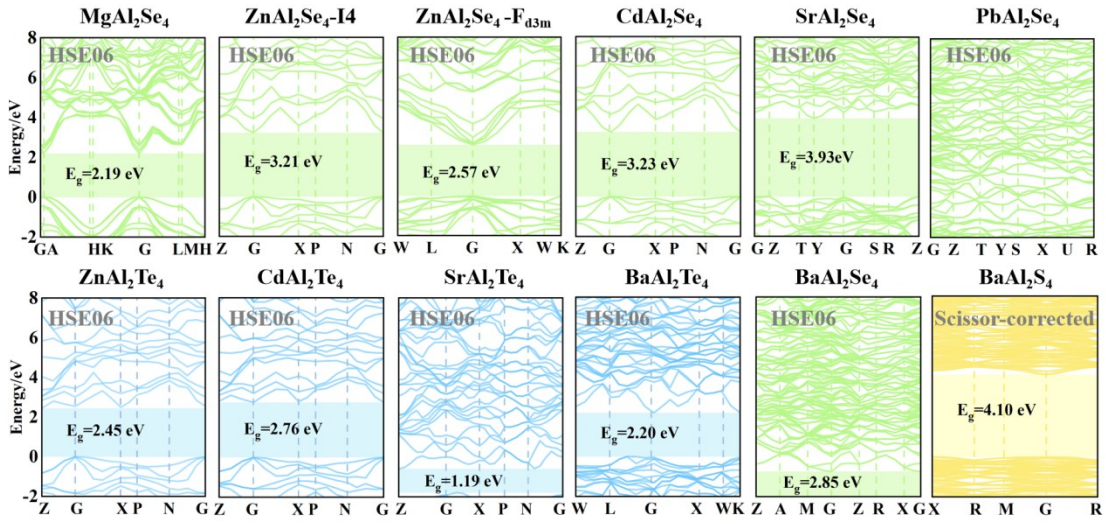


Fig. S1. The band structures of the ternary compound AB_2Ch_4 (A^{2+}=Mg^{2+}, Zn^{2+}, Cd^{2+}, Sr^{2+}, Pb^{2+}, Ba^{2+}; B^{3+} = Ga^{3+}, Al^{3+}; Ch^{2-}=O^{2-}, S^{2-}, Se^{2-}, Te^{2-}).

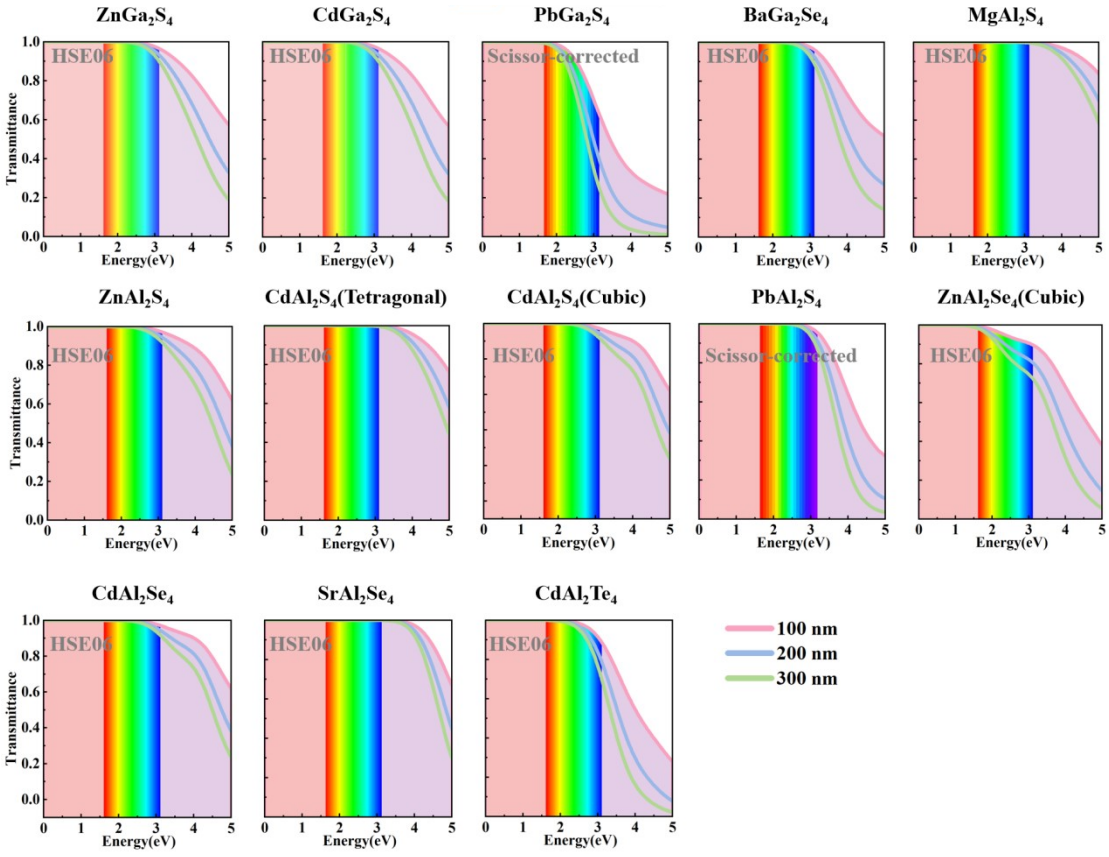


Fig. S2. The transmittance of 13 pre-screened candidates.

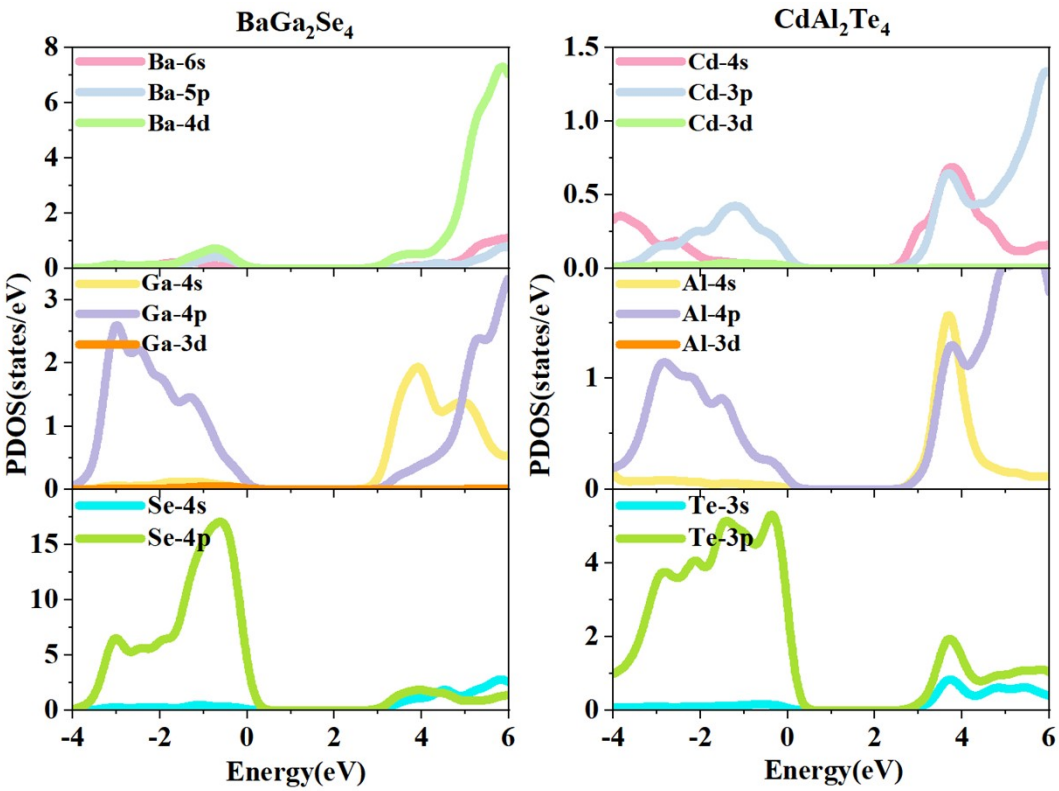


Fig. S3. The PDOS for the most promising systems $BaGa_2Se_4$ and $CdAl_2Te_4$.

(1) In order to ensure the stability of $BaGa_2Se_4$ and $CdAl_2Te_4$ during growth, the sum of the relative chemical potentials $\Delta\mu_i$ of each component element must equal the formation enthalpy (ΔH_f) of the compound:

$$\Delta H_f\left(BaGa_2Se_4\right) = \Delta\mu_{Ba} + 2\Delta\mu_{Ga} + 4\Delta\mu_{Se} \quad (1)$$

$$\Delta H_f\left(CdAl_2Te_4\right) = \Delta\mu_{Cd} + 2\Delta\mu_{Al} + 4\Delta\mu_{Te} \quad (2)$$

(2) To prevent the precipitation of elemental elements in the synthesis process, the $\Delta\mu_i$ of each element should satisfy:

$$\Delta\mu_{Ba}<0,\ \Delta\mu_{Ga}<0,\ \Delta\mu_{Se}<0 \quad (3)$$

$$\Delta\mu_{Cd}<0,\ \Delta\mu_{Al}<0,\ \Delta\mu_{Te}<0 \quad (4)$$

(3) To avoid the formation of competitive phases, the following inequality constraints must be satisfied:

$$\Delta\mu_{Ba}+\Delta\mu_{Se} \leqslant \Delta H_f(\mathrm{BaSe}) \quad (5)$$

$$\Delta\mu_{Ba}+2\Delta\mu_{Se} \leqslant \Delta H_f(BaSe_2) \quad (6)$$

$$\Delta\mu_{Ga}+\Delta\mu_{Se} \leqslant \Delta H_f(\mathrm{GaSe}) \quad (7)$$

$$2\Delta\mu_{Ga}+3\Delta\mu_{Se} \leqslant \Delta H_f(Ga_2Se_3) \quad (8)$$

$$\Delta\mu_{Cd}+\Delta\mu_{Te} \leqslant \Delta H_f(\mathrm{CdTe}) \quad (9)$$

$$2\Delta\mu_{Al}+3\Delta\mu_{Te} \leqslant \Delta H_f(Al_2Te_3) \quad (10)$$

$$2\Delta\mu_{Al}+5\Delta\mu_{Te} \leqslant \Delta H_f(Al_2Te_5) \quad (11)$$

(4) To prevent the formation of impurity phases between the doping elements and the matrix material, additional constraints on chemical potential must also be satisfied:

$$m\Delta\mu_D + n\Delta\mu_X \leqslant \Delta H(D_m X_n) \tag{12}$$

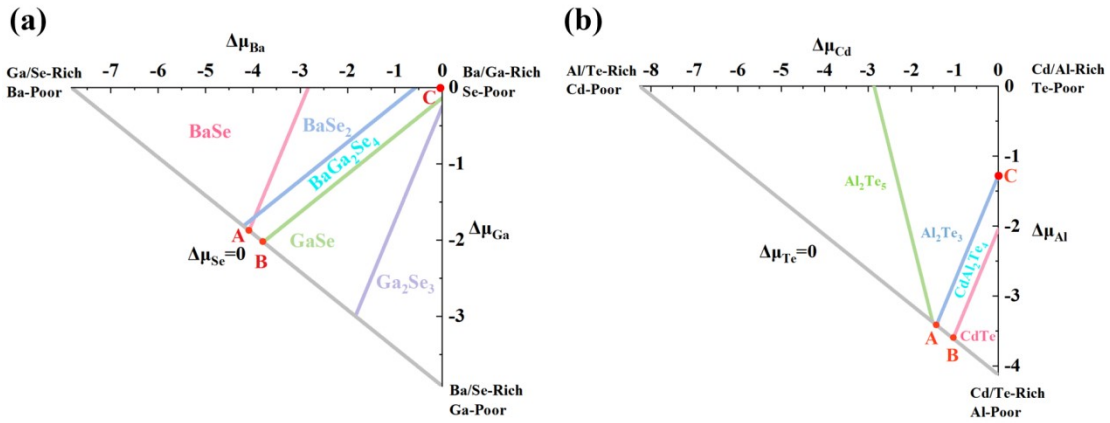


Fig. S4. The calculated phase diagrams for (a) $BaGa_2Se_4$ (A: Ba-Poor; B: Ga-Poor; C: Se-Poor) and (b) $CdAl_2Te_4$ (A: Cd-Poor; B: Al-Poor; C: Te-Poor).

Table S1. The optimized structural parameters and the experimental values of the ternary compound AB_2Ch_4 (A^{2+}=Mg^{2+}, Zn^{2+}, Cd^{2+}, Sr^{2+}, Pb^{2+}, Ba^{2+}; B^{3+} = Ga^{3+}, Al^{3+}; Ch^{2-}=O^{2-}, S^{2-}, Se^{2-}, Te^{2-})

	Sapace	a	b	c	α	β	γ	Reference
$MgGa_2O_4$	$Fd\bar{3}m$	8.48	8.48	8.48	90	90	90	This work
		8.29	8.29	8.29	90	90	90	Exp.[1]
$ZnGa_2O_4$	$Fd\bar{3}m$	8.55	8.55	8.55	90	90	90	This work
		8.33	8.33	8.33	90	90	90	Exp.[2]
$CdGa_2O_4$	$Fd\bar{3}m$	8.76	8.76	8.76	90	90	90	This work
		8.60	8.60	8.60	90	90	90	Exp.[3]
$SrGa_2O_4$	$P2_1/C$	8.46	9.13	10.86	90	94.13	90	This work
		8.38	8.99	10.68	90	93.93	90	Exp.[4]
$PbGa_2O_4$	*Ama*2	8.72	9.53	5.36	90	90	90	This work
		8.59	9.42	5.35	90	90	90	Exp.[5]
$BaGa_2O_4$	$P6_3$	19.07	19.07	8.87	90	90	120	This work
		18.65	18.65	8.67	90	90	120	Exp.[6]
$MgGa_2S_4$	*C*2/*c*	13.09	22.99	6.50	90	108.40	90	This work
		12.74	22.54	6.43	90	108	90	Exp.[7]
$ZnGa_2S_4$	$I\bar{4}$	5.47	5.47	10.55	90	90	90	This work
		5.34	5.34	10.45	90	90	90	Exp.[8]
$CdGa_2S_4$	$I\bar{4}$	5.71	5.71	10.36	90	90	90	This work
		5.73	5.73	10.07	90	90	90	Exp.[8]
$SrGa_2S_4$	*Fddd*	21.32	20.88	12.45	90	90	90	This work
		20.93	20.55	12.30	90	90	90	Exp.[9]
$PbGa_2S_4$	*Fddd*	21.05	20.67	12.40	90	90	90	This work
		20.71	20.38	12.16	90	90	90	Exp.[10]
$BaGa_2S_4$	*Pa*3	12.90	12.90	12.90	90	90	90	This work
		12.66	12.66	12.66	90	90	90	Exp.[11]

$ZnGa_2Se_4$	*I*$\bar{4}$2*m*	5.66	5.66	11.11	90	90	90	This work
		5.54	5.54	11.03	90	90	90	Exp.[12]
$CdGa_2Se_4$	*I*$\bar{4}$	5.93	5.93	10.92	90	90	90	This work
		5.84	5.84	10.75	90	90	90	Exp.[13]
$SrGa_2Se_4$	*Cccm*	6.35	10.99	11.19	90	90	90	This work
		6.34	10.97	11.10	90	90	90	Exp.[14]
$PbGa_2Se_4$	*Fddd*	12.83	21.63	21.76	90	90	90	This work
		12.72	21.28	21.53	90	90	90	Exp.[15]
$BaGa_2Se_4$	*Cccm*	6.39	11.46	11.65	90	90	90	This work
		6.48	11.13	11.24	90	90	90	Exp.[16]
$ZnGa_2Te_4$	*I*$\bar{4}$2*m*	6.07	6.07	11.98	90	90	90	This work
		5.92	5.92	11.81	90	90	90	Exp.[17]
$CdGa_2Te_4$	*I*$\bar{4}$	6.29	6.29	11.90	90	90	90	This work
		6.12	6.12	11.81	90	90	90	Exp.[18]
$SrGa_2Te_4$	*Cccm*	6.80	11.78	11.83	90	90	90	This work
		6.74	11.62	11.63	90	90	90	Exp.[19]
$BaGa_2Te_4$	*I*4/*mcm*	8.24	8.24	7.50	90	90	90	This work
		8.42	8.42	6.80	90	90	90	Exp.[20]
$MgAl_2O_4$	*Fd*$\bar{3}$*m*	7.96	7.96	7.96	90	90	90	This work
		8.09	8.09	8.09	90	90	90	Exp.[21]
$ZnAl_2O_4$	*Fd*$\bar{3}$*m*	8.05	8.05	8.05	90	90	90	This work
		8.09	8.09	8.09	90	90	90	Exp.[22]
$CdAl_2O_4$	*Fd*$\bar{3}$*m*	8.27	8.27	8.27	90	90	90	This work
		8.36	8.36	8.36	90	90	90	Exp.[8]
$SrAl_2O_4$	$P2_1$	8.39	8.73	5.09	90	93.17	90	This work

		8.45	8.82	5.16	90	93.42	90	Exp.[23]
$PbA1_2O_4$	*Ama*2	8.39	9.08	5.05	90	90	90	This work
		8.46	9.23	5.07	90	90	90	Exp.[23]
$BaA1_2O_4$	$P6_3$	10.34	10.34	8.73	90	90	120	This work
		10.45	10.45	8.79	90	90	120	Exp.[24]
$MgAl_2S_4$	*Pnma*	12.67	7.27	5.94	90	90	90	This work
		12.59	7.25	5.94	90	90	90	Exp.[25]
$ZnAl_2S_4$	*Fd*[illegible]*m*	10.17	10.17	10.17	90	90	90	This work
		10.01	10.01	10.01	90	90	90	Exp.[26]
$CdAl_2S_4$	*I*[illegible]	5.70	5.70	10.11	90	90	90	This work
		5.55	5.55	10.11	90	90	90	Exp.[27]
$CdAl_2S_4$	*Fd*[illegible]*m*	10.39	10.39	10.39	90	90	90	This work
		10.24	10.24	10.24	90	90	90	Exp.[28]
$SrAl_2S_4$	*Fddd*	21.15	20.57	12.18	90	90	90	This work
		21.05	20.58	12.21	90	90	90	Exp.[9]
$PbAl_2S_4$	*Cccm*	5.94	10.48	10.50	90	90	90	This work
		5.96	10.48	10.54	90	90	90	Exp.[14]
$BaAl_2S_4$	*Pa*3	12.75	12.75	12.75	90	90	90	This work
		12.65	12.65	12.65	90	90	90	Exp.[11]
$MgAl_2Se_4$	*R*[illegible]*m*	3.90	3.90	40.99	90	90	120	This work
		3.92	3.92	39.20	90	90	120	Exp.[29]

$ZnAl_2Se_4$	*I*$\bar{4}$*2m*	5.68	5.68	10.87	90	90	90	This work
		5.61	5.61	10.99	90	90	90	Exp.[8]
$ZnAl_2Se_4$	*Fd*$\bar{3}$*m*	10.71	10.71	10.71	90	90	90	This work
		10.69	10.69	10.69	90	90	90	Exp.[30]
$CdAl_2Se_4$	*I*$\bar{4}$*2m*	5.91	5.91	10.71	90	90	90	This work
		5.84	5.84	10.82	90	90	90	Exp.[8]
$SrAl_2Se_4$	*Cccm*	6.20	11.00	11.18	90	90	90	This work
		6.27	10.82	10.85	90	90	90	Exp.[31]
$PbAl_2Se_4$	*Cccm*	6.30	17.06	10.13	90	90	90	This work
		6.36	10.63	10.78	90	90	90	Exp.[32]
$BaAl_2Se_4$	*P*4/*nnc*	11.17	11.17	6.73	90	90	90	This work
		11.35	11.35	6.19	90	90	90	Exp.[16]
$ZnAl_2Te_4$	*I*$\bar{4}$*2m*	6.11	6.11	11.85	90	90	90	This work
		5.94	5.94	12.03	90	90	90	Exp.[8]
$CdAl_2Te_4$	*I*$\bar{4}$*2m*	6.31	6.31	11.78	90	90	90	This work
		6.01	6.01	12.21	90	90	90	Exp.[8]
$SrAl_2Te_4$	*I*4/*mcm*	8.41	8.41	6.72	90	90	90	This work
		8.25	8.25	6.75	90	90	90	Exp.[20]
$BaAl_2Te_4$	*P*4/*nbm*	8.63	8.63	6.72	90	90	90	This work
		8.69	8.69	6.76	90	90	90	Exp.[33]

Table S2. The carrier effective mass of AB_2Ch_4 (A^{2+}=Mg^{2+}, Zn^{2+}, Cd^{2+}, Sr^{2+}, Pb^{2+}, Ba^{2+}; B^{3+} = Ga^{3+}, Al^{3+}; Ch^{2-}=O^{2-}, S^{2-}, Se^{2-}, Te^{2-})

Compound	$m_h(m_0)$	$m_e(m_0)$
$MgGa_2O_4$	1.23	0.32
$ZnGa_2O_4$	1.36	0.33
$CdGa_2O_4$	1.30	0.32
$SrGa_2O_4$	7.69	0.41
$PbGa_2O_4$	3.70	1.09
$BaGa_2O_4$	16.67	1.28
$MgGa_2S_4$	1.30	0.35
$ZnGa_2S_4$	0.91	0.61
$CdGa_2S_4$	0.92	0.39
$SrGa_2S_4$	3.33	2.50
$PbGa_2S_4$	1.05	1.38
$BaGa_2S_4$	2.97	0.38
$ZnGa_2Se_4$	0.78	0.36
$CdGa_2Se_4$	0.73	0.27
$SrGa_2Se_4$	0.64	0.54
$PbGa_2Se_4$	0.86	1.38
$BaGa_2Se_4$	0.71	0.49
$ZnGa_2Te_4$	0.51	0.40
$CdGa_2Te_4$	0.71	0.68
$SrGa_2Te_4$	0.70	0.28
$BaGa_2Te_4$	1.32	0.72
$MgAl_2O_4$	1.59	0.48
$ZnAl_2O_4$	0.75	0.48
$CdAl_2O_4$	0.73	0.41
$SrAl_2O_4$	6.50	0.51
$PbAl_2O_4$	2.21	0.93

$BaA1_2O_4$	2.08	0.50
$MgAl_2S_4$	0.86	0.63
$ZnAl_2S_4$	0.57	0.70
$CdAl_2S_4(Fd\bar{3}m)$	0.52	0.67
$CdAl_2S_4(I\bar{4})$	1.10	0.46
$SrAl_2S_4$	4.28	3.31
$PbAl_2S_4$	0.96	0.75
$BaAl_2S_4$	1.22	0.87
$MgAl_2Se_4$	1.86	0.35
$ZnAl_2Se_4(Fd\bar{3}m)$	0.52	0.65
$ZnAl_2Se_4(I\bar{4})$	0.99	0.48
$CdAl_2Se_4$	1.03	0.44
$SrAl_2Se_4$	0.56	0.54
$PbAl_2Se_4$	-	-
$BaAl_2Se_4$	1.28	1.08
$ZnAl_2Te_4$	0.63	0.46
$CdAl_2Te_4$	0.84	0.89
$SrAl_2Te_4$	0.78	0.57
$BaAl_2Te_4$	1.26	0.55

Table S3. The elastic constants for 12 candidate materials.

C_{12}	C_{13}	C_{16}	C_{23}
27.02	32.07	-0.89	-
23.69	29.09	1.98	-
15.40	9.63	-	15.13
33.29	21.67	-	25.89
69.39	-	-	-
70.38	-	-	-
25.28	32.03	2.07	-
26.83	24.95	-	19.88
56.01	-	-	-
21.20	27.98	1.40	-
15.85	14.88	-	19.90
15.58	21.62	0.52	-

Compounnd	System	C_{11}	C_{22}	C_{33}	C_{44}	C_{55}	C_{66}
$ZnGa_2S_4$	Tetragonal	59.98	-	64.68	35.31	-	34.27
$CdGa_2S_4$	Tetragonal	53.14	-	49.16	28.97	-	26.55
$BaGa_2Se_4$	Orthorhombic	70.78	31.83	20.93	6.86	10.49	18.57
$MgAl_2S_4$	Orthorhombic	96.8	127.84	117.89	24.18	19.67	19.43
$ZnAl_2S_4$	Cubic	115.68	-	-	61.53	-	-
$CdAl_2S_4$	Cubic	101.96	-	-	52.65	-	-
$CdAl_2S_4$	Tetragonal	59.30	-	52.23	31.39	-	27.47
$PbAl_2S_4$	Orthorhombic	125.98	57.31	58.96	5.38	22.70	23.16
$ZnAl_2Se_4$	Cubic	92.43	-	-	49.20	-	-
$CdAl_2Se_4$	Tetragonal	51.49	-	47.36	28.24	-	24.12
$SrAl_2Se_4$	Orthorhombic	90.85	47.74	49.13	10.14	17.43	21.87
$CdAl_2Te_4$	Tetragonal	40.47	-	40.79	23.10	-	19.89

These 12 materials, covering cubic, tetragonal and orthorhombic crystal systems, must satisfy the following mechanical stability conditions, respectively:

$$C_{44} > 0,\ C_{11} - C_{12} > 0,\ C_{11} + C_{12} > 0$$

$$C_{44} > 0,\ |C_{11} - C_{12}| > 0,\ C_{33}(C_{11} + C_{12}) - 2C_{13}^2 > 0,\ C_{66}(C_{11} - C_{12}) - 2C_{16}^2 > 0$$

$$\begin{cases} C_{11} > 0,\ C_{44} > 0,\ C_{55} > 0,\ C_{66} > 0,\ C_{11}C_{22} - C_{12}^2 > 0, \\ C_{11}C_{22}C_{33} + 2C_{12}C_{13}C_{23} - C_{11}C_{23}^2 - C_{22}C_{13}^2 - C_{33}C_{12}^2 > 0 \end{cases}$$

Table S4. The enthalpy of formation for 12 candidate materials.

Compound	Group	E_f（eV/Cell）
$ZnGa_2S_4$	$I\bar{4}2m$	-0.73
$CdGa_2S_4$	$I\bar{4}2m$	-0.72
$BaGa_2Se_4$	*Cccm*	-1.11
$MgAl_2S_4$	*Pnma*	-1.82
$ZnAl_2S_4$	$Fd\bar{3}m$	-1.61
$CdAl_2S_4$	$Fd\bar{3}m$	-1.64
$CdAl_2S_4$	$I\bar{4}3m$	-1.58
$PbAl_2S_4$	*Cccm*	-1.66
$ZnAl_2Se_4$	$Fd\bar{3}m$	-1.64
$CdAl_2Se_4$	$I\bar{4}2m$	-1.51
$SrAl_2Se_4$	*Cccm*	-1.92
$CdAl_2Te_4$	$I\bar{4}2m$	-1.18

Table S5. The calculated deformation potential constant E and hole mobility μ_h for 12 screened candidates.

Compound	Space Group	E(eV)	μ_h
Ga_2O_3	*C*2/*m*	7.50	1.30
$CuAlO_2$	$R\bar{3}m$	-	10.40
ZGa_2S_4	$I\bar{4}3m$	30.98	5.21

$CdGa_2S_4$	*I*$\bar{4}2m$	24.82	6.50
$BaGa_2Se_4$	*Cccm*	1.41	1521.87
$MgAl_2S_4$	*Pnma*	4.40	198.08
$ZnAl_2S_4$	*Fd*$\bar{3}m$	13.09	20.71
$CdAl_2S_4$	*Fd*$\bar{3}m$	2.42	502.86
$CdAl_2S_4$	*I*$\bar{4}$	14.76	11.54
$PbAl_2S_4$	*Cccm*	3.51	283.84
$ZnAl_2Se_4$	*Fd*$\bar{3}m$	13.09	20.71
$CdAl_2Se_4$	*I*$\bar{4}2m$	13.23	16.71
$SrAl_2Se_4$	*Cccm*	3.01	1156.99
$CdAl_2Te_4$	*I*$\bar{4}2m$	9.59	41.62

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