Supplementary information

Quasi-1D Selenohalides: The First-principles Insights into Thermoelectric and Optoelectronic Applications

Prakash Govindaraj, Hern Kim[*]

Department of Energy Science and Technology, Environment Waste Recycle Institute, Myongji University, Yongin, Gyeonggi-do 17058, Republic of Korea.

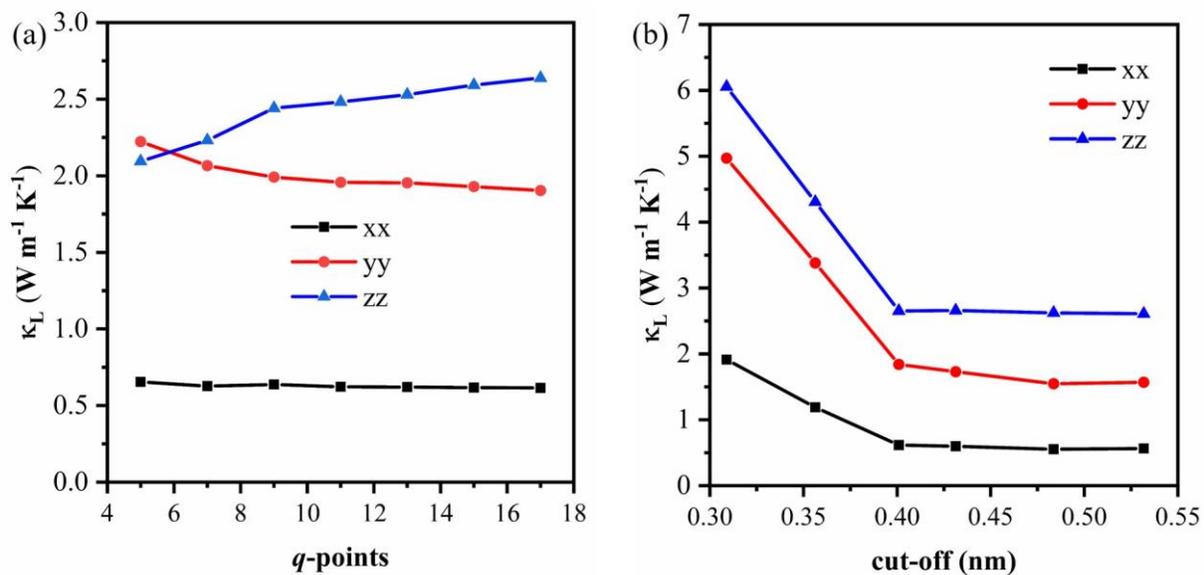* Corresponding Author E-mail: hernkim@mju.ac.kr

Figure S1 Convergence test of (a) q-points and (b) cutoff radius with respect to lattice thermal conductivity.
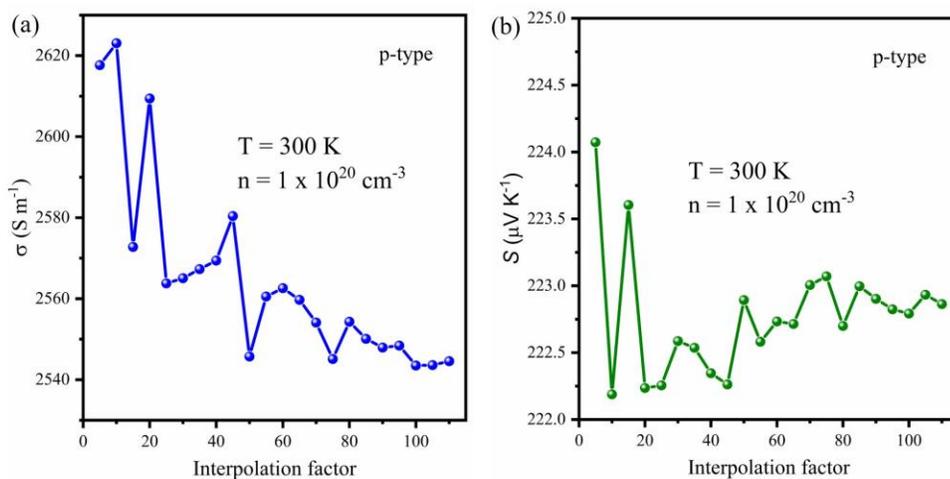
Figure S2 Convergence test of (a) electrical conductivity and (b) Seebeck coefficient with respect to interpolation factor using AMSET.
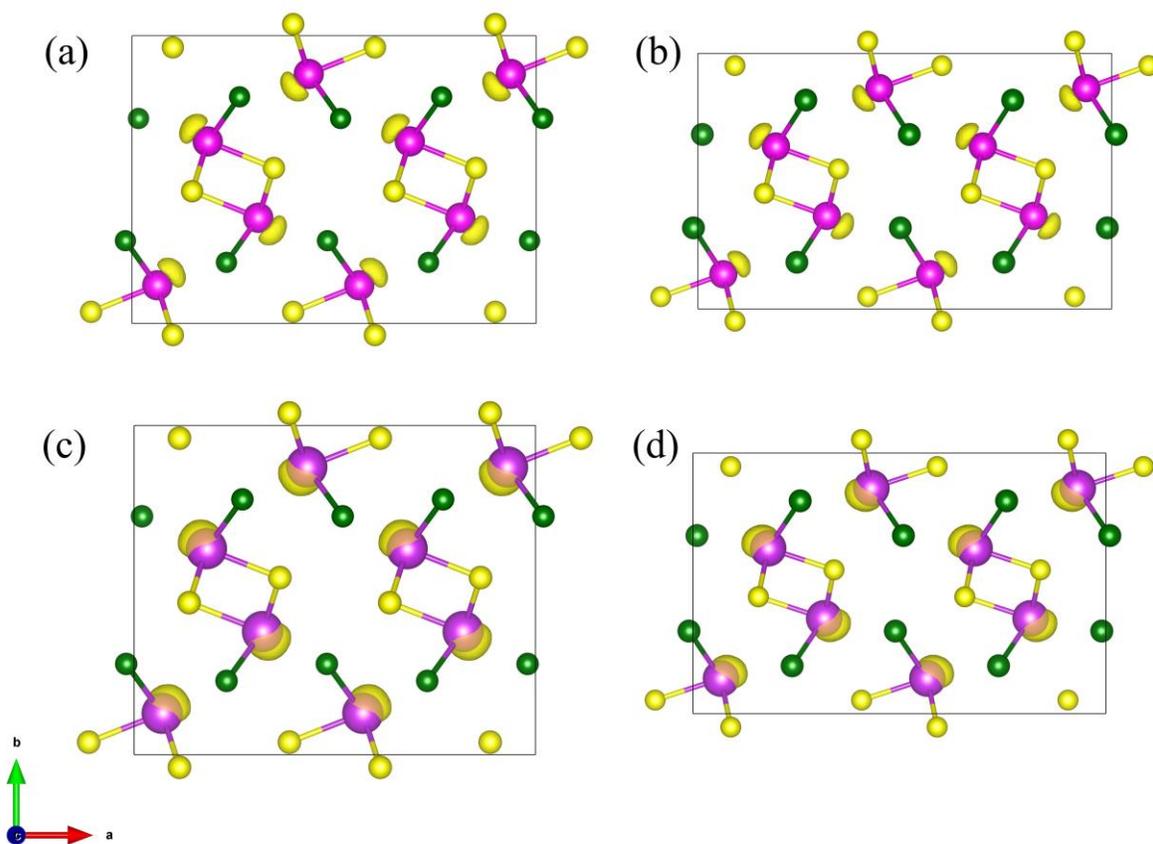

Figure S3 3D electron localization function of (a) SbSeCl, (b) SbSeBr, (c) BiSeCl and (d) BiSeBr.

Table S1. Bond lengths between atoms in the square pyramids. Here X is Sb/Bi atoms, Ha is Cl/Br atoms. b1, b2, b3, b4 and b5 are the bonds indexed in the main text.

	b1 (X-Ha)	b2 (X-Ha)	b3 (X-Se)	b4 (X-Se)	b5 (X-Se)
SbSeCl	2.84	2.84	2.66	2.77	2.77
SbSeBr	2.97	2.97	2.65	2.78	2.78
BiSeCl	2.94	2.94	2.78	2.83	2.83
BiSeBr	3.07	3.07	2.76	2.84	2.84

S1. Mechanical Properties

The bulk (B), shear (G) and Young's modulus (Y) is calculated using the following relation,

$$B = \frac{B_V + B_R}{2} \; ; G = \frac{G_V + G_R}{2} \; ; Y = \frac{9BG}{3B+G}$$

Here,

$$B_V = (1/9)[C_{11} + C_{22} + C_{33} + 2(C_{12} + C_{13} + C_{23})]$$

$$G_V = (1/15)[C_{11} + C_{22} + C_{33} + 3(C_{44} + C_{55} + C_{66}) - (C_{12} + C_{13} + C_{23})$$

$$B_R = \Delta /[C_{11}(C_{22} + C_{33} - 2C_{23}) + C_{22}(C_{33} - 2C_{13}) - 2C_{33}C_{12} + C_{12}(2C_{23} - C_{12}) + C_{13}(2C_{12} - C_{13}) + C_{23}(2C_{13} - C_{23})]$$

$$G_R = 15/\{4[C_{11}(C_{22} + C_{33} + 2C_{23}) + C_{22}(C_{33} + C_{13}) + C_{33}C_{12} - C_{12}(C_{23} + C_{12}) - C_{13}(C_{12} + C_{13}) - C_{23}(C_{13} + C_{23})]/\Delta + 3[(1/C_{44}) + (1/C_{55}) + (1/C_{66})]\}$$

$$\Delta = C_{13}(C_{12}C_{23} - C_{13}C_{22}) + C_{23}(C_{12}C_{13} - C_{23}C_{11}) + C_{33}(C_{11}C_{22} - C_{12}C_{12})$$

Cauchy's pressure is

$$CP = C_{11}\text{-}C_{44}$$

Poisson's ratio is given by,

$$\nu = \frac{3B - 2G}{2(3B + G)}$$

Table S2. Mechanical properties of the selenohalides.

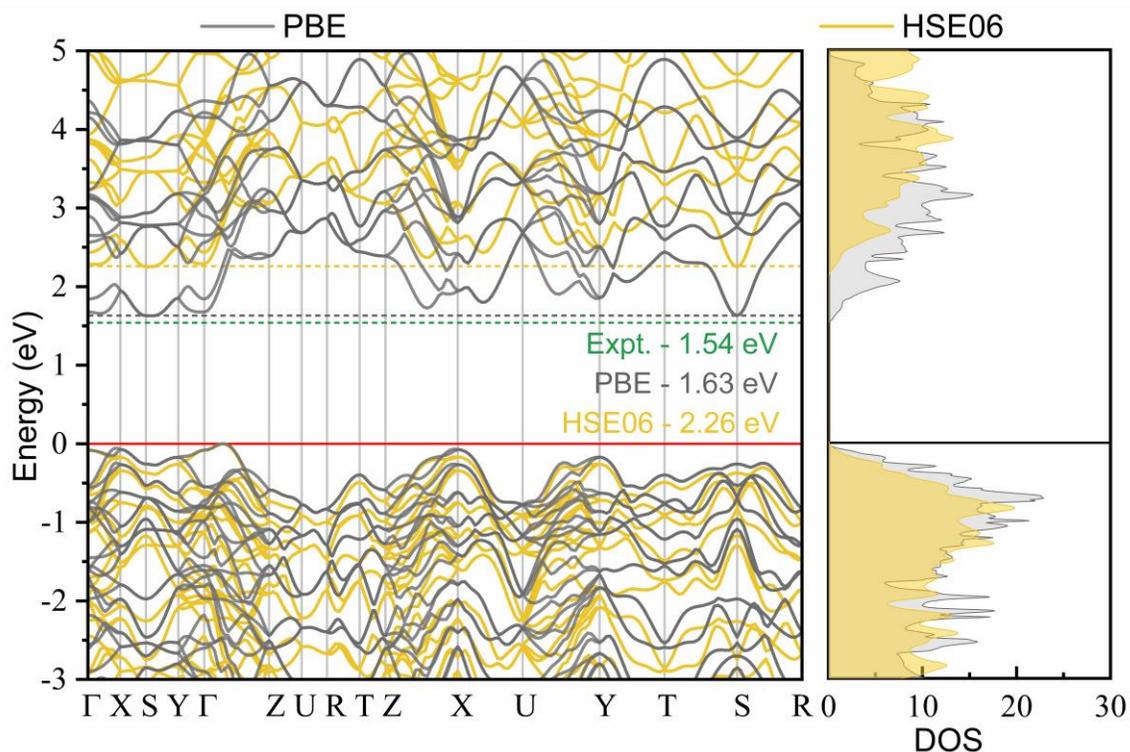|	SbSeCl	SbSeBr	BiSeCl	BiSeBr
B (GPa)	22.52498	21.93781	23.99261	22.79986
G (GPa)	8.850752	9.404773	10.21016	8.299202
Y (GPa)	23.47728	24.68659	26.82529	22.20356
B/G	2.54498	2.332625	2.349875	2.747235
CP (GPa)	7.21	14.91	4.68	18.4
ν	0.326287	0.31245	0.313656	0.337692

Figure S4 Calculated electronic band structures and corresponding DOS of BiSeBr obtained using the PBE and HSE06 exchange–correlation functionals. van der Waals corrections are included in both calculations.
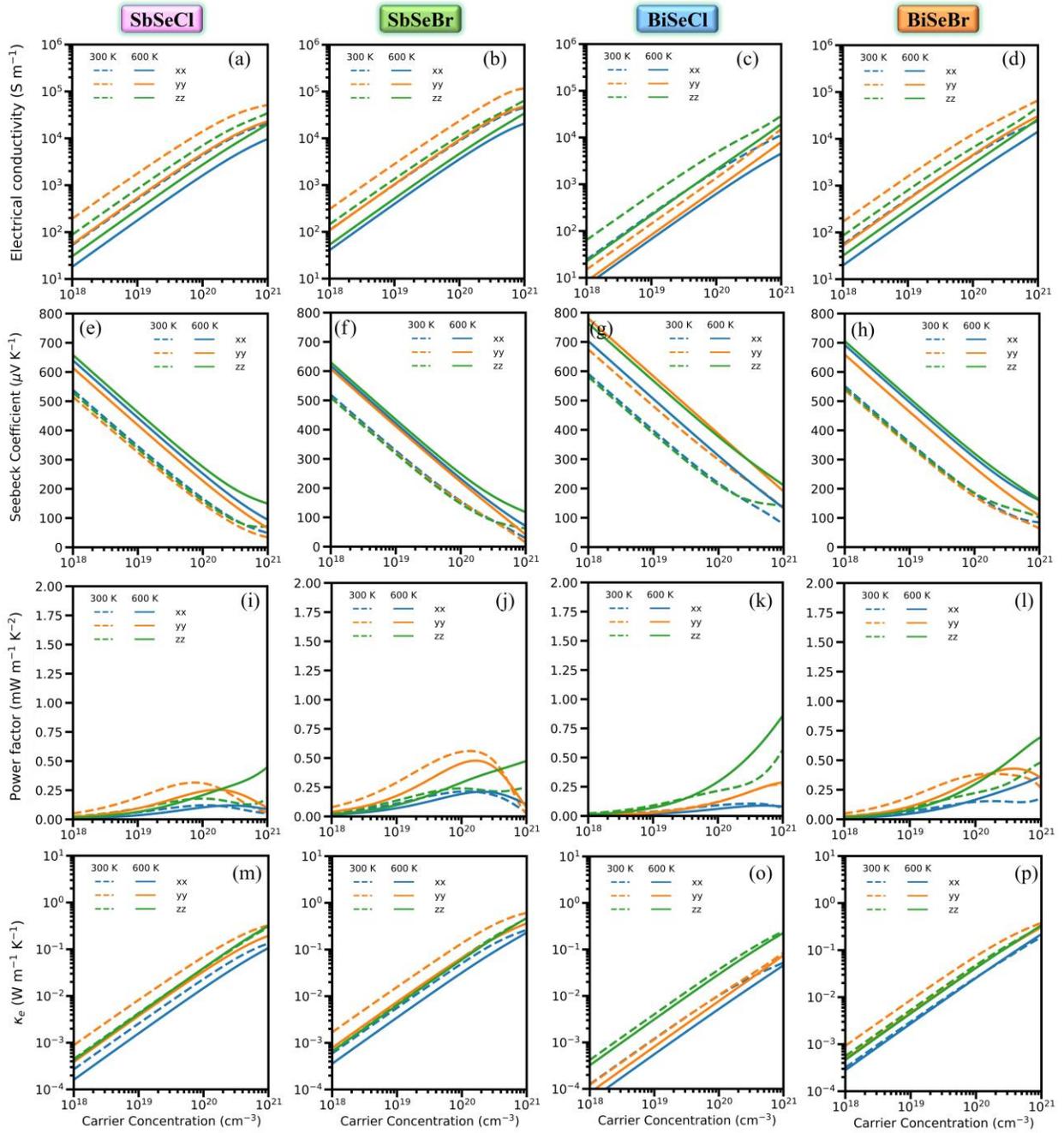
Figure S5 (a–d) Electrical conductivity, (e-f) Seebeck Coefficient, (i-l) power factor and (m-p) electronic thermal conductivity of p-type SbSeCl, SbSeBr, BiSeCl, and BiSeBr.

Table S3. Electrical conductivity for n-type selenohalides at the specific carrier concentration of 1×10^{20} cm^{-3}. The unit is S m^{-1}.

	300 K				600 K			
	SbSeCl	SbSeBr	BiSeCl	BiSeBr	SbSeCl	SbSeBr	BiSeCl	BiSeBr
x	8825.61	13323.97	15684.72	7229.224	3800.773	5931.317	7113.71	4335.728
y	8972.985	26009.83	20452.48	9141.523	4416.675	11597.3	9304.783	5112.293
z	57909.89	78757.27	42244.15	33968.34	30027.03	36834.56	21064.67	17742.3

Table S4. Electrical conductivity for p-type selenohalides at the specific carrier concentration of 1×10^{20} cm^{-3}. The unit is S m^{-1}.

	300 K				600 K			
	SbSeCl	SbSeBr	BiSeCl	BiSeBr	SbSeCl	SbSeBr	BiSeCl	BiSeBr
x	4331.316	8786.744	1914.624	4378.492	1611.724	3568.371	623.198	1762.869
y	14019.87	23531.04	1365.86	12520.69	4661.285	9282.936	800.314	4521.407
z	7023.693	11137.03	4772.11	6475.148	2733.624	4670.411	2062.668	2909.118

Table S5. Seebeck coefficient for n-type selenohalides at the specific carrier concentration of 1×10^{20} cm^{-3}. The unit is μV K^{-1}.

	300 K				600 K			
	SbSeCl	SbSeBr	BiSeCl	BiSeBr	SbSeCl	SbSeBr	BiSeCl	BiSeBr
x	-144.09	-83.0496	-114.422	-220.351	-192.549	-133.292	-196.633	-307.949
y	-153.85	-85.6708	-120.939	-213.934	-218.981	-135.658	-200.43	-299.805
z	-150.04	-84.3828	-136.671	-219.778	-205.307	-145.599	-219.326	-276.542

Table S6. Seebeck coefficient for p-type selenohalides at the specific carrier concentration of 1×10^{20} cm^{-3}. The unit is μV K^{-1}.

	300 K				600 K			
	SbSeCl	SbSeBr	BiSeCl	BiSeBr	SbSeCl	SbSeBr	BiSeCl	BiSeBr
x	166.9248	155.6958	218.4061	183.4347	252.0852	232.3713	311.5361	306.7411
y	148.9529	153.4066	298.0491	173.439	227.5969	221.3923	385.9259	272.5047
z	159.2332	146.7559	211.987	185.5563	275.2649	249.0351	376.6631	320.6294

Table S7. Power factor for n-type selenohalides at the specific carrier concentration of 1×10^{20} cm^{-3}. The unit is mW m^{-1} K^{-2}.

	300 K				600 K			
	SbSeCl	SbSeBr	BiSeCl	BiSeBr	SbSeCl	SbSeBr	BiSeCl	BiSeBr
x	0.18326	0.0919	0.20535	0.35101	0.14091	0.10538	0.27505	0.41117
y	0.2124	0.1909	0.29914	0.41839	0.21179	0.21343	0.37379	0.45951
z	1.30361	0.56079	0.78908	1.64075	1.26566	0.78086	1.01329	1.35685

Table S8. Power factor for p-type selenohalides at the specific carrier concentration of 1×10^{20} cm^{-3}. The unit is mW m^{-1} K^{-2}.

	300 K				600 K			
	SbSeCl	SbSeBr	BiSeCl	BiSeBr	SbSeCl	SbSeBr	BiSeCl	BiSeBr
x	0.12069	0.213	0.09133	0.14733	0.10242	0.19268	0.06048	0.16587
y	0.31106	0.55377	0.12133	0.37664	0.24146	0.455	0.1192	0.33575
z	0.17809	0.23986	0.21445	0.22295	0.20713	0.28965	0.29264	0.29907

Table S9. Electronic thermal conductivity for n-type selenohalides at the specific carrier concentration of 1×10^{20} cm^{-3}. The unit is W m^{-1} K^{-1}.

	300 K				600 K			
	SbSeCl	SbSeBr	BiSeCl	BiSeBr	SbSeCl	SbSeBr	BiSeCl	BiSeBr
x	0.042745	0.072197	0.105493	0.046576	0.026829	0.059058	0.078249	0.051414
y	0.051038	0.143625	0.133637	0.054713	0.041404	0.107536	0.101952	0.060775
z	0.32403	0.473347	0.297675	0.181895	0.262113	0.390439	0.236621	0.145122

Table S10. Electronic thermal conductivity for p-type selenohalides at the specific carrier concentration of 1×10^{20} cm^{-3}. The unit is W m^{-1} K^{-1}.

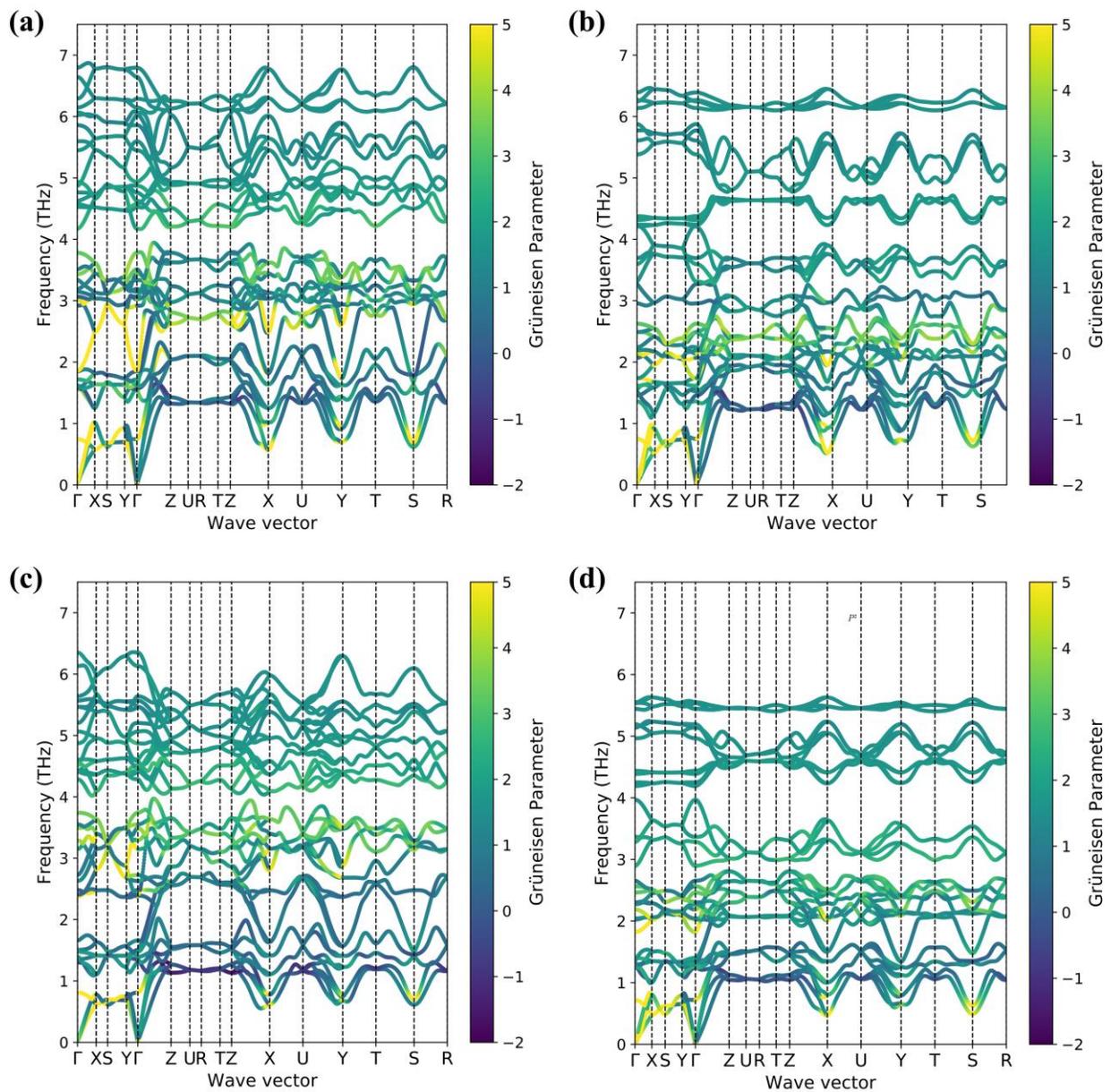|	300 K				600 K			
	SbSeCl	SbSeBr	BiSeCl	BiSeBr	SbSeCl	SbSeBr	BiSeCl	BiSeBr
x	0.02304	0.05028	0.010208	0.026275	0.014691	0.031875	0.005238	0.025645
y	0.068971	0.135538	0.01044	0.074233	0.033563	0.066155	0.007753	0.042469
z	0.038833	0.066385	0.03701	0.048057	0.039528	0.060446	0.029242	0.041744

Figure S6 Grüneisen-projected phonon dispersion for (a) SbSeCl, (b) SbSeBr, (c) BiSeCl and (d) BiSeBr.
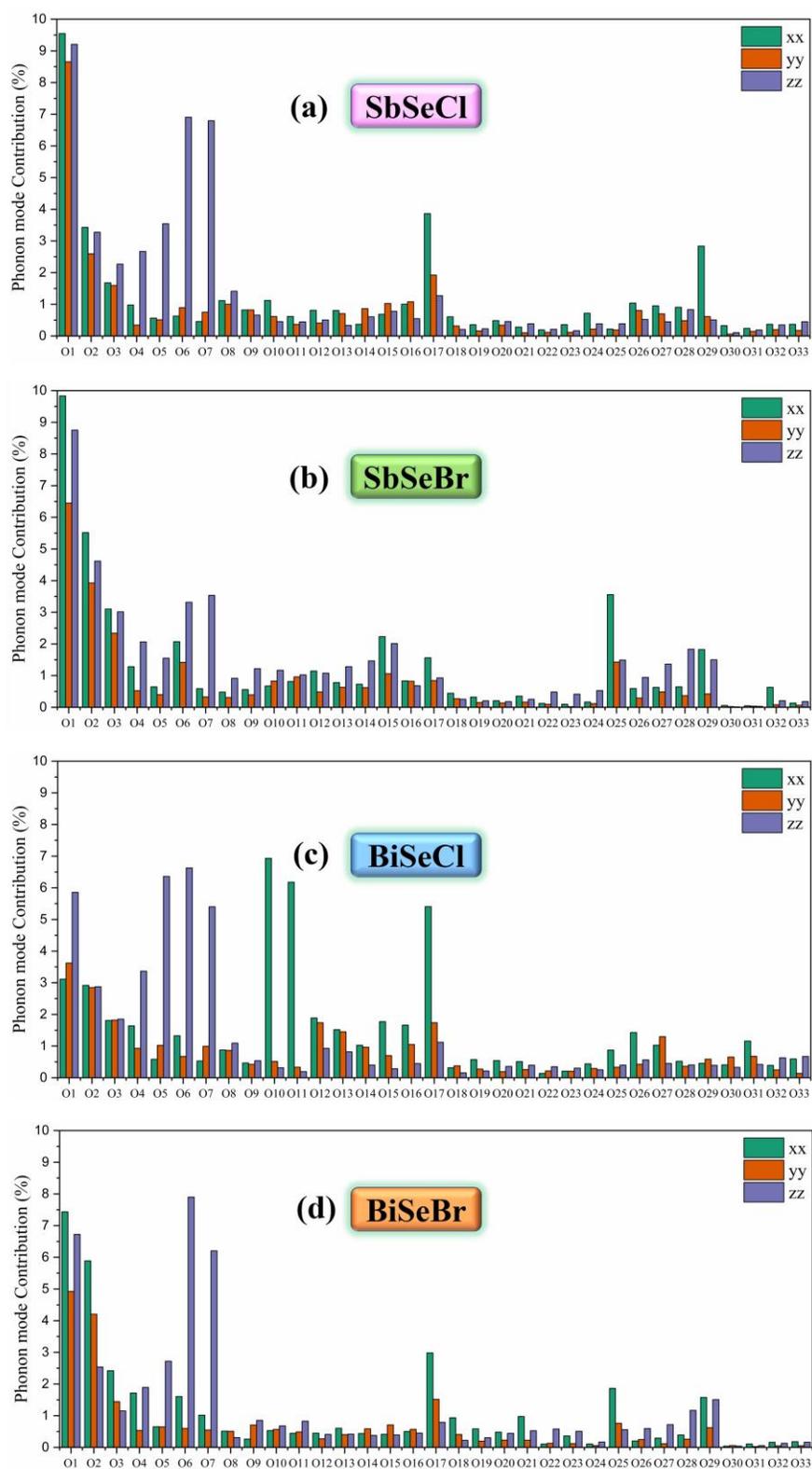
Figure S7 Percentage of phonon mode contribution to κ_L for (a) SbSeCl, (b) SbSeBr, (c) BiSeCl, and (d) BiSeBr.

Table S11 maximum zT of the selenohalides and corresponding carrier concentrations.

n-type

	T=300 K								T = 600 K							
	SbSeCl		SbSeBr		BiSeCl		BiSeBr		SbSeCl		SbSeBr		BiSeCl		BiSeBr	
	zT	n	zT	n	zT	n	zT	n	zT	n	zT	n	zT	n	zT	n
x	0.08	7.00E+19	0.05	3.00E+19	0.08	5.00E+19	0.18	3.00E+20	0.24	9.00E+20	0.17	4.00E+19	0.4	1.00E+20	0.81	3.00E+20
y	0.05	9.00E+19	0.05	3.00E+19	0.07	9.00E+19	0.11	2.00E+20	0.19	1.00E+20	0.19	5.00E+19	0.32	1.00E+20	0.53	3.00E+20
z	0.11	8.00E+19	0.07	3.00E+19	0.08	1.00E+20	0.22	2.00E+20	0.42	1.00E+20	0.27	5.00E+19	0.4	1.00E+20	0.71	2.00E+20

p-type

	T=300 K								T = 600 K							
	SbSeCl		SbSeBr		BiSeCl		BiSeBr		SbSeCl		SbSeBr		BiSeCl		BiSeBr	
	zT	n	zT	n	zT	n	zT	n	zT	n	zT	n	zT	n	zT	n
x	0.05	1.00E+20	0.08	1.00E+20	0.05	3.00E+20	0.07	1.00E+20	0.19	2.00E+20	0.28	1.00E+20	0.15	4.00E+20	0.42	8.00E+20
y	0.07	7.00E+19	0.11	1.00E+20	0.06	8.00E+20	0.09	1.00E+20	0.22	1.00E+20	0.38	1.00E+20	0.25	9.00E+20	0.36	3.00E+20
z	0.02	1.00E+20	0.02	1.00E+20	0.06	1.00E+21	0.06	1.00E+21	0.14	1.00E+21	0.14	8.00E+20	0.34	1.00E+21	0.29	8.00E+20