Exploring the A-Site Chemistry of $A_2LiTlCl_6$ (A= K, Rb, and Cs) Double Perovskites: Structural Stability, Electronic Structure, and Transport Properties

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Supplementary Information

**Table. S1** The calculated elastic constant and related parameters derived from elastic constants using GGA-PBEsol.

Parameters	$K_2LiTlCl_6$	$Rb_2LiTlCl_6$	$Cs_2LiTlCl_6$	Other work
C_{11}	47.6	43.7	42.6	42.7 [1], 66.9 [2], 39.3 [3]
C_{12}	16.1	15.8	15.12	16.52 [2], 16.8 [1], 13.7 [3]
C_{44}	12.1	11.6	12.2	21.98 [2], 9.10 [1], 8.7 [3]
$B = (C_{11}+2C_{12}) / 3$	26.5	25.1	24.29	32.5 [1], 29.9 [2], 22.2 [3]
$Y = 9BG_v / (3B + G_v)$	34.8	32.2	32.8	28.7 [1], 56.5 [2], 26.4 [3]
$G = (G_v + G_R) / 2$	13.5	12.5	12.8	23.2 [2], 7.29 [1], 10.1 [3]
B/G	1.97	2.01	1.89	1.43 [2], 3.10 [1], 2.19 [3]
$C_p = C_{12} - C_{44}$	4.0	4.18	2.92	5.05 [3], -5.46 [2]
$v = 3B - 2G / 2(3B + G)$	0.28	0.28	0.27	0.35 [1], 0.21 [2]
$A = 2 C_{44} / (C_{11}-C_{12})$	0.76	0.72	0.88	0.70 [1], 0.87 [2]
$T_m= (553K + (5.91\ K/GPa)\ C_{11}) \pm 300K$	(834 ± 300) K	(811.26 ± 300) K	(804.76 ± 300) K	(785.6 ± 300) K [3], (959.7 ± 300) K [4]
$V_t = \left(\dfrac{\rho}{G}\right)^{-\frac{1}{2}}$	2124.99	1905.8	1832.9	1581 [3], 2259.6 [4]
$V_l = \left(\dfrac{3B + 4G}{3\rho}\right)^{\frac{1}{2}}$	3865.12	3486.97	3290.5	2969 [3], 4086.7 [4]
$V_m = \left[\dfrac{1}{3}\left(\dfrac{2}{V_t^3} + \dfrac{1}{V_l^3}\right)\right]^{-\frac{1}{3}}$	2368.6	2125.3	2040.9	1766 [3], 2514.6 [4]
$\theta_D(K) = \dfrac{h}{k_B}\left[\dfrac{3nN_A\rho}{4\pi M}\right]^{\frac{1}{3}}V_m$	232.3	206.6	195.6	274 [3], 249.5 [4]
$\gamma = 3(1+v) / 2(2-3v)$	1.65	1.65	1.60	0.94 [4], 0.91 [4]

Table S2. The optical and transport properties such as; (σ/τ) electrical conductivity, (S) Seebeck coefficient, (κ_e/τ) electronic thermal conductivity, and (ZT) Figure of merit of $K_2LiTlCl_6$, $Rb_2LiTlCl_6$ and $Cs_2LiTlCl_6$ were computed mBJ-GGA-PBEsol functional

	Materials Properties		$K_2LiTlCl_6$	$Rb_2LiTlCl_6$	$Cs_2LiTlCl_6$	Other work
Optical Properties	$\varepsilon_1(0)$		2.31	2.32	2.40	2.93 [5], 2.41 [6]
	n (0)		1.52	1.52	1.54	1.71 [5], 1.55 [6]
	R (0)		0.042	0.043	0.046	0.07 [5], 0.05 [6]
Transport properties	S (μV/K)	300 K	222.67	208.53	202.71	170 [7], 250 [8]
		800 K	193.61	185.69	189.13	238 [8], 147 [7]
	$\kappa_e\ 10^{14}$ (W/mKs)	300 K	0.98	0.96	0.93	0.11 [8], 0.7 [7]
		800 K	5.68	5.11	5.28	2.1 [7], 3.8 [8]
	$\sigma/\tau\ 10^{18}$ $(\Omega ms)^{-1}$	300 K	0.50	0.55	0.56	0.46 [7], 0.15 [8]
		800 K	1.39	1.32	1.32	0.89 [7], 0.59 [8]
	$S^2\sigma/\tau$ $(10^{11}W/K^2ms)$	300 K	2.51	2.43	2.31	1.5 [7], 1.25 [8]
		800 K	5.24	4.56	4.73	1.9 [7], 3.8 [8]
	ZT	300 K	0.76	0.75	0.74	0.75 [8],0.62 [7]
		800 K	0.73	0.71	0.71	0.78 [8], 0.21 [7]
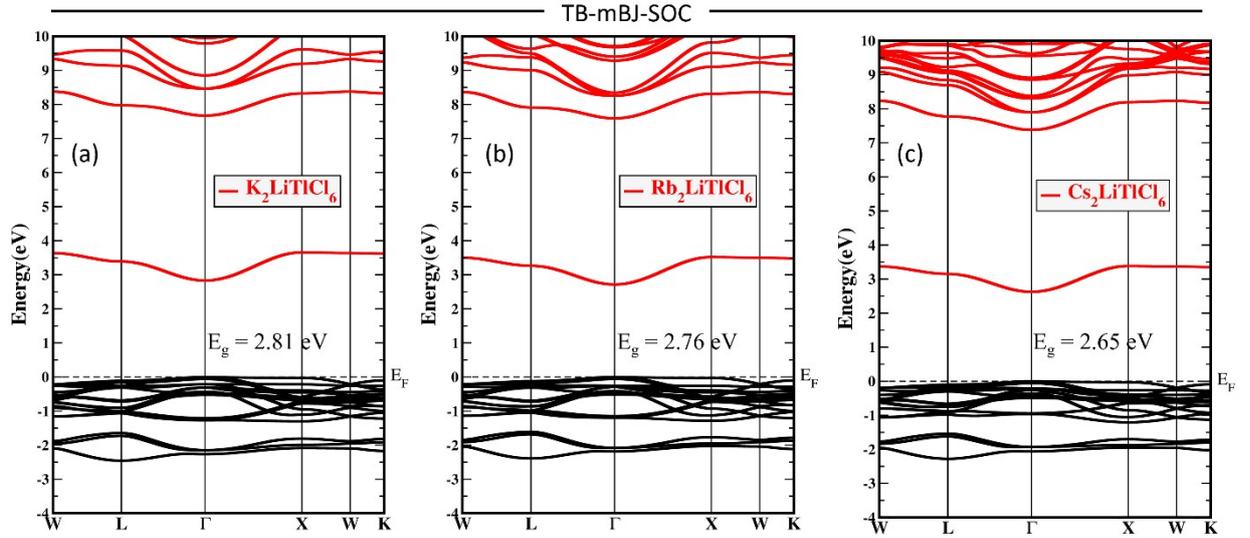
Figure. S1 (a–c) TB-mBJ-SOC calculated band structures of $K_2LiTlCl_6$, $Rb_2LiTlCl_6$, and $Cs_2LiTlCl_6$ with band gaps of 2.81, 2.71, and 2.62 eV, respectively.
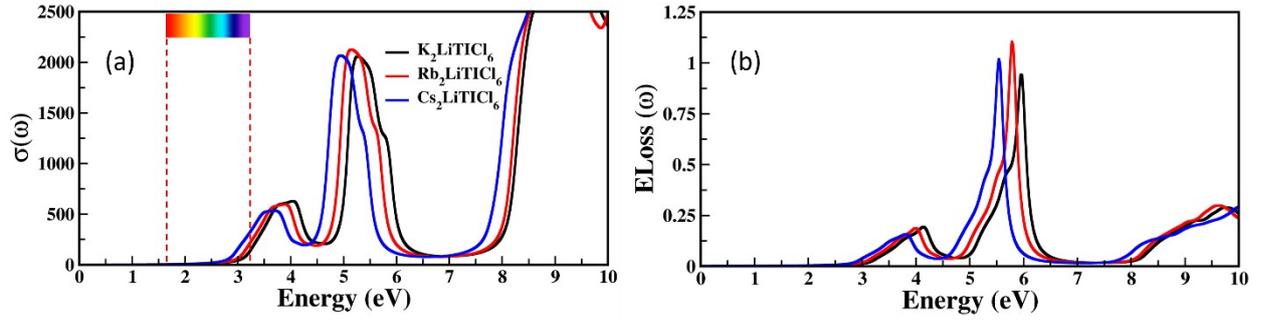
Figure. S2 The plot of (a) Optical conductivity: $\sigma(\omega)$, (b) Energy Loss: ELoss (ω) of $A_2LiTlCl_6$ (A= K, Rb, and Cs).
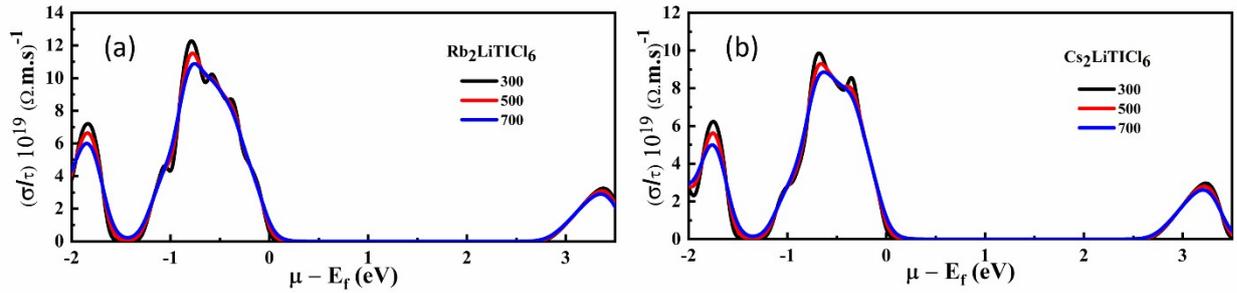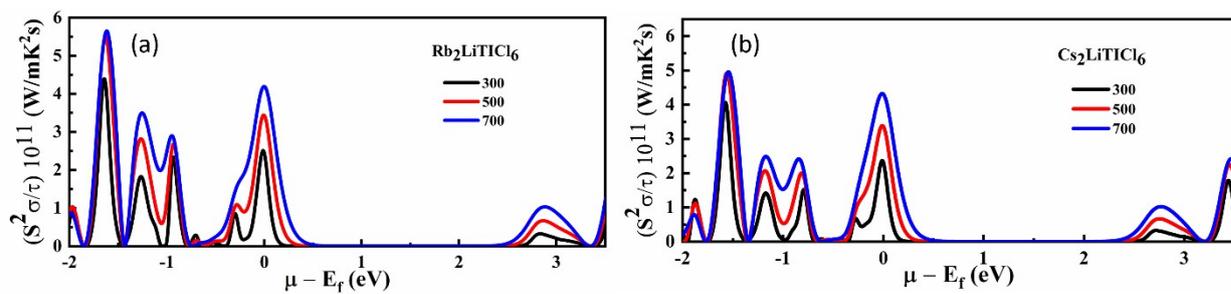

Figure. S3 Calculated electrical conductivity presented as a function of chemical potential between -2 and +3.5 eV of $A_2LiTlCl_6$ (A= Rb, and Cs).
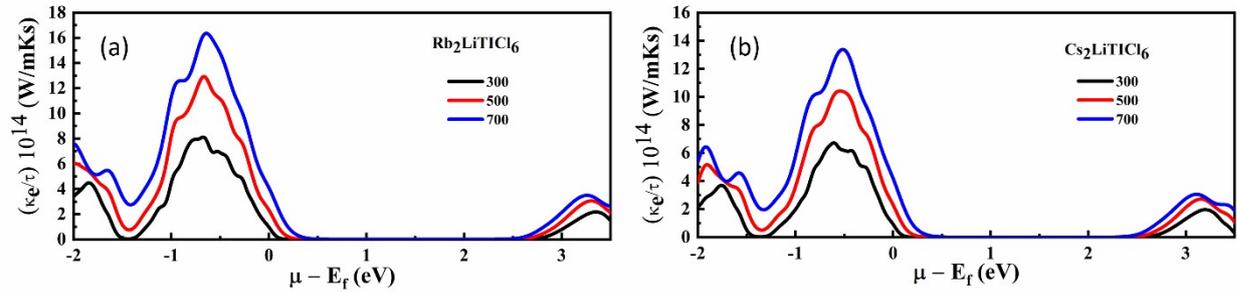

Figure. S4 Calculated electronic thermal conductivity presented as a function of chemical potential between -2 and +3.5 eV of $A_2LiTlCl_6$ (A= Rb, and Cs).
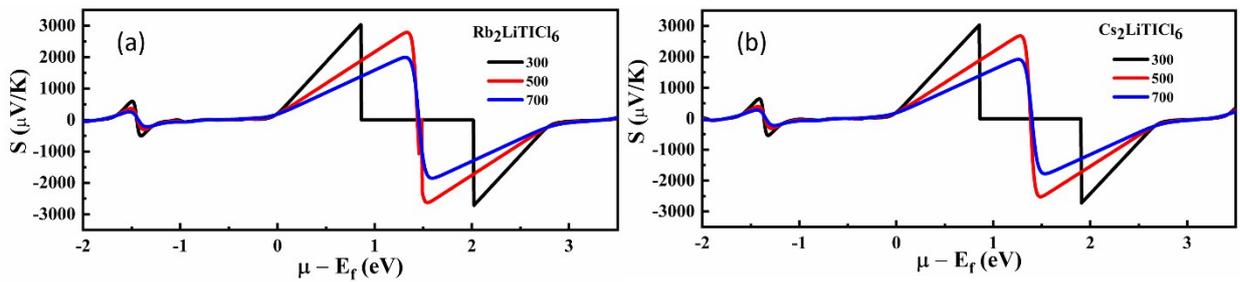
Figure. S5 Calculated Seebeck coefficient presented as a function of chemical potential between -2 and +3.5 eV of $A_2LiTlCl_6$ (A= Rb, and Cs).

Figure. S6 Calculated power factor presented as a function of chemical potential between -2 and +3.5 eV of $A_2LiTlCl_6$ (A= Rb, and Cs).
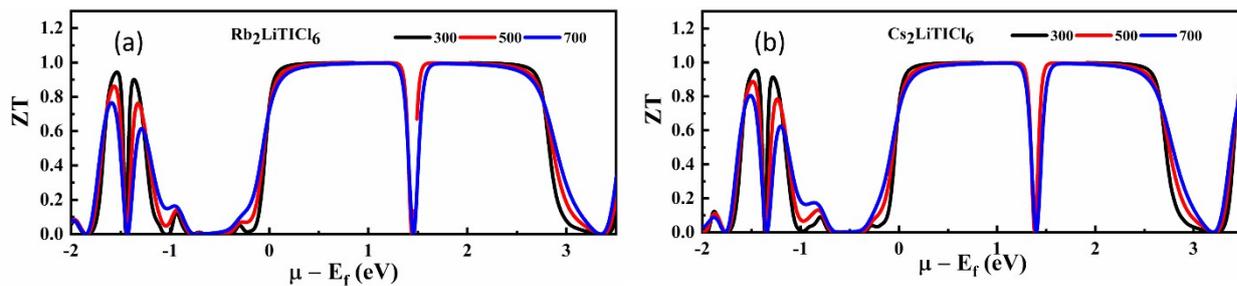
Figure. S7 Calculated Figure of merit presented as a function of chemical potential between -2 and +3.5 eV of $A_2LiTlCl_6$ (A= Rb, and Cs).

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