Relativistic electronic structure and photovoltaic performance of K_2CsSb

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S1 Lattice parameter comparison

Table S1: Comparison of lattice parameters with experimental and calculation results from other works

a (Å)	Diff (%)	Method	Ref.
8.610	—	Experiment	1
8.553	-0.670	PBEsol	This work
8.560	-0.584	FP_LAPW	2
8.508	-1.199	FP_LAPW	3
8.730	1.375	PBE	4
8.530	-0.938	mBJ+soc	4
8.760	1.712	GW	5

S2 Phonon band structure

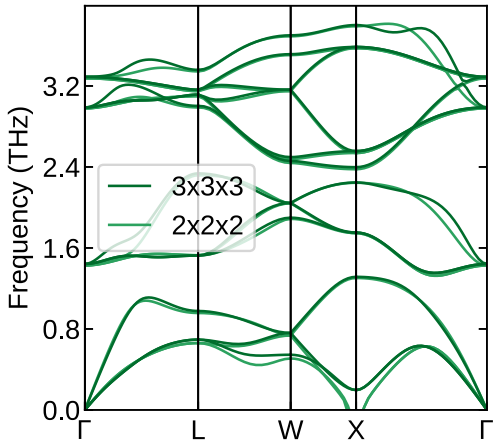

Figure S1: Convergence of the phonon band structure of K_2CsSb with supercell size, calculated using the PBEsol exchange–correlation functional.

S3 Secondary phases

Table S2: Competing phases and total energies used to calculate the energy above hull. All materials were relaxed using the PBEsol exchange-correlation functional

Compound	**Energy (eV/f.u.)**
Cs	-0.938
Cs_2Sb_4	-7.774
Cs_3Sb	-8.934
Cs_5Sb_8	-45.801
CsSb	-6.430
K_3Sb	-9.498
K_5Sb_4	-27.669
K	-1.121
KSb	-6.528
KSb_2	-11.115
Sb	-4.557
K_2CsSb	-9.433

S4 Density of states

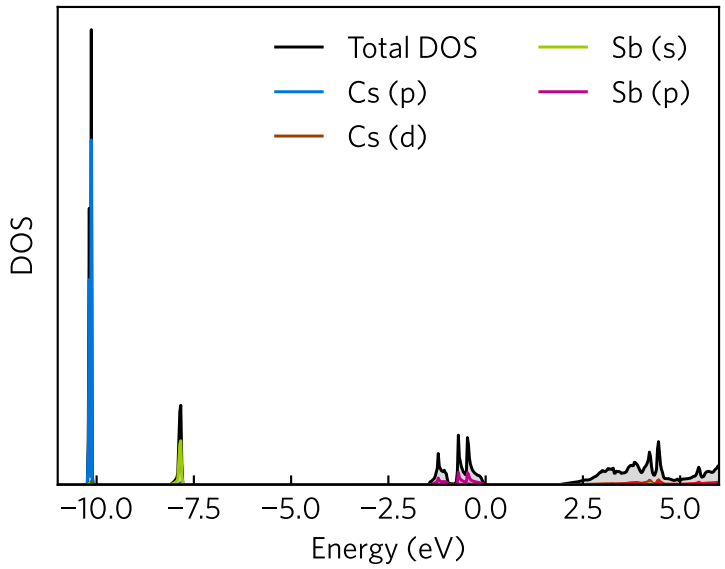

Figure S2: Density of states of K_2CsSb calculated using HSE+SOC plotted to deeper energies.

S5 Electron affinity constrained detailed balance metric

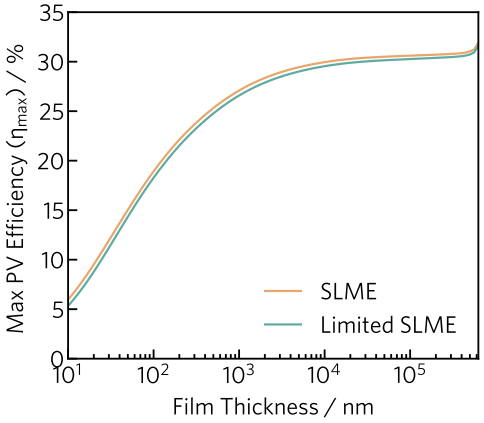

Figure S3: Spectroscopic limited maximum efficiency (SLME) metric for K_2CsSb, adjusted such that the absorption at energies greater than the ionisation potential (3.4 eV) is set to zero.

S6 Optical absorption convergence

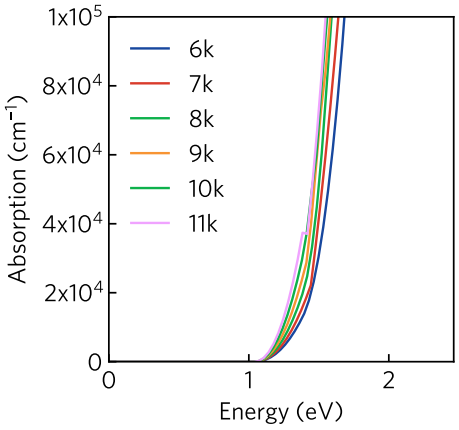

Figure S4: Convergence of optical absorption with respect to k-point mesh. Convergence was performed using the PBEsol functional.

S7 Thickness-dependent efficiency estimation by the Blank method

The detailed balance metric proposed by Blank et al.[6] is an extension of the Shockley-Queisser (SQ) detailed balance limit in which the complex refractive index and non-radiative recombination are taken into account. The SQ limit assumes a step absorption $A(E) = 0$ for $\leq E_g$ and $A(E) = 1$ for energies above the band gap, E_g, an ideality factor of $n_{id} = 1$, and gives the radiative efficiency limit as

$$\eta^{rad} = \frac{\max_V(V\{J_{SC} - J_0^{rad}\left[\exp\left(\frac{qV}{kT}\right) - 1\right]\})}{\int_0^\infty E\phi_{sun}(E)dE} \tag{S1}$$

, where J_0^{rad} is the SQ radiative saturation current, J_{SC} is the SQ short-circuit current, V is the voltage of the solar cell, q is the elementary charge, k is the Boltzmann constant, T is the temperature, and ϕ_{sun} is the AM1.5g solar spectrum.

The Blank metric differs from the SQ limit by using the calculated absorbance rather than a step function. Furthermore, two light-trapping schemes are introduced. The flat model simulates an idealised structure with flat front and back surfaces, with the reflectance set to zero and unity, respectively. The Lambertian model is identical to the flat approach but assumes a Lambertian scatter at the front surface. The full details are provided in the work by Blank et al.[6]

References

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