Supporting Information

Unraveling the Influence of Organic Cations on Tuning Electronic Structures and Spin-Splitting in Two-Dimensional Layered Organic−Inorganic Tin-Iodine Perovskites

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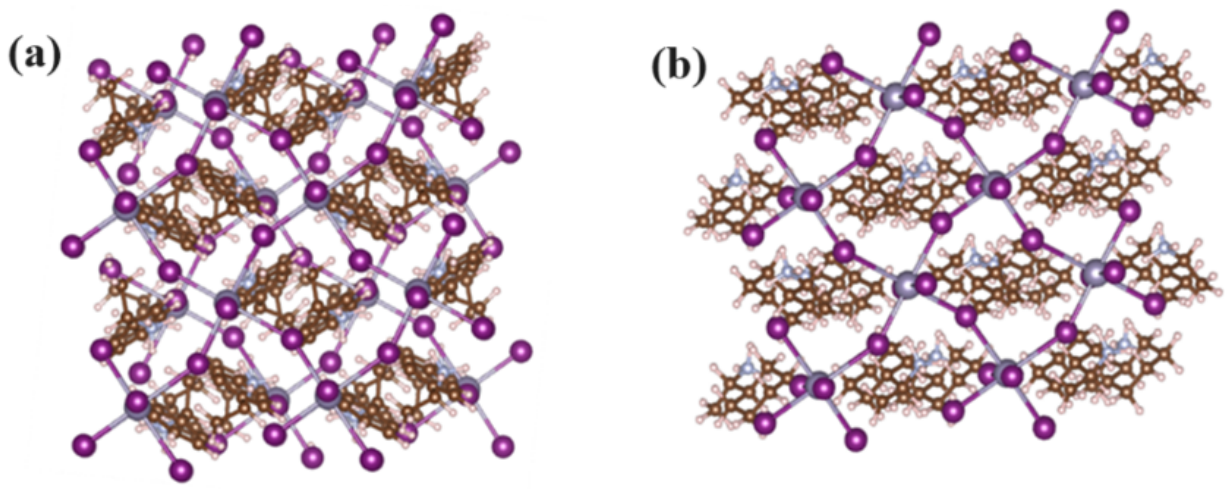

Figure S1. a-b) Top view of $(S\text{-}MBA)_2SnI_4$, and $(S\text{-}CYHEA)_2SnI_4$ crystals, respectively, showing staggered configuration for $(S\text{-}MBA)_2SnI_4$, and eclipsed configuration for $(S\text{-}CYHEA)_2SnI_4$.

Table S1: Sn-I bond lengths for $(S\text{-}MBA)_2SnI_4$, and $(S\text{-}CYHEA)_2SnI_4$ and their average of distortions.

Compound	Bond	Length (A˚)	Δd
$(S\text{-}MBA)_2SnI_4$	$Sn\text{-}I_{ax(1)}$	3.186 A˚	2.701×10^{-3}
	$Sn\text{-}I_{ax(4)}$	3.156A˚	
	$Sn\text{-}I_{eq(2)}$	3.100A˚	
	$Sn\text{-}I_{eq(3)}$	3.048A˚	
	$Sn\text{-}I_{eq(5)}$	3.514A˚	
	$Sn\text{-}I_{eq(6)}$	3.408A˚	
$(S\text{-}CYHEA)_2SnI_4$	$Sn\text{-}I_{ax(1)}$	3.143A˚	2.124×10^{-3}
	$Sn\text{-}I_{ax(4)}$	3.430A˚	
	$Sn\text{-}I_{eq(2)}$	3.265A˚	
	$Sn\text{-}I_{eq(3)}$	3.365A˚	
	$Sn\text{-}I_{eq(5)}$	3.043A˚	
	$Sn\text{-}I_{eq(6)}$	3.054A˚	

Table S2: I-Sn-I bond angle for $(S\text{-}MBA)_2SnI_4$, and $(S\text{-}CYHEA)_2SnI_4$ and their average of distortions.

Compound	Bond	Angle (deg)	σ^2 (deg^2)
$(S\text{-}MBA)_2SnI_4$	$I_{(1)} - Sn - I_{(2)}$	85.33°	9.10
	$I_{(1)} - Sn - I_{(3)}$	91.64°	
	$I_{(1)} - Sn - I_{(5)}$	92.60°	
	$I_{(1)} - Sn - I_{(6)}$	87.53°	
	$I_{(4)} - Sn - I_{(2)}$	86.79°	
	$I_{(4)} - Sn - I_{(3)}$	92.22°	
	$I_{(4)} - Sn - I_{(5)}$	95.15°	
	$I_{(4)} - Sn - I_{(6)}$	88.55°	
	$I_{(2)} - Sn - I_{(3)}$	92.71°	
	$I_{(2)} - Sn - I_{(6)}$	86.97°	
	$I_{(3)} - Sn - I_{(5)}$	89.05°	
	$I_{(5)} - Sn - I_{(6)}$	91.23°	
$(S\text{-}CYHEA)_2\text{-}SnI_4$	$I_{(1)} - Sn - I_{(2)}$	88.79°	10.79
	$I_{(1)} - Sn - I_{(3)}$	95.86°	
	$I_{(1)} - Sn - I_{(5)}$	89.45°	
	$I_{(1)} - Sn - I_{(6)}$	85.03°	
	$I_{(4)} - Sn - I_{(2)}$	87.83°	
	$I_{(4)} - Sn - I_{(3)}$	93.77°	
	$I_{(4)} - Sn - I_{(5)}$	93.71°	
	$I_{(4)} - Sn - I_{(6)}$	85.33°	
	$I_{(2)} - Sn - I_{(3)}$	91.37°	
	$I_{(2)} - Sn - I_{(6)}$	88.76°	
	$I_{(3)} - Sn - I_{(5)}$	89.86°	
	$I_{(5)} - Sn - I_{(6)}$	90.01°	

Table S3: Hydrogen bond for (S- MBA)$_2$SnI$_4$ and (S- CYHEA)$_2$SnI$_4$

Compound	H-bond	Length (A˚)
(S-MBA)$_2$SnI$_6$	NH···I$_{eq}$	2.89 A˚
	NH···I$_{eq}$	3.06 A˚
	NH···I$_{ax}$	2.49 A˚
	NH···I$_{ax}$	2.65 A˚
	NH···I$_{ax}$	2.48 A˚
	NH···I$_{ax}$	2.61 A˚
(S-CYHEA)$_2$SnI$_6$	NH···I$_{eq}$	2.85 A˚
	NH···I$_{eq}$	2.69 A˚
	NH···I$_{ax}$	2.45 A˚
	NH···I$_{ax}$	2.60 A˚
	NH···I$_{ax}$	2.57A˚
	NH···I$_{ax}$	2.41A˚

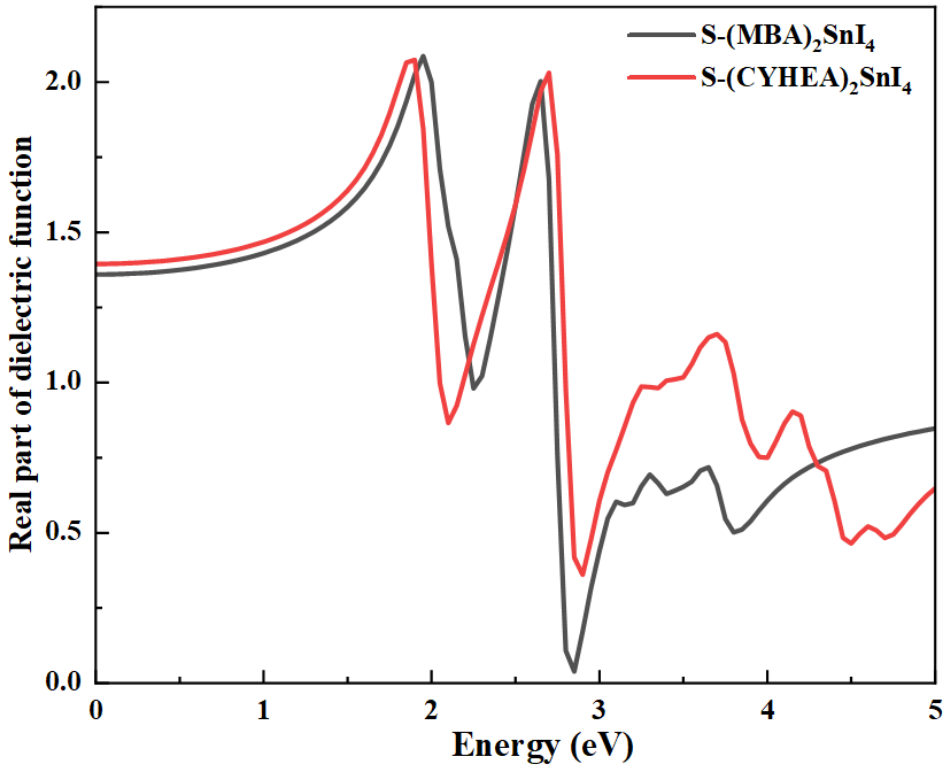

Figure S2. The real part of the dielectric function of S-$(MBA)_2SnI_4$ and S-$(CYHEA)_2SnI_4$

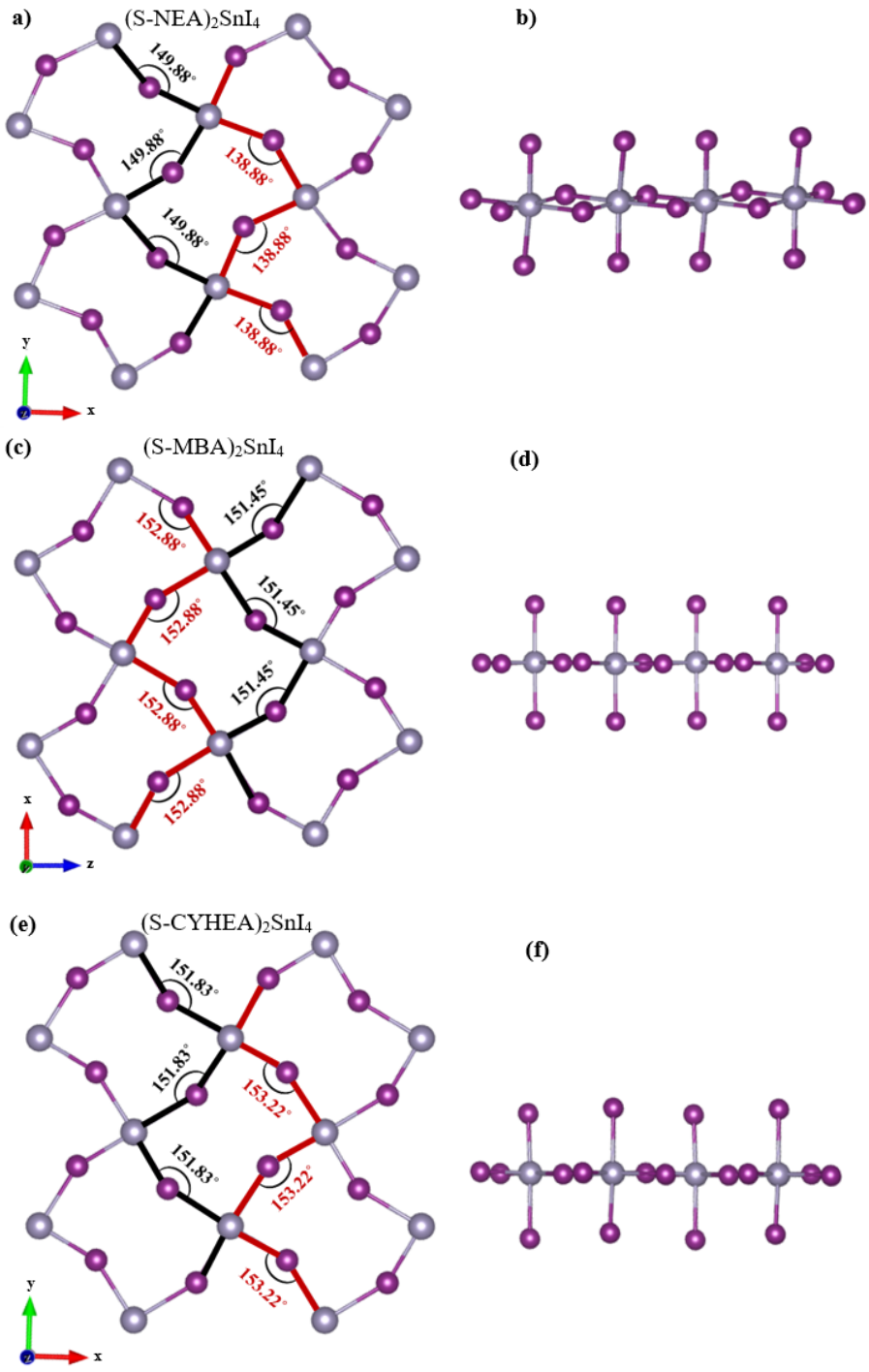

Figure S3: Illustration of the bond angle labels used in geometry comparison between perovskites in a) S-$(NEA)_2SnI_4$ show two different bond angles. In contrast (c) S- $(MBA)_2SnI_4$ and (e) S-$(CYHEA)_2SnI_4$ exhibit two very similar bond angles, denoted as red and black colors. b, c and f) Side view of the inorganic layer.

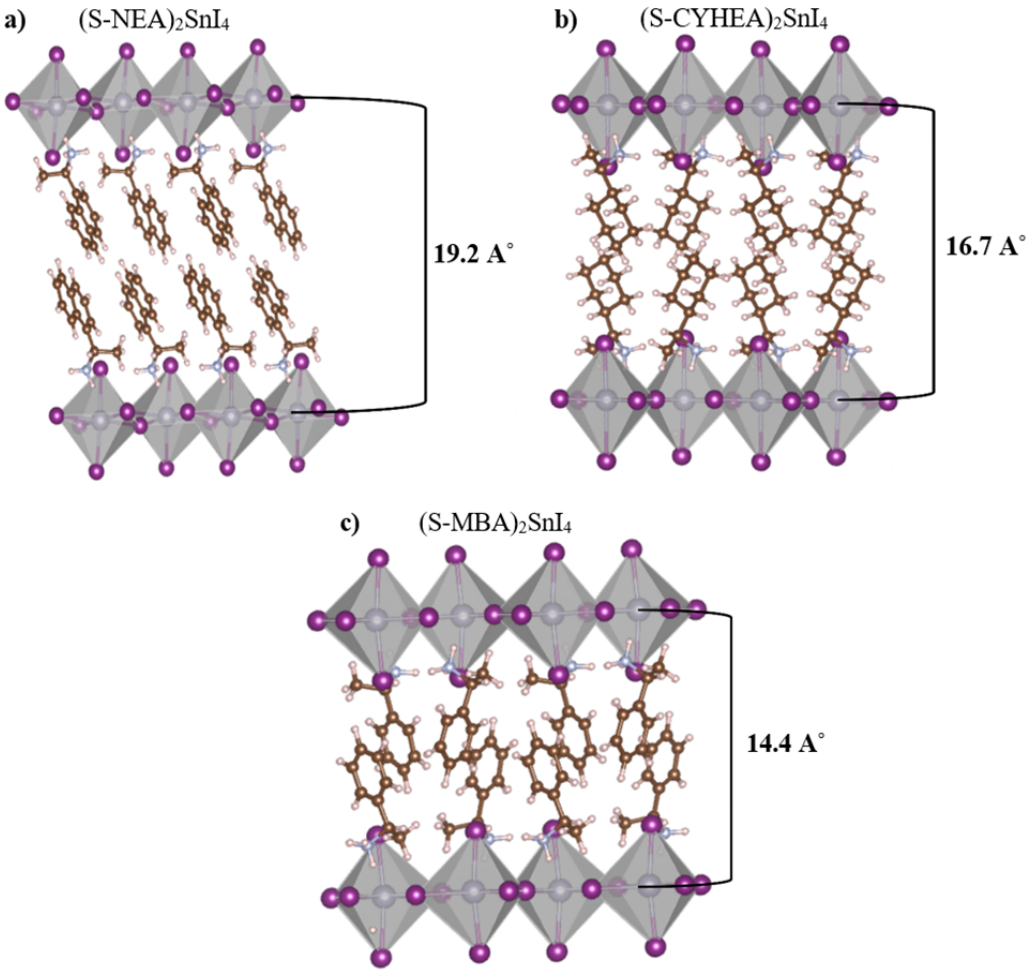

Figure S4: Schematic illustration of d-spacing between inorganic layers for a) $(NEA)_2SnI_4$; b) S-$(CYHEA)_2SnI_4$; and c) S-$(MBA)_2SnI_4$.