Supplemental Material for "Functionality-directed Screening of Pb-free Hybrid Organic-inorganic Perovskites with Desired Intrinsic Photovoltaic Functionalities"

Dongwen Yang,[†,‖] Jian Lv,[†,‖] Xingang Zhao,[†] Qiaoling Xu,[†] Yuhao Fu,[†] Yiqiang Zhan,[‡] Alex Zunger,[*,¶] and Lijun Zhang[*,†,§]

†Department of Materials Science and Engineering and Key Laboratory of Automobile Materials of MOE, Jilin University, Changchun 130012, China

‡State Key Laboratory of ASIC and System, Department of Microelectronics, SIST, Fudan University, Shanghai 200433, China

¶University of Colorad.13097(d..3097(2 111)0.13097(n)0.3083 21(d)-349.928(K)-R.14(3 2)-1.217 1(S)0.30


APbI3
APbBr3
APbCl3
45
30
(a)

Figure Band structures of the se ected $AM^{IV}X_3^{VII}$ perovs ites having indirect band gaps he va ence band axi u and conduction band ini u are ar ed by red circ es he actua band gaps E_g and direct band gaps E_g^d

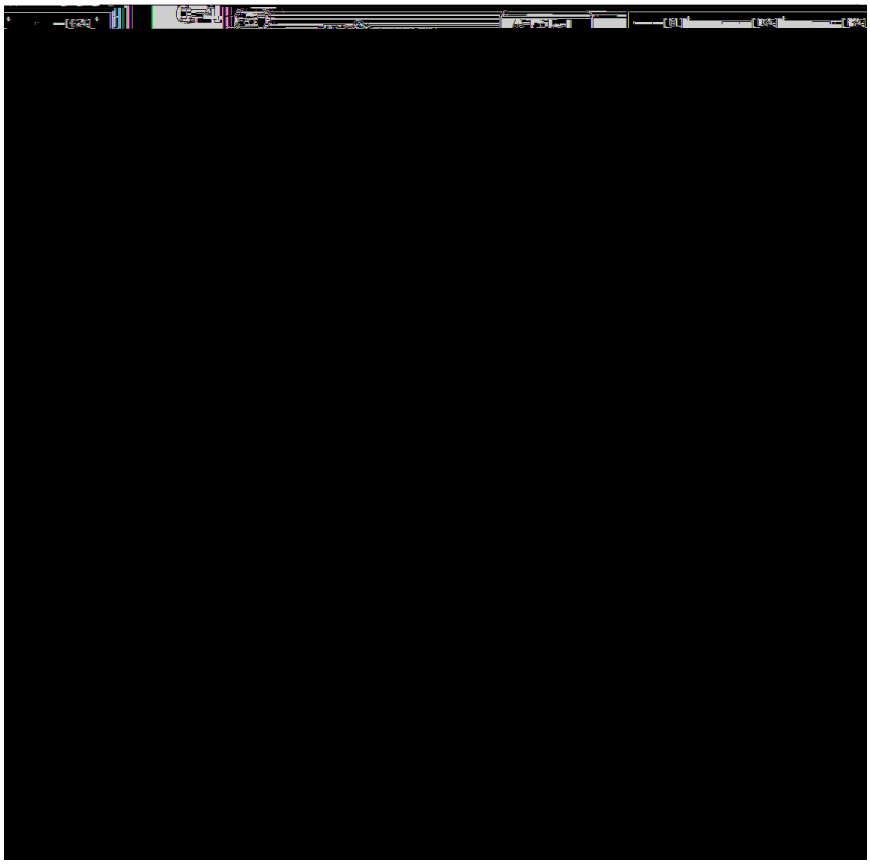
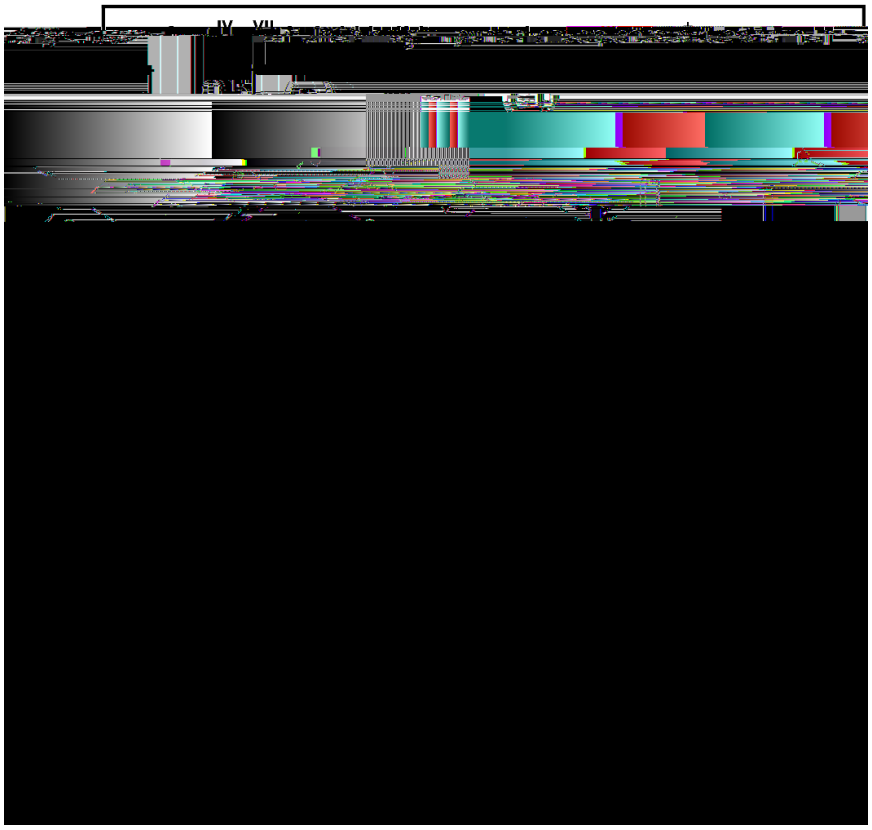


Figure Crysta orbita over ap popu ations COOP of the $AM^{IV}X_3^{VII}$ perovs ites For co parison, the CBM of each ateria is set to energy zero



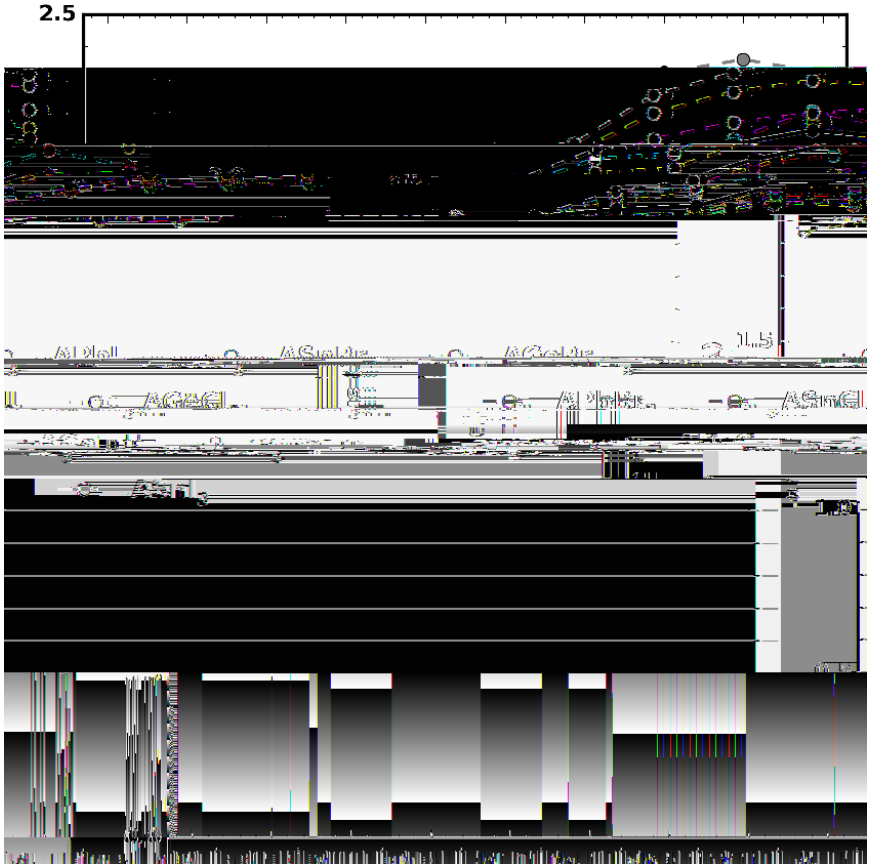


Figure 0 Eva uation of the steric sizes of organic o ecu ar cations within the idea ized so id sphere ode see the Experi enta ection iv

ab e 1 Ca cu ated deco position entha pies H of the candidate $\mathrm{AM}^{IV}\mathrm{X}_3^{VII}$

ab e Ca cu ated direct band gaps E_g^d of the candidate $AM^{IV}X_3^{VII}$ perovs ites he green shading indicates the co pounds passing the current DM $E_g^d <$ e , as we as the DM in ab e ′ he ightb ue shading indicates the co pounds passing on y the current DM

E_g^d e	PbI_3	$PbBr_3$	PbC_3	nI_3	nBr_3	nC_3	GeI_3	$GeBr_3$	GeC_3
[M $^+$									

ab e Ca cu ated exciton binding energies E_B of the candidate $AM^{IV}X_3^{VII}$ perovs ites he green shading indicates the co pounds passing the current DM $E_B <$ e , as we as the DMs in ab es , and he ightb ue shading indicates the co pounds passing on y the current DM

E_B e	PbI_3	$PbBr_3$	PbC_3	nI_3	nBr_3	nC_3	GeI_3	$GeBr_3$	GeC_3
M^+									
Cs^+									
HA^+									
DA^+									
MA^+									
FM^+									
FA^+									
EA^+									
GA^+									
DEA^+									

ab e Ca cu ated various DMs for the $AM^{IV}X_3^{VII}$ perovs ites containing pseudo ha ogen anions, AM^{IV} $BF_{4\ 3}$ and AM^{IV} $CN_{\ 3}$ with A Cs^{+}, MA^{+}, FA^{+} and M^{IV} Pb^{2+}, n^{2+}

Materia s	H	E_g^d	$^{*}_{e}$	$^{*}_{h}$	E_B e	α_{ex} n
	Co pounds based on $BF_4{}^{-}$					
CsPb $BF_{4\ 3}$						
Cs n $BF_{4\ 3}$						
MAPb $BF_{4\ 3}$						
MA n BF4						
	C pounds based on CN^{-}					
CsPb $CN_{\ 3}$						