## Recent progress in organic-inorganic halide perovskite solar cells: mechanisms and materials design

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## Supplementary information



Figure S1. Scheme of energy level of electrodes: FTO,<sup>1, 2</sup> ITO,<sup>3-5</sup> Al,<sup>3, 5</sup> Au,<sup>1</sup>  $Ag^2$  and  $Carbon^{6, 7}$ ; ETMs:  $TiO_2$ ,<sup>1, 8-11</sup>  $ZnO^{10, 12}$  and  $PCBM^{3-5}$ ; perovskites:  $CH_3NH_3SnI_3$ ,<sup>13</sup>  $NH_2CHCH_2PbI_3$ ,<sup>14</sup>  $CH_3NH_3PbI_3$ ,<sup>8, 10, 15</sup>  $CH_3NH_3PbI_3$ ,<sup>2</sup> $xCI_x$ ,<sup>10, 11</sup>  $CH_3NH_3SnBr_3$ ,<sup>13</sup>  $NH_2CHNH_2PbBr_3$ <sup>1</sup> and  $CH_3NH_3PbBr_3$ ,<sup>9</sup>; HTMs: spiro-OMeTAD,<sup>8, 10, 12, 13</sup> P3HT,<sup>10, 12, 14</sup> PTAA,<sup>15</sup> PEDOT:PSS,<sup>3, 4</sup>  $CuSCN^{10, 12, 16}$  and  $NiO^{2, 5}$ .

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Table S1. A summary of hybrid perovskite solar cells performance parameters with different anodes. (In order of the appearance in main text)

	Metho	J <sub>SC</sub>	V <sub>OC</sub>	FF	PCE
Structure (from substrate to back electrode)	ds	[mA/c	[V]	[%]	[%]
		m <sup>2</sup> ]			
Glass-FTO/c-TiO <sub>2</sub> /m-ZrO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /spiro/Ag <sup>153</sup>	SDM	17.3	1.07	59	10.8
Glass-FTO/c-TiO <sub>2</sub> /nr-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /spiro/Au <sup>154</sup>	OSPD	15.6	0.955	63	9.4
Glass-FTO/c-TiO <sub>2</sub> /nw-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /spiro/Au <sup>155</sup>	OSPD	10.67	0.74	54	4.29
Glass-FTO/c-TiO <sub>2</sub> /nf-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /spiro/Au <sup>156</sup>	SDM	15.88	0.98	63	9.82
Glass-FTO/c-TiO <sub>2</sub> /nt-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /I <sup>-</sup> ,I <sup>3-</sup> /Pt-FTO <sup>157</sup>	SDM	17.9	0.63	57.8	6.52
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> (15nm)	OSPD	19.6	1.04	66	12.8
/CH <sub>3</sub> NH <sub>3</sub> Pb(I <sub>0.9</sub> Br <sub>0.1</sub> ) <sub>3</sub> /PTAA/Au <sup>158</sup>					
Glass-FTO/c-TiO <sub>2</sub> /ns-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /spiro/Au <sup>159</sup>	SDM	19.25	0.963	65	12.30
Glass-FTO/hc-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /spiro/Au <sup>161</sup>	SDM	18.74	0.93	72	12.56
Glass-FTO/np-TiO <sub>2</sub> ,TiAcAc	OSPD	21.5	1.02	71	15.9
/m-Al <sub>2</sub> O <sub>3</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-x</sub> Cl <sub>x</sub> /spiro/Ag <sup>22</sup>					
Glass-FTO/np-TiO <sub>2</sub> ,graphene/m-Al <sub>2</sub> O <sub>3</sub> /	OSPD	21.9	1.04	73	15.6
CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-x</sub> Cl <sub>x</sub> /spiro/Au <sup>21</sup>					
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> , graphene quantum dots	OSPD	17.06	0.937	63.5	10.15
/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /spiro/Au <sup>162</sup>					
Glass-FTO/m-TiO <sub>2</sub> /ALD-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /spiro/Au <sup>163</sup>	SDM	17.64	0.969	67	11.5
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> ,HOCO-(CH <sub>2</sub> ) <sub>3</sub> -NH <sub>3</sub> <sup>+</sup> I <sup>-</sup> /	SDM	19.2	1.00	62	12.0
CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /spiro/Ag,Au <sup>164</sup>					
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /Y <sub>2</sub> O <sub>3</sub> /	OSPD	16.55	0.79	-	7.53
CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-x</sub> Cl <sub>x</sub> /spiro/Ag,Au <sup>165</sup>					
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /Sb <sub>2</sub> S <sub>3</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /CuSCN/Au <sup>166</sup>	OSPD	17.04	0.56	53	5.03
Glass-FTO/rutile c-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /spiro/Au <sup>167</sup>	SDM	19.8	1.05	64	13.7
Glass-ITO/np-TiO <sub>2</sub> , TiAcAc/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-x</sub> Cl <sub>x</sub> /P3HT/Ag <sup>168</sup>	OSPD	21.0	0.936	69.1	13.6
Glass-FTO /c-TiO <sub>2</sub> /rutile m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /spiro/Au <sup>169</sup>	SDM	20.02	1.022	71	14.46
Glass-FTO/c-TiO <sub>2</sub> /m-Y-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /spiro/Au <sup>170</sup>	OSPD	18.1	0.945	66	11.2
Glass-FTO/c-TiO <sub>2</sub> /m-Mg-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /P3HT/Au <sup>171</sup>	OSPD	10.4	0.802	50	4.17
Glass-FTO/c-Al-TiO <sub>2</sub> /m-Al <sub>2</sub> O <sub>3</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-x</sub> Cl <sub>x</sub> /spiro/Ag <sup>172</sup>	OSPD	20.00	1.07	65	13.8
Glass-ITO/PEIE/c-Y-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-x</sub> Cl <sub>x</sub> /spiro/Au <sup>4</sup>	OSPD	22.75	1.13	75.01	19.3
Glass-FTO /c-ZnO/nr-ZnO/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /spiro/Ag <sup>173</sup>	OSPD	12.7	0.68	58	5.0
Glass-FTO /c-ZnO/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /spiro/Au <sup>174</sup>	SDM	11.27	1.08	45.44	5.54
Glass-FTO/c-ZnO/nr-ZnO/CH3NH3PbI3/spiro/Au174	SDM	16.98	1.02	51.11	8.9

DET ITO/a 7nO/nr 7nO/CH NIL DbL /mira/Aul74	SDM	7.50	0.80	42.14	262
$\frac{PE1-I10/C-ZII0/III-ZII0/CH_3NH_3P0I_3/Spilo/Au^{22}}{Cl}$	SDM	10.60	0.80	45.14	2.02
$\frac{\text{Glass-F1O/c-ZnO/m-Al}_{2}\text{O}_{3}/\text{CH}_{3}\text{NH}_{3}\text{Pbl}_{3}/\text{spiro}/\text{Au}^{132}}{\text{Glass-F1O/c-ZnO/m-Al}_{2}\text{O}_{3}/\text{CH}_{3}\text{NH}_{3}\text{Pbl}_{3}/\text{spiro}/\text{Au}^{132}}$	SDM	19.68	0.975	41.66	/.86
Glass-FTO/c-ZnO/nr-ZnO/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /spiro/Au <sup>175</sup>	SDM	20.08	0.991	56	11.13
Glass-FTO/c-TiO <sub>2</sub> /nr-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /spiro/Au <sup>175</sup>	SDM	20.92	0.869	55	10.02
Glass-FTO/c-TiO <sub>2</sub> /nc-ZnO/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /spiro/Au <sup>176</sup>	SDM	16	0.718	41.2	4.8
Glass-ITO/np-ZnO/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /spiro/Ag <sup>177</sup>	SDM	20.4	1.03	74.9	15.7
PET-ITO/np-ZnO/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /spiro/Ag <sup>177</sup>	SDM	13.4	1.03	73.9	10.2
Glass-FTO/c-Al-ZnO/m-Al-ZnO/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /spiro/Ag <sup>178</sup>	SDM	15.1	1.045	76	12.0
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /C <sub>60</sub> SAM/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-x</sub> Cl <sub>x</sub> /spiro	OSPD	19.6	0.84	72	11.7
/Ag <sup>180</sup>					
Glass-ITO/PEDOT:PSS/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /PCBM/BCP/Al <sup>179</sup>	OSPD	10.32	0.60	63	3.9
Glass-ITO/PEDOT:PSS/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /PCBM/Al <sup>38</sup>	OSPD	8.74	0.92	76	6.16
Glass-ITO/PEDOT:PSS/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /PCBM/Al <sup>103</sup>	SDM	10.829	0.905	75.6	7.41
Glass-ITO/PEDOT:PSS/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-x</sub> Cl <sub>x</sub>	OSPD	15.8	0.94	66	9.8
/PCBM/c-TiO <sub>2</sub> /Al <sup>181</sup>					
Glass-ITO/PEDOT:PSS/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-x</sub> Cl <sub>x</sub> /PCBM/Al <sup>181</sup>	OSPD	17.2	0.84	-	11.5
Glass-ITO/PEDOT:PSS/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-x</sub> Cl <sub>x</sub> /PCBM/Al <sup>32</sup>	OSPD	14.3	0.86	60.9	7.5
Glass-FTO/PEDOT:PSS/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-x</sub> Cl <sub>x</sub> /PCBM/Bis-	OSPD	17.5	0.92	73	11.8
C <sub>60</sub> /Ag <sup>36</sup>					
Glass-ITO/PEDOT:PSS/polyTPD /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /PCBM/Al <sup>182</sup>	DSVD	16.12	1.05	0.67	12.04
Glass-ITO/PEDOT:PSS/polyTPD/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /PCBM/Au <sup>183</sup>	DSVD	18.8	1.07	0.63	12.7
Glass-ITO/PEDOT:PSS/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /ICBA/C <sub>60</sub> /BCP/Al <sup>184</sup>	OSPD	15.7	0.97	80.1	12.2
Glass-ITO/PEDOT:PSS/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /PCBM/LiF/Al <sup>185</sup>	OSPD	20.7	0.866	78.3	14.1
Glass-ITO/PEDOT:PSS/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /ICBA/C <sub>60</sub> /BCP/Al <sup>49</sup>	IDM	19.6	0.99	79.3	15.4
Glass-ITO/PEDOT:PSS/polyTPD/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /PCBM/Au <sup>186</sup>	DSVD	18.2	1.09	75	14.8
Glass-ITO/PEDOT:PSS/polyTPD/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub>	DSVD	14.2	0.7	56	5.5
/3TPYMB/Au <sup>186</sup>					
Glass-ITO/polythiophene/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /C <sub>60</sub> /BCP/Ag <sup>187</sup>	SDM	16.2	1.03	70.7	11.8
PET-ITO/PEDOT:PSS/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-x</sub> Cl <sub>x</sub> /PCBM/Al <sup>181</sup>	OSPD	16.5	0.86	64	9.2
PET-ITO/PEDOT:PSS/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-x</sub> Cl <sub>x</sub>	OSPD	14.4	0.88	51	6.4
/PCBM/c-TiO <sub>2</sub> /Al <sup>189</sup>					
PET/AZO/Ag/AZO/PEDOT:PSS/polyTPD	DSVD	14.3	1.04	47	7
/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /PCBM/Au <sup>190</sup>					

Notes: For structures: spiro is short for spiro-OMeTAD while c-, hc-, n-,m-, nr-, nw-, nf-, nt- and ns- of TiO<sub>2</sub> refer to compact, high compact, nanoparticle, mesoporous, nanorod, nanowire, nanofiber, nanotube and nanosheet structure, respectively. For perovskite synthesis methods: SDM and OSPD mean sequential deposition and one step precursor deposition methods, respectively; while DSVD and IDM mean dual-source vapour deposition and interdiffusion deposition methods, respectively.

Table S2. A summary of hybrid perovskite solar cells performance parameters with different cathodes. (In order of the appearance in main text)

	Methods	J <sub>SC</sub>	V <sub>OC</sub> [V]	FF	PCE
Structure (from substrate to back electrode)		[mA/c		[%]	[%]
		m <sup>2</sup> ]			
Glass-FTO /c-TiO <sub>2</sub> /ns-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /Au <sup>17</sup>	OSPD	16.1	0.6316	57	5.5
Glass-FTO /c-TiO <sub>2</sub> /np-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /Au <sup>191</sup>	OSPD	18.8	0.712	60	8
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /Au <sup>192</sup>	SDM	19	0.84	68	10.85
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /Au <sup>194</sup>	SDM	17.8	0.905	65	10.49
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /Au <sup>50</sup>	MTSD	16.0	0.948	69	10.47
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /Al <sub>2</sub> O <sub>3</sub> /Au <sup>195</sup>	-	10.67	0.789	-	5.07

Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>2</sub> NH <sub>2</sub> PbI <sub>2</sub> /TPB/Au <sup>196</sup>	MTSD	14.1	0 786	61	6 71
FTO-glass/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /Al <sub>2</sub> O <sub>3</sub> /spiro/Au <sup>198</sup>	OSPD	11.11	0.86	46	4.6
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>2</sub> NH <sub>2</sub> PbJ <sub>2</sub> /CuI/Au <sup>199</sup>	OSPD	17.8	0.55	62	6.0
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /CuSCN/Au <sup>51</sup>	OSPD	14.5	0.63	53	4.85
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /CuSCN/Au <sup>200</sup>	MTSD	19.7	1.016	62	12.4
Glass-FTO/c-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-v</sub> Cl <sub>v</sub> /CuSCN/Au <sup>201</sup>	OSPD	14.4	0.727	61.7	6.4
Glass-FTO/NiO/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-v</sub> Cl <sub>v</sub> /PCBM/Ag <sup>202</sup>	DSVD	14.2	0.786	65	7.26
Glass-FTO/c-NiO/m-NiO/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /PCBM/Al <sup>203</sup>	OSPD	4.9	0.83	35	1.5
Glass-ITO/c-NiO/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /PCBM/Al <sup>46</sup>	SDM	15.4	1.05	47	7.6
Glass-ITO/c-NiO/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /PCBM/BCP/Al <sup>186</sup>	OSPD	12.43	0.92	68	7.8
Glass-ITO/c-NiO/m-NiO/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /PCBM/BCP/Al <sup>205</sup>	SDM	13.24	1.040	68	9.51
Glass-FTO/c-NiO/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /PCBM/Au <sup>206</sup>	SDM	16.27	0.882	63.5	9.11
Glass-ITO/c-NiO <sub>x</sub> /m-NiO/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /PCBM/BCP/Al <sup>207</sup>	SDM	19.8	0.96	61	11.6
Glass-ITO/GO/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-x</sub> Cl <sub>x</sub> /PCBM/ZnO/Al <sup>209</sup>	OSPD	1.00	17.46	71	12.40
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /P3HT/Au <sup>210</sup>	OSPD	12.6	0.73	73.2	6.7
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /PCPDTBT/Au <sup>210</sup>	OSPD	10.3	0.77	66.7	5.3
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /PCDTBT/Au <sup>210</sup>	OSPD	10.5	0.92	43.7	4.2
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /PTAA/Au <sup>210</sup>	OSPD	16.5	0.997	72.7	12.0
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /PDPPDBTE/Au <sup>211</sup>	OSPD	14.4	0.8553	74.9	9.2
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /PCBTDPP/Au <sup>212</sup>	OSPD	13.86	0.83	48	5.55
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /PTB-DCB21/Au <sup>213</sup>	OSPD	15.35	0.888	64	8.7
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> :	inject	14.48	0.78	65	7.34
Li-salt/PANI/FTO-glass <sup>214</sup>	U				
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /PTAA/Au <sup>24</sup>	OSPD	21.3	1.04	73	16.2
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> Pb(I <sub>1-x</sub> Br <sub>x</sub> ) <sub>3</sub> /PTAA/Au <sup>24</sup>	OSPD	19.64	1.11	74.2	16.15
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /C <sub>60</sub> SAM	OSPD	14.9	0.81	-	6.7
/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-x</sub> Cl <sub>x</sub> /P3HT/Ag <sup>180</sup>					
Glass-FTO /c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /C <sub>60</sub> SAM	OSPD	15.6	0.88	51	6.84
/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-x</sub> Cl <sub>x</sub> /PCPDTBT/Ag <sup>180</sup>					
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /P3HT-MWNT/Au <sup>215</sup>	OSPD	14.8	0.76	57	6.45
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-x</sub> Cl <sub>x</sub> /P3HT/Au <sup>216</sup>	OSPD	12	0.93	58	9.3
Glass-ITO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-x</sub> Cl <sub>x</sub> /P3HT/Ag <sup>39</sup>	OSPD	20.8	0.921	54.2	10.4
Glass-ITO/c-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-x</sub> Cl <sub>x</sub> /P3HT+Li,D-TBP/Ag <sup>52</sup>	OSPD	19.1	0.98	66.3	12.4
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub>	OSPD	18.3	0.865	66.0	10.4
/spiro:FK209+Li,TBP/Au <sup>219</sup>	2514	160	1.000	- 1	11.0
Glass-FTO/c-T1O <sub>2</sub> /m-T1O <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub>	SDM	16.8	1.000	71	11.9
$\frac{(\text{spiro:MY11+L1, 1BP/Au^{220})}{(\text{spiro:MY11+L1, 1BP/Au^{220})}}$	OCDD	15.00	1.0(4		10.0
$Glass-FIO/c-IIO_2/CH_3NH_3POI_{3-x}CI_x$	OSPD	15.90	1.064	64	10.8
Class ETO/s TiO /m TiO /CIL NUL Dh	SDM	16.26	0.97	56	7.01
$\sqrt{\text{gnire} \cdot \text{Ru} \text{Pu} \text{Im} \text{TESI} \cdot \text{TRP} / \Lambda \text{g}^{222}}$	SDM	10.20	0.87	50	/.91
Glass-FTO/c-TiOe/m-TiOe/CH-NH-PhI	SDM	18	0.0	60	10.1
/spiro+spiro(TFSI).·L i TBP/Au <sup>223</sup>	SDIVI	10	0.9	00	10.1
Glass-FTO/c-TiO2/m-TiO2/CH2NH2PhJ2/no-	OSPD	21.2	1.02	77.6	16.7
spiro:Li TBP/Au <sup>25</sup>	ODID	21.2	1.02	//.0	10.7
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>2</sub> NH <sub>2</sub> PbI <sub>2</sub> /2TPA-2-DP/Au <sup>224</sup>	OSPD	16.8	0.91	67.1	91
Glass-FTO/c-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-x</sub> Cl <sub>x</sub> /DR3TBDTT/Au <sup>225</sup>	OSPD	15.3	0.95	60	8.8
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>2</sub> NH <sub>2</sub> PbI <sub>2</sub>	SDM	13.0	1.08	78.3	11.0
/KTM3+FK269+Li,TBP/Au <sup>226</sup>					
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /TTF-1/Ag <sup>227</sup>	SDM	19.9	0.86	64.4	11.03
Glass-FTO/c-TiO2/m-TiO2/CH3NH3PbI3/PNBA/Au <sup>228</sup>	SDM	17.5	0.945	68.9	11.4

Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /HTM1/Au <sup>229</sup>	SDM	18.1	0.921	68	11.34
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /HTM2/Au <sup>229</sup>	SDM	17.9	0.942	69	11.63
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub>	OSPD	20.2	0.89	69.4	12.4
/Py-C+FK269+Li,TBP/Au <sup>230</sup>					
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub>	SDM	20.74	0.92	66	12.51
/Triazine-Th-OMeTPA+FK102+Li,TBP/Au <sup>231</sup>					
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /Fused-F/Au <sup>232</sup>	SDM	17.9	1.036	68	12.8
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub>	SDM	19.1	1.05	65	13.2
/H101+FK102+Li,TBP/Au <sup>233</sup>					
Glass-FTO/c-TiO <sub>2</sub> /m-	OSPD	23.38	1.06	67	15.7
TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-x</sub> Cl <sub>x</sub> /IPFB/spiro/Ag <sup>234</sup>					
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /MMT/spiro/Au <sup>235</sup>	SDM	21.2	0.88	64	11.9
Glass-FTO/c-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-x</sub> Cl <sub>x</sub> /spiro/MoO <sub>x</sub> /Al <sup>236</sup>	OSPD	19.55	0.990	59.0	11.42
Glass-FTO/c-TiO <sub>2</sub> /m-ZrO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /graphite <sup>237</sup>	Drop	12.4	0.878	61	6.64
	coating				
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /m-ZrO <sub>2</sub>	Drop	13.23	0.841	63	7.02
/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /OMC+flaky graphite <sup>238</sup>	coating				
Glass-FTO/c-TiO <sub>2</sub> /ns-TiO <sub>2</sub> /m-ZrO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /graphite <sup>239</sup>	SDM	20.1	0.868	61	10.64
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /m-ZrO <sub>2</sub> /	SDM	20.9	0.921	67	12.9
$(CH_3NH_3)_{0.4}(NH_2CHNH_2)_{0.6}PbI_3/graphite^{240}$					
Glass-FTO/c-TiO <sub>2</sub> /m-ZrO <sub>2</sub> /(5-AVA) <sub>x</sub> (MA) <sub>1-x</sub> PbI <sub>3</sub> /graphite <sup>241</sup>	Drop	22.8	0.858	66	12.84
	coating				
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-x</sub> Cl <sub>x</sub> /spiro/CNT <sup>242</sup>	SDM	18.1	1.00	55	9.90
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-x</sub> Cl <sub>x</sub> /CNT <sup>242</sup>	SDM	15.46	0.88	51	6.87

Notes: For structures: c-, m-, np- and ns- refer to compact, mesoporous nanoparticle and nanosheet structure. For perovskite synthesis methods: SDM and OSPD mean sequential deposition and one step precursor deposition methods, respectively; while DSVD and MTSD are short for dual-source vapour deposition and modified two-step deposition method.

Table S3. A summary of hybrid perovskite solar cells' perform	mance parameters with different perovskite. (In
order of the appearance in	n main text)

	Method	J <sub>SC</sub>	V <sub>OC</sub>	FF	PCE
Structure (from substrate to back electrode)	S	[mA/c	[V]	[%]	[%]
		m <sup>2</sup> ]			
Glass-FTO /m-TiO <sub>2</sub> /Z907/CH <sub>3</sub> NH <sub>3</sub> PbBr <sub>3</sub> /I <sup>-</sup> ,I <sup>3-</sup> /Pt-FTO <sup>243</sup>	OSPD	11.7	0.54	54	3.4
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /PbS QDs/Au <sup>244</sup>	OSPD	24.63	0.3438	43	3.6
Glass-FTO/c-TiO <sub>2</sub> /m-Al <sub>2</sub> O <sub>3</sub> :Au@SiO <sub>2</sub>	OSPD	16.91	1.02	64	11.4
/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-x</sub> Cl <sub>x</sub> /spiro/Ag <sup>245</sup>					
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbBr <sub>3</sub> /PCBTDPP/Au <sup>212</sup>	OSPD	4.47	1.16	59	3.04
Glass-FTO/c-TiO <sub>2</sub> /m-Al <sub>2</sub> O <sub>3</sub> /CH <sub>3</sub> NH <sub>3</sub> PbBr <sub>3</sub> /PDI/Au <sup>249</sup>	OSPD	1.08	1.30	40	0.56
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbBr <sub>3</sub> /PIF8-TAA/Au <sup>23</sup>	OSPD	6.1	1.40	79	6.7
Glass-FTO /c-TiO <sub>2</sub> /nw-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>2</sub> Br/spiro/Au <sup>155</sup>	OSPD	10.12	0.82	59	4.87
Glass-FTO/np-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-x</sub> Br <sub>x</sub> /Au <sup>250</sup>	SDM	16.2	0.77	68	8.54
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-x</sub> Br <sub>x</sub> /PTAA/Au <sup>251</sup>	OSPD	19.3	0.91	70.2	12.3
Glass-FTO/c-TiO <sub>2</sub> /m-Al <sub>2</sub> O <sub>3</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-x</sub> Cl <sub>x</sub> /CBP/Au <sup>252</sup>	OSPD	4.0	1.50	46	2.70
Glass-FTO/c-TiO <sub>2</sub> /m-Al <sub>2</sub> O <sub>3</sub>	OSPD	19.63	1.015	62.6	12.5
/CH <sub>3</sub> NH <sub>3</sub> Pb(Br <sub>x</sub> I <sub>1-x</sub> ) <sub>3-y</sub> Cl <sub>y</sub> /spiro/Au <sup>253</sup>					
Glass-ITO/PEDOT:PSS	OSPD	14.9	0.99	68	10.0
/CH <sub>3</sub> NH <sub>3</sub> Pb(I <sub>0.8</sub> Br <sub>0.2</sub> ) <sub>3-x</sub> Cl <sub>x</sub> /PCBM/Bis-C60/Ag <sup>254</sup>					
Glass-FTO /np-TiO <sub>2</sub> /CH <sub>3</sub> CH <sub>2</sub> NH <sub>3</sub> PbI <sub>3</sub> /I <sup>-</sup> ,I <sup>3-</sup> /Pt-FTO <sup>256</sup>	OSPD	5.2	0.660	70.4	2.4
Glass-ITO/PEDOT:PSS/Cs <sub>0.10</sub> MA <sub>0.90</sub> PbI <sub>3</sub> /PCBM/Al <sup>257</sup>	OSPD	10.10	1.05	73	7.68

Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /20%SnF <sub>2</sub> -CsSnI <sub>3</sub> /m-MTDATA/Au <sup>258</sup>	OSPD	22.7	0.24	37	2.02
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /NH <sub>2</sub> CHNH <sub>2</sub> PbI <sub>3</sub> /P3HT/Au <sup>259</sup>	in-situ	18.3	0.84	50	7.5
	dipping				
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /NH <sub>2</sub> CHNH <sub>2</sub> PbI <sub>3</sub> /spiro/Au <sup>260</sup>	SDM	6.45	0.97	68.7	4.30
Glass-FTO/c-TiO <sub>2</sub> /NH <sub>2</sub> CHNH <sub>2</sub> PbI <sub>3</sub> /spiro/Au <sup>14</sup>	OSPD	23.3	0.94	65	14.2
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub>	SDM	21.2	1.003	70	14.9
/(CH <sub>3</sub> NH <sub>3</sub> ) <sub>0.6</sub> (NH <sub>2</sub> CHNH <sub>2</sub> ) <sub>0.4</sub> PbI <sub>3</sub> /spiro/Au <sup>261</sup>					
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub>	SDM	20.97	1.032	74	16.01
/NH <sub>2</sub> CHNH <sub>2</sub> PbI <sub>3</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /spiro/Au <sup>262</sup>					
Glass-FTO/c-TiO <sub>2</sub> /NH <sub>2</sub> CHNH <sub>2</sub> PbBr <sub>3</sub> /spiro/Au <sup>263</sup>	SDM	6.6	1.35	73	6.5
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> Sn <sub>0.5</sub> Pb <sub>0.5</sub> I <sub>3</sub> /P3HT/Ag/Au <sup>264</sup>	OSPD	20.04	0.42	50	4.18
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> Sn <sub>0.5</sub> Pb <sub>0.5</sub> I <sub>3</sub> /spiro/Au <sup>265</sup>	OSPD	20.64	0.584	60.32	7.27
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> SnI <sub>3</sub> /spiro/Au <sup>266</sup>	OSPD	16.30	0.68	48	5.23
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> SnIBr <sub>2</sub> /spiro/Au <sup>266</sup>	OSPD	11.73	0.82	57	5.73
Glass-FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> SnI <sub>3</sub> /spiro/Au <sup>267</sup>	OSPD	16.8	0.88	42	6.4

Notes: For structures: c-, m- and np- refer as compact, mesoporous nanoparticle structure. For perovskite synthesis methods: SDM and OSPD mean sequential deposition and one step precursor deposition methods, respectively.